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CARBONACEOUS MATTER IN METEORITES (ORGANIC COMPOUNDS, DIAMONDS, GRAPHITE)

by G. P. Vdovykin

"Nauka" Press, Moscow, 1967

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION • WASHINGTON, D. C. • APRIL 1970

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CARBONACEOUS MATTER IN METEORITES

(ORGANIC COMPOUNDS, DIAMONDS, GRAPHITE)

By G. P. Vdovykin

Translation of "Uglerodistoye Veshchestvo
Meteoritov (Organicheskiye Soyedineniya, Almazy,
Grafit)"
"Nauka" Press, Moscow, 1967

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

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CARBONACEOUS MATTER OF METEORITES: ORGANIC COMPOUNDS, DIAMONDS, AND GRAPHITE

G. P. Vdovykin

ANNOTATION: This book gives the first systematized generalization of the results of recent studies of the carbonaceous matter in meteorites (organic compounds, diamonds and graphite). Their distribution, chemical composition and structural characteristics are examined and the possible conditions for the formation of extraterrestrial matter and diamonds are discussed in relation to the origin of the meteorites containing them.

The book is interesting for various specialists and individuals concerned with the space chemistry of carbon and the structures of meteorites.

134 figures and 58 tables

Responsible Editor Academician A. P. Vinogradov

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FROM THE EDITOR

The matter in meteorites, both stony and iron, is constantly subjected to cosmic irradiation. This gives rise to nuclear reactions, resulting in a change in the composition of meteorite matter. The isotopic compositions of many chemical elements making up meteorites are changed. New isotopes, stable and radioactive, especially long-lived isotopes, etc., appear as a result of the spallation reaction. /5*

Thus, the matter in meteorites retains indicators of its origin. Meteorite matter is a sort of record of cosmic events over an enormous interval of time from the pre-planetary stage over the course of billions of years of their existence in space and down to the present day. We have every basis for believing that a detailed study of the matter in meteorites will make it possible to reconstruct the history of our Galaxy.

This book is concerned with one of the very interesting aspects of this problem; it deals with organic matter, diamonds and graphite in meteorites. Studies in this field are developing rapidly in many laboratories throughout the world. With each passing year more and more new data are appearing concerning the organic matter in meteorites. Data on the so-called carbonaceous meteorites which contain several percent of carbon are particularily interesting.

Quite recently much stir was caused by a number of articles devoted to the discovery of "organized elements" in stony meteorites; these were described by some authors as bacteria, by others by microalgae, etc. In other words, there are those who say that living organisms are transported to the earth with meteorite matter. However, a very competent group of scientists recently demonstrated that these microorganisms are the result of contamination of the matter of meteorites under terrestrial conditions, that is all these organisms were shown to be ancient inhabitants of the earth. No sooner had this stir ended, when another arose: it was stated that complex organic compounds, seemingly of biogenous origin, had been found in meteorites.

Prior to saying a few words on this subject, I wish to recall for the reader that the carbon in meteorites, particularily carbonaceous (rich in carbon) meteorites, is found in several stages: mixtures in organic compounds, bituminous matter, graphite, and sometimes diamond. Apparently, the primary matter of carbon was carbon in the graphite form. Diamonds, as shown experimentally, are formed as a result of impact, for example the impact of a meteorite against the earth or the collision of asteroidal bodies, when

*Numbers in the margin indicate pagination in the foreign text.

pressures and temperatures arise which correspond to the field of existance of a diamond. The organic compounds extracted from carbonaceous meteorites were complex, frequently aromatic and other compounds. They were found to include hydrocarbons, amino acids, multinuclear aromatic compounds and other complex organic substances. A common characteristic for all of them was the absence of optical activity. Many investigators have postulated that these organic compounds have a biogenous origin. However, it was soon demonstrated that mixtures of H_2O with different gases such as CO_2, CO, CH_4, SO_2, HCl and others, which appeared first at the earths surface in the form of volcanic gases (even now they are ejected by colcanos), after being irradiated by ultraviolet rays, electrons, neutrons or protrons, form extremely complex organic substances similar to those found in carbonaceous meteorites, that is, that in these experiments there was a radiogenic synthesis of complex organic compounds.

Thus, the nature of the organic compounds discovered in carbonaceous meteorites was clarified. All of this of particular interest for understanding the conditions under which life on earth first developed.

In actuality, if we assume that in the distant past, 3 or 4 billion years ago, the earth was either subjected to irradiation by a more intense cosmic ray source or that it had a less dense atmosphere (which is very probable, since oxygen and nitrogen in the present day atmosphere had a secondary origin, having developed later by a biogenous process), then at its surface, nuclear reactions transpired under the influence of cosmic radiation, as in meteorites, do so to speak "to the cosmic window". As a result of radiogenic synthesis a great many varieties of complex organic substances could appear at the earth's surface. This was the substrate on which life later developed. The thick gas envelope created by it put an end to radiogenic synthesis at the earth's surface.

All these thoughts arise when one begins to become better acquainted with the chemistry of carbons in different cosmic bodies.

Therefore, it seems to me that this book by G. P. Vdovykin, being a primary source of new data on carbons in meteorites, will be read by a great many researchers.

Academician A. P. Vinogradov

FORWARD

Meteorites have long attracted the attention of researchers. Their mineral and chemical composition and structure have been clarified as a result /7 of many years work and classifications of meteorites have been proposed. The science of meteoripics is concerned with their study.

Meteorites are being studied at the present time very intensively. Meteorites are being studied with the use of the most modern methods: X-ray spectro microanalysis, the mass spectrometer method, gamma-resonance spectroscopy, etc. As a result, information has been obtained concerning the chemical composition of individual mineral components of meteorites, the isotopic composition of elements, etc. This information is of importance not only for clarifying the evolution of meteorite matter. It is also important because it provides us with information concerning conditions in space where meteorites were prior to their falling to the earth.

On the basis of all these and other data a new science, space chemistry, is beginning to develop more intensively. One of its branches, organic space chemistry, includes the study of organic compounds in meteorites. These organic components, as is now known, are of chemical origin rather than of biogenous origin. In contrast to terrestrial organic matter, the organic compounds of meteorites have not passed through the stage of living matter. However, with respect to the composition of individual components, they are somewhat similar to the organic compounds in rocks.

Academician V.I. Vernadskiy classified such substances as primary carbonaceous compounds. He particularily emphasized the importance of investigating primary organic compounds for understanding the prophecies of evolution of living matter. However, earlier there was no possibility to do this, even though organic compounds were known to exist in meteorites even during the last century. Only in recent years have they been studied in greater detail.

The study of other forms of carbon, especially diamonds and graphite, in meteorites also is of definite interest.

Diamonds have long been known to exist in meteorites. However, earlier no systematic study of the diamonds in meteorites was made. By analogy with terrestrial diamonds, it was assumed that the diamonds in meteorites were formed at high static pressures and temperatures such as have existed in the central parts of the planetary body from which the meteorites were formed.

Beginning in 1955, investigators began experimental studies of the conditions of synthesis of diamonds. It was found that diamonds can be formed when subjected to a brief dynamic load. Study of the peculiarities of struc- /8 ture of meteorites containing diamonds and the structural peculiarities of

their diamonds revealed that the diamonds in meteorites were formed as a result of impact, either at the time of falling of the Canyon Diablo diamond-bearing meteorite to the earth, or at the time of collision of astroidal bodies, when diamond-bearing ureilites were formed.

In the detailed investigation of the diamonds found in meteorites, the existence of a hexagonal diamond was recently established, in addition to diamonds with a cubic structure. Hexagonal diamonds are not found on earth, but they were obtained experimentally as a result of impact.

On the other hand, in addition to graphite with a hexagonal structure meteorites are known to contain graphite having a cubic morphology, cliftonite. It is believed that cliftonite was formed during the decay of iron carbide (cohenite) under low-pressure conditions. Study of the multiple types of carbon in meteorites is of great importance for clarifying the problem of the origin of meteorites.

Although the structure of mineral composition of meteorites has been studied rather well, the origin of meteorites still remains unclear. In studies of their origin, it is customary to take into account their structure and mineral composition; thus silicate metallic and other phases are taken into account. Only recently have researchers begun to take into account the carbonaceous substance, especially organic compounds which are present in meteorites. In contrast to the mineral components of meteorites, organic compounds are extremely sensitive to environmental changes. The composition of the organic compounds can be used to judge the physical chemical conditions under which the matter in meteorite was formed.

Clarification of the origin of meteorites is also important for explaining the processes of evolution of matter on the earth. Academician A. P. Binogradov formulated the idea of phase smelting of the matter in the earth's crust. He assumed that the matter in the earth's metal is similar in composition to the silicate fraction of chondrites, corresponding to the matter of the solar system.

Some authors feel that meteorites which are most enriched with organic compounds (so-called carbonaceous chondrites) represent the initial matter from which ordinary chondrites were formed. The chemical composition of carbonaceous chondrites, except for volatile elements, is very close to the composition of other chondrites. It was important to make a more detailed study of these meteorites containing an increased quantity of volatile elements, in particular, to study the organic components of these meteorites.

The study of the organic compounds in meteorites involves great difficulties. Carbonaceous chondrites containing organic compounds are so few in number that the problem has repeatedly arisen of establishibg international control of the use of these meteorites for research purposes.

The principal difficulty in studying the organic components was the small quantity of meteorite matter of this type, but finally dispersed distribution of organic matter in meteorites, making difficult its separation

for study, and also the possibility of terrestrial contamination. Accordingly, /9
their study became possible only recently when methods had been developed for investigating microquantities of organic compounds.

The author presented his first report on the minerology of meteorites in 1952.

The author has been studying carbonaceous matter in meteorites since 1957. Such studies have been made in foreign countries since that time. However, until recently foreign scientists studied primarily that part of the organic components which is extracted from meteorites by organic solvents. This bituminous matter is somewhat similar to the organic matter in terrestrial rocks which is extracted in much the same way. On this basis some researchers surmised that the organic compounds in meteorites, like the similar compounds in terrestrial rocks, also have a biogenous origin. Microparticles in carbonaceous chondrites, morphologically similar to spores were regarded as remnants of such forms of life in space. However, it was found that these particles for the most part are mineral formations.

A discussion of the nature of meteorite microparticles similar to extra terrestrial life forms ("organized elements") was held at a special symposium in 1962; the results of the discussion were published in the journal Nature. The nature of the "organized elements" was examined at a conference called New York Academy Sciences on 30 April - 1 May 1962, at the International Congress on Geochemistry of Organic Matter in 1962 in Milan and a conference in 1965 in Washington.

The nature of the organic matter in meteorites has been discussed at a number of conferences. A special session at the Eleventh Meteorite Conference held in Moscow in May 1964, was devoted to this problem.

The results of study of organic compounds and diamonds in meteorites were discussed at a special session of the Section on Space Chemistry at the Twentieth International Congress on Theoretical and Applied Chemistry in July 1965 (in Moscow).

The importance of carbonaceous chondrites containing organic matter for clarifying problems involved in the origin of life and the origin of meteorites was discussed at a special conference of the Commission on Cosmogony of the Astronomical Council Academy of Sciences USSR in 1962 (Moscow), at the Twelveth Meteorite Conference in 1966 (Novosibirsk), at a session of the Society Conference Biochemical Division of the Adademy of Sciences USSR in 1966 (Moscow), and at the Meteorite Conference in 1966 (Washington). Carbonaceous chondrites and ureilites containing diamonds were discussed in particularily great detail at the International Congress on the Origin of Elements held in Paris in May 1967.

The carbonaceous matter in meteorites is of interest to astronomers, specialists working in the fields of descriptive meteoritology, space chemistry, geochemistry, geology, biochemistry, exobiology and even micropaleontology and microbiology. The results of study of diamonds and graphite

in meteorites can be used by geologists studying the conditions under which diamonds and graphite were formed in nature by specialists who are studying /10 the structure of diamonds, by physicists engaged in synthesizing diamonds, by specialists studying shock waves, and the results of study of the graphite in iron meteorites can be used by specialists in the field of casting, etc.

Recently the need has arisen for the most thorough examination of the results which have been obtained from studying the carbonaceous matter in meteorites.

In this book the author has presented his own findings and data in the literature concerned with different methods for studying the carbonaceous matter in meteorites, that is the organic matter in carbon chondrites, diamonds in ureilites, and the graphite in iron meteorites. These studies made it possible to obtain a clear answer to the question as to whether the organic matter in meteorites has an extraterrestrial abiogenous nature and to make an attempt to examine the possible conditions under which carbonaceous matter and the meteorites containing it are formed.

The author expresses appreciation to everyone who manifested an interest in this study and who rendered assistance.

INTRODUCTION

Concise Information on Meteorites

Meteorites have been studied for a long time and many investigations have been devoted to this subject. Cohen (1894, 1903, 1905) published a good summary of investigations of meteorites. Detailed studies of the structure of meteorites, their mineral composition and metamorphism have been given by Wahl (1907, 1910, 1952), Prior (1916, 1920), Merrill (1921, 1930), and other investigators. The petrography of many meteorites, including pallasites, was investigated by P. N. Chirvinskii (1949a). The literature on meteorites published between 1491 and 1950 has been assembled into a bibliography published in 1953 edited by Brown (1953). A catalogue of meteorites was published in this same year by Prior, supplemented by Hey (1953). In our country a large number of papers concerned with meteorites is published in a special collection of articles Meteoritika whose individual numbers have appeared since 1941, and also in the journal Geokhimiya. /11

A number of monographs on meteorites have been recently published. Ye. L. Krinov (1948, 1955, 1963a) has collected and systematized a great amount of data on the conditions of falling, morphology, mineral composition and structure of meteorites. The results of a detailed mineralogical investigation of many meteorites have been presented in studies by A. N. Zavaritskiy and L. G. Kvash (1952) and A. N. Zavaritskiy (1956). The petrography of meteorites is covered in articles by L. G. Kvash (1948, 1950, 1956, 1958, and others). Many of the studies by A. P. Vinogradov (1954, 1958, 1959b, 1962a, b, c, 1964a, 1965) deal with the chemical composition of meteorites. The chemistry of meteorites in relation to their origin has been considered in many studies by A. A. Yavnel' (1955-1965, and others). The effect of cosmic radiation on the matter in meteorites has been investigated by A. K. Labrukhin (1965a, b, 1966; Labrukhin, Kolesov, 1965). A number of meteorites have been investigated mineralographically by I. A. Yudin (1956, 1958, 1965, and others). Recently a number of reviews on meteorites have been published by Wood (1963b), Anders (1964), and B. Yu. Levin (1965), as well as a book by Mason (1963a), translated in the Russian language.

At the present time there are three commonly used classifications of meteorites: 1) The Roze - Chermak - Brzhezina and 2) The A. N. Zavaritskiy classification, based on the mineral composition and structure of meteorites, and 3) Prior classification, based on the chemical composition of meteorites. We now will give, an abbreviated form, the A. N. Zavaritskiy classification (Zavaritskiy, Kvasha, 1952), which is used in our book as a matter of convenience in reverse order. The following types of meteorites are defined in this classification.

1. Stony meteorites:

a) Chondrites (unaltered, crystalline, and carbonaceous);

b) chondrites [nonfeldspathic - ureilites, peridotitic (chassinites, amphoterites, rodites, naclites) and pyroxenitic (chladnites, bustites, angrites) and feldspathic - eucrites, sherghottites, howardites].

2. Iron-stony meteorites - achondritic (mesosiderites and lodranites) and ferroginous chondrites (grahamites).

3. Stony-iron meteorites (siderolites) - pallasites, siderophyres. /12

4. Iron meteorites - hexahedrites and nickel-poor ataxites, octahedrites, and nickel-rich ataxites.

By 1963 1,684 meteorites were known (Prior, Hey, 1953; Krinov, 1964). According to the data published by A. P. Vinogradov (1965), by 1965 the total number of meteorites had reached 1,759. Less than half of them are meteorites whose falling was observed (764 meteorites); the others were found (995 meteorites). Most of the meteorites are of the stony type (they account for about 93% of the falls); among the stony meteorites most are chondrites (approximately 86%). Iron meteorites account for about 6% of the falls (Wood, 1963b). The largest of all the known meteorites is the Hoba[1] iron meteorite which was found in Southwest Africa. It weighs about 60 tons. The lower limit of weight for meteorites can vary. Ye. L. Krinov (1963b) found a piece which weighed only a few milligrams among the individual parts of the giant Sikhote-Alinskiy meteorite shower. It should be noted that in addition to meteorites a great quantity of cosmic matter falls on the earth in the form of spherelets with a diameter of a few microns; this is known as cosmic dust. In addition to meteorites and cosmic dust, tektites possibly have an extra - terrestrial origin (Tektity, 1966).

With respect to chemical composition iron meteorites consist for the most part of iron (on the average 90.8%, but also nickel (8.5%). A higher nickel content is characteristic only of meteorites; this distinguishes them from terrestrial objects. In stony meteorites there is a predominance of silicon (on the average 21.0%), iron (15.5%), magnesium (14.3%), and also oxygen (41.0%) (Krinov, 1955; Heide, 1957, and others).

The principal mineral components of meteorites are ferronickel, found in iron meteorites, and the ferromagnesial silicates in stony meteorites, represented primarily by olivine and pyroxenes, and also plagioclases. The number of minerals (about 50) found in meteorites is limited in comparison with the number found in rocks. Most of the minerals in meteorites are also known on earth.

Stony meteorites are classified as chondrites and achondrites. The predominant type of meteorites, chondrites consist of rounded mineral

[1] For a list of meteorites see the end of the book.

formations called chondrules (Figures 1 and 2) [1], whose diameters range from 0.5 to 1-2 mm and which do not exceed 3-4 mm. Chondrites sometimes make up entire meteorites; examples of such natural aggregates of chondrules are the Mikol'skye and Saratov chondrites (Dvasha, 1965). However, in most cases chondrules are distributed among fragments of mineral grains. Most chondrites have been recrystallized (Figures 3 and 4) a great many chondrites have a breccia-like appearance.

The chondrules in meteorites, frequently polymineral, consist of olivine and pyroxene, and ferronickel as well. Monomineral chondrules are encountered less frequently. Many studies and review articles have been devoted to the structure of chondrules. A. N. Zavaritskiy (1948b, c) defines the following structural types of chondrules: completely crystalline, gratelike, complex gratelike, radio, microporphyritic, microlithic, and also glassy and iron.

In 1916 Prior (1916) in his chemical classification distinguished among chondrites those of the following types: carbonaceous enstatitic and ordinary (bronzite and hypersthene). He noted an interrelationship between the /14
content of iron in silicates and the content of nickel in chondrites. This was expressed in Prior's rule: "the smaller the ferronickel content in chondrites, the richer it is in nickel and the richer is the iron content in magnesian silicate" (Prior, 1916).

In 1953 Urey and Craig published a summary of chemical analysis for 350 meteorites. They demonstrated that on the basis of the total iron content all chondrites can be divided into two groups: ferronickel (L-group) with a total iron content averaging 22.33% and a high-iron group (H-group) in which the total iron content averages 28.58%.

The chemical interrelationships between different types of chondrites
were investigated by A. A. Yavnel' (1958a), Ringwood (1963), Mason (1963a, b, /15
1965), Keil and Fredriksson (1964), and others. Recently VanSchmus and Wood (1967) proposed that chondrites be divided into a number of groups on the basis of their petrological characteristics and chemical composition.

A common characteristic of achondrites is their lack of achondritic structure and usually, a low ferronickel content. These meteorites are represented by the nonfeldspathic and feldspathic groups. Among the feldspathic achondrites we find real tuff (eucrites, howardites). The basic minerals in nonfeldspathic achrondrites are olivine and rhombic pyroxenes, whereas in ureilites the principal minerals are olivine and monoclinic pyroxenes. Feldspathic achondrites consist primarily of plagioclases and monoclinic pyroxenes of the enstatite - augite series. L. G. Kvasha (1959)

---

[1] The petrographic section (Figure 2) was prepared from a sample of the Krymka meteorite of 1701 on a polybutyl methacrylate cement which was non-luminescent in ultraviolet light. In ultraviolet light (λ = 365 mμ) olivine grains are slightly luminescent; this was probably caused by effects in the crystal lattice.

Figure 1. Olivine Chondrule of Microlitic Structure from the Unmodified Saratov Chondrite. The prepared chondrule was mounted in epoxy resin and polished. Reflected light.

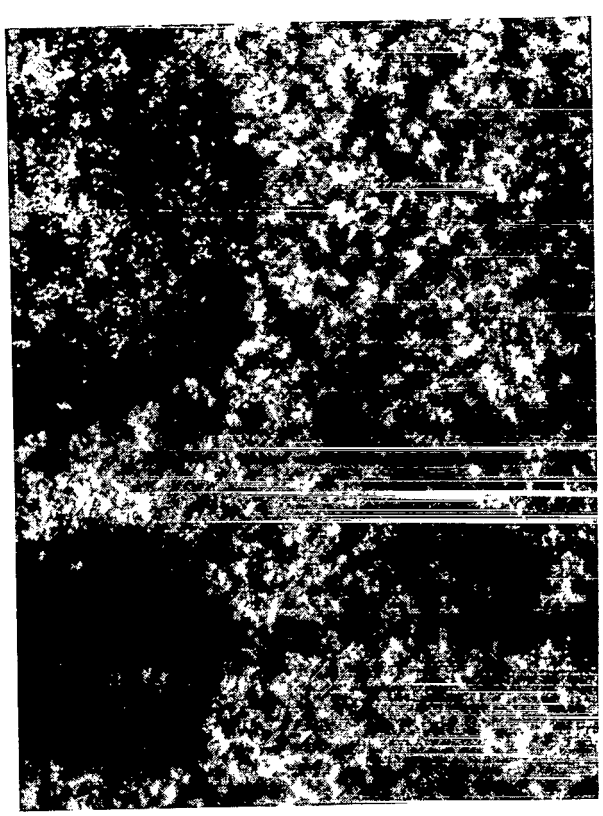


Figure 2. Microphotograph of a section of the Krymka Meteorite, a Slightly Modified Chondrite of a Dark Variety. The greater part of it consists of a fine grain aggregate of mineral grains. Traces of chondrules which have fallen out during polishing can be seen. Magnification 280

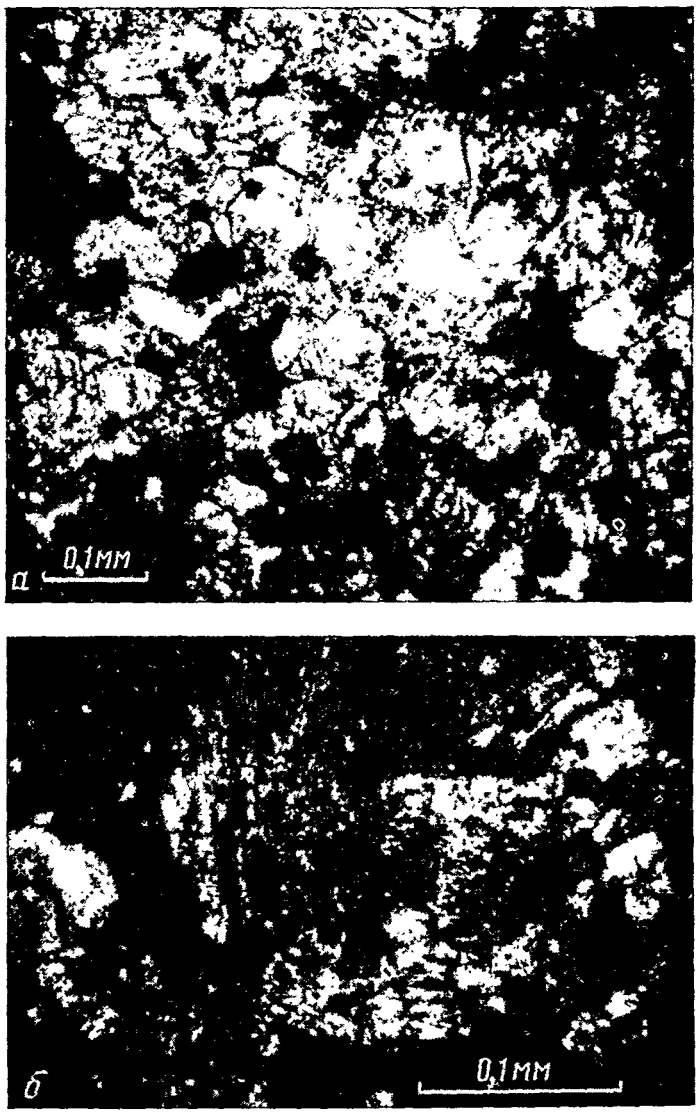


Fig. 3. Kulp Crystalline Chondrite. Section 95. a) nonuniform granular aggregate of silicates and opaque minerals; relicts of chondrules are distinguishable; b) silicate chondrule with a complex - grated structure.

and Moore (1962) have considered the petrochemistry of achondrites.

Stony-iron and iron-stony meteorites, of which only 79 are known to exist (Vinogradov, 1965), consist of silicates and ferronickel. They also contain troilite, schreibersite and other minerals. Plagioclase appears in

mesosiderites. The olivine in these meteorites is found in the form of angular or rounded grains up to 10 mm in diameter or more. In some of them (for example, in pallasites, rather well circumscribed olivine crystals have been observed.

Iron meteorites consist of kamacite, the α-phase of a ferronickel alloy with a body-centered lattice and taenite, the γ-phase with a face-centered lattice. In a fine mixture kamacite and taenite produce plessite. Iron meteorites can be classified into different types. The greater part of these meteorites is represented by octahedrites, characterized by a Widmanstatten structure (named in honor of the Four Austrian Widmanstatten), a regular coalescence of platelets of kamacite, oriented at a definite angle in an octahedron with taenite. In octahedrite it is the nickel content which is responsible for the coarseness of the Wedmanstatten structure.

The structure of iron meteorites, their classification and origin, have been examined by many researchers, in particular by A. A. Yavnel' (1961, 1964b, and others).

Fig. 4 Gilgoin Crystalline Chondrite. A silicate chondrule, broken into individual pieces, is present in the opaque basic mass. Section 2286, magnification 280.

Other peculiarities in the composition, structure, and radiogenic and radiation age of meteorites, discussed in the appropriate literature, are evidence of the quite complex processes which transpire in the history of development of meteorite matter. /17

Fig. 5 Total Carbon Content in Chondrites and Ureilites.
The petrological types of chondrites are plotted along the x-axis. In conformity to the classification formulated by Van Schmus and Wood (1967); C-total carbon; a-enstatic chondrites; b-carbonaceous chondrites; c-chondrites of H-group; d-chondrites of L-group; e-chondrites of LL-group; f-nonequilibrium chondrites of the corresponding groups; g-ureilites (Moore, Lewis, 1966a-c; to be reproduced from Hayes, 1967).

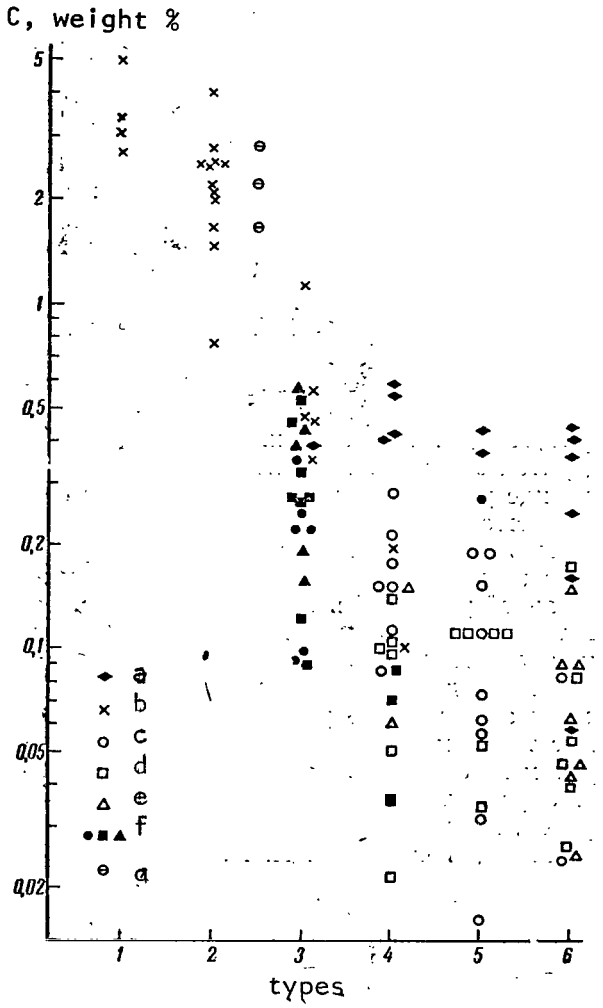
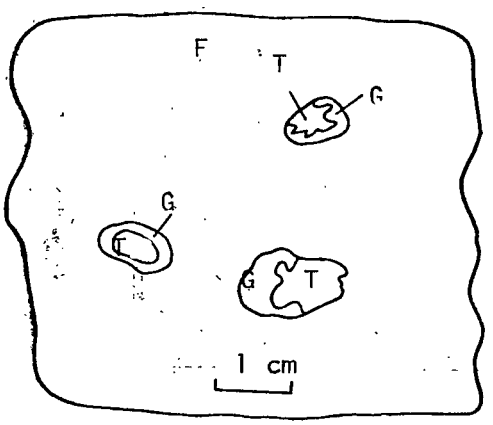


Fig. 6 Relationship between Graphite (G) and Trilite (T) in Carbonaceous-sulfide Nodules in Coarse-structured Odessa Octahedrite (drawing from polished section 2,270).
F (ferronickel).



Carbon Content in Meteorites

The mean carbon content in the most common meteorites, chondrites, has been estimated at 0.04% by A. P. Vinogrodov (1962c). This same figure is used by A. A. Yavnel' and M. I. D'Yakonova (1958) as the calarke of carbon concentration in the silicate phase of meteorites. B. Yu. Ledin, et al (1956) cite data on the carbon content in the silicate phase of meteorites (0.034%) and in the metallic phase (0.108%); the mean carbon content in meteorites, according to their data, is 0.044%.

It should be noted that determination of the mean carbon content in chondrites, and in meteorites in general, is a time-consumming task, since the carbon in different types of meteorites has a nonuniform distribution; moreover, it appears in different forms.

Carbonaceous chondrites have the highest carbon content (up to 4.8%). In these meteorites the carbon is present primarily as organic compounds.

The least carbon is found in the ordinary chondrites, from 0.01% (Rich Mountain meteorite) to 0.37% (Breitscheid meteorite). In these meteorites the carbon is represented for the most part by graphite. In enstatitic chondrites the carbon content is greater, from 0.10% (Saint Sauveur meteorite) to 0.43% (Indarkh meteorite) (Chirvinskiy, 1949b; Mason, 1966, and others). In these meteorites it is also represented by graphite, and in some cases such as in the Abee chondrite, it is found in cohenite.

Moore and Lewis (1965, 1966a-c) recently determined the total carbon content in different types of chondrites (Figure 5).

Among the achondrites it is ureilites which have the highest carbon content (averaging about 2%). In ureilite the carbon is represented primarily by diamond and graphite.

In pallasites the carbon content is from 0.01% (Springwater meteorite) to 0.45% (Bragin meteorite). The mean carbon content in these meteorites is 0.09% (data from P. N. Chirvinskiy (1949a) based on 22 determinations). For the most part the carbon was concentrated in the metallic phase.

In iron meteorites the mean carbon content according to Buddhue's computations (1946) is 0.08%, whereas according to Perry's data (1944) it is from 0.08% (in nickel-poor ataxites to 0.60% (in octahedrites); the mean content in iron meteorites is 0.11% (based on 360 analyses). Carbon is represented primarily by graphite, and also is present in carbides (cohenite and moissanite). In addition, diamond is known in the Canyon Diablo meteorite.

In octahedrites, which have the highest carbon content, it is found in the form of small platelites or grains of graphite which are present

for the most part between kamacite formations. In these meteorites, particularily in the coarse-structured octahedrites, carbon sometimes forms nodules up to 10 cm in diameter. Such nodules, containing carbonaceous matter, are known in the Yardynlinskiy Burgavli, Odessa, Magura, Toluca, Cosby's Creed, Canyon Diablo and other meteorites. In these nodules the carbon is frequently associated with troilite and other accessory minerals: schreibersite, cohenite, and others. Such nodules in individual parts of octahedrites (for example, in the Toluca meteorite) account for as much as 30% of the volume. Figure 6 shows examples of different relationships between graphite and troilite in carbonaceous-sulfide nodules of the Odessa meteorite.

ORGANIC MATTER IN CARBONACEOUS CHONDRITE

CHAPTER ONE

HISTORY OF THE STUDY OF ORGANIC COMPOUNDS IN METEORITES

As early as 1806 the presence of carbon was established in the Aláis chondrite by Thénard (1806); this was the first such find in meteorites. Thirty years later the Swedish chemist J. J. Berzelius (1834) for the first time found in this same meteorite organic compounds whose quantity attained 0.95% of the weight of this meteorite. Later many researchers noted that carbonaceous chondrite has a specific "bitumen-like" odor immediately after falling. Organic matter was extracted from a number of meteorites and sometimes a more detailed analysis of this matter was made. /18

In 1858-1859 Wohler (1858, 1859) studied bitumen-like compounds in the Cold Bokkeveld and Kaba carbonaceous chondrites. He noted that when the pulverized samples of these meteorites are boiled in alcohol the solvent containing an extract of the Cold Bokkeveld meteorite was colored a pale yellow color whereas the solvent with an extract of the Kaba meteorite remained without color. After the solvent had been dried, Wohler noted that yellowish and colorless, resin-and wax-like substances with an aromatic odor remained. Its content attained 0.25% of the weight of the meteorite. After this substance was dissolved in ether and the solvent was subsequently evaporated a crystalline substance was formed. During heating in air it became volàtile: in a sealed glass tube it fused with the formation of a petroleum-like, and then a coal-like substance. It did not dissolve in alkali. Wohler compared this organic substance with ozokerite and scheererite and Shepard proposed for it the name kabaite and described it as meteoritic petroleum.

The bituminous matter in the Aláis meteorite was investigated in 1863 by Roscoe (1863). After extracting this meteorite with ethyl ether he obtained 0.64% organic matter; this recrystallized into acicular crystals with a melting point of 114°. This matter was dissolved in ether and carbon bisulfide; with heating it was sublimated with the formation of a carbon-like residue. An elementary microanalysis of 7.8 mg of matter revealed the following composition; C 28.57%, H 5.29%, S 66.14%. Roscoe felt that the extract represented a mixture of hydrocarbons and free sulfur. After excluding the sulfur the composition of the matter actually corresponded to hydrocarbons: C 84.37%, H 15.63%.

In 1876 and 1879 Smith (1876, 1879) used ether for extracting organic matter from the Aláis and Orgueil meteorite. After the solvent was evaporated a colorless organic substance remained which recrystallized into acicular crystals. It also had a aromatic odor and dissolved in carbon disulfide. In a sealed glass tube it fused at 115-120°, but with greater heating part of it /19 became volàtile, leaving behind a coal-like substance; yellow sulfur droplets

were formed during condensation of the vapors. An elementary analysis of the organic matter found in the Orgueil meteorite revealed the following composition: C 15.00%, H 3.00%, S 79.65%. Smith, in contrast to Roscoe, believed that the sulfur in the extract of this meteorite was not free but enters into composition with sulfur-bearing organic compounds hypothetically of the composition $C_4H_{12}S_5$. Smith proposed that this compound be called celestialite.

An ozocerite substance with a specific odor was extracted with alcohol in 1889 by Meunier (1889) from the Migei carbonaceous chondrite[1]. The content of the extract was 0.056%. However, in the chemical analysis Meunier noted that in actuality the content of organic matter in the Migei meteorites was higher -- 4.72%. At the same time, Yu. I. Simashko (1890) also demonstrated a high content of carbonaceous matter in this meteorite (about 4%), as well as the presence of bituminous matter in a quantity 0.230%, soluble in alcohol and ether, but insoluble in water and hydrochloric acid. The extract which Simashko obtained from this meteorite was a viscous yellowish mass; it fused readily in the air and in part became volatile leaving behind a dark oily substance. Morphologically the extract represented aggregates of tiny needles and platelets distributed among small viscous lumps. Simashko called this substance erdelite. He postulated that it was formed chemically in the meteorites under conditions of relatively low temperature. Later, P. G. Melikov and L. D. Krshishanovskiy (1896) in a chemical analysis found that the Migei meteorite contained 2.63% amorphous carbonaceous matter and they also noted the presence of organic compounds similar to the heavy fractions of petroleum. In addition, they extracted 10 g of pulverized meteorite matter with distilled water and in the water extract they found 0.53% organic compounds, primarily fatty acids, volatile with water vapor.

In 1888 Friedheim (1888), after extraction with ether, obtained from the Nogoya carbonaceous chondrite a yellowish mass with an aromatic odor (its content in the meteorite was 0.21%). With cooling of this bituminous matter to 0° it congealed and resembled paraffin, but with heating above 200° it was carbonized.

Plon (Tschermak, 1878), in a study of the Grozmaya carbonaceous chondrite, found that it contained 0.68% C and 0.17% H; he assigned these two organic compounds. Tschermak (1870) noted that the dark color of the Renazzo and Ornans carbonaceous chondrite was caused, at least in part, by carbon compound.

Thus, in a number of carbonaceous chondrites organic compounds were detected during extraction with organic solvents. The bituminous compounds

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[1]R. A. Prendel (1897) noted that when this meteorite was pulverized in a mortar there was a singular "bituminous" odor. A specific "garlic" odor was also sensed by witnesses of the falling of carbonaceous chondrites (Krinov, 1931).

extracted from meteorites sometimes had a hydrocarbon composition and with certain degree were similar to ozocerite.

Some researchers attempted to extract organic compounds from carbonaceous chondrite by other methods. Cloez (1864a, b) after processing the Orgueil meteorite with concentrated HCl and a weak solution of KOH, obtained a carbonaceous residue (6.41%); he stated that it was apparently of an amorphous structure. A composition of this matter was: C 63.45%, H 5.98%, O 30.57%. Cloez felt that it was similar to terrestrial humus substances.

The carbonaceous matter of the Orgueil meteorite was also investigated by Berthelot (1868). He noted that this substance when processed with KCl and concentrated H_2SO_4 does not form graphitic acid and with respect to its /20
composition is an organic substance containing hydrocarbons Berthelot felt that the organic substance in the Orgueil meteorite is rather similar to a terrestrial organic substance.

In addition to carbonaceous chondrites, during the last century organic matter was also noted in meteorites of other types. For example, the Goalpara ureilite, according to an analysis made by Teclu (1870), contained 0.73% C and 0.13% H. Teclu felt that the carbon and hydrogen in this meteorite form organic compounds which had the compositon: C 84.88%, H 15.12% (Cohen, 1894).

In rare cases organic compounds were observed in ordinary chondrites. Will and Pinnow (1890) used ether in separating out an organic substance from the ordinary Carcote chondrite, which when heated was carbonized, and then completely combusted. Organic solvents were used in separating out from the Hessle chondrite a viscous substance of a brownish color with the content 72.6% C, 5.3% H and 22.1% O (Nordenskiöld, 1879). Lindstrom (Cohen, 1894) postulated that its composition corresponds to the formula $NC_8H_9O_2$. Bituminous matter of a yellowish-white color (0.56%), soluble in benzene, was also extracted from the Collescipoli chondrite (Trottorelli, 1891).

Organic compounds were also observed in iron meteorites, in their carbonaceous nodules. Smith (1876, 1879) after using ether to extract carbonaceous nodules of the Cosby's Creek, Cranbourne, and other iron meteorites, obtained a bituminous substance with a specific "garlic" odor. This substance formed acicular crystals. It dissolved readily in carbon bisulfide. Konig (Cohen, 1894) noted that the carbonaceous nodules of the Canyon Diablo iron meteorite apparently also contained similar organic compounds. Konig processed the carbonaceous matter from this meteorite using aqua regia; after such processing the alkaline extract had a dark brown color.

The results obtained during the last century in determining gases (H_2, CO_2, CO, N_2, CH_4), released during the heating of meteorites are also of interest (Cohen, 1894). When meteorites were heated to 500° the greatest quantity of gases, among which carbon dioxide and methane were most permanent,

Table 1

Content and Average Composition of Gases Released from Stony and Iron Meteorites When Heated to 500°

(In % of Total Volume of all Released Gases)

	Number of analyzed samples of meteorites	Volume of released gas, cm^3	H_2	CO_2	CO	N_2	CH_4
Stony meteorites	10	0.49-3.49	27.85	58.14	9.30	2.12	3.16
Carbonaceous chondrites:							
Orgueil.....	1	9.80	-	76.05	11.67	3.33	8.93
Cold Bokkeveld.....	1	25.23	0.38	93.11	2.42	0.84	3.25
Iron meteorites	9	0.97-6.38	57.70	7.93	26.32	7.41	0.77
Magura meteorites	1	47.13	18.19	12.56	67.71	1.54	-
Toluca meteorite (graphite)	1	7.25	2.50	91.81	-	0.10	5.40

was released from carbonaceous chondrites (Table 1). Some of these gases were formed during the compositon of organic compounds during the process of meteorite heating.

Thus, even during the last century organic compounds were noted in a number of meteorites. The extracts obtained from these meteorites by using various organic solvents (ether, alcohol, benzene, etc.) had a viscous consistency, a yellowish or brownish color, frequently formed acicular crystals and had a "bituminous" odor. An elementary analysis revealed that they had a hydrocarbon composition. However, many researchers erroneously assumed that this bituminous substance in meteorites represents an individual compound and on the basis of an elementary composition attempted to compute an empirical formula for this compound. However, as early as 1894, Cohen, in reexamining the results of these studies, postulated that the organic compounds in meteorites are represented by at least three types: Organic compounds consisting of carbon and hydrogen. Compounds containing carbon, hydrogen and sulfur, and compounds containing carbon, hydrogen and oxygen. /21

Only recently did it become known that the organic substance in meteorites in reality is a very complex mixture of individual compounds belonging to various classes.

In addition to data on the quantitative content, and in individual cases on the elementary compositon of bituminous compounds, the publication appearing in the past century also contained some information on the part of the carbonaceous matter in meteorites which could not be extracted with

solvents. However, information on the bituminous matter and particularly on the remaining part of the carbonaceous matter in meteorites for the most part was of a qualitative nature (Vdovykin, 1964a).

With respect to the origin of the organic compounds in meteorites, Wöhler (1859), and Cloëz (1864) assumed that these compounds have a biogenous nature. However, as early as 1834 Berzelius emphasized that the presence of carbon-bearing compounds in meteorites gives no basis for postulating their organic origin. Berthelot (1868) and Yu. I. Simashko (1890) also assumed that the organic compounds characteristic of carbonaceous chondrites have a chemical, abiogenous origin.

At that time it was already known that organic compounds can be abiogenously synthesized. We recall that as early as 1828 Wöhler for the first time produced urea (carbamide) in a chemical process by the heating of a mixture of CO_2 and NH_3. In 1868 A. I. Bazarov developed the synthesis of urea from CO_2 and NH_3 without heating under pressure (Gonikberg, 1960).

Much later, Spielman (1924) postulated that the organic compounds in meteorites could be formed after the falling of the meteorites to earth as a result of the effect of ground water on the carbides believed present in carbonaceous chondrites. This hypothesis found no support for the following reasons. For formation of approximately 5% carbonaceous matter in meteorite it is necessary that the carbide content be almost 25% of the meteorite weight. The decomposition of such a quantity of carbides would result in destruction of the sample. Moreover, carbonaceous chondrites do not contain any carbide at all, although many of the chondrites were investigated immediately after their falling.

V. I. Vernadskiy devoted much attention to study of organic compounds in meteorites. A. P. Vinogradov (1948, p. 8) writes: "Enlisting the support of his fellow workers, Vladimir Ivanovich formulated a broad plan for the study of the meteorite content of gases, organic substances, and water. However, the work initiated by him in this direction had to be suspended during the time of the [Great Patriotic] war".

For all practical purposes, there was a long interruption which lasted until 1950 in the history of the study of organic compounds in meteorites which have begun in the 1890's. This can be attributed to the lack of fully developed methods for studying microquantities of organic compounds. /22
Individual studies only indirectly touched on this problem. For example, in 1942 Buddhue (1942), in checking old information concerning the presence of nitrogen in meteorites, found that chondrites contain nitrogen in the ammonium form in a quantity 6-33 $\times 10^{-4}$%, among the eight meteorites which he investigated one ordinary chondrite (La Lande) contained organic matter.

A. B. Trofimov (1950) determined the isotopic composition of carbon in a number of meteorites, including four carbonaceous meteorites for which he

obtained the ratio C^{12}/C^{13} = 89.9-90.6[1]. Due to the insufficiently high accuracy in the determination (±0.3%), this ratio differed little from the C^{12}/C^{13} ratio for other chondrites. The isotopic composition of carbon, and also the composition of hydrogen for a number of carbonaceous chondrites was determined by Boato (1954a, b); like Trofimov, he determined the isotopic composition of total carbon. On the basis of isotopic composition of carbon, Boato defined two groups of carbonaceous chondrites for which δC^{13}(‰)was: for the first group, from -3.7 to -11.4, for the second group from -15.7 to -18.8. Boato felt that the organic compound in carbonaceous chondrites are natural in these meteorites. Wiik (1956) made a chemical analysis of many carbonaceous chondrites and classified them into three types. He found that they contained a high content of C, S and H_2O. Losses in calcination, which sometimes exceeded 5%, were attributed by Wiik primarily to losses of organic compounds.

The real study of organic compounds in meteorites began in 1953, when Mueller (1953) published the results of his studies of the bituminous matter in the Cold Bokkeveld carbonaceous chondrites. He found that this meteorite contains 2.20% C, 1.53% H, 1.99% S. Mueller used a mixture of HCl, HF and water in obtaining the carbonaceous matter from 12 gm of a pulverized meteorite sample. The composition took place during heating in a water bath. Graphite was not found in the residue. Mueller processed 20 gm of a meteorite in water during heating in a closed flask. The water extract had a specific aromatic odor. With 12.5 gm of pulverized meteorite he extracted a bituminous substance by using organic solvents in a Soxhlet apparatus. The content of the extract was: In the case of extraction with methanol -- -- 0.508%, with ethanol -- 0.204%, with chloroform -- 0.160%, with benzene -- -- 0.242%, with carbon bisulfide -- 0.002%. In all, 1.116% bituminous matter was extracted using organic solvents at their boiling point. Using a different weight sample (10 gm), extraction with organic solvents was done in a different sequence; bituminous components were extracted (in %) with the following -- with benzene 0.655, with chloroform 0.017, with ethanol 0.424, with methanol 0.029; the organic compounds extracted constituted 1.125% of the total matter.

The extracts did not dissolve in water, but they dissolve well in organic solvents. They all had an aromatic odor, their consistancy was viscous, and the chloroform extracts were semifluid. The benzene extracts were dark brown in color and in ultraviolet light they had a brownish luminescence. The color of the other extracts was light brown and they had a greenish yellow luminescence. Acids exerted no effect on the extracts but in alkali these organic components yielded brownish solutions.

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1 According to A. B. Trosimov the C^{12}/C^{13} for meteorites containing carbon is: for the Migei carbonaceous chondrite 90.9, for the Staroye Boriskino 89.9, Cold Bokkeveld 90.6, and Grosnaya 90.3, for the Kainsas chondrite containing carbon it is 90.6, and for the Novyy Urey ureilite it is 90.9.

The extracts were combined and reprecipitated with alcohol. Mueller separated out tiny crystals of sulfur (10-12% of the entire extract) from this combined sample. An elementary microanalysis of the organic matter (scaled to the ash-free part) revealed the following composition (in %): C 24.26, H 8.12, N 4.00, S 8.78, Cl 5.89, O 48.95; total 100.0. Such a high chlorine content as was found in the organic matter of this meteorite is not found in terrestrial bituminous compounds. Making note of this, Mueller proposed that the bituminous matter in the meteorite be called chlorobitumen and that the nonextracted part of the carbonaceous matter be called retigen, in contrast to the kerogen of terrestrial rocks. After comparing the meteorite extracts with terrestrial organic compounds, Mueller concluded that they are very close to dopplerite. /23

When heating the bituminous matter of the Cold Bokkeveld meteorite to 350 °under atmospheric conditions Mueller noted a gradual decomposition; the residue contained a black carbonaceous substance. In a 5% benzene solution the bituminous matter did not reveal optical rotation. Mueller felt that this substance was formed in the meteorite as a result of polymerization of simple initial compounds at temperatures of 350-600 °. The macromolecules of organic compounds forming chemically during the course of development of the meteorite parent bodies were precipitated, Mueller speculates, armed with the surfaces of condensing cosmic bodies. For these cosmic bodies the ratio of carbon to the sum of volatile elements (H, O, N, S) can serve as a natural thermometer. Mueller feels that a close value of the ratio is observed for the organic matter in rock formed at high temperatures (for example, in Derbyshire pegmatites containing bituminous compounds).

In 1957 a study of the organic compounds in meteorites was begun by the author of this book, but until 1962 this study was of a sporatic character and was carried on simultaneously with other studies of terrestrial organic compounds. Since 1962 the author has been making a systematic study of the organic matter in meteorites. A detailed study of the organic compound in meteorites was begun by foreign researchers in 1962-1963.

In 1957-1958 a preliminary study was made of the bituminous compounds in four carbonaceous chondrites, Migei, Staroye Boriskino, Cold Bokkeveld and Groznay, by the luminescent bituminological method (Ddovykin, 1960). These compounds were extracted without heating and using the following solvents: petroleum ether, chloroform, alcohol-benzene (1:1) and a 2% solution of KOH. The total content of extracts separated out was (in % by weight): Migei 0.064, Staroye Boriskino 0.071, Cold Bokkeveld 0.045, Groznay 0.016. However, later the author became convinced that these figures were too low due to inadequately complete extraction. It was shown (Veodykin 1960) that the bituminous substance in meteorites is not an individual compound, but a complex mixture of organic components which include reduced compounds of a hydrocarbon nature, "resins", and also humuslike substances, although an insignificant quantity. The ratio of the individual components in the organic matter extracted under the same conditions differed somewhat in the different investigated carbonaceous chondrites.

Both the extracts themselves, as well as their capillary extracts were luminescent in ultraviolet light (λ = 365 mμ). However, significant luminescence could not be observed in the meteorite samples themselves prior to the removal of the organic compounds from them. The author postulated that the bituminous compounds in carbonaceous chondrites are in a finally dispersed state and therefore they cannot be seen under an ordinary ultraviolet light. This was actually confirmed in an examination of polished sections of carbonaceous chondrites under an ultraviolet microscope. It was found that these compounds are in the form of very tiny inclusions from 1 to 16 μ in diameter. They exhibited an appreciable luminescence in ultraviolet /24 light of a yellowish-greenish color (Vdovykin, 1962b, 1963b).

K. I. Sztrókay, et al (1958), who studied the chemical and mineral composition of the Kaba carbonaceous chondrite, made computations which revealed that the meteorite contains 4.03% organic compounds the C_{org} content in this meteorite, according to their computations, was 1.99%. In studying the hydrocarbon part of the organic matter they used a fraction of the meteorite which had been processed with HCl (Sztrókay, et al, 1961). The infrared absorption spectrum of this fraction (Figure 7) revealed the presence of aliphatic hydrocarbons: The spectrum had two absorption bands with maxima in the regions 2939 and 1455 cm^{-1}, and also at 900 and 1100 cm^{-1}. The maximum absorption at 1200 cm^{-1} was attributed to the presence of sulfur.

Calvin (Calvin, Vaughn, 1960; Calvin, 1961) examined the possible direction in chemical evolution of organic compounds and reported the results of their studies of the organic matter in the Murray carbonaceous chondrite. He found that this meteorite contains the following quantities of volatile elements (% by weight): C 2.07, H 0.90, N 0.08, S 0.375, P 0.02, and also SO_4 5.72; residue 87.0. An analysis of the gas released during processing of the samples HCl revealed the presence of CO_2 and H_2S with a molecular ratio 2:1. Subsequent extraction of 9 gm of the meteorites with solvents made it possible to extract from it 1.21 % by weight of organic compounds, consisting of (in %): with extraction by CCl_4 - 0.22%; $CHCl_3$ - 0.04; benzene 0.04; water 0.81; ethanol 0.10.

Calvin studied the extracts obtained using CCl_4 and water in greater detail. In the infrared absorption spectrum of the CCl_4 extract (Figure 8) Calvin found C-H bonds: methyl (2960 cm^{-1}) and methylene (2930 cm^{-1}) groups. The presence of C-H bonds was confirmed by absorption bands with maxima at 1380 and 1460 cm^{-1}. The spectrum also revealed a carbonyl bond - C=O (~1700 cm^{-1}) and aromatic hydrocarbons were noted (800 cm^{-1}). Calvin assumed that the hydrocarbons in this meteorite contained at least 12 or more carbon atoms in the chain.

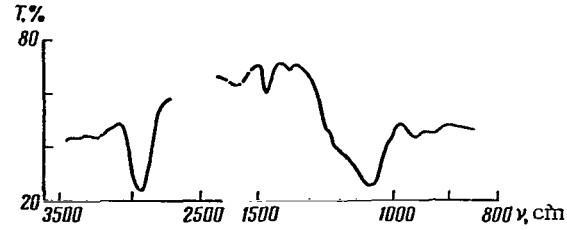
The ultraviolet spectra of the extracts which Calvin obtained using CCl_4, $CHCl_3$, C_6H_6 and CH_3OH revealed absorption in the region 2500-3000 Å

caused, in his opinion, by elementary sulfur. True maxima (at 2750 and 2800 Å) were noted on the spectrum of extracts soluble in water eluviated with HCl in an ion-exchange column. The behavior of these absorption maxima was dependent on change in pH. Calvin postulated that they were caused, in all probability, by the presence of heterocyclic(bases very similar to cytosine However, Oró (1963c) later investigated control eluates from ion-exchange chromatographic columns, when only 0.1 n. HCl and 4 n. ascetic acids or eluated, and demonstrated that these eluates give similar absorption bands on the ultraviolet spectra. On the basis of the identity of the absorption maxima and the similar character of their change for different pH Oró postulated that the organic compounds in a water extract of the Murray meteorite, studied by Calvin, were for the most part contamination, and that Calvin possibly separated them out from ion-exchange resins during the elution process.

In 1961 Briggs (1961) studied water-soluble organic matter which he separated out from the Mokoia carbonaceous chondrite by using hot water. Briggs extracted this matter in a quantity of about 2% of the meteorite weight; it had a light brown color and the following composition: ~6% C, 1.3% H and traces of N. He investigated this abstract by the ultraviolet method, infrared spectroscopy and paper chromotography. The infrared spectrum of a fraction of this extract, soluble in $CHCl_3$, is shown in Figure 9. /26 In the infrared spectrum Briggs noted the presence of C-H, C=0 groups and others. In the water-soluble extract of this meteorite Briggs postulated the presence of at least four individual organic compounds, one of which was similar to purine or was close to imidazole. On the ultraviolet absorption spectrum of this extract it was possible to note the change of the curves as a function of pH (Figure 10); in neutral solutions there was an absorption increase in the region 260 mμ.

The hydrocarbon components of the organic matter in the Orgueil meteorite were studied by the mass-spectrometer method by Nagy, Meinschein and Hennessy (1961). They extracted organic matter from this meteorite by using a mixture of CCl_4 and trimethylpentane (1:1); on the spectrum in the infrared region for this substance they noted absorption at 3.46 and 10.0 microns. In the mass-spectrometer analysis, when heating was at 150-250 and 250-400°, the authors identified in the Orgueil meteorite the presence of saturated hydrocarbons containing from 15 to 29 carbon atoms in their chain (Table 2). The distribution of hydrocarbons in the meteorite, according to the data of these authors, resembled the distribution of hydrocarbons in biogenous products and recent sediments. On this basis they concluded that the organic compounds in the Orgueil meteorite have a biogenous nature (Nagy, Meinschein, Hennessy, 1962). In discussing this conclusion, Anders (1962) made a more detailed study of the mass spectrograms of these organic compounds from different sources and established a rather distinct difference between them, particularly the fraction C_nH_{2n+2} (Figure 11a). The least difference was noted for the fraction C_nH_{2n-6} (Figure 11b).

Fig. 7 Infrared Absoption Spectrum of Fraction Obtained from the Kaba Meteorite after Processing Pulverized Meteorite Matter with HCl (Sztrokay, et al, 1961).
T -- transmission, ν -- wave number



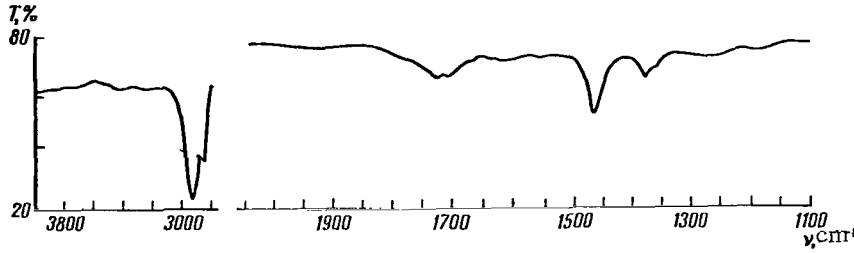
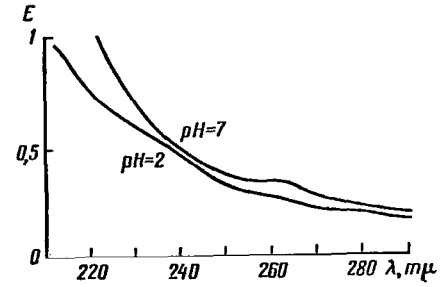


Fig. 8 Infrared Absorption Spectrum of Bituminous Matter Obtained from the Murray Meteorite using CCl_4 (Calvin, 1961).

Fig. 9 Infrared Absorption Spectrum of Part of Extract Soluble in Chloroform; extract obtained from the Mokoia meteorite using hot water (Briggs, 1961).

Fig. 10 Ultraviolet Spectrum of Extract obtained from Mokoia Meteorite using Hot Water (Briggs, 1961).
E -- absorption, λ -- wave length



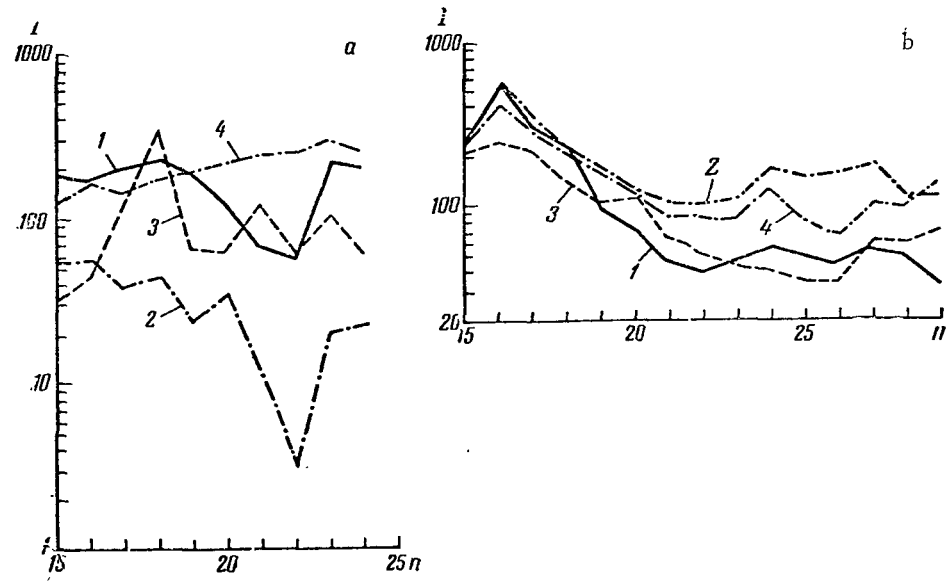


Fig. 11 Distribution of Limiting Hydrocarbons Separated out from Orgueil Meteorite and Hydrocarbons of Terrestrial Biogenous Objects.
I - lower intensity of peaks; n - number of carbon atoms; a - hydrocarbons of type C_nH_{2n+2}; b - hydrocarbons of the C_nH_{2n-6}; 1 - bituminous matter of Orgueil meteorite;

2 - hydrocarbon fraction of the same meteorite; 3 - oil; 4 - Organic matter in recent sediments.
The mass spectra were obteined by B. Nagy, et al (1961); diagrams are reproduced from Anders (1963c).

Thus, recent research (using infrared spectroscopy, mass spectrometry, and other methods) not only have confirmed the report of presence of organic compounds in meteorites published during the last century, but have also yielded some information on their composition. /27

In the course of discussions of the collected data, researchers repeatedly have commented on their importance, especially for clarifying problems involved in the origin of life (Bernal, 1961a; Anders, 1962). Interest in carbonaceous chondrites containing organic matter has increased greatly. The danger has arisen of a rapid expenditure of these meteorites in research (Nagy, 1961). For this reason Bernal (1961a, b) has proposed the creation of a commission for controlling the expenditure of carbonaceous chondrites and studying these meteorites. A group was recently established in the United States, headed by Dr. Libby concerned with the analysis of organic compounds in carbonaceous chondrites (Group Organized to Analyse..., 1964).

Table 2

Relative Distribution of Hydrocarbons in Orgueil Meteorites.
Based on Data from a Mass Spectrometer Analysis
(Nagy, Meinschein, Hennessy, 1961)

Number of carbon atoms	Relative Intensity of Peaks			
	n-paraffins	Monocycloalkanes	Bicycloalkanes	Tetracycloalkanes
15	180	667	267	231
16	170	563	215	578
17	198	498	225	302
18	221	445	288	230
19	176	356	149	99
20	113	281	137	70
21	65	185	103	46
22	55	155	102	39
23	205	135	64	45
24	186	100	61	56
25	-	-	-	50
26	-	-	-	44
27	-	-	-	53
28	-	-	-	46
29	-	-	-	29

CHAPTER TWO

MINERAL COMPOSITION AND STRUCTURE OF CARBONACEOUS CHONDRITES

Characteristics of Structure of Carbonaceous Chondrites

Carbonaceous chondrites are few in number, but in many respects they constitute an interesting type of meteorites. They are distinguished from other meteorites in having a high content of carbonaceous matter, by the presence of minerals with a low-temperature origin, and a high degree of iron oxidation, and therefore there is a low content or complete absence of ferronickel and other peculiarities. Some researchers are inclined to regard these meteorites as primary matter from which other types of stony meteorite, as well as the planets of the solar system, were formed. /28

The mineralogical composition and structure of carbonaceous chondrites after the presence of water in them was established in a form bonded in silicates (Kvasha, 1948; A. N. Zabaritskiy, 1948a, Zabaritskiy, Kvasha, 1952), followed by a detailed study of organic compounds, have recently been studied by many authors (Wiik, 1956; Mason, Wiik, 1962a, b; Mason, 1963d; Anders, 1963c; Kvasha, 1963; Kvasha, Wiik, 1964; Vdovykin, 1964 c, 1965a, and others). Information on the composition of carbonaceous chondrite is given in studies written in the last century as well; a bibliography of these studies has been given in review (Wülfing, 1897; Brown, 1953), and the most complete of these was in a book by Cohen (1894).

Table 3

Quantitative Relationship between Carbonaceous Chondrite and Other Meteorites

	Number of falls and finds (Vinogradov, 1965)	Carbonaceous Chondrites, %	Number of falls and finds (Vinogradov, 1965)	Carbonaceous Chondrites, %
All Meteorites	1759	1.70	764	3.93
Stony Meteorites	1082	2.77	704	4.26
Chondrite	919	3.26	596	5.03

Reviews of recent studies of the organic matter in carbonaceous chondrites have been published by Briggs and Mamikunian (1963), Vdovykin (1964a, 1966b), Nagy (1966b), Urey (1965b, 1966a), Hayes (1967).

In our book we will give only concise information concerning carbonaceous chondrites and a brief description of those carbonaceous chondrites in which the author studied organic matter.

About 30 carbonaceous chondrites are now known. This represents 3.26% of the total number of falls and finds of chondrites or 5.03% of all falls of chondrites (Table 3).

The falls of all these meteorites were observed. The first carbonaceous chondrite (Alais in France) fell in 1806. During recent years the following carbonaceous chondrites have fallen: Essebi in Congo (Corin, 1960) and /29
Al Rais in Arabia in 1957, Bells in the United States in 1961 (Monnig, 1963) and Revelstoke in Canada in 1965 (Meteoritnyy Byullètin'..., 1965; Shoemaker, Lowery, 1966). In addition, in 1963 the Pollen meteorite, which had fallen in Norway in 1942, was recognized as a carbonaceous chondrite (Wolff, 1963a, b). When carbonaceous chondrites fall, a bolide is almost always observed, and often it is rather bright (Prior, Hey, 1953; Kvasha,1963). The total weight of carbonaceous chondrites which are now found in museums is about 132 kg. The largest carbonaceous chondrite (Lancé) weighs 52 kg; the smallest (Revelstoke) weighs about 1 g.

Like other (ordinary) chondrites, carbonaceous chondrites are covered by a thin black and dull fused crust less than 1 mm thick[1]. These meteorites are black; usually they are very brittle and are rather easily pulverized with the fingers. According to Mason (1963c, d), their specific gravity is lower (2.2 - 3.6) than for ordinary chondrites (3.7 - 3.8). The structure of all these meteorites is chondritic (crumblike) and the chondrules account for from 10-20 to 90 percent of the volume or more (Kvasha, 1963). The diameter of a chondrules is usually 0.1-0.5 mm. However, some carbonaceous chondrites which are considerably enriched with carbon (for example, the Orgueil meteorite) do not contain chondrules at all. In individual cases (in the Migei and Staroye Boriskino carbonaceous chondrites) L. G. Kvasha (1963) observed a clastic structure and inclusion. Mueller (1966) recently reported the presence of petrographically nonhomogeneous sectors in a number of carbonaceous chondrites.

On the basis of a number of peculiarities in chemical composition, carbonaceous chondrites can be divided into three classes; these were defined by Wiik (1956) on the basis of some old chemical analysis which he reviewed and his own analysis of 13 carbonaceous chondrites. Wiik used the term "types"

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[1] In contrast to the external part of meteorites, covered by a fused crust the inter part of meteorites is not heated during their flight through the atmosphere. Houtermans (1958) used the thermoluminescent analysis method to demonstrate that during the flights of the Holbrook meteorite through the atmosphere heating was propagated beneath its fused crust only to a depth of 10-15 mm.

for these classes. At the same time A. A. Yavnel' (1956) defined three classes of carbonaceous chondrites on the basis of the quantitative content of different forms of iron in these meteorites.

Table 4 lists all the presently known carbonaceous chondrites of the three types, as well as general information concerning them. General information concerning almost all the meteorites listed in this table, as well as their specific gravity, is given in a study by Mason (1963d). The total content of volatiles was determined by Mueller, et al., (1966; 1965) by a thermal balance analysis during the heating of meteorite samples to 1000° in a nitrogen atmosphere. The values which they computed are given in parenthesis. The sequence of arrangement of the carbonaceous chondrites proposed by Mueller on the basis of the determined sum of volatiles or his computed figure has almost not been changed in the table.

The three types of carbonaceous chondrites (Orgueil, Cold Bokkeveld and Felix types) are interrelated to one another with respect to the peculiarities of their composition. The first type of carbonaceous chondrites corresponds to the ornansites defined by Meunier (1884). Mason (1963a, d) defines the third type into an independent group of olivine - pigeonite chondrite. The St. Caprais chondrite (which fell in France in 1883) and the Chainpur meteorite (which fell in India in 1907) are close to the third type. These meteorites contain a high carbon quantity: 0.64 and 0.36% respectively. However, they differed somewhat in the peculiarities of their mineral composition.

The Karoonda chondrite, which fell in South Australia in 1930, with respect to the peculiarities of its mineral composition, somewhat resembles carbonaceous chondrite but contains little carbon (0.10%).

The Renazzo meteorite contains 1.44% carbon, but Akaiwa (1966), on the basis of the peculiarities of the mineralogical composition of this meteorite, assigns it to a type intermediate between carbonaceous and olivine-bronzite chondrites. Hey (1966), on the basis of the K, Cu and Ti contents in the Renazzo meteorite, included this meteorite in the group of enstatic chondrite. /30

A. N. Zabaritskiy (Zabaritskiy, Kvasha, 1952), on the basis of the mineralogical composition, we find two groups of carbonaceous chondrites: Meteorites containing fluorite and those not containing fluorite. According to Zabaritskiy, the first group corresponds to types I and II, which were later defined by Wiik (1956); the second group corresponds to type-III carbonaceous chondrites, designed by Wiiks. /31

Carbonaceous chondrites differ considerably from other meteorites in their mineral and chemical composition. A very important peculiarity of their mineral composition is the co-existence of two mineral associations, low- and high-temperature associations, whose stability is determined by thermal conditions. "Low-temperature" minerals are represented by carbonates, sulfates, chlorides, free sulfur, etc.: Anders (1963c, 1964, 1965) also includes magnetite in this list. Other minerals that can be included are hydrous

layered-lattice silicates of the chlorite-serpentine type, first found in carbonaceous chondrites in 1946 by L. G. Kvasha in the Staroye Boriskino meteorite (Kvasha, 1948), and later in the Migei meteorite (Kvasha, 1950). "Low-temperature" minerals are characteristic only of carbonaceous chondrite; they are not found in other meteorites. This low-temperature association of minerals corresponds to a hydrothermal association which was described for carbonaceous chondrites by Academician A. N. Zavaritskiy (1956).

"High-temperature" minerals in carbonaceous chondrites are the following. Olivine, pyroxenes, plagioclases, ferronickel, troilite, pentlandite, spinel, and others. The relationship between "low-" and "high-temperature" mineral components in three types of carbonaceous chondrites shown in Figure 12, will be considered further, as an indication of the origin of carbonaceous chondrites. We note only that with a transition from type-III carbonaceous /32 chondrite to type-I carbonaceous chondrites the quantitative content of "low-temperature" components increases.

Carbonaceous chondrites, in addition to having a nonequilibrium between the two groups of minerals, are characterized by a nonequilibrium in the chemical composition between the chondrules and the inter-chondrule matrix. The chondrules in these meteorites are in a more reduced state than the matter filling the inter-chondrule space. In addition, there is a nonequilibrium in the chondrules themselves (Mason 1963d; Anders, 1964; Ledin, 1965; Wood, 1967, and others). In general, all three types of carbonaceous chondrites, as demonstrated by Wiik (1956), in contrast to other meteorites, are characterized by high degree of oxidation of their matter, the absence of ferronickel or a very low content of ferronickel, but a high quantitative content of C, H_2O, S[1]. Whereas the content of sulfur, carbon and hydrogen in carbonaceous chondrites increases in transition from type III carbonaceous chondrites to type I, the content of iron, silicon and magnesium (the basic chemical elements making up the mineral phase), on the other hand, decreases (Table 5). However, if chemical analyses of carbonaceous chondrite are considered without taking into account the volatile elements (C, H, S, N, O), the differences in chemical composition between the three types of carbonaceous chondrites are smoothed out. The data in Table 6 shows that in this case the differences between carbonaceous chondrite and chondrites of other types become less conspicuous. Indeed, this is the high content of volatile elements which is responsible for the peculiarities in the chemical composition of carbonaceous chondrites. The forms of appearance of all these volatile elements in carbonaceous chondrites are widely varied. Some of these elements enter into organic compounds.

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[1] In all his chemical analysis of carbonaceous chondrites, Wiik (1956) gives all sulfur as FeS. However, the forms of appearance of sulfur in carbonaceous chondrites, in contrast to other meteorites, are highly varied. Therefore, for a comparison of the chemical composition of carbonaceous chondrites and other meteorites Wiik proposes use of chemical analysis which have been scaled after excluding the volatile elements.

Table 4

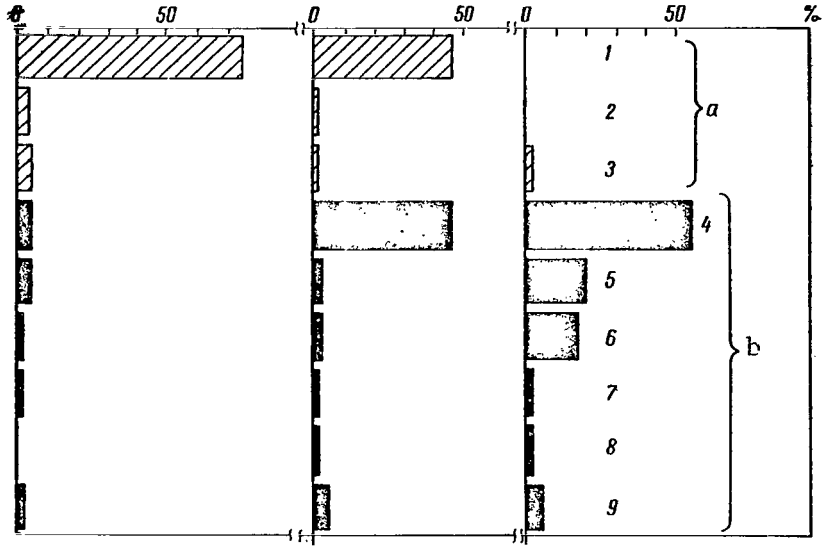
General Information on Carbonaceous Chondrites

Meteorite	Place of Fall	Date of Fall	Weight, kg	Specific Gravity, g/cm^3	Sum of volatiles, %
		Type I (Orgueil type)			
Ivuna	Tanzania	16.XII 1938	0.70	2.23	28.42
Alais	France	15.III 1806	0.26	2.23	28.10
Orgueil	France	14.V 1864	11	2.24	28.02
Tonk	India	22.I 1911	0.01	2.20	(27.15)
Essebi	Congo (Capital Kinshasha)	28.VII 1957	-	-	-
Revelstoke	Canada	31.III 1965	0.001	-	-
		Type II (Cold Bokkeveld type)			
Haripura	India	17.I 1921	0.32	2.72	(21.19)
Hawapali	India	6.VI 1890	0.06	2.57	(20.96)
Nogoya	Argentina	30.VI 1879	2.5	2.66	18.18
Pollen	Norway	6.IV 1942	0.25	2.57	-
Migei	USSR	18.VI 1889	8	2.70	16.23
Staroye Boriskino	USSR	20.IV 1930	1.17	2.74	15.69
Cold Bokkeveld	South Africa	13.X 1838	4	2.65	18.18
Erakot	India	22.VI 1940	0.11	2.66	(15.98)
Santa Cruz	Mexico	3.IX 1939	0.05	2.57	(15.31)
Murray	United States	20.IX 1950	13	2.86	15.20
Bells	United States	9.IX. 1961	0.03	2.84	-
Al Rais	Arabia	10.XII 1957	2.92	2.49	(12.62)
Crescent	United States	17.VIII 1936	0.78	2.82	-
Renazzo	Italy	15.1 1824	1.1	3.29	8.52
		Type III (Felix type)			
Groznaya	USSR	28.VI 1861	3.3	3.49	7.96
Lance	France	23.VII 1872	52	3.64	5.55
Mokoia	New Zealand	26.XI 1908	4	3.41	4.36
Kaba	Hungary	15.IV 1857	3	3.40	(3.92)
Vigarano	Italy	22.I 1910	16	3.42	3.90
Bali	Cameroons	22.XII 1907	0.01	3.40	-
Felix	United States	15.V 1900	3	-	(1.63)
Ornans	France	11.VII 1868	6	3.61	1.34
Warrenton	United States	3.I 1877	1.6	-	0.93

(Table 4 Continued)

Chondrites Containing Carbon

Karoonda	South Australia	25.XI	1930	37.5	3.57	-
St. Caprais	France	28.I	1883	0.14	-	5.54
Chainpur	India	9.I	1907	8	-	(2.69)
Kainsaz	USSR	13.IX	1937	210	3.93	3.42
Effemovka	USSR	Prior to	1962	21	3.53	



Ornans (Type III) Migei (Type II) Orgueil (Type I)

Fig. 12 Diagram of Mineral Composition of Three Types of Carbonaceous Chondrites.
a) "High-temperature" minerals; b) "Low-temperature" minerals; 1) olivine, pyroxenes (and glass); 2) ferro-nickel; 3) sulfides (troilite, pentlandite); 4) hydrous ferromagnesian silicates; 5) iron-oxide (magnetites); 6) sulfites (epsolmite); 7)sulfur; 8) carbonates; 9) carbonaceous matter. Diagram compiled by Anders (1964; 1965). Supplemented by data from L. G. Kbasha (1948, 1963) and by the results obtained by the author with respect to carbonates in Type-II carbonaceous chondrites.

Table 5

Variations in the Content of Some Elements in Three Types of Carbonaceous Chondrites (% by Weight) (See Mason, 1963d)

Types of Carbonaceous Chondrites	Fe	Si	Mg	S	C	H
I Type	17-20	9-11	8-10	5.0-6.7	2.7-4.8	1.8-2.4
II Type	20-24	12-14	10-14	2.6-5.4	1.3-4.0	1.0-1.8
III Type	24-26	14-16	13-15	2.1-2.5	0.4-1.1	0.2-0.5

Recently, Mueller, et al (1965, 1966), by a thermal balance analysis with the heating of carbonaceous chondrites to 1000° in a nitrogen atmosphere determined the sum of volatiles in many carbonaceous chondrites. According to his data, the sum of volatiles also decreases with transition from type-I carbonaceous chondrites (28.42% in the Ivuna meteorite) to type-III (0.93% in the Warrenton meteorite) (see Table 4). On the basis of the "sums of volatiles" Mueller determined the order of carbonaceous chondrites by Wiik types, which has almost not been changed in Table 4[1].

During recent years, with the many-sided study of carbonaceous chondrites, they were found to contain many admixture elements. It was found that some of them, relatively highly volatile, considerably enrich the matter of carbonaceous chondrites in comparison with ordinary chondrites. Carbonaceous chondrites are enriched with the following admixture elements: F, Cl, Cu, Zn, Ga, Ge, Br, In, Te, I, Hg, Tl, Pb, Bi (Table 7). Incidently, under terrestrial conditions /33 these elements are concentrated in the earth's upper layers during the course of degassing and melting out of the matter in the mantel (Vinogradov, 1962b; Tarasov, 1963). In carbonaceous chondrites, particularily in types-I, the occurrence of these elements is close to that in space.

Carbonaceous chondrites like other meteorites contain inert gases. Their distribution in carbonaceous chondrites was recently discussed by Pepin and Signer (1965).

[1] In some carbonaceous chondrites Mueller (1964b) did not directly determine the sum of the volatiles. He computed the content emperically.

Table 6

Chemical Composition of Carbonaceous and Other Chondrites
(In Atom %); Scaled After Excluding Volatile Elements

Meteorites	Fe	Ni	Co	Si	Ti	Al	Mn	Mg	Ca	Na	K	P	Cr
Carbonaceous chondrites:													
I Type													
vuna.....	27.41	1.44	0.07	30.43	0.09	2.56	0.26	32.14	2.71	1.95	0.11	0.47	0.36
Alais.....	27.00	1.58	0.08	29.34	0.08	3.84	0.28	32.95	2.04	2.03	0.08	0.33	0.34
Orgueil...	28.48	1.42	0.07	31.26	0.08	1.27	0.28	32.61	1.76	1.95	0.08	0.34	0.36
II Type													
Haripura...	27.00	1.63	0.08	31.19	0.09	3.16	0.19	31.76	2.54	1.60	0.10	0.23	0.43
Nawapali...	26.76	1.47	0.06	32.07	0.10	2.92	0.17	31.56	2.79	1.24	0.24	0.26	0.36
Migei.....	26.18	1.41	0.06	31.85	0.09	2.90	0.19	33.19	2.04	1.40	0.07	0.29	0.33
Staroye Boriskino.	27.35	1.43	0.07	31.65	0.09	3.10	0.21	31.56	1.97	1.74	0.05	0.31	0.37
Cold Bokkeveld.	26.28	1.41	0.08	32.02	0.09	3.17	0.19	32.67	1.96	1.37	0.07	0.30	0.39
Erakot.....	27.51	1.42	0.07	31.16	0.08	2.64	0.23	32.49	1.97	1.64	0.05	0.31	0.38
Santa Cruz.	26.56	1.41	0.07	31.39	0.12	2.12	0.17	33.69	2.63	1.04	0.17	0.30	0.33
Murray.....	26.13	1.38	0.08	32.70	0.10	2.26	0.19	33.56	2.34	0.49	0.05	0.32	0.40
Al Rais....	26.82	1.32	0.07	31.70	0.06	2.43	0.21	33.10	2.25	1.27	0.04	0.24	0.47
Renazzo....	25.71	1.32	0.10	32.32	0.13	2.66	0.20	33.94	1.85	1.02	0.05	0.23	0.43
III Type													
Groznaya..	26.65	1.27	0.06	31.84	0.09	2.54	0.18	32.83	2.53	1.28	0.04	0.33	0.36
Lance.....	25.97	1.45	0.07	31.36	0.11	3.25	0.16	33.10	2.67	1.07	0.17	0.26	0.36
Mokoia....	24.82	1.29	0.06	32.08	0.09	2.84	0.16	34.32	2.64	0.95	0.05	0.31	0.39
Vigarano..	25.43	1.29	0.07	31.38	0.17	3.72	0.16	33.62	2.64	0.89	0.07	0.20	0.38
Felix.....	26.17	1.31	0.07	31.85	0.09	3.22	0.17	33.18	2.21	1.07	0.06	0.27	0.33
Ornans....	26.13	1.31	0.08	31.38	0.13	2.83	0.18	34.05	1.97	1.01	0.23	0.28	0.42
Enstatic Chondrites	28.52	1.65	0.11	34.98	0.08	1.91	0.02	29.43	1.67	1.13	0.18	-	0.32
Olivine-Bronzite Chondrites	26.32	1.74	0.12	33.17	0.10	2.04	0.25	31.72	2.34	1.37	0.23	0.23	0.37
Olivine-Hypersthene Chondrites	21.85	1.27	0.04	36.78	0.09	2.01	0.26	33.92	1.67	1.30	0.17	0.25	0.39

Carbonaceous chondrites -- data from Wiik (1956) and Mason (1963d).
Mean analysis of other chondrites are given by Mason (1963a, b).

Table 7

Content of Some Admixture Elements in Carbonaceous Chondrites ($n \cdot 10^{-4}$ % by weight)

Types of carbonaceous chondrites	Meteorites	F	Cl	Cu	Zn	Ga	Ge	Br	In	Te	J	Hg	Tl	Pb	Bi
I	Ivuna	—	310 (1)* 280 (2)	260 (3)	566 (3)	12,2 (4)	—	11,0 (2)	—	4,21 (2)	1,125 (2)	—	—	—	—
	Orguei'	405 (5) 206 (6) 190 (1)	290 (1) 210 (2)	247 (1) 198 (3)	469 (3) 228 (1)	14,3 (4)	36,7 (1)	2,0 (2)	0,53 (4) 0,115 (7) 0,082 (2,8)	3,35 (7) 2,30 (2) 1,04 (8) 0,97 (4)	0,365 (2)	8,63 (9) 22; 114 (15)	0,141(9)	2,94 (9)	0,13 (9)
II	Migei	220 (5)	350 (2)	143 (3)	210 (3)	10,4 (4)	—	3,5 (2)	0,29 (4) 0,07 (10) 0,050 (7) 0,049 (2,8)	2,25 (11) 1,91 (7) 1,88 (2,8) 1,2 (4)	0,550 (2) 0,310 (11)	6,82(15)	0,097(9)	1,55 (9)	0,18 (9)
	Cold Bokkeveld	—	—	—	—	—	—	2,2 (12)	—	—	—	—	—	—	—
	Murray	—	—	205 (3)	155 (3)	9,6 (4)	17,3 (10)	—	—	1,59 (11)	0,227 (11)	5,0(13) 1,57(15)	—	4,6 (14)	—
	Renazzo	—	—	—	—	—	4,7 (7)	—	0,023 (7)	—	—	—	—	—	—
III	Lance	170 (1) 66 (6)	350 (1) 125 (2)	267 (3) 87 (1)	162 (3) 57,4 (1)	10,4 (4)	18,4 (1)	2,6 (2)	0,19 (4) 0,031 (7) 0,025 (2,8)	1,17 (7) 1,01 (2,8) 0,15 (4)	0,110 (2)	—	—	—	—
	Mokoia	170 (1)	370 (1)	159 (3) 77 (1)	169 (3) 64,2 (1)	6,8 (4)	8,3 (1)	—	—	—	—	2,0 (13) 0,69 7,3 (15)	—	0,93 (14)	—
	Vigarano	—	—	228 (3)	174 (3)	6,0 (4)	—	—	—	—	—	—	—	—	—
	Felix	—	—	201 (3)	122 (3)	10,6 (4)	—	—	—	—	—	—	—	—	—
	Warrington	160 (1)	360 (1)	193 (3) 103 (1)	158 (3)	10,6 (4)	—	—	—	—	—	—	—	—	—
Containing Carbon	Karoonda	190 (1)	310 (1) 117 (2)	103 (1)	49,9 (1)	—	11,8 (1)	1,5 (2)	—	1,46 (2)	0,220 (2)	1,28(15)	—	—	—
	Chainpur	—	—	—	186 (3)	8,9 (4)	—	—	—	—	—	—	—	—	—
Mean content in ordinary chondrites (From A.P. Vinogradov, 1962c)		28	70	100	10	3	10	0,5	0,001	0,5	0,04	3	0,001	0,2	0,003

*The references are given in parenthesis: 1—Greenland, 1963; Greenland, Lovering, 1965; 2—Reed, Allen, 1966; 3—Greenland, Goles, 1965; 4—Greenland, 1965; 5—Fisher, 1963, 1964; 6—Reed, 1964; 7—Akaiwa, 1966; 8—Smales, Mapper, 1965 (Akaiwa, 1966); 9—Reed et al., 1960; 10—Vinogradov 1965; 11—Goles, Anders, 1962; 12—Filby, Ball, 1965 (Reed, Allen, 1966); 13—Mueller 1966; 14—Marshall, 1962; 15—Elmann, Lovering, 1967.

*Tr. Note: Commas indicate decimal points.

Table 8

Content and Isotopic Composition of Inert Gases in Carbonaceous Chondrites

($n \cdot x\ 10^{-8}\ n \cdot x\ cm^2/gm$)

Types of Carbonaceous Chondrites	Meteorites	He^3	He^4	Ne^{20}	Ne^{21}	Ne^{22}	Ar^{36}	Ar^{38}	Ar^{40}	Kr^{84}	Xe^{132}	Authors
I	Ivuna	—	—	21,3	0,4	2,5	96,5	18,0	600	—	—	Stauffer, 1961
	Orgueil	7,7	24 800	47,3	0,9	5,1	67	12	400	0,90	1,0	Zähringer, 1962
II	Migei	2	3 700	12,3	0,65	2,2	58	10,5	850	0,92	1,0	Zähringer, 1962
	Staroye	<1	2 760	27,5	1,22	3,60	56	10,4	900	—	—	Vinogradov
	Boriskino	<1	11 000	82	<0,05	7,4	56	11	1 100	—	—	Задорожный, 1964
	Cold Bokkeveld	0,5	2 800	10,0	0,08	1,3	43	8	600	0,55	0,60	Zähringer, 1962
	Murray	4	8 900	40,7	1,1	5,1	68	12,2	315	0,70	0,86	Zähringer, 1962
		—	—	54,7	1,0	6,0	137	25,6	500	—	—	Stauffer, 1961
		6,8	14 200	65,2	2,2	9,1	76,5	14,2	650	0,27	0,43	Reynolds, 1960
	Renazzo	17,3	6 800	18,2	2,5	4,6	27,3	5,4	1 570	0,6	1,56	Reynolds, Turner, 1964
III	Groznaya	<1	1 620	2,2	0,82	1,76	24	4,7	920	—	—	Vinogradov, Zadorozhnyy
	Mokoia	—	—	312	3,25	26,2	34,6	6,7	1 490	—	—	Stauffer, 1961
	Felix	78	2 400	16,5	12	14,2	150	32	2 475	—	—	Kirsten et al., 1963
Chondrites containing carbon	Karoonda	74	2 540	12,5	12,9	13,9	4,15	2,58	1 670	—	—	Hintenberger et al., 1964
	Kainsav	30	2 000	9,1	6,3	7,0	170	30,5	2 600	—	—	Vinogradov, Zadorozhnyy

Tr Note: Commas indicate decimal points.

It is well known that meteorites contain three components of inert gases which are nonuniformly distributed in each sample: primary cosmogenic and radiogenic (Vinogradov, Zadorozhnyy, 1964, 1965; Mazor, et al, 1966). The isotopic composition of inert gases in meteorites varies in a wide range, which is dependent on many circumstances, especially on the degree of irradiation of a meteorite in space. In contrast to many meteorites carbonaceous chondrites contain primarily the fractionated component of primary gases, i.e., the light gases in them have a low ratio to the heavy gases in comparison with the relationship applying for the sun. This is similar to the atmospheric distribution of inert gases (Stauffer, 1961; Signer, Suess, 1963; Zähringer, /36
1966). Carbonaceous chondrites are enriched with the following isotopes of inert gases: He^4, Ne^{20}, Ar^{36}, Kr^{84}, Xe^{132} (Table 8). The ratio Ne^{20}/Ar^{36} in them is 0.02-0.07, the ratio Ar^{36}/Ar^{38} is 5.36-5.47 (Signer, 1964).

Table 9

Radiogenic and Radiation Age of Carbonaceous Chondrites

Types of Carbonaceous Chondrites	Meteorite	Age $n \cdot 10^6$ yrs Radiogenic (K-Ar)	Radiation	Authors
I	Ivuna	1400	1,6	Stauffer, 1961
	Orgueil	1300	3	Zähringer, 1962
II	Migei	4300	—	Gehrling, Rik 1955
		2400	2,4	Zähringer, 1962
	Staroye	2700	3,7	Vinogradov, Vadorozhnyy, 1951
	Boriskino	3400	0,6	
	Cold Bokkeveld	1200	0,2	Zähringer, 1962
	Murray	2500	4	Stauffer, 1961
		1600	4	Zähringer, 1962
		—	2—3,5	Fisher, 1965
	Renazzo	<3500	—	Reynolds, Turner, 1964
III	Groznaya	3500	2,5	Vinogradov, Vadorozhnyy, 1961
	Lancé	<3900	5	Stauffer, 1961
	Mokoia	3400	13	
	Felix	4500	56	
		4100	48	Zähringer, 1962
Chondrites containing carbon	Karoonda	2520	—	Hintenberger et al., 1964
	Kainsoz	3900	—	Gehrling, Rik 1954
		2100	15,3	Vinogradov, Vadorozhnyy, 1961

Tr Note: Commas indicate decimal points.

The radiation age of carbonaceous chondrites (the time during which the meteorites were subjected to irradiation by cosmic rays) is given in Table 9. The radiation age was computed by using the mean values of the rates of accumulation of cosmogenic isotopes, which were: for He^3 $1.96 \cdot 10^{-8}$ $cm^3/g \cdot 10^6$ years, for Ne^{21} $0.328 \cdot 10^{-8}$ $cm^3/g \cdot 10^6$ years (Vinogradov, Zadorozhnyy 1965). The radiation age of stony meteorites is from $1 \cdot 10^6$ to $122 \cdot 10^6$ years, for most of the studied carbonaceous chondrites it usually is much less than 10 million years. It is assumed that the low radiation age of stony meteorites is governed for the most part by two factors: 1) the formation of fragments with fresh surfaces arising during collisions and 2) errosion in space by cosmic dust, micrometeores, corpuscular and electromagnetic radiation.

The radiogenic age, i.e. the age of hardening and crystallization of the matter in meteorites was determined by the K-Ar method; for carbonaceous chondrites it varies from $1.2 \cdot 10^9$ to $4.5 \cdot 10^9$ years. A. P. Vinogradov (1965) attributes the low radiogenic age or carbonaceous chondrites with a high carbon content to the fact that argon can be lost more easily in these meteorites than in others. Type III carbonaceous chondrites, being denser, lose less argon; their radiogenic age is close to the radiogenic age of many ordinary chondrites (Zähringer, 1962, 1964; Vinogradov, Zadorozhnyy, 1964, 1965). It was found for ordinary chondrites that for most bronzitic chondrites the radiogenic age is higher than for most hypersthenic chondrites. /37
These differences, caused by losses of inert gases, are attributed by A. P. Vinogradov and I. K. Zadorozhnyy (1964) to blithe heating during the collision of astroidal bodies and long-term, possibly periodic, heating of the outer layers as a result of fuller radiation during the passage of the parent bodies near the sun, and in the case large bodies, there was heating of the inner domes as the result of radioactive decay.

Recently Murthy and Compston (1965) used the Rb-Sr method to determine the age of hardening of matter in the Orgueil, Murray, Mokoia and Lancé carbonaceous chondrites, as well as the age of the chondrules and all the matter in the Peace River olivine hypersthenic chondrite. According to their data, the age is 4.5-4.7 billion years. This agrees with the data which were obtained earlier by Marshall (1962) by the U-Pb method for carbonaceous chondrites: Orgueil (4.6 billion years), Murray (4.7 billion years) and Mokoia (4.6 billion years).

The author of this study has investigated the organic matter in the following carbonaceous chondrites: Orgueil (Type-I carbonaceous chondrites), Migei, Staroye Boriskino, Cold Bokkeveld, Murray (Type-II) and Groznay (Type-III). These meteorites have been described in considerable detail in the literature. Below we give basic information concerning these meteorites and also data on the Kainsaz chondrite, which contains carbon, which the author studied as a comparison with carbonaceous chondrites.

Type-I Carbonaceous Chondrite

Orgueil

The Orgueil meteorite fell in Southern France on 14 May 1864. A bolide was observed at the time of its falling; the meteorite flight was accompanied by the sounds of an explosion. After falling about 20 samples with a total weight of about 12 kg were collected. A large part of the meteorite (more than 9 kg) is now in the Paris Museum of Natural History (Hey, 1966).

This meteorite was described by Daubrée, (1864) and was analyzed by Cloëz (1864b) and Pisani, (1864) several days after its falling. Later the meteorite was analyzed many times. A chemical analysis of the meteorite was made by Wiik (1956), Nagy, et al (Nagy, Meinschein, Hennessy, 1961) and Briggs (1963a). Recently Wiik (Mason, 1963d) made a new analysis of the meteorite, and Nagy (1966b) described its petrographic characteristics. During recent years this meteorite has been used in making most of the foreign investigations of organic compounds in meteorites.

In his study, the author was provided a small piece from a sample of a meteorite which was supplied by Professor G. Mueller in 1964 to the Committee on Meteorites Academy of Sciences USSR.

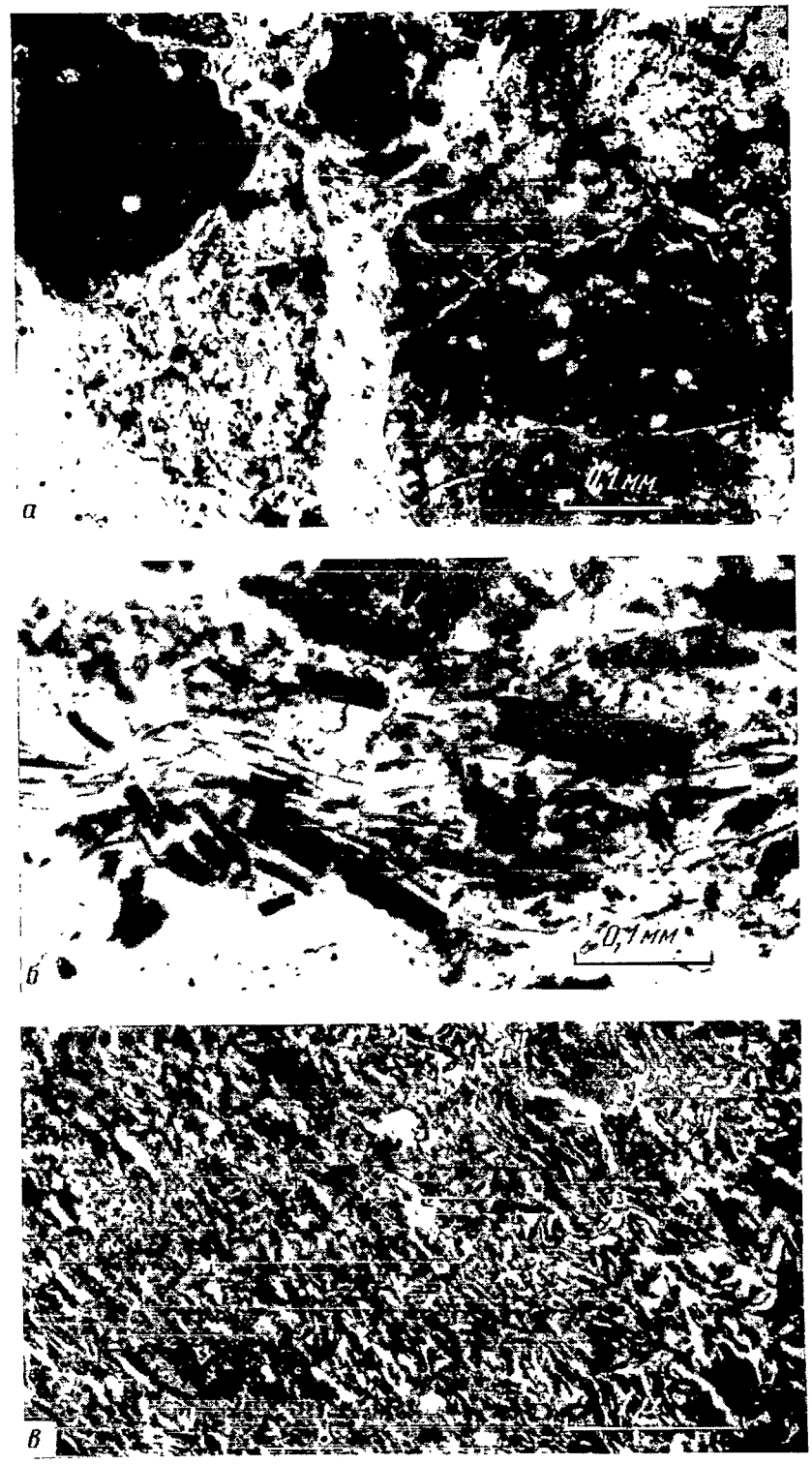
The Orgueil meteorite is a rather homogeneous black mass. Its structure is granular, without any indication of layering. The meteorite is light (specific gravity 2.24, see Table 4), very brittle and soils the hands. It is easily pulverized into angular fragments of irregular configuration. Along tiny cracks one can see a great many intrusions of tiny crystals of a white color up to 0.04 mm in diameter which are readily soluble in water. Silicate grains of an irregular shape, whose diameter rarely attains 0.2 mm, are found amidst the black matrix. This meteorite does not contain visible chondrules, which are characteristic for all chondrites. This has been noted by many authors (Figure 13).

The author of this study investigated the meteorite with an electron microscope (Vdovoykin, 1965a) (Figure 14). The meteorite matrix consists of a mixture of chlorite-serpentine minerals and carbonaceous matter. The /39
chlorite-serpentine minerals are present in the meteorite primarily in the form of platy particles of the irregular form with a diameter of about 0.1 μ. In many cases they form aggregates of particles of an irregular form. Magnetite grains and infrequent hexagonal troilite platelets also are visible. In individual cases droplets of free sulfur were observed forming under the electron beam in the microscope[1].

[1]A similar phenomenon of the formation of sulfur droplets under an electron beam was noted recently by Sciacca and Lipschultz (1964) in a study of the carbonaceous matter in the Canyon Diablo meteorite.

Fig. 13 Orgueil Carbonaceous Chondrite (Nagy, 1966b).

a - Microphotograph of part of a polished section; mineral aggregates are visible; b - microphotograph of part of polished section; a predominant orientation of opaque minerals is visible; c - Electron microscope photograph of an ultrathin section of the meteorite processed with 6 n. HCl.



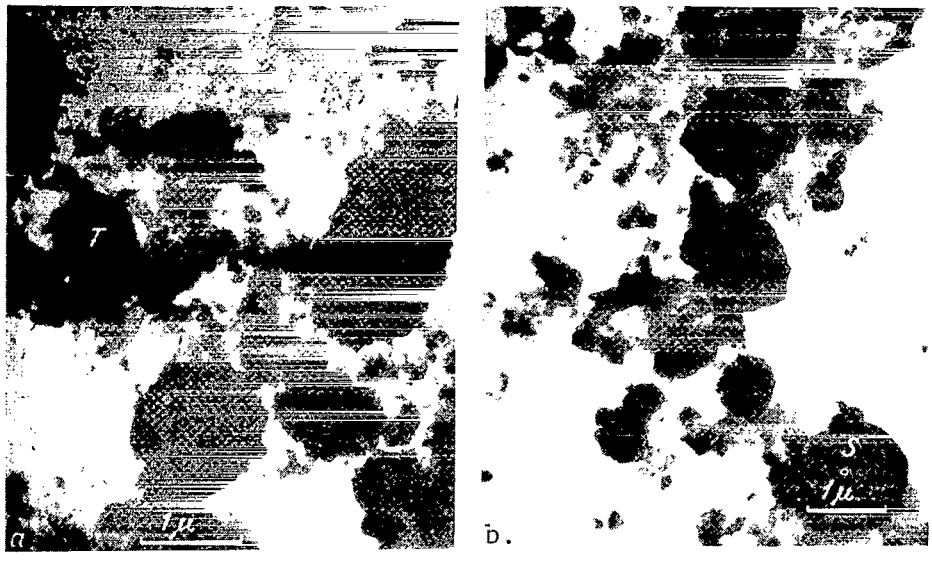


Fig. 14 Electron Microscope Photographs of Dispersed Sample of the Orgueil Meteorite. Translucent platelets of irregular configuration are visible; these are aggregates of chlorite--serpentine minerals and other grains.
a - Hexagonal troilite platelet;
b - Steroidal sulfur; c - Magnetite

N. N. Stulov (1960) has published an x-ray photograph of the Orgueil meteorite and has noted that it contains clorite-serpentine minerals and magnetite. However, epsomite also can be clearly noted from this x-ray photograph (Table 10). Water-soluble magnesium sulfate salts of a white color occur very widely in the meteorite: up to 15% by volume (DuFresne, Anders, 1962b). /41

Accordingly, the meteorite consists for the most part of chlorite-serpentine minerals, magnetite, epsomite, and carbonaceous matter; free sulfur and troilite are also present. This composition is confirmed by many earlier studies of this meteorite.

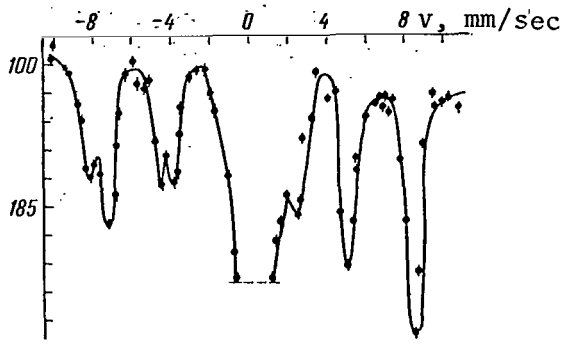


Fig. 15 Gamma-resonance (Mössbauer) Spectrum of Magnetite Fe_3O_4 in the Orgueil Meteorite (Gérard, Delmelle, 1964).

The presence of magnetite in the Orgueil meteorite was pointed out by Cloëz as early as 1864 and recently by Mason (1963d), DuFresne and Anders (1962b), and Nagy, et al (Nagy, Meinschein, Hennessy, 1963). The magnetite in this meteorite was confirmed by Mössbauer (gamma-resonance) spectroscopy (Gérard, Delmelle, 1964) (Figure 15). Jedwab (1965) recently described in the Orgueil meteorite a magnetite with a raspberry structure, consisting of individual rounded grains (0.3-3 μ in diameter). An electron microscope /42
study of the magnetite in this meteorite was made by Orcel and Alpern (1966).

Chlorite-serpentine minerals have been observed repeatedly in the Orgueil meteorite. Nagy, et al (Nay, Meinschein, Hennessy, 1961) feel that these minerals are similar to chlorite; these authors cite the following parameters of its unit cell, computed on the basis of data from a roentgenometric analysis: a = 7.6 Å, b = 9.26 Å. Later these authors studied the Orgueil meteorite using an electron microscope and observed in it a great many platy particles of hydrous silicates with particles having mean diameters of 0.1-0.2 μ (Nagy, Meinschein, Hennessy, 1963). They postulated that this is chlorite or possibly montmorillonite. The authors note that chlorite is confirmed by a thermogravimetric analysis. Platy particles of hydrous silicates were also observed in the Orgueil mete rite by Kerridge (1962-1964). He defines them as minerals similar to sepiolite; in individual cases Kerridge determined Clinochrysolite. According to his data, the parameters of the cell for these silicates are: a = 7.4 Å, b = 9.2 Å.

Breunnerite, up to 0.3% (Mason, 1963d), was noted in the Orgueil meteorite; it was also observed by Pisani (1864). Nagy and Andersen (1964) used the x-ray spectromicro analysis method to show that in this meteorite the

Table 10*

Mineral Composition of Orgueil Carbonaceous Chondrite

Orgueil Meteorite (Stulov, 1960)						Interpretation					
Light Fraction			Dark Fraction			Pennine (Mikheyed, Kalinin, 1958)		Magnetite (Stulov, 1960)		Epsomite (Mikheyed, 1957)	
I	$\frac{d_\alpha}{n}$	$\frac{d_\beta}{n}$	I	$\frac{d_\alpha}{n}$	$\frac{d_\beta}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$
—	—	—	1	4,89	4,43	10	4,80	—	—	3	5,3
—	—	—	2	4,53	4,10	4	4,60	—	—	1	4,50
1	4,26	3,86	1	4,28	3,88	—	—	1	4,21	10	4,22
2	3,91	3,55	—	—	—	5	3,96	—	—	2	3,76
1	3,52	3,19	—	—	—	10	3,58	—	—	2	3,41
1	3,29	2,981	—	—	—	1	3,31	3	(3,31)	1	3,17
2	3,06	2,771	1	3,06	2,774	3	3,15	—	—	—	—
7	2,973	2,694	7	2,968	2,690	—	—	6	2,99	3	2,96
1	2,880	2,611	1	2,876	2,606	9	2,864	—	—	3	2,87
7	2,785	2,524	5	2,785	2,524	3	2,809	5	(2,807)	2	2,74
1	2,637	2,390	—	—	—	4	2,705	—	—	7	2,66
10	2,532	2,295	10	2,532	2,295	10	2,538	10	2,541	—	—
4	2,471	2,240	1	2,451	2,224	10	2,445	3	2,428	1	2,48
1	(2,314)	2,098	1	(2,310)	2,094	—	—	3	(2,310)	2	2,38
4	2,268	2,056	—	—	—	7	2,286	—	—	1	2,27
1	2,163	1,961	—	—	—	—	—	—	—	2	2,21
7	2,099	1,902	8	2,094	1,898	—	—	7	2,098	2	2,10
1	1,938	1,757	—	—	—	10	2,008	—	—	1	1,96
1	1,880	1,704	1	1,880	1,712	6	1,893	2	1,884	2	1,88
7	1,751	1,587	3	1,780	1,614	6	1,741	4	1,785	2	1,80
7	1,711	1,551	4	1,711	1,551	7	1,694	5	1,710	1	1,72
1	1,687	1,529	—	—	—	4	1,670	—	—	2	1,64
1	(1,635)	1,482	1	(1,635)	1,482	—	—	2	(1,632)	—	—
8	1,613	1,462	8	1,614	1,463	—	—	9	1,612	—	—
5	1,534	1,390	7	1,532	1,388	10	1,535	—	—	—	—
9	1,480	1,342	9	1,482	1,343	4	1,462	9	1,479	—	—
1	1,442	1,307	—	—	—	4	1,432	—	—	—	—
1	1,417	1,284	—	—	—	—	—	2	1,411	—	—
1	1,394	1,263	—	—	—	—	—	—	—	—	—
3	1,351	1,225	—	—	—	—	—	—	—	—	—
3	1,327	1,203	3	1,328	1,203	7	1,320	3	1,325	—	—
1	1,315	1,192	—	—	—	—	—	—	—	—	—
6	1,278	1,159	6	1,279	1,159	7	1,295	5	1,277	—	—
1	1,264	1,146	2	1,265	1,146	—	—	2	1,264	—	—
3	1,210	1,096	2	1,211	1,098	—	—	3	(1,209)	—	—
6	1,119	1,014	5	1,121	1,016	—	—	4	1,119	—	—
9	1,091	0,989	9	1,092	0,989	—	—	8	1,091	—	—
7	1,047	0,949	6	1,049	0,950	—	—	6	1,047	—	—

*Tr Note: Commas indicate decimal points.

Table 11

Chemical Composition of Orgueil Carbonaceous Chondrites

Components	A				B			C	
	1	2	3	4	Components	4	5	Components	4
SiO_2	26,08	24,48	24,16	22,56	Fe	18,42	19,47	Mg	32,61
MgO	17,00	8,16	16,25	15,81	Si	10,53	10,75	Si	31,26
FeO	22,93	31,24	12,05	11,39	Mg	9,53	9,71	Fe	28,48
Al_2O_3	0,90	1,18	1,31	1,65	S	5,49	5,01	Al	1,27
CaO	1,85	2,18	1,58	1,22	C	3,10	3,33	Ca	1,76
Na_2O	2,26	1,24	0,98	0,74	H	2,23	1,80	Ni	1,42
K_2O	0,19	0,31	0,11	0,07	Al	0,87	0,42	Na	1,95
P_2O_5	—	Сл.	0,30	0,28	Ni	0,97	1,02	Cr	0,36
Cr_2O_3	0,33	0,23	0,34	0,36	Ca	0,87	0,86	P	0,34
MnO	0,36	1,82	0,21	0,19	Na	0,55	0,55	Mn	0,28
TiO_2	—	—	0,05	0,07	Cr	0,24	0,23	K	0.08
Fe	0,00	—	0,00	0,00	Mn	0,15	0,185	Ti	0,08
Ni	0,00	—	0,00	0,00	P	0,12	0,13	Co	0,07
Co	0,00	—	0,00	0,00	Co	0,05	0,052		
NiO	—	0,45	1,31	1,23	Ti	0,04	0,045		
CoO	—	2,09	0,08	0,06	K	0,058	0,038		
FeS*	13,43	—	14,85	15,07	Cu	—	0,033		
H_2O	—	13,31	19,25	19,89	V	—	0,009		
C	4,00	—	4,10	3,10	N	—	0,24		
Others	—	—	—	6,96	(O)	46,78	46,12		
Total	89,33	86,69	96,93	100,65		100,00	100,00		99,96

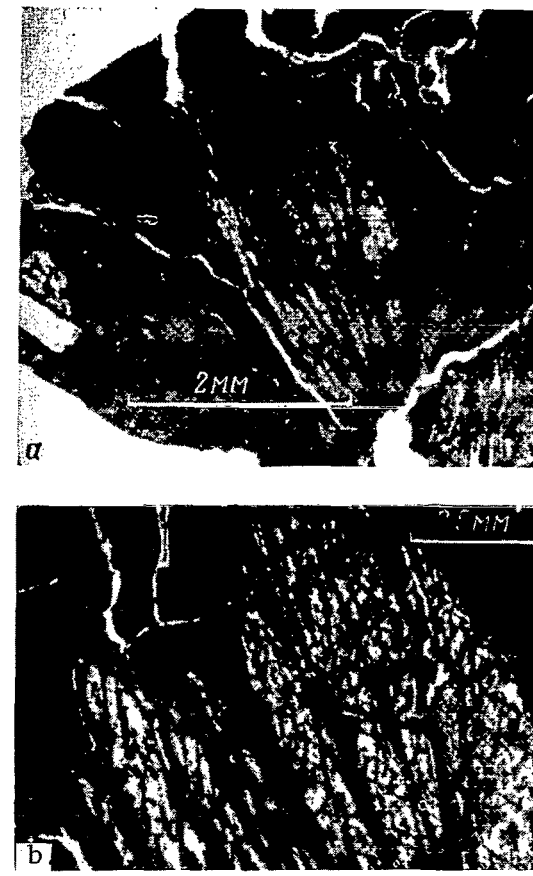
A and B -- % by weight, C -- atom. % (volatile elements excluded); 1 - Cloëz, 1864b; 2 - Nagy, Meinschein, Hennessy, 1961; 3 - Briggs, 1963a; 4 - Wiik, 1956 (sample from the Chicago Museum); 5 - Wiik, 1963 (sample from the New York Museum, Mason, 1963d).

*See footnote 1 on page 34.

breunnerite $(Mg_{0.8}Fe_{0.2})\ CO_3$ (Figure 16) contains: 12-20% Mg and ~12% Fe; in addition, they found Mn (2%), Ca and Si (0.4% each). These investigators found in the meteorite gypsum containing 19% Ca, 15.5% S, 3.1% Fe, and a phosphatic mineral similar to merrillite. The composition of the latter mineral (in %) is: Ca 27.4, P 14.0, Fe 2.3, Si 4.0; the Ca/P ratio = 1.96. The phosphatic mineral obviously incorporates that 0.079% phosphorous was found in this meteorite by a chemical analysis (Nagy, Meinschein, Hennessy, 1963).

Fig. 16 Carbonate (breunnerite) Crystals in Orgueil Meteorite (Nagy, 1966b).

a - general appearance, b - sector of crystals under large magnification.



The Orgueil meteorite was found to contain sulfide (troilite) and free sulfur (DuFresne, Anders, 1963b); it possibly contains traces of olivine (Kerridge, 1964) ferronickle is lacking.

Working independently during the last century, Pisani (1864) and Cloëz (1864b) noted that this meteorite contains water-soluble ammonium salts NH_4Cl. However, in 1956 Wiik (1956) no longer found them. Urey (1966a) /43 postulates that these salts were hydrolyzed; the HCl forming in this process reacted with other mineral components. This actually can be confirmed by some data. Nagy, et al (Nagy, Fredriksson, Urey, et al, 1963) used the x-ray spectro microanalysis method to demonstrate that the Orgueil meteorite contains rounded particles which in their composition correspond to limonite, but containing up to 3% chlorine.

Table 11 gives the chemical composition of the Orgueil meteorite. The most representative analysis are those of Wiik (1956), Mason (1963d) and

Briggs (1963a). According to the chemical analysis made by Wiik, two samples of the Orgueil meteorite differ somewhat from one another. This can probably be attributed to the degree of averaging of the initial analyzed weighted samples. The meteorite is characterized by a high content of volatile elements. The carbon content in it, according to data published by various authors, vary somewhat and are 2.8% (Boato, 1954a), 3.10% (Wiik, 1956), 3.33% (Mason, 1963d), 3.52% (Mueller, 1966), 4% (Cloëz, 1864a, b), and 4.10% (Briggs, 1963a). This is due at least in part to some nonuniformity in the carbon distribution in the samples. Thus, the same author, Wiik (Mason, 1963d) sights a carbon content for a sample of the Orgueil meteorite from the Chicago Museum of 3.10%, whereas for the samplein the New York Museum the value given is 3.33%.

Type II - Carbonaceous Chondrite

Migei

The Migei meteorite fell on 21 June 1889in the area of the present-day Odesskaya Oblast (Ukranian SSR). Its falling was preceded by a bolide and sound defects. The total weight of the meteorite is about 8 kg. Most of the meteorite is in the Chicago Museum (2.2 kg) and in the meteorite collection of the Academy of Sciences USSR (about 1.5 kg). Pieces of it are found at Odessa University (0.8 gm) and other places.

The meteorite was analyzed for the first time by Meunier (1889) and Yu. I. Simashko (1890) soon after its falling, and later by P. G. Melikov and L. V. Krshishanovskiy (1896). Later the meteorite was repeatedly studied. In particular, it was investigated by L. G. Kvasha (1950). She established that it contained florite-serpentine minerals, as was later confirmed by the roentgenometric studies of V. I. Mikheyev and A. I. Kalinin (1958) and I. A. Yudin and N. F. Obodnin (1961). The meteorite has been described by A. N. Zavaritskiy and L. G. Kvasha (1952). The Migei meteorite was also studied by DuFresne and Anders (1962b, 1963). A modern chemical analysis of the meteorite has been made by Wiik (1956).

The meteorite has a black color and an earthy fracture. The meteorite is rather friable and light. Its specific gravity is 2.70. It has a chondritic structure. In this meteorite L. G. Kvasha (1963) noted a clastic structure[1]. The black color of the Migei meteorite (Figure 17) is caused by a high content of carbonaceous matter which is present in the form of small lots in the matrix separating the chondrules. Scattered among these lumps are brightly colored chondrules, as well as grains of silicates and troilite of an irregular shape. The mineral grains present within such small lumps are very fine.

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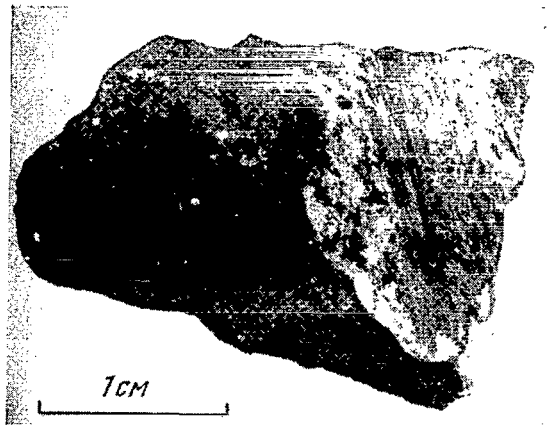
[1]DuFresne (Anders, Lipschutz, 1966a), in a roentgenometric study, discovered asterism for the olivine in this meteorite. The clastic structure and the asterism, apparently are evidence that the Megei meteorite experienced a collision in space.

Fig. 17 General Appearance of a piece of the Migei Carbonaceous Chondrite. The dark coloring of the meteorite was caused by a high content of carbonaceous matter.



The lumpy structure is clearly visible under the microscope (section 1,856). The edges of the lumps are of a dark gray color, with sectors having a greenish hue caused by the presence of light-green florite-serpentine minerals. The lumpy structure of the carbonaceous matter can be seen particularily well in reflected light, in which this matter has a dirty brown color (Figure 18, a). Under a large magnification it is noted that the dark matrix of a meteorite has a flaky structure. A finer distribution of this matter can be seen among the opaque clusters. Here it is somewhat translucent and also of a brownish color. /44

The configuration of such translucent sectors is irregular, but for the most part it is nearly round (usual diameter 0.2 mm). Sometimes microfissures can be seen.

In the carbonaceous matter there is a rather uniform distribution of chondrules 0.08 - 0.3 mm in diameter. They consist for the most part of olivine, but also of chlorite-serpentine minerals, which replace olivine. In the chondrules the olivine grains are separated by opaque carbonaceous matter, frequently observed also in the central parts of the chondrules, and by fluorite-serpentine minerals. The dimensions of the tiny olivine crystals making up the chondrules are usually not greater than 0.05 mm (Figure 18, b). This replacement is frequently developed along the periphery of the chondrules and therefore the chondrules have a disconnected appearance. Disconnected chondrules, made up of florite-serpentine, surrounded by an envelope of carbonaceous matter are observed. Individual elongated grains of chlorite-serpentine of an irregular configuration, duplicating the outlines of the chondrule, are distributed outside, beyond this envelope. Such chondrules apparently represent the inner part of earlier existing large chondrules which have been destroyed and replaced during subsequent processes.

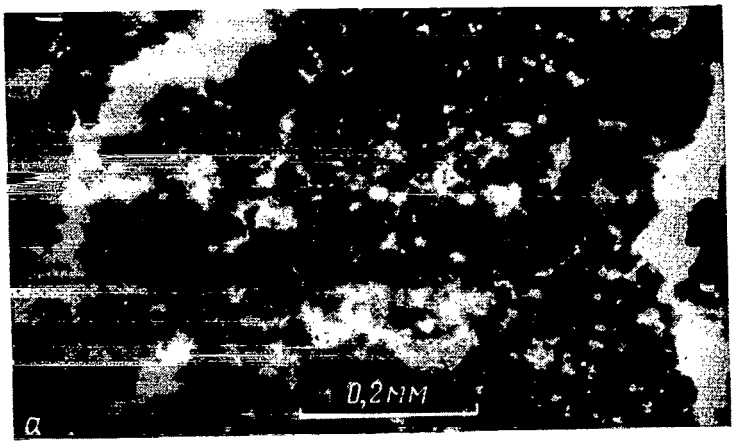


Fig. 18 Migei Carbonaceous Chondrite.

a - General appearance of section; the dark lumps consist of carbonaceous matter; b - Chondrules and grains of silicates (within carbonaceous matter); c - Monomineral olivine chondrule in carbonaceous matter; gas bubble cavities (?) can be seen in the chondrule.

Table 12

X-Ray Photograph and Mineralogical Composition of Migei and Murray Carbonaceous Chondrites

Migei		Murray		Olivine		Pennine		Magnetite	
I	$\frac{d_a}{n}$	I	$\frac{d_a}{n}$	I	$\frac{d_a}{n}$	I	$\frac{d_a}{n}$	I	$\frac{d_a}{n}$
2	7,93	—	—	—	—	2	7,92	—	—
9	7,12	9	7,19	—	—	9	7,18	—	—
2	6,95	—	—	—	—	—	—	—	—
1	5,86	3	6,02	1	5,84	1	5,84	—	—
1	5,36	2	5,39	—	—	5	5,29	—	—
—	—	1	4,71	—	—	10	4,79	—	—
1	4,54	—	—	—	—	4	4,60	—	—
1	4,40	1	4,32	2	4,29	—	—	1	4,21
—	—	1	4,13	—	—	—	—	—	—
1	3,84	4	3,85	7	3,871	—	—	—	—
9	3,53	8	3,59	6	3,491	10	3,585	—	—
1	3,29	1	3,36	—	—	1	3,311	3	(3,31)
1	3,11	1	3,19	—	—	3	3,152	—	—
3	2,985	3	2,965	5	2,995	—	—	6	2,99
1	2,841	4	2,878	—	—	9	2,864	—	—
2	2,787	—	—	—	—	—	—	—	—
—	—	1	2,757	9	2,770	—	—	—	—
1	2,686	1	2,707	—	—	4	2,705	—	—
10	2,530	10	2,557	—	—	10	2,538	10	2,541
4	2,445	5	2,463	10	2,455	10	2,445	—	—
—	—	1	2,393	—	—	6	2,385	—	—
2	2,263	1	2,295	9	2,253	7	2,286	—	—
7	2,145	7	2,172	5	2,165	—	—	—	—
—	—	2	2,114	—	—	—	—	7	2,098
—	—	1	2,058	2	2,032	—	—	—	—
1	2,014	—	—	—	—	—	—	—	—
1	1,988	—	—	—	—	10	2,008	—	—
1	1,917	1	1,944	—	—	6	1,893	—	—
1	1,861	1	1,887	3	1,877	—	—	2	1,884
6	1,785	6	1,798	2	1,788	—	—	4	1,785
—	—	4	1,742	10	1,747	—	—	—	—
6	1,734	—	—	—	—	6	1,741	—	—
—	—	2	1,684	—	—	—	—	5	1,710
1	1,673	—	—	5	1,671	4	1,670	—	—
1	1,612	4	1,612	6	1,619	—	—	9	1,612
—	—	3	1,599	—	—	—	—	—	—
6	1,571	5	1,576	4	1,572	9	1,575	—	—
4	1,537	3	1,539	3	1,539	10	1,535	—	—
—	—	3	1,503	8	1,493	7	1,503	—	—
3	1,489	2	1,488	9	1,481	—	—	—	—
2	1,477	—	—	—	—	4	1,462	9	1,479
—	—	4	1,444	2	1,437	4	1,432	—	—
1	1,429	—	—	—	—	—	—	2	1,411
2	1,390	2	1,384	9	1,397	9	1,405	—	—
1	1,337	—	—	9	1,350	—	—	—	—
1	1,326	1	1,315	9	1,316	7	1,320	3	1,325
2	1,309	—	—	—	—	—	—	—	—
—	—	1	1,295	3	1,295	7	1,295	—	—
2	1,262	2	1,269	2	1,267	—	—	2	1,264
1	1,239	1	1,245	3	1,238	—	—	—	—
—	—	2	1,193	1	1,209	—	—	3	(1,209)
—	—	1	1,181	7	1,188	—	—	—	—
1	1,164	—	—	—	—	—	—	—	—
1	1,137	1	1,141	7	1,137	—	—	—	—
1	1,122	1	1,116	5	1,125	—	—	4	1,119
—	—	2	1,103	—	—	—	—	—	—
1	1,096	—	—	8	1,098	—	—	8	1,091
—	—	3	1,072	—	—	—	—	—	—
—	—	2	1,061	—	—	—	—	—	—
—	—	4	1,036	—	—	—	—	—	—
—	—	3	1,019	—	—	—	—	—	—

Table 13

Chemical Composition of the Migei Carbonaceous Chondrite

Components	A 1	A 2	B 1	B 2
SiO_2	31,00	5,63	24,29	54,27
MgO	29,73	2,83	18,10	29,68
FeO	24,44	2,10	22,63	3,42
Al_2O_3	—	0,11	1,38	4,08
CaO	—	0,31	1,85	4,35
Na_2O	—	—	1,25	2,02
K_2O	—	—	0,13	0,62
MnO	—	Trace	0,55	1,73
Fe_{met}	—	—	2,94	—
Ni+Co	—	—	1,01	—
FeS*	—	—	0,46	—
$S_{free}+SO_3$	—	—	3,52	—
Organic matter	—	4,72	—	—
Total	85,17	15,70	78,11	100,17

C Components	3	C Components	4	C Components	5
SiO_2	27,81	Fe	21,24	Mg	33,19
MgO	19,46	Si	12,98	Si	31,85
FeO	19,13	Mg	11,73	Fe	26,18
Al_2O_3	2,15	S	3,66	Al	2,90
CaO	1,66	C	2,48	Ca	2,04
Na_2O	0,63	H	1,44	Ni	1,41
K_2O	0,05	Al	1,14	Na	1,40
P_2O_5	0,30	Ni	1,20	Cr	0,33
Cr_2O_3	0,36	Ca	1,18	P	0,29
MnO	0,21	Na	0,47	Mn	0,19
TiO_2	0,08	Cr	0,24	K	0,07
Fe	0,00	Mn	0,16	Ti	0,09
Ni	0,00	P	0,13	Co	0,06
Co	0,00	Co	0,05		
NiO	1,53	Ti	0,05	Total	100,00
CoO	0,07	K	0,042		
FeS*	10,05	(O)	41,81		
H_2O	12,86				
C	2,48	Total	100,00		
Others	0,36				
Total	99,19				

A--Analysis by Meunier (1889), B--Analysis by P. G. Melikov and L. V. Krshishanovskiy (1896); 1 - Part dissolved in HCl, 2 - Part insoluble in HCl (% by weight); C--Analysis by Wiik (1956); 3 and 4 - % by weight, 5 - atom.% (volatile elements excluded).

*See footnote 1 on page 34.

Monomineral olivine chondrules (Figure 18, c) are also sometimes observed. Chondrules with a microporphyritic structure sometimes consist of tiny crystals of pyroxene and glass of a brownish color. The carbonaceous matter contains grains of sulfide, most of which have an irregular configuration, with diameters up to 0.2 mm, and grains of ferronickel. In addition, I. A. Yudin (1958) noted that this meteorite contains infrequent grains of chromite. According to I. A. Yudin and N. F. Obotnin (1961), the composition of the mineral phase of the Migei meteorite is as follows (in % by volume): Silicate part 99.63, troilite 0.29, ferronickel 0.08, chromite -- infrequent grains. DuFresne and Anders (1962b) noted that this meteorite contains pentlandite, spinel, free sulfur, gypsum.

In addition, the meteorite was found to contain devitrified glass which was not stable when heated above 300° (DuFresne, Anders, 1961).

The Migei meteorite was studied by the author using an electron microscope and by the roentgenometric method. It was found in the study under the electron microscope that the meteorite matrix is a fine mixture of carbonaceous /47 matter and mineral particles. Chlorite-serpentine minerals which are present in the interchondrule space have the appearance of fibrous (needle-shaped) formations (Figure 19, a) and also platelet-forms (Figure 19b). As a comparison, Figure 20 shows an electron microscope photograph of chrysotile from the serpentinite of the Southern Urals.

The roentegnometric survey was made in a chamber 57.3 mm in diameter with iron radiation by an ion tube with a manganese filter.

Figure 21 shows X-ray photographs of the Migei meteorite, the Murray carbonaceous chondrite and the Kainsaz chondrite, which contains carbon.

The results of this roentegnometric study (Table 12) confirm the conclusion that the greater part of the meteorite consists of allodium and hydrous ferromagnesium silicates. In addition, the X-ray photograph reveals magnetite.

Table 13 gives the chemical composition of the Migei meteorite. Meunier (1889) noted that the meteorite contains 4.72% organic matter. The carbon in the Migei meteorite was found to be 2.48% (Wiik, 1956), 2.5% (Trofimov (1950), 2.6% (Boato, 1954a), 2.76% (Mueller, 1966).

Staroye Boriskino /49

The meteorite fell on 20 April 1930 near the village of Staroye Boriskino in Orenburgskaya Oblast (Russian Soviet Federative Socialist Republic). It weighed 1.34 km. Most of the meteorite is in the meteorite collection of the Committee on Meteorites Academy of Sciences USSR. A sample is also preserved at Odessa University (365 grams); other samples are available elsewhere.

The circumstances of falling of this meteorite are described in studies by Ye. L. Krinov (1931, 1932). A detailed mineralogical description of the Staroye Boriskino carbonaceous chondrite has been given by L. G. Kvasha (1948).

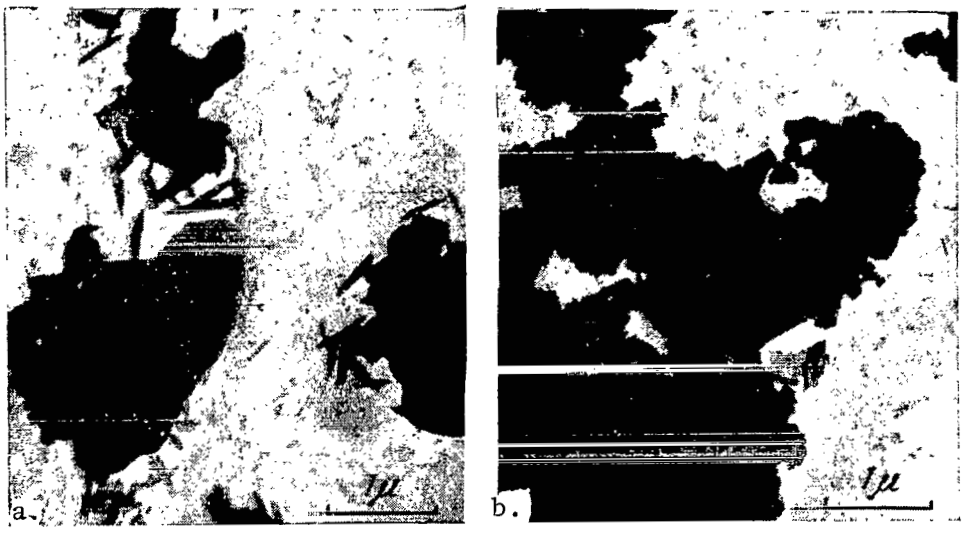


Figure 19. Electron Microscope Photograph of a Dispersed Sample of the Migei Meteorite (Mineral grains with contiguous needle-shaped formations (a) and platelets (b) of hydrous silicates).

Figure 20. Electron Microscope Photograph of Chrysotile Fibers (Serpentine from the Southern Urals). Magnification 3,800.

For the first time for meteorites she established microscopically the presence of hydrous silicates of the phyritic type; this was confirmed by chemical and thermal analyses of the meteorite (Kvasha, 1948; Zabaratskiy, 1956; Zabarastkiy, 1952). In addition, Kvasha also found that the meteorite contained calcite. She recently studied this meteorite again, devoting her principal attention to secondary minerals, the structure of their aggregates and the nature of secondary changes in the meteorite (Kvasha, Wiik, 1964); this same article gives a new analyses of the meteorite made by Wiik.

Data from a roentgenometric study of the Staroye Boriskino meteorite, presented in a study by N. N. Stulov (1960), confirmed that the meteorite contained minerals of the chlorite-serpentine type. The same results were obtained by I. A. Yudin and N. F. Obotnin (1961). The author (Vdobykin, 1960) established that this meteorite contains organic compounds; this was confirmed by Wiik (Kvasha, Wiik, 1964).

In the meteorite sample obtained by the author of this book for his study, a small part of the fused crust with piezoglyptes was preserved. The crust had a spongy structure. The color of the crust and the upper sub-crustal part of the sample was a golden bronze.

Under the microscope a clastic structure can be noted (Section 191); this was described earlier by L. G. Kvasha (Zavaritskiy, Kvasha, 1952). The fragments are up to 7 mm in diameter. Most of the fragments are black in color. Angular sectors of a lighter color are sometimes noted.

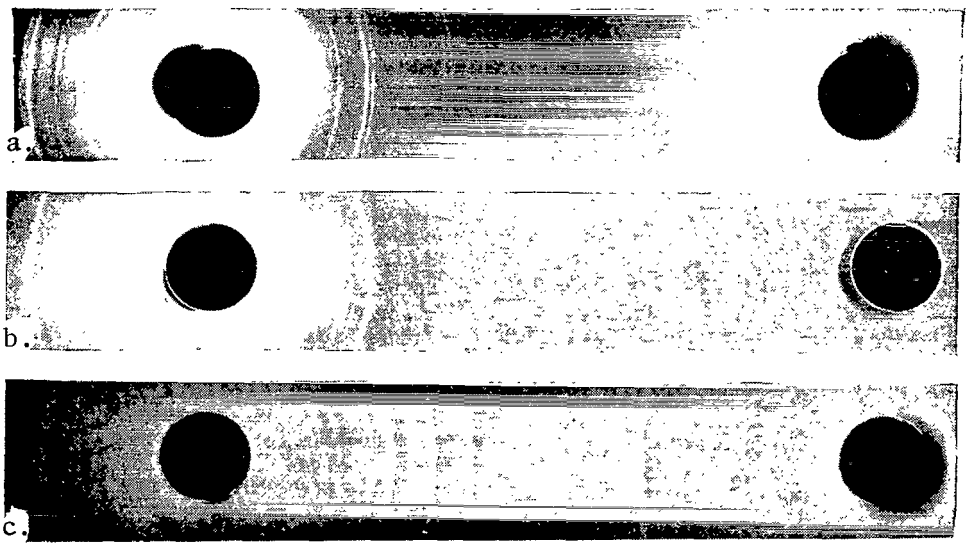


Figure 21. X-Ray Photographs of Pulverized Matter from Meteorites. a - Migei carbonaceous chondrite; b - Murray carbonaceous chondrite; c - Kainsaz chondrite containing carbon; FeK_{α}, manganese filter.

The matter separating the chondrules, as in the Migei meteorite, has a lumpy structure. Chondrules of a rounded or oval configuration measuring 0.1-0.5 mm are scattered at random in the matrix. The structure of the chondrules is frequently microporphyritic or microlithic. The chondrules consist for the most part of small crystals of olivine, but also pyroxenes--clinopyroxenes, according to L. G. Kvasha (1948), and glassy matter. In many cases they contain chlorite-serpentine minerals. /51

Mason (1962a, b) believes that in carbonaceous chondrites the chlorite-serpentine minerals were formed earlier than the olivine. However, in the example of the Staroye Boriskino meteorite, the secondary nature of hydrous ferramagnesial silicates can be seen very clearly.

It can be seen in the sections of the Staroye Boriskino meteorite that chlorite-serpentine minerals not only replace small grains in the space separating the chondrules, but also frequently form the substance of chondrules.

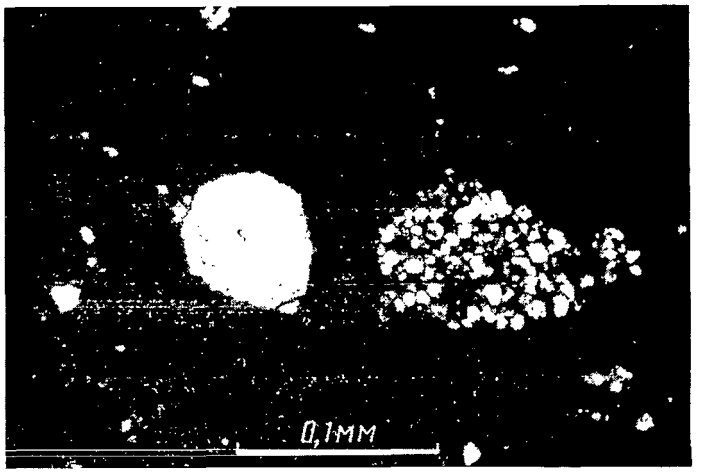


Figure 22. Olivine Chondrules in the Carbonaceous Matter of the Staroye Boriskino Meteorite.

At left - completely crystallized chondrule surrounded by a narrow rim of chlorite-serpentine minerals; at right - a chondrules consisting of individual rounded grains of silicate. Nicols +.

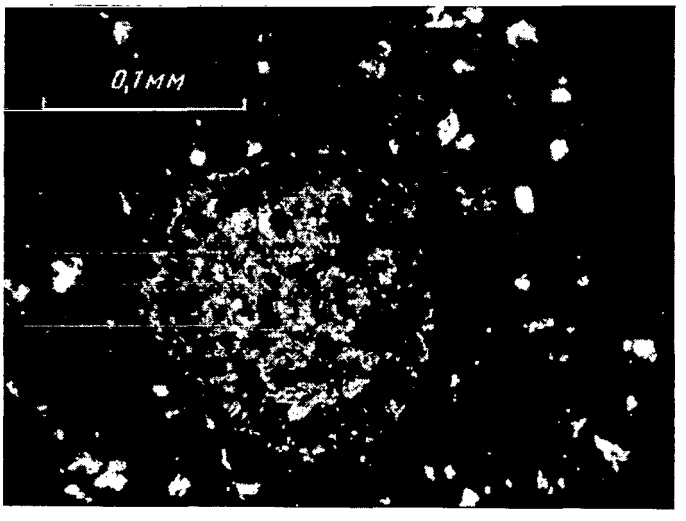


Figure 23. Chondrule Consisting of Chlorite-serpentine of Veined Structure. Staroye Boriskino Meteorite. Without analyzers.

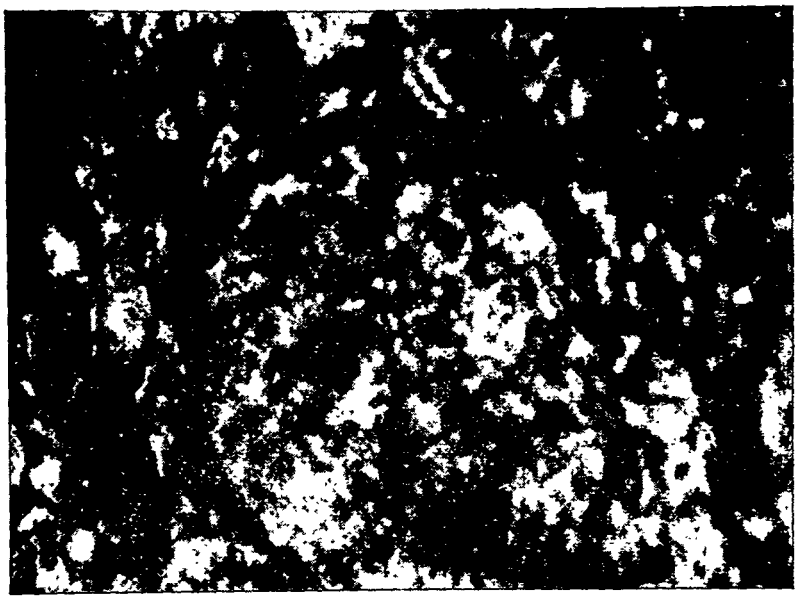


Figure 24. Chondrule Partially Replaced by Chlorite-Serpentine Minerals. Staroye Boriskino Meteorite. Magnification 280, without analyzer.

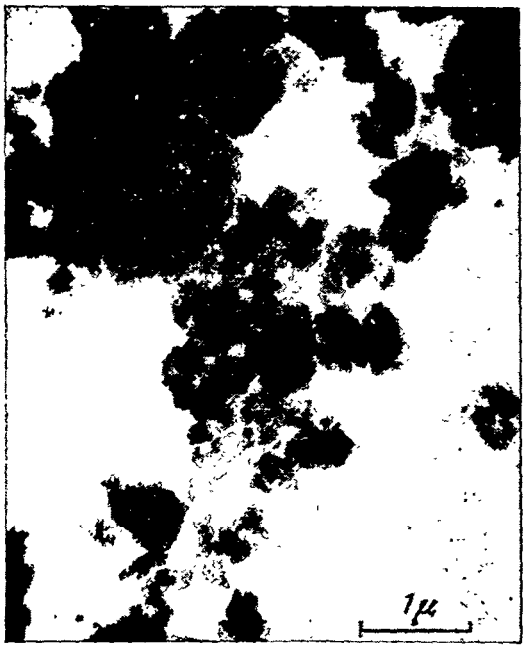


Figure 25. Microscope Photograph of a Dispersed Sample of the Staroye Boriskino Meteorite.

The characteristic replacement of chondrules is highly varied. Sometimes they only rim the olivine chondrules in the form of a narrow shell (Figure 22) or in the form of veinlets are developed within individual tiny olivine crystals. There are some chondrules consisting of chlorite-serpentine, but with a tiny olivine grain in the center. Some chondrules have been completely replaced by chlorite-serpentine, frequently having a veined structure (Figure 23).

Hydrous silicates are particularly abundant in the brighter sectors where there are entire zones which are enriched with chlorite-serpentine; here these silicates frequently completely replace the chondrules. Replacement by chlorite-serpentine was apparently accompanied by a process of destruction of the chondrules from their peripheries. Figure 24 shows a chondrule which has been partially replaced by hydrous silicates. It is surrounded by individual

Table 14

Chemical Composition of the Staroye Boroskino Carbonaceous Chondrite

Components	A 1	A 2	B Others	B 2	C Total	C 2
SiO_2	26,73	27,61	Fe	22,15	Mg	31,56
MgO	19,35	18,46	Si	12,89	Si	31,65
FeO	—	18,00	Mg	11,13	Fe	27,35
Al_2O_3	3,11	2,29	S	3,23	Al	3,10
CaO	2,22	1,64	C	2,06	Ca	1,97
Na_2O	1,07	0,78	H	1,34	Ni	1,43
K_2O	0,20	0,036	Al	1,21	Na	1,74
P_2O_5	—	0,33	Ni	1,21	Cr	0,37
Cr_2O_3	0,30	0,42	Ca	1,17	P	0,31
MnO	0,15	0,22	Na	0,58	Mn	0,21
TiO_2	0,15	0,10	Cr	0,28	K	0,05
Fe	—	0,00	Mn	0,17	Ti	0,09
Ni	1,15	—	P	0,14	Co	0,07
Co	0,03	—	Co	0,059		
Cu	—	0,03	Ti	0,06		
Fe_2O_3	29,79	4,77	K	0,030		
NiO	—	1,54	Cu	0,030		
CoO	—	0,08	V	0,013		
V_2O_5	—	0,023	N	0,07		
FeS*	—	7,59	(O)	41,93		
SO_3	—	1,46				
H_2O^+	8,72	11,97				
H_2O^-	2,96					
N	—	0,07				
C	—	2,06				
Trace ($C+CO_2$)	3,75	—				
S	1,02	—				
Total	100,70	99,48				

A, B - % by weight; C - atom. % (volatile elements excluded): 1 - analysis made by O. A. Alekseyeba (Kvasha, 1948, first individual sample); 2, analysis by Wiik (Kvasha, Wiik, 1964 (second individual sample).

*See footnote 1 given earlier.

grains of silicate incorporated into the carbonaceous matter and arranged in such a way that they duplicate the configuration of the chondrule. This chondrule in actuality is possibly the central part of an earlier existing large chondrule which was destroyed during the secondary replacement of its matter.

In the Staroye Boriskino meteorite well-identified calcite grains are associated with the carbonaceous matter and the chlorite-serpentine minerals; this was noted earlier by L. G. Kvasha (1948; Kvasha, Wiik, 1964). For the time being this carbonate has not been found in other carbonaceous chondrites.

Sulfides are among the opaque minerals which can be seen under the microscope. L. G. Kvasha (1948) notes that the sulfides are represented by torilite and pyrrhotite. The configuration of these grains is frequently isometric and they are up to 10 μ in diameter. They are usually distributed through the /52 space separating the chondrules, but sometimes they are found in the chondrules themselves. Ferronickel grains up to 0.1 mm in diameter are visible in rare cases. Their configuration is nearly round. Ferronickel grains hundreds of a millimeter in diameter are also found within olivine chondrules. Sometimes they are somewhat replaced along the periphery by a gray mineral goethite with the formation of a corroded structure (I. A. Yudin and N. F. Obotnin, 1961).

An electron microscope study of this meteorite (Vdovykin, 1965a) shows that in the meteorite matrix platey transparent grains of chlorite-serpentine minerals occur in a mixture with carbonaceous matter (Figure 25).

With respect to its chemical composition the Staroye Boriskino meteorite is rather similar to the Migei carbonaceous chondrite (Table 14). Its carbon content is 2.06% (Mason, 1963d; Kvasha, Wiik, 1964), or 2.65% (Trofimov, 1950).

Cold Bokkeveld

This meteorite fell in South Africa on 13 October 1838. Its falling was /53 preceeded by a bolide and explosive effects. Several samples were collected, the largest of which weighed about 2 kg.(Prior, Hey, 1953). This meteorite is represented in many museums throughout the world. In the meteorite collection of the Academy of Sciences USSR there is a sample weighing approximately 103 gr.

The meteorite was investigated earlier by a number of scientists. The literature on this meteorite has been compiled by Wülfing (1897). The chemical analysis of the meteorite was made by Wöhler, Hörnes, 1859); a modern analysis has been made by Wiik (1956). Recently it was also analyzed by Briggs, 1963a). The results of a roentgeometric study revealing the presence of chlorite-serpentine minerals in the meteorite are given by N. N. Stulov (1960). The mineralogical composition of the meteorite was also investigated by DuFresne and Anders, 1962b)[1]. L. G. Kvasha (1963) recently found carbonates in this meteorite. A modern study of the organic compounds in meteorites was begun in a study by Mueller (1953) on the Cold Bokkeveld meteorite.

[1] DuFresne and Anders observed asterism for the olivine of the Cold Bokkeveld meteorite.

The Cold Bokkeveld meteorite is rather friable and has a low specific gravity (2.65).

It can be seen under the microscope (Section 26) that the meteorite mass separating the chondrules has a nonuniform lumpy structure of a brownish color, sometimes with a greenish hue caused by the presence of hydrous silicates. The carbonaceous matter is not only scattered through the mass separating the chondrules, but is sometimes found in the chondrules themselves and frequently replaces the chondrules along the perphipery (Figure 26). Small chondrules up to 0.2-0.4 mm in diameter, as well as small individual olivine crystals, are scattered through the matrix. Olivine chondrules for the most part have a microlithic structure, frequently of round configuration, but sometimes somewhat elongated (Figure 27). Less frequently the chondrules consist of pyroxene; their structure is frequently microvorphyrithic (Figure 28). Glassy brown matter is quite abundant.

An electron microscope study (Vdovykin, 1965a) confirms the presence of chlorite-serpentine minerals in this meteorite (Figure 29).

Infrequent grains of ferronickel and sulfides can be seen in the sections in reflected light. Magnetite has been discovered in the meteorite using the roentgenometric method (Stulov, 1960; DuFresne, Anders, 1962b, 1963), and also the electron-diffraction method (Vdobykin, 1965a). Carbonates (Kvasha, 1963) and gypsum (DuFresne, Anders, 1963) have also been found in this meteorite.

Wood (1965, 1967) recently made a study of grains of ferronickel and sulfide in a number of carbonaceous chondrites, including the Cold Bokkeveld meteorite, by the X-ray spectral microanalysis method. He noted that the ferronickel range in this meteorite in most cases is round and has a diameter up to 50 μ. The ferronickel is represented by kamacite with a low nickel content (about 5-6% by weight); sulfides are represented by troilite and pentlandite; the troilite contains 1-3% nickel. The pentlandite of this meteor contains about 10% nickel. Wood found that in the matter of the Cold Bokkeveld present among the chondrules the mean nickel content is 1.3% and the mean iron content is 21.2%.

Table 15 gives the chemical composition of the Cold Bokkeveld meteorite; it can be seen that meteorite is a typical representative of type-II carbonaceous chondrites. The results of the three analysis of the Cold Bokkeveld meteorite agree rather well with one another but in the analysis made by Wöhler, the Na_2O and MnO contents are exaggerated.

The carbon content in this meteorite is: 1.30% (Wiik, 1956), 1.32% /56
(Briggs, 1963a), 1.52% (Wöhler, Hörnes, 1859), 1.6% (Boato, 1954a), 1.95% (Trofimov, 1950).

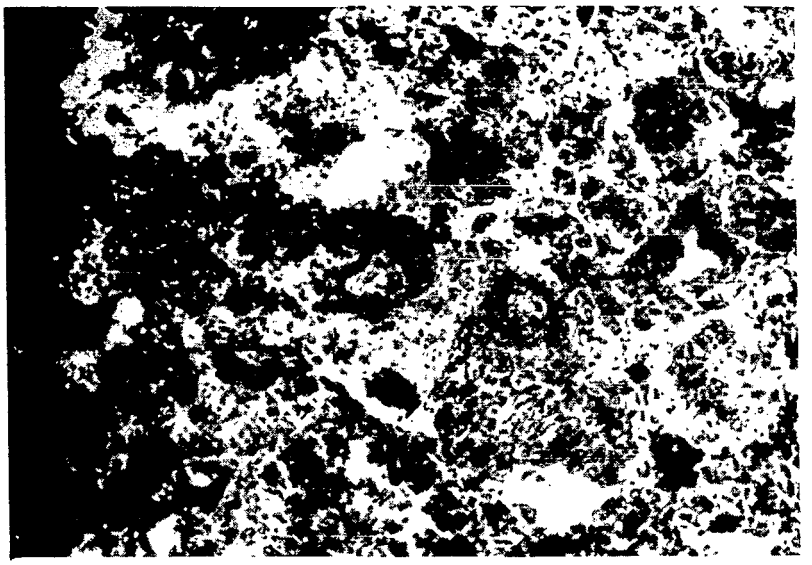


Figure 26. Substitution of Periphery of Chondrule by Carbonaceous Matter. Cold Bokkeveld Meteorite. Magnification 280, without analyzer.

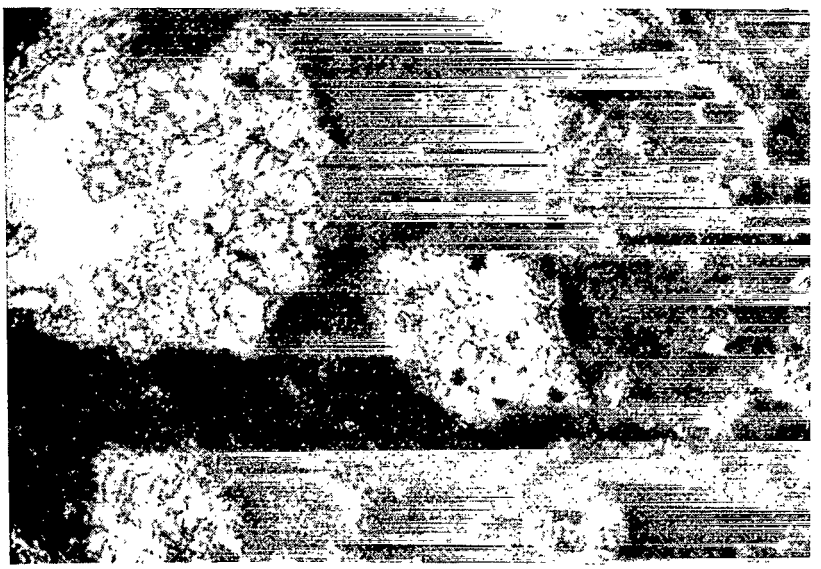


Figure 27. Distribution of Chondrules and Olivine Grains in Carbonaceous Matter of Lumpy Structure. Cold Bokkeveld Meteorite. Magnification 270, without analyzer.

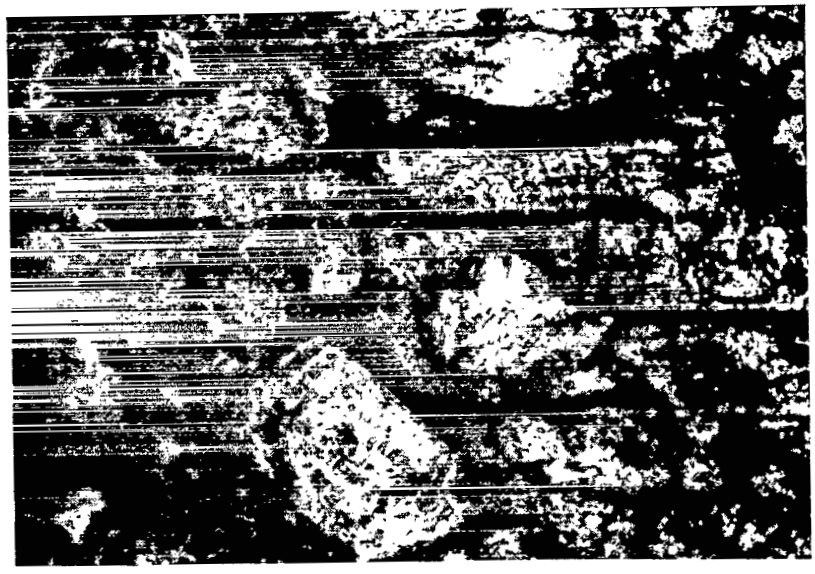


Figure 28. Fragment of Chondrule with a Microporphyritic Structure. Cold Bokkeveld Meteorite. Magnification 280, without analyzer.

Figure 29. Electron Microscope Photograph of Dispersed Sample of Cold Bokkeveld Meteorite. Translucent Platelets of Chlorite-Serpentine Minerals can be seen adjoining large grains and aggregates.

Table 15

Chemical Composition of Cold Bokkeveld Carbonaceous Chondrite

	A			B		C	
Components	1	2	3	Others	3	Others	3
SiO_2	28,09	27,51	27,33	Fe	20,85	Mg	32,67
MgO	20,24	19,01	18,73	Si	12,76	Si	32,02
FeO	23,34	20,51	20,17	Mg	11,29	Fe	26,28
Al_2O_3	1,87	2,21	2,29	S	2,98	Al	3,17
CaO	1,55	1,55	1,56	C	1,30	Ca	1,96
Na_2O	1,12	0,63	0,61	H	1,70	Ni	1,41
K_2O	—	0,05	0,05	Al	1,21	Na	1,37
P_2O_5	Сл.	0,31	0,30	Ni	1,17	Cr	0,39
Cr_2O_3	0,69	0,45	0,42	Ca	1,11	P	0,30
MnO	0,88	0,20	0,19	Na	0,45	Mn	0,19
TiO_2	—	0,08	0,08	Cr	0,29	K	0,07
Fe	0,00	0,00	0,00	Mn	0,15	Ti	0,09
Ni	0,00	0,00	0,00	P	0,13	Co	0,08
Co	0,00	0,00	0,00	Co	0,06		
NiO	1,50	1,50	1,49	Ti	0,05	Total	100,00
CoO	Сл.	0,09	0,08	K	0,042		
FeS*	8,44	8,23	8,16	(O)	44,46		
H_2O	10,50	15,91	15,17				
C	1,52	1,33	1,30	Total	100,00		
Trace	0,23	—	2,23				
Total	99,97	99,57	100,16				

A and B - % by weight; C - atom. % (volatile elements excluded); 1 - Wöhler, Hörnes (1859); 2 - Briggs (1963a); 3 - Wiik (1956).

* See footnote 1 given earlier.

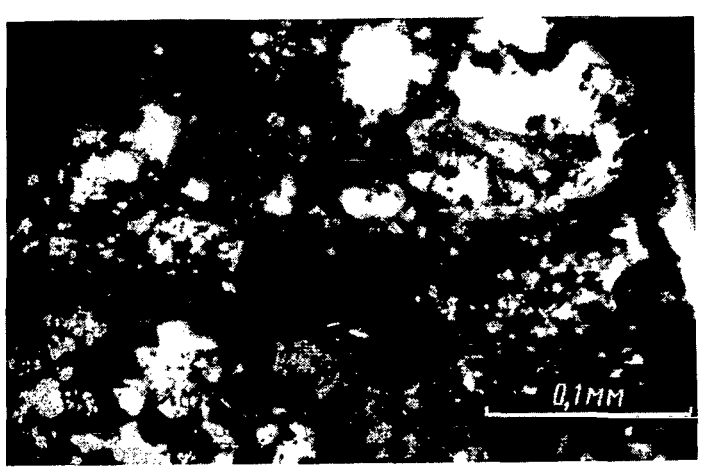


Figure 30. Chondrules and Individual Silicate Grains Distributed in the Matrix of the Groznaya Carbonaceous Chondrite. Without analyzer.

Murray

This meteorite fell in the form of a meteoritic rain on 20 September 1950 in Kentucky in the United States. The falling was accompanied by a bolide. Several individual samples with a total weight of about 13 kg were found. The largest sample weighed 3.4 kg. Most of the meteorite is now in the Washington National Museum.

The circumstances of the falling of the meteorite, its description and chemical analysis are described in an article by Horan (1953). According to his data, the geocentric velocity of the meteorite was 71 km/sec. This meteorite has been studied by DuFresne and Anders (1962b). L. G. Kvasha (1963) found that it contained carbonates. The meteorite was analyzed by Wiik (1956) and Briggs (1963a).

This meteorite is almost black in color, also caused by the high content of carbonaceous matter. Under the microscope (Section 2309) it can be seen that the carbonaceous matter of the meteorite is opaque; it is black in color with a brownish hue and it has a lumpy structure. In the matrix, which constitutes about 70% of the meteorites volume (Fredriksson, Keil, 1964), and which, like other carbonaceous chrondrites, is a fine mixture of carbonaceous matter and mineral components, there is a uniform distribution of silicate /57 chrondrules, most of which are polycrystalline, of rounded form, 0.05-0.5 mm in diameter, but also finer grains of silicate. The structure of the olivine chrondrule is microlithic. Some chondrules have ill-defined outlines and along the periphery are replaced by carbonaceous matter which sometimes is also found within the chrondrule. The replacement of olivine by chlorite-serpentine can be noted. Chondrules of microporphyrithic structure consists of tiny crystals for clinopyroxene, represented by enstatite and clinoenstatite (Kerridge, 1964). The meteorite also contains carbonate (Kvasha, 1963), calcium sulfate (Kerridge, 1964), and sulfur (DuFresne, Anders, 1963). Sulfides and ferronickel are found both within the cementing mass of the chondrule and within the chondrules. Their grains attain 0.1 mm in diameter (Fredriksson, Kiel, 1964).

Using the roentgenometric method, this meteorite was found to contain olivine and chlorite-serpentine minerals, the basic components of the meteorite, as well as magnetite (Vdovykin, 1965a). The X-ray photograph of this meteorite is comparable to the X-ray photograph of the Migei carbonaceous chondrite (see Figure 21, Table 12).

Fredriksson and Keil (1964) used the X-ray spectral microanalysis method for studying the distribution of Fe, Mg, Ca and Ni in silicates (olivine, pyroxene and chlorite-serpentine minerals) and also determined the composition of troilite, rich in nickel, pentlandite and ferronickel in this meteorite. The authors noted a variability in composition of the olivine and pyroxene. In the olivine grains they found tiny grains of ferronickel in which the nickel /58 content varied in the range 5-9%. These researchers noted a high nickel content and 1-4% sulfur in the matrix separating the chondrules, where they found finely dispersed pentlandite, as well as grains of other sulfides and iron oxides less than 1 μ in diameter.

Table 16

Chemical Composition of the Murray Carbonaceous Chondrite

A			B		C	
Components	1	2	Others	2	Others	2
SiO_2	27,56	28,69	Fe	21,25	Mg	33,56
MgO	19,91	19,77	Si	13,39	Si	32,70
FeO	20,95	21,08	Mg	11,92	Fe	26,13
Al_2O_3	2,21	2,19	S	2,80	Al	2,26
CaO	1,78	1,92	C	2,78	Ca	2,34
Na_2O	0,41	0,22	H	1,39	Ni	1,38
K_2O	0,06	0,04	Al	1,16	Na	0,49
P_2O_5	0,34	0,32	Ni	1,18	Cr	0,40
Cr_2O_3	0,46	0,44	Ca	1,36	P	0,32
MnO	0,20	0,21	Na	0,16	Mn	0,19
TiO_2	0,10	0,09	Cr	0,30	K	0,05
Fe	0,00	0,00	Mn	0,16	Ti	0,10
Ni	0,00	0,00	P	0,14	Co	0,08
Co	0,00	0,00	Co	0,06		
NiO	1,58	1,50	Ti	0,054	Total	100,00
CoO	0,08	0,08	K	0,033		
FeS*	7,51	7,67	(O)	41,86		
H_2O^+	12,56	9,98	Total	100,00		
H_2O^-		2,44				
C	2,91	2,78				
Others	—	0,62				
Total	98,62	100,04				

A and B - % by weight; C - atom. % (volatile elements excluded); 1 - Briggs (1963a); 2 - Wiik (1956).

* See footnote 1 on page 34.

Wood (1967), who made an analysis of the Murray meteorite by this same X-ray spectral microanalysis method, also demonstrated the presence of troilite, containing up to 2% nickel, and pentlandite (about 25% nickel). According to his data, the nickel content in the ferronickel is up to 15%.

Table 16 gives the chemical composition of the Murray meteorite. The carbon content in this meteorite is: 1.9% (Boato, 1954a), 1.99% (Horan, 1953), 2.57% (Mueller, 1966), 2.78% (Wiik, 1956), 2.91% (Briggs, 1963a).

Type III Carbonaceous Chondrite

Groznaya

This meteorite fell on the territory of the present-day Severo-Osetinskaya ASSR on 28 June 1861. The falling was accompanied by an explosive effect. Only one individual sample weighing 3.25 kg was found; the other fragments fell in the Terek River (Krinov, 1948). Most of the meteoritesis now in the meteorite collection of the Academy of Sciences USSR (about 1.8 kg). Samples of this meteorite can be found in the British museum and other museums as well.

The meteorite has been described by Rose (1862, 1864) and Tschernak (1878). A microscopic study of the meteorite has been made by A. N. Zavaratsiky and L. G. Kvasha (1952); a roentgenometric analysis has been made by I. A. Yudin and M. F. Obotnin (1961). The chemical composition of the meteorite was studied by Plon (Tschernak, 1878). A new chemical analysis of the Groznaya meteorite was recently made by Wiik (Mason, 1963d).

The meteorite is dark grey in color. It is denser and heavier than the meteorite described above; its specific gravity is 3.49. Silicate chondrules and mineral grains are distributed nonuniformly in the matrix. The chondrules are up to 2 mm in diameter, that is, they are larger than in Type II carbonaceous chondrites.

The nonuniform distribution of olivine and pyroxene chondrules and a large number of mineral grains in the matrix, having a brownish-dirty grey color, can be clearly seen under the microscope (Section 3,213, Figure 30). The chondrules frequently have a regulated rounded form. The structure of the chondrule is microporphyritic, fully crystalline, less frequently the structure is gratelike or eccentrically radial. Carbonaceous matter is sometimes found within the chondrule (Figure 31). The chondrules with an eccentrically radial structure consist of rhombic pyroxene. Olivine grains are visible along the edges of such chondrules with transition to the matrix. A glassy substance of a brown color is observed in some cases.

Grains of ferronickel and troilite from 0.01 to 0.05 mm in diameter are distributed in the space surrounding the chondrule and also inside the silicate chondrule (Zavaritskiy, Kvasha, 1952); magnetite is also found there. I. A. Yudin (1958), notes that tiny chromite grains are present in aggregates with ferronickel and troilite.

Wood (1967), using the X-ray spectral microanalysis method, found that the troilite in the Groznaya contains 1-3% nickel. In addition Wood noted pentlandite containing up to 25% nickel. According to his data the space surrounding the chondrules on the average contains 26% iron and 1% nickel.

The author of this book, using the chloroform extraction method, was able to separate out sulfur from the Groznaya meteorite in addition to organic matter (Vdovykin, 1962b). The tiny sulfur crystals, upon drying out of the solvent, formed droplike aggregates of a yellow color 0.2-1 mm in diameter

(Figure 32). The hardness of this sulfur, determined by the author by the microhardness method (Vdovykin, 1967a), varied in the range 24-35 kg/mm^2 with a mean value H_{mean} = 30 kg/mm^2. With respect to hardness it does not differ from sulfur of terrestrial origin. /60

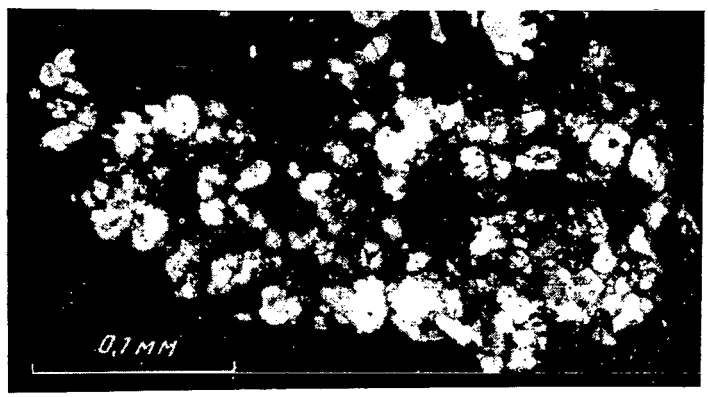


Figure 31. Olivine Chondrules Present within Carbonaceous Matter. Groznaya Meteorite. Nicols +.

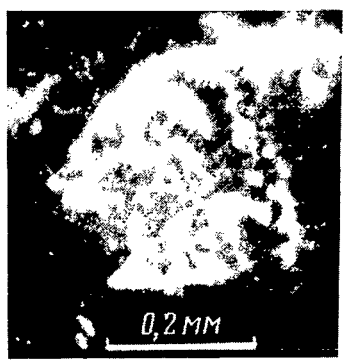


Figure 32. Aggregate of Tiny Sulfur Crystals Extracted by the Chloroform Method from the Groznaya Carbonaceous Chondrite (fixed in epoxy resin and polished). Reflected light.

With respect to its chemical composition the Graznaya meteorite is a typical representative of type III carbonaceous chondrite (Table 17). The carbon content is 0.56% (Mason, 1963d), 0.68% (Tschernak, 1878), 0.78% (Trofimov, 1950).

Kainsaz Chondrite Containing Carbon

This meteorite is classified as an unmodified black clinoenstatitic chondrite (Chirvinskiyi, Ushakova, 1946) or as an unmodified chondrite (dark variety) (Zavaritskiy, Kvasha, 1952; Kvasha, 1962).

The meteorite fell in the form of an abundant meteoritic rain on the territory Tatriskiya ASSR on 13 September 1937. Fifteen samples with a total weight of more than 210 kg were collected. The largest sample (102.5 kg is in the meteorite collection of the Academy of Sciences USSR.

Table 17

Chemical Composition of the Groznaya Carbonaceous Chondrite

A		B		C	
Components	1	Components	2	Components	2
SiO_2	33,78	Fe	24,80	Mg	32,83
MgO	23,55	Si	14,91	Si	31,84
FeO	28,8	Mg	13,31	Fe	26,65
Al_2O_3	3,446	S	2,10	Al	2,54
CaO	3,22	C	0,56	Ca	2,53
Na_2O	0,63	H	0,45	Ni	1,27
K_2O	0,30	Al	1,14	Na	1,28
FeS	5,37	Ni	1,24	Cr	0,36
C	0,68	Ca	1,69	P	0,33
H	0,17	Na	0,49	Mn	0,18
Total	100,00	Cr	0,31	K	0,04
		Mn	0,16	Ti	0,09
		P	0,17	Co	0,06
		Co	0,061	Total	100,00
		Ti	0,072		
		K	0,024		
		Cu	0,042		
		V	0,013		
		N	0,22		
		(O)	38,23		
		Total	100,00		

A and B = % by weight; C - atom. % (volatile elements excluded); 1 - analysis by Plon (Tschernak, 1878); 2 - Analysis by Wiik (Mason, 1963d).

The circumstances of falling of the meteorite have been described by L. S. Selivanov (1941) and N. N. Sytinskiya (1941). A chemical and petrographic study of this meteorite was made by P. N. Chirbinskiy and V. G. Ushakova (1946). The meteorite was studied by A. N. Zavaritskiy and L. G. Kvasha (1952) and I. A. Yudin (1958, 1965). The age of hardening of the meteorite matter was determined by E. K. Gerling and K. G. Rik (1954). A. V. Troflimov (1950) studied the isotopic composition of the carbon. Mueller (1966) recently /61 determined the content of volatile elements in the meteorite. M. I. D'yakonova (1964) made a chemical analysis of the Kainsaz meteorite.

The meteorite is of a dark grey color; it contains white chondrules, sometimes translucent, with a brownish hue. The chondrules are dense and do not break in fracture. The specific gravity of the meteorite is 3.93.

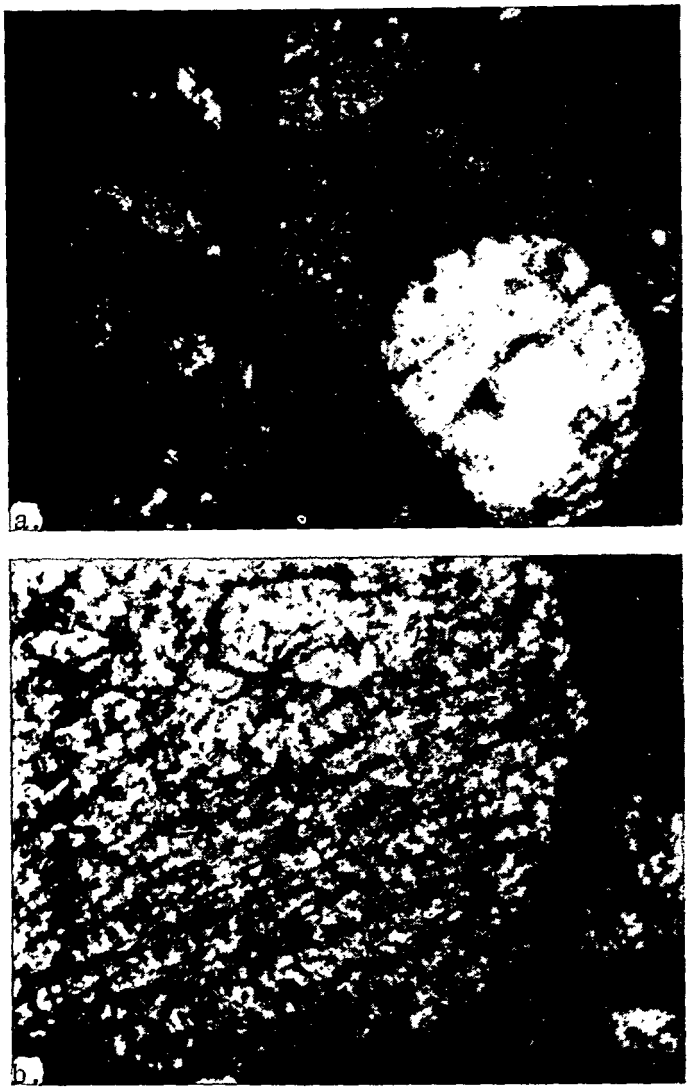


Figure 33. Olivine Chondrules, Fully Crystalline (a) and Gratelike (b) in Structure in Opaque Matrix of the Kainsaz Carbon-bearing Chondrite. Magnification 280, without analyzer.

Under the microscope it can be seen (Section 1093) that chondrules and grains of silicate are distributed rather uniformly in the matrix. The structure of the matter surrounding the chondrules is more homogeneous than in carbonaceous chondrites. There is no evidence of lumpiness (Figure 33). Most of the chondrules are above 0.3 mm in diameter, but there are some chondrules up to 1 mm or more in diameter. They consist of olivine and pyroxenes. The olivine chondrules have a fully crystalline structure (Figure 33a) and a gratelike stru- /63 cture (Figure 33b). Sometimes the grains in the chondrules are separated by a glassy substance of a light brown color; the structure of such chondrules is microporphyritic.

Pyroxenes are observed in the form of individual tiny crystals or form chondrules of an eccentrically radial structure (Figure 34). P. N. Chirbinskiy and Z. G. Ushakovaka (1946) noted clinoenstatite, clinobronzite and augite among the pyroxenes. Rounded grains of ferronickel up to 0.2-0.3 mm in diameter, which sometimes are also observed within the pyroxene chondrules, are scattered in the cementing mass surrounding the chondrules, representing some of the oqaque minerals (Zabaritskiy, 1956). However most of the ferronickel, as well as the troilite, is represented by small grains of irregular shape. In addition, I. A. Yudin noted native copper and chromite in the Kainsaz meteorite (Yudin, 1958; Keil, Fredriksson, 1963). According to I. A. Yudin (1965), the ferronickel content in the Kainsaz chondrite averages 10%; it is represented by kamacite and taenite; native copper is found in the form of individual small grains (0.01-0.015 mm); the troilite content is 4.3%. Chromite is found in the form of individual grains; there is also an unknown mineral; I. A. Yudin feels that it may possibly be osbornite.

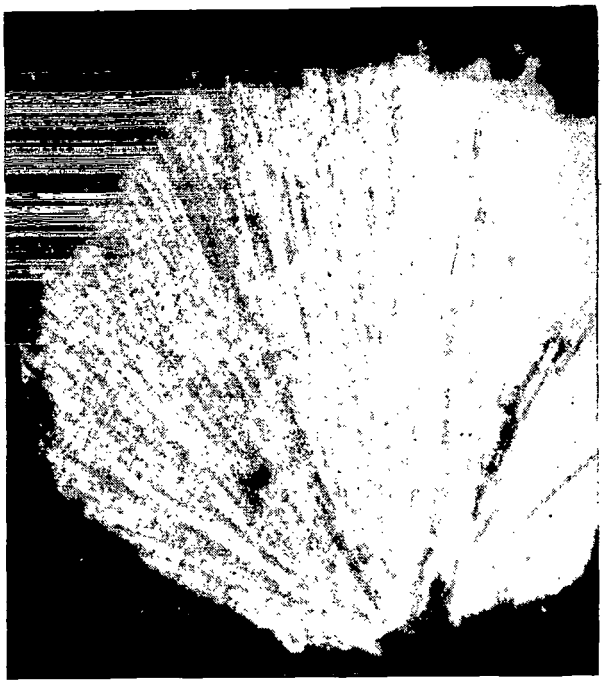


Figure 34. Pyroxene Chondrule with an Eccentrically Radial Structure. Kainsaz Meteorite. Magnification 280, without analyzer.

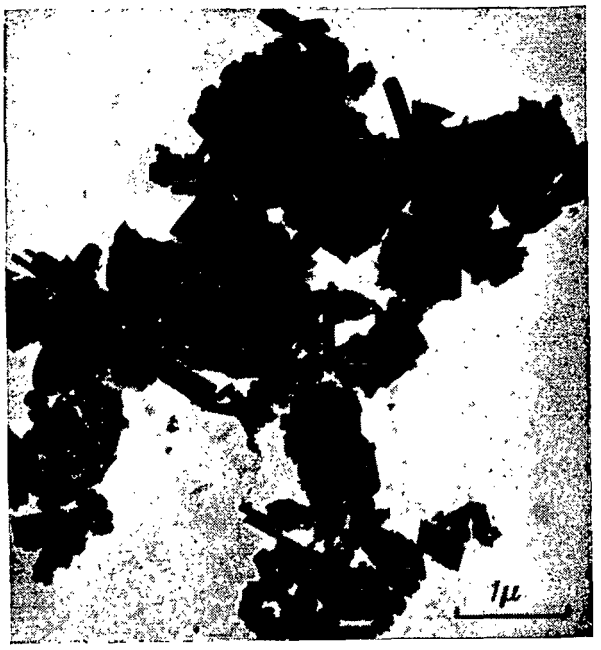


Figure 35. Electron Microscope Photograph of Dispersed Sample of Kainsaz Meteorite (platey small crystals and aggregates of pyroxene grains are visible).

An electron microscope study (Vdovykin, 1965a) revealed platey tiny crystals and aggregates of pyroxene grains (Figure 35).

A roentgenometric method analysis (see Figure 21, Table 18) reveals that this meteorite has an olivine-pyroxene composition. The X-ray photograph clearly also shows lines characteristic of ferronickel. /64

With respect to its chemical composition (M. I. D'yakanova, 1964). The Kainsaz meteorite differs from carbonaceous chondrites in having a high degree of reducibility of its matter; this meteorite contains much metallic Fe and Ni (ferronickel 20.01% with a nickel content in the metal 8.24%; Table 19). M. I. D'yakanova (1964) notes a low FeO and FeS content. The meteorite has a carbon content of 0.46% (Trofimov, 1950), 0.66% (Mueller, 1966), 0.81% (Chirvinskiyi, Ushakova, 1946). G. Mueller (1966) gives the sum of volatiles in the meteorite as 3.42%. The author of this book has shown that the carbonaceous matter in this meteorite contains graphite and that there are traces of bituminous compounds (Vdovykin, 1964d).

Table 18

Mineralogical Composition of the Kainsaz Carbon-Bearing Chondrite

Kainsaz		Olivine		Ferro-nickel		Kainsaz		Olivine		Ferro-nickel	
I	$\frac{d_a}{n}$	I	$\frac{d_a}{n}$	I	$\frac{d_a}{n}$	I	$\frac{d_a}{n}$	I	$\frac{d_a}{n}$	I	$\frac{d_a}{n}$
1	5,16	4	5,07	—	—	1	1,682	—	—	—	—
1	4,30	2	4,29	—	—	1	1,661	5	1,671	—	—
4	3,86	7	3,87	—	—	1	1,603	—	—	2	(1,578)
2	3,76	4	3,72	—	—	2	1,488	9	1,481	—	—
4	3,47	6	3,49	—	—	8	1,426	2	1,437	7	1,430
1	3,29	3	(3,33)	—	—	2	1,410	—	—	—	—
2	(3,16)	—	—	—	—	2	1,392	9	1,397	—	—
1	(3,08)	4	(3,06)	—	—	3	1,370	1	1,370	—	—
5	2,970	5	2,995	—	—	4	1,351	1	1,350	—	—
3	2,855	—	—	—	—	3	1,318	9	1,316	—	—
7	2,787	9	2,770	—	—	2	(1,287)	—	—	3	(1,289)
1	(2,711)	3	(2,715)	—	—	5	1,260	2	1,267	—	—
1	2,650	—	—	—	—	1	1,209	1	1,209	—	—
10	2,516	10	2,515	—	—	1	1,194	—	—	—	—
10	2,455	10	2,455	—	—	10	1,166	7	1,167	8	1,168
2	2,387	—	—	—	—	1	1,137	7	1,140	—	—
4	2,271	9	2,253	—	—	1	1,114	5	1,117	—	—
2	2,230	—	—	4	(2,333)	1	1,104	—	—	—	—
2	2,160	5	2,165	—	—	2	1,101	8	1,098	—	—
2	2,087	—	—	—	—	3	1,079	7	1,080	—	—
2	2,053	2	2,032			1	1,067	7	1,063	—	—
10	2,014	—	—	10	2,022	4	1,044	4	1,052	—	—
3	(1,922)	7	(1,931)	—	—	2	1,029	—	—	—	—
10	1,747	10	1,747	—	—	1	1,018	—	—	—	—
2	1,716	—	—	—	—	8	1,012	—	—	—	—

Table 19

Chemical Composition of Carbon-Bearing Kainsaz

Components	% by weight	Elements	% by weight
SiO_2	34,68	Fe	26,73
MgO	24,72	Si	16,18
FeO	7,37	Mg	14,83
Al_2O_3	3,72	S	1,51
CaO	2,50	Al	1,97
Na_2O*	0,52	Ni	1,65
K_2O*	0,05	Ca	1,78
P_2O_5	0,18	Na	0,38
Cr_2O_3	0,72	Cr	0,49
MnO	0,30	Mn	0,23
TiO_2	0,22	P	0,08
Fe	18,36	Ti	0,13
Ni	1,65	K	0,04
FeS	4,15	(O)	33,14
Total	99,14	Total	99,14

* A new determination of Na_2O and K_2O by M. I. D'yakonova (1967, personal communication).

CHAPTER THREE

MORPHOLOGY OF ORGANIC INCLUSIONS IN CARBONACEOUS CHONDRITE

The morphology of tiny particles of organic matter in the matrix of carbonaceous chondrites was investigated by the author under electron and ultraviolet microscopes. Below we also will describe the morphology of a bituminous substance separated out of meteorites using organic solvents and the morphology of the remaining part of the carbon phase of carbonaceous chondrites. /65

In order to obtain additional confirmation of the presence of bituminous compounds in meteorites the microbiological method was also used.

This chapter also gives the results of study of tiny particles in carbonaceous chondrites obtained by the author and other researchers. These microparticles (so-called "organized elements") until recently have been regarded by some researchers as remnants of cosmic forms of life on the basis of their morphological similiarity to fossil microorganisms.

Morphology of Organic Inclusions of Meteorites and Morphology of Fractions of Organic Matter Extracted from Meteorites

The matrix, filling the space separating the chondrules, has a lumpy structure in carbonaceous chondrites. On the basis of studies using a polarization microscope, L. G. Kvasha (1948; Kvasha, Wiik, 1964) and A. N. Zavaritskiy (1948a; Zavaritskiy, Kvasha, 1952) pointed out that this matrix is a mixture of carbonaceous matter and mineral grains. However since the interchondrule substance is opaque it was difficult to study it.

The author (Vdoykin, 1964d) used an electron microscope in studying the character of the interrelationship between the carbonaceous matter and very tiny mineral grains. A replica was prepared from a section of the Migei carbonaceous chondrite. A carefully polished section of the meteorite was processed from the surface with HF and HCL with great care for the destruction and removal of silicates from the surface of the section. After careful etching the section was rinsed in doubly distilled water. The carbonaceous particles remained at the surface of the section in the form of microprominances as a result of this processing.

On the collodion pseudoreplica, toned with chromium, obtained from this sample, it can be seen that the carbonaceous matter within the meteorite matrix has a nonuniform distribution and formed tiny lumps of irregular configuration (up to 2-3 μ in diameter or more). It is in a mixture with mineral grains of approximately the same size (Figure 36).

Within such tiny lumps of carbonaceous matter in the meteorite matrix there are extremely tiny inclusions of bituminous compound. These inclusions

were observed under the ultraviolet microscope in polished sections of a number of carbonaceous chondrites (Vdovykin, 1962b, 1963b, 1965). They had an oval /66 configuration and were luminescent in ultraviolet light (λ = 365 mμ) with a yellowish-greenish color. Similiar inclusions were noted in polished sections of bituminous rocks and in thin sections of meteorites. A special petrographic section of the Migei meteorite was prepared with a cement nonluminescent in ultraviolight light (Vdovykin, 1964d). Instead of the Canadian Balsam usually used in preparing petrographic sections and which is luminescent in untraviolet light, a nonluminescent solution (cement) of polybutylmetacriolate in xylene was prepared by a method proposed by T. E. Baranov and B. N. Bylalov (1961) for preparing nonluminescent sections of rock. When the section was examined under the untraviolet microscope it was possible to see a large number of very tiny inclusions of oval or rounded shape, luminescent with a yellowish-greenish color. They were distributed throughout the carbonaceous matter; individual sectors of the carbonaceous matter (up to 20 μ) seemingly were saturated by them. The luminescence of individual inclusions also causes a general luminescence of the enriched sectors. However, since the inclusions were distributed within the carbonaceous matter, the nature of the luminescence of these sectors was nonuniform. When drops of organic solvent (chloroform) were applied to the sections the area of the luminescent sectors was reduced somewhat and the bituminous compounds were partially dissolved.

Figure 36. Electron Microscope Photograph of a Section of the Migei Carbonaceous Chondrite. Collodion Pseudoreplica, Toned with Chromium (Tiny lumps of carbonaceous matter present in the interchondrule space, mixed with mineral grains, are visible).

The capacity for bituminous compounds to be dissolved in organic solvents was used for their extraction from meteorites and subsequent study. Other /67 researchers usually employed the method of extraction with organic solvents in a Soxhalet apparatus at the boiling point of the solvents for separating out these compounds. This makes it possible to attain a more complete extraction of bituminous compounds, but it is not impossible that new formations may appear, that is, some changes in the composition of the bituminous matter. Accordingly, in the studies made by the author he used the cold extraction method without heating of the solvents; this method was developed by V. N. Florovskay (1954, 1957) for extracting rocks. In this method the bituminous matter present in samples in a free state is extracted.[1] However, is is possible that new formations also appear when this method is employed? In particular, are bituminous compounds initially present in carbonaceous chondrites or are they the result of chemical transformation of the solvent itself during the extraction process as a result of its polymerization and condensation?

Figure 37. Tiny Crystals of Bituminous Matter in a Chloroform Extract of the Groznaya Carbonaceous Chondrite. Magnification 80.

G. P. Vdovykin and N. V. Pomortseva (1962) sought to answer these questions by using the microbiological method; they sowed cultures of hydrocarbon-oxidizing bacteria in a nutrient medium on samples of carbonaceous chondrites, on bituminous matter extracted from a carbonaceous chondrite, and also on ozocerite, and observed the development of this culture.

In this experiment they used samples of the Migei and Groznaya carbonaceous chondrites (small pieces from the innerparts of large samples), a chloroform extract from the Groznaya carbonaceous chondrite and a sample of ozocerite containing paraffin hydrocarbons (from the Dzvinyach deposit, Ukranian SSR). The culture of hydrocarbon-oxidizing microorganisms used for the sowing was extracted from the soil in a mineral medium with hexane added. These microorganisms, oxidizing, especially those paraffin hydrocarbons with 6-8 or more carbon atoms in the chain, are small, mobile bacilli with negative Gram reddening. The culture does not form a pigment. A mineral medium of the following composition was used for the sowing; $(NH_4)_2SO_4$ 1 g, Na_2HPO_4 0.1 g, KH_2PO_4 0.1 g, $MgSO_4$ 0.1 g, NaCl 0.1 g, tap water 1 liter; pH 7.0.

[1] Murphy and Nagy (1965, 1966) used solvents to extract bituminous matter from the Orgueil meteorite, also at room temperature.

The mineral medium was poured into soil beakers and the culture of microorganisms was introduced. The volume of the culture was 1% of the volume of the medium. The samples to be investigated were introduced into these beakers with the sowed material. A mineral medium with sowed material served as a control. The incubation temperature was 30°.

Already on the third day after sowing a good growth of microorganisms was observed on the medium containing the chloroform extract of bituminous matter from the Groznaya carbonaceous chondrite and on the medium with ozocerite. A strong bacterial "cloud" appeared on the medium with a chloroform extract, whereas a clouding and slight overgrowth of small pieces of ozocerite with bacteria occured on the medium with ozocerite. /68

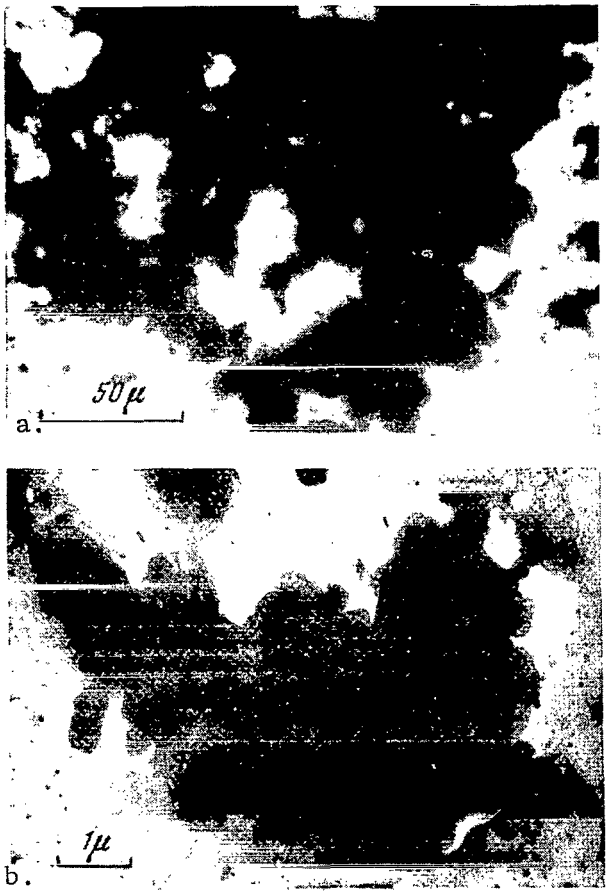


Figure 38. High-molecular Organic Matter Extracted from the Migei Carbonaceous Chondrite.
a) in polarization microscope, b) under electron microscope.

On the seventh day after the sowing on the medium with the chloroform extract from the Groznaya chondrite a bacterial film was formed on the surface which later settled. An overgrowth of hydrocarbon-oxidizing bacteria appeared on the samples of carbonaceous chondrites; it was stronger on the Migei sample. By the tenth day after sowing the overgrowth of bacteria on the Migei sample became thicker. However, small pieces of the Groznaya carbonaceous chondrite were covered by a rather thin bacterial growth until the end of the experiment.

In the control tests (mineral medium to which no organic substances had been added) the growth of microorganisms did not occur. This evidence that this hydrocarbon-oxidizing culture was unable to grow at the expense of compounds present in the air.

Thus, the organic matter in the Migei and Groznaya carbonaceous chondrites, the bituminous matter extracted with chloroform from the Groznaya chondrite and the ozocerite are used identically as the sole nutrient source by the culture of hydrocarbon-oxidizing microorganisms, supporting their growth. The results of this experiment revealed that the organic matter extracted from carbonaceous chondrites with organic solvents is present originally in meteorites and is not a new formation arising during the extraction process. /69

The bituminous matter extracted from meteorites with organic solvents is somewhat similar morphologically to the organic compounds extracted in much the same way from some rocks, such as from serpentinites of the Southern Urals (Kokpekty). Its color is usually light brown. A specific "aromatic" odor is characteristic. The consistency of this matter is viscous, but sometimes, such as in the case of the Migei, Murray and Groznaya meteorites, platey or acicular tiny crystals (up to 0.3 mm; Figure 37) are formed after the solvents dry.

The remaining part of the organic matter in carbonaceous chondrites, that is, the overwhelming fraction of the carbonaceous matter, does not dissolve in any solvents. It can be extracted from carbonaceous chondrites only by decomposition and removal of the mineral components after processing with HF and HCl. The part of the carbonaceous matter, high-polymer organic matter, extracted in this way is black in color and has a flakey structure. (Figure 38).

Thus, the organic compounds in carbonaceous chondrites are found primarily in the matrix, the interchondrule space. There they seemingly envelope very tiny mineral components and also form independent lumpy sectors. Within these sectors, which are black in color and consist for the most part of high-polymer organic matter there is a thin distribution of bituminous inclusions of rounded configurations, luminescent in ultraviolet light with a yellowish-greenish color. The bituminous compounds, as demonstrated by the experiment with the sowing of microorganisms, are natural in meteorites and are not the result of chemical transformation of the solvents used in extracting them from the meteorites. This bituminous matter, extracted from meteorites with organic solvents, frequently forms tiny crystals. The remaining part of the carbonaceous matter extracted from carbonaceous chondrites after being freed from the mineral components is black in color and has a flakey structure.

"Organized Elements" in Carbonaceous Chondrites

It is well known that under terrestrial conditions during the course of transformation of a sediment into sedimentary rock the organic remains varied in it are subjected to fossilization. Fossilized remains are bound in sedimentary rocks from the Paleozoic time to the recent era. They have even been found in rocks of Precambrian age which were formed 0.6-3 billions years ago (Timofeyev, 1959; Vologein, 1962, 1963; Barghoorn, Schopf, 1965, 1966, and others).

During recent years there has been discussion of the following problem: do meteorites also contain fossilized remains? In 1961 Claus and Nagy (1961), on the basis of morphological similarity described in carbonaceous chondrites microinclusions which were luminescent in ultraviolet light as being remnants of cosmic microorganisms. They, and later other authors as well, called these microinclusions "organized elements", "organic microstructures", "fossilized microorganisms", "lifelike forms", etc. Without question, the presence of /71
remnants of extraterrestrial life forms in meteorites would be a fact of extreme importance in many respects. Accordingly, these particles were studied in detail and their nature was discussed until recently. Some researchers felt that these forms represented an extraterrestrial biological organization.

On this basis they postulated that the parent body of the meteorites was of a planetary size and that conditions existed on it which were favorable for the development of life. Other researchers demonstrated very clearly that the "organized elements" in carbonaceous chondrites are mineral grains natural in meteorites, but also the result of contamination of the meteorite after their falling onto the earth.

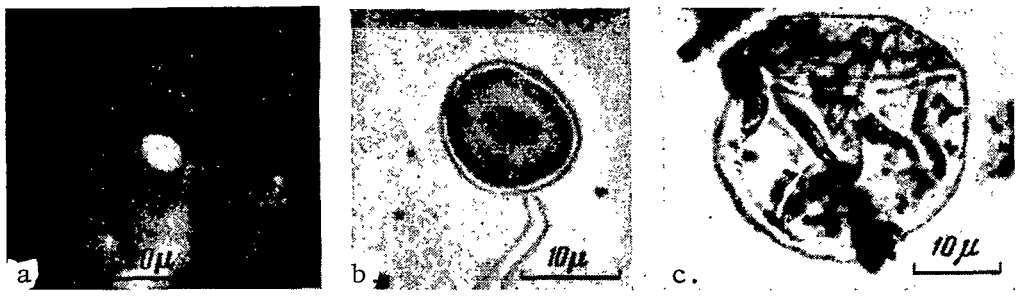


Figure 39. "Organized Elements" in the Orgueil Carbonaceous Chondrite.
a - particles visible under an ultraviolet microscope ("organized element" type I); b - particles visible under a polarization microscope ("organized element", type II);(Claus, Nagy, 1961); c - "organized element" of complex morphology (Nagy, 1966b).

Now we will consider the results of the morphological study of the "organized elements" in carbonaceous chondrites, taking into account these two opposite points of view concerning their nature.

"Organized Elements" as Remnants of Extraterrestrial Life Forms

Claus and Nagy noted microinclusions morphologically similar to algae in pulverized samples of the Ivuna and Orgueil meteorites. They mentioned five types of such "organized elements", having rounded, cylindrical or hexagonal configurations and dimensions up to 4-30 μ. One of these types (hexagonal form) had no similarity, as noted by Claus and Nagy, to any known terrestrial biogenesis form. The number of these particles was as great as 1,700 per mg of meteorite sample. The authors did not find such inclusions in the ordinary Holbrook and Bruderheim chondrites. In addition, a small number of bacteria was observed in preparations made from carbonaceous and ordinary chondrites. In polarized light the tiny particles did not have birefringence, but in ultraviolet light they were luminescent with a yellowish color (Figure 39). In the case of staining which is usually employed for the purpose of characterizing biogenesis compounds, color reactions were obtained with hematoxylin, eosin, ninhydrin, and others. On the basis of morphological characteristics, the authors concluded that forms of extraterrestrial life are present in the meteorites which they had studied.

However, the results of these observations were subjected to criticism. Fitch, Schwarcz and Anders (1962a, b) checked preparations made from the Ivuna and Orgueil meteorite and confirmed that rounded particles were present. The authors felt that if "organized elements" contain a considerable quantity of organic matter in addition to silicates and other mineral components, the density of these particles should be some mean between the density of the minerals and the density of the organic compound. The authors concentrated the "organized elements" by the centrifuging of pulverized meteorites matter in a bromoform-acetone mixture ($\rho = 2.31$). Almost all the "organized elements" were found to be in the heavy fraction. After the heavy fraction was again fractionated in bromoform ($\rho = 2.88$) and methylene iodide ($\rho = 3.33$) the greater part of the "organized elements" were again found to be in the heavy fraction.

Such a high density for microorganisms seemed to be unlikely. In addition, a roentgenometric study made by the authors revealed that the fraction having $\rho > 3.3$ consists for the most part of troilite and magnetite. They concluded that Claus and Nagy could mistake these minerals for "organized elements". In actuality, in addition to spherical particles, Fitch and other researchers observed thin platelets. With approach of a magnet, these particles, much like the "organized elements", shifted. Moreover, in reflected light the tiny /72 particles had a characteristic bronze color or troilite. Fitch, Schwarcz and Anders also checked the light fraction for the presence of "organized elements". They did this by separating the pulverized matter of meteorites in chloroform during centrifuging. It was found that the light fraction as well contains a large number of spherical particles which are luminescent in ultraviolet light (Figure 40,a). However, the authors were inclined to interpret them as being inclusions of hydrocarbons or sulfur, rather than "organized elements".

Briggs and Kitto (1962) observed similar particles in prepared sampled of the Mokoia carbonaceous chondrite. They checked the meteorite for the presence of terrestrial microorganisms. They did this by placing meteorite samples in nutrient media where the samples were pulverized. No growth of microorganisms was observed, although the authors used the nutrient media for the growth of cultures in a wide variety, both aerobic and anaerobic bacteria and fungi. For checking the conclusions drawn by Claus and Nagy, Briggs and Kitto processed pulverized matter from the Mokoia meteorite with coloring substances, safarinine, fuchsin, Gram's stain, and others. Some particles were stained and they were stained uniformly. These particles had a rounded configuration (Figure 40b), but sometimes they were very thin (Figure 40c). In size they varied from 1 to 25 μ. Briggs (1962b) postulated that these are probably organic compounds adsorbed on mineral grains and also found in the form of individual fine inclusions.

Later, Nagy, Claus and Hennessy (1962) made a number of additional studies. They subjected pulverized samples of the Ivuna and Orgueil meteorites to exposure to concentrated HF during fifteen-minute heating. "Organized elements" were found thereafter in the meteorite samples. After separating the pulverized matter of the Orgueil meteorite by density, the authors found "organized elements" in the fractions $\rho < 1.50$ and $\rho = 2.3$-2.4. However, they were not able to achieve a clear separation of the "organized elements" in the mineral grains. Nagy, et al, attribute this to the fact that the "organized elements"

are either durably bonded to the mineral particles, or that probably some of the "organized elements" were mineralized to some extent. Organic solvents in some cases have an effect on the "organized elements", but do not completely dissolve them. The tiny particles seemingly become shriveled under the influence of these solvents. These researchers found similar particles in the Aláis and Tonk carbonaceous chondrites. Among the "organized elements" they identified *Diadophore berzelii* and *Apolinarisphaera meteoriticola* (Claus, Nagy, 1962a, b).

These observations were confirmed in part by Staplin (1962a,b; Staplin et al, 1962). He observed that the Orgueil meteorite contained fossilized microparticles that were not destroyed by hydrofluoric acid. In a study of two grams of a meteorite sample Staplin found that it contained three types of biogenesis inclusions, present-day microorganisms, individual inclusions of fossilized microorganisms of Cretaceous age and unknown fossilized microorganisms. Among the latter he found several inclusions 10-100 μ in diameter having a similarity to unicellular arganisms, some of them contained tinv mineral grains. Among the microinclusions which he found, Staplin identified new forms: *Caelestites sexangulatus* and *Clausisphaera fisa*.

In the course of a microphytological study of five grams of a sample of the Migei meteorite, B. V. Timofeyev (1962) also found sporelike forms which withstood rather high temperatures. He observed more than 20 different forms of cell casings. They measured 10-60 μ in diameter and had a yellowish-grey /73
and dark grey color; the surface was smooth, less frequently it was slightly bumpy, occasionally with stomata. Some of these forms were similiar to the forms characteristic of spores of the Precambrian rocks of the Baltic shield. B. V. Timofeyev (1963) defined a number of new forms among them: *Protoleiosphaeridium migeum, P. tichovi, Prototrachysphaeridium staplini*, and others.

The small piece of the Orgueil meteorite sample which had been studied by Claus and Nagy was obtained by Palik (1962), who checked it for the presence of "organized elements" under sterile conditions. Palik observed fibrous formations; he wrote that they apparently were of a biogenesis nature, and some of them would be natural to the meteorite. However, P. Morrison (1962) noted that flakes of dendritic and platey form can be mistaken for biogenesis inclusions.

At a conference for the discussion of the nature of "organized elements", held during the period 30 April-1 May 1962 in New York (Life-like..., 1963), Claus, et al (Claus, Nagy, Europa, 1963) reported on the results of their further studies of the Ivuna, Orgueil, Murray, and other meteorites. They also studied the soil from the site of falling of the Orgueil meteorite, checked the reaction of staining with 19 stains, and concluded that the "organized elements" in meteorites have no similarity to terrestrial forms and are characteristic only of type-I carbonaceous chondrite (Nagy, Claus, 1962, 1964a, b).

The "organized elements" in carbonaceous chondrites have also been noted by other researchers (Panel Discussion, 1963). The literature describes more

than 30 different types of "organized elements" in carbonaceous chondrites. Maminkunian and Briggs (1963a, b) compiled a catalog of such microinclusions which they observed in preparations from carbonaceous chondrites prepared under sterile conditions.

In 1964 Tasch (1964) made a study of the "organized elements" in preparations of the Ivuna and Orgeuil meteorites. Among the microinclusions he found particles which are found only in meteorites and are not found in control preparations. Some of them were similar to forms described by other researchers. Tasch concluded that these forms are either natural to meteorites or entered the meteorites under terrestrial conditions but could not be the result of contamination while the meteorites were being prepared for study.

In order to clarify the extent of occurrence of "organized elements" as a function of their size, Claus and Suba-C (1964) studied the Orgueil meteorite. They demonstrated that "organized elements" in most cases are 3, 4.5 and 9 μ in diameter; this corresponds to the species *Chlorococcites minima*, *Apollinarispaera meteoriticola* and *Apollinarisphaera ellipsoidae*. The authors believe that if the "organized elements" were mineral grains, such a pattern of occurrence would not be observed.

In 1965 VanLandingham (1965a, b) studied the Aláis and Orgueil meteorite. In some preparations he noted modern forms of spores and pollen, as well as about 30 forms of "organized elements", apparently natural in meteorites. They were platelike, disk-shaped, spherical and elongated and measured 10-31 μ .

Botan (1965) undertook a check of the "organized elements" in the Orgueil meteorite by the fluorescent microscopy method for the purpose of finding a possible correlation between the number of "organized elements" and their content of tyrosine, which is determined from the fluorescence of complex compounds forming with the introduction of 1-nitroso-2-naphthol. In preparations prepared from small pieces of meteorite measuring 0.5-5 mm Botan found cell-like microstructures fluorescent after processing with a yellow-green color. /74
Their number was 33, which is in order of magnitude lower than in comparison preparations (smears from peat, as well as bacterial, fungal, yeast and algal smears). In individual cases a picture was observed which is similar to mitotic division. However, the tyrosine content in these cells was less than in laboratory culture; the author therefore concludes that the "organized elements" did not arise from contamination, but possibly have a biogenesis nature.

Various types of "organized elements" were recently found in the ordinary Saritov chondrite by A. S. Lopukhin.

In review devoted to these "organized elements" in carbonaceous chondrites, Staplin (1965a, b) and Nagy, et al (Nagy, Meinschein, Hennessy, 1965) assume, as before, that these particles possibly are natural in meteorites.

The importance of the "organized elements" for clarifying the possible existence of extraterrestrial life has been discussed in the

literature[1]. There have also been discussions of the problem of "export" and "import" of life (Imshenetskiy, 1963, 1964, 1966). However, it is difficult to explain the origin of carbonaceous chondrites containing these "organized elements". Some researchers have calculated that carbonaceous chondrites are the remnants of a cosmic body having planetary dimensions and that conditions were favorable for the development on this parent body (Timofeyev, 1962; Staplin, 1962a). Nevertheless, the peculiarities of composition and structure of meteorites do not confirm the hypothesis of a planetary size of the parent body from which meteorites originated (Urey, 1966a). Urey (1962b, c, 1965a, b, 1966a) refutes the possibility of ejection of carbonaceous chondrites containing "organized elements" from Mars. As a more likely possibility he advances the hypothesis which he formulated earlier (Urey, 1959b) that carbonaceous chondrites and other meteorites originated as a result of their ejection from the moon. However, conditions favoring the development of life could not have existed on the moon over a long period. In order to explain the presence of "organized elements" in carbonaceous chondrites Urey postulated that during an early period in the earth's history, when life had appeared on it, a hypothetical mechanism existed in which water and the biogenetic substances within it were ejected from the earth to the moon where these substances were preserved. Later, as a result of an explosion, the "organized elements" present in carbonaceous chondrites were again returned to the earth.

It is difficult to visualize this series of hypothetical events, "whose probability becomes real with their increase" (Bernal, 1962. Similar interpretations follow from the possibility that the "organized elements" may be biogenetic inclusions natural in carbonaceous chondrites. However, many studies of "organized elements" show that these tiny particles in carbonaceous chondrites are not inclusions of biogenetic origins, but in all probability are mineral grains and the result of contamination under terrestrial conditions.

"Organized Elements" as Mineral Grains and the Result of Contamination

Soon after publication of the article by Claus and Nagy (1961) the identification of "organized elements" as remnants of extraterrestrial life-form drew objections from many researchers. In particular, Pearson (1962) noted /75 that many of the forms of tiny particles observed in carbonaceous chondrites are similar to the present-day pollen of anemophylic plants (*Corylus*, *Alnus* and others). Other forms, as pointed out by Gregory (1962) are similar to *Asperigillus glancus*, the pollen of *Uretica*, etc.

A detailed study of "organized elements" was made by Anders et al (Anders, Fitch, 1962; Fitch, Schwartz, Anders, 1962a, b; Fitch, Anders, 1963a, b). They made a further separation of "organized elements" by density, checked their fluorescence, stained them with dyes and exposed them to acids. Their

[1] See articles; Berland, 1961; Bernal, 1961a, 1962; Deflandre, 1962; Gaskell, 1962; M.R., 1962; Nagy, Claus, Meinschein, Hennessy, 1962; Possible Presence..., 1962; Price, 1962; Sagan, 1962; Welsh, 1962; Briggs, 1963a; Hey, 1963; Mamikunian, Briggs, 1963c; Mason, 1963c; Pokrzywnicki, 1963a, b; G. Cohen, 1964; Hutchinson et al., 1964; Elias, 1965; Vagera, 1966 and others.

studies revealed that the tiny particles of simpler morphology are mineral grains. However, in ultraviolet light there was a luminescence of particles morphologically dissimilar to the "organized elements"; these particles were of irregular configuration and measured more than 2 μ. In this case staining also was nonspecific, since the authors obtained a similar effect in control preparations of kimberlite.

Fitch and Anders (1963b) made a particularly detailed study of the "organized elements" of hexagonal form in the Orgueil meteorite; as noted by Claus and Nagy (1961), these elements were not known under terrestrial conditions. In actuality, according to the results of the study by Fitch and Anders, these inclusions are the pollen of present-day ragweed; the pollen of ragweed has the same morphology and size and a similar appearance when stained.

Some carbonaceous chondrites, including the Orgueil meteorite, were studied by Mueller (1962a, b). He described three types of mineral grains which other researchers had regarded as "organized elements": hexagonal platelets of troilet, magnetite rains and pseudomorphs of limonite, rounded particles of glass, grains of olivine with a rounded configuration--microchondrules. Mueller, in particular, demonstrated that the size of microchondrules in different meteorites decrease with a transition from carbonaceous chondrites with a lesser carbon content to high-carbon meteorites. The histogram of the distribution of microchondrules has a function of their size, constructed by Mueller (1964b, 1965), was similar to the histogram of the distribution of "organized elements" constructed by Claus and Suba-C (1964).

In a morphological study of the "organized elements" in carbonaceous chondrites, the author (Vdovykin, 1964e) used a sample from the Migei meteorite weighing 0.4 grams, after first freeing it from any possible contaminants. The meteorite sample was pulverized in an agate mortar and separated by centrifuging into a number of fractions in fluids with various densities: in chloroform, in a bromoform-acetone mixture (1:1), and in bromoform. The centrifuging was continued for 15 minutes at a rate of 3,000 rpm.

After centrifuging in chloroform the fraction of the sample with a density $\rho < 1.5$ contained not only carbonaceous matter, but also olivine microchondrules of a regular rounded form measuring up to 64 μ, very similar to the earlier described "organized elements", and also resembling aggregates of chlorite-serpentine and olivine particles (Figure 41,a). Associated with the carbonaceous matter in this fraction was an insignificant quantity of bituminous compounds which were luminescent with a yellowish-greenish color in ultraviolet light. The fraction having $\rho > 1.5$ was dessicated and centrifuged in a bromoform-acetone mixture (1:1). The light part of this fraction ($1.5 < \rho < 2.31$; Figure 41,b) also consisted of olivine grains, sometimes with a rounded configuration, and tiny lumps of carbonaceous matter with luminescent bituminous compounds.

The heavy part of the fraction, with a density $\rho > 2.31$ after dessication, /77
was centrifuged in bromoform ($\rho = 2.88$). In the fraction $2.31 < \rho < 2.88$

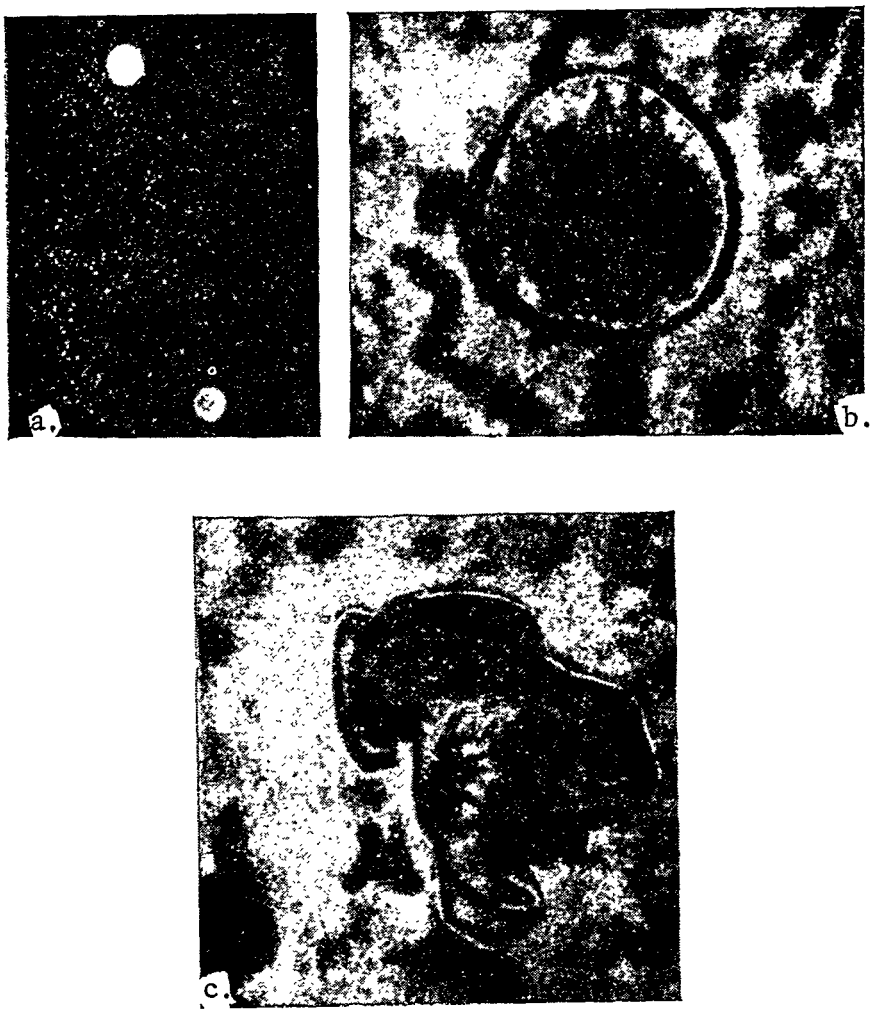


Figure 40. Particles in Carbonaceous Chondrites.
a - Particles luminescent in ultraviolet light (preparation from the Orgueil meteorite; $\rho \leq 1.5$; Fitch, et al, 1962b), Magnification 600; b - Particle of rounded configuration from the Mokoia meteorite, Magnification 6000; c - Particle of irregular configuration, Mokoia meteorite, Magnification 6000 (Briggs, Kitto, 1962).

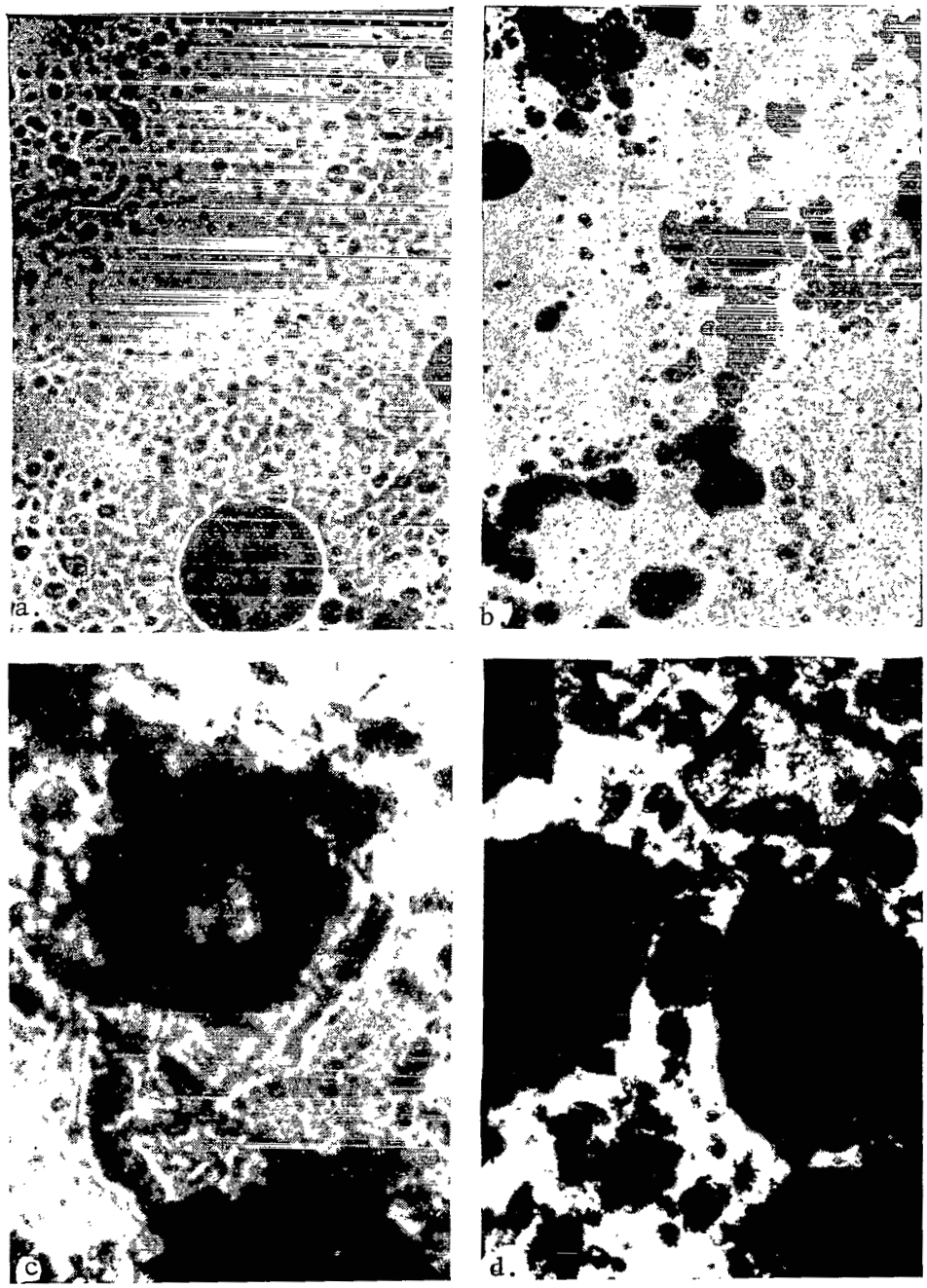


Figure 41. Pulverized Preparations from the Migei Carbonaceous Chondrite (Vdoykin, 1964e). Magnification 280.
a - $\rho < 1.5$; silicate grains and microchondrules; b - fraction $1.5 < \rho < 2.31$; olivine grains with adjoining particles of chlorite-serpentine minerals in carbonaceous matter; c - fraction $2.31 < \rho < 2.88$; olivine microchondrule with envelope of carbonaceous matter; d - fraction $< \rho <$ grains of magnetite and sulfides; transluscent tiny olivine crystals can be seen at the top.

(Figure 41c) there were a large number of loivine microchondrules, surrounded by an envelope of carbonaceous matter1. In the fraction $\rho > 2.88$, making up about 30% of the initial weight of the meteorite sample, the number of carbonaceous inclusions was much less than in the earlier fractions. This fraction contains iron pompounds (magnetite, troilite, and others) and silicate grains (Figure 41,d). In most cases there were very few luminescent inclusions in the latter two fractions.

After staining with eosin[1] in all four fractions it was possible to note a number of particles which stained a rose color. Most of the particles of this type were concentrated in the fractions $1.5 < \rho < 2.88$. The staining for microchondrules surrounded by chlorite-serpentine, as well as for aggregates of grains consisting of carbonaceous matter and chlorite-serpentine minerals, is particularly characteristic. Assuming that in this case it is the chlorite-serpentine which is stained, the author similarly prepared preparations of serpentinite (from the Southern Urals) and kimberlite from the Siberian Chomur pipe). In both cases the chlorite-serpentine was stained by the eosin in a rose color. A similar staining for kimberlite was noted by Fitch and Anders (1963a). Since the chlorite-serpentine in the meteorite surrounds olivine grains, and is also found in a mixture with carbonaceous matter, these aggregates are stained heterogeneously when stained by organic dyes. This apparently explains the nonuniformity of staining which has been noted by other researchers (Claus, Nagy, 1961; Briggs, 1962b) when describing "organized elements".

The tiny particles luminescent in ultraviolet light are also concentrated primarily in the fractions $1.5 < \rho < 2.88$. For the most part these are bituminous components, and since they are distributed in carbonaceous matter, when drops of chloroform are applied, as already noted, there is some decrease in the size of the luminescent inclusions, as was also observed for "organized elements" by Nagy, et al (Nagy, Claus, Hennessy, 1962).

Possible contaminations of the terrestrial type have approximately the same density. Therefore, in all probability in the studies made by other authors tiny particles of contamination also entered the fraction with the same density. Possible sources of meteorite contamination would be spores[2], pollen and even paper fibers, which have been described by other authors as "organized elements" (Staplin, 1962a; Mamikunian, Briggs, 1963a), as well as microorganisms whose growth in a nutrient medium has sometimes been noted in meteorites[3].

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[1] A tetrabrom deriviation of fluorescien; it is related to the rhodamine dyes.
[2] It is notable that Staplin (1962a), who found Cretaceous forms in the Orgueil meteorite, simultaneously with his study of the meteorite investigated rocks of Cretaceous age; B. V. Timofeyev, a specialist on Precambrian forms of the Baltic shield, found Precambrian forms in the Migei meteorite (Timofeyev, 1963).
[3] Terrestrial microorganisms in meteorites have been noted by Roy (1935), Lipman (1936), and Sisler (1961). Recently Oró and Tornabene (1965) made a microbiological check of three carbonaceous chondrites, Orgueil, Murray and Mokoia. In the Orgueril meteorite, after incubation of a period of two weeks at room temperatures, they found no microorganisms. (Continued on next page)

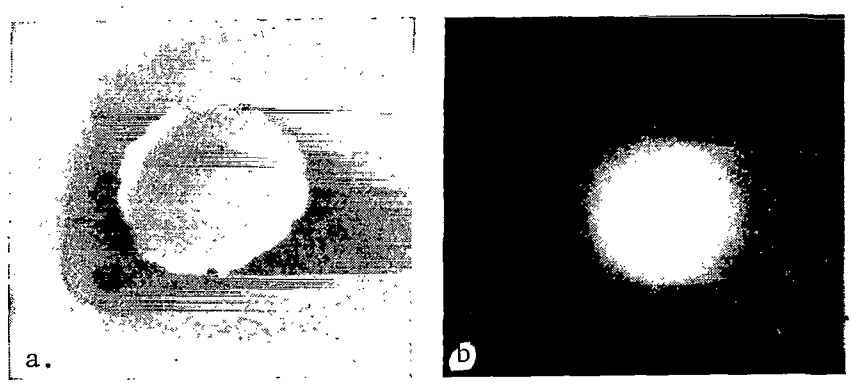

Figure 42. Fungus in a Preparation of a Fraction of the Cold Bokkeveld Carbonaceous Chondrite.
a - electron microscope image; b - diffraction spectrum. Diameter of the selector diaphragm 1 μ. Obtained using a 400-kV electron microscope electronograph.

Microorganisms were not observed in the course of our research. However, in a special moistened preparation of a fraction from the Cold Bokkeveld, which was left for one day in the open, individual fungal forms appeared, after having entered the preparation from the air. Figure 42 shows such a fungus with three inclusions, two of which are situated along the edges, and one in the center. Its configuration is oval, while the edges are irregular. The fungus measures ~2 μ. This same figure shows a diffraction spectrum which is evident of an amorphous character of this electron-microscope object.

With the partial decomposition of fractions of the Migei carbonaceous chondrite as a result of 15-minute processing with HF-acid, the studies of the author of this book revealed that the microchondrules are not completely destroyed; this was noted by Claus and Nagy (1961) also for "organized elements". However, with the decomposition of the mineral components in the meteorite the

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This meteorite had been preserved prior to the study in a sealed glass ampule. In small pieces of two other meteorites they found a relatively low content of microorganisms: up to 6,000 and 1,800 cells per gram of meteorite respectively. In the Murray meteorite microorganisms were represented by *Bacillus cereus* and *B. badius*, and in the Mokoia meteorite--*Staphylococcus epidermidis*. After two months the experiments were repeated with similar results. The literature also contains reports of the presence of microorganisms in the iron Sikhote-Alin meteorite; as was later found, this meteorite had been contaminated under terrestrial conditions (Martynov, 1962; Inshenetskiy, et al, 1963; Yavnel', 1963a). In such studies one must take into account that the contamination of meteorites can occur not only during their storage but even at the time at which they fall to earth, during their impact against it (Mueller, 1964a).

microchondrules disappeared, although the composition of the organic matter present in the meteorite does not significantly change at this time. If the "organized elements" were not mineral in their composition, but instead were organic, they also probably would be preserved after the destruction of the mineral components. The carbonaceous matter remaining after decomposition also failed to reveal particles that could be stained by eosin.

The author (Vdovykin, 1964e) also observed similar rounded silicate particles morphologically similar to "organized elements" in an electron microscope study of pulverized material from a number of carbonaceous chondrites. These ultramicrochondrules were more than 2 μ in diameter.

Thus, it can be concluded that the rounded particles observed in a number of carbonaceous chondrites in all probability are not biogenetic forms, "organized elements", but primarily siliceous micro- and ultramicrochondrules[1], in large parts surrounded by an envelop of carbonaceous matter and frequently /79 by material consisting of chlorite-serpentine minerals[2], as well as by tiny particles of troilite, magnetite, and in some cases limonite. Some of the particles, particularly the chlorite-serpentine minerals are stained by organic dyes. In ultraviolet light there is luminescence for the most part from the bituminous component, genetically related to the remaining part of the carbonaceous matter in the meteorite.

This interpretation of the nature of the rounded microparticles in carbonaceous chondrites agrees with many investigations made recently by other authors, including studies which were made by Nagy. In particular, Nagy, Fredriksson, Urey, et al (1963) used the X-ray spectral microanalysis method to show that the microparticles morphologically similar to "organized elements" (Figure 43) contain (in percent): Fe up to 40, Mg 6, Si 10, Ca 20, Ni 2, S up to 10, Cl 3 and for the most part are grains of limonite and hydrous silicates of the chlorite-serpentine type. Some of the particles do not contain elements with $Z > 11$; these are tiny lumps of carbonaceous matter. These researchers, noting the presence of chlorine and nickel in the limonite particles, felt that this confirms the extraterrestrial origin of the rounded particles, possibly forming as a result of mineralization of extraterrestrial biogenetic remains. However, Anders, et al (1964) noted that the presence of chlorine in this case is not unusual because limonite of oxidized iron meteorites, according to data published by Buddhue (1957), also contains chlorine and nickel. Later, Nagy, /80 et al (Nagy, Fredriksson, Kudynowski, Carlson, 1963) also confirmed the fact that the luminescence of the "organized elements" is caused by the presence of organic compounds in the rounded microparticles. However, as before they admitted the possibility that some part of these microparticles would be "organized elements", if not in all carbonaceous chondrites, at least this was true for the Orgueil meteorite.

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[1] During the last century, Hahn (1880, 1882) erroneously regarded meteorite chondrules as fossilized organisms

[2] L. G. Kvasha (1963; Kvasha, Wiik, 1964) notes that the aggregates of chlorite-serpentine minerals replacing the chondrules in carbonaceous chondrites are very similar to the "organized elements"; morphologically they are similar to organic remains.

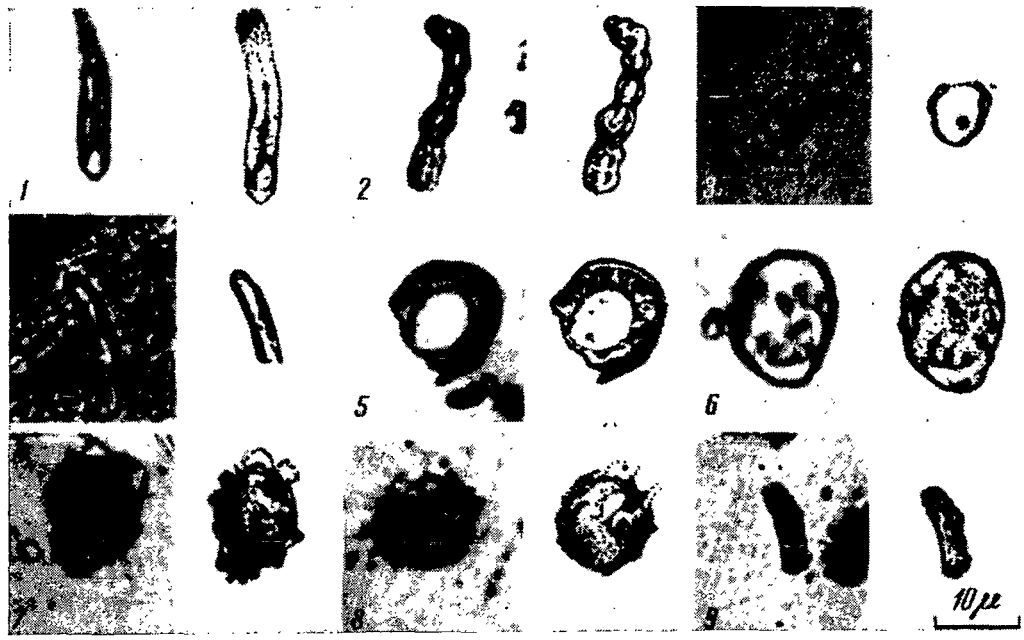

Figure 43. Photograph and Drawings of Particles in the Orgueil Meteorite (studied by X-ray spectral microanalysis) (Nagy, Fredriksson, Urey et al, 1963).
Particles 1-5 and 9 contain iron in chlorine; particle 6 contains iron in nickel; particle 7 and 8 contain magnisium-ferruginous silicates.

The sample of the Orgueil meteorite which was studied by Nagy, was again examined by Anders, et al (Anders, DuFresne et al., 1964). They confirmed their earlier conclusion that this type of "organized elements", which, as Nagy wrote, had no similarities to terrestrial forms, is in actuality spores of present-day ragweed. In this meteorite sample Anders and other researchers found not only a large number of spores, but even a tiny branchlet of this composite plant. They postulate that this meteorite sample was possibly contaminated during the last century.

Now it can be considered quite firmly established (Fitch, Anders, 1965; Orcel, Alpern, 1966, and others) that the so-called "organized elements" in carbonaceous chondrites morphologically similar to the remnants of microorganisms, in actuality are formations of a different type. For the most part they are particles natural in meteorites, but there are also particles appearing as inclusions as a result of contamination.

1. The particles natural to meteorites have the following nature:

a) Ultramicrochondrules and microchondrules consisting for the most part of olivine, glass, chlorite-serpentine minerals, frequently enveloped by carbonaceous matter;

b) Tiny lumps of carbonaceous matter in which bituminous compounds are distributed;

c) Grains of troilite, magnetite, limonite, and others.

2. Contamination inclusions of a terrestrial type, such as microorganisms, spores, mechanical contaminations, including even paper fibers, etc. The sources of this contamination have a wide range;

a) Contamination during the falling and storage of meteorites, that is, from the air, from the soil, and from the rock on which the meteorite fell, etc.;

b) Contamination which could occur during operations performed with the meteorite by other individuals at an earlier date;

c) Involuntary contamination of the samples during the course of research.

Thus, the results of a morphological study reveal that the organic compounds present, primarily in the interchondrule space in meteorites, in their matrix, with which carbonaceous chondrites are especially enriched, have nothing in common with "organized elements". The latter in all probability are not "exterterrestrial life-forms" but are instead various types of formations, for the most part mineral grains in such meteorites.

CHAPTER FOUR

RESULTS OF STUDY OF CHEMICAL COMPOSITION OF ORGANIC COMPOUNDS IN CARBONACEOUS CHONDRITES

Extraction, Luminescent-Bituminological Analysis and Separation of High-Molecular Fraction of Organic Matter

The author of this book separated bituminous compounds from meteorites by extraction with organic solvents without heating, using the method proposed by V. N. Florovskaya (1954, 1957). /81

The samples for extraction were taken from the inner part of the meteorites. In those cases when the initial samples had a very low weight they were mechanically cleansed of possible contaminates, after which the small pieces were washed in cloroform and then ground in an agate mortar. The weighed samples were placed in clean test tubes with a ground glass stopper and a certain quantity of solvent was added. The extraction was done successively using the following organic solvents: petroleum ether, benzene, ethanol, alcohol-benzene (1:1), carbon tetrachloride and chloroform; isobutyl alcohol was also used for some weighed samples of the Migei and Groznaya meteorites. Using each solvent extraction continued for ten days at room temperature; during this time the test tubes were periodically shaken.

The solvents used in the extraction process were pure. They did not yield luminescence in ultraviolet light. All other fluids and containers used were also clean; prior to use they were carefully cleansed.

Under ultraviolet irradiation (λ = 365 mμ) all the extracts in the organic solvents were clearly luminescent with a bluish or slightly cinnamon or brownish color. The author determined the quantitative content of bituminous components in them by the standard method using the procedures formulated by V. N. Florovskaya (1954).

In all six of the investigated samples of carbonaceous chondrites it was possible to detect bituminous compounds. Their total content of extracted organic matter ranged from 0.097% (Groznaya meteorite) to approximately 0.5% (Orgueil meteorite, chloroform extract[1]) (Table 20). In the Kainsaz chondrite, which contains carbon (its initial weighed sample was 1.6880 g), the content of bituminous compounds was 0.007%. It can be noted in passing that results which are in general good agreement are obtained when the luminescent-bituminological method is used. For example, in the extraction of the Migei carbonaceous chondrite the quantitative content of bituminous components was 0.156%.

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[1] In another extraction method, extraction in a Soxhlet apparatus with a series of solvents, Briggs (1963c) determined the following content of organic compounds in two carbonaceous chondrites: in the Haripura meteorite 16.3 mg/g, in the Mokoia meteorite 3.6 mg/g.

(Vdovykin, 1964d). Earlier the author had found this meteorite to contain 0.157% of bituminous components (Vdovykin, 1962b) in an equal weight of another sample of the Migei meteorite under similar extraction conditions. /82

Table 20

Contents of Bituminous Matter Extracted from Six Carbonaceous Chondrites by Organic Solvents

Types of Carbonaceous Chondrites	Meteorite	Place of Falling	Date of Falling	Investigated meteorite sample*	Total weight of sample used in this study, g	Weight of extracted sample, grams	Content of Bituminous Matter, % by weight
I	Orgueil	France	14 V 1864	2478	0.115	0.1152	~0.5
II	Migei	Ukranian SSR	18.VI 1889	1856	6.905	0.2374	0.156
	Staroye Boriskino	Russia	20.IV 1930	192	1.327	0.1988	0.276
	Cold Bokkeveld	S.Africa	13.X 1838	26	1.362	0.6255	0.156
	Murray	Kentucky, U.S.A.	20.IX 1950	2306	0.260	0.2600	0.437
III	Groznaya	Russia	28.VI 1861	73	2.189	0.1650	0.097

*The number of the sample is from the catalog compiled from the meteorite collection of the Committee on Meteorites Academy of Sciences, USSR.

As a comparison, organic compounds were extracted from two types of chondrites. The initial weight of the sample for these meteorites varied in the following range (weight in grams): unmodified chondrites, Krymka (0.5304), Manych (1.7758), Liksna (0.2635), Sarapov (2.7843), crystal in chondrites, Chuvashskiye Kissy (1.0456), Sevryukovo (0.2425, Farmington (0.0275), Ghubara (0.2065), Kul'p (0.7965), Mezö-Madaras (0.0570), Gilgoin (0.0833), Kunashak, light variety (1.7275), dark variety (0.3950), Pilastvere enstatic chondrite (0.4030). Bituminous compounds were found in the following chondrites (in %): Krymka 0.010, Saratov 0.002 (CCl_4 extract), Sevryukovo 0.003, Ghubara 0.035, Kul'p 0.081[1]. In a repeated similar extraction with six solvents using a different sample of the Kul'p meteorite (0.1399 gr) it was found to contain 0.077% bituminous components.

[1] No organic compounds were found in the Pilistevere enstatic chondrite. Oró and Nooner (1967) extracted 1 μg/g of hydrocarbons from the Indarkh enstatic chondrite.

Earlier the author studied the organic matter in rocks of the northwestern Ciscaucasia region, the southern Emba region and the southern Urals (Vdovykin, 1961, 1962a, 1963a, c). In a comparison of these data with the results of extraction performed for the meteorites it was found that with respect to the content in qualitative characteristics the extracts of the meteorites were similar to the extracts of serpentinite from the southern Urals.

After extraction a luminescent-capillary analysis was made; as is well known, this makes it possible to obtain in a first approximation some idea concerning the qualitative composition of bituminous compounds. Strips of pre-purified filter paper 6-7 mm wide and up to 15 cm long were placed in test tubes with the extract and were left there until the solvent had dried out. Then they were examined in ultraviolet light and the luminescent zones were noted; these revealed that the weakest solvents (such as petroleum ether) extract the more reducible organic compounds. They are luminescent with a bluish color. Stronger solvents first extract the neutral compounds and then /83 more acidic organic compounds.

The bituminous substances from the five carbonaceous chondrites extracted with organic solvents were studied by the luminescent-component analysis method using the procedures described by V. N. Floraskaya (1954) applicable to microquantities of bituminous matter.

The luminescence of capillary extracts of the "oily" fraction of carbonaceous chondrites is blue; the "resins" have a yellowish luminescence; the "asphaltine" have a brownish luminescence. Table 21 gives the results of the component analysis.

Table 21

Component Composition of Bituminous Matter in Carbonaceous Chondrites (% by weight)

Meteorite	"Oils"	"Resins"	"Asphaltines"
Migei	47	33	20
Staroye Boriskino	74	17	9
Cold Bokkeveld	68	23	9
Murray	61	27	12
Groznaya	45	34	21

The bituminous matter in carbonaceous chondrites is only about 10% of the total organic matter. The remaining part of the carbonaceous matter in the samples is not extracted by the solvents and therefore it has been studied very poorly, not only for meteorites, but also for rocks. The extraction of this substance from rocks and its subsequent study is usually done after the decomposition of the mineral components which it contains (Degens, Reuter, 1964). After checking this method using samples of rocks, the author used a similar method for separating carbonaceous matter from meteorites (Vdovykin, 1964d).

After extracting the bituminous components the residue was processed with 5% HCl by the cold method and with weak heating for decomposition of carbonates, traces of which are present in some carbonaceous chondrites, as well as troilite and other mineral components. Then the processing was done in a platinum bowl or crucible with HF acid for decomposing the silicates and iron compounds. At first decomposition was done without heating, but later with weak heating (up to 30-40°) the residue was processed with weak HCl for converting the undissolved salts into volatile chlorite, was washed with doubly distilled water (decantation), dried at 30° and freed from the possible presence of bituminous compounds present in meteorites in a bonded state by means of extraction with organic solvents.

After the decomposition of carbonaceous chondrites the remaining fraction enriched with carbonaceous matter represents up to 5-7% of the initial weight of the meteorites. Meteorites with a higher carbon content (Orgueil meteorite) are the best type to use with this type of processing; this is because of the peculiarities of their mineral composition. It is believed that when this method for separating out high-molecular organic matter is used it is not modified significantly (Degens, Reuter, 1964; Manaskaya, Drozdova, 1964).

A similar method was recently used by Schulz and Elofson (1965) and also by Murphy and Nagy (1966) for separating out the high-molecular organic matter /84 from the Orgueil carbonaceous chondrite. According to the data published by Murphy and Nagy, the content of polymer organic matter in this meteorite is 6.9%.

In order to separate out the unextractable carbonaceous matter from the Orgueil meteorite, Raia (1966) processed a weighed sample of this meteorite with concentrated HCl and then with a 48% solution of HF and 6 m HCl. He also demonstrated that this substance represented high-molecular organic compounds.

Thus, the organic matter in meteorites is represented by bituminous matter and high-molecular compounds which are unextractable with solvents. Later, the author of this book studied these fractions separately. They were separated from the meteorites directly prior to each type of analysis.

Elementary Microanalysis

The possibilities of using elementary microanalysis in the study of the organic compounds in meteorites are limited to the very small initial weights of the meteorite samples. Accordingly, it is necessary to determine the composition of the extract separated out by organic solvents as a whole, rather than the elementary composition of individual components. Moreover, it is known that the composition of the extracts separated out from natural samples (rocks) by use of different solvents differs somewhat in each case. This can be attributed to the fact that each solvent extracts more or less definite groups of organic compounds, for example: ether extracts reducible compounds primarily of a hydrocarbon type, benzene and chloroform extract more acidic components, etc. The extraction method also exerts an effect on the elementary composition.

An elementary microanalysis of the high-molecular organic matter in meteorites also cannot completely reflect the actual elementary composition of this matter because in many cases it is difficult to free the matter completely from microadmixtures remaining after the decomposition of the mineral phase. Therefore, using this method one can judge the actual composition with a greater or lesser approximation. In an elementary microanalysis made on samples of meteorites without separating the organic compounds from them one obtains still more approximate data, since the volatile elements determined in meteorites and particularly in carbonaceous chondrites by the use of this method have very different forms of manifestation.

The author determined the elementary composition of bituminous matter in the Groznaya meteorite (Vdovykin, 1962b) and a fraction of the Staroyo Boriskino meteorite, which is enriched with high-molecular organic matter (Vdovykin, 1965b). In an elementary microanalysis the bituminous matter from the Groznaya carbonaceous chondrite was extracted with chloroform without heating. The elementary composition of this matter, determined by the method of combustion in a reaction tube (by the method described by M. O. Korshun and N. E. Gel'man (1949), is as follows (in % of the weight of the bituminous matter: C 35.19, H 7.80, N + S + Cl + O 49.12, ash 7.89, and when scaled to the ash-free part (in %): C 38.21, H 8.54, heteroatoms 53.25 (Table 22).

With respect to its elementary composition this substance differs somewhat from the bituminous matter extracted by Mueller (1953) from the Cold Bokkeveld carbonaceous chondrite by extraction in a Soxhlet apparatus using a series of solvents. In the latter case the composition was as follows (in % by weight): C 19.84, H 6.64, N 3.18, S 7.18, Cl 4.81, O 40.02, ash 18.33, and when scaled to the ash-free part: C 24.26, H 8.12, Cl 5.89, N 4.00, S 8.78, O 48.95.

The elementary composition of the bituminous matter separated out using /85
a series of organic solvents in the Soxhlet from the Haripura and Mokoia carbonaceous chondrites was determined by Briggs (1963c). Scaled to the ash-free part, the composition of this bituminous matter from the Haripura meteorite was as follows (in % by weight): C 48.05, H 5.27, Cl 1.74, N 4.87, S 2.75, O 37.32; from the Mokoia meteorite: C 46.43, H 5.03; Cl 1.06, N 4.13[1], S 3.99, O 39.36.

Thus, according to the elementary composition, the bituminous matter in carbonaceous chondrites is characterized by a high degree of oxidation, this brings it close to the oxidized fractions of terrestrial ozocerites (Figure 44).

Another peculiarity of the composition of this bituminous matter is its high content of sulphur. nitrogen, and particularly chlorine. The latter is characteristic only for organic matter in meteorites.

It is improbable that these results do not correspond to the actual com- /86
position of bituminous matter in meteorites. All the researchers mentioned above removed free sulphur from the extracts prior to the elementary analysis.

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[1] In the article by Briggs the nitrogen content given is 2.13%; this is a misprint.

Table 22

Elementary Composition of Organic Matter in Carbonaceous Chondrites
Ash-Free Part; in % by Weight

Elements	Bituminous matter				Fraction containing high-molecular organic matter (separated out after freeing from the mineral part of meteorites	
	Separated out by cold extraction by chloroform from the Groznaya meteorite (Vdovykin, 1962b)	Separated out by extraction in a Soxhlet apparatus using a series of organic solvents from the meteorites				
		Haripura	Mokoia	Cold Bokkeveld (Mueller, 1953)	Staroye Boriskino (Vdovykin, 1965b)	Orgueil (Schulz, Elofson, 1965)
		(Briggs 1963c)				
C	38,21	48,05	46,43	24,26	17,18	64,5
H	8,54	5,27	5,03	8,12	5,47	3,9
Cl		1,74	1,06	5,89	2,56	
N	53,25	4,87	4,13	4,00	74,79	31,6
S		2,75	3,99	8,78		
O		37,32	39,36	48,95		
Total	100,0	100,0	100,0	100,0	100,0	100,0

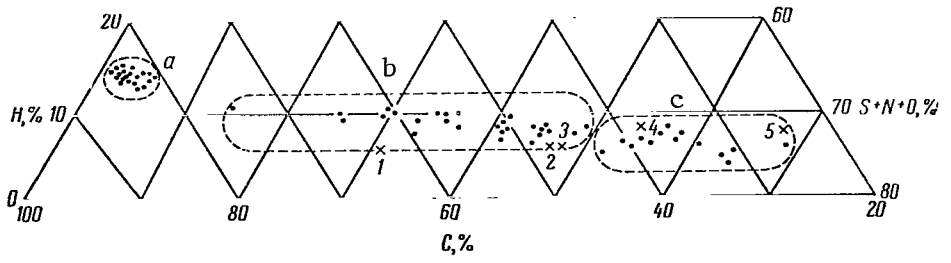


Figure 44. Diagram of the Elementary Composition of Ozocerites and Organic Compounds in Carbonaceous Chondrites.
Fractions of ozocerites from the Minusinskaya and Tuvinskaya depressions (according to Putsillo, et al, 1958): a - light paraffins; b - dark paraffins; c - resinous part. Organic compounds or carbonaceous chondrites: 1 - Orgueil (from Schulz, Eloison, 1965); 2 - Mokoia meteorite; 3 - Haripura meteorite (from Briggs, 1963a, c); 4 - Groznaya meteorite (from Vdovykin, 1962b); 5 - Cold Bokkeveld meteorite (from Mueller, 1953).

For example, Mueller (1953) and Vdovykin (1962b) made a manual separation of the elementary sulphur. Briggs (1963c) eliminated it by using colloidal copper. It could be surmised that the chlorine originates from the solvents ($CHCl_3$, CCl_4). However, Mueller, using $AgNO_3$, demonstrated that the chlorine present in extracts from the Cold Bokkeveld meteorite is organic. On the other hand, if this was really the case, there should be a similar phenomenon when extracting organic matter from terrestrial rocks, and in actuality this is not the case.

The initial presence of heteroatoms, especially Cl and S, in the organic matter in meteorites was recently demonstrated by Studier, Hayatsu, Anders, 1965a-c). They used the mass spectrometer method for a study of the organic matter in carbonaceous chondrites without prior separation of this matter from the meteorites; heating in the mass spectrometer system was up to 140°. In the Orgueil and Cold Bokkeveld carbonaceous chondrites, and in some cases in the Murray meteorite as well, they identified organic compounds containing chlorine, sulphur and oxygen.

Among the compounds containing chlorine these authors noted the presence of chlorobenzene in the Orgueil meteorite, dichlorobenzene in the Cold Bokkeveld meteorite and alkyl chlorides of the series C_{12}-C_{18} in both meteorites. Hayes and Biemann (1967), in a mass spectrometer study of the Murray meteorite, also established the presence of heteroatoms (N, S, O, Cl) in the organic matter; the chlorine was found in dichlorobenzene. Alkyl chlorides were not found.

For an elementary microanalysis of the fraction enriched with high-molecular matter, the author extracted from the Staroye Boriskino meteorite two weighed samples of this fraction (Vdovykin, 1965b). The elementary composition of this fraction (average of two determinations) was as follows (in %): C 17.18, H 5.47, Cl 2.56, N + S + O 74.79 (see Table 22). Naturally, this analysis cannot completely reflect the actual composition of the high-molecular matter in the meteorites. However, even in the first approximation it can be seen that this matter also contains an increased quantity of chlorine and is characterized by a high degree of oxidation.

The elementary analysis of this fraction of organic matter is dependent on the degree of its purification from mineral components. It was mentioned above that the Orgueil is the most readily subject to such purification. Schulz and Elofson, 1965), who used a similar method in extracting high-molecular organic matter from this meteorite, determined the following composition for this fraction of the Orgueil meteorite (scaled to the ash-free part, in %): C 64.5, H 3.9, Cl + N + S + O 31.6 (see Figure 44, *l*).

The polymeric organic matter in the Orgueil meteorite was analyzed by Raia (1966) and Hayes (1967). According to Raia's data, this substance contains (in %): C 70.39, H 4.43, Cl 1.22, F 1.25, N 1.59, S 6.91, O 9.80, residue (ash) 4.58.

Thus, according to an elementary analysis, the organic matter in carbonaceous chondrites (bituminous and high-molecular organic matter) is characterized by a high degree of oxidation and an increased content of heteroatoms, among which chlorine is of particular interest.

In the future, after further developing the method of elementary ultramicroanalysis of organic compounds, it will be interesting to determine the composition of the individual fractions of the bituminous matter in carbonaceous chondrites, as well as the composition of all the bituminous matter extracted by the same method from different meteorites. Moreover, it is important to make a detailed study of the composition of the ash residue which remains after combustion of the bituminous matter. /87

Study of Organic Compounds in Carbonaceous Chondrites by the Infrared, Ultraviolet and Nuclear Magnetic Resonance Spectroscopy Methods

Infrared Spectroscopy

The infrared absorption spectroscopy method used for studying organic substances is based, as is well known, on the interaction between matter and electromagnetic radiation. The absorption of infrared rays during radioscopic study of a thin layer of matter is associated with definite changes transpiring within a molecule, transitions between the energy (vibrational levels) within it

We can differentiate the near-infrared region, which in the spectrum of electromagnetic waves occupies the region up to 2 μ, the fundamental infrared region (region of fundamental frequencies) 2-50 μ, and the far-infrared region (from 50 μ to 1 mm). The latter already corresponds to rotational (band) spectra. On the infrared spectrograph it is customary to work in the region of fundamental frequencies.

This method is being successfully used in the study of organic compounds. When a study is not confined to individual compounds, but a complex mixture of compounds, as is ordinarily encountered under natural conditions, approximate conclusions can be drawn concerning the composition of the compounds on the basis of the presence of characteristic frequencies in the absorption spectrum. In our case, naturally, the study of organic compounds in individual fractions is of the greatest importance.

The author used infrared spectroscopy in a study of bituminous matter extracted with alcohol-benzene from the Migei, Staroye Boroskino and Groznaya carbonaceous chondrites, and also the high-molecular organic matter in the Migei and Groznaya carbonaceous chondrites. The survey was made using a dual beam quartz spectrograph UR-10, produced at Jena in the region 700-3400 cm^{-1}. The bituminous matter was applied without solvents on a KCl prism in the form of thin strips measuring approximately 3 × 20 mm. The spectra were interpreted by L. Bellami (1963). /88

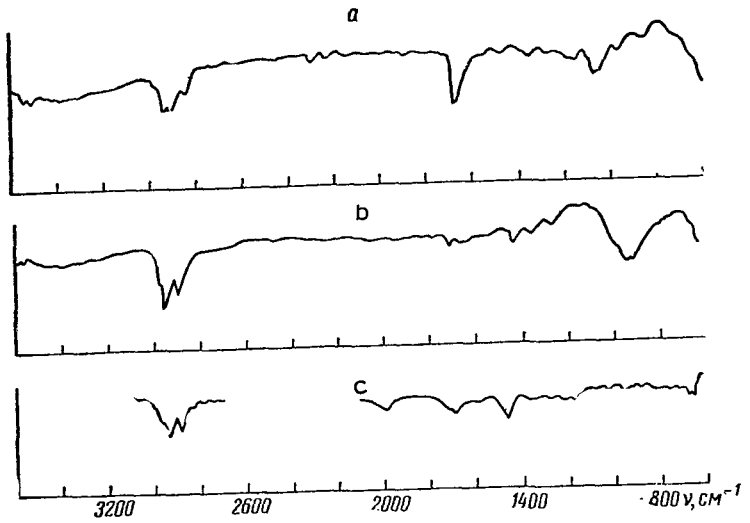


Figure 45. Infrared Absorption Spectra of Bituminous Matter in Carbonaceous Chondrites.
a - Migei; b - Staroye Borinski; c - Groznaya.

The absorption spectra of bituminous matter extracted from carbonaceous chondrites (Figure 45) are rather similar to one another (Vdovykin, 1962b, 1965a, d). These spectra contain absorption bands with maxima at 3400, 2960, 2930, 2850, 1730, 1460, 1380 (?), 1075 (Migei), 970 (Staroye Borinskino) 780 (Groznaya) cm^{-1}. This spectra are evidence of the presence of CH-groups /89 at 2960-2850 cm^{-1}, and also 1460 cm^{-1} ($-CH_3$) and 1380 cm^{-1} ($=CH_2$). The carbonyl group -C = 0 (at 1730 cm^{-1}) is noted. In the Staroye Borinskino meteorite the absorption band at 970-980 cm^{-1} is possibly caused by the carboxyl group COOH (organic acids). The extract from the Groznaya meteorite probably also contains aromatic hydrocarbons. The bituminous matter in carbonaceous chondrites constitutes a mixture of organic compounds of both a hydrocarbon (aliphatic, and also apparently aromatic) and nonhydrocarbon nature.

These results agree with data published by other researchers. In the study of the bituminous matter in the Haripura and Mokoia carbonaceous chondrites by the infrared spectroscopy method Briggs (1963c; Briggs, Mamikunian, 1963) noted absorption bands with maxima at 3400 cm^{-1} (-OH), 2960 ($-CH_3$), 2930 ($=CH_2$), 1740 and 1735 (?), 1700 (-C=O), 1460 ($-CH_3$), 1380 ($=CH_2$), 810 and 790 (aromatic), 750 (C-Cl) cm^{-1}. He concluded that this substance is a mixture of compounds, among which there are hydrocarbons, carbonic acids, phenols, and possibly saturated compounds containing chlorine.

Kaplan, et al (1963) used this method for studying the organic matter in a number of meteorites. In the case of the bituminous matter in the Orgueil, Cold Bokkeveld and Mokoia meteorites, dissolved in CCl_4, they noted on the

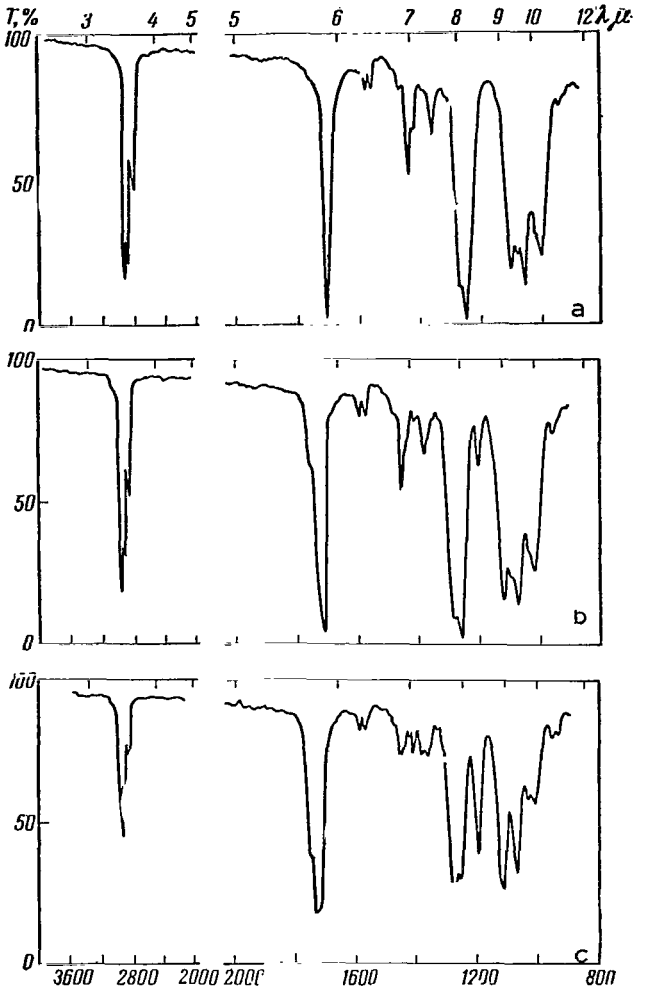


Figure 46. Infrared Absorption Spectra of Bituminous Matter in Carbonaceous Chondrites.
a - Migei; b - Murray; c - Felix. Solvent CCl_4 (Kaplan, et al, 1963).

spectra that absorption maxima occurred at 2900, 1750 and 1400-1500 cm^{-1}. They obtained the clearest spectra for the Migei, Murray and Felix meteorites (Figure 46). The absorption bands on these spectra fall in the regions 3000 - 2900, 1750 - 1730, 1610 - 1590, 1470 - 1370, 1300 - 1270, 1210, 1130, 1075, 970 - 940 cm^{-1}. Kaplan and other researchers believe that the meteorite extracts contain aliphatic and aromatic groupings, and probably unsaturated compounds (of the alkene type). The authors note that the intensity of the absorption band at 1750 cm^{-1} apparently characterizes the carbonyl (aldehydes or ketones) or carboxyl groups (unsaturated or halogen-substituted aliphatic acids).

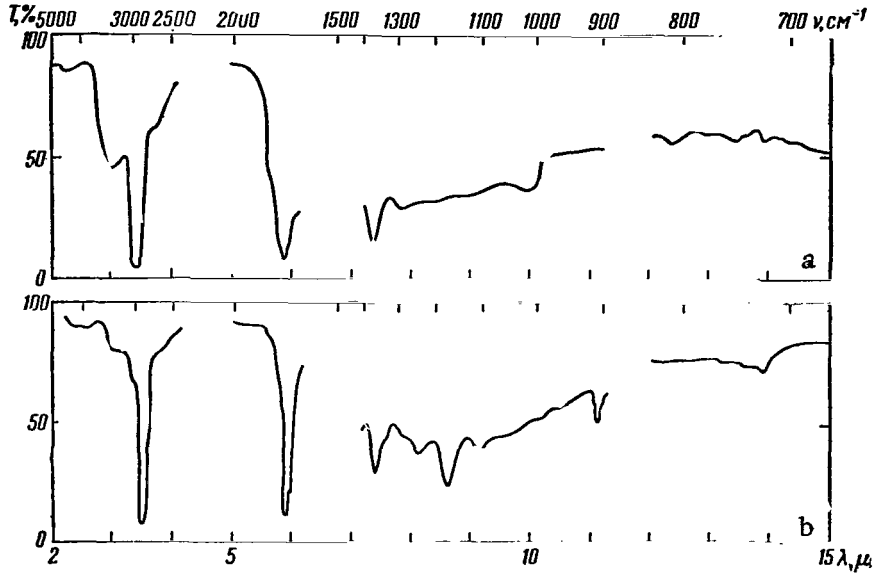


Figure 47. Infrared Absorption Spectra of Bituminous Matter Extracted from the Orgueil meteorite (a) with Benzene and a Benzene Extract of a Product of Abiogenous Synthesis Obtained by the Fischer-Tropsch (b) Netted Shown as a Comparison. Solvent CS_2 (Meinschein et al, 1963).

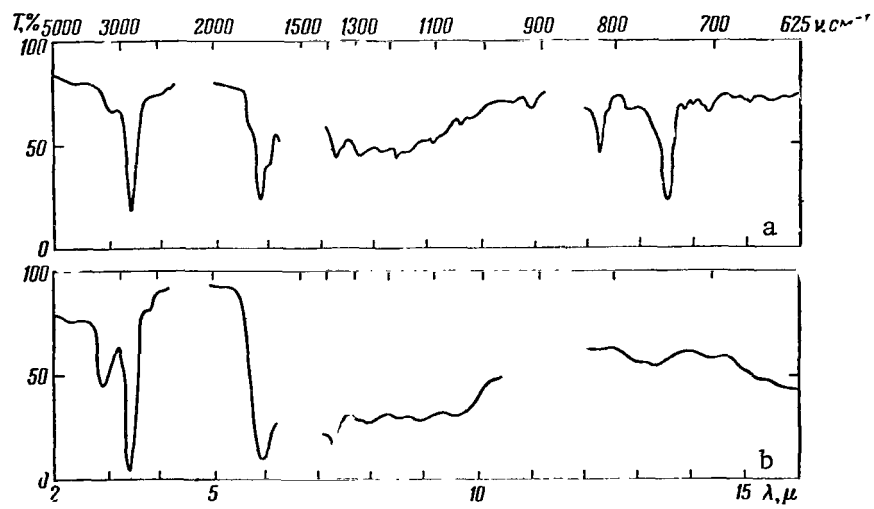


Figure 48. Infrared Absorption Spectra of Bituminous Matter Extracted from the Orgueil (a) and Murray (b) Meteorites Using a Benzene-Methanol Mixture. Solvent CS_2 (Meinschein et al, 1963).

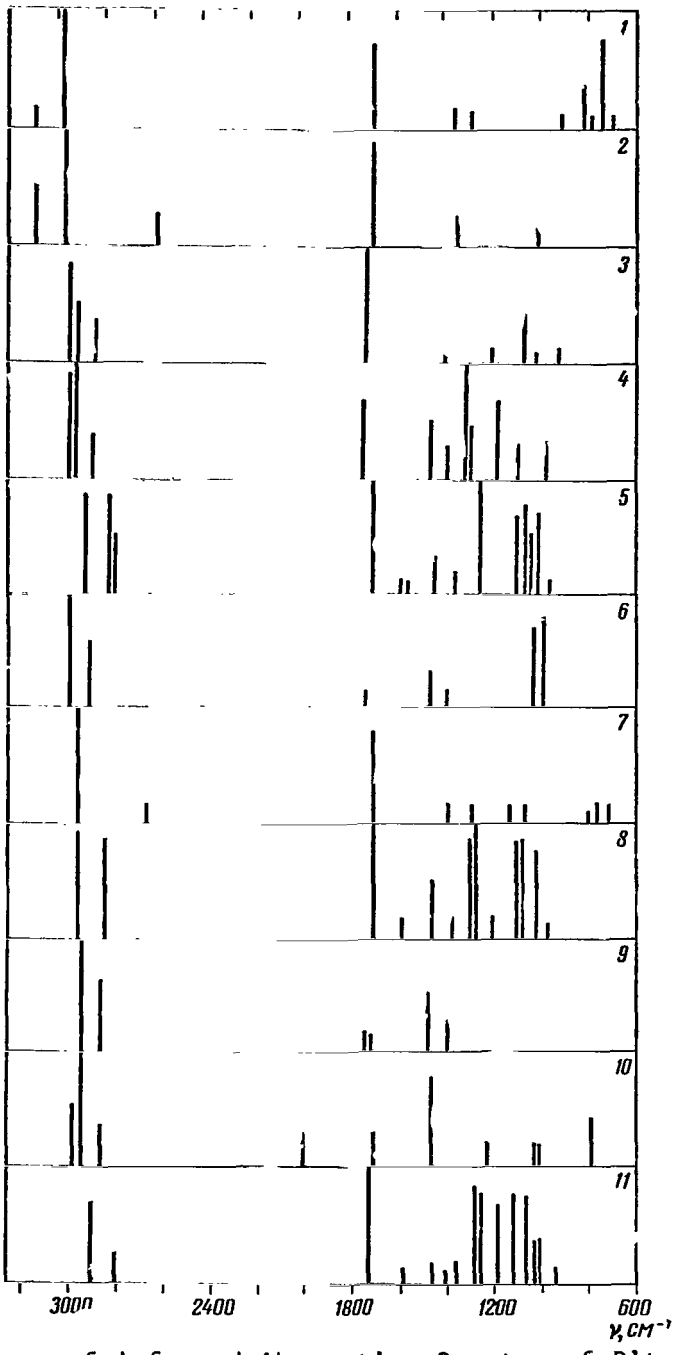


Figure 49. Comparison of Infrared Absorption Spectra of Bituminous Matter Extracted from Carbonaceous Chondrites by Various Researchers.
Along x-axis: wave number, along y-axis: relative intensity of maxima of absorption bands. Meteorites: 1 - Orgueil (from Meinschein et al, 1963); 2 - Orgueil (from Meinschein, 1963a); 3 - Migei (from Vdovykin, 1962b); 4 - Migei (from Vdovykin, 1965a); 5 - Migei (from Kaplan, et al, 1963); 6 - Staroye Boriskino (from Vdovykin, 1965a); 7 - Murray (from Meinschein et al, 1963), 8 - Murray (from Kaplan et al, 1963); 9 - Murray (from Calvin, 1961); 10 - Groznaya (from Vdovykin, 1962b); 11 - Felix (from Kaplan, et al, 1963).

Meinschein (Meinschein, 1963b; Meinschein et al, 1963) used the infrared spectroscopy method in studying a benzene extract from the Orgueil meteorite. From the absorption spectrum he noted the presence of CH-groups (2775 - 3000 and 1330 - 1430 cm^{-1}), OH- or NH-groups ($\approx$3300 cm^{-1}), aromatic groups (1030, 815 and 745 cm^{-1}) and ether carbonyl (1730 cm^{-1}) and concluded that the extract consists of a complex mixture of carbons, among which, in particular, there are saturated hydrocarbons. As noted by Meinschein, this absorption spectrum is similar in part to the spectra for the organic matter extracted from recent sediments and also to the absorption spectrum of a product of a biogenous synthesis obtained by the Fischer-Tropsch (Figure 47).

Meinschein et al (1963) also studied extracts obtained using benzene-methanol from the Orgueil and Murray meteorites. They found that the infrared absorption spectra of these extracts are also rather similar to one another (Figure 48). They determined the presence of OH- and NH-(2.9 - 3.1 μ), CH-(3.3 - 3.6 μ and 7.1 - 7.9 μ) and -C = O-(5.6 - 6.0 and 8.9 - 9.0 μ) -groups, aromatic or olefin compounds (14 - 14.5 μ) and a C-C bond (13.8 - 13.9 μ).

The infrared absorption spectra of bituminous matter extracted by different researchers from the same or from different carbonaceous chondrites in general are in reasonable agreement with one another (Figure 49). Some similarity is noted in a comparison of these spectra with the infrared absorption spectra of bituminous compounds extracted by the author from rocks (Vdovykin, 1963b), ground water (Vdovykin, 1962a), and other natural objects (Figure 50). /92
A definite similarity can also be found when they are compared with the absorption spectra of some terrestrial organic minerals (Figure 51).

It was mentioned above that infrared spectroscopy gives the best results when studying individual fractions, rather than a complex mixture of organic compounds. W. Meinschein, et al (1963) investigated fractions of bituminous matter from the Orgueil carbonaceous chondrite. An extract of bituminous matter weighing 75 mg, extracted by benzene-methanol from 14.5g of this meteorite was defractionated by the eluant chromatography method in a silica-gel column into a number of fractions whose quantitative content was (in % of weight of the entire extract): eluate of n-heptane (n-C_7) 10.0, CCl_4 - 5.4, benzene 3.2, methanol 63.1. The authors studied these fractions by the infrared (Figure 52) and ultraviolet spectroscopy methods. The infrared spectrum of the chromatographic fractions of n-C_7 and CCl_4 revealed the presence of /93
C - C and C - H bonds. At the same time the n-C_7 eluate contained primarily n-paraffins (their absorption maximum was 720 cm^{-1}), and in the CCl_4 fraction, aromatic hydrocarbons (795 cm^{-1}) as well, that is, the n-C_7 fraction in a large part of the CCl_4 eluate consisted of saturated hydrocarbon. The benzene eluate consisted for the most part of aromatic hydrocarbons; its absorption spectrum revealed bands with maxima at 1030, 815 and 745 cm^{-1}, which are usually noted in the aromatic fractions of petroleums. The absorption spectrum for the methanol eluate revealed OH- or NH-(3300 cm^{-1})- and -C = O-(1730 cm^{-1}) groups. It contained alcohols or amino compounds and esters.

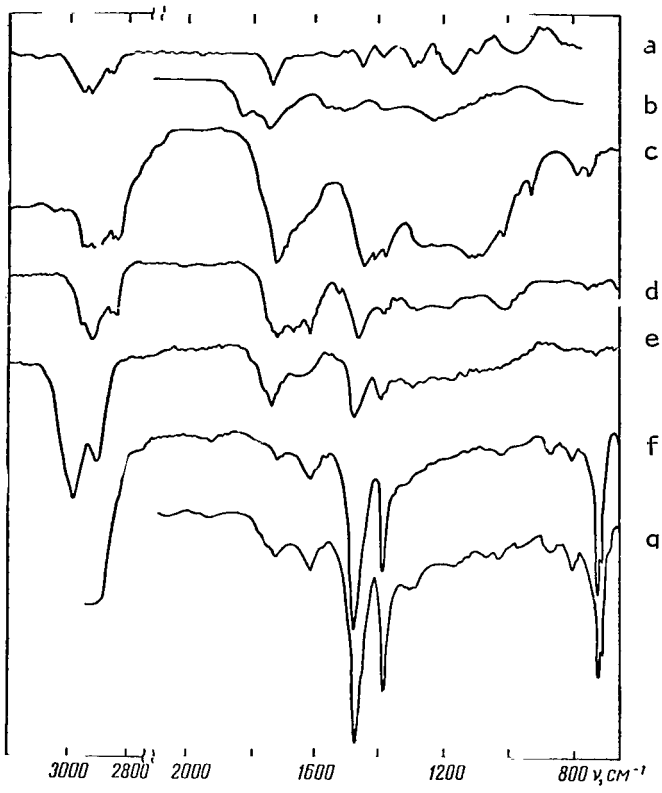


Figure 50. Comparison of Infrared Absorption Spectra of Bituminous Matter in the Migei Meteorite (a) and Terrestrial Bituminous Substances Extracted by the Author from Serpentinite (Southern Urals) (b), Clay Shale (Southern Urals) (c), Ground Water (Northwestern Ciscaucasia) (d), Combustible Shale (Estonia, Kukruze Mine) (e), and also from Ozocerite (Ukranian SSR) (f), an Oily Fraction of this Ozocerite (g).

The infrared spectroscopy method was used by the author in studying the high-molecular organic matter in the Migei and Groznaya carbonaceous chondrites which had been extracted after freeing it from the mineral components. The spectrum of the Migei meteorite (Figure 53) revealed absorption at 1080-1175 cm^{-1}, caused by C-H bonds (aromatic), at 1440 cm^{-1} (deformed group of OH carboxyl or alcohols) and at 1660 cm^{-1}, apparently -C = O vibrations in the carboxyl group or vibrations associated with the aromatic group. A wide absorption band in the region 3000 - 3600 cm^{-1} was responsible for the vibrations in hydroxyl OH; in this case it was caused by the moisture content of the fraction. The absorption spectrum of the similarly extracted high-molecular organic matter in the Groznaya meteorite has a similar form. This spectrum also revealed the presence of absorption bands with maxima at 1130, 1660, 2370, 3590 cm^{-1}, but they were less clearly expressed than in the case of the Migei meteorite. Soot was studied for comparative purposes, but noticeable absorption bands were not detected in its infrared spectrum.

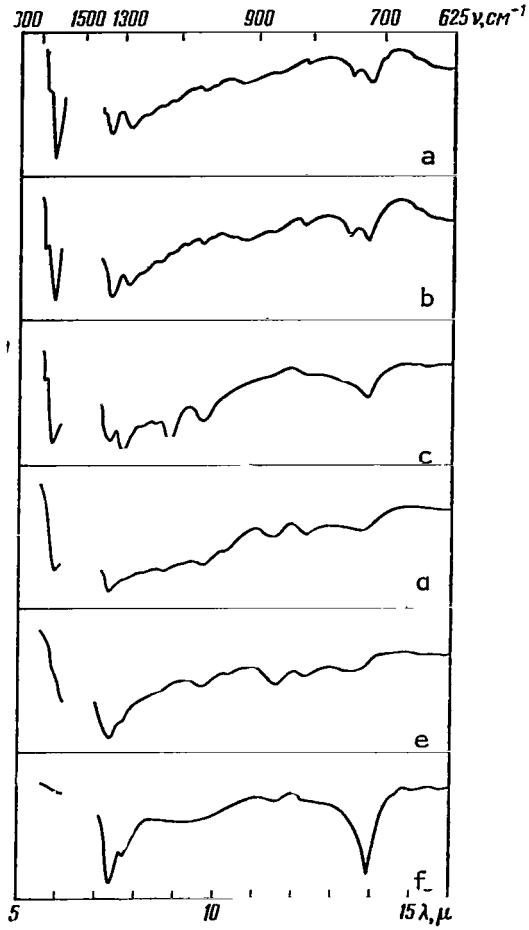


Figure 51. Infrared Absorption Spectra of some Terrestrial Organic Minerals (Bell, Hunt, 1963). a - Ingramite; b - Albertite; c - Wurtzlite; d - Gilsonite; e - Tabdeyite; f - Ozocerite.

The infrared absorption spectra of high-molecular organic matter in carbonaceous chondrites are somewhat similar to the infrared spectra of humic compounds (Kasatochkin, et al, 1956, 1958; Petrova, 1963; K. V. D'yakonova, 1964, and others). Like the humic substances, the high-molecular organic matter in meteorites apparently has an aromatic condensed structure of a carbon skeleton with a peripheral part where functional groups are arranged. Moreover, the degree of this polymerization in different carbonaceous chondrites in all probability is different. In the case of the Migei meteorite, judging from the infrared spectrum, the functional groupings are more clearly expressed than for the Groznaya meteorite. However, in the structure of the high-molecular organic matter in the Groznaya meteorite there is /95 a predominance of the nuclear component.

Thus, on the basis of data from infrared spectoscopy, the bituminous matter in meteorites constitutes a complex mixture of organic compounds of hydrocarbon and non-hydrocarbon character.

In the infrared absorption spectrum the overwhelming part of the organic matter (high-molecular) reveals C-H bonds and OH- and -C = O-groups. It probably constitutes an aromatic condensed system with side groupings, somewhat similar in structure to humic substances.

Spectroscopy in the Ultraviolet and Visible Regions

In the spectrum of electromagnetic waves the ultraviolet region extends from 50 Å to 400 mμ[1] and the visible region extends from 400 to 700 mμ. In a radioscopic study of a thin layer or solution of the investigated organic matter using such rays changes occur in the electron levels of the molecules which are registered in the spectrum. In studying aromatic polynuclear hydrocarbons the most effective method is spectroscopy in the ultraviolet and visible regions. However, when studying complex natural mixtures of organic compounds, particularly when the contents of these compounds are small, absorption spectroscopy is not sufficiently effective.

[1] Studies usually are made in the middle of the ultraviolet region (200-400 mμ).

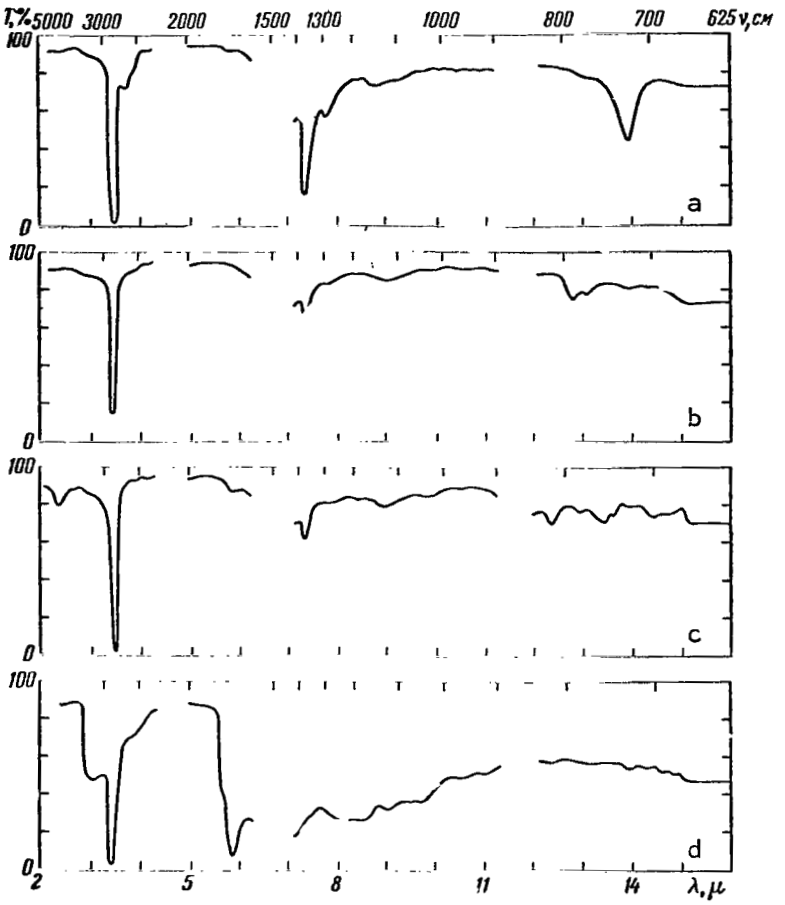


Figure 52. Infrared Absorption Spectra of Chromatographic Fractions of Bituminous Matter Extracted from the Orgueil Carbonaceous Chondrite Using Benzene-Methanol (Meinschein, et al, 1963).
The eluates were separated out with the following solvents:
a - *n*-heptane; b - CCl_4; c - Benzene; d - Methanol.

The author could not obtain any information on the absorption spectrum in the ultraviolet region or the bituminous matter extracted from the Staroye Borinskino meteorite. In the absorption spectrum of the bituminous matter extracted by ethanol from the Migei meteorite, in the visible region there was one very weak broad absorption band with a maximum at 360 mμ (Figure 54) (Vdovykin, 1966b). It is assumed (Levshin, 1951) that this absorption band is most characteristic for anthracene. Figure 54 shows the absorption spectrum of high-molecular organic matter extracted from the Migei meteorite. No noticeable absorption bands appear in this spectrum.

Kaplan, et al (1963) used spectroscopy in the ultraviolet and visible regions for studying the bituminous matter in a number of carbonaceous chondrites, including the Orgueil, Migei, Cold Bokkeveld and Murray meteorites. On the

spectra obtained in the visible region these researchers could not detect noticeable absorption bands. However, in the ultraviolet spectra (Figure 55) absorption bands could be noted in the region 270-280 mμ; these could be evidenced of the presence of aromatic hydrocarbons, conjugated olefins, and possibly carbonyl groups. In some cases there was a broad absorption band with a maximum at 290 mμ; Kaplan feels that this may indicate the presence of aromatic polynuclear hydrocarbons.

In a comparative study of ultraviolet spectroscopy of fractions of aromatic hydrocarbons in the Orgueil carbonaceous chondrite and samples of terrestrial origin (recent sediments, sedimentary rocks, etc.), Meinschein (1963b) noted that the fraction of the Orgueil meteorite is rather similar to the fraction of aromatic hydrocarbons in recent sediments (Figure 56).

Meinschein, et al (1963) made a more detailed study of the chronatal fractions of bituminous matter in the Orgueil and Murray carbonaceous chondrites (Figure 57). They used ultraviolet spectroscopy in confirming the information which they obtained in work with the infrared spectroscopy method, that is, that the n-C_7 eluate of the Orgueil meteorite consists of saturated hydrocarbons. In the CCl_4 eluate there was some absorption; it contains not only saturated hydrocarbons, but also aromatic or conjugated olefins. The aromatic hydrocarbons are concentrated in the strongly absorbing benzene eluate and the authors feel that the absorption maxima can be attributed to phenanthrenes, pyrenes and chrysenes. The methanol eluate contains alcohols and amino compounds and esters. /97

The ultraviolet spectra of the eluates of bituminous matter from the Murray meteorite (Figure 57), according to Meinschein's data, confirmed interpretation of the composition of the eluates in this meteorite as well, but in the Murray meteorite the distribution of organic compounds, and especially aromatic hydrocarbons is different than in the Orgueil meteorite.

The most effective method now available for studying individual polynuclear aromatic hydrocarbons is molecular emission spectroscopy at low temperatures -- quasiline florescence spectra. This method was developed in 1952 by E. V. Sheol'skiy, et al (1959, 1960, 1962, and others).

With dissolving in individual neutral solvents, in n-paraffins (n-hexane, n-heptane and others), and freezing at a temperature of 77.3°K aromatic hydrocarbons give spectra which are so well-structured that identification from them can be made as successfully as from atomic emission spectra. This method differs from absorption spectroscopy in its high sensitivity, specificity and selectivity and makes it possible to detect individual organic compounds in a complex mixture.

At the request of the author, in 1964 T. A. Teplitskaya obtained fluorescence spectra of bituminous matter which the author had extracted from the Migei, Staroye Borinskino and Cold Bokkeveld carbonaceous chondrites. The comparitive results were published in an article by B. N. Florovskaya, G. P. Vdovykin, T. A. Teplitskaya and R. B. Zezin (1965).

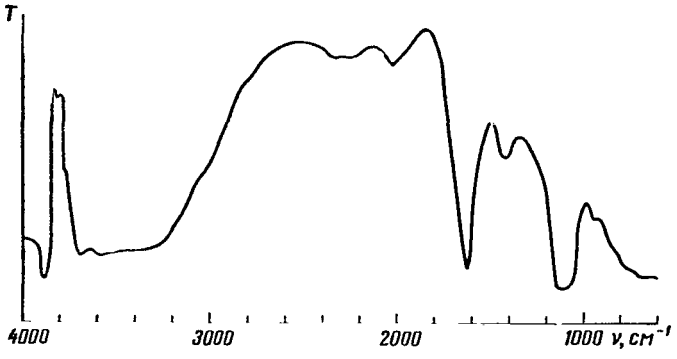


Figure 53. Infrared Absorption Spectrum of High-Molecular Organic Matter in the Migei Carbonaceous Chondrite (Vdovykin, 1964d).
T - transmission; ν - wave number.

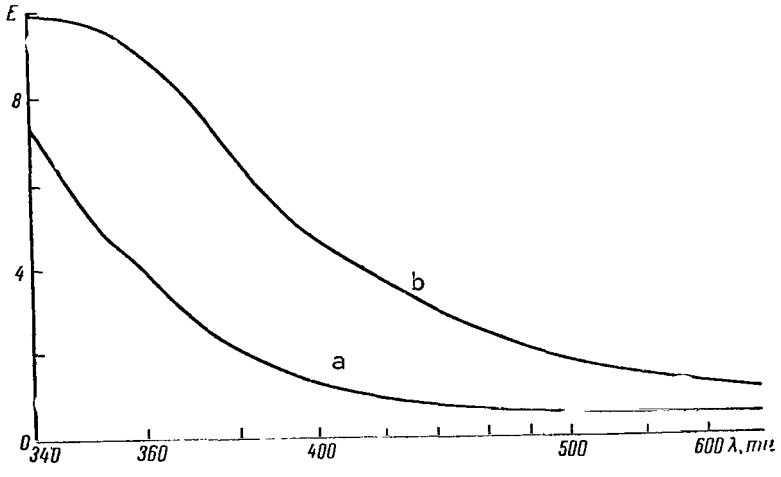


Figure 54. Absorption Spectrum in the Visible Region for an Alcohol Extract of the Migei Carbonaceous Chondrite (a) and the High-molecular Organic Matter in this same Meteorite (b).

The bituminous compounds for study were separated out using carbontetrachloride from four carbonaceous chondrites in the following quantity (in mg): Migei 0.80 (meteorite sample 0.5153 g), Staroye Borinskino 1.14 (sample 0.4123 g), Cold Bokkeveld 0.47 (sample 0.3050 g). The fluorescence spectra were obtained in the Physics Faculty at Moscow Pedagogic Institute imeni V. I. Lenin using a ISP-73 spectrograph in *n*-hexane and *n*-heptane at the temperature of liquid nitrogen (77.3°K). /99

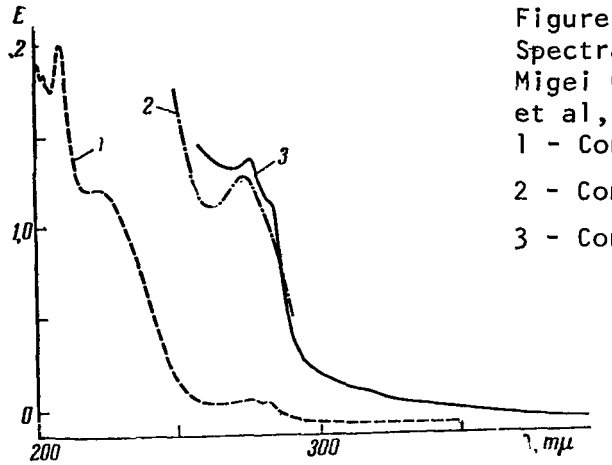


Figure 55. Ultraviolet Absorption Spectra of Bituminous Matter in the Migei Carbonaceous Chondrite (Kaplan, et al, 1963).
1 - Concentration 1 μl in 0.5 ml CH_3OH;
2 - Concentration 5 μl in 0.5 ml CH_3OH;
3 - Concentration 1 μl in 0.5 ml CCl_4.

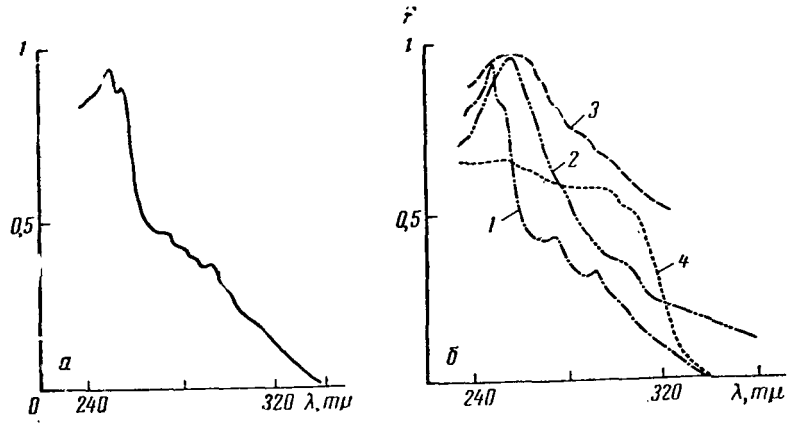


Figure 56. Ultraviolet Absorption Spectra of Fraction of Aromatic Hydrocarbons in the Orgueil Carbonaceous Chondrite (a) and Other Samples (b) (Meinschein, 1963b).
1 - Recent sediments; 2 - Sedimentary rocks; 3 - Petroleum; 4 - Product of Abiogenous synthesis by the Fischer-Tropsch method. Solvent -- isooctane.

In all study cases the fluorescence spectra for the bituminous compounds yielded a number of narrow lines, from which with an accuracy to 1-2 Å it was possible to identify the following individual polynuclear aromatic hydrocarbons: anthracene, 1,12-benzoperylene, 3,4-benzoprylene, and traces of perylene and protofluorine (Figure 58, Table 23).

The individual polycyclic aromatic hydrocarbons in the Orgueil and Cold Bokkeveld carbonaceous chondrites were recently investigated by Commins and Harington (1966). From a sample of the Orgueil meteorite (weight 5.50 g) and two samples of the Cold Bokkeveld meteorite (weight 9.26 and 4.54 g) they used /100

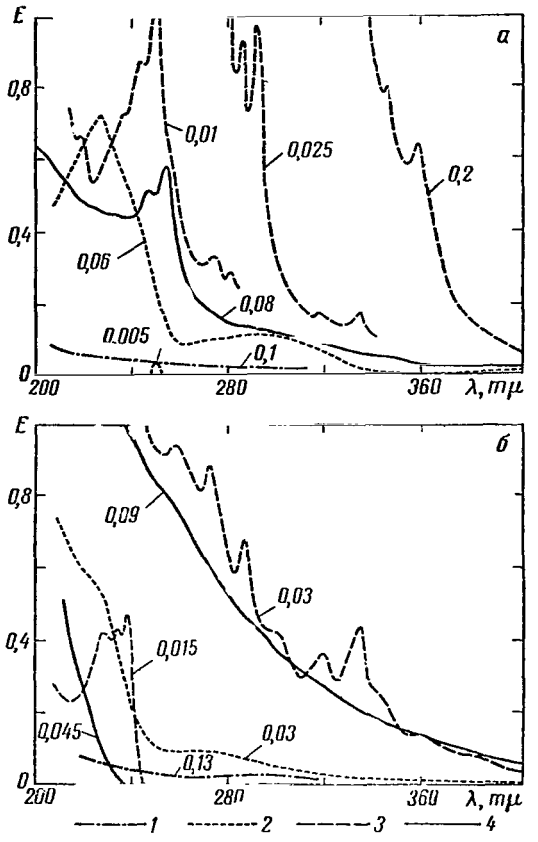


Figure 57. Ultraviolet Absorption Spectra of Chomatographic Fractions of Bituminous Matter in Carbonaceous Chondrites (Meinschein et al, 1963). a - Orgueil; b - Murray. Eluates were separated out by solvents; 1 - *n*-heptane; 2 - CCl_4; 3 - benzene; 4 - methanol. The numbers on the curves represent the concentration in solution (mg/ml).

benzene to separate out the bituminous matter; the quantity of the latter in the three meteorite samples was 164.0, 23.4 and 17.9 mg respectively. Polycyclic aromatic hydrocarbons in the extracts were studied by the chromatography and ultraviolet spectroscopy methods The Orgueil meteorite was found to fine fluorene, as well as traces of 1,2-and 3,4-benzopyrenes. The extract contained much free sulphur, and therefore it could not be studied in greater detail. In benzene extracts of two samples of the Cold Bokkeveld meteorite Commins and Harington identified and quantitatively determined the following individual polycyclic aromatic hydrocarbons: fluorene, phenanthrene, fluoranthen, pyrene, chrysene, 1,2-benzopyren, 3,4-benzopyrene, 11,12-benzofluoranthen, 1,12-benzopyrene, anthanthrene, protofluorine; anthracene and perylene could be detected qualitatively (Table 24). Figure 59 shows an ultraviolet absorption spectrum of 3,4 -benzopyrene extracted from the Cold Bokkeveld.

Nuclear Magnetic Resonance Spectroscopy

Resonance absorption in the radio wavelength range of about 10 m, caused by the magnetic properties of atomic nuclei, is used in the nuclear magnetic resonance method.

Kaplan, et al (1963) used the nuclear magnetic resonance method in a study of bituminous compounds in the Migei, Cold Bokkeveld, Murray, Mokoia, Felix, and other carbonaceous chondrites. /101 In the nuclear magnetic resonance spectrum for an extract of the Migei meteorite, dissolved in methanol (Figure 60), Kaplan noted methyl groups, evidence of the presence of paraffin hydrocarbons in the extract, as well as aromatic groupings. The spectra of organic extracts dissolved in CCl_4 for the Migei (Figure 60), Murray and Felix meteorites, in addition to revealing methyl and aromatic groups, indicated the possible presence of allyl groups: $-CH_2-CH-CH_2$ or compounds containing chlorine, nitrogen or oxygen. The Murray and Felix meteorites also hypothetically give evidence of presence of a vinyl group

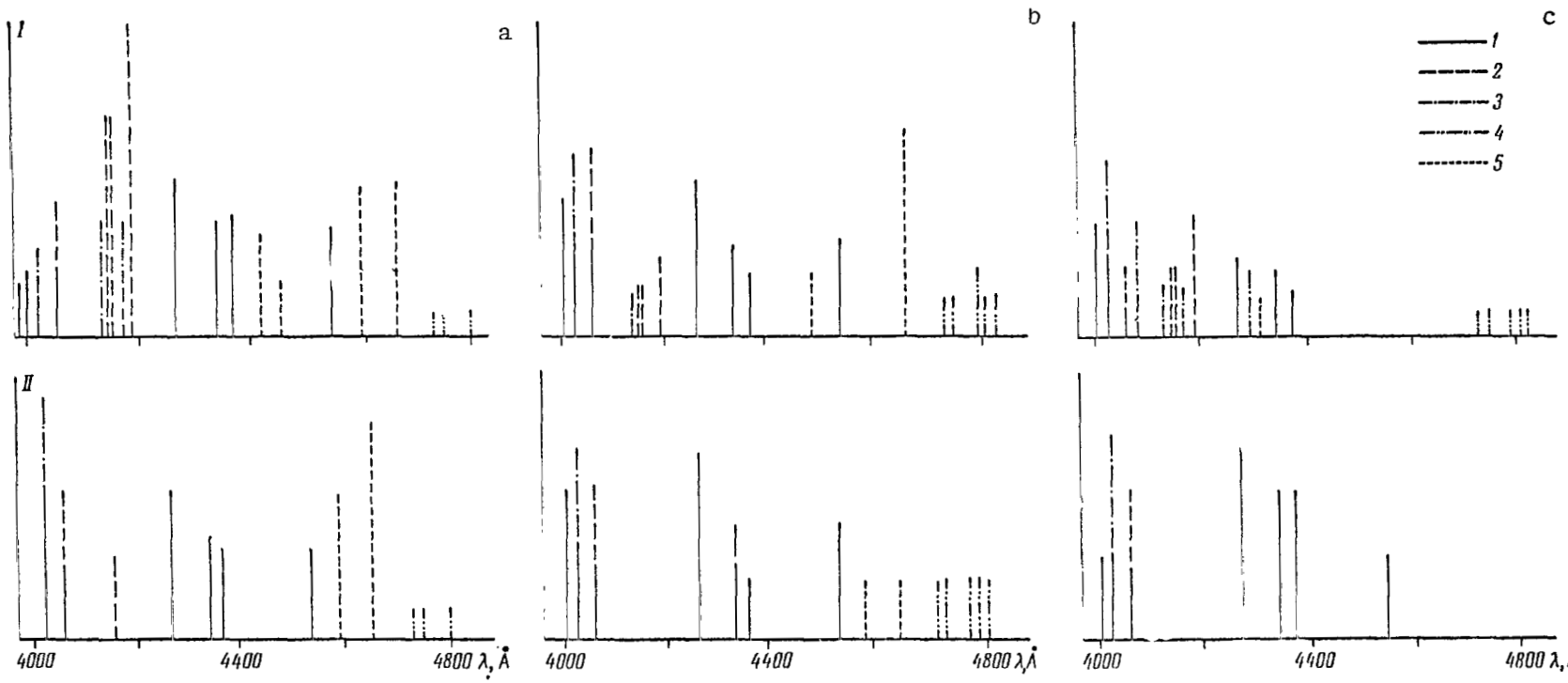
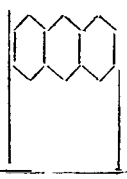
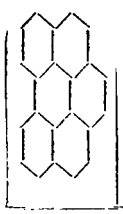
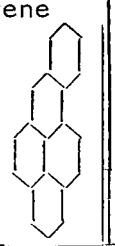
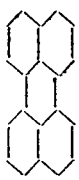


Figure 58. Diagrams of Florescence Spectra of Bituminous Matter from the Following Carbonaceous Chondrites: Migei (a), Staroye Borinskino (b) and Cold Bokkeveld (c). Obtained at T = 77.3°K in *n*-hexane (I) and *n*-heptane (II) (from Florovskaya, Vdovykin, Teplitskaya, 1964).
Lines: 1 - anthracene; 2 - 1,12-benzoperylene; 3 - 3,4-benzopyrene; 4 - perylene; 5 - other lines.

Table 23

Characteristics of Wavelengths (λ, Å) of Fluorescence Spectra of Bituminous Extracts Separated Out Using Carbontetrachloride from Three Carbonaceous Chondrites and Interpretation of Spectra

(Obtained at 77.3°K; Solvents: *n*-hexane and *n*-heptane; from Florovskaya, Vdovykin, Teplitskaya, 1964)

Extracts of Meteorites						Individual Hydrocarbons			
Migei		Staroye Borinskino		Cold Bokkeveld		Anthracene	1,12-benzoperylene	3,4 benzopyrene	Perylene
n-hexane	*n*-heptane	*n*-hexane	*n*-heptane	*n*-hexane	*n*-heptane				
3986	—	—	—	—	—	3986	—	—	—
4002	—	4002	4002	4002	4002	4002	—	—	—
4024	4024	4024	4024	4025	4024	4024	—	4024	—
4060	4061	4061	4061	4060	4061	4065	4059,8	—	—
—	—	—	—	4085	—	—	—	4085	—
4139	—	4137	—	4134	—	—	—	4133	—
4150	—	4150	—	4150	—	—	4149,4	—	—
4155	4155	4155	—	4155	—	—	4155,6	—	—
4174	—	—	—	4174	—	—	4174,9	4174	—
4191	—	4191	—	4191	—	—	4190,6	—	—
4267	4268	4268	4268	4268	4268	4268	—	—	—
—	—	—	—	4293	—	—	—	4295	—
—	—	—	—	4312	—	—	—	—	—
4341	4341	4341	4341	4339	4141	4341	—	—	—
4370	4370	4370	4370	4370	4370	4370	—	—	—
4422	—	—	—	—	—	—	—	—	—
4455	—	—	—	—	—	—	—	—	—
—	—	4491	—	—	—	—	—	—	—
4541	4541	4539	4541	—	4541	4541	—	—	—
4592	4592	—	4592	—	—	—	—	—	—
4655	4655	4655	4655	—	—	—	—	—	—
4725	4725	4727	4727	4725	—	—	—	—	4727
4744	4744	4744	4744	4744	—	—	—	—	4744
—	—	4791	4791	4791	—	—	—	—	4791
4800	4800	4809	4809	4809	—	—	—	—	4809
—	—	4827	4827	4827	—	—	—	—	4827

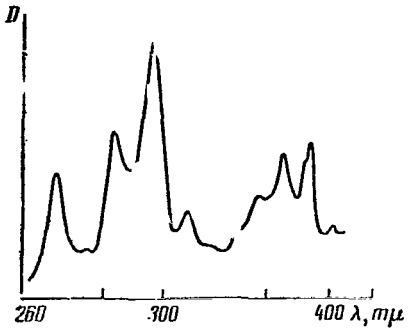


Figure 59. Ultraviolet Absorption Spectrum of 3,4-benzopyrene, Separated Out from the Benzene Extract of the Cold Bokkeveld Meteorite by Chromatographic Method. Solvent: Cyclohexane (Commins, Harington, 1966).
D - Absorption index.

$-CH = CH_2$. Extracts of the Cold Bokkeveld and Mokoia meteorites, also dissolved in CCl_4, contained functional methyl groups.

The nuclear magnetic resonance method provides some information in molecular composition of the bituminous compounds in meteorites. However, in studying the complex of these compounds and interpretation of the nuclear magnetic resonance spectrum is difficult. The author of this book observed absorption in the nuclear magnetic resonance spectrum when studying the bituminous matter in the Migei meteorite, which was dissolved in ethanol; however, the spectrum was not interpreted. This method is more effective in a study of the individual organic compounds. In the future, it will be interesting to use the

Table 24

Content of Individual Polycyclic Aromatic Hydrocarbons in Benzene Extracts of the Cold Bokkeveld Carbonaceous Chondrite (μg/g) (Commins, Harington, 1966)

Hydrocarbons	Sample 1 (from the British Museum)	Sample 2 (from Capetown Museum, South Africa	Hydrocarbons	Sample 1 (from the British Museum)	Sample 2 (from Capetown Museum, South Africa
Fluorene	2065	440			
Anthracene	Not determined	Present	1,12-benzoperylene	21	26
Phenranthrene	1873	1830	Anthranthrene	3	Present
Fluoranthen	1034	598	Protofluorine	5	15
Pyrene	880	681			
Chrysene	Present	47	Weight of extracted meteorite samples, g	9.26	4.54
1,2-benzopyrene	27	30			
3,4-benzopyrene	42	32	Weight of benzene extract, mg	23.4	17.9
11,12-benzofluoranthen	Present	5	Benzene extract, %	0.25	0.39
Perylene	Not determined	Present			

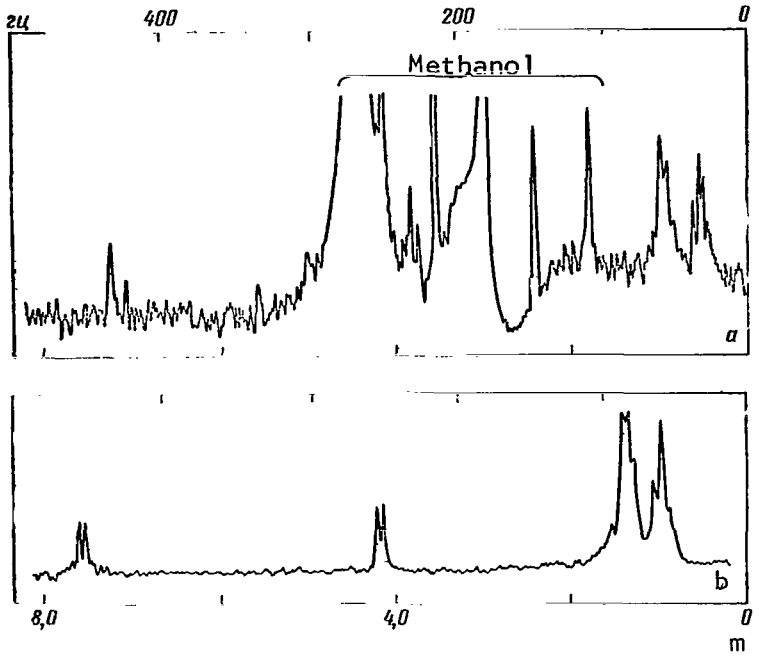


Figure 60. Nuclear Magnetic Resonance Spectra of Bituminous Matter in the Migei Meteorite (Kaplan et al, 1963). a - Solvent CH_3OH, amplification 20; b - Solvent CCl_4, amplification 12.5; m- Millionths.

nuclear magnetic resonance method for studying the organic compounds of polymeric type in carbonaceous chondrites.

Thus, by use of the infrared, ultraviolet and nuclear magnetic resonance spectroscopy methods it was demonstrated that the bituminous matter in carbonaceous chondrites has a complex composition; it contains polynuclear aromatic hydrocarbons.

Study of Organic Compounds in Meteorites by the Mass Spectrometer and Chromatography Methods

Recently various researchers have investigated individual classes of organic compounds in meteorites. In addition to spectroscopic methods, the hydrocarbons in meteorites have been studied using a mass spectrometer. Oxygen- and nitrogen-bearing and other organic compounds in carbonaceous chondrites have been studied primarily by chromatographic methods. /102

Hydrocarbons

In the analysis of the hydrocarbon composition of complex natural organic mixtures considerable information is yielded by the methos of molecular mass-spectrometric analysis; this is based on the principle of ionization of matter under the influence of electron collision.

Most available mass spectrometers do not make it possible to analyze very insignificant samples of organic matter such as we are concerned with in studying meteorites; this is all the more true because these organic compounds have a high molecular weight.

The hydrocarbons in the Orgueil meteorite were studied by the mass-spectrometer method for the first time by Nagy, et al (Nagy, Meinschein, Hennessy, 1961). The hydrocarbons in this same meteorite were recently studied by the mass-spectrometer method by Murphy and Nagy (1966) and Ryedge (Murphy, Nagy, 1966). Murphy and Nagy separated out the hydrocarbon fraction from the chloroform extract of the Orgueil meteorite by the thin-layer chromotagraphic method. This fraction was found to contain *n*-paraffins, cycloalkanes and aromatic hydrocarbon. Ryedge also identified in this meteorite *n*-paraffins of the series C_{13}-C_{26} and others, probably cycloalkanes.

Meinschein, et al (1963) used the mass spectrometer method in a study of eluates of bituminous matter in the Orgueil and Murray meteorites.

In the fractions eluated with *n*-heptane, carbontetrachloride and benzene, these researchers, on the basis of the mass spectra, determined the presence of paraffin hydrocarbons (*n*-paraffins, cycloparaffins) and aromatic hydrocarbons (Table 25). Among the paraffins they noted higher concentrations of molecules with an odd number of carbon atoms.

In the Orgueil meteorite, among the polynuclear aromatic hydrocarbons the most common were phenanthrenes, with pyrenes and chrysenes somewhat less common. In the Murray meteorite the most common hydrocarbons are pyrenes, but also chrysenes, benzofluoranthenes and phenanthrenes. As demonstrated by Meinschein, Nagy and Hennessy, the fractions of aromatic hydrocarbons in meteorites are much simpler in composition than for petroleum and the organic matter in rocks.

Hennessy (Meinschein, 1963b), using the chromatographic method, confirmed that the extract of bituminous matter from the Orgueil meteorite, whose fractions were studied by the mass-spectrometer method, contains hydrocarbons (Figure 61a). Meinschein (1965b) also studied the fraction of saturated hydrocarbons in the Orgueil meteorite using the mass-spectrometer and gas-fluid chromatography methods. The mass-spectrometric analysis revealed that this fraction contained (in %): *n*-paraffins 17.6, monocycloalkanes 28.9, bicycloalkanes 17.2, tricycloalkanes 11.8, tetrocycloalkanes 11.3, bentacycloalkanes 6.4, hexacycloalkanes 6.8. Fractions of saturated hydrocarbons were extracted from three samples of the Orgueil meteorite for a gas-fluid chromatographic analysis. The chromatograms in all three cases were similar (Figure 61b). Meinschein notes that the chromatograms reveal the presence of the isoprenoid hydrocarbons pristane (2,6,10,14-tetramethylpenthadecane) and phitane (2,6,10,14-tetramethylhexadecane) there is a predominance of hydrocarbons with /103
an odd number of carbon atoms, and the ratios of the heights of the peaks has the sequence: $C_{27}/C_{26} > C_{25}/C_{24} > C_{23}/C_{22}$. In their distribution the hydrocarbons in the Orgueil meteorite differed somewhat from petroleum hydrocarbons, particularly from the hydrocarbons of the bituminous matter in Precambrian rocks (Meinschein, 1965c) (Figure 61c).

Table 25

Hydrocarbons Identified in Eluates of Bituminous Matter in the Orgueil and Murray Meteorites by the Mass-Spectrometer Method (Meinschein, Nagy, Hennessy, 1963)

Eluating Solvent	Classes of Eluated Compounds	Eluated Hydrocarbons	Hydrocarbons in Eluated Fraction of Meteorites, %	
			Orgueil	Murray
n-heptane (n-C_7)	Saturated hydrocarbons	*n*-paraffins	17,57	16,49
		Monocycloalkanes	28,86	17,28
		Bicycloalkanes	17,16	18,06
		Tricycloalkanes	11,83	13,87
		Tetracycloalkanes	11,25	13,18
		Pentacycloalkanes	6,41	10,12
		Hexacycloalkanes	6,91	10,99
Carbontetrachloride (CCl_4)	Saturated and unsaturated hydrocarbons with traces of aromatics	*n*-paraffins	16,78	—
		Monocycloalkanes	22,52	—
		Bicycloalkanes	9,86	—
		Tricycloalkanes	15,87	—
		Tetracycloalkanes	17,00	—
		Pentacycloalkanes	7,10	—
		Hexacycloalkanes	10,87	—
Benzene (C_6H_6)	Aromatic hydrocarbons	Alkylbenzenes	5,11	4,73
		Naphthalenes	16,56	3,65
		Acenaphthenes	17,75	7,72
		Acenaphthalenes	11,10	2,81
		Phenanthrenes	20,95	6,36
		Diacenaphthalenes	4,01	4,75
		Pyrenes	1,43	13,14
		Chrysenes	0,55	7,57
		Acepyrenes	1,09	2,93
		Benzopyrenes and others	21,45	46,34

The Orgueil, Cold Bokkeveld and Murray carbonaceous chondrites were studied by the mass-spectrometer by Studier, Hayatsu, Anders (1965a). They studied not only the organic extracts, but also organic compounds and other components of the carbonaceous chondrites without their prior extraction from the meteorites. The weights of the meteorite samples were as great as 50 g, and heating in the mass spectrometer system was as high as 140°. The meteorites were found to contain a considerable content of aromatic hydrocarbons, among which compounds of the following types were identified: benzene,

toluol, naphthalene, anthracene, including trimethylanthracene. Researchers also noted the presence of alcohols, compounds of the type $C_nH_{2n+1}Cl$ (n = = 12-18), $R\text{-}SO_3\text{-}CH_3$, and others. Among the gases there was a rather considerable concentration of methane, but ethane was absent; the presence of CO, CO_2. C_4H_8, CS_2, COS, and others was established. As demonstrated by the authors, the organic compounds found in the meteorites differ in distribution from the organic compounds found in bituminous shales.

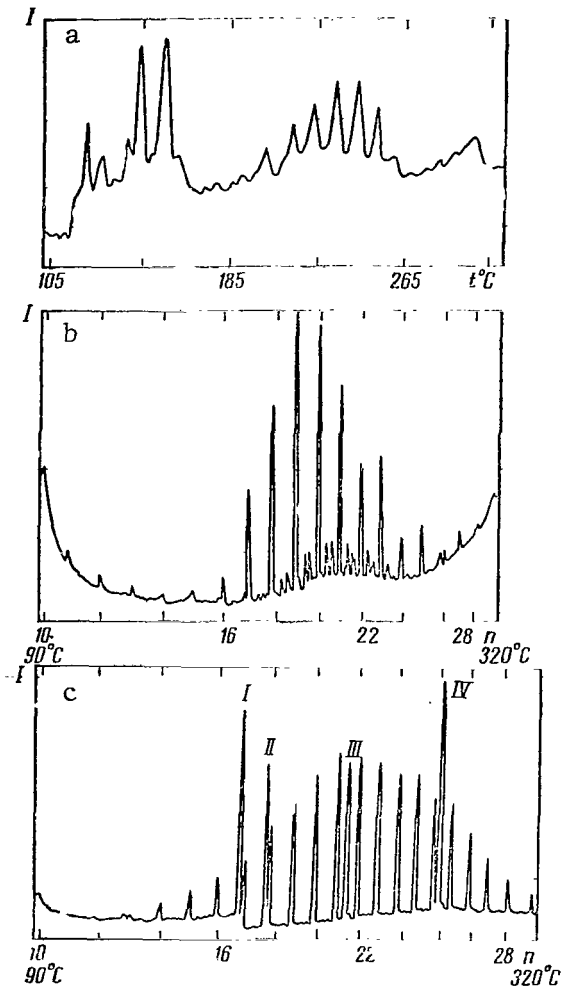


Figure 61. Chromatograms of Bituminous Compounds in the Orgueil Carbonaceous Chrondrite (a) and b) and Bituminous Matter in Rocks (c).
a - benzene extract of Orgueil meteorite; chromatogram obtained by Hennessy (Meinschein, 1963b); b - fraction of saturated hydrocarbons from the Orgueil meteorite (Meinschein, 1965b); c - fraction of *n*-paraffins of bituminous substance in Precambrian rocks (age $1 \cdot 10^9$ years; Meinschein, 1965b); I - pristane; II - phitane; III - androstane; IV - squalane; I - relative height of peak; n - number of carbon atoms.

Recently Oró and Nooner (1965) /104
and Oró, et al (1966; Oró, Nooner (1967) studied the bituminous matter extracted by organic solvents from 20 carbonaceous chondrites. They fractionated the extracts from these meteorites in a silica-gel column. The fractions of the paraffin-hydrocarbons, eluated with *n*-heptane, were investigated by the gas-fluid chromatography and mass-spectrometer methods. In all the meteorites these researchers found normal alkanes and their isomers with the number of carbon atoms in the chain from 15 to 30, in different relations. Among them they identified pristane and phitane, as well as norpristane (2,6,10-trimethylpentadecane). The quantitative content of isoprenoid hydrocarbons was less in type-I carbonaceous chondrites. Oró and Nooner established a good similarity in the

distribution of hydrocarbons in all type-II carbonaceous chondrites; this probably is evidence, as they mention, of a universality of the origin of the hydrocarbons in this group of meteorites. In the Orgueil meteorite, all Oró, et al (1966) identified cyclohexylalkanes among the cycloalkanes. and Nagy (1966b) also identified 18,19-dimethyl cyclopentanoperhydrophenanthrenes.

Meinschein (1965a) used the gas-fluid chromatography and mass spectrometer methods in studying extracts obtained using benzene-methanol from the ordinary Holbrook, Homstead and Yacanda chondrites. These meteorites were found to contain alkanes from C_{16} to C_{28}; these apparently are almost identical in composition in all three chondrites. Meinschein also identified pristane and phytane among the alkanes.

Aromatic acids and Phenols. In work on geochemical studies of organic matter it is rather common to use chromatographic methods, especially the paper chromatography method (Degens, Reuter, 1964; Manskaya, Drozdova, 1964). This method was first developed applicable to the biochemical study of the so- /105
called biochemically important organic compounds (amino acids, carbohydrates, carbonic acids, etc.) (Blok, et al 1954).

The paper chromatography method was used by Briggs (1963c) in studying water-soluble organic matter from the Haripura and Mokoia carbonaceous chondrites. He found that they contained aromatic acids and phenols, among which he identified the following acids: 3,5-dehydroxybenzoic, 2-, 3- and 4-hydroxybenzoic, 4-hydroxyphenylacetic, vanillic, and also urea and acetamide (Table 26) and acids 3-methoxy-5-hydroxybenzoic, 3,4-dehydroxybenzoic, 3-methoxybenzoic, and others (Briggs, Manikunian, 1963).

Aromatic acids in the organic matter of a number of meteorites have been noted by Kaplan, et al (1963); they used the paper chromatography method. They found compounds sensitive to diazo processes on the chromatograms of extracts from the Orgueil, Murray, Mokoia and Felix meteorites. On the basis of R_f values and qualitative reactions these authors identified organic acids; 3- and 4-hydroxybenzoic, 4-hydroxyphenylacetic and vanillic. They found that 3-hydroxybenzoic acid was most characteristic. The chromatograms also revealed about 15 other compounds which were difficult to identify.

Mass-spectrometer studies of the Murray meteorite by Hayes and Biemann (1967) revealed that this meteorite contains alkyl phenols as well as furans and alkylbenzofurans.

Fatty acids and compounds containing sulphur. In a mass-spectrometer study of the bituminous matter in the Orgueil meteorite, Meinschein, et al (1963) demonstrated that in addition to hydrocarbons the mass spectra apparen- /107
tly indicate a rather significant concentration of fatty acids. These acids (organic acids of a saturated series with an open carbon chain) in the Orgueil meteorite were the object of a special study by Nagy and Bitz (1963). In their study they used two small pieces of the meteorite weighing 12.9 and 11.3 g. In the benzene-methanol extracts of two meteorite samples, after saponification and extraction of methyl carbonates, they found the content of the

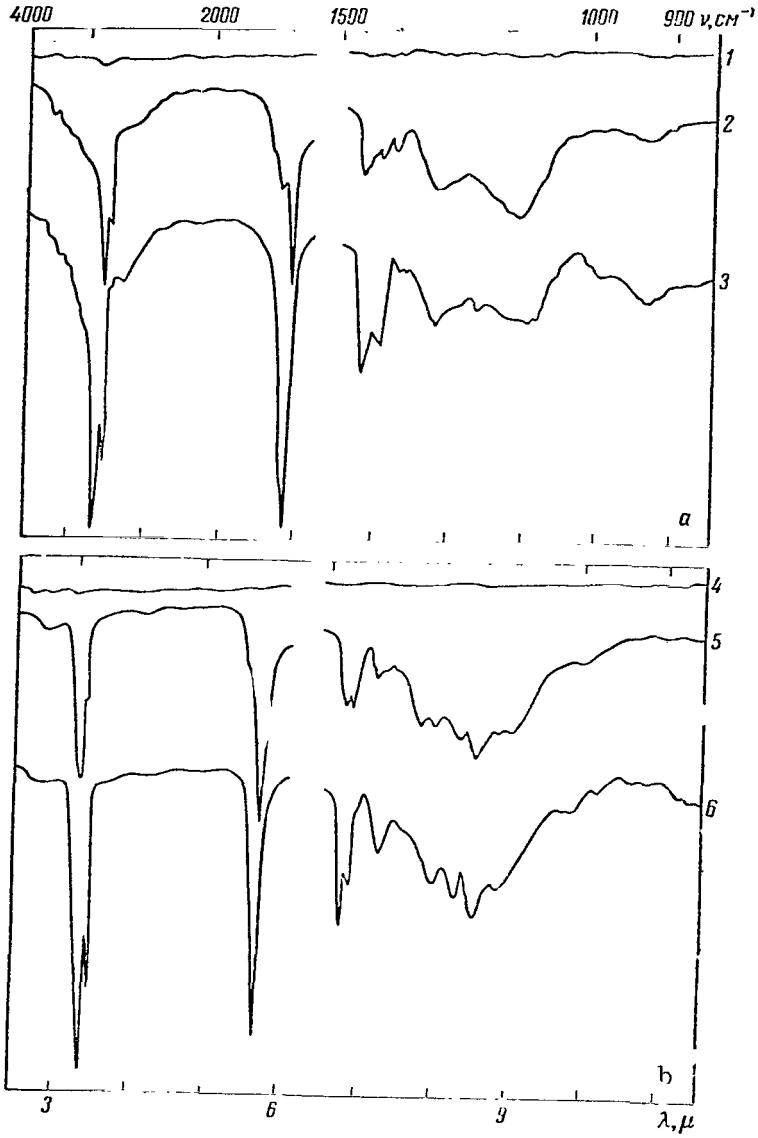


Figure 62. Infrared Absorption Spectra of Saponified Alcohol-Benzene Extract of Orgueil Carbonaceous Chondrite and Other Objects Before and After Esterification (Nagy, Bitz, 1963). a - extract of carbonic acids prior to esterification; 1 - ordinary Holbrook chondrite; 2 - Orgueil carbonaceous chondrite; 3 - fatty acids (standard); b - esterified carbonic acids; 4 - Holbrook meteorite; 5 - Orgueil meteorite; 6 - methyl carbonates (standard).

Table 26

Organic Compounds Indentified in Haripura and Makoia Carbonaceous Chondrites by the Paper Chromatography Method (Briggs, 1963c)

Chromatographic Data		Luminescence in uv	Reaction to staining	Identified Compounds
Solvent	R_f			
n-butanol-acetic-acid-water (4:1:1)	0.8	-	Dioxidized sulfanilic acid and hydrous sodium carbonate	3,5-dehydroxybenzoic acid
i-propanol-ammonia-water (8:1:1)	0.2	-	Dioxidized sulfanilic acid and hydrous sodium carbonate	
n-butanol-acetic acid-water (4:1:1)	0.9	Blue	Dioxidized sulfanilic acid and hydrous sodium carbonate	2-hydrobenzoic acid
i-propanol-ammonia-water (8:1:1)	0.6	Blue		
m-cresol-acetic acid-water (50:2:48)	0.7	Blue	Dioxidized sulfanilic acid and hydrous sodium carbonate	3-hydroxybenzoic acid
i-propanol-ammonia-water (8:1:1)	0.3	-		
n-butanol-acetic acid-water (4:1:1)	0.9	Absorption	Dioxidized sulfanilic acid and hydrous sodium carbonate	4-hydroxybenzoic acid
i-propanol-ammonia-water (8:1:1)	0.2	Absorption		
n-butanol-acetic acid-water (4:1:1)	0.8	-	Dioxidized sulfanilic acid and hydrous sodium carbonate	4-hydroxyphenylacetic acid
i-propanol-ammonia-water (8:1:1)	0.3	-		
m-cresol-acetic acid-water (50:2:48)	0.8	Blue		Vanillic acid

(continued)

Table 26 (continued)

Chromatographic Data		Luminescence in uv	Reaction to staining	Identified Compounds
Solvent	R_f			
n-butanol-acetic acid-water (4:1:1)	0.9	Blue		
buffer phenol	0.78	-	5% sodium hypochlorite	Urea
buffer phenol	0.3	-	5% sodium hypochlorite	Acetamide

Table 27

Occurrence of Monobasic Fatty Acids of the Series $C_nH_{2n}O_2$ in the Orgueil Meteorite (data from gas-fluid chromatography; (Nagy, Bitz, 1963)

Number of carbon atoms in chain	Empirical formula	Acid	Relative intensity of peak on chromatogram	Number of carbon atoms in chain	Empirical formula	Acid	Relative intensity of peak on chromatogram
14	$C_{14}H_{28}O_2$	Myristic	4	22	$C_{22}H_{44}O_2$	Behenic	7
15	$C_{15}H_{30}O_2$	Pentadecylic	3	23	$C_{23}H_{46}O_2$	Trecosanic	4
16	$C_{16}H_{32}O_2$	Palmitic	10	24	$C_{24}H_{48}O_2$	Lignoceric	5
17	$C_{17}H_{34}O_2$	Septadecylic	8	25	$C_{25}H_{50}O_2$	Heptasanic	4
18	$C_{18}H_{36}O_2$	Stearic	9	26	$C_{26}H_{52}O_2$	Ceratinic	6
19	$C_{19}H_{38}O_2$	Nonodecylic	5	27	$C_{27}H_{54}O_2$	Heptacosanic	7
20	$C_{20}H_{40}O_2$	Arachidic	5	28	$C_{28}H_{56}O_2$	Montanic	5
21	$C_{21}H_{42}O_2$	Heneicosanic	3				

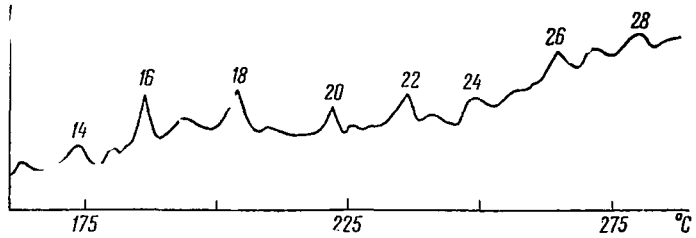


Figure 63. Chromatogram of Methyl Carbonates for Orgueil Meteorite (Nagy, Bitz, 1963).

latter to be 675 and 580 μg/g respectively; the quantity of methyl ethers of fatty acids with a straight chain was 125 μg/g in the first sample.

In the infrared spectrum (Figure 62) these fractions revealed absorption bands characteristic for carbonic acids. The authors noted absorption with maxima at 5.85, 3.7, 3.3 μ and a broad band with a maximum at 10.7 μ. After esterification (Figure 62b) the absorption maximum at 5.85 μ shifted to 5.7 μ (ether-carbonyl bond), the maximum at 3.3 μ (OH group) decreased and the maximum at 10.7 μ disappeared. However, ether bonds appeared clearly at 8.0, 8.3 and 8.5 μ. Nagy and Bitz investigated methyl carbonates by the gas-fluid chromatography method and demonstrated that the extracts contained fatty acids with the number of carbon atoms being between 14 and 28 (Figure 63, Table 27). Among them acids with an even number of carbon atoms predominated.

As a comparison, Nagy and Bitz investigated the ordinary Holbrook chondrite on samples of granite; fatty acids were not found in these objects.

The presence of fatty acids in the Orgueil meteorite was soon confirmed by Hayatsu (1964) and Ryedge (Murphy, Nagy, 1966).

Murphy and Nagy (1965, 1966) made a special study of the lipid fraction of the Orgueil meteorite to determine its content of organic compounds containing sulphur by the thin-layer chromatography, ultraviolet and infrared spectroscopy methods and by mass-spectrometer and ultramicro elementary analysis. Free sulphur, a hydrocarbon fraction containing alkanes and a fraction which stained a yellowish brown color were separated out from the chloroform extracts. The meteorite sample weighing 6.29 g yielded 285 μg of stained fraction. It contained 1.6% sulphur. Data from infrared spectroscopy revealed the presence of methyl and methylene groups (3.42 and 3.52 μ), as well as carbonyl bonds (5.7-5.9 μ) characteristic for ethers or organic acids. In the ultraviolet region the spectrum for this fraction revealed absorption /108
with maxima at 310, 285 and 278 mμ. This fraction did not have absorption bands at 260 and 280 mμ characteristic for free sulphur. The authors therefore conclude that the sulphur in the fraction is present in organic compounds containing sulphur. The fraction contained not less than five compounds. Murphy and Nagy note that the ultraviolet spectra resemble the absorption spectra of 2.2'-dicarboxydiphenyldisulfide, 2-mercaptobenzoic acid and diphenylsulfoxide (or β-phenylmercaptohydrocinnamic acid).

The organic matter extracted from another sample of the Orgueil meteorite (sample weight 2.46 g) was studied by Murphy and Nagy in the presence of compounds containing sulphur, in particular, thiols (mercaptans), by the mass-spectrometer method. The mass spectrum had a complex form (Figure 64), evidence of a multicomponent composition of the compounds, which included *n*-paraffins, cycloalkanes, monocyclic aromatic compounds, as well as sulfides with an open chain and heterocyclic compounds containing sulphur. Thiols were apparently absent, at any rate, the mass spectrum of 1-octadecanthiol, which the authors investigated as a comparison, was somewhat different.

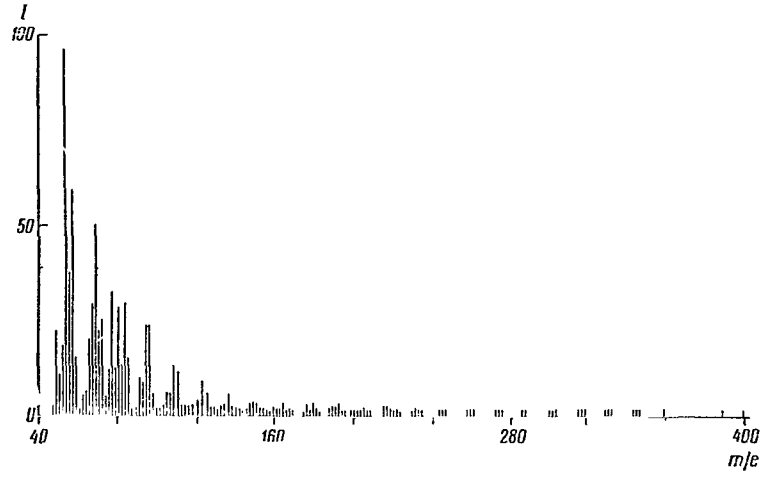


Figure 64. Mass-spectrum of Chloroform Fraction of Organic Matter in the Orgueil Carbonaceous Chondrite Containing Sulfides and other Organic Compounds Containing Sulphur (Murphy, Nagy, 1966) I - relative intensity of peaks, m/e (ratio of mass two charge).

Nagy notes that the presence of organic compounds containing sulphur in the Orgueil meteorite agrees with the results of the mass-spectrometer studies of the Murray meteorite made recently by Biemann (Murphy, Nagy, 1966), who discovered substituted thiophenes in this meteorite.

Among the thiophenes Hayes, Biemann (1967) established the presence of alkylbenzothiophenes, dibenzothiophenes and possible thiophenols. They did not detect aliphatic compounds containing sulphur in the Murray meteorite.

Amino acids in carbohydrates. Degens and Bajor (1962), and later Kaplan, Degens and Reuter (1963) used the paper chromatography method in checking a number of meteorites for their content of amino acids and carbohydrates. In carbonaceous chondrites, including the Orgueil, Migei, Cold Bokkeveld and Murray meteorites, they identified and determined, with an accuracy ±25%, the quantitative content of 17 amino acids. Their total content in these meteo- /109
rites varied from 30 μg/g in the Migei and Cold Bokkeveld to 112 μg/g in the Orgueil meteorite. Amino acids were found both in a free (in alcohol extracts) and in bonded (after hydrolysis) states in the ratio 1:3. The most common of these were: serine, glycine, alanine, leucines, glutamic acid, aspartic acid and threonine. Basic, aromatic and sulphur-bearing amino acids were detected in very small quantities (Table 28). The ultraviolet spectra for the fraction containing amino acids revealed clearly expressed absorption in the region 245-250 mμ and weak absorption at 285 mμ, which coincides with the maximum usual for aromatic amino acids (Figure 65). In the Mokoia carbonaceous chondrite the authors found amino acids also in the chondrules; only glycene was identified here in a free state. After hydrochloric acid hydrolysis the chondrules were found to contain glycene, alanine, serine, threonine, leucines, glutamic and asparatic acids. Their content attained 80 μg/g. These

researchers noted that the Orgueil, Murray and Mokoia carbonaceous chondrites contained carbohydrate (5-24 μg/g); traces of carbohydrates were also found in the Migei and Cold Bokkeveld meteorites. The carbohydrates were represented by mannose and glucose, and in individual cases, by arabinose as well. The authors combined the carbohydrate fractions of the four carbonaceous chondrites. In the ultraviolet spectrum of this combined fraction there was some absorption in the region 225-275 mμ (Figure 66). Carbohydrates were not found in the chondrules.

Table 28

Content of Amino Acids in Some Carbonaceous Chondrites Determined Using the Paper Chromatography Method After Hydrochloric Acid Hydrolysis (found Amino Acids (Kaplan, Degens, Reuter, 1963)

Class of Amino Acids	Amino Acids	Amino Acids in Meteorites in μg/g				
		Orgueil	Migei	Cold Bokkeveld	Murray	Felix
Basic	Arginine	7.3	-	-	Traces	-
	Ornithine	6.5	-	-	1.5	-
	Lyzine	4.4	0.8	1.4	1.0	-
	Histidine	2.9	-	-	0.5	-
Acidic	Aspartic	2.9	1.9	2.6	1.4	-
	Glutamic	1.8	3.3	3.1	1.7	4.9
Neutral	Glycene	9.1	3.8	4.9	8.8	6.0
	α-alanine	6.9	3.0	2.1	5.4	4.9
	β-alanine	1.5	-	-	0.6	-
	Serine	12.3	3.4	2.4	7.4	11.2
	Proline	-	1.6	-	0.5	-
	Valine	6.5	0.2	-	1.9	1.0
	Threonine	4.0	1.6	2.0	1.3	1.8
	Leucines	9.8	2.1	2.7	2.6	3.1
Atomatic	Tyrosinine	5.1	0.7	0.6	0.7	1.6
	Phenylalanine	6.9	0.5	-	0.4	1.4
Continuing sulphur	Cystine	-	-	-	-	-
	Methionine	-	-	-	Traces	-
Total		87.9	22.9	21.8	35.7	35.9

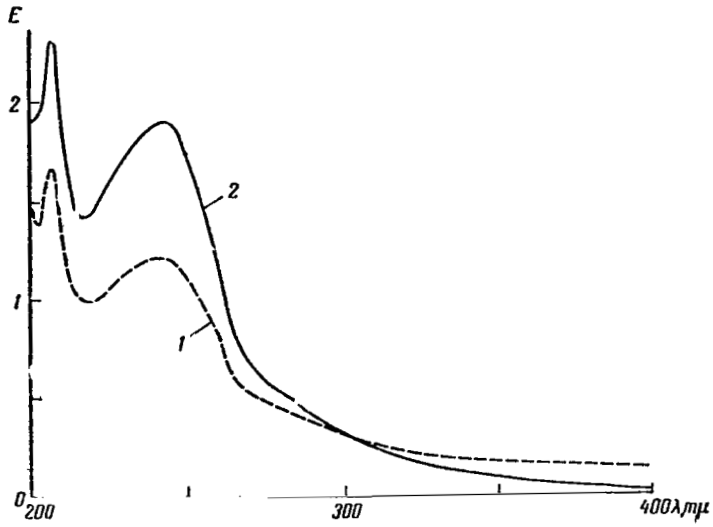


Figure 65. Ultraviolet Absorption Spectra of Fractions Containing Amino Acids, Cold Bokkeveld Meteorite (Kaplan, et al, 1963).
E - absorption; λ - wavelength; 1 - bound amino acids; 2 - free amino acids.

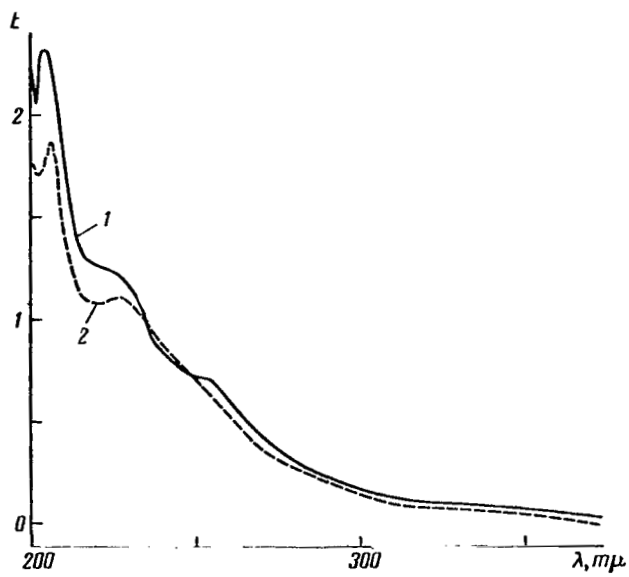


Figure 66. Ultraviolet Spectra of Combined Fractions Containing Carbohydrates from the Orgueil, Murray, Felix and Mokoia Carbonaceous Chondrites (Kaplan, et al, 1963). Concentration 75 μl in 0.5 ml 10% isopropanol; 1 - pH 7; 2 - pH 1.

In some of the investigated meteorites, particularly in the Lancé meteorite, Kaplan, et al found a very high content of amino acids. Oró and Skewes (1965) postulated that some of the amino acids in meteorites could be the result of contamination under terrestrial conditions. They used the chromatographic method for studying amino acids which would be transferred into meteorites from human hands, or by dust from the air. The composition of the amino acids in these possible sources of contamination, as demonstrated by Oró and Skewes, is rather constant in qualitative respects and in their relative distribution. The conclusion that some of the amino acids would be the result of contamination of meteorites after their falling through the earth is confirmed by the results obtained by Hamilton (1965) and Avapar (Oró, Skewes, 1965). Kaplan (1965) agreed with this assumption but noted that the presence of amino acids in chondrules carefully extracted from meteorites is evidence that most of the amino acids are natural in carbonaceous chondrites.

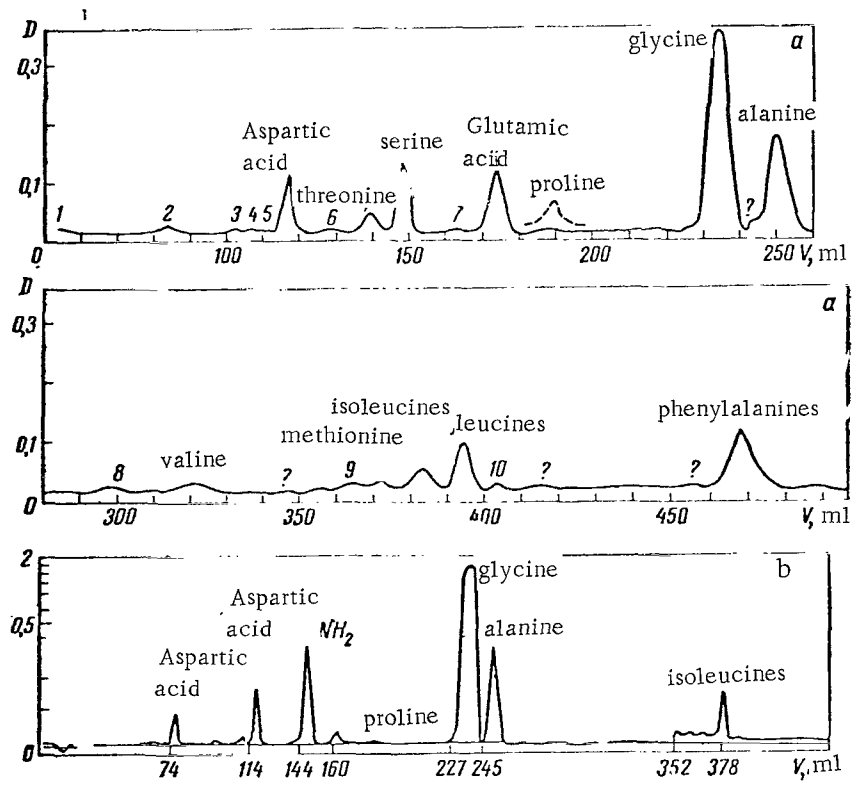


Figure 67. Chromatograms of Amino Acids from the Orgueil Carbonaceous Chondrite and Product Abiogenous Synthesis. a - acidic hydrolysate of amino acids for the Orgueil meteorite, extracted from meteorite samples weighing 7.30 g; absorption at λ = 440 mμ; dashed line represents absorption at λ = 570 mμ; temperature 50°; 1 - cysteic acid (?); 2 - urea; 3 and 4 - methionine sulfoxide; 5-10 - unidentified compounds (Vallentyne 1965); b - product of abiogenous synthesis, formed by the effect of electrical discharges on a mixture CH_4-C_2H_6-NH_3-H_2O (Oró, 1965a); D - absorption index; V - volume of eluates.

Anders, et al (Anders, DuFresne, et al,1964) investigated uncontaminated and obviously contaminated samples of the Orgueil meteorite. They demonstrated that the uncontaminated sample of this carbonaceous chondrite contained amino acids in approximately the same quantity as observed by Kaplan, et al (1963). However, in another sample of this meteorite, which was obviously contaminated /112 and contained a large quantity of modern pollen, Anders and his collegues found a considerably greater content of amino acids.

A sample of the Orgueil meteorite weighing 11.387 g was recently studied for its amino acid content by Vallentyne (1965). He used the chromatographic method for separating the hydrochloric acid hydrolysate in an amino acid analyzer (Figure 67). For the most part he studied the concent of acidic and neutral amino acids (the sum of free and bound acids). In the Orgueil meteorite he found the following amino acids (in μg/g): aspartic 1.3, glutamic 2.4, glycene 6.3, alanine 3.0, serine 1.5, traces of proline, valine 0.1, threonine 0.5, leucines 1.3, traces of isoleucines, traces of tyrosine (?), phenylalanine 3.9 traces of cystine and methionine. The total amount of amino acids in this meteorite was 20.4 Mg/g (0.38 μmol/g and, in addition, 3.0 μg/g urea. The amino acid extract was checked for the presence of optical rotation at wavelengths 436 and 546 mμ. The results were negative but they could be attributed to the small content of amino acids, below the sensitivity of the polarmeter.

The total content of amino acids in the Orgueil meteorite, according to Vallentyne's data, is below the quantity which was found in this meteorite by Kaplan and his associates. Vallentyne feels that the difference possibly can be attributed to the different research methods. For example, in Vallentyne's method, some of the amino acids (cystine, methionine, serine, threonine) can be up to 50% destroyed during the analytical process.

As a comparison with the Orgueil meteorite, Vallentyne investigated the distribution of amino acids in the ordinary Holbrook chondrite, and also in samples of dust and a sample of artificial rock on which fingerprints had been specially impressed. The amino acid content in these samples varied; in the dust samples it was 1,000 times greater than in the Orgueil meteorite.

Vallentyne was inclined to relate the formation of amino acids in meteorites to abiogenous synthesis in space. Laboratory abiogenous synthesis, as can be seen from Figure 67b, gives in part a similar picture of the distribution of amino acids, especially a high yield of glycene and alanine.

Nitrogenous cyclic compounds. Kaplan, et al (1963) combined carbohydrate extracts for a number of meteorites in checking for the presence of purine and pyramidine basis. The ultraviolet spectrum of these extracts (see Figure 66) with a pH 7 revealed the presence of weak absorption in the region 280 mμ, which disappeared when pH = 1. However, in the latter case a maximum appeared at 252 mμ. As noted by the authors, it is difficult to judge the presence of nitrogenous bases with assurance on the basis of these data.

The nitrogenous compounds in the Orgueil carbonaceous chondrite were recently the object of a special study by Hayatsu (1964); in contrast to Calvin (1961), he did not use ion-exchange resins. Hayatsu processed a meteorite

sample with 3 n. HCl, and then with acetic anhydride; the acetylized residue was extracted with butanol. The alcohol extract was broken down further. Infrared and ultraviolet spectroscopy confirmed that the fraction contained nitrogenous heterocyclic compounds. Then the fractions were separated by elution chromatography. The fractions were found to contain melamine-like substances. Then the eluates were studied by thin-layer chromatography. They were found to contain 7-8 compounds; among these, by comparison with 38 standards, it was possible to identify and quantitatively determine (with an accuracy of ±5 μg/g) the following nitrogenous compounds (in μg/g): melamine 35, ammeline 28, adenine 15, guanine 20. In all, Hayatsu extracted 1-2 mg of nitrogenous compounds from 16 g of this meteorite. He believes that this high content of nitrogenous compounds in the meteorite gives no basis for assuming that they originated from contamination. Moreover, this researcher points out that since the derivatives of triazine which he discovered are not biochemically important compounds, their presence in the Orgueil meteorite can indicate an abiogenous formation of nitrogenous organic compounds. 113

Hayes and Biemann (1967) studied the nitrogenous compounds in the Murray meteorite using a mass spectrometer with a high resolution. On the mass spectra these authors noted the probable appearance in the Murray meteorite of the following: picoline, alkylpyrroles, alkylpyridines (or anilines). Benzonitrile and indole (or methylbenzonitrile).

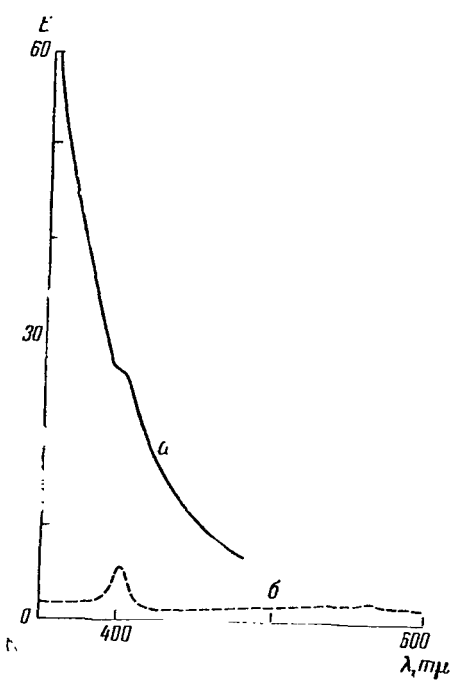


Figure 68. Spectrophotometric Curves of Benzene Eluate of Bituminous Matter from the Orgueil Meteorite (a) and Porphyrin Standard (b) (Hodgson, Baker, 1964). E - absorption; λ - wavelength.

Porphyrins. Many researchers have shown that petroleum and dispersed organic matter of rocks contain vanadic and nickeliferous porphyrins; chlorins have been identified in recent sediments. Bituminous substances with a high sulphur content are particularly enriched with vanadic porphyrins (Radchenko, Sheshina, 1955; Radchenko, 1960). Porphyrins and chlorins are products of the decomposition of hemin and chlorophyl.

The basis of the structure of porphyrins, a large porphin grain, consists of 45-member pyrrole rings, connected by carbon bridges.

Solutions of porphyrins on the spectrum revealed four absorption bands in the visible region of the spectrum and one in the ultraviolet region (Glebovskaya, Vol'kenshteyn, 1948; Pringsgeyn, 1951, and others).

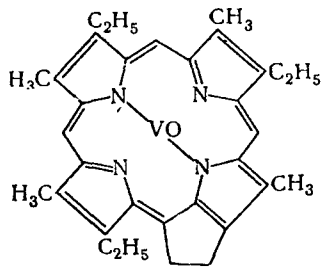
It was of interest to check carbonaceous chondrites for the presence of porphyrins in order to clarify the

nature of organic compounds in meteorites.

Hodgson and Baker (1964) investigated the organic matter in the Orgueil carbonaceous chondrite and demonstrated that it contains vanadic-porphyrinic complexes. They used samples of this meteorite weighing 21, 9 and 7 g for their study. These researchers chromatographically separated the bituminous matter in the Orgueil meteorite in a silica-gel column, the eluates were studied spectroscopically.

In the benzene eluates the authors noted a small absorption at $\lambda = 412$ mμ (Figure 68); they felt that this was caused by the presence of vanadic-porphyrinic complexes having a structure such as shown below (Nagy, 1966b).

Hodgson and Baker felt that the porphyrins in the Orgueil meteorite have a content of $0.010 \cdot 10^{-4}$% of the meteorite weight (with an accuracy to $\pm 0.003 \cdot 10^{-4}$%), or about $0.16 \cdot 10^{-4}$% of the weight of the carbonaceous matter. No chlorins were found in the meteorite The authors do not deny that the porphyrins could be the results of meteorite contamination. However, Mellor (1966) postulated that the absence of chlorins can be evidence that the porphyrins in the Orgueil meteorite has an abiogenous origin.



Kaplan, et al (1963) also checked for the presence of porphyrins in the organic matter of carbonaceous chondrites including the Orgueil meteorite, and could not detect porphinic pigments. The author (Vdovykin, 1965d, 1966b) made an unsuccessful attempt to find vanadic porphyrins in the Migei carbonaceous chondrite while making a study of an alcohol extract of this meteorite. The concentration of the extract from a sample of the Migei meteorite weighing 1.2435 g was 1.3 mg/ml. On the spectrum obtained in the visible region (see Figure 55) there were no noticeable absorption bands characteristic for porphyrins. Naturally, it is not impossible that the results could be effected by the small samples of initial matter. However, the presence of porphyrins in carbonaceous chondrites, as before, cannot be regarded as finally settled.

High-molecular organic matter. Recently some authors have investigated the products of oxidation or pyrolysis of high-molecular organic matter in meteorites by the mass-spectrometer method.

The results of study of the products of oxidation of polymeric matter in the Orgueil carbonaceous chondrite have been published by Bitz and Nagy (1966). They processed the polymeric organic matter extracted from 10.1 g of this meteorite with hydrogen peroxide and ozone. The products of oxidation were separated into fractions. In a mass-spectrometer study these fractions were found to contain benzenetri-, benzenetetra- and benzenepentacarboxylic acids, as well as pyridinearbonxylic and adipic acids.

Hayes and Biemann (1967) demonstrated that in the ordinary Holbrook chondrite the polymeric organic matter is present in a quantity 100 μg/g of the meteorite. Without extracting this substance from the meteorite, Hayes and Biemann carried out a pyrolysis of this matter in a vacuum. In a mass-spectrometer study the products of pyrolysis were found to contain for the most part aromatic hydrocarbons, but also compounds containing oxygen, nitrogen and sulphur. The authors feel (Hayes, 1967) that if some of the compounds containing sulphur could be formed during the course of pyrolysis during secondary reactions (for example, with FeS), compounds containing oxygen and nitrogen were initially present in the polymeric matter.

Study of Free Organic Radicals in Carbonaceous Chondrites by the Electron Paramagnetic Resonance Method

The electron paramagnetic resonance method is based on resonance absorption in the range of centimeter wavelengths; this is caused for the most part by the rotational movements of molecules and the magnetic properties of electron gels of some atoms and ions. This method makes it possible to detect some impurities in minerals and also to study free radicals, organic compounds having unshared electrons, which are responsible for their paramagnetic properities (Blyumenfel'd, Voyevodskiy, Seminov, 1962).

It has been made clear in many studies that when free radicals participate various processes of transformations of organic compounds transpire (Semenov, 1958, 1959). Free radicals are usually present in a reaction system for only a very short time. However, there are free radicals which persist for a long time. The stability of stable free radicals is related to a high degree of delocalization of unshared π-electrons within complex aromatic structures. Using the electron paramagnetic resonance method free radicals have been detected during recent years in various carbonaceous substances such as petroleums, coal, peat, etc.

Free radicals are not present in terrestrial objects alone. They have been found by spectroscopic studies in the sun and other stars, in comets and in interplanetary matter.

Urey and Donn (1956) postulated that in the primary matter of the solar system the concentration of free radicals was quite considerable; they could play some role in the heating of matter at the time of agglomeration of cosmic bodies. Under terrestrial conditions, even prior to the appearance of living systems on the earth, free radicals probably also had some importance in the chemical transformation of carbonaceous compounds (Vdovykin, 1963d). It was of definite interest to check meteorites for the presence of free radicals. In particular, this was pointed out by Bernal (1961a).

Duchesne and Depireux (1958) used the electron paramagnetic resonance method for studying the Aláis, Orgueil and Cold Bokkeveld carbonaceous chondrites. They took intact pieces of the meteorite without extracting the carbonaceous matter from them. However, the meteorites contain rather considerable concentrations of ferromagnetic particles, which, as is well known, produce a broad line on the electron paramagnetic resonance spectrum which

overlaps all the other possible absorption lines. The authors obtained these broad lines on spectra and the width of these lines (in oersteds) was: for the Aláis meteorite 2320, Orgueil 2250, Cold Bokkeveld 2460, with an absorption maximum for all samples at H_0 = 3100 oersteds and an approximate g-factor 2.19. Signals caused by the free radicals if they were present, were thus covered by the ferromagnetic absorption lines.

We repeated this observation using the Migei meteorite. On the electron paramagnetic resonance spectrum of a small piece of this meteorite we also found a broad absorption line (~2500 oe) with a maximum value H_0 = 3100 oe. It was caused by ferromagnetic particles in the meteorite, primarily by magnitite, for which absorption in this region is characteristic (Figure 69).

Therefore, later we investigated the high-molecular organic matter extracted from meteorites after being freed from the mineral components. Prior to this, the possibility of formation of free radicals during the course of extraction of carbonaceous matter was checked. Individual samples of humic matter, whose structure to some degree is similar to high-molecular organic matter in meteorites, processed in this way, failed to produce strong signals /116 on the electron paramagnetic resonance spectrum.

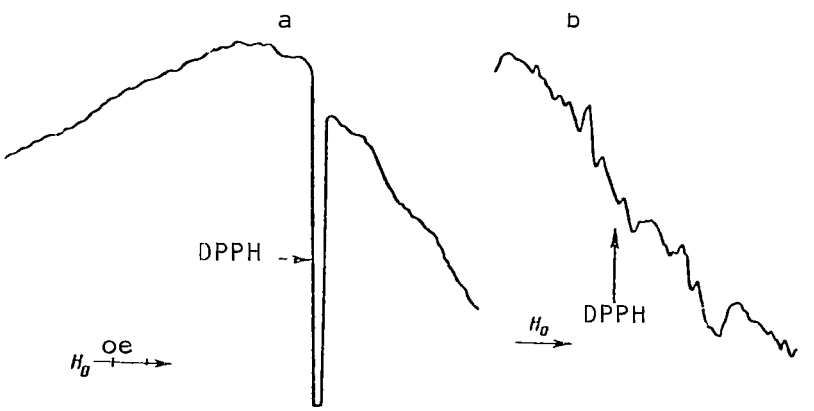


Figure 69. Electron Paramagnetic Resonance Spectra for Migei Carbonaceous Chondrite Without Extraction of Carbonaceous Matter (a) and Terrestrial Magnetite (b).

The measurements on the electron paramagnetic resonance spectrometer were made at room temperature. The capillaries which contain the matter being studied were made from quartz or molybdenum glass; they did not produce a noticeable signal on the electron paramagnetic resonance spectrum. The standard used was 1,1-diphenyl-2-picrylhydrazyl (DPPH), which in a polycrystalline state produces a strong narrow paramagnetic absorption line with a g-factor 2.0036.

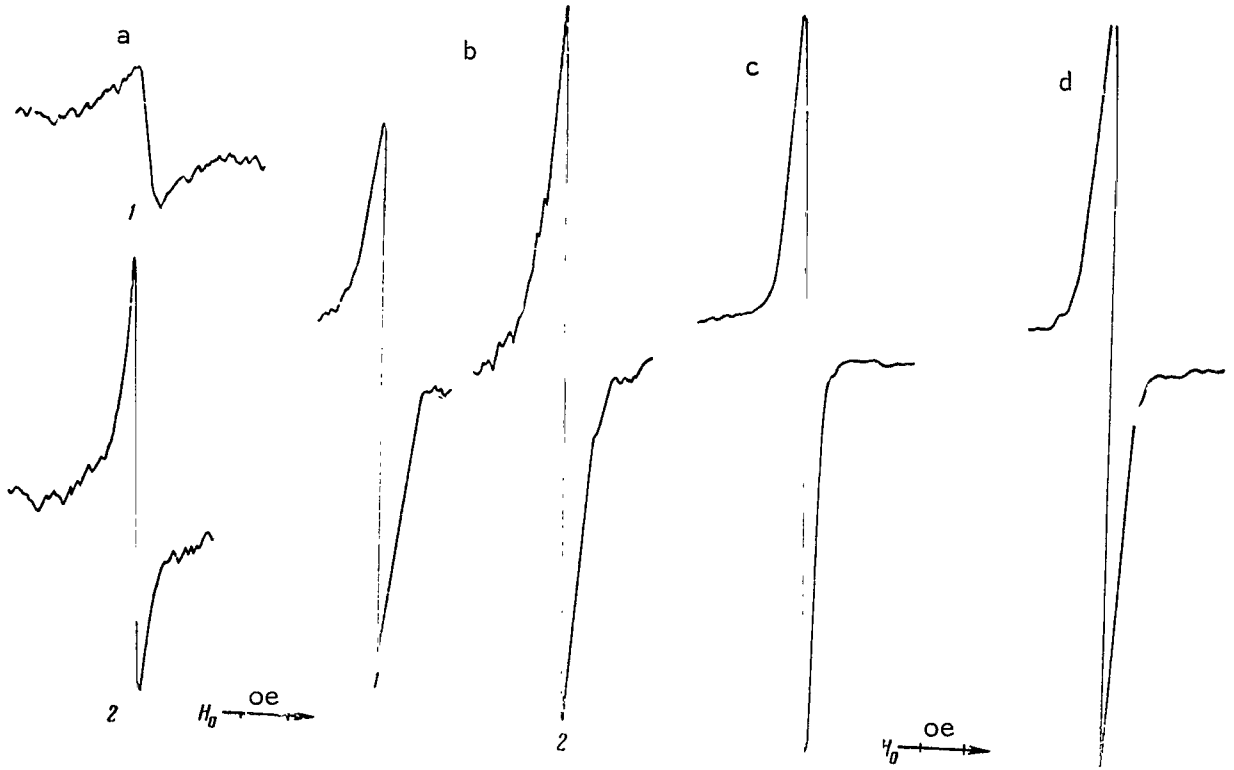


Figure 70. Electron Paramagnetic Resonance Spectra of High-molecular Organic Matter in Carbonaceous Chondrites.
a - Orgueil; b - Migei; c - Staroye Borinskino; d - Cold Bokkeveld;
1 - without standard; 2 - with DPPH.

In all the investigated samples of high-molecular organic matter from carbonaceous chondrites it was possible to detect stable free organic radicals (Vinogradov, Vdovykin, Marov, 1964, 1966; Vinogradov, Vdovykin, 1964; Vdovykin, 1965a). Figure 70 shows electron paramagnetic resonance spectra for carbonaceous matter from the Orgueil, Migei, Staroye Borinskino and Cold Bokkeveld carbonaceous chondrites. The spectra show rather strong paramagnetic absorption lines having similar parameters; their g-factor (2.002-2.003) is close to the g-factor for DPPH; $\Delta H \approx 6$ oe. This is evidence that in the complex aromatic structure of high-molecular organic matter the unpaired π-electrons are delocalized.

We made a detailed check of the free radicals for the Migei meteorite. Using six different initial samples of this meteorite, we successively extracted high-molecular matter and made a study with the electron paramagnetic resonance method. The spectra for all six extracted fractions had one rather strong absorption line whose parameters in all cases were similar, but also similar to the parameters of similarly extracted fractions of other investigated carbonaceous chondrites; the g-factor of these signals was close to the g-factor for DPPH. There was found to be a dependence between the signal amplitude being a function of the quantitative content of free radicals, on the initial weighed sample of high-molecular organic matter.

Then a study was made of the behavior of free radicals of high-molecular organic matter for the Migei carbonaceous chondrite as a function of heating. Heating was gradual, for one hour, from room temperature to a temperature of 256° for two individual weighed samples of this substance (tests 1 and 5)) placed in quartz ampules, filled with nitrogen and then sealed. In both cases ΔH the signal for the test samples present in the inert medium was the same as in the air.

The intensity of the electron paramagnetic resonance signals for sample 1 /119
decreased after heating to 150° With cooling to 23° the signal intensity increased somewhat, but after heating to 250° its intensity decreased insignificantly. This is evidence of a recombination of particles with an unshared electron. After heating to 250° the signal could not disappear and its ΔH increases somewhat, approximately to 8 oersteds.

Then this sample was extracted from the ampule and processed with HF-acid by a method similar to that which was used for extracting high-molecular organic matter from the meteorite. This was done for checking the possible formation of complex compounds during the course of processing the sample with acids. After processing this sample the signal on the electron paramagnetic resonance spectrum did not change, which does not confirm the possibility of formation of complex compounds in this process.

A similar study was made of the carbonaceous matter in the Migei meteorite extracted from another sample, test 5 (Figure 71). A recording of the electron paramagnetic resonance signals was made continuously each 0.5 minute. In the course of heating to 150-200° the signal amplitude decreases, but at a higher temperature it remains virtually constant. After heating to 256°

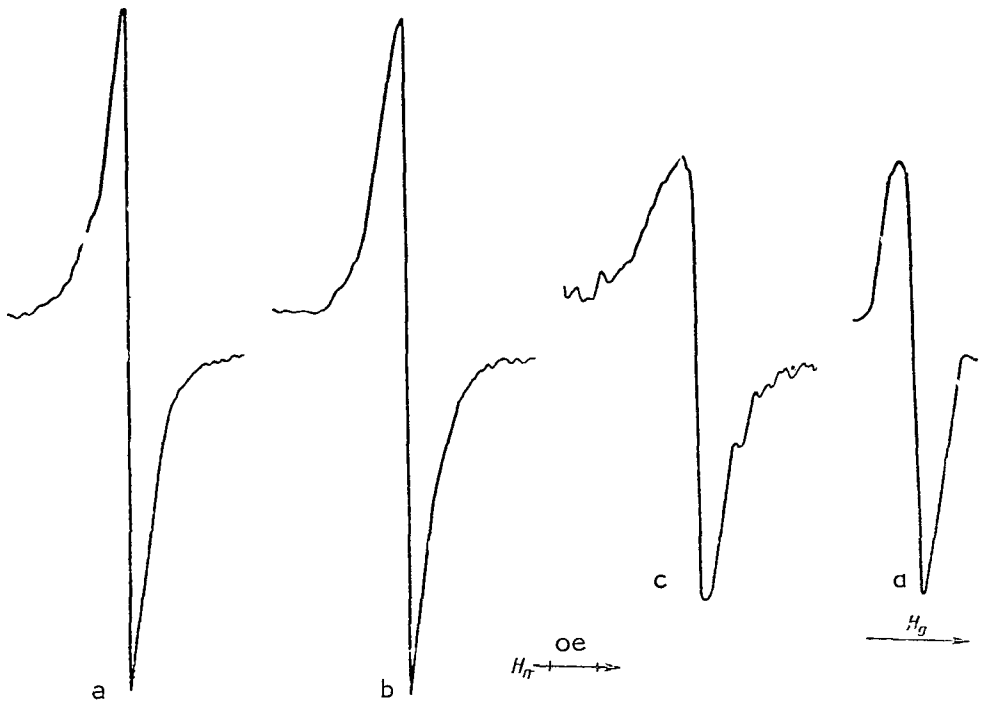


Figure 71. Electron Paramagnetic Resonance Spectra of High-molecular Organic Matter in the Migei Meteorite, Obtained Under Various Conditions.
a - in the air, 22°; b - in a nitrogen atmosphere, 22°; c - after heating in a nitrogen atmosphere to 256° and subsequent cooling to 24°; d - after neutron irradiation.

ΔH for the signal was, as in test 1, about 8 oersteds. This sample was irradiated by neutrons, but during the course of irradiation the ampule was broken and therefore it is difficult to say anything about the character of the possible change in the signal of this sample irradiated in the air.

The bituminous matter in the Migei, Staroye Borinskino, Murray, Cold Bokkeveld and Groznaya carbonaceous chondrites was also checked for the presence of free radicals. However, the spectrum did not reveal clear paramagnetic absorption lines. Three radicals in the bituminous matter were either absent or their content at the concentration of the bituminous matter in the CCl_4 solution of 0.11 mg/ml was below the sensitivity of the apparatus (10^{13} spins).

As a comparison with the organic matter of meteorites a study was made of various terrestrial carbonaceous formations: petroleums of different age from the northwestern Ciscaucasia and Volga region, samples of coals from the Don Basin and the Moscow area brown coal basin, peat from Shatura and Belorussia and combustible shale from Estonia and other samples.

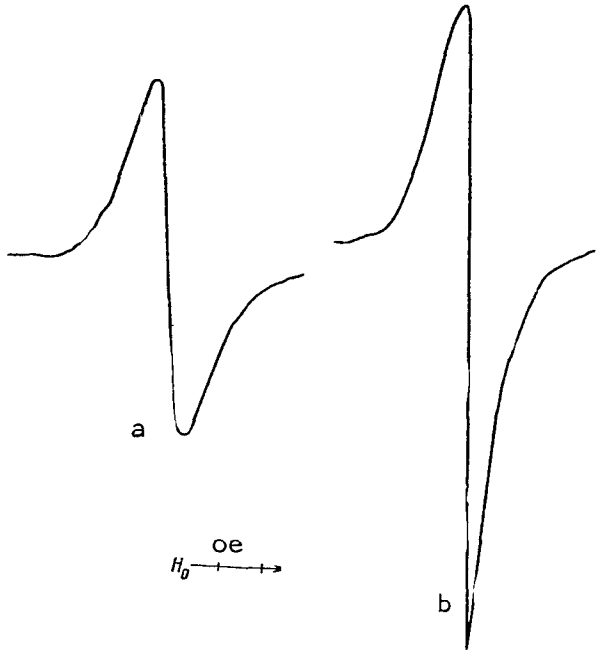


Figure 72. Electron Paramagnetic Resonance Spectra of PZH-Brand Coal (Don Basin).
a - without DPPH; b - with DPPH.

As is well known, the minerals making up coal usually have a condensed aromatic structure. The high-molecular organic matter in meteorites apparently has a structure to some extent similar to it. The presence of free radicals in coals is also well known. On the other hand, in petroleum the free radicals are associated with asphaltenes, condensed aromatic systems.

Figure 72 and 73 show electron paramagnetic resonance spectra of some coal samples which we studied (Figure 72), as well as petroleums, and also bituminous matter from a diabase dike in Siberia (Figure 73). The electron paramagnetic resonance signals of these samples resemble in their parameters the electron paramagnetic resonance spectra of high-molecular organic matter in meteorites.

It is assumed that in coals, peats, and shales the free radicals have a character inherited primarily from living matter. The nature of the free radicals in petroleums has not been clarified sufficiently well. It can be mentioned in passing that according to our studies, the formation and behavior of free radicals in petroleum is influenced by the physical chemical conditions of the environment in which petroleum developed (Vdovykin, Dubrov, Marov, 1967). The petroleums existing in deep horizons were affected by the degree of radioactive irradiation, heating, etc. For another group of petroleums, lying at a relatively shallow depth, the concentration of free radicals decreases with the depth at which the petroleum lies. This concentration in general is in direct dependence on the specific gravity of the petroleum /121

(Figure 74) and its content of asphaltenes, but in inverse dependence on the content of fractions in the petroleums with a low boiling point. In addition, for the petroleums of the Maykop suite in the northwestern Ciscaucasia there is an inverse dependence between the concentration between free radicals in the petroleums and the content of bituminous compounds dissolved in the stratal waters of these same samples (Vdovykin, 1962a; Figure 75).

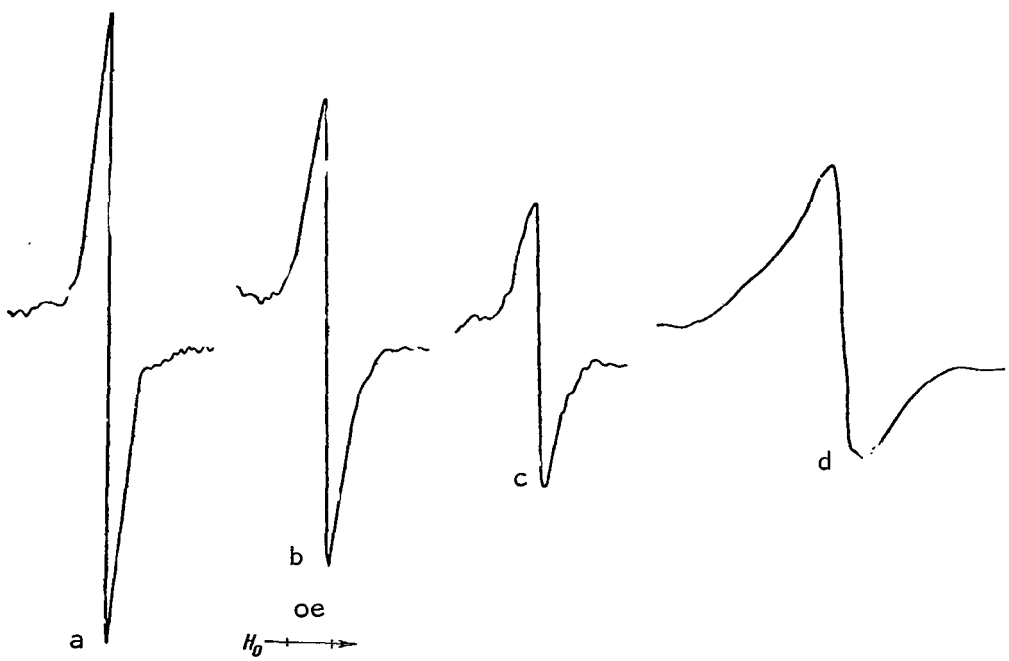


Figure 73. Electron Paramagnetic Resonance Spectra of Some Bituminous Substances.
a - petroleum of the Neftegorskoye deposit from northwestern Ciscaucasia, hole 574, Maykop VI; b - petroleum from the Anastas'yevsko-Troitskoye deposit in northwestern Ciscaucasia, hole 571, Maykop VI; c - petroleum from the Korobovskoye deposit (lower Volga region), hole 190, Bobrikovskiy horizon; d - bituminous matter from diabase dike, Siberia, Tunguska River.

The literature contains reference to numerous experiments which show that the concentration of free radicals increases during irradiation; it is also known that free radicals in different organic compounds are formed under various kinds of irradiations. The author used humic matter for irradiation. Prior to irradiation it did not produce a strong signal on the electron paramagnetic resonance spectrum. After irradiation by neutrons over a period of two weeks ($E = 0.025$ eV, $I = 1.2 \cdot 10^{13}$ neutrons/cm^2·sec) the electron paramagnetic resonance spectrum revealed a rather strong signal (Figure 76) which

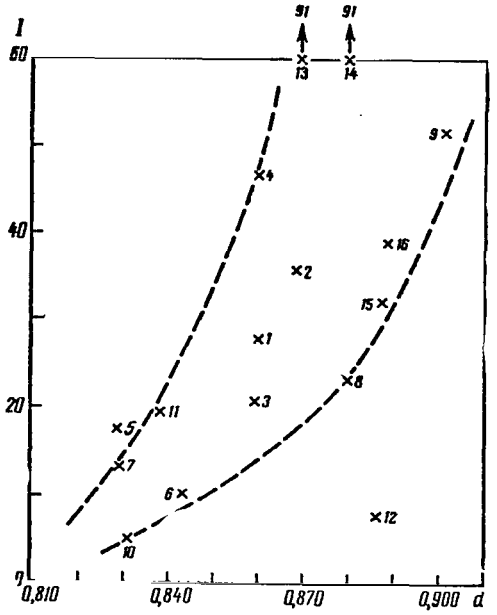


Figure 74. Dependence of Relative Intensity (I) of Electron Paramagnetic Resonance Signal on Specific Gravity (d) of Petroleums from Maykop Suite in Northwestern Ciscaucasia.
1 - Khadyshenskoye deposit, hole 169, Maykop I; 2 - same, hole 202, Maykop II; 3 - same, hole 166, Maykop III; 4 - Central field, hole 657, Maykop IV; 5 - same, hole 574, Maykop, VI; 6 - same, hole 285, Maykop, VI; 7 - same, hole 265, Maykop, VI; 8 - Khopry, hole 586, Maykop, V; 9 - Pablova Gora, hole 797, Maykop, I; 10 - same, hole 688, Maykop, I; 11 - same, hole 668, Maykop, VII; 12 - Kadyshenskaya area, hole 535, Maykop, II; 13 - same, hole 179, Maykop, III; 14 - same, hole 195, Maykop, III; 15 - Akhtyrsko-Bubundyrskoye deposit, hole 262, Maykop, 16 - Neftegorsk, kir (solidified petroleum), Maykop, IV.

was identical in its parameters to the electron paramagnetic resonance signals of polymeric matter in carbonaceous chondrites.

In the first approximation these data do not give a basis for denying the hypothesis that in meteorites, whose high-molecular organic matter has a structure partially similar to the structure of humic substances, the free radicals could be formed as a result of irradiation to which the meteorites were subjected in space (Vinogradov, Vdovykin, 1964; Vdovykin, 1966).

Independently and simultaneously with our studies, a study of the free radicals in a number of carbonaceous chondrites was made by Duschesne and his associates. They established the presence of free radicals in the Aláis and Cold Bokkeveld meteorites (Villée, Duchesne, Deprieux, 1964). These authors used a highly sensitive electron paramagnetic resonance spectrogram for studying samples of meteorites (weighing about 1 g) in intact form, without extracting the carbonaceous matter. Against the background of broad lines caused by virile magnetic particles, the electron paramagnetic resonance spectrum revealed weak signals which were evidence of the presence of three organic radicals (Figure 77). The width of these signals was < 10 oersteds; the value of the g-factor for the Cold Bokkeveld meteorite (~1.96), as the authors note, was too low because of an internal field caused by the ferromagnetic particles; in actuality, it was close to 2.0023. The concentration of free radicals in the Aláis meteorite, according to the data published by these authors, is approximately 10^{16} centers per gram of carbonaceous matter. It attains the same value, as the authors later demonstrated (Duschesne, Depireux, Litt, 1964a), in the Cold Bokkeveld meteorite. Duschesne investigated the behavior of the electron paramagnetic resonance signal in the Cold Bokkeveld

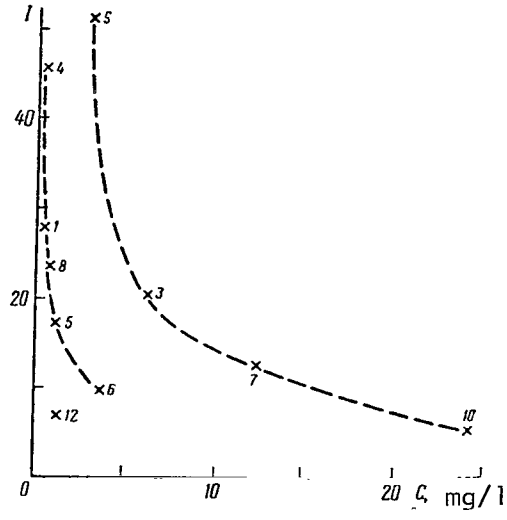


Figure 75. Dependence of Relative Intensity (I) of Electron Paramagnetic Resonance Signal for Petroleum on the Content of Water-Soluble Bituminous Matter (C) in Waters Underlying Petroleum for the Maykop Suite in the Northwestern Ciscausia (number of samples -- see annotation to Figure 74.

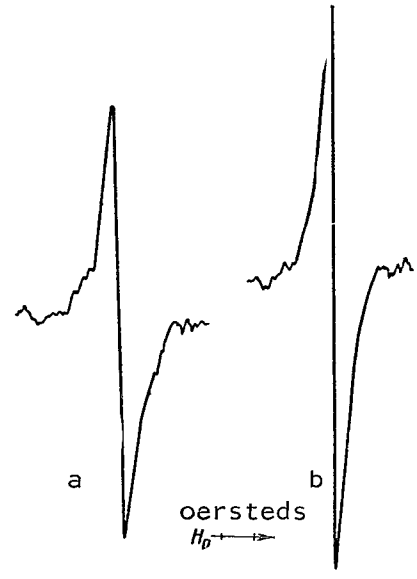


Figure 76. Electron Paramagnetic Resonance Spectra of Humic Matter Irradiated by Neutrons.
a - without standard; b - with standard (DPPH).

meteorite in a vacuum and after irradiation and demonstrated that for a meteorite sample exposed to a vacuum there is some narrowing of the signal, but after meteorite irradiation by X-rays the signal amplitude increases; the radiation output, reduced to the carbon part, is 10^{-4} (Figure 77b).

Duschesne, et al (Duschesne, Depireaux, Litt, 1964b) also established the presence of free organic radicals in the Migei and Nogoya carbonaceous chondrites. The electron paramagnetic resonance signal was similar to the signals for other meteorites The authors estimated the concentration of free radicals in the Migei meteorite at 10^{17} and in the Nogoya meteorite at approximately 10^{15} centers per gram. In the study of the Migei meteorite in a vacuum these authors noted that there was some narrowing of the electron paramagnetic resonance signal. /122

The parameters of the electron paramagnetic resonance signals of meteorites and the peculiarities of behavior of the signals under various conditions made it possible for Duschesne to postulate that the electrons responsible for the signals were delocalized, apparently in complex aromatic structures similar to the structure of humic substances.

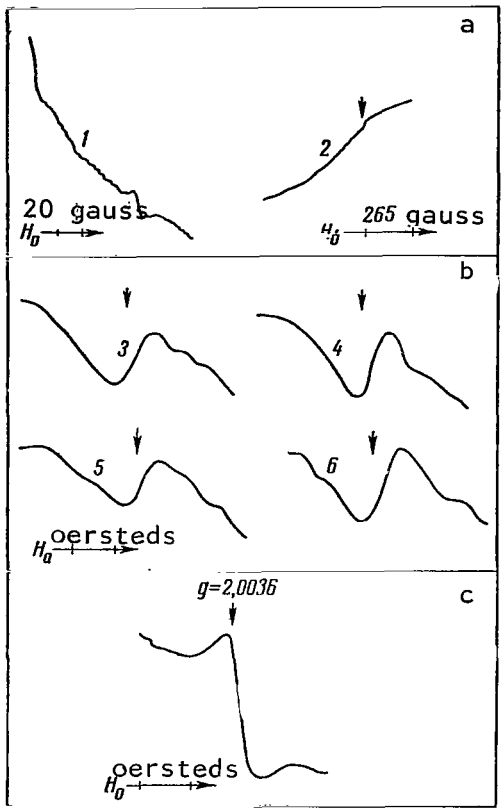


Figure 77. Electron Paramagnetic Resonance Spectra of Carbonaceous Chondrites (from data in the literature).
a - electron paramagnet resonance spectra of meteorites; 1 - Aliás, 2 - Cold Bokkeveld; the arrow corresponds corresponds to g ~1.96 (Villée, et al, 1964); b - electron paramagnet resonance spectra of Cold Bokkeveld meteorite; 3 - in air; 4 - in vacuum; 5 - prior to irradiation; 6 - after irradiation by X-rays; arrows correspond to g = 2.002 (Duschesne, et al, 1964a); c - electron paramagnet resonance spectrum of Migei meteorite; arrow corresponds to g = = 2.0036 (Duschesne, et al, 1964b).

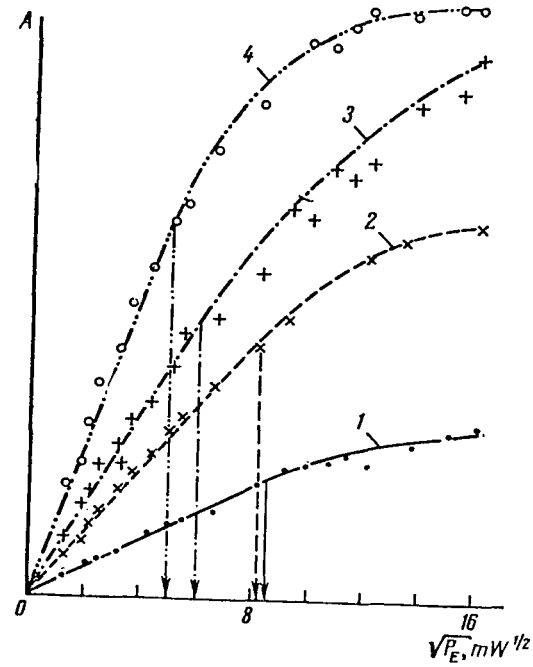


Figure 78. Curves of Saturation of Electron Paramagnetic Resonance Signals of Migei Meteorite and Terrestrial Carbonaceous Substances (Duschesne, et al, 1965).
1 - meteorite; 2 - coal; 3 - lignite; 4 - saccharose. The arrows correspond to the region of appearance of saturation. A - ratio of signal amplitude to weighted sample (in g).

Duschesne, et al (Duschesne, Cornil, et al, 1965) made a comparative study of the effects of saturation of electron paramagnetic resonance signals. They demonstrated that for the meteorite and coal the saturation virtually coincides, but differ from the saturation for saccarose. The saturation for lignite occupies an intermediate position between coal and saccharose (Figure 78). This was also confirmed by the similarity of structures of carbonaceous matter in meteorites and coal.

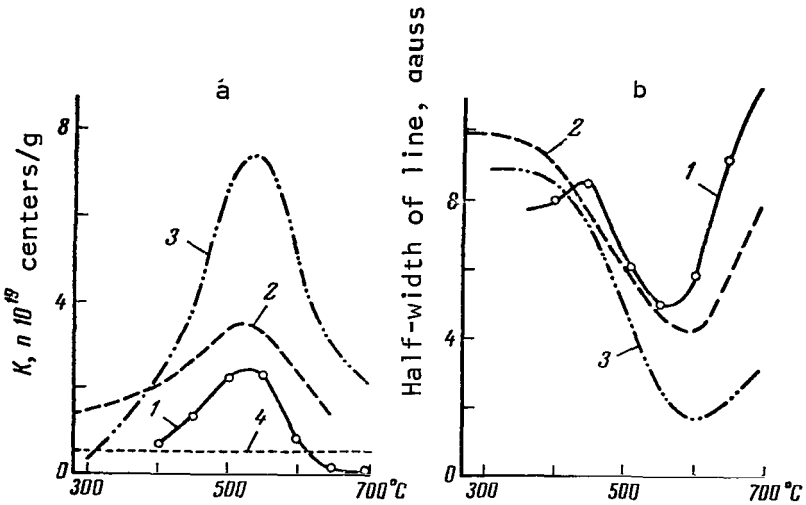


Figure 79. Dependence of Concentration of Free Radicals (a) and Width of Electron Paramagnet Resonance Signal (b) on Carbonization Temperature for High-molecular Organic Matter of the Orgueil Meteorite and Terrestrial Carbonaceous Substances (Schulz, Elofson, 1965).
1 - meteorite; 2 - coal (89% C); 3 - saccharose; 4 - graphite.

On this basis Duschesne (1965) postulated that the free radicals in meteorites, like the free radicals in coals, are caused by biogenitic processes occurring on the parent body from which meteorites originated. However, Urey (1966a) correctly noted that free radicals of a similar nature can also arise in other ways without any participation of life.

The free organic radicals in carbonaceous chondrites recently have been studied by other researchers. Schulz and Elofson (1965) discovered free radicals in the Orgueil meteorite. In their study they did not use the meteorite in intact form, like Duschesne and his fellow workers, but only the high-molecular fraction of the organic matter extracted after being freed from the mineral components. The electron paramagnetic resonance spectrum of this fraction revealed a rather strong signal with a width of 6.1 gauss with a g-factor of 2.0025. The authors estimate the concentration of free radicals at /124
1.3·10 centers per gram.

Schulz and Elofson studied the behavior of free radicals of organic matter in this meteorite when it was carbonized during the course of heating in a vacuum from 400 to 700° and noted a rather similar behavior of the electron paramagnetic resonance signals for the high-molecular organic matter in the Orgueil meteorite and coal (Figure 79).

In a report at the Twentieth International Congress of Theoretical and Applied Chemistry Schieldkancht (1965) reported on the free organic radicals in the Essebi carbonaceous chondrite which recently (1957) fell. Schieldkancht demonstrated that the behavior of the free radicals during carbonization of the high-molecular organic matter in the Essebi meteorite was very similar to the behavior of radicals in the Orgueil meteorite which had been studied by Schulz and Elofson.

Thus, free organic radicals have been found in a number of carbonaceous chondrites. They are apparently present in all meteorites of this type. The presence of free radicals in carbonaceous chondrites also is confirmed by other research results which show that the greater part of the carbonaceous matter in these meteorites is high-molecular organic matter. The stability of the free organic radicals can be attributed to delocalization of unshared π-electrons in complex aromatic structures of the high-molecular organic matter in meteorites.

The free radicals were possibly formed as a result of irradiation in space to which the meteorites were subjected while in space. In the future, with a more detailed study of the free radicals in carbonaceous chondrites, they may be used in judging the degree of cosmic irradiation of meteorites. However, another possibility is equally probable; the formation of free radicals under the conditions prevailing on the parent bodies from which meteorites originated. In the latter case they can be evidence of the presence of some energy source during the transformation of organic compounds.

CHAPTER FIVE

STRUCTURAL ANALYSIS OF CARBONACEOUS MATTER IN CARBONACEOUS CHONDRITES

For a more precise determination of the phase composition of carbonaceous matter, determination of the degree of crystallization of carbonaceous particles and other parameters of carbonaceous matter in carbonaceous chondrites we made a roentgenometric analysis, an electron diffraction and microdiffraction study, as well as a thermal analysis. The author studied the Kainsaz chondrite for comparison with carbonaceous chondrites.

Roentgenometric Analysis

X-ray methods of structural analysis are based on the phenomenon of X-ray diffraction. These methods make use of the special properities of X-rays; their great penetrating capacity and short wavelength (10^{-10}-10^{-6} cm). Several roentgenometric methods for studing matter are known: X-ray spectral micro-analysis, X-ray structural analysis, roentgenometric (X-ray phase) analysis, and others. The latter method is widely used. It makes it possible to identify matter, make a qualitative and sometimes a quantitative phase analysis of mixtures, and determine the size of a unit cell and the degree of crystallinity, which is especially important in a study of organic polymeric compounds.

The roentgenometric method has been used by many researchers in studying various terrestrial carbonaceous compounds: Schungite (various soots, coals, kerogen and fractions of bituminous matter. The author used the roentgenometric method in a study of the bituminous matter in the Groznaya carbonaceous chondrite and fractions of high-molecular carbonaceous matter in the Migei, Staroye Boriskino, Cold Bokkeveld, Murray and Groznaya carbonaceous chondrites. As a comparison the author studied the carbonaceous matter in the Kainsaz chondrite which comtains carbon. The survey was made eith a Fe anticathode. Figure 80 shows some X-ray photographs with chloroform with Groznaya carbonaceous chondrite for making a roentgenometric analysis.

A bituminen-like substance, after extraction from carbonaceous chondrite and removal of the solvent, crystallizes into discoid or acicular crystals up to 0.3 mm in diameter. For the purpose of X-ray analysis, this substance was extracted from the Groznaya carbonaceous chondrite with chloroform.

The X-ray photograph revealed 54 lines (Figure 80, Table 29). The strongest lines were 3.83, 3.12, 2.113 and 1.909 Å. The X-ray photograph of this substance is comparable to the X-ray photographs of ozercerite (Mikheyev, 1957) and organic minerals of hydrothermal origin: evenkite (Mikheyev, 1957) and hatchettine (Giller, 1955; Giller, Spitkovskaya, 1963).

Some of the information concerning the structure of the high-molecular fraction of carbonaceous matter in carbonceous chondrites has been contradictory. L. G. Kvasha (1948) found it to be present in the Staroye Borinskino meteorite, /126

defining it as graphite. Mueller (1953), in a roentgenometric study of carbonaceous matter extracted from the Cold Bokkeveld meteorite noted that the X-ray photograph revealed no lines characteristic for graphite. V. I. Mikheyev and A. I. Kalinin (1958), in a roentgenometric study of the Migei meteorite in intact form, without extraction of carbonaceous matter, also found no lines on the X-ray photograph which were characteristic for graphite. In their opinion, the carbonaceous matter of this meteorite was amorphic coallike matter.

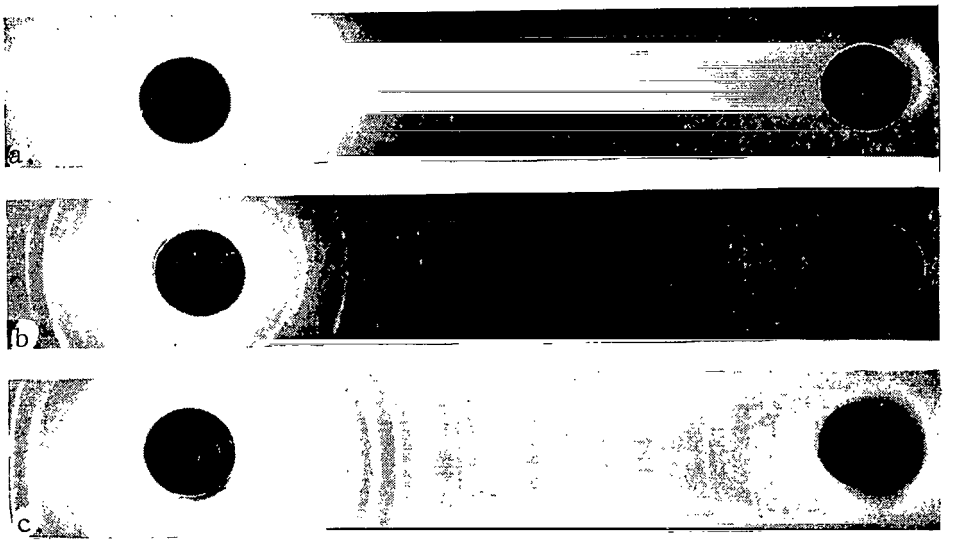


Figure 80. Some X-ray Photographs of Carbonaceous Substances Extracted from Chondrites.
a - tiny crystals of bituminous matter extracted with chloroform from the Groznaya carbonaceous chondrite; b - high-molecular fraction of carbonaceous matter from the Migei carbonaceous chondrite; c - high-molecular fraction of carbonaceous matter from the Kainsaz chondrite, which contains carbon.

The author used the roentgenometric method for studying the high-molecular fraction of carbonaceous matter in the Migei, Staroye Borinskino, Cold Bokkeveld, Murray and Groznaya meteorites after extracting it from the meteorite by means of careful decomposition of the mineral components. The X-ray photographs of these fractions of carbonaceous chondrites are rather similar to one another (Table 30). They are comparable to the X-ray photographs of terrestrial humic substances (Sedletskiy, 1937; Kashatochkin, et al, 1956, 1958). Evidently, the overwhelming part of this fraction is organic matter having a condensed aromatic structure which is also characteristic for terrestrial humic substances.

The X-ray photograph of the carbonaceous matter extracted from the Kainsaz chondrite, which contains carbon (Table 31), not only shows lines characteristic for carbonaceous matter, but clearly expressed lines for graphite. In the

carbonaceous matter of this meteorite the carbon atoms are more ordered than in the carbonaceous matter of carbonaceous chondrites.

It is assumed that in chondrites of other types graphite is the principal component of their carbonaceous matter. In the remnants from decomposition of meteorites it is found in a mixture with chromite and other minerals which are not destroyed by acids (Ponomarev, 1951; Hoops, 1964). Sarma and Mayeda (1961) used the roentgenometric method and found graphite in a mixture with chromite in a number of ordinary chondrites. A. F. Sandrea (1965) observed graphite in petrographic of the Saint Sauveur enstatic chondrite.

Electron Diffraction and Microdiffraction Studies

By using the electron microscope and electron diffraction methods it is possible to study the morphology and structure of very tiny inclusions of various nature. The principles of these methods and many results obtained using them have been described in monographs by Z. G. Pinsker (1949), B. K. Vaynshteyn (1956), B. M. Lyk'yanovach (1960), G. S. Gritsayenko, et al (1961), B. B. Zvyagin (1964), and others. /128

In a study of the structure of finely dispersed substances the most promising instrument is the use of the 400-kV electron microscope-electron diffraction camera constructed by N. M. Popov (1959). This instrument combines the electron microscope and the electron diffraction method. With an increase in the energy of electrons to relativistic velocities (600 keV) the admissible thickness of the particles increases in comparison with the ordinary electron microscope and there is an increase in selectivity when obtaining the diffraction spectrum from an observed particle. Highly selective microdiffraction, involving the use of high-energy electrons, has been called microdiffraction of ultrafast electrons by N. M. Popov (1959; Popov, Svyagin, 1958, 1959, and others). With this instrument it is possible to study objects with a thickness up to 1 μ with a microdiffraction selectivity to 0.05 μ. /129

Figure 81. Electron Diffraction Pattern of Bituminous Matter in the Groznaya Carbonaceous Chondrite (obtained using a 400-kV electron microscope-electron diffraction camera).

It becomes possible to study smaller inclusions than with an ordinary electron microscope or study individual parts of inclusions separated out using a special selector diaphragm.

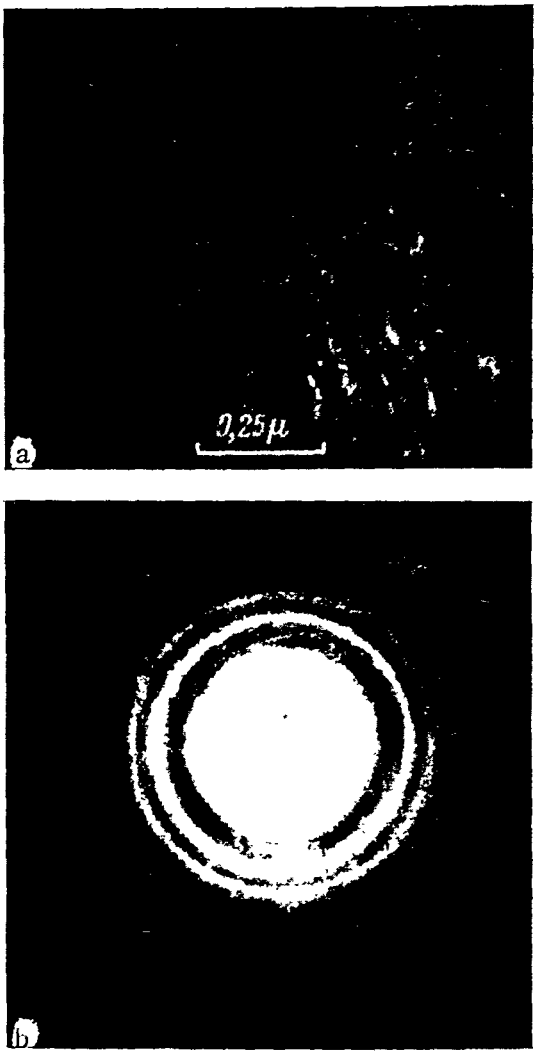


Figure 82. Electron Microscope Image (a) and Diffraction Spectrum (b) of High-molecular Organic Matter in the Orgueil Carbonaceous Chondrite (obtained using 400-kV electron microscope-electron diffraction camera).

N. M. Popov, V. I. Kasatochkin and V. M. Luk'yanovach (1960) used this method in studying the structure of soot particles. They demonstrated that in graphitized soot, between the individual units there are intercalations of nongraphitized carbonaceous matter of an amorphous nature. The atomic-molecular structure of these particles, in the opinion of the authors is similar to the structure of lustrous carbon, but differs in the symmetry of packing of plane carbon lattices.

The author (Vdovykin, 1964d) first studied the carbonaceous matter in meteorites using an ordinary EM-7 electron microscope. In this way he confirmed roentgenometric data indicating that the greater part of the carbonaceous matter in carbonaceous chondrites is represented by a high-molecular organic substance; it had an amorphous and crystalline structure.

A. P. Vinogradov, G. P. Vdovykin and N. M. Popov (1965) made a more de- /130
tailed study of the structure of different forms of carbonaceous matter in a number of meteorites, including carbonaceous chondrites, using the 400kV electron microscope-electron diffraction camera. The study was made using the electron diffraction and micro-diffraction methods. The bituminous matter found in the Groznaya carbonaceous chondrite and the high-molecular part of the carbonaceous matter found in the Orgueil, Migei, Staroye Borinskino and Cold Bokkeveld carbonaceous chondrites were studied.

The preparations were dispersed in distilled water by the ultrasonic method at a frequency of 1 mc/sec and then applied to a film base. Aluminum was used as a standard. About 300 photographs were taken using this instrument, thereby making it possible to select the most typical photographs.

Table 29

X-Ray Photographs of Bituminous Matter of the Groznaya Carbonaceous Chondrite and Terrestrial Organic Minerals

Bituminous Substance in Graznaya Meteorite		Ozocerite		Evenkite		Hatchettine	
I	$\frac{d_a}{n}$	I	$\frac{d_a}{n}$	I	$\frac{d_a}{n}$	I	$\frac{d_a}{n}$
2	4,33	10	4,26	—	—	7	(4,65)
2	4,10	—	—	10	4,18	—	—
10	3,83	8	3,30	—	—	—	—
1	3,78	—	—	9	3,74	8	3,79
1	3,61	—	—	—	—	—	—
5	3,50	2	3,41	—	—	—	—
4	3,34	—	—	1	3,35	2	(3,32)
7	3,25	—	—	—	—	—	—
8	3,12	4	3,07	5	3,02	4	3,01
8	2,850	3	2,809	—	—	—	—
1	2,783	—	—	3	(2,783)	3	(2,761)
1	2,709	—	—	—	—	—	—
6	2,635	—	—	1	2,627	—	—
1	2,589	—	—	—	—	1	(2,606)
6	2,515	6	2,527	7	2,515	6	2,508
6	2,443	—	—	2	2,461	—	—
6	2,365	—	—	4	2,415	4	2,371
4	2,302	5	2,270	8	2,245	—	—
1	2,219	5	2,244	—	—	6	2,237
1	2,172	3	2,147	3	2,150	4	2,141
9	2,113	4	2,099	6	2,124	—	—
1	2,072	—	—	3	2,079	4	2,076
2	2,000	—	—	3	2,015	1	1,984
2	1,944	4	1,945	—	—	—	—
8	1,909	2	1,901	5	1,914	3	(1,927)
4	1,833	—	—	3	1,866	3	1,871
6	1,784	—	—	—	—	1	(1,788)
3	1,760	5	1,745	6	1,751	5	1,751
5	1,715	—	—	1	1,718	—	—
4	1,706	4	1,678	—	—	2	1,697
2	1,656	—	—	5	1,658	2	(1,666)
3	1,648	—	—	—	—	—	—
3	1,621	—	—	3	1,623	3	1,621
2	1,613	4	1,619	—	—	—	—
2	1,563	—	—	1	1,571	—	—
3	1,536	—	—	—	—	—	—
1	1,510	4	1,510	4	1,515	3	1,515
1	1,476	—	—	2	1,460	—	—
4	1,444	3	1,443	—	—	—	—
5	1,422	—	—	2	1,426	2	(1,426)
2	1,394	3	1,388	3	1,386	3	1,382
6	1,353	—	—	—	—	—	—
1	1,304	2	1,300	4	1,299	3	1,295
4	1,283	3	1,277	—	—	2	1,280
1	1,249	2	1,243	4	1,243	—	—
3	1,232	—	—	—	—	2	1,239
1	1,207	4	1,211	4	1,214	3	1,215
2	1,169	—	—	—	—	—	—
2	1,150	—	—	4	1,116	3	1,107
1	1,095	3	1,105	2	1,102	2	1,092
1	1,056	—	—	—	—	—	—
1	1,031	—	—	3	1,026	—	—
2	1,012	—	—	—	—	—	—

The carbonaceous matter of the Pilistere enstatic chondrite, according to the roentgenometric data obtained by the author of this book, is graphite.

Table 30

X-Ray Photographs of Fractions Containing High-Molecular Carbonaceous Matter in Carbonaceous Chondrites

Migei		Staroye Boriskino		Cold Bokkeveld		Murray		Groznaya	
I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$
1	7,68	1	7,49	—	—	—	—	1	7,28
1	5,66	10	5,72	10	5,66	—	—	10	5,72
1	5,14	—	—	4	4,92	4	4,92	—	—
3	4,68	4	4,88	—	—	—	—	4	4,88
10	3,91	5	3,85	7	3,85	6	3,88	4	3,90
2	3,66	3	3,57	4	3,54	2	3,51	4	3,59
1	3,34	7	3,32	3	3,34	4	3,32	8	3,32
1	3,16	3	3,21	5	3,21	3	3,23	—	—
10	3,09	10	3,09	10	3,06	10	3,07	10	3,07
1	2,921	3	2,850	4	2,836	3	2,850	3	2,850
2	2,658	—	—	1	2,646	1	2,670	—	—
1	2,423	2	2,356	—	—	2	2,347	2	2,347
4	2,227	9	2,227	9	2,227	8	2,211	9	2,227
1	2,212	3	2,135	—	—	2	2,120	2	2,135
4	2,072	4	2,078	4	2,079	4	2,085	3	2,072
—	—	6	1,968	2	2,037	3	2,011	6	1,968
3	1,950	5	1,944	2	1,944	—	—	—	—
5	1,828	10	1,813	—	—	—	—	9	1,824
5	1,760	—	—	9	1,760	—	—	—	—
4	1,693	1	1,693	2	1,689	1	1,693	1	1,695
1	1,545	6	1,546	4	1,556	5	1,566	5	1,556
—	—	—	—	5	1,529	4	1,522	5	1,522
2	1,513	3	1,519	4	1,516	—	—	—	—
—	—	3	1,456	4	1,458	3	1,458	3	1,456
—	—	2	1,436	2	1,428	4	1,425	—	—
1	1,375	9	1,384	9	1,389	9	1,386	10	1,384
—	—	1	1,348	1	1,348	1	1,348	—	—
2	1,298	—	—	2	1,309	—	—	—	—
3	1,296	1	1,288	1	1,288	1	1,288	1	1,286
1	1,236	3	1,228	1	1,230	—	—	3	1,230
1	1,145	1	1,145	1	1,130	—	—	1	1,130
2	1,101	2	1,101	1	1,101	2	1,100	2	1,101
1	1,079	1	1,073	2	1,077	—	—	1	1,065
1	1,051	1	1,057	—	—	1	1,055	1	1,057
2	1,010	—	—	2	1,009	1	1,014	—	—

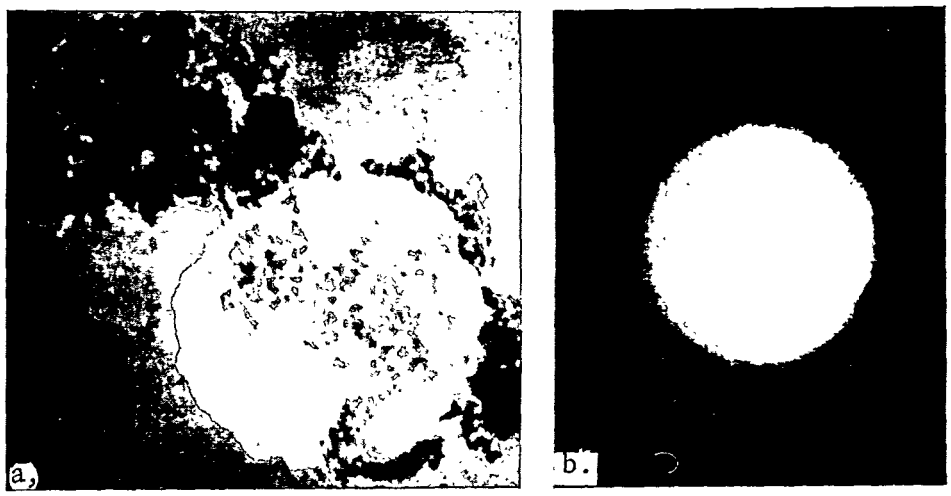


Figure 83. Bright-Field Image (a) and Diffraction Spectrum (b) of the Morphic High-Molecular Organic Matter in the Cold Bokkeveld Carbonaceous Chondrite (effective diameter of selector diaphragm 1 μ; obtained using 400-kV electron microscope-electron diffraction camera).

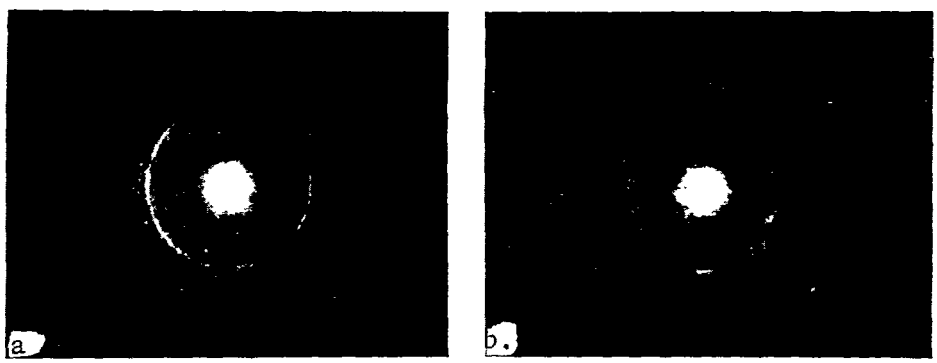


Figure 84. Electron Diffraction Pattern of High-Molecular Organic Matter in the Migei Carbonaceous Chondrite (obtained using 400-kV electron microscope-electron diffraction camera). a - $\phi = 0°$; b - $\phi = 30°$.

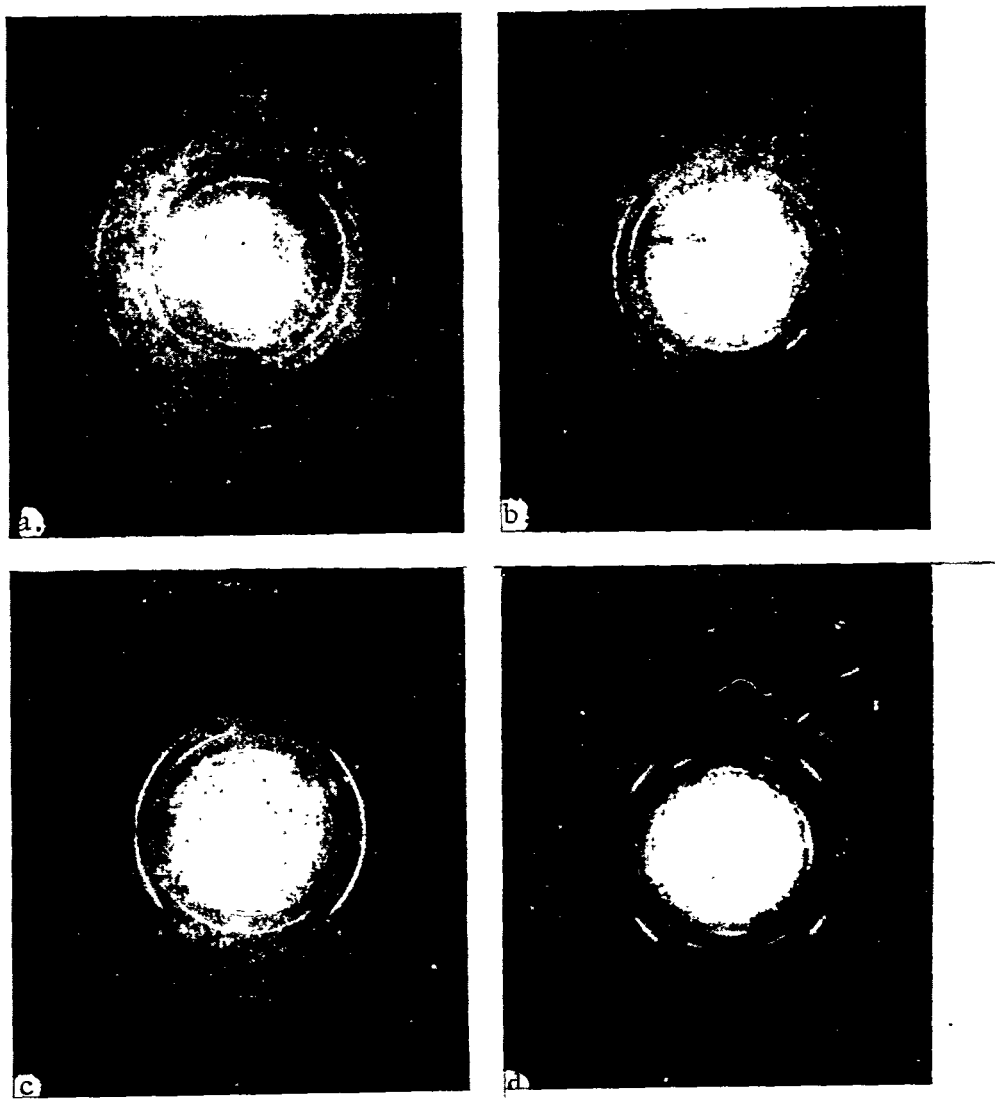


Figure 85. Electron Diffraction Patterns of Oblique Textures of Particles of Carbonaceous Matter in Staroye Borinskino Carbonaceous Chondrite (obtained using 400-kV electron microscope-electron diffraction camera).
Angle of inclination ϕ; a - 20°; b - 30°; c - 40°; d - 50°.

Table 31

Interplanar Spacings of Fraction Containing High-Molecular Carbonaceous Matter in the Kainsaz Chondrite, Containing Carbon

Kainsaz		Graphite		Kainsaz		Graphite	
I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$
1	7,31	—	—	1	1,578	—	—
1	4,69	—	—	3	1,543	6	1,545
3	3,84	—	—	1	1,517	—	—
10	3,51	10	3,38	3	1,454	—	—
9	3,22	—	—	9	1,370	5	1,350
1	2,736	—	—	1	1,349		
1	2,387	—	—	2	1,227	9	1,228
1	2,271	—	—	1	1,157	9	1,153
9	2,222	—	—	5	1,114	4	1,116
3	2,130	5	2,129	1	1,103	—	—
2	2,020	2	2,019	2	1,078	—	—
1	1,940	—	—	1	1,069	—	—
2	1,780	3	1,783	3	1,041	—	—
7	1,752	—	—	1	1,032	—	—
1	1,734	—	—	1	1,012	1	1,015
9	1,703	8	1,672				

The electron diffraction pattern of bituminous matter for the Groznaya meteorite (Figure 81) gives values for the interplanar spacings similar to roentgenometric data. It is comparable to the electron diffraction patterns of solid high-molecular paraffins in petroleums which were studied by B. A. Anurov, Yu. M. Korolev and G. I. Nosov (1962).

The polymers of the greater part of the carbonaceous matter in carbonaceous chondrites had an amorphous and crystalline structure. However, in individual meteorites, in addition to the high-molecular organic matter, there was an insignificant admixture of particles with a more ordered structure. In the Orgueil meteorite (type-I carbonaceous chondrite) the carbonaceous matter was completely represented by high-molecular organic matter (Figure 82). In the Cold Bokkeveld carbonaceous chondrite the organic polymers had primarily an amorphous structure (Figure 83). In the Migei meteorite, in addition to amorphous polymers (Figure 84) there were inclusions of high-molecular organic matter with a crystalline structure and also very tiny inclusions of finely dispersed matter with traces of graphitization of individual particles[1]. In electron diffraction study of the high-molecular organic matter in the Staroye Borinskino carbonaceous chondrite revealed the presence of polymers with a crystalline nature. Figure 85 shows the textured polycrystals of the

[1] This agrees with the data obtained by I. V. Grinberg (1964) that some part of the carbonaceous matter in the Migei carbonaceous and also in the Groznaya meteorite is represented by elementary carbon.

polymers in this meteorite rotated from a position perpendicular to the electron beam by different angles ϕ.

Thus, in carbonaceous chondrites the carbonaceous matter is represented by a mixture of compounds with different degrees of crystallinity, from amorphous high-molecular organic matter to partially graphitized matter in which good order of carbon atoms in the chain is observed.

In a type-I carbonaceous chondrite (Orgueil meteorite) it is completely represented by high-polymeric organic matter. In type-II carbonaceous chondrites, this matter either completely makes up the fraction (Cold Bokkeveld meteorite) or quantitatively predominates (Migei and Staroye Borinskino meteorites).

Thermal Analysis

Thermal analysis, one of the structural analysis methods, is rather frequently used in the study of natural objects. However, since the latter constitute a complex mixture, this analysis gives only qualitative information concerning the phase composition and therefore it is used as a supplementary method.

Thermal analysis is frequently used in the study of clayey minerals. /132
As is well known, clays contain finely dispersed organic matter. Therefore, the thermograms of clays yield some information concerning the organic compounds which they contain. For example, thermograms of clays of Maykop age show exothermic effects with maxima at ~ 380 and ~ 600°; these correspond to bituminous humic substances respectively (Logvinenko, Shumenko, 1961).

Some researchers have used these methods in a special study of organic compounds such as petroleums, coal, shales, peat, etc.

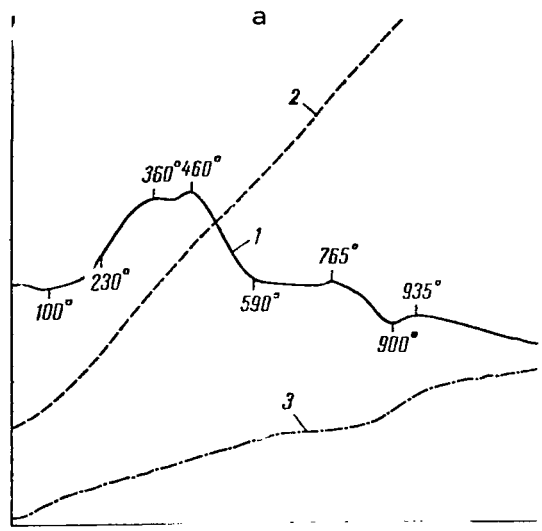
The author used thermal analysis in a study of the Migei and Groznaya carbonaceous chondrites, as well as the Kainsaz chondrite, in order to obtain additional confirmation of the interpretation of the composition of carbonaceous compounds without extracting them from the meteorite matrix. A differential thermal analysis was made using an ATVU-8 automatic thermal balance apparatus with heating for a period of 1 hour under atmospheric conditions up to 1100°. Figure 86 shows the thermograms of these meteorites.

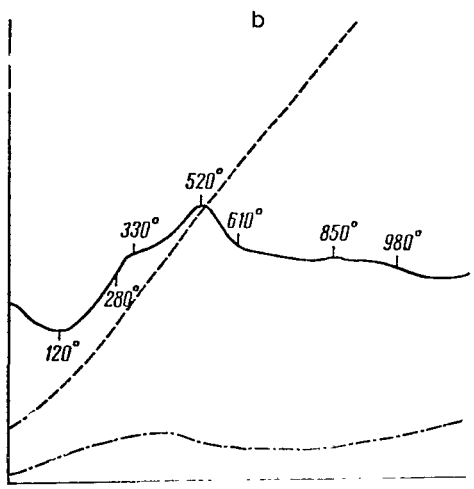
The differential thermal curves for the Migei carbonaceous chondrite (Vdovykin, 1965a) shows a broad exothermal effect in the temperature range from 230 to 590° with 2 maxima at 360 and 460°. These maxima also correspond to the changes on the temperature curve. In all probability, the first maximum is caused by the presence of bituminous compounds, whereas the second is caused by the presence of high-molecular organic matter. Naturally, the influence of other mineral components also could exert an effect.

The very weak exothermic rise at 765° can have a number of explanations. It possibly is caused by the presence of very tiny inclusions of carbon with a more ordered structure. On the other hand, it is known, for example that coal

(Bressler, et al, 1952) when heated to 500-600° is transformed into semicoke, and with further heating it is changed into coke with the release of gaseous products. A similar process could also occur in the carbonaceous matter in meteorites. Other transformations, which require special study, also are possible.

The differential curve for the Migei meteorite shows an endothermic effect at ~ 100°, which corresponds to the elimination of absorption water, and at ~900°, caused by separating out of the hydroxyl group from the chlorite-serpentine minerals[1]. The elimination of the hydroxyl group had a marked effect on increase in weight loss, as is clearly seen on the curve for weight loss. The total weight decrease for this meteorite was 14.5%.





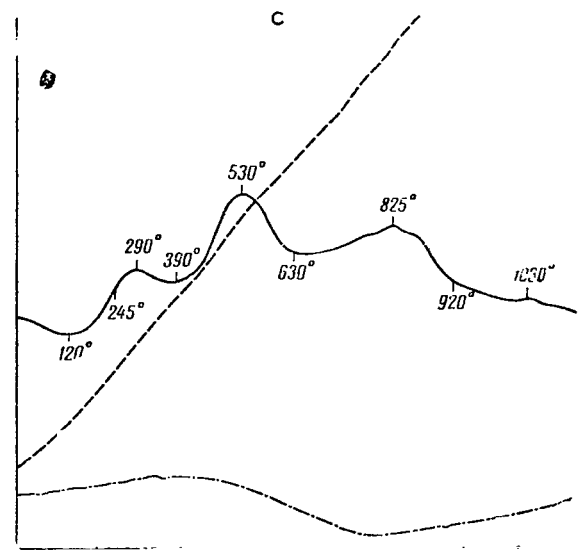


Figure 86. Thermograms of Migei (a) and Groznaya (b) Carbonaceous Chondrites and Kainsaz (c) Chondrite Containing Carbon.
1 - differential curve; 2 - temperature curve; 3 - curve of change in weight.

[1] A. M. Davaritskiy and L. G. Kvasha (1952) give a heating curve for the pulverized Staroye Borinskino carbonaceous chondrite, enriched with hydrous silicates. The curve revealed endothermic effects characteristic for chlorite; at ~680 and ~820°, which can be attributed to the setting free of the hydroxyl group.

The differential curve for the Groznaya meteorite also reveals the presence of bituminous compounds; there is an exothermic effect at 330° and high-molecular organic matter (~520°).

It is notable that the first exothermic peak has a far lesser height in comparison with the second than in the case of the Migei meteorite. This agrees with the data given above on the higher concentration of bituminous compounds in the Migei meteorite in comparison with the Groznaya chondrite.

The behavior of the curve of weight change for the Groznaya meteorite is different than for the Migei meteorite. Approximately to 500° there is a /134 gradual loss in weight, then to ~800° the weight increases somewhat, after which there is again rise of the curve. The final weight loss for the Groznaya meteorite is only 1.8% of the initial weight. This behavior of the curve can be attributed to the presence of ferronickel and sulphide in the meteorite, whose oxidation during heating leaps to an increase in weight of the meteorite.

The thermogram for the Kainsaz chondrite, which contains carbon, is in part similar to the thermogram for the Groznaya carbonaceous chondrite. The differential curve reveals exothermic effects with maxima at ~290 and ~530°. The rather high exothermic peak at a temperature of ~ 825° can be attributed in part to the presence of graphite particles. The behavior of the curve for weight change is also somewhat similar to the character of the curve for the Groznaya meteorite. Since the content of ferronickel and sulphides in the Kainsaz meteorite is higher than in the Groznaya chondrite, the weight after heating even increased insignificantly for the Kainsaz chondrite.

As a comparison with the thermogram of the investigated chondrite, Figure 87 shows a thermogram of PZh coal from the Don Basin obtained by Yu. P. Girin. The strongest exothermic effect in this case appears at 620°.

Nagy, et al (1963) made a thermobalance analysis of the Ivuna, Orgueil and Murray carbonaceous chondrites, as well as the St. Marks enstatic chondrite and the ordinary Bruderheim and Holbrook chondrites. For the carbonaceous chondrites they noted a considerable weight loss when heated to 1000° (Figure 88). These authors prepared a mixture of compounds which in composition corresponded somewhat to the mineral composition of the Orgueil meteorite. The composition of the mixture was as follows (% by weight): chlorite 68, magnetite 14, epsomite 8, sulphur 4, humic acid 6. The curve for the thermobalance analysis of this mixture was rather similar to the curve for the Orgueil meteorite. Nagy, et al, also reported that Faust investigated the Orgueil and the Migei meteorites by the differential thermal analysis method. On the thermograms for these meteorites Faust obtained broad exothermic effects caused by the presence of organic compounds.

The character of the change in the weight loss curve with heating under atmospheric conditions to 360° was investigated by Mueller (1963a) for the Orgueil and Cold Bokkeveld carbonaceous chondrites and for bituminous matter extracted from the Cold Bokkeveld meteorite (Figure 89). According to his data, the Orgueil meteorite loses weight gradually; at 350° the loss is about 18%.

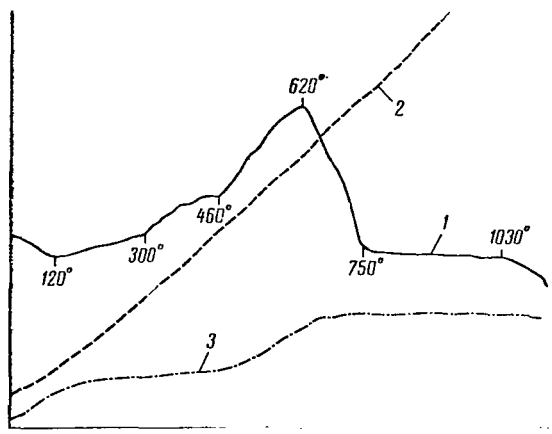


Figure 87. Thermogram of PZh Coal (5% coal plus 95% Al_2O_3; from Yu. P. Girin, 1964).
For symbolization see Figure 86.

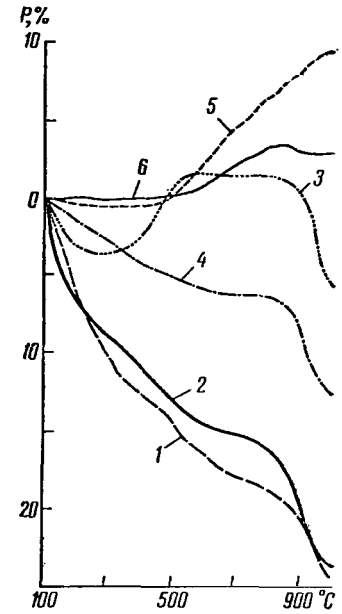


Figure 88. Curves of Thermal Balance Analysis of Carbonaceous Chondrites and Other Meteorites (Nagy, Meinschein, Hennessy, 1963).
P - change of weight during heating (upward from 0-- weight increase, downward -- loss); 1 - Ivuna; 2 - Orgueil; 3 - Orgueil (after heating to 510°); 4 - Murray; 5 - St. Marks enstatic chondrite; 6 - ordinary Bruderheim and Holbrook chondrites.

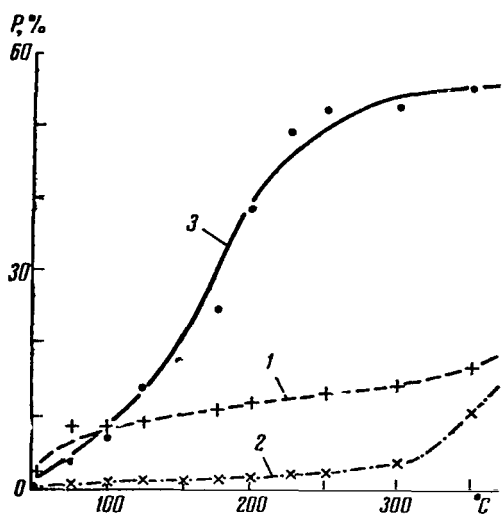


Figure 89. Curves of Weight Loss with Heating to 360° for the Orgueil (1), and Cold Bokkeveld (2) Carbonaceous Chondrites and Bituminous Matter from the Cold Bokkeveld Meteorite (3)(G.Mueller, 1963a).

The Cold Bokkeveld meteorite loses~ 3% weight at~300°, but at 350° its weight loss is already 14%. The bituminous matter extracted by solvents in a Soxhlet apparatus from the Cold Bokkeveld meteorite is rapidly oxidized.

With heating to 100° the loss was 7%, but with heating to 250° it was already ~53%. This was followed by a very slow weight loss, and at 350° it had attained 55%.

Thus, a thermal analysis confirms that the carbonaceous matter in carbonaceous chondrites has a complex composition: it is bituminous matter and high-molecular organic matter, although in individual cases there is an admixture of carbon with a more ordered structure.

There is a quantitative predominance of high-molecular organic matter. In the Kainsaz chondrite, which contains carbon, the carbonaceous matter is represented to a large extent by graphite.

The high-molecular fraction of carbonaceous matter in carbonaceous chondrites is a complex mixture of polymers of aromatic carbon with different degrees of order of the atoms in the structure (from amorphous high-molecular /135 matter to partially graphitized). Therefore, for the carbonaceous matter in meteorites it is difficult to find an analogy among terrestrial organic minerals. Since the greater part of the carbonaceous matter in meteorites is represented by high-molecular organic matter whose structure to a certain degree approaches the structure of coals, henceforth it apparently seems justified to refer to the carbonaceous matter in carbonaceous chondrites as coaly matter.

The admixture of particles with a more ordered structure of carbon atoms, insignificant in quantitative respects, apparently should scarcely be related to graphite. At this stage in research, it possibly is admissible to identify such particles arbitrarily with the black carbon which was extracted by P. Ramdor (1962), although in the mineralogy of caustobiologists (Orlov, Uspenstiy, 1936) this form of aromatic carbon is not defined.

DIAMONDS IN UREILITES

CHAPTER SIX

CARBONACEOUS MATTER IN UREILITES

History of Study of Diamonds in Meteorites

Diamonds were first discovered in meteorites by M. V. Yerofeyev and P. A. Lachinov in 1888 in the Novyy Urey achondrite-ureilite. By processing the meteorite with aqua regia, HF and H_2SO_4 Yerofeyev and Lachinov extracted from it a residue of black color (2-2.5% of the meteorite weight) which did not decompose when exposed to acids. This residue contained 89.56% carbon and 10.44% ash. After fusion of this black fraction, which could not be decomposed by acids, with $KHSO_4$ about 40% of its initial weight remained. The fraction remaining after fusion consisted of very hard, white, slightly greyish grains of diamond, which Yerofeyev and Lachinov classified as black diamonds. The hardness of the grains was greater than the hardness of corondum and its specific gravity was 3.3. The carbon in this fraction was 95.40%, ash 3.23%. The authors noted that of the 2.26% carbon in the Novyy Urey meteorite 1% constituted diamonds and 1.26% was accounted for by carbonaceous matter which they felt was amorphous carbon (Yerofeyev, Lachinov, 1888a, b; Jerofejeff, Latchinoff, 1888). The presence of diamonds in the Novyy Urey meteorite was soon confirmed by Kunz (1888) and was recently confirmed by the roentgenometric method by Ringwood (1960a); the latter author noted that the fraction in the Novyy Urey meteorite which would not be decomposed by acids consisted of diamond, graphite and an admixture of kamacite. /136

Diamonds were known not only in the Novyy Urey and Goalpara achondrite-ureilites, but also in an iron meteorite, the coarse-structured Canyon Diablo octahedrite. Diamonds were first identified in this meteorite by Foote in 1891. The diamonds in the Canyon Diablo meteorite had a specific gravity of 3.3 and according to chemical analysis data, contained 99.36% carbon and 1.29% Fe_2O_3 (Farrington, 1915).

Ksanda and Henderson (1939) checked the diamonds in the Canyon Diablo meteorite by the roentgenometric method. After processing pieces of this meteorite with HNO_3 and HCl acids, Ksanda and Henderson extracted about 50 tiny black grains with a grain size ranging between 0.1-0.6 and 0.9 mm, which they decided were black diamonds. The roentgenometric method analysis (CuK) which they made revealed that these aggregates contained diamond. Table 32 gives the roentgenometric characteristics of the diamonds in the Canyon Diablo meteorite. The unit cell of the diamond is a = 3.557 Å. /137

Nininger, 1939, 1956) noted that the diamond in this meteorite was a black diamond which is in aggregates with graphite. He noted a certain pattern in the distribution of diamonds: diamonds were present only in meteorite

samples found on the wall of a crater; diamonds were not discovered in samples of the meteorite found on the plain surrounding the crater. A more detailed study of samples of the Canyon Diablo meteorite containing diamonds was made later by Lipschutz and Anders (1961a, b)

Table 32

Roentgenometric Characteristics of Diamond in Canyon Diablo Iron Meteorite (Ksanda, Henderson, 1939)

hkl	*I*	$\frac{d_\alpha}{n}$	*a*, A
111	10	2,062	3,572
220	6	1,255	3,550
311	4	1,069	3,545
400	1	0,889	3,556
331	1	0,817	3,561
		a_{mean}, A	3,557

A roentgenometric study of the diamonds in the Canyon Diablo and Novyy Urey meteorites was made by Wentorf and Bovenkerk (1961). After comparing them with synthetic diamonds, these researchers concluded that the diamonds in the meteorites were formed from graphite at a pressure greater than 55kbar and at a temperature at about 1200° During this process iron played the role of a catalyst.

Weinschenk (1889) believes that another iron meteorite, the Magura octahedrite, also contains diamonds. However, this meteorite has not yet been checked for the presence of diamonds.

The literature contains reports on the possible presence of diamonds in chondrites. A very small grain, possibly containing a diamond, was found in the Carcote crystalline olivine-bronzite chondrite (Sandberger, 1889). This meteorite was checked by G. Urey, et al (1957), but they apparently did not find diamonds in it (Lipschutz, Anders, 1961b).

Sarma and Mayeda (1961) checked a number of chondrites for possible presence of diamonds. They took weighed pulverized samples of meteorites, each 10 g, processed them with aqua regia and HF, H_2SO_4 and $HClO_4$ acids. The black residue which was insoluble in acids was centrifuged in heavy fluids and studied by the roentgenometric method. In two chondrites (Peetz and Taiban) Sarma and Mayeda noted the possible presence of diamonds. The fraction of the Peetz meteorite was represently primarily by a spinel phase. The X-ray photograph of this fraction shows a number of lines, five of which were comparable to the lines for diamonds. However, the line intensity was different than the intensity of the roentgenometric lines for a diamond. The authors estimate

the possible presence of diamond at 0.015%. On the X-ray photograph of the fraction from the black crystalline Taiban chondrite, among the lines characteristic of diamond, there was only one (2.063 Å). Its intensity was comparable to the intensity of a similar line for diamonds. However, the quantity of residue which could not be decomposed, in the opinion of these authors, was too small for other lines to appear on the X-ray photograph, particularly since /138 the fraction was very finely dispersed. Sarma and Mayeda note that in order to draw more reliable conclusions concerning the presence of diamonds in this meteorite it will be necessary to investigate a larger piece. In their other studies they failed to find diamonds in chondrites; only the presence of a spinel phase was observed, although sometimes graphite was also quoted.

Thus, diamonds were definitely established in three meteorites, in two achondrite-ureilites, the Novyy Urey and Goalpara, and in the Canyon Diablo iron meteorite-octahedrite.

The Dyalpur ureilite, the third of all the known meteorites classified as ureilites, had not been checked earlier for the presence of diamonds. In this ureilite diamonds were found independently by Lipschutz (1962) and in our investigation (Vinogradov, Vdovykin, 1963). In addition, we noted definite evidence indicating the possible presence of diamonds in the Ghubara chondrite and then in the Kul'p chondrite. The initial weighed samples of these chondrites were 0.2065 and 0.7965 g respectively. The X-ray photographs of the fractions extracted after decomposition of samples of these meteorites by acid were in part similar to the X-ray photograph published by Sarma and Mayeda (1961) for a fraction of the Peetz chondrite. However, Lipschutz (1965a) feels that on these X-ray photographs the lines characterizing diamonds in all probability correspond to the X-ray lines of chromite. We undertook a detailed check of the diamonds in the Ghubara chondrite. We checked the possibility of a partial oxidation of carbonaceous matter during fusion with Na_2O_2, extracted the fractions from the nonmagnetic part of this meteorite by means of acids, and then investigated them, studied a synthetic mixture of prepurified terrestrial chromite and pulverized diamond with a particle size of micromonocrystals which was postulated for the Ghubara chondrite (~0.01 μ), etc. No final answer could be obtained. Accordingly, it must be agreed that at the present time diamonds are known most reliably in four meteorites, of which three are ureilites and the fourth is an iron meteorite, the Canyon Diablo.

Researchers have repeatedly emphasized the importance of studying diamonds in meteorites because diamonds could be evidence of the conditions under which the meteorites themselves were formed. Primarily on the basis of the presence of diamonds in meteorites, Urey (1956) advanced the idea of so-called primary and secondary objects. He postulated that meteorites underwent two stages in their development. Initially there was a cosmic body of lunar size. In its central part, where sufficiently high pressures[1] and temperatures prevailed, diamonds were formed. After the destruction of this primary body there was an agglomeration of matter with the formation of cosmic bodies of small dimensions, the size of asteroids. Milledge, et al (1965) recently postulated that if the

[1] The pressure in the lunar interior attains 45kbar.

diamonds in the Canyon Diablo meteorite were formed in space, they would have radiation defects in their structure caused by cosmic irradiation.

Lipschutz and Anders (1961a, b), who made a detailed study of the occurrence of diamonds in the Canyon Diablo meteorite, concluded that the diamonds in this meteorite were formed at the time of its impact against the earth. These researchers postulated that the diamonds in ureilites were also formed at the time of some catastrophic event, during the destruction of the parent bodies where the meteorites originated at the time of a collision. In the latter case, the diamonds in ureilites could therefore yield information on the character of such a destructive collision in space.

Mineralogicial Composition and Structure of Ureilites

Structural Characteristics of Ureilites

Ureilites as a special type of stony meteorites were defined by M. V. Yerofeyev and P. A. Lachinov (1888a, b). In the Roze-Chermak-Brzhezina classification ureilites were considered chondrites. However, Prior (1920) later classified them as calcium-poor achondrites. A. N. Zavaritskiy (Zavaritskiy, Kvasha, 1952) defines these meteorites as carbonaceous nonfeldspathic achondrite-ureilites. Mason (1963a), in conformity with Prior, classified ureilites in a group of olivine-pigeonite achondrites. /139

Only three meteorites, Novyy Urey, Dyalpur and Goalpara, are classified as ureilites. This constitutes 0.17% of all the meteorites, approximately 0.28% of the stony meteorites, or 4.92% of all achondrites. The ureilites have not been sufficiently studied. This entire group of meteorites has been studied by L. G. Kvasha (1958, 1959; Kwascha, 1958). Wiik (1966) recently made a chemical analysis of all three ureilites. Allen determined their carbon content.

Ureilites have a dark grey, almost black color and a granular fracture. The specific gravity of the Novyy Urey meteorite is 3.46[1] (Yerofeyev and Lachinov, 1888b); the specific gravity of the Goalpara meteorite is 3.44 (Teclu, 1870). Ureilites are permeated by macroscopically visible, slightly sinuous, elongated voids and pores; the elongated voids do not intersect mineral grains. Tschermak (1870) described them as "fissure-honeycombs".

Macroscopically and under the microscope in petrographic sections all three meteorites are very similar to one another, particularly the Novyy Urey and Dyalpur meteorites. Tschermak described the structure of the Goalpara meteorite as porphyric. Ureilites do not contain chondrules. They consist of grains of silicates of irregular configuration, frequently rather large (up to 4 mm). The large grains are separated by carbonaceous matter.

Ureilites consist of olivine, monoclinic pyroxyene, which according to A. N. Zavaritskiy and L. G. Kvasha (1952) is close in its optical properties

[1] According to a recent determination made by M. I. D'yakonova (1964), the specific gravity of the Novyy Urey meteorite is 3.29 g/cm^3.

to pigeonite, ferronickel (kamacite) and also sulphides (troilite) and carbonaceous matter. According to L. G. Kvasha (1958), the mineral composition of ureilites is as follows (% by volume):

Novyy Urey: 89 Ol_{20}, 9 Py_m, 2 Fe_4, (C);
Dyalpur: 88 Ol_{20}, 10 Py_m ~ 2 Fe, (C)
Goalpara: 85 Ol_{20}, 11 Py_m 3.5 Fe_6, (C).

In chemical composition ureilites also differ from other meteorites. They contain little FeS and metallic iron. In addition, ureilites have a very low content of alkali elements.

Table 33

Content and Isotopic Composition of Neon and Argon in Novyy Urey and Goalpara Ureilites (Stauffer, 1961)

Meteorite	Ne^{20}	Ne^{21}	Ne^{22}	Ar^{36}	Ar^{38}	Ar^{40}	Ne^{20}/Ne^{21}	Ne^{21}/Ne^{22}	Ne^{20}/Ne^{22}	Ar^{40}/Ar^{36}	Ar^{36}/Ar^{38}
	$n \cdot 10^{-8}$ n. cm^3/g										
Novyy Urey	3,5	2,25	2,58	238	44,7	260	1,55	0,87	1,33	1,09	5,33
Goalpara	9,28	8,55	9,71	39,6	7,7	⩽440	1,08	0,88	0,956	⩽11,1	5,14

Ureilites, like carbonaceous chondrites, are enriched with primary inert gases, argon and neon. The content and isotopic composition of the inert gases in the Novyy Urey and Goalpara ureilites are given in Table 33 on the basis of data published by Stauffer (1961). The isotopic composition of the neon in these meteorites also was studied by the Eberhardts (P. Eberhardt, A. Eberhardt, 1960, 1961). According to their data, the radiation age of the Novyy Urey meteorite is 10 million years and the radiation age of the Goalpara meteorite is 35 million years. /140

Fleischer, et al (1967) recently studied the tracks of heavy cosmic particles in the olivine of the Goalpara ureilite. They found from the tracks that while in space the meteor body had a radius of more than 48 cm and the 12-centimeter meteorite which fell to earth, when penetrating into the earth's atmosphere, was at a depth of more than 38 cm from the original surface of the meteorite body.

The author of this book had small samples of all three ureilites at his disposal.

Novyy Urey

The Novyy Urey ureilite fell in the territory of the present-day Gor'kovskaya Oblast (Russian Soviet Federative Socialist Republic) on 4 September 1886. Its falling was preceded by a bright bolide and explosive effects. Three individual pieces fell, but only one was preserved; it weighed approximately

1.9 kg. A second individual piece fell into a swamp and the third was destroyed by the local population. The greater part of the meteorite is now in the Leningrad Mining Museum.

The circumstances of falling of the meteorite and its description have been given in studies by M. V. Yerofeyev and P. A. Lachinov (1888a, b), who found diamond in this meteorite. The meteorite was studied by Daubrée (1888) and Kunz (1888). This meteorite has now been studied more thoroughly than the other ureilites. It was studied microscopically by A. N. Zavaritskiy and L. G. Kvasha (1952), L. G. Kvasha (1958, 1959) and I. A. Yudin (1958) and by X-ray methods by V. I. Mikheyev and A. I. Kalinin (1958). A roentgenometric method study of the diamonds was made by Ringwood (1960a), the isotopic composition of the carbon was studied by A. V. Trofimov (1950), the isotopic composition of oxygen was investigated by A. P. Vinogradov, et al (1958) and the inert gases were the object of research by Stauffer (1961). The meteorite was chemically analyzed by P. A. Lachinov (Yerofeyev, Lachinov, 1888a, b) and a partial analysis was made by Ringwood (1960a). A complete chemical analysis of the ureilite was recently made by M. I. D'yakonov (1964) and Wiik (1966).

It can be seen in a polished section of the meteorite that it is permeated by a system of voids and pores (Figure 90). The voids do not penetrate into the large grains of silicates, which are up to 4 mm in diameter. These large grains are seemingly surrounded by voids. The grains of silicates are easily sorted out in the pulverized matter of the meteorite. In the silicate fraction the grains are pure, without carbon admixtures (Figure 91), in contrast to carbonaceous chondrites. This is evidence, in particular, of a more compact structure of the carbonaceous matter in ureilites.

The meteorite matrix also contains rather infrequent platelets of kamacite and troilite, sometimes present in aggregate with one another. Frequently they are oriented along the voids. The platelets measure less than 0.3 mm, but sometimes measure even as much as 1 mm. In a carefully pulverized sample of the meteorite weighing 40 mg the author found only one kamacite platelet 1 mm in diameter (Figure 92) having a semicrystalline structure[1]. In most cases the kamacite and sulfides are found in the matrix in the form of very /145
small grains. Under an electron microscope the author observed infrequent hexagonal platelets of troilite up to 2 μ in diameter in pulverized matter from the Novyy Urey meteorite.

The irregular configuration of the grains of silicates distributed in the opaque matrix of this meteorite can be seen particularly well in a petrographic section of the Novyy Urey meteorite (Section 1176). The silicate grains for the most part are olivine, less frequently they are pyroxene. They are broken by a system of very small cracks (Figure 93). Smaller grains of olivine (with a diameter up to 0.5 mm) are scattered among the large grains of

[1] A semicrystalline character was also noted for the troilite in the Canyon Diablo meteorite (Heymann, et al, 1966) and for the cohenite in this same meteorite (Walker, 1966). It is assumed that the semicrystallinity is the result of recrystallization at the time of impact.

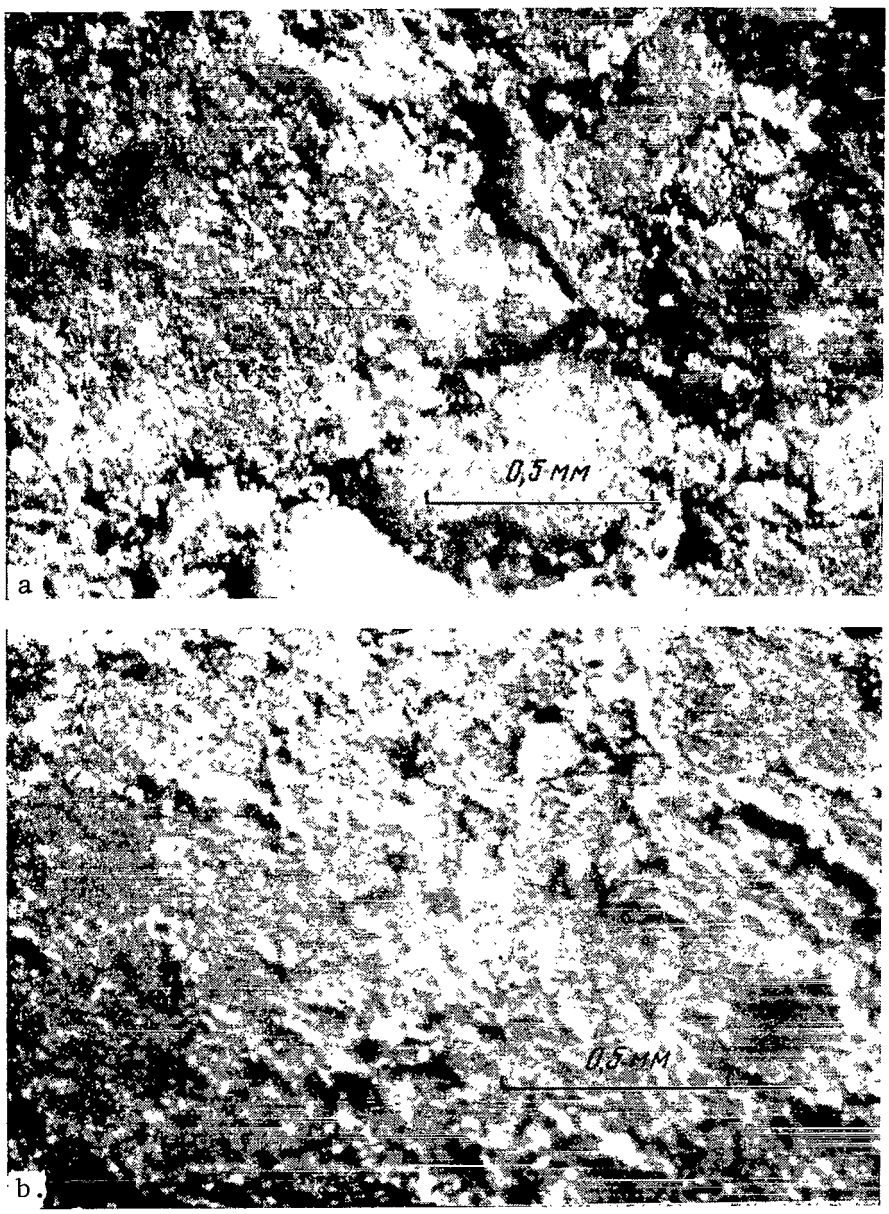
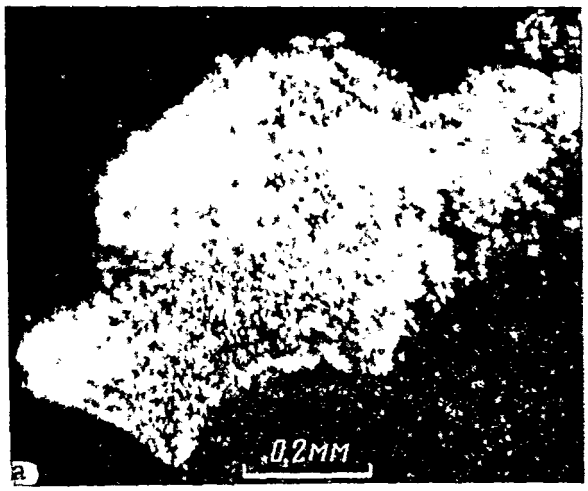


Figure 90. Elongated Voids in Pores in the Novyy Urey Ureilite. Polished Section, Reflected Light.
a - voids bordering on large grains of silicates; b - voids in more uniformly structured part of meteorite.

Figure 91. Silicate Fraction of Novyy Urey Meteorite.



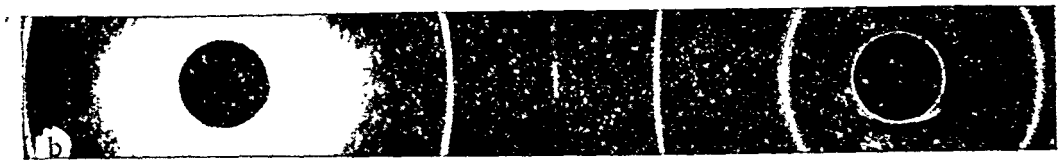


Figure 92. Large Platelet of Semicrystalline Kamacite from Novyy Urey Meteorite (a) and its X-Ray Pattern (b) (Fe-anticathode, Mn-filter; see Table 35.

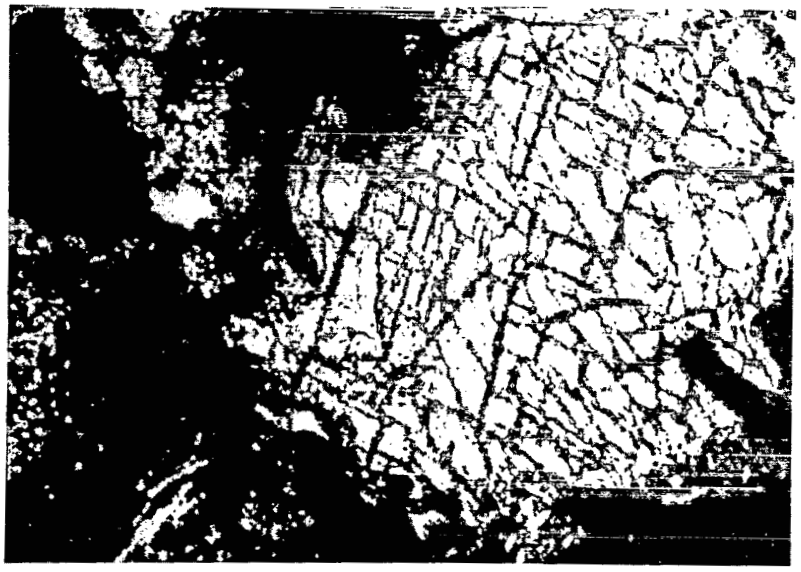


Figure 93. Large Grain of Olivine Broken by a Network of Cracks in the Novyy Urey Ureilite. Magnification 280, Without Analyzer.

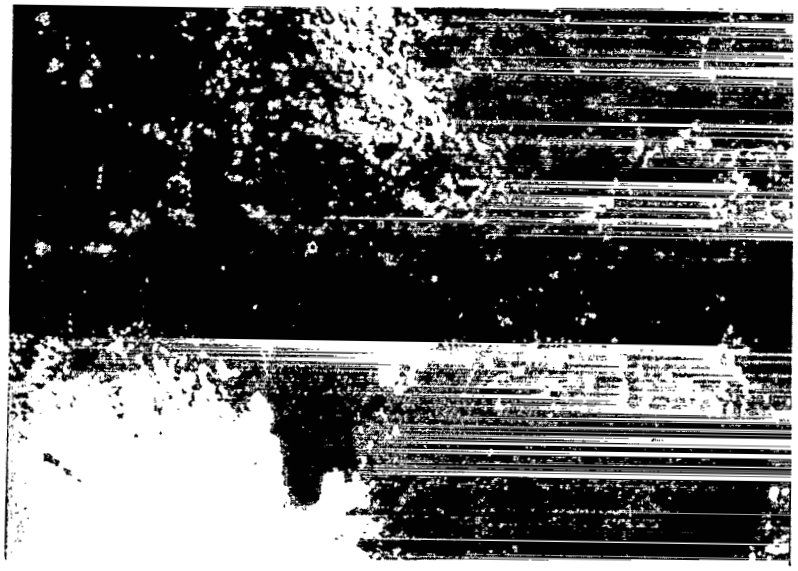


Figure 94. Grains of Silicates Separated by Carbonaceous Matter in the Novyy Urey Ureilite. Magnification 280, Without Analyzer.

Table 34

X-Ray Photographs and Mineral Composition of Ureilites

Meteorite								Interpretation					
Novyy Urey (Mikheyev, Kalinin, 1958)		Dyalpur		Goal-para		Fraction of Sili-cates from Goalpara meteor-ite		Olivine (Mikhe-yev, 1957)		Pigeonite (Ginzburg, et al, 1964)		Ferronickel (Mikheyev, Kalinin, 1958)	
I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$
2	5,11	2	5,10	2	5,10	—	—	4	5,07	—	—	—	—
1	4,59	—	—	—	—	—	—	1	4,60	—	—	—	—
—	—	1	4,35	2	4,32	1	4,35	2	4,29	—	—	—	—
1	4,17	—	—	—	—	—	—	1	(4,04)	—	—	—	—
7	3,88	—	—	6	3,88	—	—	7	3,87	—	—	—	—
4	3,72	—	—	2	3,75	—	—	4	3,72	—	—	—	—
6	3,49	2	3,48	4	3,50	—	—	6	3,49	3	(3,556)	—	—
6	3,31	2	3,36	2	3,32	2	3,34	3	(3,33)	5	3,321	—	—
6	3,19					5	3,19	—	—	—	—	—	—
—	—	2	3,16	3	3,17	9	3,16	—	—	10	3,204	—	—
7	2,998	5	2,995	5	2,996	—	—	5	2,995	10	2,993	—	—
6	2,883	4	2,906	4	2,906	10	2,880	—	—	9	2,898	—	—
—	—	3	2,796	—	—	—	—	—	—	—	—	—	—
9	2,776	—	—	7	2,770	—	—	9	2,770	—	—	—	—
—	—	—	—	1	2,719	—	—	3	(2,715)	—	—	—	—
—	2,587	—	—	—	—	4	2,567	—	—	7	2,567	—	—
9	2,518	6	2,525	7	2,525	—	—	10	2,515	6	2,491	—	—
10	2,458	7	2,441	8	2,463	5	2,463	10	2,455	—	—	—	—
—	—	—	—	1	2,384	—	—	—	—	—	—	—	—
1	2,359	—	—	1	2,347	—	—	2	2,350	—	—	—	—
—	—	—	—	4	2,277	—	—	—	—	—	—	—	—
8	2,257	4	2,252	—	—	—	—	9	2,253	1	2,256	—	—
—	—	—	—	4	2,244	1	2,219	—	—	3	2,245	—	—
2	2,161	—	—	2	2,157	—	—	5	2,165	—	—	—	—
1	2,126	3	2,128	2	2,128	3	2,135	—	—	6	2,148	—	—
1	2,084	—	—	—	—	—	—	—	—	5	2,055	—	—
10	2,026	10	2,024	10	2,024	4	2,031	2	2,032	—	—	10	2,022
—	—	—	—	1	1,999	—	—	—	—	—	—	—	—
5	(1,933)	2	1,944	3	1,938	2	1,944	7	(1,931)	1	1,954	—	—
1	1,879	2	1,887	2	1,887	—	—	3	1,877	—	—	—	—
1	1,844	—	—	—	—	—	—	—	—	—	—	—	—
3	1,814	—	—	1	1,823	—	—	2	1,812	2	1,808	—	—
3	1,786	—	—	2	1,779	1	1,783	2	1,788	2	1,788	—	—
9	1,748	7	1,741	8	1,741	1	1,747	10	1,747	1	1,759	—	—
3	1,726	—	—	—	—	—	—	—	—	—	—	—	—

(Table continued)

Table 34 (continued)

X-Ray Photographs and Mineral Composition of Urellites

Meteorite								Interpretation					
Novyy Urey (Mikheyev, Kalinin, 1958)		Dyalpur		Goal-para		Fraction of Sili-cates from Goalpara meteor-ite		Olivine (Mikhe-yev, 1957)		Pigeonite (Ginzburg, et al, 1964)		Eerronickel (Mikheyev, Kalinin, 1958)	
I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$
4	1,671	3	1,664	4	1,668	—	—	5	1,671	—	—	—	—
3	1,639	—	—	3	1,632	10	1,621	6	1,639	8	1,626	—	—
4	1,604	8	1,617	5	1,613	—	—	6	1,619	—	—	—	—
3	1,567	2	1,563	3	1,567	—	—	4	1,572	—	—	—	—
3	1,537	3	1,532	3	1,536	3	1,536	3	1,539	3	1,539	—	—
—	—	—	—	2	1,522	—	—	2	1,515	—	—	—	—
6	1,499	4	1,503	5	1,500	—	—	8	1,498	2	1,494	—	—
7	1,481	5	1,479	6	1,479	4	1,485	9	1,481	—	—	—	—
—	—	1	1,453	1	1,453	—	—	1	1,462	—	—	—	—
8	1,431	6	1,425	6	1,430	—	—	2	1,437	—	—	7	1,430
—	—	3	1,401	—	—	—	—	—	—	—	—	—	—
8	1,398	3	1,391	3	1,396	1	1,401	9	1,397	3	1,395	—	—
7	1,387	—	—	4	1,384	—	—	—	—	—	—	—	—
—	—	1	1,371	—	—	10	1,379	1	1,370	—	—	—	—
9	1,351	5	1,348	6	1,350	—	—	1	1,350	—	—	—	—
—	—	2	1,332	—	—	1	1,330	—	—	1	1,340	—	—
—	—	4	1,323	—	—	—	—	—	—	—	—	—	—
8	1,315	—	—	5	1,319	—	—	9	1,316	—	—	—	—
5	1,291	2	1,294	3	1,290	—	—	3	1,295	2	1,280	3	(1,289)
2	1,268	4	1,265	2	1,269	4	1,267	2	1,267	—	—	—	—
2	1,258	—	—	—	—	—	—	3	1,256	—	—	—	—
2	1,243	2	1,243	2	1,247	—	—	3	1,238	—	—	—	—
2	1,226	3	1,220	2	1,225	4	1,222	3	1,226	2	1,231	—	—
5	1,188	4	1,189	4	1,189	—	—	7	1,188	—	—	—	—
9	1,168	10	1,169	9	1,170	3	1.176	7	1,167	—	—	8	1,168
—	—	1	1,156	2	1,156	—	—	3	1,157	—	—	—	—
—	—	—	—	1	1,154	—	—	—	—	—	—	—	—
6	1,140	—	—	—	—	1	1,148	7	1,140	—	—	—	—
6	1,135	2	1,136	2	1,137	—	—	—	—	—	—	—	—
4	1,126	3	1,121	3	1,124	—	—	5	1,125	—	—	—	—
4	1,116	—	—	4	1,118	1	1,113	5	1,117	2	1,117	—	—
6	1,102	1	1,107	3	1,107	—	—	—	—	—	—	—	—
—	—	2	1,097	2	1,100	—	—	8	1,098	—	—	—	—
6	1,079	1	1,088	1	1,083	—	—	7	1,080				
5	1,074	3	1,073	4	1,075	4	1,072	7	1,074	4	1,076		
6	1,065	2	1,066	4	1,067	—	—	7	1,063				
5	1,054	3	1,051	4	1,052	3	1,053	4	1,052	5	1,059		

(Table continued)

Table 34 (continued)

X-Ray Photographs and Mineral Composition of Ureilites

Meteorite								Interpretation					
Novyy Urey (Mikheyev, Kalinin, 1958)		Dyalpur		Goal-para		Fraction of Sili-cates from Goalpara meteor-ite		Olivine (Mikhe-yev, 1957)		Pigeonite (Ginzburg, et al, 1964)		Ferronickel (Mikheyev, Kalinin, 1958)	
—	—	1	1,044	1	1,043	2	1,041	—	—	—	—	—	—
—	—	4	1,037	4	1,035	—	—	—	—	2	1,035	—	—
—	—	1	1,023	—	—	—	—	—	—	—	—	—	—
—	—	1	1,019	2	1,020	1	1,018	—	—	—	—	—	—
—	—	7	1,012	7	1,012	—	—	—	—	3	1,012	—	—
—	—	1	1,007	1	1,007	4	1,004	—	—	—	—	—	—
—	—	1	1,002	2	1,002	—	—	-	—	—	—	—	—

silicates (usually 1-3 mm in diameter). Among the smaller grains are groups of olivine grains resembling greatly fragmented chondrules. Such grains are separated primarily by carbonaceous matter (Figure 94). Here it forms independent sectors of irregular configuration, which seemingly have adapted themselves to the space free of silicates.

In reflected light it is possible to see very tiny infrequent grains of sulfides (troilite) and ferronickel. L. G. Kvasha (1958) notes that the ferro- /146
nickel is found in the form of a reticular distribution and in large olivine grains.

Carter and Kennedy (1964), in a study of two small sections of the Novyy Urey meteorite determined the following composition of this ureilite (in %): olivine 50, clinopyroxene 30, kamacite 10, troilite 4, graphite 5, diamond 1-2. A composition corresponding to Fo_{76} was determined for the olivine.

According to data from a roentgenometric analysis made by V. I. Mikheyev and A. I. Kalinin (1958) (Table 34), the Novyy Urey meteorite has an olivine-pyroxene composition; ferronickel is also present. According to Mikheyev and Kalinin, the olivine in this meteorite has a unit cell with the parameters: a = 4.752 Å, b = 10.279 Å, c = 5.996 Å. Its composition is expressed by the formula: $(Mg_{0.75}Fe_{0.25})_2 SiO_4$.

Table 35

Roentgenometric Characteristics of Kamacite from the Novyy Urey Ureilite (FeK_α)
(See Figure 92)

hkl	*I*	$\frac{d_\alpha}{n}$	*a*, Å
110	10	2,037	2,880
200	6	1,436	2,872
211	10	1,170	2,865
220	7	1,012	2,863
		a_{mean}, Å	2,870

Table 36

Chemical Composition of Novyy Urey Ureilite
(% by weight)

Components	1	2	3	4	Components	3	4
SiO_2	39,51	40,76	39,65	39,76	Fe	15,81	15,64
MgO	35,80	36,91	34,45	36,87	Si	18,54	18,57
FeO	13,35	16,58	17,56	15,09	Mg	20,78	22,23
Al_2O_3	0,60	0,39	1,97	0,41	S	0,25	0,58
CaO	1,40	1,05	1,19	0,81	C	—	2,23
Na_2O	—	0,45	0,05	0,18	Al	1,04	0,26
K_2O	—	—	0,04	0,04	Ni	0,09	0,12
P_2O_5	0,04	0,07	0,12	0,10	Ca	0,85	0,57
Cr_2O_3	0,95	0,45	0,62	0,68	Na	0,04	0,13
MnO	0,43	0,36	0,26	0,40	Cr	0,42	0,47
TiO_2	—	0,09	0,11	0,14	Mn	0,20	0,31
Fe	5,25	2,73	1,73	2,90	P	0,05	0,04
Ni	0,20	0,16	0,09	0,12	Co	—	0,05
Co	—	—	—	0,05	Ti	0,07	0,08
FeS	0,41	—	0,68	1,59	K	0,03	—
C	2,26	—	—	2,23	(O)	41,86	38,72
Total	100,20	100,00	98,52	101,37	Total	100,00	100,00

1 - M. V. Yerofeyev; P. A. Lachinov (1888); 2 - Ringwood (1960a); 3 - M. I. D'yakonova (1964); 4 - Wiik (1966).

The unit cell of ferronickel has a = 2.863 Å; kamacite contains approximately 4 atom. % nickel. According to Ringwood (1960a), the olivine in this meteorite contains 21 mol. % Fe_2SiO_4 (Fo_{79}) and kamacite has a = 2.8672 Å and contains about 2% nickel in solid solution. According to our roentgenometric data, the kamacite in the Novyy Urey meteorite has a = 2.870 Å (Table 35).

In its optical properties pyroxene is close to pigeonite (Zavaritskiy, Kvasha, 1952; Carter, Kennedy, 1964). Here it can be stated in passing that Reid, et al (1964) checked the luminescence of pyroxenes in this meteorite and found that the pyroxenes are not luminescent in ultraviolet light. /147

DuFresne (Anders, Lipschutz, 1966a) noted asterism in Laue diffraction patterns of the silicates of the Novyy Urey meteorite. He feels that this is characteristic for silicates which have experienced small impact.

The Novyy Urey meteorite apparently contains traces of chromite. I. A. Yudin (1958) cites data from a spectral analysis of the magnetic fraction of this ureilite in which he found (in %); Ni 0.03, Co and Ti traces, Mn 0.1, Al 0.1, Cu 0.01, Cr 0.1.

Table 36 gives a chemical composition of the Novyy Urey meteorite. According to an analysis made by M. I. D'yakonova (1964), the meteorite is characterized by a high degree of reducibility of its matter and a low content of FeS (0.68%) and the metallic Fe (1.82%). The metal contains 4.94% Ni, but according to Wiik's data (1966) the nickel content is 3.97%.

The carbon content in the meteorite is 2.13% (Trofimov, 1950), 2.23% (Wiik, 1966), or 2.26% (Yerofeyev, Lachinov, 1888a, b).

Dyalpur

The Dyalpur meteorite fell in the province of Sultanpur (India) on 8 May 1872. Only one individual piece weighing about 280 g fell. Almost the entire meteor is now in the British Museum of Natural History.

General brief information concerning the meteorite is given in the catalog published by Prior and Hey (1953), Ye. L. Krinov (1962b) and L. G. Kvasha (1962). In contrast to the Novyy Urey meteorite, this ureilite has remained almost completely unstudied. It has been investigated only by L. G. Kvasha (1958, 1959) by the microscopic method and by Wiik (1966), who determined its chemical composition. L. G. Kvasha noted a similarity of the mineral composition and structure of the Dyalpur ureilite and the Novyy Urey meteorite. A roentgenometric method study of the Dyalpur ureilite, which also established its similarity to the Novyy Urey meteorite, was made by the author (Vdovykin, 1964c).

The similarity of these meteorites can be seen particularly well under the microscope in petrographic sections; they are virtually indistinguishable from one another.

The Dyalpur ureilite consists (Section 2161) of large grains of silicates, olivine and monoclinic pyroxene, 1-3 mm in diameter, having irregular angular outlines, and smaller fragments (up to 0.5 mm in diameter).

Between the fragments of silicates there is carbonaceous matter which also fills the fissures in the large mineral grain. The irregular configuration of tiny grains of ferronickel and sulfides, frequently present in aggregates with one another, can be seen.

A roentgenometric analysis (Figure 95, Table 34) reveals that in this meteorite there is a predominance of olivine and pyroxene of the enstatite-augite type (pigeonite). Ferronickel is also noted. Asterism appears in the X-ray photograph. Asterism was also noted for the silicates in the Dyalpur meteorite by Lipschutz (Anders, Lipschutz, 1966a). Like DuFresne, Lipschutz feels that the asterism is caused by the mosiac structure of crystals of the silicates, which was caused by the impact. He noted a similar asterism for dunite, which was subjected to a pressure of 600 kbar during an impact.

Table 37 gives the chemical composition of the Dyalpur and Goalpara ureilites. Vogt and Ehmann (1965) made a neutron-activation analysis for the Dyalpur meteorite and found a silicon content of 19.0%, which is comparable /148
with Wiik's data (1966). Ehmann, et al (Ehmann, Lieberman, et al, 1967) found that the Dyalpur meteorite contained bromine and some other easily volatile elements. In the metallic phase of this ureilite they found $0.03 \cdot 10^{-4}$% gold and $0.15 \cdot 10^{-4}$% iridium (Ehmann, Baedecker, 1967).

The carbon content in this meteorite was 2.92% (Wiik, 1966).

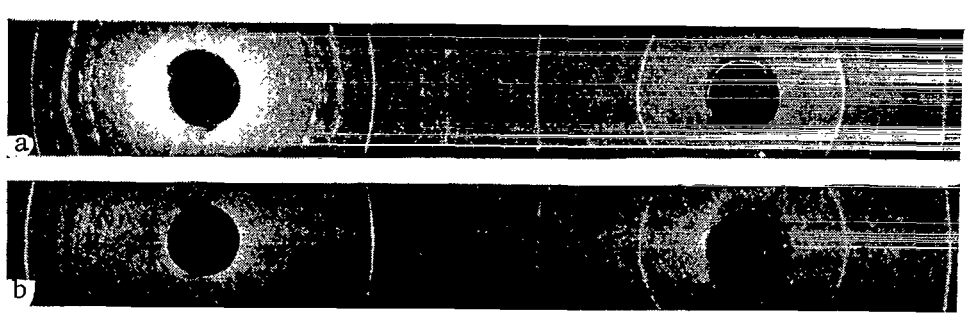


Figure 95. X-Ray Pattern of Pulverized Matter from the Dyalpur (a) and Goalpara (b) Ureilites; FeK_2, Manganese Filter.

Table 37

Chemical Composition of Dyalpur and Goalpara Ureilites
(% by weight)

Components	Dyalpur 1	Goalpara 2	Goalpara 1	Components	Dyalpur 1	Goalpara 1
SiO_2	41,92	40,36	40,82	Fe	11,31	16,40
MgO	38,91	37,42	36,52	Si	19,57	19,06
FeO	9,20	13,32	12,19	Mg	23,46	22,02
Al_2O_3	0,32	Сл.	0,88	S	0,63	0,61
CaO	1,36	0,60	0,43	C	2,92	1,54
Na_2O	0,09	—	0,07	Al	0,17	0,47
P_2O_5	0,07	—	0,06	Ni	0,13	0,09
Cr_2O_3	0,93	—	0,93	Ca	0,97	0,31
MnO	0,41	—	0,40	Na	0,06	0,05
TiO_2	0,18	—	0,12	Cr	0,64	0,64
Fe	3,07	8,49	5,86	Mn	0,32	0,31
Ni	0,13	—	0,09	P	0,03	0,03
Co	0,06	—	0,05	Co	0,06	0,05
FeS	1,72	сл.	1,67	Ti	0,11	0,07
C	2,92	0,72	1,54	(O)	39,62	38,35
H	—	0,13	—			
Total	101,29	101,04	101,63	Total	100,00	100,00

1 - Wiik (1966), 2 - Teclu (1870).

Goalpara

The Goalpara ureilite was found in the province of Assam in India in 1868. The weight of the piece found there was about 2.6 kg (Prior, Hey, 1953). The greater part of the meteorite is now in the British Museum of Natural History and in the Indian Geological Museum (in Calcutta).

The meteorite has been described by Tschermak (1870) and has been analyzed by Teclu (1870) and recently by Wiik (1966).

Urey, et al (1957) found diamonds in this meteorite. A microscopic study /149
of the meteorite was made by L. G. Kvasha (1958). The content and isotopic composition of the inert gases in the meteorite was determined by Stauffer (1961). Taylor, et al (1965) recently studied the isotopic composition of the hydrogen in the pyroxenes of this meteorite.

Like the two preceding ureilites, the Goalpara meteorite is permeated by a system of elongated voids and pores which do not penetrate the silicate grains (up to 2-4 mm).

The irregular configuration of the silicate grains also can be seen clearly in a transparent section of this meteorite (Section 2160). The edges of the grains are uneven and gradually merge into the matrix. In many cases the silicate grains are broken by fissures filled with carbonaceous matter (Figure 96). Sometimes groups of silicate grains resemble broken chondrules up to 0.5 mm in diameter (Figure 97). Still finer grains of silicates are distributed in the meteorite matrix in a large quantity (Figure 98). The matrix, which surrounds these small and larger silicate grains, has a black color which is completely opaque under the microscope; it has a homogeneous structure. Individual independent sectors of a black color and irregular configuration (up to 0.9 mm) are visible.

In large silicate grains, and also in the matrix, it is easy to distinguish small grains of troilite and ferronickel. Platelets of ferronickel (kamacite) usually have an elongated form and frequently surround individual grains of olivine, causing, as pointed out by L. G. Kvasha (1958), a sideronitic structure.

According to Tschermak (1870), the Goalpara meteorite contains 61.72% olivine, which has a composition corresponding to the formula $(Mg_{0.75}Fe_{0.25})_2SiO_4$, 30.01% pyroxenes (Tschermak determined pyroxenes as enstatite) and 8.49% ferronickel.

Lipschutz (Anders, Lipschutz, 1966a) recently noted asterism for the silicates in the Goalpara ureilite. Geake and Walker (1966) observed luminescence in the red region.

A roentgenometric method study of the Goalpara ureilite (Vdovykin, 1965a) (See Figure 95 and Table 34) shows that the meteorite consists of olivine and pyroxene of the enstatite-augite type (pigeonite).

Olivine and pigeonite stand out well in a roentgenometric survey of the silicate fraction extracted by the author from this meteorite (Figure 99, Table 34).

An X-ray pattern of a sample of the Goalpara ureilite also reveals ferronickel and troilite. The X-ray pattern of this meteorite is very similar to X-ray patterns of the other two ureilites, which is evidence of a similarity in their mineral composition.

In its chemical composition the Goalpara meteorite differs little from the other ureilites (Table 37). Like the other ureilites, this meteorite is characterized by a low content of alkali elements. Edwards (1955b) and Edwards and Urey (1955) determined the sodium content (0.04%) and potasium content (0.006%) in the Goalpara ureilite. The low content of alkali elements in this meteorite was recently confirmed by Easton and Lovering (1964). Using the flame photometry method, they determined the sodium content (0.018%) and potasium contents (0.007%). The meteorite contains $< 6 \cdot 10^{-8}$% uranium (Morgan, Lovering, 1967).

Figure 96. Filling of Fissures in Large Olivine Grain in Goalpara Ureilite with Carbonaceous Matter. Magnification 280, Without Analyzer.

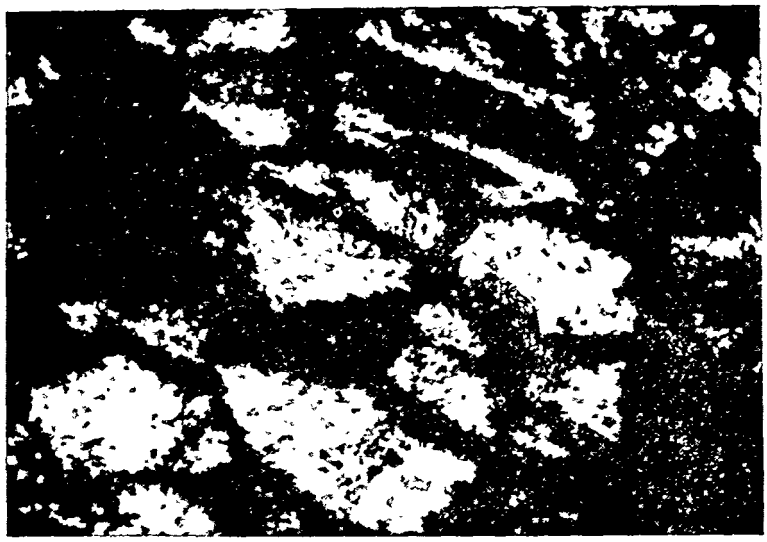


Figure 97. Sectors of Fragmented Olivine Chondrule Separated by Carbonaceous Matter in the Goalpara Ureilite. Magnification 280, Without Analyzer.

According to N. Teklu, the carbon content in the Goalpara meteorite is 0.72%; according to Allen (Wiik, 1966) it is 1.54%.

Mueller (1966) recently determined the total of volatiles for this meteorite (3.25% of the total meteorite weight).

Thus, in achondrite-ureilites, like in carbonaceous chondrites, the carbonaceous matter making up the matrix is distributed among the silicate fragments. /151 The carbonaceous matter fills the fissures in the mineral grains or surround the grains, around the periphery, in the form of a thin envelope. However, this substance for the most part fills the space between the blocks of large broken silicate grains. This character of the relationship between the carbonaceous matter and the silicate grains and the structural characteristics of these meteorites probably are evidence that the ureilites were subjected to a strong impact, as a result of which the carbonaceous matter in them was redistributed.

Results of Study of Carbonaceous Matter in Ureilites /152

Bituminous Matter

The author studied small pieces (about 0.1 g) of three ureilites taken from the inner parts of large samples; the luminescent-bituminological method was used. The pieces were without contamination. The author pulverized them in an agate mortar and successively extracted bituminous compounds from them at room temperature using the following organic solvents; petroleum ether, benzene, ethanol, alcohol-benzene (1:1), carbon tetrachloride and chloroform. It was found that the ureilites contain bituminous components in the following quantities (in %): Novyy Urey 0.066, Dyalpur 0.023, Goalpara 0.036 (Table 38), which is 2-5% of all the carbonaceous matter. This bituminous matter had a reduced character. In ultraviolet light (λ = 365 mμ) the extracts were luminescent in a blue color. The extracts dissolved in organic solvents and also the capillary extracts had the same color of luminescence. The reduced character of these extracts distinguished them from the bituminous matter in carbonaceous chondrites. Bituminous compounds were extracted with chloroform from 4 g of the Novyy Urey meteorite for a study of the organic matter by the infrared absorption spectroscopy method. The infrared spectrum of the extract is shown in Figure 100 (Vinogradov, Vdovykin, Karyakin, Zubrilina, 1966).

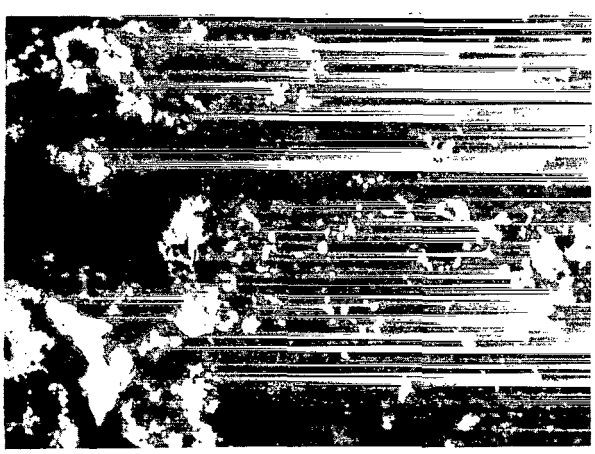


Figure 98. Distribution of Small Grains of Silicates in Goalpara Ureilite. Magnification 280, Without Analyzer.

The spectrum revealed a number of absorption bands. A broad absorption band in the region 2800-3000 cm^{-1} with

Figure 99. Silicate Fraction Extracted from Goalpara Ureilite.

Table 38

Content of Bituminous Matter in Ureilites

Meteorite	Place of falling	Date of falling	Sample of meteorite used*	Weight of extracted sample in g	Content of Bituminous Matter, % by wt.
Novyy Urey	R.F.S.F.R.	4.IX 1886	1174	0.1025	0.066
Dyalpur	India	8.V 1872	2161	0.1215	0.023
Goalpara	India	1868**	2160	0.0765	0.036

*Number of samples from the catalog of the meteorite collection of the Committee on Meteorites Academy of Sciences USSR.
**Date of meteorite find.

maxima at 2880, 2940, 2960 cm^{-1} corresponds to stretching vibrations of $-CH_3$ and $=CH_2$ groups (Bellami, 1963). This is also confirmed by the presence of /153 absorption bands in the regions 1400, 1140, 800 and 750 cm^{-1} which correspond to deformation vibrations of $-CH_3$ and $=CH_2$ groups and their chains. The presence of absorption in the region 3600, 3200 cm^{-1}, as well as weak absorption in the region from 1500 to 1400 cm^{-1}, makes it possible to postulate the presence of C-N-H groups in the organic matter of this meteorite. Oxygen groupings are completely absent. The organic matter in the Novyy Urey meteorite is represented primarily by hydrocarbons of the paraffin series.

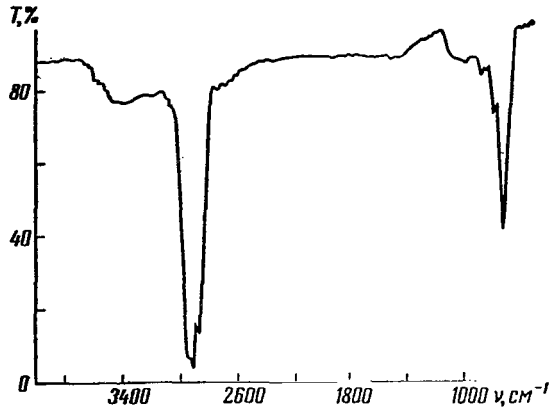


Figure 100. Infrared Absorption Spectrum of Bituminous Matter Extracted by Chloroform from the Novyy Urey Ureilite.

We used the infrared spectroscopy method for studying the organic matter in the Goalpara ureilite. Bituminous compounds from this meteorite were extracted with carbon tetrachloride for study; the meteorites weight was 0.52 g. The infrared absorption spectrum of the extract was therefore less clear. However, an absorption band at 2940 cm^{-1} which corresponds to stretching vibrations of CH groups stood out quite well on the spectrum.

The hydrocarbon character of the bituminous matter in ureilites can be confirmed by the earlier research by Teclu (1870); in the Goalpara meteorite he determined 0.73% carbon and 0.13% hydrogen. Teclu felt that these elements in the meteorite are included in organic compounds which had the compo sition (Cohen, 1894); C 84.88%, H 15.12%.

Polymineral Aggregates Containing Diamond and Graphite

Samples of pulverized matter from three ureilites (about 0.1 g), from which the bituminous components had first been extracted, were subsequently decomposed by the author by means of acids. First this pulverized matter was processed with aqua regia (3 parts HCl + 1 part HNO_3) in glass beakers with a subsequent boiling off of the acids and washing of the precipitate. Then the samples were processed in platinum vessels with HF acid supplemented by small portions of $HClO_4$.

The remaining fractions, which were undecomposed by acids, constituted approximately 2-3% of these meteorites. Later a similar method was used in decomposing larger samples of the Novyy Urey and Goalpara meteorites.

The initial sample of the Novyy Urey meteorite weighed 2.65 g. The weight of the fraction remaining after its decomposition was 0.06 g. Therefore, the fraction of this meteorite which could not be decomposed by acids was approximately 2.5%. After decomposition with acids, a sample of the Goalpara meteorite weighing 0.52 g had an undecomposed fraction weighing 0.01 g. Its content in the meteorite is approximately 2.0%. Diamonds were present in the fractions of all three ureilites (Vinogradov, Vdovykin, 1963).

The fractions of ureilites undecomposed by acids consisted of individual black grains, irregular in shape, sometimes of an angular configuration. The surface of the grains is uneven. In the Novyy Urey and Dyalpur ureilites the grains have a diameter up to 0.3 mm. In the Goalpara meteorite the grains had the largest size (up to 0.9 mm). Under a binocular it could be seen that these

grains constitute an aggregate of smaller grains. Some of these aggregates extracted by the author from three ureilites are shown in Figures 101 and 102.

An X-ray pattern of all the fraction of the Novyy Urey meteorite undecomposed by acids (Table 39) shows that it contains diamond, graphite, chromite, as well as traces of ferronickel, and apparently a coaly substance. The fraction contained not only aggregates containing diamond, but individual chromite grains as well.

Individual grains which were studied by the roentgenometric method were selected from the ureilite fractions. The survey was made on a fiber with FeK (Figure 103, Table 40).

The X-ray patterns of these grains for the Novyy Urey and Dyalpur meteorite are very similar. They revealed the presence of diamond, which corresponds to three lines. Lines characteristic for graphite are also clearly visible. The lines for kamacite, which is present in these aggregates, apparently in very insignificant quantity, appear very faintly. Some of the lines are comparable to the roentgenometric lines of carbonaceous matter. /155

The polymineral aggregates in the Goalpara ureilite produce a large number of lines on the X-ray pattern. On the basis of these lines it is also possible to identify all three carbonaceous forms: diamond, graphite, and coaly substance. There is evidence of presence of traces of kamacite, as well as troilite and chromite; there very tiny grains are found within semicrystalline aggregates in which diamond and graphite predominate quantitatively.

The diamonds in ureilites are similar to ordinary natural diamonds with respect to the size of a unit cell (Table 41).

Thus, the black grains in fractions of ureilites are very fine-grained polymineral aggregates. They consist not only of diamond, but also other forms of carbonaceous matter. Diamond and graphite predominate quantitatively. In the aggregates grown single crystals of diamond are also cemented by graphite and a coaly substance. In the spaces between the single crystals there are very tiny grains of kamacite, and in the Goalpara ureilite, there is also troilite and chromite. /156

This interpretation of the composition of polymineral aggregates containing diamonds is confirmed by a microdiffraction study which we made for the Novyy Urey meteorite using the 400-kV electron microscope-electron diffraction camera (Vinogradov, Vdovykin, Popov, 1965). Figure 104 shows some microdiffraction spectra. The diamond gives a system of concentric rings on the spectra in which it is very easy to see individual reflections, evidence of the degree of crystallinity of the diamonds. Graphite sometimes forms good micromonocrystals. The Laue diffraction pattern from such a single graphite crystal, revealing traces of residual deformation, is shown in Figure 105.

Strong, et al, recently obtained a new form, a hexagonal modification of diamond, by synthetic means. They demonstrated that this modification of diamond is present in the Canyon Diablo meteorite (Strong, et al, 1966) and the

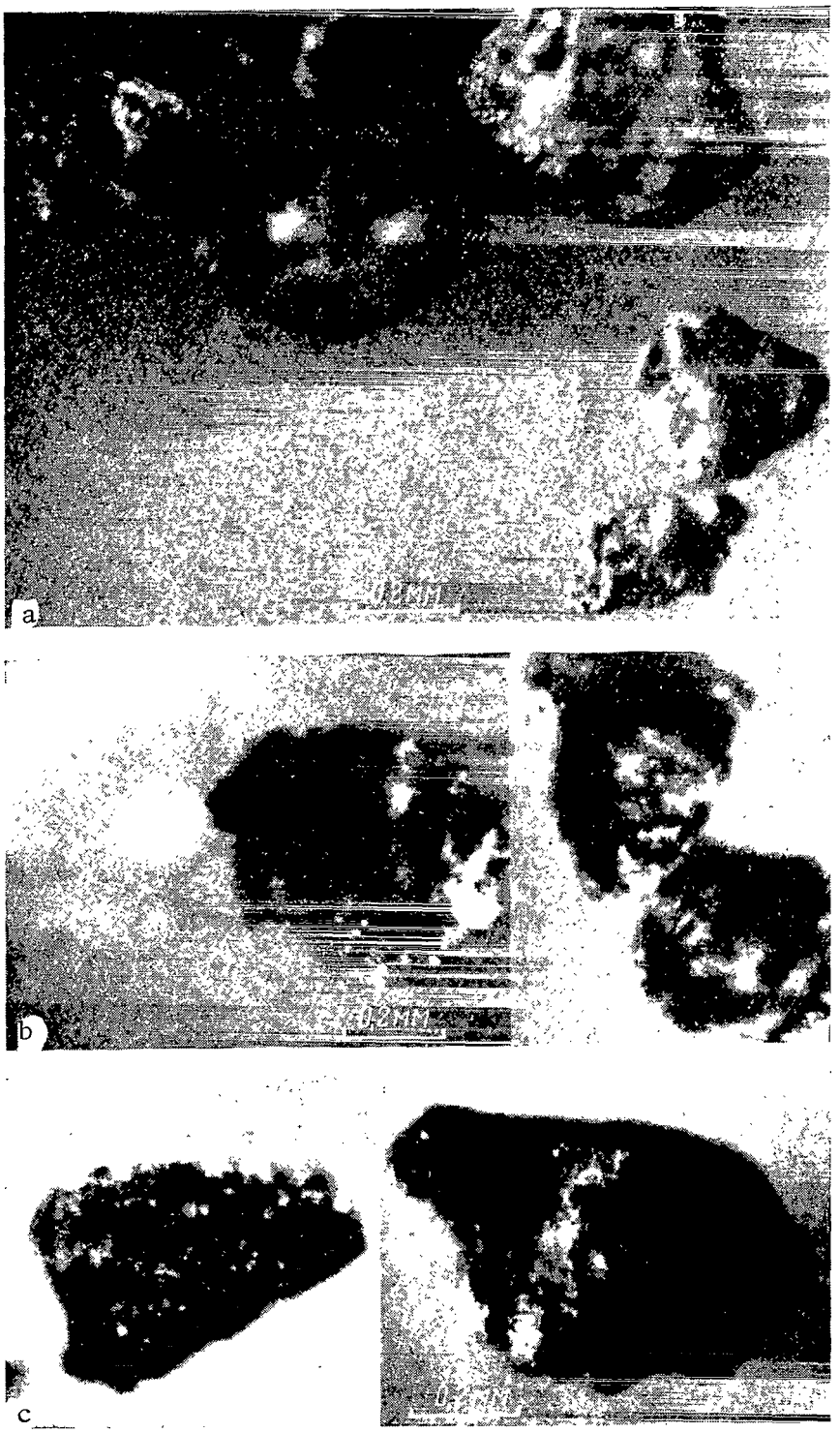


Figure 101. Polymineral Aggregates of Diamond and Graphite in Novyy Urey (a), Dyalpur (b) and Goalpara (c) Ureilites.

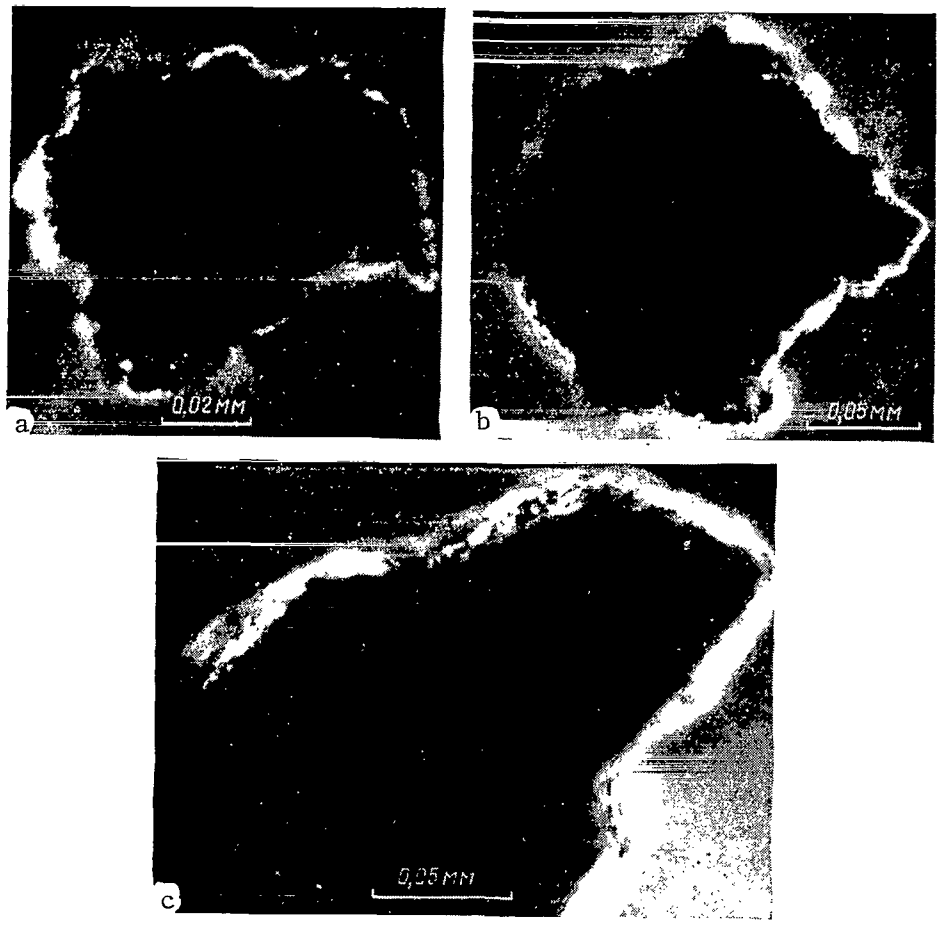


Figure 102. Polymineral Aggregates of Diamond and Graphite Having an Angular Configuration.
a - Novyy Urey; b - Dyalpur; c - Goalpara.

Goalpara meteorite (Hanneman, et al, 1967). According to the data of these authors the parameters of the lattice of the hexagonal diamond are a = 2.52 Å, c = 4.12 Å and its specific gravity is 3.51 g/cm^3. The diamond of the Canyon Diablo meteorite contained about 30% of the hexagonal modification; the diamond of the Goalpara meteorite contained less than 10%. The authors estimated the mean size of the micromonocrystals of the hexagonal and cubic diamonds in the Canyon Diablo meteorite: 50 and 90 Å respectively; the maximum size attained 200 Å. The hexagonal diamond was named Lonsdalite (Urey, 1967).

According to our data, a hexagonal modification is also present in the diamonds of the Novyy Urey meteorite (Figure 104).

Our studies confirmed the earlier known presence of diamond in the Novyy Urey and Goalpara ureilites, and also established the presence of diamonds in the Dyalpur ureilite. The diamonds in the Dyalpur meteorite, as already /157

Table 39

Interpretation of Mineralogical Composition of Fraction Extracted from Novyy Urey Ureilite Undecomposed by Acids (CuK_α)

Fraction of Novyy Urey Meteorite		Diamond (Mikheyev, 1957)		Graphite (Mirkin, 1961)		Chromite (Steulov 1960)		Kamacite (Mikheyev, Kalinin, 1958)	
I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$
4	4,79	—	—	—	—	—	—	—	—
2	4,39	—	—	0,5	4,19	—	—	—	—
10	3,36	10	3,36	10	3,34	—	—	100	3,38
7	3,18	0,5	3,18	0,5	2,995	—	—	—	—
4	2,547	—	—	2	2,547	—	—	—	—
1	2,252	2	2,235	10	2,235	—	—	5	2,12
10	2,051	10	2,058	10	2,074	10	2,05	10	2,02
—	—	1	1,719	9	1,715	—	—	—	—
2	1,672	1	1,680	4	1,648	—	—	10	1,69
2	1,602	—	—	—	—	—	—	—	—
—	—	4	1,539	3	1,519	—	—	—	—
—	—	0,5	1,461	—	—	—	—	—	—
—	—	5	1,384	9	1,381	—	—	—	—
—	—	4	1,307	2	1,325	—	—	—	—
9	1,262	9	1,262	9	1,263	8	1,26	—	—
—	—	0,5	1,247	—	—	—	—	—	—
—	—	5	1,230	5	1,230	—	—	18	1,227
—	—	3	1,152	2	1,128	—	—	—	—
—	—	—	—	4	1,117	—	—	—	—
—	—	—	—	3	1,098	—	—	—	—
8	1,074	8	1,076	8	1,075	7	1,072	—	—
—	—	—	—	5	1,057	—	—	—	—
—	—	6	1,015	4	1,018	—	—	—	—

mentioned, were noted by Lipschutz (1962). He also demonstrated that the fractions of the Dyalpur meteorite which remained undecomposed by acids, contained not only diamond, but also graphite and kamacite. Later, Lipschutz (1964b) investigated all three ureilites and also independently confirmed the presence of diamond in these meteorites. He found that the diamond in the Goalpara meteorite have a very clearly expressed predominant orientation. The X-ray pattern for the Novyy Urey meteorite was more complex, but also gave evidence of a predominant orientation of the diamonds.

Lipschutz noted a predominant orientation as well in the synthetic diamond formed under impact. All this confirmed the earlier expressed hypothesis /158 by Anders (1961) that if the diamonds in meteorites were formed during impact, they would have to have a predominant orientation.

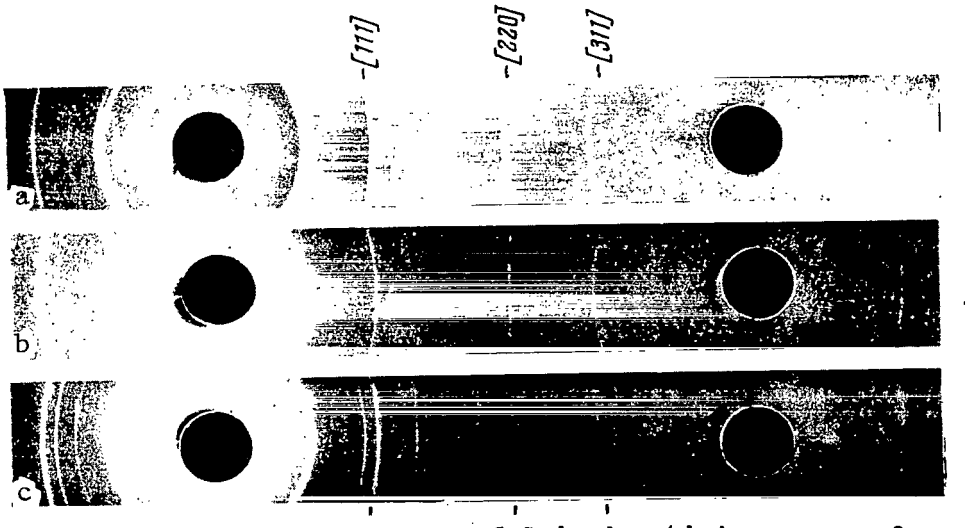


Figure 103. X-Ray Patterns of Polymineral Aggregates Containing Diamond Extracted from the Novyy Urey (a), Dyalpur (b) and Goalpara (c) Ureilites; FeK_{α}, Manganese Filter.

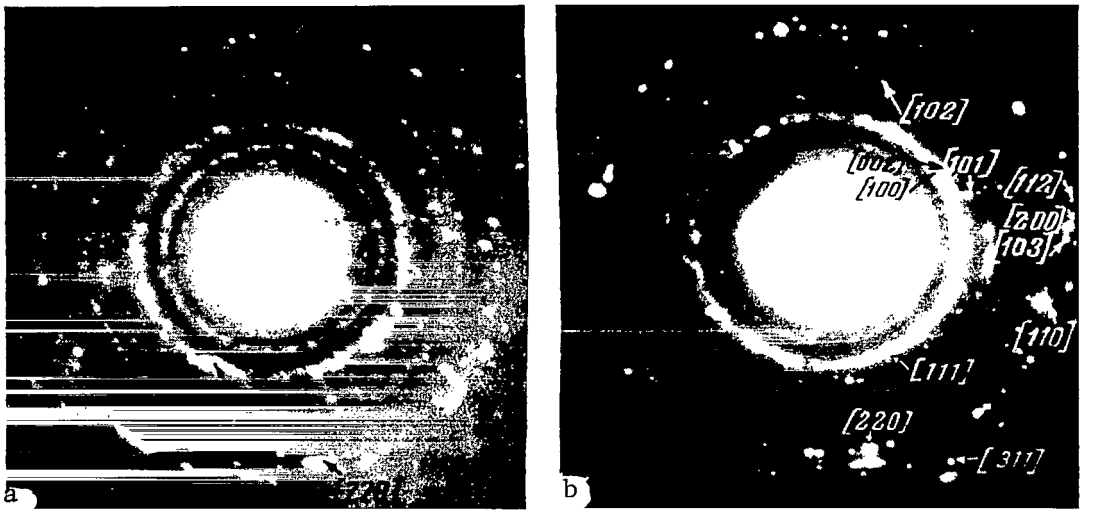


Figure 104. Microdiffraction Spectra of Particles Containing Diamond Extracted from the Novyy Urey Ureilite (Obtained using 400-kV electron microscope-electron diffraction camera). Reflections of Cubic (a), Cubic and Hexagonal (b) Diamonds are Indicated.

Table 40

Debye Powder Diagrams of Polymineral Aggregates in Ureilites Containing Diamond (FeK_α)

Novyy Urey		Dyalpur		Goalpara		Diamond		Graphite	
I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$
4	4,79	—	—	—	—	—	—	—	—
2	4,39	—	—	0,5	4,19	—	—	—	—
10	3,36	10	3,36	10	3,34	—	—	100	3,38
7	3,18	0,5	3,18	0,5	2,995	—	—	—	—
4	2,547	—	—	2	2,547	—	—	—	—
1	2,252	2	2,235	10	2,235	—	—	5	2,12
10	2,051	10	2,058	10	2,074	10	2,05	10	2,02
—	—	1	1,719	9	1,715	—	—	—	—
2	1,672	1	1,680	4	1,648	—	—	10	1,69
2	1,602	—	—	—	—	—	—	—	—
—	—	4	1,539	3	1,519	—	—	—	—
—	—	0,5	1,461	—	—	—	—	—	—
—	—	5	1,384	9	1,381	—	—	—	—
—	—	4	1,307	2	1,325	—	—	—	—
9	1,262	9	1,262	9	1,263	8	1,26	—	—
—	—	0,5	1,247	—	—	—	—	—	—
—	—	5	1,230	5	1,230	—	—	18	1,227
—	—	3	1,152	2	1,128	—	—	—	—
—	—	—	—	4	1,117	—	—	—	—
—	—	—	—	3	1,098	—	—	—	—
8	1,074	8	1,076	8	1,075	7	1,072	—	—
—	—	—	—	5	1,057	—	—	—	—
—	—	6	1,015	4	1,018	—	—	—	—

Thus, all three ureilites contain diamonds. The polymineral aggregates of irregular configuration in which the diamonds are found are distributed rather uniformly through the volume in each ureilite and are found among the silicate grains. These black polymineral aggregates, up to 0.3 mm in diameter in the Novyy Urey and Dyalpur meteorites and up to 0.9 mm in the Goalpara meteorite consist primarily of diamond and graphite, but contain an insignificant admixture of kamacite, and in the Goalpara meteorite, troilite and chromite are also present. In their X-ray characteristics, the diamonds in ureilites are indistinguishable from terrestrial diamonds. However, they contain an admixture of a hexagonal modification of diamond.

Researchers engaged earlier in the study of diamonds in ureilites, other than M. V. Yerofeyev and P. A. Lachinov (1888) and Wentorf, Bovenkerk, (1961), apparently actually did not describe diamonds, but polymineral aggregates containing diamond. Some of these researchers defined these aggregates as black diamonds. /1

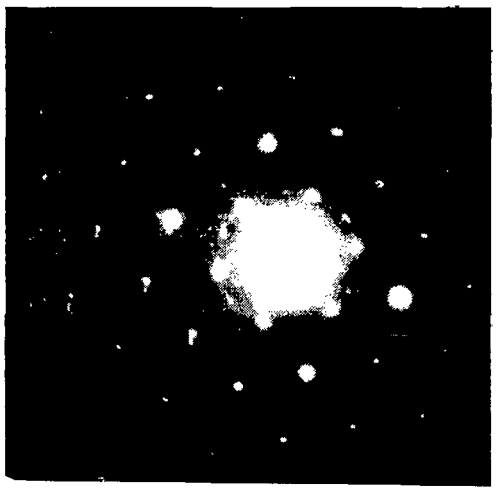


Figure 105. Laue Diffraction Pattern of Graphite Particles Extracted from the Novyy Urey Ureilite (Obtained using 400-kV electron microscope-electron diffraction camera.

A black diamond, as is well known, is a fine-grained diamond (Orlov, 1963; Gomon, 1966, and others). The grains in black diamonds have various colors, including black. In many cases they are rather porous (Figure 106) and contain some quantity of mineral impurities. However, it is well known that these are aggregates of well-consolidated microscopic single tiny crystals of diamond. However, in meteorites we have polymineral aggregates instead. About 10% of these aggregates consist of various mineral impurities (kamacite, troilite, and others), approximately half the remaining part is accounted for primarily by graphite, and only the other half consists of diamond. The microscopic single crystals of diamond in these aggregates are cemented more solidly than in a black diamond; evidence of this is that these polymineral aggregates containing diamond, set in epoxy resin, as well as small pieces of ureilites

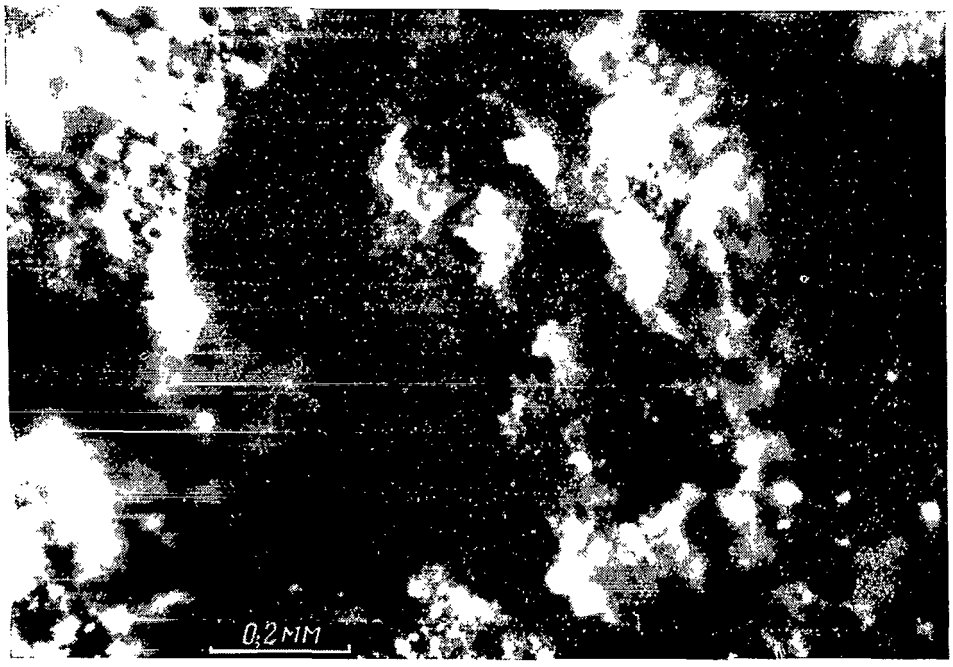


Figure 106. Porous Fine-Grained Black Diamond of Greyish Color from Brazil. The dark sectors are voids.

Table 41

X-Ray Characteristics of Diamonds in Semicrystalline Aggregates in Ureilites (FeK_α)

hkl	Diamonds in ureilites									Diamond	
	Novyy Urey			Dyalpur			Goalpara				
	I	$\frac{d_\alpha}{n}$	a	I	$\frac{d_\alpha}{n}$	a	I	$\frac{d_\alpha}{n}$	a	I	$\frac{d_\alpha}{n}$
111	10	2,051	3,552	10	2,058	3,564	10	2,074	3,593	10	2,05
220	9	1,262	3,568	9	1,262	3,568	9	1,263	3,573	8	1,26
311	8	1,074	3,561	8	1,076	3,568	8	1,075	3,568	7	1,072
a_{mean},	A	—	3,561	—	—	3,565	—	—	3,578	—	3,560

as a whole, are rather easily polished in comparison with a black diamond.

In order to separate out the monomineral fraction of diamond from ureilites it was necessary to eliminate the other mineral components.

Monomineral Fraction of Diamond

In order to obtain a monomineral fraction of diamond the author fused pulverized samples of the Novyy Urey meteorite with sodium peroxide Na_2O_2. Since the graphite and coaly particles within the polymineral aggregates are very small (< 1 μ in diameter), they experience combustion when the pulverized meteorite matter is fused with sodium peroxide in an oxygen flow at a temperature of about 600-700°. Some of the tiniest diamond crystals experience combustion at the same time. This agrees with the data published by A. A. Gumilevskiy (1964) showing that the tiniest diamond grains in a micropowder in part experience combustion at a temperature as low as 500°. For a sample of the nonmagnetic fraction of the Novyy Urey meteorite weighing 1.85 g after fusion with Na_2O_2 at a temperature of about 600°, there was a residue of 8.5 mg of diamonds, less than half of all the diamonds in this meteorite. After fusion at a temperature of about 700° of another sample of the nonmagnetic fraction of the Novyy Urey ureilite (0.985 g) the residue contained approximately 3 mg of diamonds, that is about a third of all the diamonds.

This pure fraction of diamonds from the Novyy Urey meteorite consists of tiny particles of diamond; each tiny particle in turn consists of microscopic individual crystalline grains. The color of these tiny particles is whitish-

light grey. They have an irregular configuration and it is impossible to detect tiny grains having the appearance of crystalline edges (Figure 107). Individual grains measure < 1 μ.

These tiny particles of meteorite diamond are very similar to particles of a synthetic diamond which was specially investigated by the author for comparative purposes (Figure 108); this synthetic diamond had the same morphology, and color as the diamond from the Novyy Urey meteorite. However, the size of the particles of synthetic diamond was far less, almost 0.01 μ, according to data from an X-ray structural analysis.

The X-ray pattern for the monomineral fraction of diamond from the Novyy Urey meteorite, obtained with CuK_{α}, revealed five characteristic lines for diamond (Figure 109, Table 42). The computed unit cell for this diamond has a = 3.552 Å. This is also evidence that the meteorite diamond is indistinguishable from a terrestrial diamond.

As a comparison, the author obtained an X-ray pattern for borazon, a cubic modification of boron nitride BN. This mineral, synthesized at high pressure and high temperature, in its structure and many of its properties is similar /161
to diamond. A unit cell of borazon measures a = 3.632 Å (Table 42).

A pure fraction of diamond from the Novyy Urey meteorite was studied by the electron diffraction camera method and the microdiffraction method using the 400-kV electron microscope-electron diffraction camera. The diffraction spectra revealed concentric circles. However, in most cases the spectra revealed point reflections (Figure 110). This was evidence that the preparation contained individual monocrystals of diamonds of larger sizes.

As a comparison, we made a similar study of the fine-grain powder of synthetic diamond (Figure 111). The spectra for this diamond also revealed only concentric Debye rings (Figures 111, 112), evidence of a less ordered character of the atoms in the structure of this diamond in comparison with the diamond from the Novyy Urey ureilite.

For a more detailed description of the structure of diamonds in ureilites, as well as for a possible clarification of the source of carbon in the formation of diamond, it was of interest to study a monomineral fraction of diamond from the Novyy Urey meteorite using the infrared and electron paramagnetic resonance spectroscopy methods.

It is now assumed that one of the very widely occurring defects in the crystal structure of natural diamond is the substitution of a carbon atom by a nitrogen atom; in this case the unshared nitrogen atom is localized in N-C bonds. In the electron paramagnetic resonance spectrum it is manifested (with an orientatation $H_{\parallel}(100)$) in a system of three narrow lines of equal intensity (Smith, et al, 1959). The correlation between the optical spectra and the electron paramagnetic resonance spectra for natural "nitrogenous" diamonds was recently studied by Ye. V. Sobolev, et al (1964-1965), M. D. Samsonenko (1964), G. O. Gomon (1966) and other researchers. Another type of defect in diamond

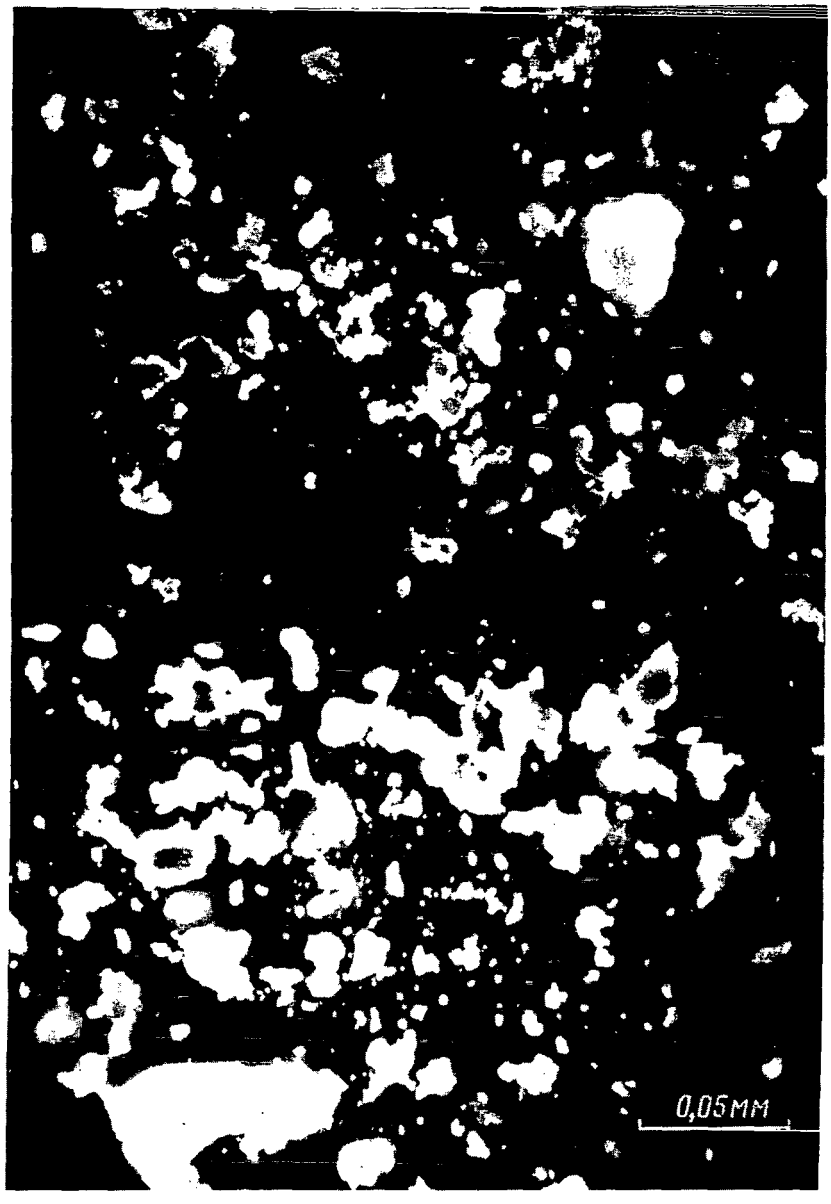


Figure 107. Pure Fraction of Diamond Extracted from the Novyy Urey Ureilite by Fusion of Pulverized Meteorites with Na_2O_2. The particles of diamonds (light) consists of microscopic individual crystalline grains.

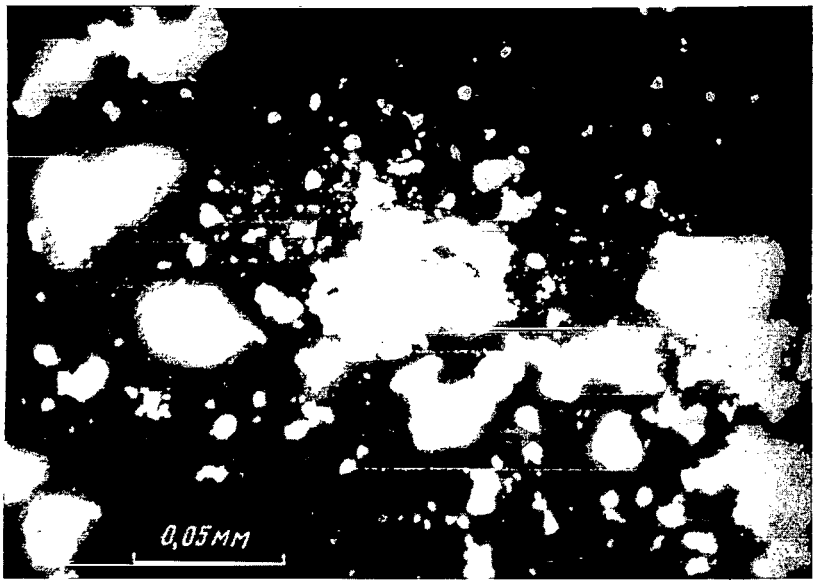


Figure 108. Particles of Synthetic Diamond.

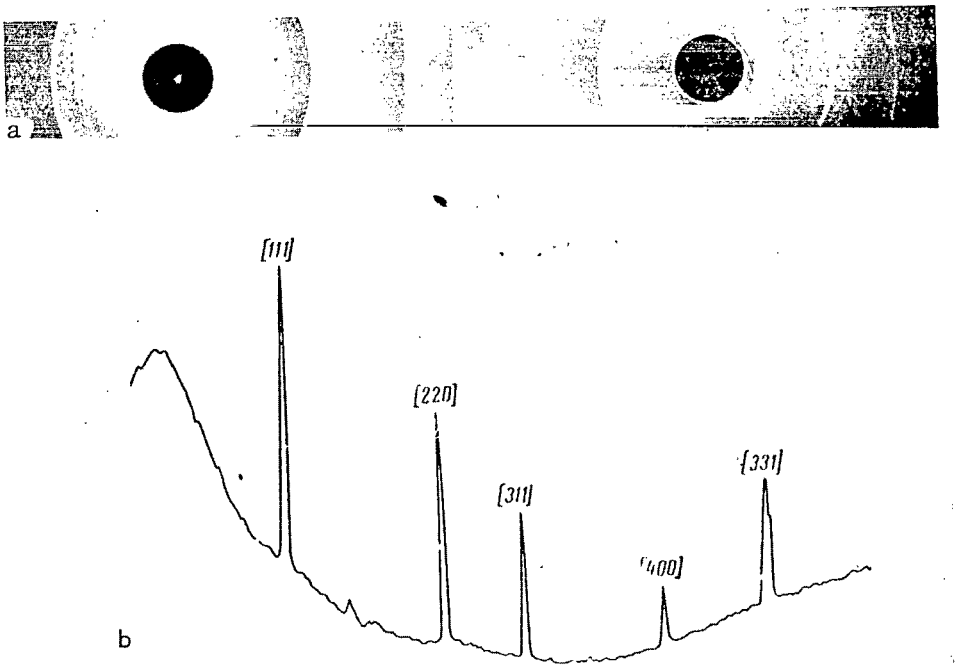


Figure 109. X-Ray Pattern (a) in Photometric Curve (b) of Monomineral Fraction of Diamond from Novyy Urey Ureilite; CuK_{α}, Nickel Filter.

when the microcrystals alternate with zones of an amorphous character, is known. These defects frequently arise during irradiation; it is postulated that in this case a carbon atom is moved from the diamond crystal lattice. On the electron paramagnetic resonance spectrum these defects are characterized by a strong central signal similar to the paramagnetic absorption line observed for graphite. The central electron paramagnetic resonance signal for both "nitrogenous" diamonds and for diamonds with a "mosaic" structure falls /162 in the region $g \approx 2$.

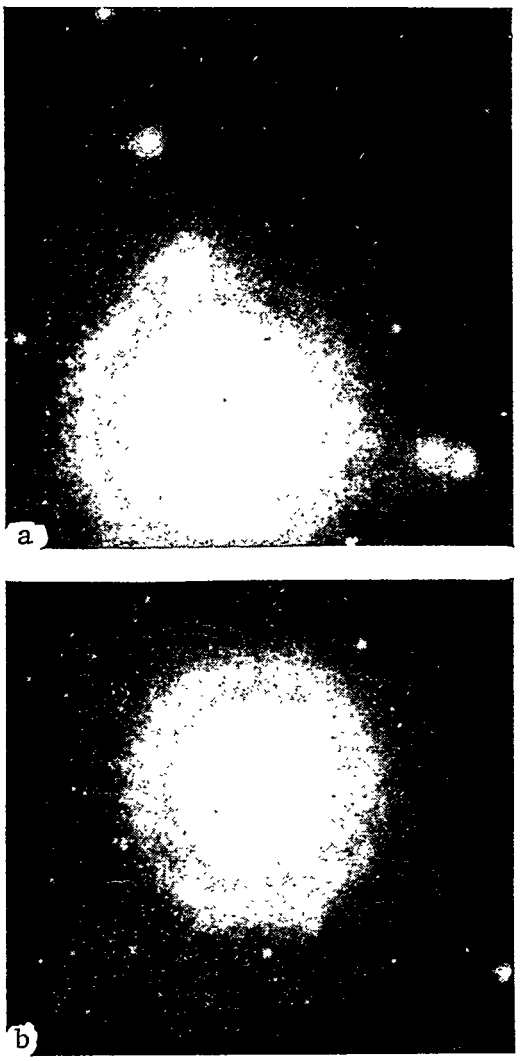


Figure 110. Microdiffraction Spectra of Particles of Diamond from the Novyy Urey Meteorite (Obtained using 400-kV electron microscope-electron diffraction camera).

During 1964 we used the electron paramagnetic resonance method for studying the polymineral aggregates /164 containing diamond and the monomineral diamond fraction weighing 3 mg which were extracted from the Novyy Urey ureilite. On the spectrum the polymineral aggregates gave an unresolved paramagnetic absorption line with a g-factor equal to approximately 2.003 (Figure 113). At its edges it was possible to see very poorly expressed additional lines. The monomineral fraction of diamond from the Novyy Urey meteorite produced on the electron paramagnetic resonance spectrum a quite well-resolved structure of three components, a strong central line and two additional lines of symmetrical configuration (Figure 113,b). The splitting ($\Delta H/2 = 33.6$ oe) has the same value as for the ordinary structure of a donor admixture of nitrogen in natural diamond, but in contrast to the latter the lines on the spectrum of the monomineral fraction of diamond from the meteorite are broader. This gives basis only for postulating with some degree of probability that the structure of the meteorite diamond was caused by a donor admixture of nitrogen. A comparative study of natural diamond from the "Mir" diamond pipe and borazon BN did not allow final conclusions to be drawn.

The nature of the central strong signal has not yet been finally clarified. As a comparison, a study was made of pulverized synthetic diamond (diameter of micromonocrystals ~ 0.01 μ). The electron paramagnetic resonance spectrum for

Table 42

Roentgenometric Characteristics of Monomineral Fraction of Diamond from the Novyy Urey Ureilite and Borazon (CuK_α)

hkl	Diamond from Novyy Urey meteorite			Diamond (Mirkin, 1961)		Borazon BN		
	I*	$\frac{d_\alpha}{n}$	a	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	a
111	100	2,074	3,592	100	2,05	10	2,112	3,657
220	74	1,261	3,564	50	1,26	9	1,284	3,631
311	46	1,068	3,544	40	1,072	8	1,093	3,625
400	19	0,887	3,548	10	0,885	6	0,907	3,628
331	42 25	0,815** 0,813**	3,544	25	0,813	7 3	0,831** 0,829**	3,618
		a Å mean	3,552	3,560				3,632

*Line intensity was determined on the basis of a photometric study of the X-ray photographs (see Figure 109).
**Doublets.

the snythetic diamond revealed a rather strong paramagnetic absorption line (Figure 114). It is not impossible that it could be caused to some extent by an insignificant admixture of graphite in diamond fractions. In order to obtain a final solution of this problem it is necessary that a more detailed study be made.

In a study of a pure fraction of diamond weighing 3 mg, extracted from the Novyy Urey meteorite, the infrared absorption spectrum was unclear because of light scattering. The spectrum revealed weak absorption bands at 985, 1180 and ~1300 cm^{-1}. Later, a better infrared spectrum was obtained in a study of a diamond fraction weighing 8.5 mg (Vinogradov, Vdovykin, et al, 1966). The spectrum of this fraction revealed an absorption band at ~ 500 cm^{-1} and a particularly strong band in the region 900-1300 cm^{-1} (Figure 115). These absorption bands, it is assumed, are characteristic for type-I diamonds containing a nitrogen admixture in their crystal lattice (Figure 116).

Naturally, for a final determination of the structure of diamonds in /165
ureilites further studies will be required. However, on the basis of the data now at our disposal it can be postulated that this diamond is characterized by inhomogeneity in the crystal structure, at least partially caused by admixtures of donor nitrogen.

Thus, the carbonaceous matter in ureilites is represented by three forms: diamond, graphite and an insignificant quantity of organic matter in these me- equilibrium state. The composition of the carbonaceous matter in these meteorites does not make it possible to classify ureilites as carbonaceous meteorites, as in the classification developed by A. N. Zavaritskiy (Zavaritskiy,

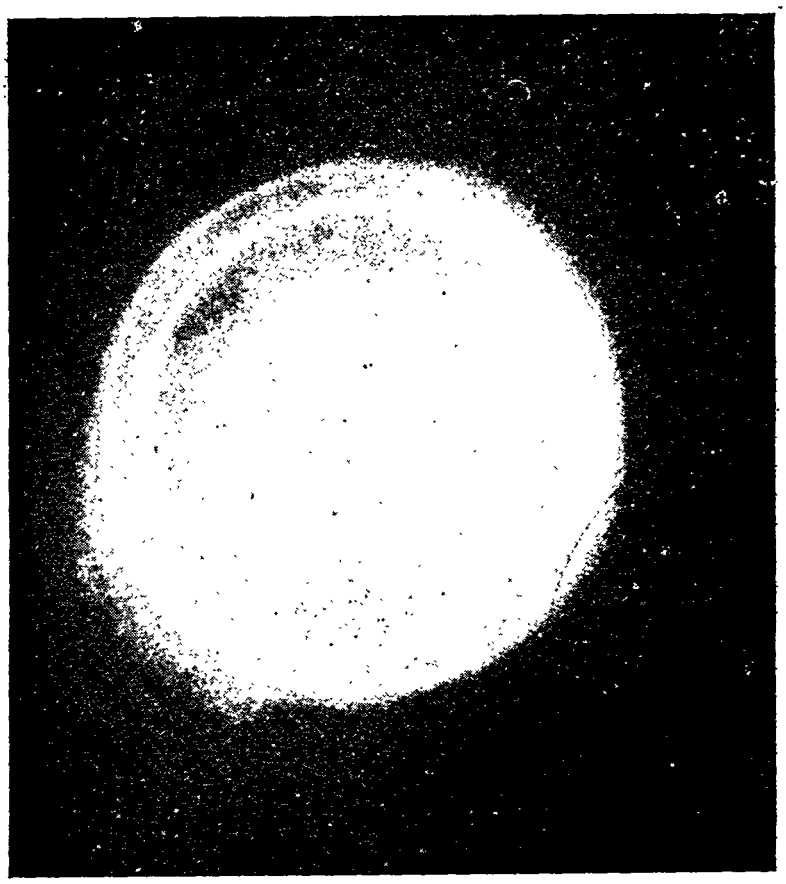


Figure 11.. Electron Diffraction Pattern of Pulverized Synthetic Diamond (Obtained using 400-kV electron microscope-electron diffraction camera).

Kvasha, 1952). It would be more correct to classify these meteorites as diamond-bearing achondrite-ureilites.

In order to confirm this composition of the carbonaceous matter in ureilites, the author studied the Novyy Urey meteorite by the differential thermal analysis method. Figure 117 shows a thermogram for the Novyy Urey ureilite. It differs from the described thermograms for carbonaceous chondrites. /166 On the differential curve for the Novyy Urey meteorite there is a poorly expressed exothermic rise at ~ 340°. It probably is caused by the combustion of bituminous matter present in an insignificant quantity in this meteorite. A small change on the temperature curve corresponds to this rise. Then, beginning at approximately 420°, the differential curve rises gently; the rise reaches a maximum at ~ 850°. In the temperature region 900-950° there is an abrupt dropoff of the curve.

The exothermic effect with a maximum at 850° differs from the effect for the Kainsaz meteorite. In the Novyy Urey achondrite it is caused by the presence of elementary carbon, primarily graphite and diamond, and to an

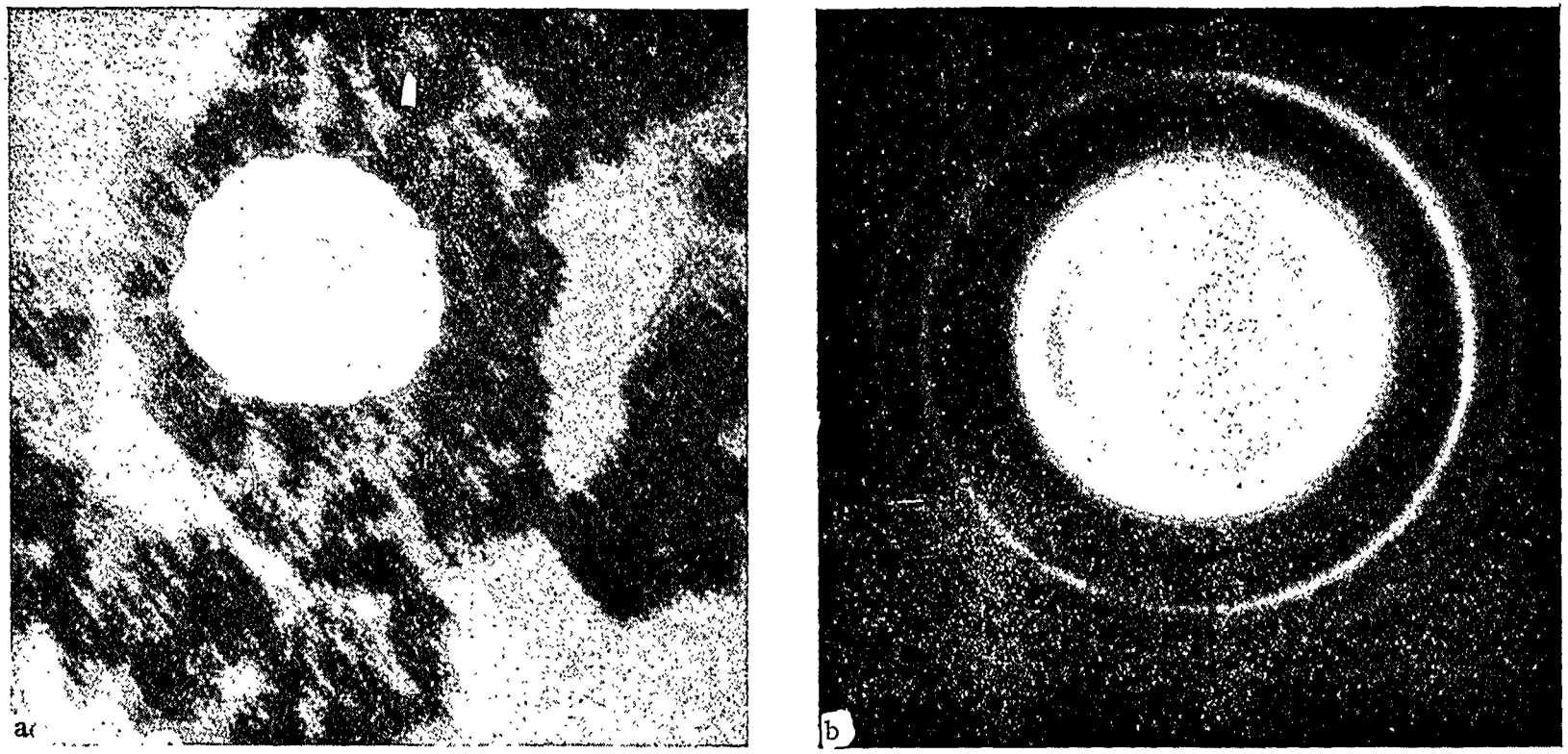


Figure 112. Bright-Field Image (a) and Microdiffraction Spectrum (b) of Synthetic Diamond (Effective diameter of selector diaphragm 1 μ; obtained using 400-kV electron microscope-electron diffraction camera).

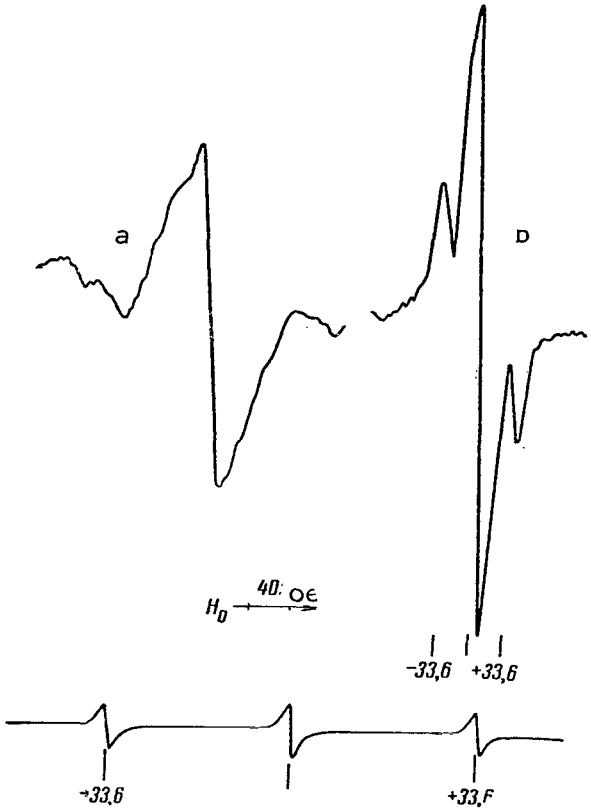


Figure 113. Electron Paramagnetic Resonance Spectra of Carbonaceous Matter in Novyy Urey Meteorite. a - polymineral aggregates of diamond and graphite; b - monomineral fraction of diamond from meteorite; as a comparison the figure shows an electron paramagnetic resonance spectrum of donor admixtures of nitrogen in natural diamond (from Smith, et al, 1959); $H_0 \parallel (100)$ (at bottom).

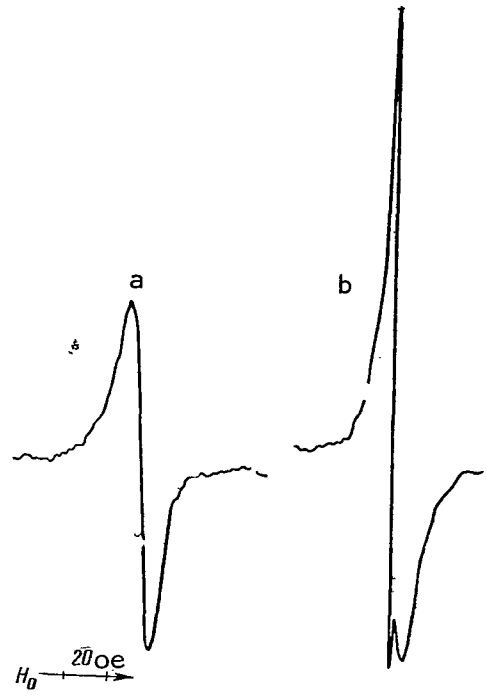


Figure 114. Electron Paramagnetic Resonance Spectrum of Fine-Grained Powder of Synthetic Diamond. a - without standard (DPPH); b - with DPPH.

insignificant degree, carbon with lesser order of the atoms in its structure. The abrupt dropoff of the curve at 900-950° is caused by a relatively rapid combustion of diamond[1], which apparently begins even at the time when the graphite grains have not yet been completely oxidized.

The high-molecular organic matter, like the bituminous matter, is present in this meteorite in only an insignificant quantity. This can be judged from the very small weight loss when the meteorite was heated to 500°, as is indicated by the curve of weight change. At 500-600° the weight increases somewhat, then it decreases very slightly with heating to 920-930°, which corresponds to combustion of the diamond fraction; thereafter it again increases somewhat. However, all these changes in the curve are very /167

[1] After heating powder from the Novyy Urey meteorite the author decomposed it with acids; no diamonds were found.

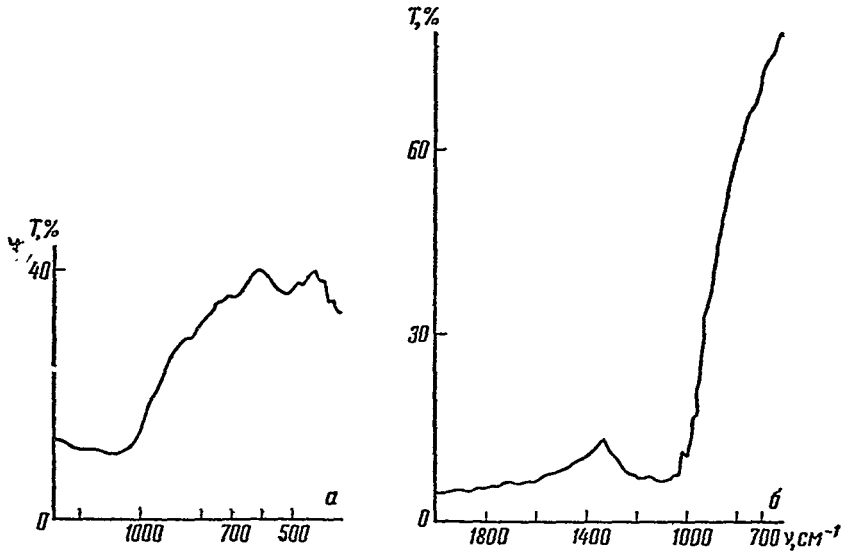


Figure 115. Infrared Absorption Spectrum of Diamond from Novyy Urey (d ≈0.2 mm).
a - under normal conditions; b - with decompensation of second beam by means of auxiliary slits.

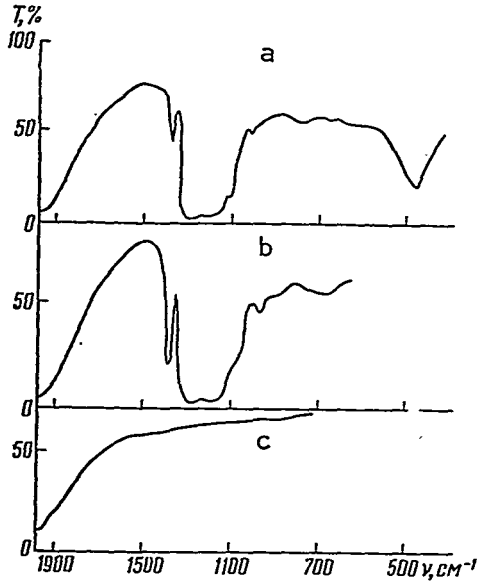


Figure 116. Infrared Absorption Spectra of Terrestrial Diamonds.
a - type-I diamond (d = 2.5 mm; Sobolev, et al, 1964); b - type-I diamond (d = 2.4 mm; Veyr, et al, 1964); c - type-II diamond (d = = 1 mm; Veyr, et al, 1964).

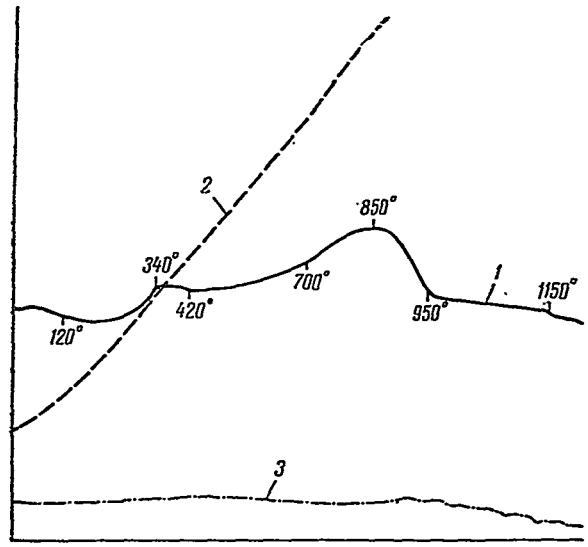


Figure 117. Thermogram of Novyy Urey Ureilite.
1 - differential curve; 2 - temperature curve; 3 - weight change curve.

insignificant; this is due to two processes: combustion of carbonaceous compounds, resulting in a decrease in meteorite weight, and oxidation of metallic grains, which in turn causes a weight increase.

Thus, diamonds in ureilites are found in aggregates with other forms of carbonaceous matter. In addition to such aggregates very tiny grains of ferronickel (kamacite), and sometimes other minerals as well. The diameter of a micromonocrystal of diamond is $< 1\ \mu$. The diamond in ureilite is characterized by inhomogeneity in its crystal structure, caused at least in part by admixtures of donor nitrogen.

GRAPHITE IN IRON METEORITES

CHAPTER SEVEN

CARBONACEOUS MATTER IN COARSE-STRUCTURED OCTAHEDRITES

Carbonaceous Nodules of Yardymlinskiy and Burgavli Octahedrites

Some iron meteorites, especially coarse-structured octahedrites, contain carbonaceous matter (graphite) in the form of individual nodules which have a nonuniform distribution in the ferronickel. The nodules frequently have a rounded configuration and are as great as several centimeters in diameter. The carbon in these nodules is usually associated with troilite and other accessory minerals. /168

Many researchers have noted such graphite nodules in octahedrites. Cohen (1894, 1903) published data to the effect that the nodules in the Cosby's Creek meteorite contained 54.59% graphite, of which 54.04% was accounted for by carbon and 0.55% was an ash residue. The carbonaceous matter in the Magura meteorite contained (in %): 91.75 C and 0.29 H, in the Cranbourne meteorite the corresponding figures were 89.66 C and 0.26 H, whereas in the Toluca meteorite the values were 76.10 C and 0.11 H, with the ash residue 23.50.

Ksanda and Henderson (1939) used the X-ray structural analysis method for investigating the graphite in such nodules in the Canyon Diablo iron meteorite. A roentgenometric study of the graphite from the Yardymlinskiy octahedrite was made by M. A. Kashkay and V. I. Aliyev (1961).

El Goresy (1965) recently made a microscopic study in reflected light and an X-ray spectral microanalysis of nodules from 15 iron meteorites, among them nodules from the Canyon Diablo, Odessa, Toluca and other octahedrites. On the basis of mineral composition he defined three types of nodules: graphite nodules, in which traces of sulfides were observed, graphite-troilite nodules and troilite nodules.

It is known that graphite in metallurgical alloys has similar forms of manifestation. On the basis of this and other peculiarities, it is assumed that iron meteorites in the course of their formation experienced a high-temperature stage in a process somewhat similar to the metallurgical process.

The author (Vdovykin, 1964c) studied the carbonaceous matter in nodules from the coarse-structured Yardymlinskiy (Arus) and Burgavli octahedrites.

Yardymlinskiy (Arus) Octahedrite

The Yardymlinskiy iron meteorite rain fell on 24 November 1959 in the Azerbaydzhan (SSR). A bolide preceded its falling. Six individual pieces weighing a total of 152.5 kg were found.

The meteorite was investigated and described in detail by M. A. Kashkay and V. I. Aliyev (1960, 1961). The mineral composition and structure of the meteorite have also been studied by I. A. Yudin and N. P. Edeleva (1963). /169
A chemical analysis of the meteorite was made by M. I. D'yakonova and V. Ya. Kharitonova (1963). I. Ye. Starik, et al (1961), studied the isotopic composition of the lead in the Yardymlinskiy meteorite, and Goel and Kohman (1962a, b) determined the content of the radioactive isotope C^{14}.

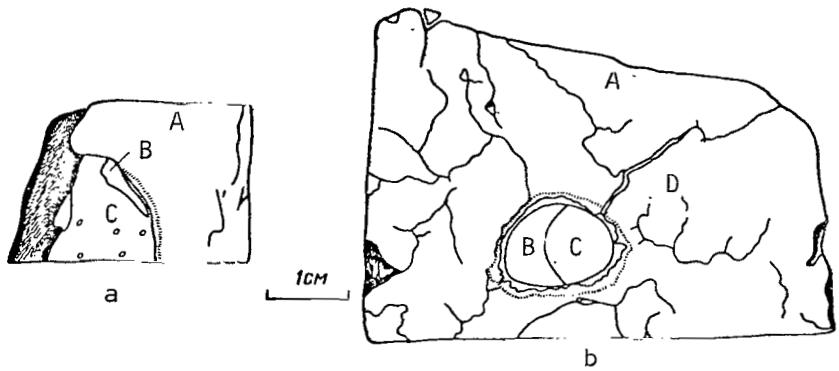


Figure 118. Carbonaceous-Sulfide Nodules in Iron Meteorites--Coarse-Structured Octahedrite Meteorite. a - Yardymlinskiy, Section 2302; b - Burgavli, Section 2260. Drawing made by polished sections. A - ferronickel; B - graphite; C - troilite; D - schreibersite.

Table 43

Chemical Composition (% by weight) of Yardymlinskiy and Burgavli Iron Meteorites (D'yakonova, Kharitonova, 1963).

Element	Yardymlinskiy	Burgavli
Fe	93,19	92,58
Ni	6,30	6,65
Co	0,42	0,65
Cu	0,01	0,03
P	0,10	0,12
Total	100,02	100,03

In mineralogical respects this meteorite consists (Section 2302, Figure 118a) of kamacite and taenite, separated by plessite. In the ferronickel there are grains of schreibersite and needles of rhabdite. Sectors of a fine mixture of graphite and troilite, which form thin veinlets and clusters of an irregular configuration, are frequently found at the joints of kamacite grains. A peculiarity of this meteorite is the presence of numerous nodules of carbon-troilite composition. The configuration of the nodules is rounded or oval. In these nodules graphite sometimes predominates over troilite, but in most cases troilite predominates in the nodules. The nodules are usually surrounded by

a narrow rim of schreibersite. Small schreibersite grains are rather uniformly distributed in the troilite as well. The graphite sectors of the nodules do not consist of carbon alone; they contain many grains of troilite and schreibersite. Table 43 gives the chemical composition of the Yardymlinskiy and Burgavli meteorite.

The author had at his disposal carbonaceous matter from one such nodule, whose chemical composition had been studied earlier by M. I. D'yakonova and V. Ya. Kharitonova (1963). According to their data, this matter contains (in %): Fe 32.56, S 18.91, traces of P and SiO_2, C 48.13.

Burgavli Octahedrite

This meteorite was found in the Burgavli goldfields in the Yakutskaya ASSR in 1941 (Krinov, 1955, 1963a). The meteorite weighed about 25 kg. Almost the entire meteorite is in the Kolymskoye Geological Museum (Magadan).

The chemical composition of the Burgavli meteorite was studied by M. I. D'yakonova and V. Ya. Kharitonova (1963). They noted a similarity between /170 this meteorite and the Yardymlinskiy octahedrite (Table 43). The isotopic composition of the lead in the Burgavli meteorite was determined by I. Ye. Starik, et al (1961).

The Burgavli meteorite, like the Yardymlinskiy octahedrite, consists (Section 2260) of kamacite, taenite and plessite. Grains of schreibersite are visible in the ferronickel and needles of rhabdite can be seen in the kamacite. Like the Yardymlinskiy meteorite, this octahedrite is fissured; the fissures are filled by schreibersite or a fine mixture of troilite and graphite. The fissures are up to 7 mm wide. The Burgavli meteorite also contains troilite-carbon nodules. In the sample obtained by the author for research (Figure 118b) such a nodule has an oval configuration and measures 7 × 15 mm. In the nodule the graphite has a crescent-shaped contact with the troilite. The contact between the graphite and the troilite is clear, with a smooth curvature. Within the troilite it is possible to see small grains of schreibersite. On the whole, the nodule is surrounded by an uneven narrow envelope of schreibersite.

The matter in the carbonaceous nodules of the Yardymlinskiy and Burgavli meteorites has a black color and a fine-grained structure; it is soft and pulverizes in the fingers. Under the microscope in reflected light it can be seen that the individual tiny pieces of these nodules are not formed completely of graphite: aggregates of graphite grains alternate with grains of other minerals, primarily troilite. The troilite seemingly grows through the graphite. The graphite sectors had an irregular configuration with a diameter from several microns to tens of microns.

Results of Study of Carbonaceous Matter in Yardymlinskiy and Burgavli Octahedrite

It is known from the literature that the carbonaceous matter in iron meteorites-octahedrites contain traces of organic compounds. Their possible

presence in the Canyon Diablo meteorite was noted by Ksanda and Henderson (1939). M. A. Kashkay and V. I. Aliyev (1961) used chloroform in extracting the carbonaceous from the Yardymlinskiy meteorite and found that it contained bituminous compounds (0.003% by weight).

The carbonaceous matter in the nodules of the Yardymlinskiy and Burgavli iron meteorite, weighing 0.387 and 0.200 g respectively, was extracted by the author using six organic solvents (Vdovykin, 1964d). It was found that this substance contains organic compounds: in the Yardymlinskiy meteorite 0.037%, in the Burgavli meteorite 0.046%. A similar extraction was performed for graphite from Southern Karelia; organic compounds were not found.

On the infrared absorption spectrum of the bituminous matter extracted from a carbonaceous nodule from the Yardymlinskiy meteorite there were absorption bands with maxima at 2930 and 2857 cm^{-1}. This was evidence that the matter contains saturated hydrocarbons. (Organic compounds in carbonaceous nodules in iron meteorites were recently discovered by Oró and Nooner (1967)).

The carbonaceous matter in the Yardymlinskiy and Burgavli meteorite was investigated by X-ray structural analysis and microdiffraction methods.

The X-ray photographs of this substance in two meteorites, obtained with FeK_{α}, are rather comparable to one another (Vdovykin, 1964c) (Figure 119, Table 44). On these photographs, in addition to the lines characteristic for the basic component of these nodules, graphite, there are lines for troilite and schreibersite.

A carbonaceous fraction was extracted from the Burgavli meteorite, purified from other mineral components by processing with HF and HCl, for a microdiffraction study which we made using the 400-kV electron microscope-electron /1
diffraction camera (Vinogradov, Vdovykin, Popov, 1965). The carbonaceous matter in the Burgavli meteorite consists almost entirely of graphite[1]. Figure 120a shows a laue diffraction pattern of a graphite particle. As a comparison with the graphite in the meteorite, the author used an ordinary electron microscope for studying terrestrial graphite (Figure 120b).

In addition to graphite, the carbonaceous matter in the Burgavli meteorite contained an insignificant quantity of aromatic carbon with a poor order of the atom in the structure[2] and high-molecular organic matter of crystalline and amorphous structure. The crystalline polymers frequently have the appearance of monolithic grains, sometimes being fiberlike. Figure 121 shows a typical example of the fibrous texture.

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[1] This was recently also demonstrated by Sciacca Lipschutz (1964), who used an electron microscope in studying the carbonaceous matter in the Canyon Diablo iron meteorite.

[2] El Goresy (1965) compares the carbon particles in iron meteorites to retort graphite, palmetto graphite and other types.

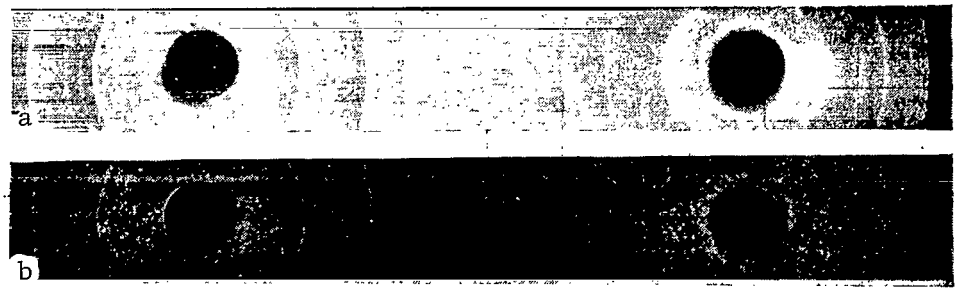


Figure 119. X-Ray Pattern of Substance in Carbonaceous Nodules of Iron Meteorites.
a - Yardymlinskiy; b - Burgavli; FeK_{α}.

Thus, the carbonaceous matter in iron meteorites is represented primarily by graphite, but there is also an insignificant quantity of other types of carbonaceous matter, especially organic compounds. In its structure this graphite does not differ from terrestrial graphite.

The author (Vdovykin, 1967a) used the microhardness method to determine the hardness of graphite in the Yardymlinskiy meteorite. On the basis of sixteen determinations on a PMT-3 instrument designed by M. M. Khrushchov and Ye. S. Berkovich the microhardness of the graphite in this meteorite was found to vary in the range 7-12 kg/mm^2; H_{mean} = 10 kg/mm^2. As a comparison, we studied the hardness of terrestrial graphite from Southern Karelia. Its microhardness was from 7-12 kg/mm^2 (21 determinations); the mean hardness, determined from the variation curve was 9 kg/mm^2. These data agree with the results of determinations of hardness of terrestrial graphite published in a book by S. I. Lebedeva (1963) (7-12 kg/mm^2, H_{mean} = 10 kg/mm^2). Therefore, the graphite in iron meteorites does not differ from the hardness of graphite in terrestrial samples.

Cliftonite of the Canyon Diablo Octahedrite

In 1846 Haidinger (1846) noted small black grains of cubic appearance consisting of carbon in the Magura iron meteorite. They were assumed to be a special variety of carbon and later were named cliftonite. Cliftonite is not found in rocks. Then cliftonite was also found in other iron meteorites; Youndegin, Cosby's Creek, Toluca, Smithville, Canyon Diablo (Anders, 1964; El Goresy, 1965). In these iron meteorites cliftonite is found in the carbonaceous zones.

According to the data cited in Cohen's book (1903), the cliftonite from /174
the Toluca contains 94.44% C and 0.33% H.

Cliftonite was recently found in other (stony) meteorites; the indarkh (Ramdohr, 1963) and Jajh deh Kot Lalu (Keil, Andersen, 1965) enstatic chondrites.

Table 44

X-Ray Patterns of Matter in Carbonaceous Nodules from the Yardymlinskiy and Burgavli Meteorites

Yardymlinskiy		Burgavli		Graphite		Troilite		Schreibersite	
I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$
1	4,69	0,5	4,89	—	—	3	4,72	—	—
1	3,77	1	3,84	—	—	3	3,82	—	—
3	3,72	4	3,71	—	—	—	—	—	—
3	3,43	—	—	—	—	—	—	—	—
10	3,31	10	3,38	10	3,37	—	—	—	—
4	2,963	6	2,970	—	—	6	2,968	—	—
1	2,916	1	2,855	—	—	—	—	—	—
7	2,642	8	2,650	—	—	7	2,644	1	2,652
—	—	4	2,538	—	—	1	2,531	—	—
2	2,511	—	—	—	—	—	—	6	2,506
4	2,301	4	2,305	—	—	5	(2,304)	1	2,270
—	—	2	2,182	—	—	—	—	10	2,190
1	2,148	1	2,145	5	2,129	—	—	7	2,137
10	2,085	10	2,094	—	—	10	2,085	7	2,104
—	—	1	2,060	9	2,019	—	—	8	2,028
—	—	1	1,976	—	—	—	—	10	1,973
3	1,921	1	1,922	—	—	4	1,921	—	—
1	1,887	1	1,900	—	—	—	—	—	—
1	1,838	1	1,830	—	—	—	—	—	—
1	1,788	1	1,775	3	1,783	—	—	8	1,779
2	1,741	—	—	—	—	2	1,755	8	1,757
7	1,715	9	1,716	—	—	2	1,720	—	—
4	1,663	7	1,673	8	1,672	—	—	7	1,678
4	1,633	1	1,637	—	—	5	1,637	—	—
2	1,588	1	1,622	6	1,545	—	—	1	1,593
1	1,498	1	1,496	—	—	—	—	4	1,501
3	1,462	1	1,465	—	—	5	1,472	3	1,465
3	1,445	1	1,445	—	—	3	1,448	—	—
2	1,418	1	1,420	5	1,350	—	—	6	1,411
—	—	3	1,326	—	—	8	1,328	—	—
5	1,324	1	1,324	—	—	6	1,319	1	1,321
3	1,313	—	—	—	—	—	—	—	—
1	1,281	1	1,279	—	—	2	1,279	9	1,277
2	1,268	—	—	—	—	—	—	—	—
—	—	3	1,229	—	—	—	—	7	1,230
5	1,227	1	1,227	9	1,228	3	1,223	—	—
2	1,220	—	—	—	—	—	—	—	—
1	1,208	1	1,201	—	—	—	—	6	1,206
1	1,195	—	—	—	—	—	—	7	1,197
4	1,182	1	1,180	—	—	4	1,182	—	—
2	1,178	—	—	—	—	—	—	—	—

(Table continued)

Table 44 (continued)

X-Ray Patterns of Matter in Carbonaceous Nodules from the Yardymlinskiy and Burgavli Meteorites

Yardymlinskiy		Burgavli		Graphite		Troilite		Schreibersite	
5	1,152	3	1,154	9	1,153	—	—	7	1,148
4	1,129	—	—	—	—	2	1,134	8	1,122
6	1,114	3	1,117	4	1,116	6	1,117	9	1,111
4	1,105	2	1,106	—	—	3	1,106	—	—
2	1,087	1	1,090	—	—	2	1,090	4	1,088
1	1,075	—	—	—	—	—	—	—	—
1	1,050	6	1,050	—	—	6	1,051	8	1,051
5	1,044	—	—	—	—	—	—	—	—
3	1,022	1	1,022	1	1,015	—	—	4	1,012

Table 45

X-Ray Patterns of Cliftonite from the Canyon Diablo Meteorite, Graphite from Iron Meteorites and Graphite of Terrestrial Origin

Cliftonite from Canyon Diablo* meteorite		Graphite from Yardymlinskiy meteorite		Graphite from Burgavli meteorite		Graphite from Burgavli meteorite (purifraction)*		Graphite from Southern Karelia	
I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$	I	$\frac{d_\alpha}{n}$
10	3,41	10	3,31	10	3,38	10	3,42	10	3,37
4	2,145	1	2,148	1	2,145	6	2,144	5	2,129
7	2,053	1	2,060	1	2,060	7	2,066	2	2,019
5	1,766	1	1,788	1	1,775	4	1,829	3	1,783
6	1,677	4	1,668	7	1,673	6	1,677	8	1,672
1	1,542	2	1,588	1	1,622	1	1,542	6	1,545
9	1,233	5	1,227	1	1,227	9	1,233	9	1,228
8	1,156	5	1,152	3	1,154	9	1,156	9	1,153
5	1,115	6	1,114	3	1,117	4	1,118	4	1,116
3	1,016	3	1,022	1	1,022	3	1,016	1	1,015
5	0,993	—	—	—	—	5	0,991	—	—
3	0,826	—	—	—	—	2	0,831	—	—
3	0,801	—	—	—	—	3	0,799	—	—

*Cu-anticathode, Ni filter; others, with Fe-anticathode, Mn filter.

Figure 120. Laue Diffraction Patterns of Meteorite and Terrestrial Graphite. a - graphite from Burgavli iron meteorite (obtained using 400-kV electron microscope-electron diffraction camera); b - graphite from Southern Karelia (Debye rings are for the standard object, aluminum).

Figure 121. Diffraction Spectrum of Fibrous Particle of Carbonaceous Matter from the Burgavli Iron Meteorite. Standard object was aluminum (obtained using 400-kV electron microscope-electron diffraction camera).

Much importance has been given to cliftonite in studies of the origin of meteorites. Some researchers regarded this form of carbon as a diamond pseudomorphism formed at high pressures in the parent body from which the meteorite originated. This point of view was recently supported by Carter and Kennedy (1964, 1966). However, it was demonstrated by Grenville-Wells (1952) that different predominant orientations existed for the graphite of an artificially graphitized diamond and for the cliftonite from the Youndegin meteorite. In the cliftonite the c axis was parallel to the (001) axis, whereas with graphitization of diamond the c axis is oriented parallel to the third-order axis of symmetry of the diamond. Anders and Lipschutz (1966a, b) also pointed out that in the case of graphitization of diamond there is an evitable volume increase by 56%. However, cliftonite is no indication of such an increase in volume; moreover, in comparison with the density of graphite it has a lesser density, approximately 2.12 g/cm^3. Therefore, a diamond cannot be a precursor of cliftonite. Most

researchers feel that cliftonite in meteorites was formed during the decomposition of cohenite.

At our request Dr. Mason supplied us with cliftonite (weighing about 0.05 g) from the Canyon Diablo iron meteorite, many samples of which are preserved in the American Museum of Natural History in New York (Mason, 1964).

The coarse-structured Canyon Diablo octahedrite was found in Arizona (United States) in 1891. Over the course of many years of search a great number of samples (fragments) of different size were collected that had a total weight of more than 30 tons. This meteorite formed a crater. The Canyon Diablo meteorite and meteorite crater have been studied by many researchers and in particularly great detail by D. M. Barringer and H. H. Nininger.

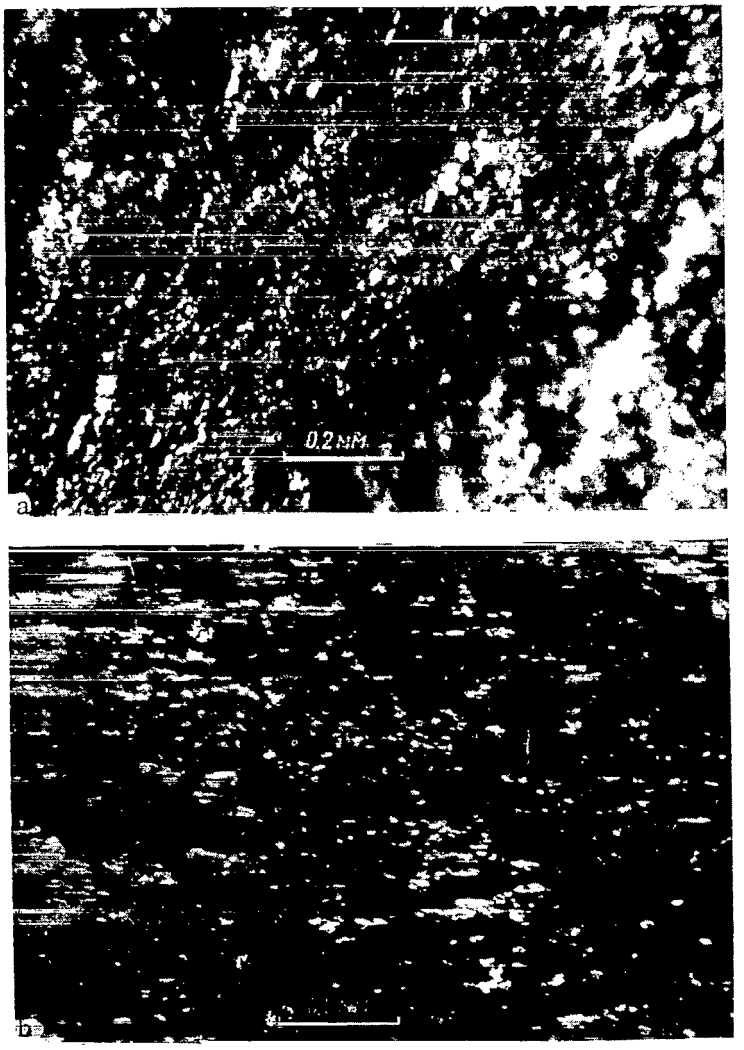


Figure 122. Microstructure of Cliftonite from the Canyon Diablo Iron Meteorite. Reflected light. a - in fracture; b - in polished section.

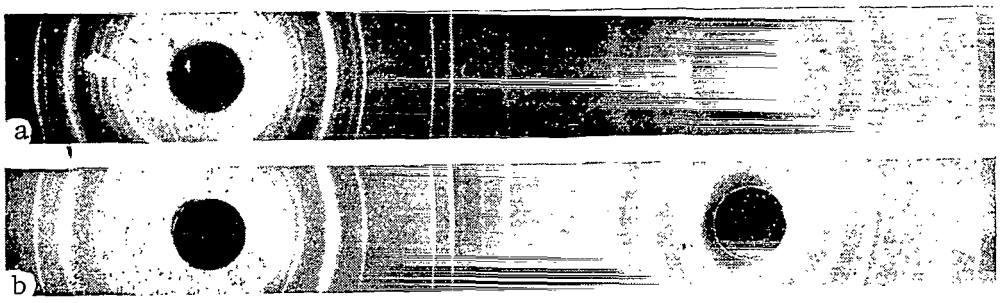


Figure 123. X-Ray Pattern of Cliftonite from Canyon Diablo Iron Meteorite (a) and Purified Graphite from Burgavli Iron Meteorite (b); CuK_{α}.

The Canyon Diablo meteorite diamonds which were formed during the meteorite collision with the earth. Carbonaceous nodules up to 5-10 cm are distributed nonuniformly in different samples of the meteorite. The Canyon Diablo meteorite contains an average of 0.6% C (Brentnall, Akhop, 1962). /17

The carbonaceous matter of this iron meteorite (cliftonite)sent to us consisted of individual small pieces measuring up to 3-4 mm. The small pieces had a black color, a resinous lustre and consisted of tiny carbon grains (Figure 122). Outwardly this matter was very similar to the carbonaceous matter in the Yardymlinskiy and Burgavli meteorites.

In a polished section under the microscope it could be seen that infrequent grains of troilite were distributed through the cliftonite.

The hardness of the cliftonite, determined by the microhardness method, was determined on the basis of ten analyses to be 7-13 kg/mm^2; H_{mean} = = 10 kg/mm^2 (Vdovykin, 1967a). This hardness is characteristic for the graphite in iron meteorites.

An X-ray structural analysis of the cliftonite in the Canyon Diablo meteorite did not reveal structural differences between this mineral and the graphite in iron meteorites (Figure 123). It can be seen from Table 45, which gives the X-ray lines for cliftonite, the graphite in meteors and terrestrial graphite from Southern Karelia, that the cliftonite in the Canyon Diablo meteorite is also similar in its structure to terrestrial graphite. This confirms the data obtained by Hey (1938) and Grenville-Wells (1952), who demonstrated, also on the basis of of roentgenometric study, that the cliftonite from meteorites is similar in its structure to terrestrial graphite.

ORIGIN OF THE CARBONACEOUS MATTER IN METEORITES

CHAPTER EIGHT

EXTRATERRESTRIAL ABIOGENOUS NATURE OF THE ORGANIC COMPOUNDS IN METEORITES

In a discussion of the problem involved in the origin of organic components in meteorites[1] it is customary to examine only the composition of the bituminous matter extracted by organic solvents from carbonaceous chondrites; the remaining part of the carbonaceous matter is rarely taken into account. In comparing the general composition of the bituminous matter in the Orgueil and Murray carbonaceous chondrite with terrestrial organic compounds of biogenous origin, Meinschein, et al (1963) found some similarity between chem and assumed by analogy that the bituminous matter in meteorites also has a biogenous nature. It was postulated that the bituminous matter in meteorites either originated from contamination or was formed as a result of extraterrestrial biogenous processes. However, many peculiarities in the composition of the bituminous matter in carbonaceous chondrites made it possible for a number of authors (Mueller, 1953, 1963a, b; Vdovykin, 1962b, 1964d; Anders, 1962; Hayatsu, 1964; Vdovykin, Vinogradov, 1965; Vdovykin, Vinogradov, 1965) to postulate that it was formed as a result of chemical processes of an abiogenous nature. /177

Obviously, sounder conclusions concerning the origin of the organic compounds in meteorites can be drawn on the basis not only of the composition of the bituminous matter, but also the nature of the interrelationship between this matter and high-molecular organic compounds, the predominant fraction of the carbonaceous matter, and also the interrelationships between the organic compounds and the other forms of carbon in meteorites.

Organic Compounds as an Integral Part of the Carbonaceous Matter in Meteorites

Organic compounds are present in all the investigated carbonaceous chondrites. They are represented by two forms: bituminous matter and high-molecular organic matter. Type-I carbonaceous chondrites (Orgueil meteorite) are most

[1] We recall that in the definition of "organic components of meteorites" the author has in mind not their origin (that is the biogenous nature of these components), but only the chemical composition, the similarity of individual components to products of biogenous activity. In the definition (organic compounds of abiogenous "in organic origin" the paradox is obvious. Many researchers, even in the last century, proposed special names for the bituminous matter in carbonaceous chondrites which were not adopted because this substance is not an individual compound. With these considerations in mind the author desisted from using the term which he had proposed in 1958 for the bituminous matter in carbonaceous chondrites (Vdovykin, 1960).

enriched with these compounds.

The content of bituminous matter extracted by solvents without heating of the latter attains 0.5% of the weight of the meteorite, but with extraction in /178 Soxhlet apparatus it attains even 1%. A component analysis, infrared spectroscopy, indicates the presence of hydrocarbons in this matter. The hydrocarbons, according to data published by Nagy, Meinschein and Hennessy (1961) and Meinschein, et al (1963), contained from 15 to 29 carbon atoms in their chain. According to data from infrared spectroscopy and particularly from a mass-spectrometer analysis (Meinschein, 1963a, b; 1965b), among the hydrocarbons it is possible to identify *n*-paraffins, cycloparaffins and aromatic hydrocarbons.

Aromatic hydrocarbons account for up to 60% of all the hydrocarbons present in the bituminous matter (Meinschein, 1963b). Among them there is a predominance of polynucleate aromatic hydrocarbons which are represented by phenanthrenes (predominant in the Orgueil meteorite) and pyrenes (predominant in the Murray carbonaceous chondrite; Meinshein, et al, 1963), and anthracene (most characteristic for the Migei, Staroye Boriskino and Cold Bokkeveld meteorites; according to V. N. Florovskaya, G. P. Vdovykin and T. A. Teplitskaya, 1964, V. N. Florovskaya, et al, 1965). Some of them, the most characteristic, are listed in Table 46; the same table also lists other organic compounds found in carbonaceous chondrites.

A number of researchers have also identified nonhydrocarbon organic compounds in meteorites; aromatic acids and phenols (among the acids, 2-, 3-, 4-hydroxybenzoic, 3,5-dehydroxybenzoic, 4-hydroxyphenylacetic, et al); fatty acids with carbon atoms numbering from 14 to 28, among which there is a predominance of acids with an even number of carbon atoms; amino acids, in which there is a predominance of serine, glycene, alanine, leucines, glutamic acid, aspartic acid and threonine; carbohydrates (predominance of mannose), as well as amino derivatives of triazine and derivatives of guanidine (melamine, ammeline, adenine, guanine). The total content of all identified organic compounds in carbonaceous chondrites averages only 10-25% of the carbonaceous matter.

According to data from elementary microanalysis, infrared spectroscopy and structural analysis, the overwhelming part of the carbonaceous matter is primarily high-molecular organic matter. As for the bituminous matter, its content increases with a transition from type-III carbonaceous chondrites (Groznaya meteorite) to type-I (Orgueil meteorite). The matter has a condensed aromatic structure; its nuclear part is more apparent than is the peripheral part where the functional groups are situated. This high-molecular substance contains stable free organic radicals, whose unpaired π-electrons are delocalized in a complex aromatic structure.

The high-molecular carbonaceous matter is apparently a complex mixture of polymers with different degrees of order of the carbon atoms. Very tiny grains of carbonaceous matter in which there is a definite order of the carbon atoms in the structure are distributed as an insignificant admixture in the carbonaceous matter. Graphite already predominates quantitatively in the Kainsaz chondrite which contains carbon. Thus, the carbonaceous matter in carbonaceous

chondrites constitutes a complex mixture of organic compounds ranging from bituminous matter and its basic component (aromatic hydrocarbons) to the high-molecular organic matter which predominates quantitatively and which has a condensed aromatic structure, and admixtures of aromatic carbons in which there is some degree of order of the atoms. In addition, carbonaceous chondrites are /183 known to contain other forms of carbon, gaseous compounds (Studier, et al, 1965a) and carbonates (Kvasha, 1948, 1963; Nagy, Andersen, 1964). The general composition of the carbonaceous matter in carbonaceous chondrites and other meteorites, as well as the probably genetic interrelationships among the individual forms of carbonaceous matter in meteorites, which are considered below, are represented in Figure 124. The organic compounds making up almost all of the carbonaceous matter, occurring in a finely dispersed state in the interchondrule space or in the chondrules themselves in carbonaceous chondrites (in the Orgueil meteorite and in some others the carbonaceous matter is represented exclusively by organic compounds), are an integral part of the carbonaceous matter in these meteorites. Obviously, the organic compounds cannot be the result of contamination. They are natural in carbonaceous chondrites.

In ureilites the carbonaceous matter is represented by diamond, graphite, and according to data from a chemical-bituminological and structural study, also by carbonaceous matter. The latter includes high-molecular organic matter and a bituminous substance. The bituminous matter in ureilites accounts for 0.023-0.066% of the weight of the meteorites. In comparison with carbonaceous chondrites, in ureilites it has a reduced character.

In the carbonaceous nodules of iron meteorites, the Yardymlinskiy and Burgavli octahedrites, contain not only graphite, but an insignificant admixture of carbonaceous matter. The high-molecular organic matter has both an /184 amorphous and crystalline structure. The bituminous matter in these meteorites is approximately 0.04%. According to data from infrared spectroscopy, hydrocarbons are present.

The carbon in iron meteorites is also represented by the mineral cliftonite, whose structure does not differ from the graphite of other iron meteorites. In the Canyon Diablo iron meteorite, an octahedrite, other forms of carbon are also known; iron carbide (cohenite) and diamond. Thus, in meteorites organic compounds are an integral part of the carbonaceous matter. They are natural in meteorites and are not the result of contamination.

Comparison of Composition of Organic Compounds in Meteorites and Rocks

The organic compounds in meteorites have some similarity to terrestrial organic matter. For example, the bituminous matter is somewhat similar to ozocerite and other organic minerals, and also similar to the bituminous matter in the serpentinites of the Southern Urals. The high-molecular organic matter has a structure somewhat similar to the structure of terrestrial humic substances.

Table 46

Some Organic Compounds Identified in Carbonaceous Chondrites

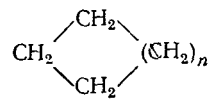
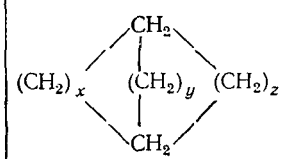
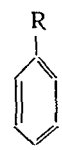
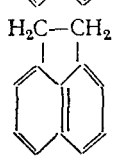
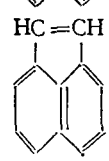
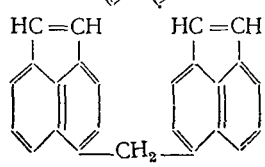
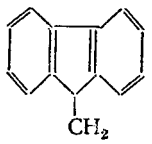
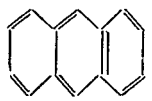
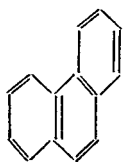
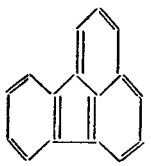
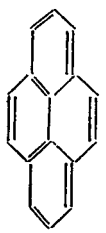
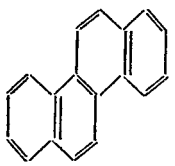
Organic Compounds (classes, groups, individual compounds)	Formula	Carbonaceous Chondrites in which Organic Compounds have been Identified		
		Type-I	Type-II	Type-III
Hydrocarbons				
Saturated Hydrocarbons				
n-paraffins.	C_nH_{2n+2}	Orgueil (1—4) *	Harioura (5) Migei (3,6) Staroye (7) Boriskino Cold Bokkeveld (3) Murray (2, 3, 4, 8)	Groznava (6) Mokoia (3, 4, 9) Kaba (10) Felix (3)
Monocycloalkanes . .		Orgueil (1, 2, 11)	Murray (2, 11)	—
Polycycloalkanes				
Aromatic Hydrocarbons				
Alkylbenzenes. . . .		Orgueil (1, 2, 11)	Cold Bokkeveld (11) Murray (2, 11)	—
Naphthalines		Orgueil (2)	Murray (2)	—
Acenapthenes				
Acenaphthylenes. . .				
Fluorene				(Continued)

Table 46 (continued)

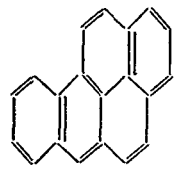
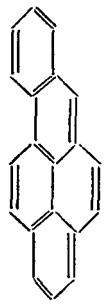
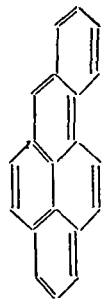
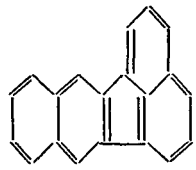
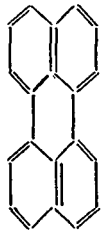
Some Organic Compounds Identified in Carbonaceous Chondrites

Organic Compounds (classes, groups, individual compounds)	Formula	Carbonaceous Chondrites in which Organic Compounds have been Identified		
		Type-I	Type-II	Type-III
Anthracene		Orgueil (12)	Cold Bokkeveld (12)	—
Phenanthrenes. . . .		Orgueil (11)	Migei (13) Staroye Boriskino (13) Cold Bokkeveld (11, 12, 13) Murray (11)	—
Phenanthrene		Orgueil (2)	Murray (2)	—
Fluoranthen . . .		—	Cold Bokkeveld (12)	—
Fluoranthen		—	Cold Bokkeveld (12)	—
Pyrenes.		Orgueil (2)	Murray (2)	—
Pyrene		—	Cold Bokkeveld (12)	—
Chrysenes		Orgueil (2)	Murray (2)	—
Chrysene		—	Cold Bokkeveld (12)	—

(Continued)

Table 46 (Continued)

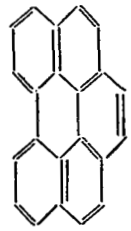
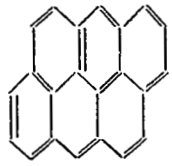
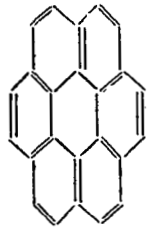
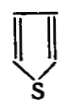
Some Organic Compounds Identified in Carbonaceous Chondrites

Organic Compounds (classes, groups, individual compounds)	Formula	Carbonaceous Chondrites in which Organic Compounds have been Identified		
		Type-I	Type-II	Type-III
Benzopyrenes		Orgueil (2)	Murray (2)	—
1,2-benzopyrene		Orgueil (12)	Cold Bokkeveld (12)	—
3,4-benzopyrene		Orgueil (12)	Migei (13) Staroye Boriskino (13) Cold Bokkeveld (12, 13)	—
11,12-benzofluoranthen		—	Cold Bokkeveld (12)	—
Perylene		—	Migei (13) Staroye Boriskino (13) Cold Bokkeveld (12, 13)	—

(Continued)

Table 46 (Continued)

Some Organic Compounds Identified in Carbonaceous Chondrites

Organic Compounds (classes, groups, individual compounds)	Formula	Carbonaceous Chondrites in which Organic Compounds have been Identified		
		Type-I	Type-II	Type-III
1,12-benzoperylene		—	Cold Bokkeveld (12)	—
Anthanthrene		—	Cold Bokkeveld (12)	—
Protofluorine.		—	Migei (13) Staroye Boriskino (13) Cold Bokkeveld (12, 13)	—
Compounds containing Oxygen				
Aromatic Acids		Orgueil (3)	Haripura (9, Murray (3)	Mokoia (3, 9) Felix (3)
Fatty Acids	$CH_3(CH_2)_nC{<}^{O}_{OH}$	Orgueil (2, 14, 15, 16)	—	—
Carbohydrates		Orgueil (3)	Murray (3, 17)	Mokoia (3)
Compounds containing Sulphur				
Sulfides		Orgueil (11, 18)	—	—
Heterocyclic (phiophenes)		Orgueil (18)	Murray (19)	—

(Continued)

Table 46 (Continued)

Some Organic Compounds Identified in Carbonaceous Chondrites

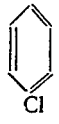
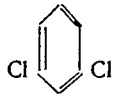
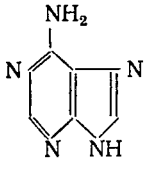
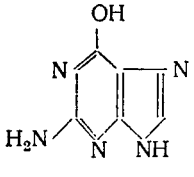
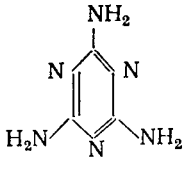
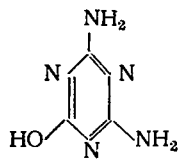
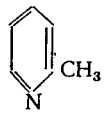
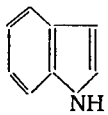
Organic Compounds (classes, groups, individual compounds)	Formula	Carbonaceous Chondrites in which Organic Compounds have been Identified		
		Type-I	Type-II	Type-III
Compounds containing chlorine				
Alkylchlorides		Orgueil (11)	Cold Bokkeveld (11)	—
Chlorbenzene		Orgueil (11)	—	—
Dichlorbenzene		—	Cold Bokkeveld (11) Murray (19)	—
Compounds containing nitrogen				
Amino acids.		Orgueil (3, 20)	Migei (3) Cold Bokkeveld (3) Murray (3, 17)	Mokoia (3) Felix (3)
Cyclic				
Purines:				
Adenine		Orgueil (15)	—	Mokoia (21)
Guanine		Orgueil (15)	—	—
Triazines:				
Melamine.				

Table 46 (Continued)

Some Organic Compounds Identified in Carbonaceous Chondrites

Organic Compounds (classes, groups, individual compounds)	Formula	Carbonaceous Chondrites in which Organic Compounds have been Identified		
		Type-I	Type-II	Type-III
Ammelin.		Orgueil (15)	—	—
Other cyclic:				
Picoline.				
Indole.		—	Murray (19)	—

• 1 — Nagy et al., 1961; 2 — Meinschein et al., 1963; 3—Kaplan et al., 1963; 4—Oró et al., 1965; Oró, Nooner, 1967; 5 — Briggs, 1963c6-Vdovykin, 1926b; 7-Vdovykin, 1964d8 — Calvin, 1961; 9 — Briggs, 1962e; 10 — Sztiókay et al., 1961; 11 — Studier, Hayatsu, Anders, 1965a; 12 — Commins, Harington, 1966; 13-Florovskaya, Vdovykin, Teplitskaya, 1964. 14 — Nagy, Bitz. 1963; 15 — Hayatsu, 1964; 16 — Ryhage, 1965; 17 — Degens, Bajor, 1962; 18 — Murphy, Nagy, 1966; 19 — Hayes, Biemann, 1967; 20 — Vallentyne, 1965; 21 — Briggs, 1961.

All the organic compounds identified in the bituminous matter of carbonaceous chondrites are known under terrestrial conditions. The occurrence of the principal classes of organic compounds in meteorites is somewhat similar to the distribution in various geological objects. Figure 125 is reproduced from Degens and Reuter (1964) with some changes and additions based on data published by Vdovykin (1962a, 1964d) for hydrocarbons and compounds with O, N and S in carbonaceous and stratal waters in petroleum deposits.

However, there are differences within the limits of each class. For /186
example, Meinschein (1963b) notes that the fraction of aromatic hydrocarbons in the Orgueil meteorite differs from the fraction of petroleums in having a simpler composition. In this respect it somewhat approaches recent sediments[1]

[1] Degens (1964) explains this similarity on the basis of the thermal history of transformation of biogenous matter in recent sediments and the chemical synthesis of hydrocarbons in meteorites present in space at a distance of approximately one astronomical unit from the sun where the temperature did not exceed 300-350°K.

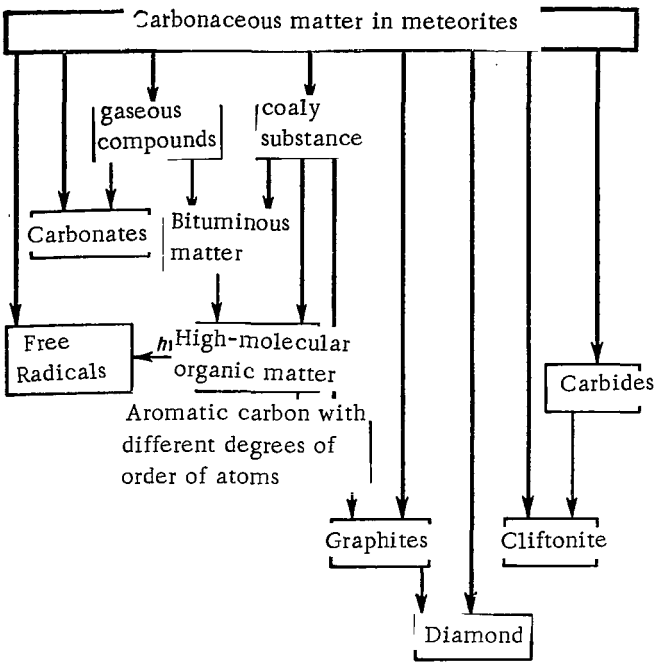


Figure 124. Composition of Carbonaceous Matter and Probable Genetic Interrelationships Among its Different Forms in Meteorites. The thick lines correspond to the composition and the fine lines to the probable genetic interrelationship.

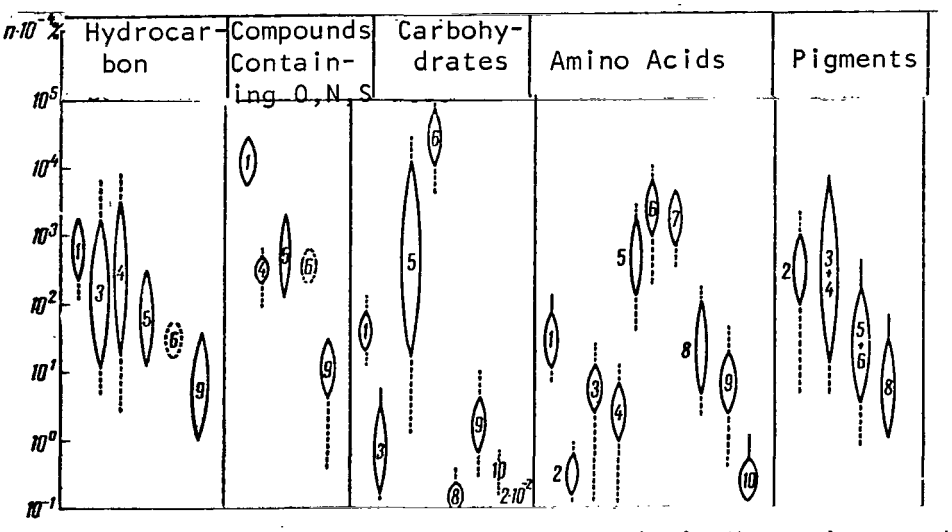


Figure 125. Distribution of Organic Compounds in Meteorites and Various Geological Objects.
1 - carbonaceous chondrites; 2 - petroleum; 3 - shales; 4 - limestones; 5 - recent sediments; 6 - soils; 7 - peat; 8 - coal; 9 - stratal waters of petroleum deposits; 10 - surface waters.

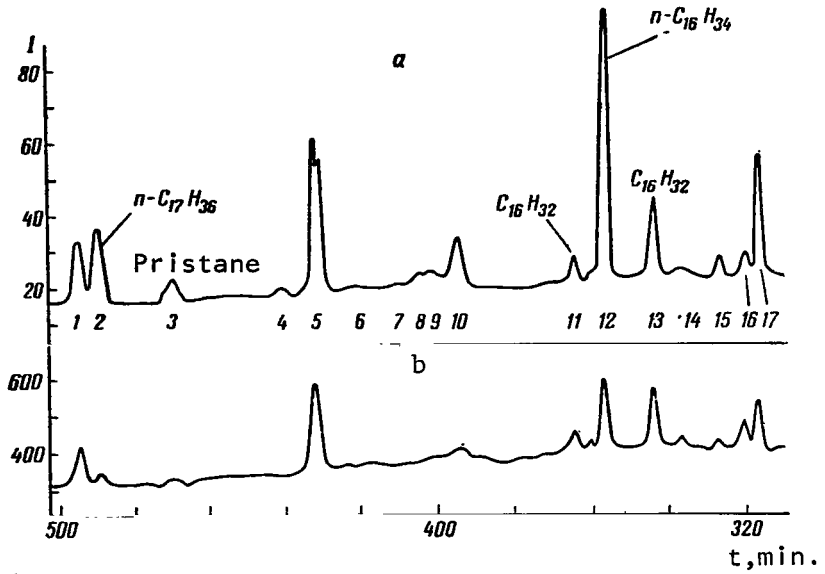


Figure 126. Mass Spectrogram of Organic Compounds of the Series C_{16}-C_{17} and Pristane Synthesized in the Fischer-Tropsch Reaction ($CO + H_2$ = 1:1) with Use of Catalyst (Canyon Diablo Meteorite) for 19 Hours at 300°.
a - m/e = 57; b = m/e = 50-56, 58 → ∞ (t = 164°); I - intensity of peak; t - scanning time. Organic compounds: 1 - ketones (?); 2 - *n*-heptadecane; 3 - pristane; 4 - branched alkanes (?); 5 - phenylalkanes (?); 6-9, not identified; 10 - branched alkanes (?); 11 - $C_{16}H_{32}$; 12 - *n*-hexadecane; 13 - $C_{16}H_{32}$; 14 - alkanes; 15 - alkanes; 16 - dibenzofuran; 17 - pentadecanol (?) (The spectrogram was sent to the author of this book by Professor E. A. Anders on 6 July 1966).

but there is a difference in the distribution of the aromatic hydrocarbons. Anthracenes and naphthacenes are not identified in the organic matter of soils and recent sediments. Soils and recent sediments contain phenanthrenes, chrysenes, pyrenes and perylenes. In petroleums, other 20 aromatic hydrocarbons, there is anthracenes, but there is a predominance of hydrocarbons--phenanthrenes. It is assumed that the aromatic hydrocarbons of petroleum are the products of transformation of isoprehnoids and steroids.

Oró and Nooner (1965) identified saturated isoprehnoid hydrocarbon pristane and phitane in many carbonaceous chondrites. These same hydrocarbons were found in coaly shales in a Precambrian formation in Michigan, whose age

was more than 1 billion years (Meinschein, Barghoorn, Schopf, 1965; Eglinton, Scott, et al, 1965), and also in the Gunflint Precambrian iron ore formation, whose age is estimated at 2 billion years (Oró, Nooner, et al, 1965). It is postulated that the pristane and phitane in these ancient rocks are the products of decomposition of chlorophyll. However, Studier, et al (Studier, Hayatsu, Anders, 1965c), found pristane and phitane in products of abiogenous synthesis by the Fischer-Tropsh method.(Figure 126).

There are differences in the distribution not only of the hydrocarbons, but also other classes of organic compounds. For example, among the aromatic acids in meteorites the most common is 3-hydroxybenzoic acids, in rocks it is vanillic, sirenic, salicylic, 4-hydroxybenzoic, and others (Degens, 1964). Nagy and Bitz (1963) noted that the Orgueil meteorite contained some predominance of fatty acids with an even number of hydrogen atoms. In the ratio of odd fatty acids to even fatty acids they somewhat resemble the organic matter in rocks (micropetroleum, see N. B. Vassoyevich, 1959), and petroleums as well. Among the amino acids found in meteorites the most common are serine, glycene and alanine, as in the organic matter in rocks. However, in different carbonaceous chondrites the relation among amino acids varies. One can note a general tendency to an increase in the content of unstable amino acids[1] in meteorites most enriched with carbon. For example, in the Orgueil meteorite there is a predominance of serine, arginine and phenylalanine. In type-II and type-III carbonaceous chondrites these amino acids are less widely distributed than are the more stable amino acids: glutanic acid, proline, etc. In contrast to the organic matter in rocks, recent sediments and petroleums, pigments, the products of transformation of living matter, from pheophytin to metalloporphyrin complexes, have not been reliably determined in meteorites.

At the present time we do not have extensive data on the composition of organic matter in rocks of endogenous origin. Available data show that it is quite indistinguishable in its composition from the organic matter of sedimentary rocks. However, the genesis of the organic matter encountered in endogenous rocks is frequently unclear.

Many geological examples of occurrence of bitumoids in endogenous rocks /187
are given in studies by N. A. Kudryavtsev (1959), P. N. Kropotkin and K. A. Shakhvarstova (1959), as well as in articles by L. N. Kapchenko (1963a, b), Sylvester-Bradley and King (1963), Robinson (1966) and others. Information on the chemical composition of many natural organic compounds, probably abiogenous in origin, is available in studies made by G. G. Lebedev (1907), N. A. Orlov and V. A. Vspenskiy (1936) and E. Dana (1937). Recently, researchers have made special studies of the chemical composition and geological conditions of appearance of bitumoids in endogenous rocks. A rather detailed study of the organic matter in igneous and metamorphic rocks of the Kola Peninsula and elsewhere has been made by I. A. Petersil'e (1963, 1964a, b; Petersil'e, et al, 1961, 1965). The bitumoids of these rocks contain paraffinic and aromatic hydrocarbons, as well as organic compounds which include oxygen, nitrogen and sulphur. According to data published by N. S. Beskrovnyy

[1] The stability of amino acids at different temperatures has been studied by Abelson (1959) and Vallentyne (1964).

and T. E. Baranova (1963), in the carbonatites from the Kovdorskiy complex of ultrabasic and alkaline rocks of the Kola Peninsula the content of organic compounds is up to 0.004-0.02%; the bituminous matter contains oily and resinified groups. The organic compounds are somewhat similar to the composition of matter found in volcanic types on the Siberian Platform (Beskrovnyy, 1960; Pikovskiy, Guseva, 1963). The results of study of organic minerals, possibly of endogenous origin, found in the Southern Urals, in Transcarpathia, and in other places, have recently been published (Florovskaya, et al, 1964; Grinberg, et al, 1964, 1965).

The pyrite ores of the Urals have a bitumoid content up to 0.2% (Ivanov, et al, 1961). They are represented primarily by paraffinic hydrocarbons. In addition, they contain aromatic structures, alkphatic and aromatic ethers. According to an elementary analysis, these bitumoids contain (in %): C 62.74-75.56, H 8.30-9.25, ash 3.2-8.16. Their component composition, according to data published by V. N. Florovskaya, et al (1964), is: oils 78-88%, resins 9-13%, asphaltenes ~ 3-9%.

Bitumoids have also been noted in extrusive rocks (hundredths and thousandths of a percent). According to data published by N. A. Kudryavtsev (1958), the bitumoids in the extrusive rocks of Armenia consist primarily of paraffinic and naphthene-aromatic hydrocarbons. According to an elementary analysis, they contain (in %): C 75.8, H 11.17, N + S + O 13.03. These organic compounds are similar to the bitumens of recent sediments[1].

Most researchers include thucholites among the most typical representatives of primary carbonaceous compounds. This is an organic mineral containing not only C (43-67%) and H (3-4.5%), but also S, N, O, As and others. Thucholites contain 8-10% hydrocarbon and up to 60% ash residue (Fersman, 1940; Soboleva, Pudovkina, 1957, and others). Thucholites are known in the Khibinyye Mountains, Sweden, Norway, Canada, South Australia, etc. They are found in pegmatites of Archean age. In the pegmatite veins the thucholites are associated with zones corresponding to very definite geochemical phases, indicating a genetic relationship between them and the matter in the pegmatite veins[2]. The thucholites in the pegmatite veins are frequently accompanied by "coaly matter" and even graphite. The thucholites have been poorly studied.

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[1] Juvenile hydrocarbons probably enter into the upper part of the earth's crust along deep vaults in regions of contacts between foredeeps and platforms (Kropotkin, Valyayev, 1965). It can be assumed that the gas from such deposits differs in its isotopic composition of carbon from the gas from ordinary gas deposits. However, it has been shown in our studies that there is no appreciable difference with respect to the carbon (Vdovykin, et al, 1964) or sulphur (Grinenko, Vdovykin, 1966).

[2] Another interesting fact is that the thucholites (Bogard, et al, 1965), as well as the Karelian schungeites (Cherdyntsev, Kolesnikov, 1965) contain high concentrations of inert gases. V. V. Cherdyntsev and Ye. M. Kolesnikov believe that their composition corresponds to the composition of the earth's ancient atmosphere.

Indications of Extraterrestrial Abiogenous Origin of Organic Compounds in Meteorites

In work on geochemical studies of organic matter, until recently there were two best characteristic indications of organic compounds of abiogenous nature: absence of optical activity and porphyrins[1]. /188

Optical activity, that is the capacity for individual fractions of organic compounds to rotate the plane of a polarized ray, is regarded as an indication of biogenous origin of these compounds. It is caused by the presence of an asymmetrical carbon atom in the structure of such matter. Most optically active biological compounds, as is well known, rotate the plane of the polarized ray to the left. However, many petroleums (Dobryanskiy, 1948, 1961; Amosov, 1955), as well as coal extracts (Krayushkin, 1966) rotated to the right. The optical activity of petroleum attains 2.44° (naphthenic petroleums) and even 4.45° (methane-naphthene-aromatic-resinous petroleums) (Starobinets, 1966).

The bituminous compounds in carbonaceous chondrites have been repeatedly checked for optical activity. Mueller (1953), in a study of a 5% benzene solution of bituminous matter from the Cold Bokkeveld meteorite did not find optical activity when checking using a polarimeter with a sensitivity of 0.02°. The author of this book (Vdovykin, 1964d) also failed to determine optical activity in a study of the bituminous matter in the Migei meteorite. Water extracts from the Haripura and Mokoia meteorites, which were studied by Briggs (1963c), were also optically inactive. Kaplan, et al (1963) also checked the organic compounds of a number of meteorites for the presence of optical activity. They investigated the bituminous matter extracted from carbonaceous chondrites by means of organic solvents, fractions containing amino acids, which were combined for a number of meteorites (Orgueil, Murray, Felix), and combined extracts of carbohydrates for the Cold Bokkeveld, Lancé and Warrenton meteorites. These researchers checked optical activity in the region from 400 to 600 mμ using a polarimeter with a sensitivity of 0.0005°; in all cases no such activity was noted.

However, recently Nagy, et al (Nagy, Murphy, et al, 1964), found that the bituminous matter of the Orgueil carbonaceous chondrite, as indicated in an examination on a spectropolarimeter with a sensitivity of 0.005°, is characterized by a rotation to the left, which in the two investigated samples of this meteorite was $\alpha = -0.023°$ (at $\lambda = 435$ mμ) and $\alpha = -0.020°$ (at $\lambda = 440$ mμ) respectively. The authors feel that this could be caused by contamination.

This meteorite was investigated in greater detail by Hayatsu (1964, 1965a,b). Extracts obtained using benzene, chloroform and methanol from 20 g of a sample from the Orgueil meteorite were studied by Hayatsu using spectropolarimeters with sensitivities of 0.001 at 0.0005°. Not one of the fractions (in the case of

[1] However, it recently was found that porphyrin can have an abiogenous formation (Chichibabin, 1957; Fox, 1963; Krasnovskiy, Umrikhina, 1964; Florovskaya, 1964).

concentrations 6-12 mg/ml) had any significant optical location in the region 440 mμ. The maximum value which was noted in this study ($\alpha = -0.0024°$, which corresponds to $[\alpha]_{440} < 0.33°$ ml/g·dm), was in actuality lower than for the control samples for which $\alpha = -0.0053°$ and $\alpha = -0.0093°$. Hayatsu (1966) found that these values were caused by "artefacts" caused by the polarimeter itself. /189
This conclusion was recently confirmed by Rouy and Carroll (1966).

Nagy (1965a, b) used a highly sensitive spectropolarimeter for checking optical activity and found that the optical activity in a saponified extract of the Orgueil meteorite in actuality is not greater than -0.004°. These values are as follows:(with an accuracy ± 0.0003°): -0.0017° (λ = 588 mμ), -0.0021° (546 mμ), -0.0026° (476 mμ), -0.0033° (445 mμ) and -0.0041° (417 mμ). Nagy (1966a) feels that optical activity is either the result of contamination or is natural to the organic matter of the Orgueil meteorite. In investigating the optical activity of the ordinary Holbrook and Homestead chondrites, Meinschein, Frondel, et al (1966) observed small concentrations of optically active compound in the organic matter of these meteorites; it was found that the organic matter for the most part is on the surface of the meteorite. Therefore, in this case the optically active substances were the result of contamination.

A sample of the Orgueil meteorite was checked for the presence of porphyrins by Hodgson and Baker (1964). They found that this meteorite contains an insignificant quantity of vanatic-porphyrinic complexes. However, Kaplan, et al (1963), in studying the Orgueil meteorite and a number of other carbonaceous chondrites, could not identify porphinic pigments in them. The author (Vdovykin, 1966b) also could not find porphyrins in the Migei carbonaceous chondrite.

Baker and Hodgson (1965) noted that the determination of porphyrinic pigments in meteorites is difficult not only due to the small quantity of initial meteorite matter, but also due to the presence of sulfur in the organic extracts.

Thus, neither the presence of optically active compounds nor the presence of porphyrins in the organic matter of carbonaceous chondrites has been confirmed; this distinguishes this matter from products from biogenous activity.

The organic matter in meteorites differs from biogenous matter also in its high content of chlorine (up to 2-3%). In a comparison of the elementary composition of the organic matter in meteorites and organic compound in rocks the author could find in the literature only data concerning the chlorine content in fuel shales, the kuckersites in Estonia, up to 0.002% (Metsik, 1961). However, the chlorine in kuckersites is not organic.

One of the peculiarities of the chemical composition of organic compounds in meteorites, which Mueller (1963a, b; Wilson, 1962) feels is an indicator of the abiogenous origin of these compounds, is a high value of the O/H ratio. For the organic compounds of meteorites, as well as for the thucholites of pegmatite veins this value, according to Mueller, is 0.5-1.0, whereas for

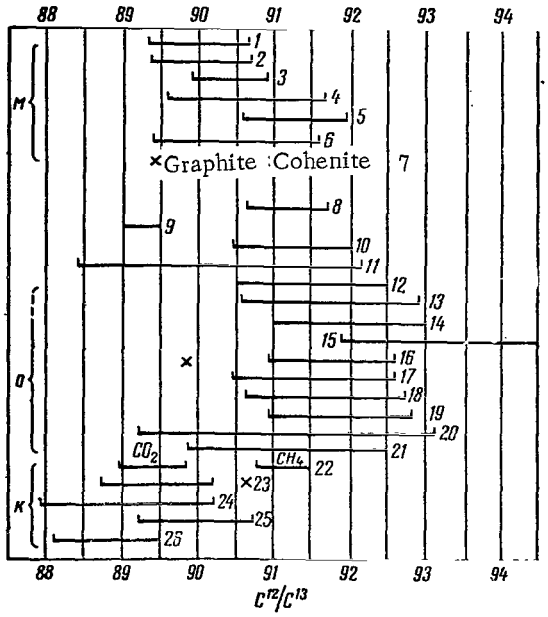

Figure 127. Variations in the Ratios of Stable Carbon Isotopes C^{12}/C^{13} in Meteorite and Terrestrial Carbon.
M - meteorite carbon; O - organic (biogenous) carbon; K - carbonaceous carbon, except for CH_4.

Carbon in meteorites: 1 - bituminous matter (Briggs, 1963b); 2 - carbonaceous chondrites (Boato, 1954); 3 - carbonaceous meteorites (Trofimov, 1950); 4 - stony meteorites; 5 - siderolites; 6 - iron meteorites; 7 - Canyon Diablo meteorite (Craig, 1953).

Terrestrial carbon: 8 - carbon in igneous rocks; 9 - diamond; 10 - carbon in metamorphic rocks; 11 - graphite; 12 - bitumens in igneous rocks; 13 - organic minerals; 14 - petroleums; 15 - fuel gases; 16 - bitumens in sedimentary rocks and recent sediments; 17 - coal; 18 - fuel shales; 19 - peat and fossilized wood; 20 - carbon in plants; 21 - carbon of animal origin; 22 - volcanic gases and the gases from fuel sources; 23 - carbonates of volcanic origin; 24 - carbon in metamorphosed and metasomatic carbonates; 25 - carbonates from zone of oxidation of sedimentary ores; 26 - carbonates of sedimentary origin.

petroleums and other products of the transformation of biogenous matter the value is 0.0-0.25.

In the organic matter of the Orgueil meteorite (Hayatsu (1964) found an increased content of nitrogenous cyclic compounds, which included derivatives of trizane. It is assumed that the latter are not characteristic for the products of transformation of biogenous compounds.

In addition, Meinschein, et al (1963), in studying the aromatic polynucleate hydrocarbons in the Orgueil and Murray meteorites by the mass spectrometer method, demonstrated that their composition is simpler than the composition of aromatic hydrocarbons in terrestrial organic compounds of biogenous origin. This probably can also be evidence of an abiogenous nature of the organic compounds in meteorites. /190

In the future, when it becomes possible to make a still more detailed investigation of the composition of the organic matter in meteorites, we obviously will find additional criteria for determining their abiogenous nature.

The cosmic abiogenous nature of the organic components in meteorites is also evidenced by the isotopic composition of the carbon and hydrogen which was determined by Briggs (1963b) for the bituminous matter in the Orgueil, Haripura, Murray and Mokoia carbonaceous chondrites extracted by means of organic solvents in a Soxhlet apparatus. According to his data, this bituminous matter is enriched in comparison with the organic matter of rocks with a heavy isotop C^{13} and especially D. The high content of D indicated an extraterrestrial origin of this matter. As early as 1954 Boato (1954a) studied the isotopic composition of the carbon and hydrogen in these four meteorites and other carbonaceous chondrites as a whole without extraction of organic compounds from the meteorite. Boato also pointed out the extraterrestrial origin of the greater part of the carbon in the meteorites which he investigated.

Figure 127, based on data in the literature, shows the limits of variations of the ratio of stable isotope C^{12}/C^{13} in meteorites and terrestrial carbon. It can be seen quite well that the carbon in meteorites differs in its isotopic composition from the carbon of terrestrial organic minerals, petroleums, coals, etc.

The results of study of the isotopic composition of carbon, hydrogen and other volatile elements for clarification of the origin of organic compounds in meteorites are considered below in greater detail.

Finally, the interrelationships among organic components and other forms of carbon and the character of the distribution of organic compounds in different meteorites also indicate that the formation of organic compounds could not be caused by secondary processes which might have occurred on the body in which the meteorites had their origin, could not be caused by biogenous processes. The organic compounds in carbonaceous chondrites are distributed not only in the interchondrule space, but also in the chondrules themselves and in the grains of silicates, that is, in the high-temperature mineral formation. In ureilites the organic compounds are found in a nonequilibrium state with other

forms of carbonaceous matter such as graphite and diamond, whose formation was /191 unrelated to biogenous processes. The possible participation of biogenous processes in the origin of the organic compounds distributed in the graphite nodules of such high-temperature formations as iron meteorites is also improbable.

Thus, some peculiarities in the chemical composition of meteorite organic compounds, the isotopic composition of their carbon and hydrogen, as well as the character of distribution of carbonaceous compounds in meteorites, are evidence of an extraterrestrial abiogenous origin of the organic components in meteorites.

Results of Experimental Studies for Synthesis of Organic Compounds

The formation of complex organic compounds by chemical methods, without any participation of biogenous processes, is confirmed by numerous studies for the synthesis of organic compounds. The studies were begun by Miller (1953, 1957, 1961, 1964; Miller, Urey, 1959, Miller, Parris, 1964) and T. Ye Pavlovskaya and A. G. Pasynskiy (1964, "Origin of Life...", 1959), and especially by studies made during recent years[1]. Using various sources of energy from simple initial compounds (CH_4, NH_3, CO_2, H_2O) it was possible to obtain many organic substances, such as hydrocarbons, aromatic acids and phenols, fatty acids, amino acids and carbohydrates, more complex nitrogenous cyclic compounds, that is, virtually all classes of organic compounds found in the organic matter of meteorites. It is well known, that some of the organic compounds (hydrocarbons, fatty acids, and others) are synthesized even in industrial quantities. Exhaustive reviews concerning the synthesis of organic compounds and new information have been published by Oró (1963b, 1965), A. G. Pasynskiy and T. Ye Povlovskaya (1964).

The energy sources used by many authors in the synthesis of organic compounds were: ionizing radiation, ultraviolet irradiation and electrical discharges; in addition, synthesis was carried out solely during heating (Oró, 1965). It became clear from this work that the formation of complex organic compounds occurs with the participation of free radicals.

Low-molecular hydrocarbons (ethane, propane, acetylene, and others) were synthesized by Glasel (1961, 1962) during bombardment of a mixture of CH_4 and NH_3 with electrons at temperatures of 6 and 20°K. Ponnamperuma and Woeller, (1964) also synthesized hydrocarbons of the series C_6/C_9 by the passage of electrical discharges through methane.

Wilson (1960) obtained high-molecular hydrocarbons by the use of electrical discharges in a mixture of CH_4-NH_3-H_2O. In this case a transparent film of

[1] So much data has been accumulated in this field that Oró (1963a, b, 1965) proposed that a special branch of knowledge be defined: experimental organic space chemistry.

hydrocarbon polymers was formed at the liquid-gas content (Figure 128); this film could not be dissolved in organic solvents, and in ultraviolet light it had a yellowish luminescence. At a temperature of 400° it decomposed slowly. The elemementary composition of this polymeric substance (in %) was: C 75.55, H 7.33, N 1.62, S 0, ash 3.79. The use of infrared spectroscopy gave evidence /192 of a polyethylene-type structure, as well as indications of the presence of C = O bonds due to polar groups. In addition, the use of gas-fluid chromatography revealed the presence of hydrocarbons of the C_1-C_5 series in the reaction mixture.

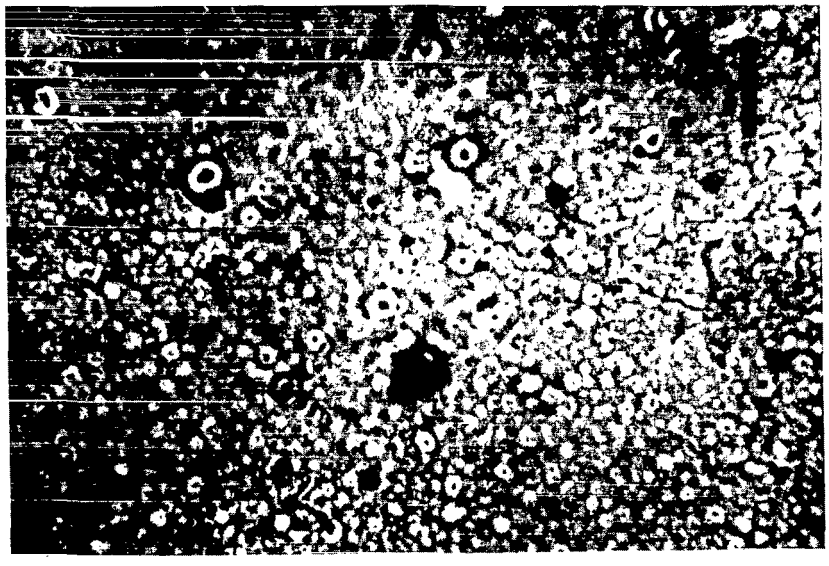

Figure 128. Microphotograph of Hydrocarbon Polymers Synthesized by Wilson (1960). The scale (at top at the right) is 20 μ (reproduced from Fitch, Anders, 1963a).

With the heating of methane to 1000° in the presence of silica gel Oró and Han (1966) synthesized many aromatic hydrocarbons, among them acenaphthylene, fluorene, anthracene, phenanthrene, fluoranthene and pyrene. Recently, Eck, et al (1966) noted polycyclic aromatic hydrocarbons (fluoranthene, pyrene, chrysene, protofluorine) in the products of plasma reactions.

Hooker (Hochstim, 1963) made an experiment with shooting through gas and water media. The velocity of the bullet was 4.6 km/sec, the composition of the gas medium was 40% CH_4, 40% NH_3, 20 H_2. After the shooting organic compounds whose molecular weight attained 200, were discovered in the water.

The industrial synthesis of hydrocarbons on the basis of CO and H_2 (Fischer-Tropsh reaction) is well known; it is carried out at a temperature of 200-300° and at increased pressure in the presence of catalysts (Ni, Co, Fe, activated Al_2O_3, and others) (Storsch, et al, 1954; Azinger, 1959). In this synthesis more than 100 individual hydrocarbons and other organic compounds are obtained[1].

A large number of carboxylic acids with a long chain (up to 40 carbon atoms) was obtained by Stoops and Furrow (1961) by the irradiation of a mixture of CO_2 and C_2H_4 with γ-rays at room temperature. Carboxylic acids have also been synthesized by Berger (1961, 1963), Oró (1963b) and other researchers by the protons irradiation of a mixture of CH_4-NH_3-H_2O at 77°K; amino acids /193 and other organic compounds were synthesized at the same time. Fatty acids have recently been synthesized by Lowe, et al (1963).

It is very easy to synthesize amino acids. They have been obtained from simple initial compounds by the use of various energy sources: irradiation by electron (E = 5 MeV) of a mixture of CH_4-NH_3-H_2-H_2O (Palm, Calvin, 1962), by protons using a mixture of CH_4-NH_3-H_2O (Berger, 1961; Palm, Calvin, 1962), by X-rays, using a mixture of CH_4, NH_3, H_2, CO_2, using electrical discharges (See Figure 67b), by ultraviolet irradiation (Oró, 1963b) and also by heating a mixture of CH_4, NH_3, H_2O to 900-1100° (Harada, Fox, 1964), etc. As a result of synthesis a total of 18 amino acids are formed, of which the most common are serine, glycene, alanine, leucine, and glutamic and aspartic acids, that is, the amino acids occurring most commonly in the organic matter of carbonaceous chondrites.

Carbohydrates are synthesized by the condensation of formaldehyde in water solutions and also under other conditions. It has been noted (Oró, 1963b) that the reaction transpires rapidly at room temperature with the participation of catalysts: calcium, magnesium and other oxides. The synthesis of nitrogenous cyclic compounds has been performed many times by Oró (1963b, 1964). He synthesized purine bases by the heating of a HCN solution to 100°; adenine, guanine and other compounds were formed from this solution during condensation (Figure 129). Synthesis has also been used in obtaining polynucleotides, polynucleosides and many other substances[2] which are also among the

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[1] The mechanism of synthesis has not yet been finally determined. However, using C^{14} it was demonstrated that in any case the intermediate formation of surface carbides of metals, if it does occur, plays a secondary role in the reaction (Brodskiy, 1952). This mechanism has recently been studied in greater detail (Studier, et al, 1965c).

[2] A. I. Oparin, et al (1964) obtained coacervates from polypeptides and polynucleotides.

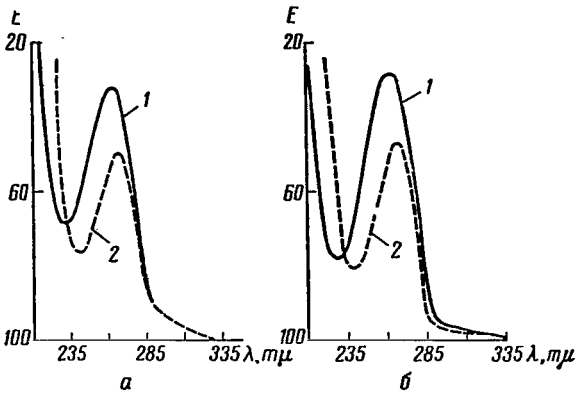

Figure 129. Spectrophotometric Curves of Purine Base-Adenine.
a - adenine obtained by synthesis from $HCN-NH_3$; b - standard adenine; 1 - in 1 n. HCl; 2 - in 0.4 n. NaOH (Orб, 1963a).

biochemically important compounds. These substances are synthesized under more specific conditions and for the time being are still not known in meteorites.

It should be noted that products of abiogenous synthesis were racemic mixtures; they had no optical activity.

High-molecular organic matter of highly condensed aromatic structure, as shown by industrial experience, is also rather easily formed from simpler initial compounds. The capacity of hydrocarbon compounds to form macromolecules is well known. Fox (1964) also synthesized macromolecules during the thermal polymerization of amino acids.

A. P. Vinogradov (1959a, c, 1963, 1964a, b) believes that during an early state of the earth's existence, prior to the appearance of life, the earth's gas envelope contained H_2, CH_4, NH_3, H_2O, and other gases. From these simple initial compounds, under the influence of various energy sources, the organic compounds now obtained in laboratory synthesis could be formed.

The general sequence of abiogenous formation of organic compounds, which could occur in nature, is also confirmed by the experimental data given in Table 47. /194

The organic compounds found in meteorites could be formed in a similar way. It is notable that during laboratory synthesis the yield of organic compounds in general is of the same character as for the organic compounds of carbonaceous chondrites. Moreover, Kaplan, et al (1963), in a study of carbonaceous chondrites, found on the chromatograms many ninhydrin-positive compounds which had been noted only in synthetic products obtained when electrical discharges were passed through simple initial compounds and which had not been observed during the study of terrestrial natural objects.

In a comparison of the hydrocarbons in carbonaceous chondrites and the hydrocarbons of a product of abiogenous synthesis in the Fischer-Tropsch reaction ($CO + H_2$) period (Briggs 1963c) noted some difference between them. For example, among the paraffins in meteorites there is a predominance of normal paraffins, whereas in the synthetic product it is cyclic paraffins which predominate. There are also definite differences in the composition of the atomatic hydrocarbon, as pointed out by Meinschein (1963b). These differences can be attributed to different conditions of formation and a more complex

Table 47

Sequence of Abiogenous Formation of Organic Compounds in Nature (Compiled by Fox, 1963) on basis of experimental data)

Initial Components		Organic Compounds		Macromolecular Compounds
CO, CO_2 and CH_4		Hydrocarbons		Polysaccharides
NH_3 of N_2	→	Organic acids	→	Proteins
H_2O	Irradiation by β- and γ-, X-rays, heating, ultraviolet irradiation (solar), electrical discharges	Amino acids	Heating	Nucleacic acids
H_2		Purines		
H_2S		Pyrimidines		
Metal		Porphyrins, etc.		
H_3PO_4				

system of initial components in the meteorites matter. The predominance of a normal series of paraffins in meteorites can be evidence of conditions for the synthesis of these paraffins somewhat different from the conditions for the synthesis of hydrocarbon polymers performed by Wilson, that is, there could be a reaction between the methyl radicals and organic molecules in the surface layer of the organic compound. This phenomenon was noted by Briggs (1963c) for organic compounds exposed in an electron beam.

However, Studier, Hayatsu and Anders (1965b, c) did not find significant differences in the composition of organic matter in meteorites and products of abiogenous synthesis obtained in the Fischer-Tropsch reaction. These investigators carried out a series of experiments for the synthesis of hydrocarbons from a $CO + H_2$ mixture; they used the Canyon Diablo iron meteorite and the ordinary Bruderheim chondrite as catalysts. The distribution of hydrocarbons in the synthetic products (See Figure 126) and in the meteorite was rather similar and agreed with the distribution computed by Dayhoff, et al (1964) for equilibrium conditions at 500° in a C-H-O-N mixture.

The results of experimental studies on the synthesis of organic compounds show that the most probable precursors of organic matter in meteorites were simple gaseous compounds. Graphite, which under the influence of cosmic and /19
solar irradiation gave rise to a complex series of organic compounds in carbonaceous chondrites, scarcely was a precursor of this substance. It is known that under the influence of irradiation defects in crystal structure are formed in graphite. Many researchers have shown that during the irradiation

of graphite the mean interplanar spacing in its crystal lattice is increased and the distance between atoms decreases within the lattice. At the same time the crystallites are pulverized[1].

If the primary form of carbon in meteorites was graphite, it would be very difficult to explain the regular change in the quantitative content and qualitative composition of organic compounds in different types of carbonaceous chondrites, the regular change in the degree of carbonization and enrichment with organic components only in the matter in carbonaceous chondrites. It is simpler to attribute all these phenomenon to metamorphism of organic compounds, the result of which is their carbonization[2]. Such metamorphism is well known under terrestrial conditions (Figure 130).

Nevertheless, the carbonaceous matter in meteorites in space undoubtedly /196
experienced exposure to cosmic and solar irradiation. The degree of this irradiation of the carbonaceous matter in meteorites for the time being is still difficult to evaluate. We do not exclude the possibility that cosmic irradiation could cause the formation of free organic radicals in carbonaceous chondrites.

However, after the agglomeration of the parent bodies from which meteorites originated only their upper zones were evidently subjected to cosmic irradiation. Under the influence of irradiation there scarcely was a significant change in the calm position of carbonaceous matter in the meteorites themselves which had formed as a result of fragmentation of the parent bodies. It does cause formation of local defects in the carbonaceous matter (Bollmann, 1961, Bollmann, Maringer, 1964).

Thus, the organic compounds in meteorites as part of the carbonaceous matter are an integral component, characteristic of meteorites. The composition of the organic compounds in meteorites in the first approximation is somewhat similar to the composition of organic matter in rocks, but it differs in a number of characteristics. These characteristics indicate an abiogenous origin of the organic components of meteorites, as is confirmed by the results of studies on the synthesis of organic compounds from simple initial substances. Some similarity in the composition of the organic compounds in meteorites and rocks is evidence that the carbon, especially the organic compounds, regardless of their initial origin, have a similar character of transformation under certain conditions.

The organic compounds in rocks, as is well known, have a biogenous origin. The process of transformation of living matter, which was the basis for these compounds, is associated primarily with the destruction of

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[1] The effect of irradiation on the structure of graphite has recently been examined in many studies (Klimenkov, Aleksenko, 1955; Klimenkov, 1961; Agranovich, Semenov, 1961; Telegina, et al, 1962; Grafit (graphite), 1964; Konstruktsionnyye...(Construction), 1964; Kurchatov, et al, 1964; Ubbelode, L'yuis, 1965, and others.

[2] This agrees with the hypothesis published by Ramdohr (1963) that the graphite in chondrites could be formed during the thermal decomposition of organic matter.

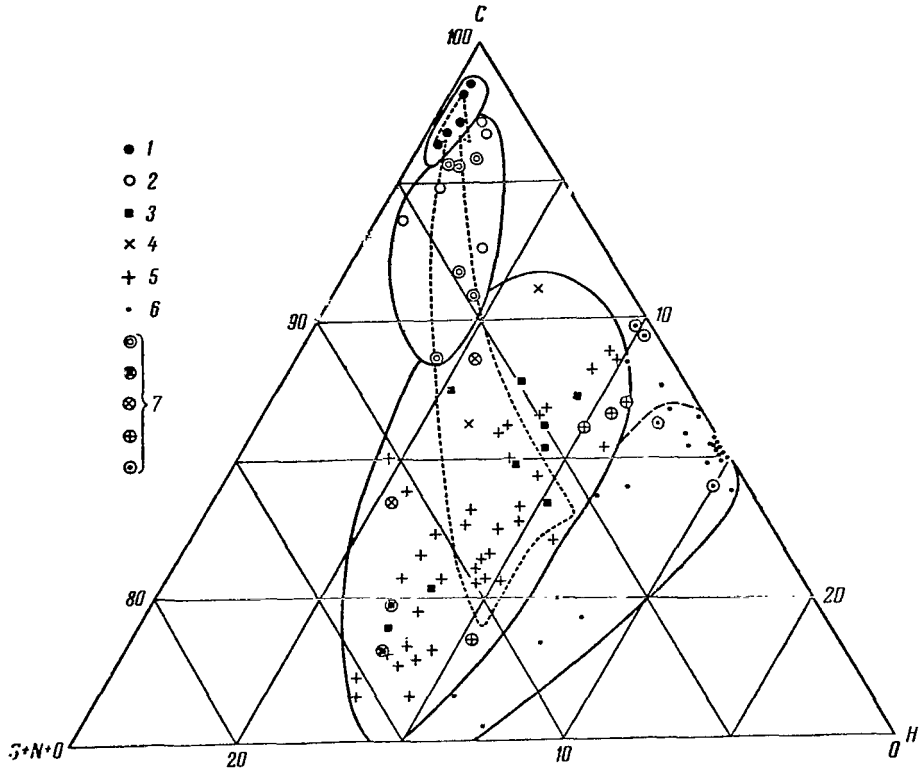

Figure 130. Sequence of Process of Carbonization of Some Organic Minerals (Diagram of elementary composition).
1 - schungeites; 2 - other anthraxolites; 3 - albertites; 4 - intsonites; 5 - asphalts and asphaltites; 6 - ozocerites and other paraffins; 7 - corresponding bitumen shows in igneous rocks.

macromolecules of living systems. This process already begins at the stage of sedimentation and continues during the lithogenesis of sediments. The forming substances later react with one another with the formation of the various organic compounds observed in rocks.

In meteorites, in contrast to rocks, this initial stage (macromolecules of living matter) is absent. The process of formation of organic components begins here directly with simple initial compounds. However, since the chemical nature of carbons is identical, regardless of its origin, biogenous or abiogenous, the end result in the first approximation is identical in both rocks and in meteorites. The only differences are in some peculiarities of the organic components and in part in the distribution of individual components within each class of organic compounds.

CHAPTER NINE

ORIGIN OF METEORITES IN RELATION TO THE ORIGIN OF THE CARBONACEOUS MATTER WHICH THEY CONTAIN

The carbonaceous matter in meteorites, particularly organic compounds, is an integral part of meteorites. Therefore, the conditions of formation of this carbonaceous matter can be regarded only when taking into account the origin and characteristics of development of all meteorite matter. It also goes without saying, that the origin of meteorites containing carbon must be considered together with the problem of the origin of all types of meteorites. /197

General Data

Any of the hypothesis concerning the origin of meteorites is based on the following fundamental initial data (Mason, 1963a).

1. Computations of the orbits of the Sikhote-Alin meteorite, made by Academician V. G. Fesenkov (1951), and also reliable determinations of the orbits of other meteorites, especially the computation made by Ceplecha, 1961) for the observed falling of the Pribram meteorite, indicate an elliptical character of the orbit. This is evidence that meteorites are related in their origin to the solar system.

2. The chemical and mineralogical interrelationship among various classes of meteorites indicate an origin of all meteorites initially from a common parent substance.

3. The atomic distribution of elements in stony meteorites and on the sun, as pointed out by Academician A. P. Vinogradov (1962a), is very similar. This can be evidence of a genetic relationship between the matter found in meteorites and on the sun.

4. Chondrites are the most common types of meteorites: they represent about 86% of all observed falls.

5. Many of the chondrites are bodies with a low density, evidence of their formation in a weak gravity field.

6. The chemical composition of all chondrites, when analyses are scaled for excluding the volatile elements C, H, S, N and O, is rather uniform (Mason, 1965).

7. The chemical composition of the minerals in chondrites is also more or less constant. The minerals in carbonaceous chondrites are an exception.

8. The matter in meteorites is characterized by a relatively high degree of reducibility. In carbonaceous chondrites the chondrules are in a reduced state, but the interchondrule matter in many cases is oxidized.

9. The mineral composition of meteorites is evidence of high-temperature conditions of formation of the minerals in the chondrules and other meteorite components. In carbonaceous chondrites there is a co-existence of "high-temperature" and "low-temperature" minerals.

10. The isotopic composition of C, S and O is rather similar for different types of meteorites, except for carbonaceous chondrites and ureilites. /198

11. Determination of the age of meteorites reveals that their matter was crystallized approximately 4.6 billion years ago.

Clarification of the origin of meteorites is an exceedingly difficult problem. Without dwelling at length on the numerous hypotheses concerning the formation of meteorites, we will mention only a few of the modern hypotheses.

Reviews of many hypotheses and their criticisms have been given in studies by Mason (1963a), Anders (1964) and B. Yu. Levin (1965) and others. These numerous hypotheses have been based on various assumptions concerning the size of the parent bodies from which meteorites originated (planatary, lunar or asteroidal sizes), the number of these parent bodies (one or more), location within the confines of the solar system, origin of the chondrules, initial matter of the meteorites, etc. However, in any hypothesis many of the peculiarities of composition or structure of meteorites will remain unexplained.

Academician V. G. Fesenkov (1965), on the basis of many data, concludes that the process of evolution of asteroidal bodies from which meteorites originated was rather complex and prolonged.

A. A. Yavnel' (1965) concluded that there were several asteroidal bodies from which the meteorites originated. He drew this conclusion on the basis of a study of the composition and structure of meteorites. The same conclusion was drawn by B. Yu. Levin (1959), who recently has given great importance to the role of diffusion in a solid state during the process of separation of phases during the formation of meteorites (Levin, 1964).

The characteristics of the structure of meteorites are evidence that they experience heating during their formation developing the ideas published by Wahl (1907, 1910, 1952) and Lacroix (1926) concerning the heating of meteorites, A. N. Zavaritskiy (1948b, 1950) and L. G. Kvasha (1948, 1956, 1961, 1963) feel that metamorphism plays an important role in the processes of transformation of meteorite matter. In the case of carbonaceous chondrites, whose matter was formed at low temperatures, an important role was played by secondary modification as a result of hydrothermal or pneumatolithic metamorphism or modification as a result of some reactions under the conditions prevailing in space. This modification could occur either after the formation of the chondrites or during the time of their formation.

I. A. Yudin (1956, 1958), on the basis of mineragraphic studies, and M. I. D'konov and V. Ya. Kharitonov (1962), on the basis of a study of the

chemical composition of meteorites, also believes that the matter in the meteorites was locally heated during its formation and this was responsible for the co-existence of light and dark sectors in the same chondrite.

On the basis of a study of the structure and mineral compositions of chondrules in the Saritov meteorite and other nonrecrystallized chondrites, L. G. Kvasha (1965) concludes that the chondrules and nonrecrystallized chondrites were formed simultaneously or at almost the same time.

Some probable characteristics of the chemical fractionation during the formation of meteorite matter was studied by K. N. Savich-Zablotskiy (1960).

It is particularly difficult to explain the peculiarities in the composition and structure of carbonaceous chondrites. Some researchers regard type-I carbonaceous chondrites (of the Orgueil type) as the primary matter from which other meteorites were formed. For example, Mason (1960a, b, 1962b) believes that ordinary chondrites were formed from type-I carbonaceous chondrites as a result of dehydration and the loss of volatile components, in this case chondrules of olivine and pyroxene were formed from hydrous ferro- /199
magnesian silicates as a result of recrystallization in a solid state. This type of metamorphism could occur in small bodies. However, this does not take into account that "high-temperature" silicate minerals are earlier in their origin than the secondary hydrous ferromagnesian silicates. Ringwood (1961, 1962) also regards carbonaceous chondrites of the Orgueil type as primary material. He postulates that the evolution of the matter in chondrites took place in a parent body of lunar size, from which the meteorites originated, and that the chondrules were formed from a silicate melt at a temperature < 1000°.

Wood (1962a, b; 1963a) feels that the Renazzo carbonaceous chondrite, whose chondrules are in a reduced state, whereas the interchondrule substance is in an oxidized state, is a representative of the primary matter from which the planets and asteroids were formed. This same composition of primary matter is accepted by Anders (1964) in his hypothesis of the origin of meteorites. He assumes that the matter in carbonaceous chondrites occupied the surface zones on parent bodies of asteroidal size; liquid water could exist in these zones.

Schmitt, et al (1966) believes that meteorites similar to the Chainpur chondrite, which contains carbon, are the most probable precursors of ordinary chondrites.

However, Urey (1958, 1959a, and others) postulates that carbonaceous chondrites were formed from ordinary chondrites. The oxidation of ferronickel and the appearance of "low-temperature" minerals is attributed by this author to the effect of H_2O, H_2S and other volatile compounds on the matter of chondrites in the H-group. However, he does not discuss the source of the volatile compounds. Urey considers meteorites to be generations of two types of cosmic bodies: primary (of lunar size) and secondary (of asteroidal size). This hypothesis has been subjected to criticism by Anders (1964), Mason (1963a) and other researchers.

As early as 1960 Fish, Goles and Anders (1960) postulated that carbonaceous chondrites correspond to the matter in the upper zone of asteroidal bodies from which meteorites originated. In examining the possible physicochemical conditions of evolution of matter in asteroidal bodies, these researchers concluded that the matter in asteroidal bodies could experience a stage of differentiation provided there was sufficiently rapid heating. They feel that in small bodies, about 500 km in diameter, situated at a distance of 2-5 astronomical units, this differentiation could be sufficiently deep, with the melting out of ferronickel ($T > 1620°K$) at the center of the body, followed by a zone of ferrosulfide eutectic ($1260° < T < 1620°K$) and a zone made up of the matter of ordinary chondrites. The principal energy source during such rapid heating, according to Fish and other researchers, were short-lived, at the present time decaying, radioactive isotopes with a half-life of 10^{-5}-10^{-8} years (Fe^{60}, Cl^{36}, Al^{26}, I^{129}, and others). Long-lived radioactive isotopes (K^{40}, U^{235}, U^{238}, Th^{232}) played an additional role in the process of differentiation of matter in asteroids as sources of energy, as did gravitational energy and the energy of exothermic oxidative-reduction reactions (for example,for formation the sulfides).

There are also other hypotheses of the origin of carbonaceous chondrites (Urey, 1966b; Ringwood, 1966; Wood, 1967; Larimer, Anders, 1967; Dodd, et al, 1967, and others). On the basis of a study of the Murray meteorite, Fredrisson and Keil (1964) postulated that carbonaceous chondrites represent a mechanical mixture of minerals. Some minerals were formed in a low-temperature medium, whereas others were formed under high-temperature conditions. Later these minerals were mixed; in this process drops of ferronickel were /200
introduced into the olivine, impoverished with FeO. Mueller (1964b; Mueller, et al, 1965; Mueller, 1966) studied the interrelationship between the total content of volatiles and some other peculiarities in the structure of carbonaceous chondrites. He feels that the regularities observed in the structure of carbonaceous chondrites can be explained on the basis of the hypothesis of the origin of carbonaceous chondrites as a result of the rapid cooling of "red-hot" particles of matter rising to the surface zones of the parent bodies from which meteorites originated.

It is entirely obvious that many of the processes which have been examined in greater or lesser detail by many researchers unquestionably transpired in a series of different transformations of the matter in meteorites. However, the general direction in their development still remains unclear because we still are unclear about one of the principal problems in any hypothesis of the origin of meteorites: whether chondrules existed even prior to the formation of the meteorite matter as a whole, or whether chondrules were formed in some way or other within the parent bodies from which the meteorites had their origin or in a single cosmic body, that is, we do not know whether chondrules are primary or secondary in relation to chondrites. Various hypotheses exist. The author of this book feels that the most convincing hypothesis has been advanced by A. P. Vinogradov; Vinogradov supports the primary nature of chondrules and believes that in comparison with other meteorites the matter in chondrites has been least modified by various kinds of secondary processes.

A. P. Vinogradov advanced a hypothesis concerning the general development of matter in meteorites. He compared the atomic distribution of elements in stony meteorites with their distribution on the sun, established spectroscopically, and demonstrated the close similarity between the curves of atomic distribution in both cases (Vinogradov, 1962a). Exceptions are H, He, Li, Be, B, whose distribution on the sun was modified by reactions in thermonuclear processes. This similarity in distribution curves indicated a genetic relationship between solar and meteorite matter. Therefore, A. P. Vinogradov (1959c, 1961, 1962d, 1963) postulated that the chondrules in stony meteorites and in iron meteorites were formed as the result of ejection of matter of solar composition during some metastable process and the subsequent separate condensations of silicates and iron. "Chondrules thus constitute liquid-drops, spheroical-state primary matter of meteorites, one of the basic forms of state of matter in cosmic transformations" (Vinogradov, 1959b, p.8). Their matter was subjected to self-purification as a result of multiple recondensation.

The chondrules forming in this way, subsequently, brought in a "cold" state (Tarasov, 1963), merged into larger bodies in which processes of differentiation of meteorite matter into different types of stony meteorites could transpire; this was responsible for the character of distribution of rare and other elements. "However, this process did not attain a great depth, that is, the degree of separation of the individual components was insignificant" (Vinogradov, 1961, p. 11).

Iron meteorites were formed from droplets of iron merging into large masses during a process simultaneous with the formation of chondrules. Siderolites were formed from droplets of silicates in iron by means of agglomeration. Achondrites represent the products of a process of a different kind: the recrystallization of matter in small bodies under conditions of an impoverishment with easily volatile compounds. The recrystallization of matter was also caused by heating and by pressures caused by collisions. This character of recrystallization has also been observed under experimental conditions.

The poorly expressed character of modification of meteorite matter is /201
evidenced by the isotopic composition of volatile elements (carbon, sulphur, oxygen), which A. P. Vinogradov, et al, determined for different types of meteorites. It was found that with respect to the isotopic composition of these elements, different meteorites are rather similar to one another with the exception of carbonaceous chondrites and ureilites. In turn, they were similar to rocks; dunites. A. P. Vinogradov feels that these rocks, similar to the silicate phase of chondrites,constitute the matter making up the earth's mantle and that the earth's crust was formed as a result of the phase melting of the mantle.

Thus, chondrites do not represent the result of an intense differentiation of the matter constituting the parent bodies from which they originated, but are the result of agglomeration of cosmic bodies whose matter was modified by secondary processes only to a certain extent.

A. P. Vinogradov demonstrated that some carbonaceous chondrites and the Novyy Urey ureilite with respect to the isotopic composition of carbon and oxygen, as well as respect to the content and isotopic composition of inert gases, differ from other meteorites and that the matter in these carbonaceous meteorites was formed under different conditions (Vinogradov, 1965). However, he does not discuss in detail the possible conditions of their formation and does not consider the behavior of the volatile components containing C, H, O and other elements during the process of meteorite formation.

The author of this book feels that the results of study of the organic compounds and other forms of carbon, as well as other peculiarities in meteorites containing carbon, confirm the hypothesis advanced by A. P. Vinogradov that the chondrules have a primary character in relation to chondrites and his hypothesis of the origin of chondrites, and also make it possible for the author to apply this hypothesis to the origin of those meteorites, carbonaceous chondrites and ureilites, whose origin is most difficult to explain.

Origin of Carbonaceous Chondrites and Their Organic Compounds

Interrelationship Between Content of Organic Compound and Volatile Elements C, H, S, N, O in Carbonaceous Chondrites

The occurrence of organic compounds in different carbonaceous chondrites has a definite pattern. With transition from type-III carbonaceous chondrites (Groznaya meteorite) to type-I (Orgueil meteorite), the content of bituminous components increased, as does the high-molecular organic matter content, whereas the degree of carbonization decreases.

In this same direction there is a clearly expressed regular change in other peculiarities in the structure of carbonaceous chondrites; this has been noted by many researchers (Wiik, 1956; Anders, 1963c; Mason, 1963; Vdovykin, 1965a, 1966a[1]). Some of these patterns have recently been described by Mueller (1966) in his comparison of the change in composition and structure of carbonaceous chondrites with a change in the total content of volatiles, which he determined or computed for many carbonaceous chondrites.

The regular changes in composition and structure of carbonaceous chondrites, correlating with the change in content of organic components, are evidence of processes of transformation which the matter in these meteorites underwent. These peculiarities must also be taken into account as initial data /202 in the study of the origin of carbonaceous chondrites and the organic compounds which they contain. Accordingly, before discussing the conditions of formation of carbonaceous chondrites we must examine the changes and content and the forms of occurrence of volatile elements (carbon, hydrogen, sulphur, nitrogen and oxygen) in greater detail than was done in the second chapter of

[1] See also the news items concerning the Twelfth Meteorite Conference (Vdovykin, Zotkin, Florenskiy, 1966; Florenskiy, Vdovykin, 1966).

this book and we must also do the same for the isotopic composition of these elements (with the exception of nitrogen) in all known carbonaceous chondrites. These volatile elements form part of the carbonaceous matter and the other components in carbonaceous chondrites. Table 48 gives their content in three types of carbonaceous chondrites.

Table 48

Content of C, H, S. N* and O** in Carbonaceous and Some Other Chondrites Containing Carbon
(% by weight)

Type of Carbonaceous Chondrites	Meteorite	C	H	S	N	O
I	Ivuna	4,83	2,09	6,07	—	42,53
	Alais	3,19	2,18	6,46	0,29	46,47
	Orgueil	3,52	1,50	5,55	0,24	46,78
	Tonk	2,70	2,42	5,87	—	46,82
	Essebi	3,71	—	—	—	—
II	Haripura	4,00	1,53	5,44	—	39,38
	Nawapali	2,50	1,84	2,80	—	43,42
	Nogoya	1,62	1,60	3,27	—	43,10
	Migei	2,76	1,30	3,91	—	41,81
	Staroye Boriskino	2,06	1,37	3,18	0,07	41,93
	Cold Bokkeveld	1,30	1,70	2,98	—	44,46
	Erakot	2,14	1,29	3,34	0,26	41,51
	Santa Cruz	2,54	1,16	3,30	—	36,76
	Murray	2,78	1,16	2,82	0,08***	41,86
	Al Rais	2,49	0,95	2,66	0,19	37,99
	Renazzo	1,44	0,63	1,31	0,06	36,27
III	Groznaya	0,56	0,45	2,10	0,22	38,23
	Lance	0,57	0,19	2,40	—	35,49
	Mokoia	0,98	0,25	2,73	—	37,12
	Kaba	1,99	0,07	2,27	—	38,56
	Vigarano	1,12	0,32	2,32	0,01	35,81
	Felix	0,45	0,02	2,00	—	35,74
	Ornans	0,35	0,15	2,23	—	35,56
	Warrenton	0,19	0,01	1,87	—	—
Chondrites Containing Carbon	Karoonda	0,10	0,09	1,58	0,01	36,22
	St. Caprais	0,64	0,12	2,20	—	—
	Chainpur	0,36	0,12	2,35	—	38,15
	Kainsaz	0,66	0,03	2,34	—	—
	Yefremovka****	0,80	—	—	—	—

* The content of C, H, S, N is based on data from Wiik (Mason, 1963d) and in part from data published by Mueller (1966).
** The oxygen content was determined by Wiik (Wiik, 1956; Mason, 1963d) by computations based on chemical analysis data.
*** The nitrogen content in the Murray meteorite is based on data from Calvin (Calvin, Vaughn, 1960).
**** See L. G. Kvasha, V. Ya. Kharitonova (1966).

Carbon. The carbon content increases regularly with transmission from type-III carbonaceous chondrites to type-I (Table 48). In type-III its content is 0.19-1.99%, in type-II it is 1.30-2.78 (and in the Haripura meteorite it is 4.00%), and in type-I it is 2.70-4.83%. According to data published by Mueller (1966), there is an increase in the content of "volatile" forms of carbon in this same direction (Table 49); this agrees with the increase in the content of organic compounds. /203

In type-III carbonaceous chondrites as well as in some type-II meteorites, the carbon is also represented by aromatic carbon with a definite order of atoms in the structure. In type-I meteorites it is absent. However, in type-II carbonaceous chondrites, and particularly in type-I, carbon is also present in the form of carbonates, which are not known in type-III meteorites.

Table 49

Content of "Volatile" Forms of Carbon, Hydrogen and Sulfur in Some Carbonaceous Meteorites and Other Meteorites Containing Carbon (Kainsaz and Goalpara) (Mueller, 1966; in %)

Type of Meteorite	Meteorite	"Volatile" C	"Bound" C	"Volatile" C in total C	"Volatile" H in total H	"Volatile" S in total S
Carbonaceous Chondrites I	Orgueil	3,34	0,18	94,7	97,6	63,2
II	Migei	2,47	0,29	89,6	98,5	36,3
	Staroye Boriskino	1,90	0,16	92,2	98,8	38,0
	Cold Bokkeveld	1,20	0,10	92,5	97,1	45,7
	Murray	2,40	0,17	93,4	100	55,2
III	Groznaya	0,33	0,23	58,9	100	57,6
Chondrite containing carbon	Kainsaz	0,30	0,36	45,6	100	44,8
Ureilite	Goalpara	0,41	0,31	57,0	100	0,0

The carbonates in type-II carbonaceous chondrites, the Staroye Boriskino meteorite, were detected by L. G. Kvasha (1948); in this meteorite the carbonates were represented by calcite. She reported the presence of carbonates in the Cold Bokkeveld and Murray meteorites as well (Kvasha, 1963). Carbonates are probably present in the Migei meteorite as well. I. V. Grinberg and A. A. Salamin (1963) determined 2.58% CO_2 in the Migei meteorite using a titrometric semimcromethod.

The content of carbonates in the type-I Ivuna and Orgueil meteorites is approximately 0.3% (Clayton, 1963). They are found in the form of grains up to 2-3 mm in diameter (Nagy, 1966b). Pisani (1864) identified the carbonates in the Orgueil chondrite as breunnerite. This was recently confirmed by Mason (1963d), who demonstrated that an isomorphic mixture $MgCO_3$-$FeCO_3$ contains about 20 mol % $FeCO_3$. DuFresne and Anders (1962b, 1963) determined the carbonates in this meteorite as dolomite and breunnerite. Nagy and Andersen (1964), by means of an X-ray spectral microanalysis confirmed the conclusion that the Orgueil meteorite contains a ferromagnesian mineral (breunnerite), whose composition is described by the formula $Mg_{0.8}Fe_{0.2})CO_3$. They noted a nonuniform distribution of iron in this mineral. In the Ivuna meteorite DuFresne and Anders (1962b) identified dolomite.

The regular change in the carbon content and the change in the forms of its occurrence in different carbonaceous chondrites indicate an interrelationship among all three types of meteorites. The isotopic composition of the carbon indicates the same. /204

A. P. Vinogradov (1958, 1959b) notes that the isotopic composition of the carbon in meteorites is approximately identical; this indicates the absence of a significant isotopic fractionation during the formation of the meteorites, but in carbonaceous chondrites the carbon is heavier.

For a number of carbonaceous chondrites the isotopic composition of total carbon has been determined by Boato (1954a, b). He demonstrated that carbonaceous chondrites are clearly separable into two groups on the basis of the isotopic composition of carbon. In the first group, Boato included meteorites with an isotopic composition δC^{13} from -3.7 to -11.4 $^0/_{00}$ (Table 50); these are carbonaceous chondrites, which, as later demonstrated by Wiik (1956), are represented by meteorites of types-I and II. In the second group Boato included meteorites with an isotopic composition δC^{13} from -15.7 to -18.8$^0/_{00}$[1]. These are type-III carbonaceous chondrites as defined by Wiik[2]. According to Boato's data, the ordinary Richardton and Forest City chondrites contain more lighter carbon (δC^{13} = -24.6 and 24.3$^0/_{00}$ respectively).

Thus, on the basis of the isotopic composition of carbon carbonaceous chondrites differ from ordinary chondrites and within the group of carbonaceous chondrites there is a general increase in the weight of carbon with transition from type-III to type-I (Table 50). Boato also noted some nonuniformity in carbon distribution with respect to isotopic composition within the same meteorite.

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[1] These results are close to the data obtained by Briggs and Kitto (1962) on the isotopic composition of total carbon in the Mokoia carbonaceous chondrite (-19.3$^0/_{00}$ δC^{13} at t = 1100°).

[2] Boata also includes the Indarkh meteorite among the carbonaceous chondrites; it is now customary to consider it an enstatic chondrite.

Boato's data, as well as the earlier results obtained by A. V. Trofimov (1950) related to the total carbon in carbonaceous chondrites, that is, to the total of carbonaceous matter and carbonates; however, since carbonates are rarely found, to all intensive purposes the results apply only to carbonaceous matter.

Clayton (1963) determined the isotopic composition of carbon in the carbonates extracted from the Ivuna and Orgueil meteorites. He publishes the results of five determinations of the isotopic composition of carbon in carbonates for the Orgueil meteorite, which in general are similar (δC^{13} from +58.6 to +64.4$^0/_{00}$), that is, according to Clayton's data, the carbon in the carbonates of the Orgueil carbonaceous chondrite and the Ivuna meteorite is 6% heavier than the carbon in terrestrial carbonates.

Briggs (1963b) determined the isotopic composition of carbon for bituminous matter extracted using a mixture of benzene and methyl alcohol (9:1) in a Soxhlet apparatus for four carbonaceous chondrites (Orgueil, Haripura, Murray and Mokoia). Briggs' data show (See Table 50 and Figure 127) that the carbon in this fraction of organic matter in meteorites in its isotopic composition virtually does not differ from the carbon in carbonaceous matter as a whole (Boata, 1954a).

According to data obtained by Nagy (1966), the bituminous matter in the Orgueil meteorite has $\delta C^{13} = -23.8^0/_{00}$.

A. P. Vinogradov, O. I. Kropotova, G. P. Vdovykin and V. A. Grinenko (1967) recently studied the isotopic composition of carbon in individual forms of hydrocarbon matter in four carbonaceous chondrites and other meteorites containing carbon (Table 51). It was found that the bituminous matter extracted from meteorites by means of organic solvents without the latter being heated is somewhat enriched with the light carbon isotopic C^{12} in comparison with the total carbon of these same meteorites. The graphite and diamond in /205
the meteorite in their isotopic composition were "heavier". No significant differences between graphite and the diamond in meteorites was established with respect to the isotopic composition of carbon.

Thus, with a transition from type-III carbonaceous chondrites to type-I the increase in the content of organic components and the decrease in the degree of their carbonization is accompanied by an increase in the content of total carbon in these meteorites; with respect to isotopic composition, the carbon in general becomes heavier in this direction. The isotopic composition of carbon in carbonaceous chondrites is evidence of an isotopic fractionation which occurred. Apparently, some isotopic fractionation of carbon in different forms of carbonaceous matter could occur also at the time of development of the matter in other types of meteorites. In particular, the data published by Craig (1953) on the different isotopic composition of carbon in graphite and cohenite in the Canyon Diablo iron meteorite can serve as evidence of this process (see Figure 127).

Table 50

Isotopic Composition of Carbon in Carbonaceous and Other Chondrites (δC^{13}, $^0/_{00}$).

Type of Meteorite	Meteorite	Total Carbon (Carbonaceous Matter and Carbonates) (Boata, 1954a)	Carbon in Bituminous Matter (Briggs, 1963b)	Carbon in Carbonates (Clayton, 1963)
Carbonaceous Chondrites Type-I	Ivuna	— 6,6	—	—
	Orgueil	—11,4	—10,1	58,6 59,1 59,8 64,3 64,4
Type-II	Haripura	— 3,7	— 3,1	—
	Nawapali	—10,0	—	—
	Migei	— 9,9	—	—
	Cold Bokkeveld (British Museum)	— 9,4	—	—
	Cold Bokkeveld (Paris Museum)	— 5,2	—	—
	Santa Cruz	— 4,3	—	—
	Murray	— 3,9	— 4,0	—
Type-III	Lancé	—15,7	—	—
	Mokoia (Naynidzher Collection)	—17,4	—	—
	Mokoia (New Zealand Museum)	—18,8	—17,9	—
	Felix	—16,4	—	—
Enstatic Chondrite	Indarkh.	—12,9	—	—
Ordinary Chondrites	Richardton	—24,6	--	—
	Forest City	—24,3	—	—

PDB Standard (Craig, 1953)

During recent years the content of radioactive carbon C^{14} has been determined for a number of meteorites; on the basis of this content one can draw /207 conclusions concerning the relative intensity of cosmic radiation and also concerning the time of falling of meteorites. The contents of C^{14} has been measured in a number of meteorites by Suess and Wänke (1962) and by Goel and Kohman (1962a, b). For the meteorite which they studied they obtained terrestrial ages up to 20,000 years or more. The activity level for the C^{14} attained 78 decays min/kg. In the Orgueil carbonaceous chondrite the specific activity was 82 decays min/kg (Tamers, 1962), whereas in the Cold Bokkeveld meteorite it was 47 decays min/kg (Fireman, et al, 1963).

Table 51

Isotopic Composition of Carbon in Meteorites Containing Carbon
(Vinogradov, Kropotova, Vdovykin, Grinenko, 1967)

Types of Meteorites	Meteorite	Total Carbon		Bituminous Matter		Unextractable Carbonaceous Matter		Graphite		Graphite and Diamond (Aggregates)		Diamond (Pure Fraction)	
Carbonaceous Chondrites		δC^{13}, %	C^{12}/C^{13}	δC^{13}, %	C^{12}/C^{13}	δC^{13}, %	C^{12}/C^{13}	δC^{13}, %	C^{12}/C^{13}	δC^{13}, %	C^{12}/C^{13}	δC^{13}, %	C^{12}/C^{13}
Type-I	Orgueil (Nagy, 1966)	—	—	—2,38	91,15	—	—	—	—	—	—	—	—
Type-II	Migei	—1,11 (247) *	89,99	—2,20 (13,3)	90,99	—	—	—	—	—	—	—	—
	Staroye Boriskino	—0,85 (508)	89,75	—	—	—	—	—	—	—	—	—	—
Type-III	Cold Bokkeveld	—1,46 (155)	90,31	—	—	—	—	—	—	—	—	—	—
	Groznaya	—2,05 (237)	90,85	—2,87 (7,4)	91,61	—	—	—	—	—	—	—	—
Chondrite containing Carbon	Kainsaz	—1,53 (1250)	90,37	—	—	—1,88 (8,5)	90,69	—	—	—	—	—	—
Ureilites	Novyy Urey	—0,71 (500)	89,62	—2,77 (9,9)	91,52	—	—	—	—	—0,63 (13,5)	89,55	—0,57 (5,6)	89,50
	Goalpara	—0,84 (300)	89,74	—	—	—	—	—	—	—0,15 (7,4)	89,12	—	—
Iron Meteorites (Course structured octahedrites)	Yardymlinskiy	—	—	—	—	—	—	—0,50 (170)	89,44	—	—	—	—
	Canyon Diablo	—	—	—	—	—	—	—0,63	89,55	—	—	—	—

The data have been reduced to the PDB standard (Craig, 1953) for which the ratio $C^{12}/C^{13} = 88.99$.
* Initial weighed sample of matter (in mg).

Hydrogen. The hydrogen content also increases simultaneously with an increase in the quantitative content of organic compound with a transition from type-III carbonaceous chondrites to type-I (See Table 48). In type-III meteorites the hydrogen content is 0.01-0.45%, in type-II it is 0.63-1.84% and in type-I it is 1.50-2.42%. An insignificant part of this hydrogen is found in hydroscopic water, as is indicated by data from a thermal analysis (See Figure 86) and the results of study made by many other authors. In addition, in most cases hydrogen is present in an insignificant quantity in water of crystallization in hydrous sulfates observed in some meteorites.

The regular increase in hydrogen content with transition from type-III carbonaceous chondrites to type-I meteorites is not caused exclusively by an increase in the content of organic components (See Table 49). The hydrogen in carbonaceous chondrites enters into a hydroxyl group in chlorite-serpentine minerals; the quantitative content of these minerals increases with transition from type-III meteorites to type-I. Thus, among many carbonaceous chondrites investigated by the roentgenometric method, DuFresne and Anders (1962b) noted only traces of these minerals in the Felix, Mokoia and Ornans type-III carbonaceous chondrites. Kerridge (1964) made an electron-diffraction study of pulverized matter from carbonaceous chondrites and demonstrated that in the Lancé, Mokoia and Ornans carbonaceous chondrites the hydrous minerals have a fibrous texture and are similar to clinochrysolite. In addition, in the Kaba meteorite he noted platelets of these minerals resembling sebiolite.

In type-II carbonaceous chondrites the chlorite-serpentine minerals already make up almost half of all the silicates (See Figure 12). Hydrous silicates in meteorites were found for the first time by L. G. Kvasha (1948) in type-II carbonaceous chondrites, in the Staroye Boriskino meteorite, and then in the Migei meteorite. According to A. N. Zavaritskiy and L. G. Kvasha (1952) these minerals in their optical properties and also with respect to data from a thermal analysis, are more closely related to chlorite than to serpentine, but evidently represent a mixture of both varieties.

In 1954 the Zahariasen (DuFresne, Anders, 1962b) used an X-ray-diffraction analysis in determining the hydrous silicates in the Migei, Cold Bokkeveld, and Murray meteorites as chrysolite. X-ray structural studies of the Migei meteorite made by V. I. Mikheyev and A. I. Kalinin (1958) and similar studies of the Staroye Boriskino and Cold Bokkeveld meteorites (Stulov, 1960), also revealed that these carbonaceous chondrites contain minerals of the chlorite-serpentine series close to both the serpentine of the antigorite series and to chlorite -- pennine.

Chlorite, or a mineral close in structure to serpentine, was determined by the roentgenometric method by Mason (1962b) in a number of type-II carbonaceous chondrites and by DuFresne and Anders (1962b) in the Haripura, Migei, Cold Bokkeveld and Murray meteorites. DuFresne and Anders note that the lattices of these minerals have the following parameters: a = 7.0 - 7.1 Å, b = = 8.0 - 9.6 A. In the Pollen carbonaceous chondrite Wolff (1963) determined the presence of hydrous silicates of the antigorite series. Kerridge (1964), on the basis of an electron-diffraction study, believes that the Staroye Boriskino meteorite contains minerals similar to sepiolite (platelike aggregates

of grains in the meteorite), and also clinochrysolite (fibrous grains). Kerridge noted the latter mineral in the Cold Bokkeveld and Murray meteorites /208 as well. In the different meteorites which he studied the b value for a unit cell varied from 8.7 to 9.3 Å; the most common values were b = 8.9 - 9.0 Å. Kerridge, like other researchers, feels that even within the same meteorite there are different kinds of hydrous silicates which are in mixture with one another. In these minerals there is a predominance of cations of magnesium and iron, with aluminum being less common. In a study of the hydrous silicates in the Migei meteorite by the spectral analysis method, Kerridge determined (in %): 17 Fe, 15 Mg, 14 Si and 1.1 Al. Nagy (1966b) assumes that in the fraction of the meteorite studied by Kerridge there are very tiny magnetite grains.

The author (Vdovykin, 1964c), on the basis of a roentgenometric study, also detected chlorite-serpentine minerals in the type-II Migei and Murray carbonaceous chondrites. It was difficult to identify these minerals in greater detail because the hydrous minerals undoubtedly actually constitute a mixture. In an electron microscope study of pulverized matter from the Migei, Staroye Boriskino and Cold Bokkeveld meteorites the author (Vdovykin, 1964e, 1965a) noted that the hydrous minerals, present in the matrix of meteorites in a mixture with carbonaceous matter, are found in the form of platelike or tabular grains of irregular configuration or aggregates of grains which are translucent under an electron beam. In addition, the Migei meteorite contains fibrous forms of these minerals, similar to grains whose photograph for the Mokoia meteorite are given by Kerridge and for the Erakot meteorite by Mason (1963a).

Thus, in type-II carbonaceous chondrites the chlorite-serpentine minerals, containing a hydroxyl group, represent a mixture even within the same meteorite. If grains of only one type are present (such as platelike chlorite-serpentine minerals), their chemical composition, in all probability is not in equilibrium, and therefore it is difficult to identify them. Hydrous minerals are found in the interchondrule space of carbonaceous chondrites in a mixture with carbonaceous matter, and also secondarily replace the chondrules. Substitution with chlorite-serpentine occurred around the periphery of the chondrules, as can be judged from the relicts of olivine grains in the middle part of chlorite-serpentine chondrules. In many cases entire sectors of meteorites have been replaced by chlorite-serpentine in these meteorites. The character of this substitution has been described in detail by L. G. Kvasha (1948, 1950; Kvasha, Wiik, 1964) for the Staroye Boriskino and Migei meteorite.

In type-I carbonaceous chondrites (of the Orgueil type) silicates have been completely replaced by chlorite-serpentine. Hydrous minerals of this type were observed by N. N. Stulov (1960) in the Orgueil meteorite, by Mason (1962b) in the Aláis, by DuFresne and Anders (1962b) in the Ivuna and Orgueil meteorites and by Nagy, et al (1963) and Kerridge (1964) in the Orgueil meteorite. Nagy feels that in the Orgueil meteorite the aggregates of this mineral are represented by chlorite or possibly by montmorillonite with parameters of a unit cell a = 7.6 and b = 9.26 Å. These parameters are close to the parameters of a unit cell which Kerridge determined for the hydrous silicates in this meteorite (a = 7.4 and b = 9.2 Å). However, Kerridge, who also observed platelike

grains, and in a few cases, fibrous grains, in the Orgueil meteorite under an electron microscope, feels that these minerals are represented by sepolite and clinochrysolite respectively. Such tabular forms of hydrous silicates were noted in the Orgueil meteorite by G. P. Vdovykin (1965a) in an electron microscope study of pulverized matter from this meteorite. The Orgueil and Cold Bokkeveld meteorite were recently investigated using Mössbauer spectralscopy by Gérard and Delmelle (1964). They feel that the hydrous silicates in /209 the Orgueil meteorite are similar to amphibole.

The chlorite-serpentine in type-I carbonaceous chondrites replace olivine to such an extent that the latter usually cannot be detected at all. DuFresne and Anders, as well as Nagy, et al, failed to detect olivine in the Orgueil meteorite. Kerridge reported that it contained only traces of olivine.

Thus, there is a clearly expressed interrelationship observed in the distribution of the principal compounds containing hydrogen (organic components and secondarily substituted silicates of chlorite-serpentine minerals); the content in both increases with transition from type-III carbonaceous chondrites to type-I.

The isotopic composition of hydrogen in a number of carbonaceous chondrites was analyzed by Boato (1954a, b, 1956). He found that in the H_2O fraction extracted from these meteorites when they were heated to 180° the hydrogen does not differ in its isotopic composition from terrestrial hydrogen; this can be attributed to the presence of hygroscopic water and water of crystallization of terrestrial origin. However, the isotopic composition of the hydrogen in H_2O extracted from carbonaceous chondrites during their heating from 180 to 800° is considerably different. The hydrogen in H_2O^+ is enriched with deuterium. In the case of the Ivuna, Orgueil, Murray and Mokoia carbonaceous chondrites this enrichment attains 9-35% of the terrestrial standard (Table 52).

This enrichment with deuterium is characteristic only for cosmic objects. The hydrogen in this fraction is total; it enters into both organic compounds and chlorite-serpentine minerals.

Briggs (1963b) determined the isotopic composition of the hydrogen in bituminous matter extracted from the Orgueil, Haripura, Murray and Mokoia meteorites. The bituminous matter in these meteorites was also enriched by deuterium (Table 52); no significant differences were found in the isotopic composition of the hydrogen in the H_2O^+ fraction, as determined by Boato, and the hydrogen in the bituminous matter. This can also indicate a simultaneity of the process of formation of organic compounds and chlorite-serpentine minerals in these meteorites.

There is little data on the isotopic composition of hydrogen for meteorites of other types. Friedman (1953) analyzed the isotopic composition of the hydrogen in the Canyon Diablo iron meteorite. According to his data, the

hydrogen extracted from the meteorite at high temperatures was characterized by an increased deuterium content. Later the isotopic composition of hydrogen in a number of iron meteorites, for the most part octahedrites, including the Canyon Diablo meteorite, was analyzed by Edwards (1955a). He found that iron meteorites, which contain carbonaceous nodules, have hydrogen in the following quantities: ($n \cdot 10^{-4}$%): Canyon Diablo 4.9, Cranbourne 7.0, Cosby's Creek 33.4. The value δD was respectively (in %): -8.1, -4.4, -9.7. This hydrogen did not differ in its isotopic composition from the hydrogen in other meteorites which were analyzed by Edwards (on the average δD was equal to -8.6%) and approached the values characteristic for the hydrogen in igneous rocks.

In order to determine the intensity of cosmic radiation and establish the terrestrial age of meteorites it is of interest to determine their tritium content. However, for the time being H^3 has not been measured in many meteorites (Bainbridge, et al, 1962, Begemann, 1966). It can be noted that in the Cold Bokkeveld the activity level is H^3 = 550 decays min/kg (Fireman, et al, 1963).

Sulfur. The change in total sulfur content in carbonaceous chondrites also agrees with the quantitative increase in organic compounds with a transition from type-III meteorites to type-I. In type-III carbonaceous chondrites /210
the sulfur content is 1.87 - 2.73%, in type-II it is 2.66 - 3.98% (the Renazzo meteorite and the Haripura meteorite are exceptions, having content of 1.31% and 5.44% respectively), whereas in type-I the content is 5.55 - 6.46% (See Table 48).

The forms of manifestation of sulfur in carbonaceous chondrites vary greatly. Approximately 0.03% of the sulfur is found in the bituminous matter. Some part of the sulfur is associated with high-molecular organic matter.

Sulfur in carbonaceous chondrites appears the same as free sulfur, whose quantitative content in the Migei meteorite is 1.58% (Kaplan, et al, 1963), whereas in the Orgueil meteorite it is 1.8% (Monster, et al, 1965). It should be noted that the content of free sulfur in different carbonaceous chondrites is difficult to reckon because it is determined only by extraction by means of organic solvents and is extracted together with the bituminous matter. In the extract sulfur forms independent droplike aggregates of tiny crystals among tiny crystals of bituminous matter (See Figure 32). In addition, sulfur is determined in a study of pulverized matter from carbonaceous chondrites under an electron microscope. In the latter case it melts under the electron beam and forms drops (See Figure 14b).

The presence of elementary sulfur has been observed in the type-III Lancé and Mokoia carbonaceous chondrites (DuFresne, Anders, 1962b) and in the Groznaya meteorite (Vdovykin, 1962b). It was found in a number of type-II meteorites: Haripura, Migei, Cold Bokkeveld, Murray (Mueller, 1953; DuFresne, Anders, 1962b; Kaplan, et al, 1963). Sulfur was noted early in type-I carbonaceous chondrites by Wöhler (1859) in the Orgueil meteorite. Recently many researchers have determined sulfur in this meteorite (DuFresne, Anders, 1962b, 1963; Kaplan, et al, 1963, and others; see also Figure 14b). DuFresne and Anders established the presence of sulfur in the Ivuna meteorite. In /211

Table 52

Isotopic Composition of Hydrogen in Carbonaceous Chondrites

Type of Carbonaceous Chondrite	Meteorite	Total H (Carbonaceous Matter and Water) (Boato, 1954a)				H in Bituminous Matter δD, % (Briggs, 1963b)
		H_2O^- ($t<180°$)		H_2O^+ ($t=180-800°$)		
		%	δD, %	%	δD, %	
Type-I	Ivuna	4,7	−1,0	7,0	+35,8	—
	Orgueil	4,0	+1,5	7,3	+29,0	+27,5
Type-II	Haripura	2,4	−1,1	7,3	− 3,3	− 1,5
	Nawapall	3,0	−4,2	9,0	− 7,2	—
	Migei	1,7	−4,2	8,6	− 6,4	—
	Cold Bokkeveld (British Museum)	2,1	−7,6	7,8	−13,0	—
	Cold Bokkeveld (Paris Museum)	2,6	−3,7	8,0	− 5,8	—
	Santa Cruz	1,8	−8,0	8,4	− 0,7	—
	Murray	2,3	−8,9	6,8	+ 9,6	+ 8,4
Type-III	Lancé	0,6	−6,3	0,9	− 7,7	—
	Mokoia (Naynidzher collection)	0,3	−6,8	0,8	+25,9	—
	Mokoia (New Zealand Museum)	0,25	−7,2	0,75	+23,5	+21,2

The standard used was water from Lake Michigan (Friedman, 1953).

carbonaceous chondrites free sulfur is found in the matrix (interchondrule space) in a fine mixture with carbonaceous matter, carbonates, chlorite-serpentine and other mineral components; this is evidence of a complex composition of the early volatile compounds, leading to the formation of these "low-temperature" minerals.

Sulfates are one of these "low-temperature" minerals which contain sulfur These water-soluble salts in type-III meteorites are present only in traces (Ornans meteorite) (Anders, 1964). Sulfates have been observed in type-II meteorites: Migei, Cold Bokkeveld (DuFresne, Anders, 1963) and Murray (Calvin, 1961; DuFresne, Anders, 1963; Kerridge, 1964). In these meteorites there were infrequent tiny crystals of calcium sulfate. Type-I carbonaceous chondrites are particularly enriched with them, up to 16% (Monster, et al, 1965). Sulfates have been detected in the Ivuna meteorite, where they are represented primarily by epsomite with admixtures of hexahydrite and apparently blödite (DuFresne, Anders, 1962b). In the Aláis meteorite magnesium sulfates were observed early by Berzelius (1834). Recently sulfates have been investigated in greater detail in the Orgueil meteorite, where they are represented

primarily by epsomite (Calvin, 1961; Kerridge, 1964, and others). Kerridge (1964) and Nagy and Andersen (1964) recently demonstrated that the Orgueil meteorite also contained calcium sulfates (Ca: S = 1.23). In type-I carbonaceous chondrites the sulfates occur in the form of light encrustations and veinlets. Data on the enrichment of type-I carbonaceous chondrites (in comparison with other meteorites) by all "volatile" forms of sulfur are given in Table 49.

In contrast to the mentioned general tendency, the distribution of "high-temperature" components containing sulfur in general decreases with transition from type-III meteorites to type-I. These minerals are represented by troilite FeS, pentlandite $(Fe, Ni)_9S_8$ and other sulfides[1]. The troilite content in type-III carbonaceous chondrites, according to Mason (1963d), attains 5%. We recall that its average content in ordinary chondrites is 5.73%. In type-II troilite has been observed in an insignificant quantity in the Migei and Staroye Boriskino meteorites (Yudin, Obotnin 1961); it is rarely encountered in type-I meteorites such as Ivuna and Orgueil (DuFresne, Anders, 1962b). Pentlandite has been observed in the Kaba carbonaceous chondrite (Sztrókay, 1960) and the Migei carbonaceous chondrite (DuFresne, Anders, 1962b). Recently Wood (1967) used the X-ray spectral microanalysis method for a study of the sulfides in a number of carbonaceous chondrites and noted that pentlandite was more characteristic for carbonaceous chondrites than was troilite.

With respect to isotopic composition, the total sulfur of carbonaceous chondrites differs virtually in no way in the sulfur of other types of meteorites, which, as is well known, has an identical isotopic composition in different meteorites (Vinogradov, Chupakhin, Grinenko, 1957; Olt, 1961; Kaplan, Hulston, 1964, and others). However, in carbonaceous chondrites, as demonstrated by recent investigations, the different forms of sulfur also had a different isotopic composition. The bituminous matter in four carbonaceous chondrites, according to data published by Briggs (1963b), contains sulfur which is heavier than the standard (Table 53). Monster, Anders and Thode (1965) investigated the isotopic composition of the total and free sulfur and the sulfur in sulfates and sulfides in the Orgueil meteorite (Table 53). According to their data, the free sulfur and the sulfur in sulfides are heavier, as is the sulfur in the bituminous matter in this meteorite. This, in particular, can indicate a synchronous process of formation of the sulfur /213
in organic compounds and the free sulfur. Hulston and Thode (1965a) recently made a detailed study of the isotopes of sulfur S^{33}, S^{34} and S^{36} in the different forms of its occurrence in many meteorites, including a number of carbonaceous chondrites. Table 53, based on their data, is the δS^{34} variations for carbonaceous and some other meteorites. Hulston and Thode concluded that the variations in the content of S^{33}, S^{34} and S^{36} which they found are evidence of an isotopic fractionation during the formation of the matter in meteorite, caused by chemical and physical processes. On the basis of isotopic analysis of the sulfur in carbonaceous chondrites, Kaplan (Kaplan, 1965; Kaplan, Hulston,

[1] The Ornans carbonaceous chondrite contains tyrrhotite, whereas the Karoonda meteorite contains pyrite and chalcopyrite, present in an aggregate with pentlandite (Mason, Wiik, 1962b).

Table 53

Isotopic Composition of Sulfur in Carbonaceous and Other Meteorites

Types of Meteorites	Meteorite	Total S	S in Bituminous Matter	Free S	S in SO_4^{2-}	S in FeS	S in MnS	S in CaS	Remaining S
Carbonaceous Chondrite Type-I	Orgueil	+0,4 (2)	+3,14	+3,04 (2)	−1,30 (2)	—	—	—	—
		—	—	+1,5 (2)	−1,5 (2)	+2,6 (2)	—	—	—
		—	—	+1,4	−0,9	—	—	—	—
		0,0	—	+1,0	−1,6	+3	—	—	—
		+0,35	—	+1,8; +3,3	−1,7	—	—	—	—
		—	—	—	0,0	—	—	—	—
Type-II	Haripura	—	+2,51	—	—	—	—	—	—
	Migei	+0,3	—	+0,4	0,0	+1,7	—	—	−1,5
	Cold Bokkeveld	+0,52	—	−0,6; +0,2	+0,7	+2,2	—	—	—
	Murray	−0,01	+2,86	—	−0,2	+0,8;	—	—	+0,3
		—	—	—	−1,7	+2,5	—	—	—
Type-III	Lancé	—	—	—	—	+0,5	—	—	—
	Mokoia	+0,3	+1,09	—	+1,3	−1,4	—	—	+0,1; +0,4
	Felix	—	—	—	—	+0,3	—	—	—
	Warrenton	—	—	—	—	+0,4	—	—	—
Chondrite containing carbon	Karoonda	+0,2; −0,2	—	−0,2	+0,4	—	—	—	−0,8
Enstatic chondrites	Abee	−0,06	—	−1,7	—	—	−0,2; +0,4	+1,6; −1,7	—
	Hvittis	−0,14	—	—	—	+0,1	—	−0,8; +2,5	+0,2
Ordinary chondrite	Richardton	+0,5	—	+0,3	—	+0,2; +0,5	—	—	—
	Bjurböle	—	—	—	—	+0,3	—	—	−1,7

* The sources in the literature are given in parenthesis: 1 - Briggs, 1963b; 2 - Monster, Anders, Thode, 1965; all the other data are from Hulston and Thode (1965) and Kaplan and Hulston (1966). The standard was the sulfur in troilite from the Canyon Diablo meteorite (S^{32}/S^{34} = 22.22).

1966) noted that there was a fractionation of the isotopes of sulfur, whose mechanism excludes biogenous processes.

With respect to the isotopic composition of sulfur for carbonaceous chondrites, one can note the same regularity which we saw for the carbon and hydrogen. Table 53 shows that with transition from type-III carbonaceous chondrite to type-I meteorites the sulfur in the bituminous matter, free sulfur and the sulfur in sulfides are enriched with the heavy isotope of sulfur S^{34}.

Nitrogen. The nitrogen content and the forms of its manifestation in carbonaceous chondrites, and in general in meteorites, have been poorly studied. However, available data indicate a general tendency to an increase in the nitrogen content simultaneously with an increase in the content of organic compounds with transition from type-III carbonaceous chondrites to type-I. In type-III meteorites the nitrogen content is 0.01-0.22%, in type-II it is 0.07-0.26%, and in type-III meteorites it is 0.24-0.29% (See Table 48).

Some part of the nitrogen in carbonaceous chondrites (approximately 0.04%) is associated with organic compounds (See Table 22). It was pointed out in the second chapter that nitrogen can also be present in water-soluble ammonium compounds NH_4Cl or $(NH_4)_2SO_4$. Clöez (1864b) determined 0.1% NH_3 in an analysis of the Orgueil meteorite.

Vallentyne (1965) recently determined the total content of nitrogen in the Orgueil meteorite; this content was 0.16% in an analysis without preliminary dessication and 0.12% when the samples had been reduced to a constant weight in a vacuum at 100°. 88% of the total nitrogen was in an acid-soluble form and 0.33% was in amino acids.

A. P. Vinogradov, K. P. Florenskiy and V. F. Volynets (1963) investigated the nitrogen content in a number of meteorites, which was determined in the form NH_3^+. According to their data, the mean N-NH_3 content in ordinary chondrites is 0.00224%, that is, far less than in carbonaceous chondrites.

Oxygen. In contrast to all other meteorites, carbonaceous chondrites are considerably enriched with oxygen. The increase in its content, like the increase in other volatile elements, also agrees with the increase in content of organic compounds with a transition from type-III to type-I carbonaceous chondrites. In type-III the oxygen content is 35-38%, in type-II it is 36-44% and in type-I it is 42-46% (See Table 48). This agrees with a general increase in the degree of oxidation in matter in carbonaceous chondrites as a whole with transition in the same direction. The organic components as well as the mineral part are oxidized.

The increase in the degree of oxidation is particularly characteristic for metal. It is well known that ferronickel is a characteristic component of all meteorites; in chondrites its average content is 13.18% (Urey, Craig, 1953). However, in carbonaceous chondrites the iron is in large part, and sometimes completely oxidized. In type-III carbonaceous chondrites its content is 0-6% and in type-II there are traces; ferronickel is absent in the

first type (DuFresne, Anders, 1962b; Mason, 1963d). However, magnetite, whose greatest quantity is present in type-I, is characteristic for carbonaceous chondrites (See Figure 12).

Among the type-III carbonaceous chondrites, magnetite has been detected in the Mokoia meteorite (Staces, et al, 1961; DuFresne, Anders, 1962b), the Kaba meteorite[1] (Mason, 1962b) and the Groznaya meteorite (Yudin, Obotnin, 1961). In type-II carbonaceous chondrites magnetite has been found in the following meteorites: Haripura, Migei, Staroye Boriskino, Cold Bokkeveld, Murray (DuFresne, Anders, 1962b, 1963; Vdovykin, 1965a). In the Orgueil meteorite (type-I carbonaceous chondrite) magnetite was observed early by Cloëz (1864a). Magnetite is also present in other meteorites of this type (Mason, 1962b). Mason (1963d) sites the parameters of a cell of this spinel mineral for the following meteorites: Ivuna (8.41 Å), Aláis (8.41 Å), Orgueil (8.43 Å), Tonk (8.41 A). He feels that this mineral in its composition is close to trevorite ($NiFe_2O_4$). /214

Spinel, $MgAl_2O_4$, has been identified in an insignificant quantity in the Kaba carbonaceous chondrite (Sztrókay, 1960) and Vigarano carbonaceous chondrite (Mason, 1962b). An ordinary admixture in carbonaceous chondrites is chromite, $FeCr_2O_4$ (Kvasha, 1963).

We have already examined the distribution of mineral components in carbonaceous chondrites in which oxygen is accompanied by other volatile elements, such as carbon (carbonates), hydrogen (chlorite-serpentine minerals) and sulfur (sulfates). In contrast to these minerals, the silicate "high-temperature" components decrease quantitative with transition from type-III carbonaceous chondrites to type-I; they are completely absent in type-I. They are represented by olivine and pyroxenes, which are among the principal minerals in carbonaceous chondrites. The chemical composition of these minerals is nonequilibrium.

In type-III carbonaceous chondrites olivine totals 65-70% by weight (Mason, 1963d). It forms chondrules and is encountered in the form of individual grains or irregular configuration in the matrix. The composition of olivine in carbonaceous chondrites has been studied by Mason (1963b, d). According to his data, in the olivine of the Groznaya and Mokoia meteorite there is 0.60 mol. % Fe_2SiO_4. It is most common to observe olivine containing about 42 mol. % of this component. In the Kaba meteorite the most common form of olivine has 40 and 7 mol. % Fe_2SiO_4, whereas in the Vigarano meteorite the olivine content is 40 mol. % Fe_2SiO_4 (with variations from 0 to 60 mol. %). According to Mason's data, the olivine of many type-II carbonaceous chondrites contain about 5 mol. % Fe_2SiO_4 in solid solution, and sometimes (such as in the Haripura meteorite) up to 30 mol. % Fe_2SiO_4.

The pyroxenes in carbonaceous chondrites are represented by orthopyroxenes and, to a lesser degree, clinopyroxenes (Kvasha, 1963). In type-III meteorites

[1]Sztrókay, et al (1961) noted magnetite containing nickel in the Kaba meteorite.

in which the pyroxene content is 5-10%, Mason (1963d) determined them as pigeonite, but in type-II meteorites, as enstatite or clinoenstatite. By means of an electron diffraction analysis, Kerridge (1964) confirmed the presence of enstatite and clinoenstatite in the Murray meteorite, in which the pyroxene content, according to Mason, is up to 10-15%.

In type-III carbonaceous chondrites plagioclase (oligoclase) has been noted in a quantity up to 5-10% by weight (Mason, 1963d). Structurally, glass with a silicate composition plays some role in type-II and type-III carbonaceous chondrites. Among the other compounds containing oxygen we should also mention particles of opallike silica, which were observed by Dopre (Kvasha, 1963) in the Orgueil meteorite, and especially particles in the Migei carbonaceous chondrite, classified as glass, which were found by DuFresne and Anders (1961). These particles were not stable; they were sintered at a temperature above 300°. DuFresne and Anders classified them as mineral-thermometers and postulated that these particles were found under the influence of hydrous solutions in the parent body from which the meteorites originated.

Table 54

Isotopic Composition of Oxygen in Meteorites and Igneous Rocks (Vinogradov, Dontsova, Chupakhin, 1958)

Samples		No. of Determinations	δO^{18}, %	O^{18}/O^{16}
Meteorites				
Carbonaceous Chondrites (nonmagnetic fraction)	Migei	1	—0,41	489,0
	Staroye Boriskino. .	1	—0,40	488,9
Ureilite (nonmagnetic fraction	Croznaya . .	1	—0,39	488,9
	Novyy Urey. .	2	—0,39	488,8
Mean		5	—0,40	488,9
Ordinary chondrite.		7	—0,68	490,3
Achondrites		10	-0,64	490,1
Pallasites (silicate phase) . .		6	—0,70	490,4
Igneous Rocks				
Ultrabasic (dunite)		10	—0,61	490,0
Basic (basalt).		12	—0,46	489,2
Acidic		7	—0,16	488,0
Quartz (standard)				487±1

Thus, in type-III carbonaceous chondrite the matter in the interchondrule /215 space is oxidized and to some extent or another secondarily replaced under the influence of volatile compounds; in type-II meteorites,in addition, there is a clearly expressed substitution of the chondrules; the matter in type-I carbonaceous chondrite has been substituted to the greatest degree and is most oxidated.

With respect to isotopic composition of oxygen, carbonaceous chondrites also differ considerably from other meteorites. A. P. Vinogradov, Ye. I. Dontsova and M. S. Chupakhin (1958) have shown that whereas for meteorites the O^{16}/O^{18} ratio is in general a constant value and approaches the O^{16}/O^{18} ratio for ultrabasic rocks, the Migei, Staroye Boriskino and Groznaya carbonaceous chondrites are enriched with a heavy isotope of oxygen O^{18} (Table 54). For these meteorites the O^{16}/O^{18} ratio is 488.9-489.0. According to their date the Novyy Urey ureilite, like carbonaceous chondrites, is also enriched with the heavy isotope of oxygen O^{18}.

Taylor and Duke, et al, (1965) recently investigated the isotopic composition of the oxygen in six carbonaceous chondrites, the Goalpara ureilite and other meteorites. They also noted that the total oxygen in carbonaceous chondrites is enriched with the isotope O^{18} in comparison with other meteorites. A gradually increasing enrichment with the heavy isotope of oxygen O^{18} was noted within the group of carbonaceous chondrites with a transition from type-III to type-I.

In type-III carbonaceous chondrites the value δO^{18} varies from -0.8 to $5.5^0/_{00}$, in type-II it varies from 8.4 to $11.3^0/_{00}$ and type-I (Orgueil meteorite) it is $12.2^0/_{00}$ (Table 55). In addition, Taylor and Duke investigated the isotopic composition of oxygen in individual mineral varieties from meteorites and demonstrated that in its isotopic composition the oxygen in the olivine of carbonaceous chondrites differs from the olivine of ordinary chondrites. In carbonaceous chondrites the olivine differs with respect to the isotopic composition of oxygen in having a greater scatter of δO^{18} values, from -2 to $3.6^0/_{00}$, which also distinguishes the meteorite from other chondrites. On this basis the author postulates that the olivine in carbonaceous /216 chondrites has a different origin than the olivine in ordinary chondrites. The pyroxene in the Murray carbonaceous chondrite also differs from the pyroxene in other meteorites, including the Goalpara ureilite, with respect to the isotopic composition of its oxygen. The Goalpara ureilite, it might be mentioned, differs from all other pyroxenes analyzed by the authors. They believe that the Goalpara ureilite was somehow related in its origin to carbonaceous chondrites, and that possibly this meteorite is a recrystallized carbonaceous chondrite.

All the meteorites investigated by Taylor and his colleagues were classified into three types on the basis of the isotopic composition of oxygen: 1) carbonaceous chondrites of three types and ureilites: in this group the isotopic composition of the oxygen in the pyroxenes varies greatly; 2) achondrites, hypersthenic achondrites and mesosiderites rich in calcium; the δO^{18} of the

Table 55

Isotopic Composition of Oxygen in Carbonaceous Chondrites as a Whole, Olivine and Pyroxene in Carbonaceous Chondrites and the Pyroxene in the Goalpara Ureilite (Taylor, Duke, Silver, Epstein, 1965)

Type of Meteorite	Meteorite	Weight of Samples		Olivine		Pyroxene	
		No. of Determinations	δO^{18}, ‰	No. of Determinations	δO^{18}, ‰	No. of Determinations	δO^{18}, ‰
Carbonaceous Chondrites							
Type-I	Orgueil	2	12,2±0,3	—	—	—	—
Type-II	Murray	3	8,4±0,4	2	−2±0,3	—	—
	»	2	10,6±0,2	—	—	1	−0,5
	Al Rais	3	11,3±0,3	1	3,6	—	—
Type-III	Mokoia	1	5,5	—	—	—	—
	Vigarano	4	3,0±0,3	2	2,4±0,5	—	—
	Felix	3	−0,8±0,2	—	—	—	—
Ureilite	Goalpara	—	—	—	—	3	8,6±0,3

The standard is ocean water.

pyroxenes varies from 3.7 to 4.4‰; 3) olivine-hypersthene, olivine-bronzite and enstatic chondrites, as well as enstatitic achondrites (aubrites) and augites: δO^{18} for the pyroxenes varies from 5.3 to 6.3‰. The authors believe that carbonaceous chondrites are possibly the initial substance for other types of meteorites.

In the future it will be of interest to study the isotopic composition of the oxygen in organic compounds and in other "low-temperature" components in carbonaceous chondrites. Clayton (1963) found that the δO^{18} value for the oxygen in the carbonates found in the Orgueil meteorite is from -1.0 to -3.3‰.

In the second chapter we mentioned that in addition to C, H, S, N and O the matter in carbonaceous chondrites is enriched in comparison with ordinary chondrites by volatile admixture elements (F, Cl, and others) (See Table 7). Some of them are associated with the carbonaceous matter. Thus, for example the chlorine content in carbonaceous chondrites is approximately 0.03%. A third of the chlorine is present in organic compounds (See Table 22). The chlorine is also associated with chlorides, found in carbonaceous chondrites

among other water-soluble salts (DuFresne, Anders, 1963). As indicated by Table 7, there is a general regularity in the distribution of volatile admixture elements: there is an increase in their content with transition from type-III carbonaceous chondrites to type-I meteorites. This increase occurs /217 in general synchronously with an increase in the content of organic compounds.

Thus, the quantitative content of "low-temperature" mineral components containing volatile elements increases with transition from type-III carbonaceous chondrites to type-I, parallel with an increase in the quantitative content of organic compounds[1]. The isotopic composition of the volatile elements (C, H, S, O) also changes in this direction. These elements are enriched with heavy isotopes.

The character of distribution and interrelationship between "low-" and "high-temperature" components in carbonaceous chondrites evidences a secondary origin of the "low-temperature" components. The volatile compounds exert an influence. In this process many of the "low-temperature" components, and probably all of them, which are now observed in carbonaceous chondrites were formed synchronously. The temperature at which the volatile compounds exerted an effect on the earlier "high-temperature" matter in carbonaceous chondrites and replaced this matter apparently did not exceed 300-600°, and possibly was much lower. The effect of volatile compounds occurred in the direction from type-III carbonaceous chondrites to type-I meteorites.

All this is evidence that the matter in carbonaceous chondrites, enriched with organic matter and other compounds containing volatile elements, corresponds to the matter found in the upper zones of the parent bodies from which meteorites originated.

Obviously, the highly volatile compounds which exerted an effect on the matter found in carbonaceous chondrites existed in some form in the early matter even prior to the formation of the parent bodies from which meteorites originated. However, the process of their effect probably occurred not only after the formation of the parent bodies from which the meteorites originated, but even during the formation of these parent bodies, during the time of their agglomeration.

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[1] We indicated above that with transition from type-III to type-I carbonaceous chondrites there is a gradual change in all the peculiarities of their composition. This probably indicates that with respect to genesis all three types of carbonaceous chondrites are interrelated, that is, their secondary modification occurred in the direction from type-III to type-I. Therefore, the author believes that in the future it evidently is irrational to separate type-III carbonaceous chondrites into an individual group of meteorites, olivine-pigeonite chondrites. The same conclusion has been drawn by B. Mason (Amer. Mineral., 52, 307, 1967).

Importance of Volatile Components in the Evolution of Matter in Carbonaceous Chondrites

The volatile elements entering into the organic compounds of meteorites, as well as in terrestrial organic matter, occur widely in different cosmic objects. It is well known that in the solar system H and He occur most abundantly, followed by O, Ne, C, N, Si, Mg, S (Suess, Urey, 1956; Merrill, 1959, Aller, 1963, and others) (Table 56). We have no basis for assuming that these elements were also absent in the primary matter from which the parent bodies from which meteorites originated were formed.

Probably, prior to the formation of asteroidal bodies in the solar system there was some spatial differentiation of volatiles, as can be judged, in particular, from the different composition of the atmospheres of the inner[1] and outer planets. The volatile elements in the primary matter probably formed part of simple compounds (CH_4, CO_2, NH_3, H_2O, and others), existing in a frozen state, and also apparently in stable active compounds of the type H_2O_2, C_2H_2, C_2H_4, etc. The greater part of these compounds must be in an adsorbed state /218 on chondrules and their fragments scattered in space.

Table 56

Distribution of Chemical Elements in Space
(in atoms per 10^3 Si atoms)
(Suess, Urey, 1956 and Cameron, 1959)

Sequence of Distribution	Chemical Elements and Their Atomic Distribution
10^6	H (25), He (3,8)
10^3	O (25), Ne(14), C(9,3), N(2,4), Si(1,0), Mg(0,91), S(0,38), Ar(0,15), Fe(0,15)
10^1	Al(9,5), Ca(4,9), Na(4,4), Ni(2,7), P(1,0), Cr(0,78), Mn(0,68), K(0,32), Cl(0,26), Co(0,18), Ti(0,17), F(0,16)
10^{-2}	V, Cu, Zn(20), Li(10), Sr(6), Kr(4), Sc, Ge(3), Be, B, Se, Pb(2), Zr(1,4), Ga, Y(0,9), Rb(0,6), Br, Ba(0,4), Te, Xe(0,3), As, Mo(0,2), Sn, Pt(0,1)
10^{-4}	Ru, Cd, Nd(9), Nb(8), Pd, Dy(7), J, Ce, Er, Os(6), Cs, La, Gd, Ir(5), Yb, Hg(4), Ag, Tl, Bi(3), Rh, Sb, Pr, Sm, Ho(2), In, Eu, Hf, W, Au(1)
10^{-5}	Tb, Tm(9), Re(5), Lu(4), Th(3), Ta(2), U(0,8)
—	Tc, Pr, Po, At, Rn, Fr, Ra, Ac. Pa, Np, Pu, Am, Cm, Bk, Cf, Es, Fm, Mv, No, Lr, Ku

[1] It should be noted, in passing, that the earth's present-day atmosphere was formed by secondary processes.

It is not impossible that these ices contained free radicals, CN, OH, and others (Donn, 1962; Vdovykin, 1966a). This can be attributed to the peculiarities of the physical conditions in space. The temperature of solid particles in the asteroidal ring in any case did not exceed 150°K. The pressure in space is estimated at approximately 10^{-16} mm Hg.

The free radicals could appear during ultraviolet solar irradiation and irradiation by cosmic rays. It is known that the photon energy necessary for breaking a bond in compounds must exceed or be equal to the energy of bond dissociation. The energy of dissociation of compounds, which could exist even prior to the formation of asteroidal bodies, has, according to N. N. Semenov (1958), the following values (in Cal/mole):

$H_2 \rightarrow 2H$	103,24
$CH_4 \rightarrow CH_3 + H$	101,0
$NH_3 \rightarrow NH_2 + H$	102,0
$H_2O \rightarrow H + OH$	116,0
$C_2H_6 \rightarrow 2CH_3$	83,32.

These bonds can be broken by light with a wavelength λ = 300 mμ or greater (Minkov, 1962).

Under conditions of a low temperature and a deep cosmic vacuum, the free radicals in an unrecombined state could exist for quite a long time. Urey and Donn (1956) postulated the presence of free radicals in this primary matter from which meteorites originated. However, they assumed that the free radicals could be stable if at an earlier time the solar thermal flux was less strong than it is at the present time. Rejecting these hypotheses of an increase in solar heat, they actually rejected their own idea of a major role of free radicals during the heating of the parent bodies from which meteorites were formed. However, the experiments described in such sources as a book by /219
Minkov (1962) on the formation of free radicals from a gas, as well as during irradiation of solid bodies under conditions of low temperatures and pressures, as well as other data, show that many frozen free radicals do not recombine at quite low temperatures. In laboratory experiments with low temperatures it was possible to identify OH, NH, and others, from the optical spectra. During irradiation of methane at a low pressure and a temperature of 4.2°K it was possible to identify CH, C_2, CH_2; CHO and others were identified when a mixture of CH_4 and O_2 was irradiated. It was noted at the same time that irradiation of frozen compounds by hard rays or ultraviolet light is more effective for obtaining radicals than is the irradiation of gas, but that in laboratory experiments a relatively small part of the radical is stabilized.

The possible presence of free radicals in the primary matter from which meteorites was formed can be concerned by the existence of hydroxyl OH in space (Bowers, 1963). The earliest members of the solar system, now existing at the present time, are comets (See Glased, 1964). According to the model devised by Whipple, Urey and Donn (Whipple, 1950; Urey, Donn, 1956; Donn, Urey, 1956; Donn, 1962) they constitute a conglomerate of solid mineral particles

of ices of frozen gases and frozen free radicals.

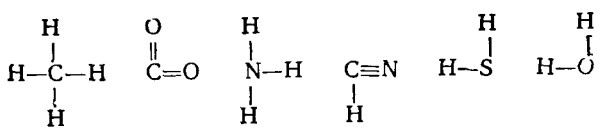
During the agglomeration of the primary cold matter into asteroidal bodies it was sufficient that there be a weak heating caused by some gravitational compression in order to cause the melting of particles consisting of volatile elements and also the recombination of free radicals possibly present. In the latter case some quantity of energy could be released. For example, Urey and Donn (1956) give the following energies released during the recombination of free radicals (at 298°K, in cal/g):

$$2OH_{\text{solid}} = H_2O_2 \ 1300,$$
$$4OH_{\text{solid}} = 2H_2O_{\text{gas}} + O_{2\ \text{gas}} \ 1700,$$
$$2CH_{\text{solid}} = C_2H_{2\ \text{gas}} \ 8000,$$
$$2CH_{\text{solid}} = 2C_{\text{solid}} + H_{2\ (\text{r.})} \ 10\,000,$$
$$C + O_{2\ \text{gas}} = CO_{2\ \text{gas}} \ 1900,$$
$$NH_{\text{solid}} = NH_4N_3 \ 3000,$$
$$2NH_{\text{solid}} = N_2 + H_2 \ 3300.$$
$$2CH_{\text{solid}} + 10OH_{\text{solid}} = 2CO_2 + 6H_2O \ 4000,$$
$$C_2H_{2\ \text{solid}} + 5H_2O_{2\ \text{fluid}} = 2CO_2 + 6H_2O \ 1800$$

etc. The energy release could cause some melting of the matter in the parent bodies from which the meteorites originated. Naturally, such an energy source does not eliminate the possibility of other sources of energy which could be released by decaying short-lived radioactive isotopes (Fe^{60}, Cl^{36}, Al^{26}, I^{129}, Pd^{107}, and others), radioactive isotopes with a large half-life (K^{40}, U^{235}, U^{238}, Th^{232}), etc.

At the present time it is difficult to estimate the contribution of any kind of energy. Obviously, they all could exert an effect on the processes of development of the matter in asteroids, but at different stages in this development.

Even at an early stage in agglomeration of asteroidal bodies gaseous compounds could rise to the surface zones of these bodies, on the way reacting with one another and with the stable active compounds present in the intermediate zones. This process as well could be accompanied by the release of heat and local heating of the matter in asteroids. Under such conditions there was a breaking of some of the bonds in the molecules of gaseous compounds (Calvin, 1964): /220



with their subsequent chemical reactions. The possible direction of many such reactions and the yield of forming organic compounds have been studied experimentally.

In this complex natural process reactions of the following types are possible:

1) Dehydrogenization, for example:

$$2CH_4 \rightarrow CH \equiv CH + 3H_2 \quad (-57{,}8 \text{ Cal} \; ; \; C_2H_6 \rightarrow CH_2 = CH_2 + H_2;$$

2) Thermal radical polymerization (with the formation of active centers), such as:

$$CH_2 = CHR \rightarrow \cdot CH_2 - CHR$$
$$CHR - CHR + CH_2 = CHR \rightarrow \cdot CH_2 - CHR - CH_2 - CH_2R$$

3) Catalytic polymerization, initiated by the presence of haloid, nitrile or carboxyl group;

4) Polycondensation, in particular, heteropolycondensation, cyclization, etc.

The catalytic effect of the mineral medium apparently was of great importance in these transformations. The forming organic compounds and the mineral medium surrounding them were closely interrelated, exerting a mutual effect on one another.

The process of formation of organic compounds inevitably must have been accompanied by the formation of more high-molecular substances and with an increase of the molecular weight of the hydrocarbon compounds their mobility, as is well known, decreases. Therefore, some quantity of hydrocarbon matter remained in a thermally bound state in the intermediate zones of asteroids, being adsorbed on the surfaces of mineral grains. Here there could be a further cyclization and ordering of the carbon atoms, which to a considerable degree favored further transition to a crystalline order.

This process, possibly transpiring in the intermediate zones of asteroids can be confirmed by the presence of graphite in a number of chondrites. However, the greater part of the volatile components enriched the upper zones of the asteroidal bodies where temperature and pressure were minimum. There these volatiles were condensed, adding to the highly volatile substances initially present in the upper zones of the asteroids. The compounds initially present there in turn became more complicated as a result of dehydrogenization, polymerization, etc.

Thus, the organic components of the upper zones in asteroids probably are a result of two processes, the transformation of compounds initially present there and subjected to cosmic and solar irradiation, and transformations of volatile compounds, the products of distillation from the deeper zones of asteroids. Naturally, during degassing the greater part of the volatiles dissipated, evaporating into space.

This outline of the general process of transformation of hydrocarbon compounds in the forming asteroidal bodies is confirmed by the pattern of distribution of organic components in the three types of carbonaceous chondrites. The matter in type-I carbonaceous chondrites, most enriched with organic compounds was formed in a medium richest in volatile elements and under conditions of minimum temperature effects. Such conditions could exist in the surface part of an asteroidal body. /221

Deeper (from the surface) parts of the upper zones in asteroidal bodies, which probably correspond to the matter found in type-II and type-III carbonaceous chondrites, experienced some heating during the course of development of the asteroids. Here the cyclization of hydrocarbons, their compaction with the formation of polynucleate aromatic hydrocarbons, occurred simultaneously with the arrival of new quantities of gaseous compounds containing C, H, N, Cl, S and O from below. The macromolecules of aromatic carbon from the surface could react with these new quantities of simpler compounds, which possibly contained free radicals. This probably was responsible for the presence of functional groups observed in the structure of high-molecular organic matter in carbonaceous chondrites. This character of the reactions of free radicals at the surface of already existing organic macromolecules can be confirmed by the results of a synthesis of macromolecules performed by Wilson (1960). Wilson (1962), incidently postulates that the formation of organic macromolecules by the same mechanism which he observed in his experiment could have occurred on the moon.

Thus, in the course of evolution of the matter in carbonaceous chondrites a major role was played by the volatile components, forming organic compounds in these meteorites. Among the early volatiles methane and other hydrocarbons probably were of considerable importance. It is probable that a major role was also played by compounds containing hydrogen: CO_2, or possibly CO and H_2O. This can be judged from the high degree of oxidation of the organic compounds. In addition, the gases contained H_2S, HCl, NH_3 and others, since components containing S, Cl and N are noted among the organic compounds of meteorites. Such a complex mixture of primary gases also was responsible for a great variety of organic compounds in carbonaceous chondrites.

Obviously, the volatile components entering into the upper zones of the asteroidal body with some increased temperature naturally exerted effect on the mineral components of these zones. The effect of volatiles on the mineral phases of carbonaceous chondrites has been discussed by DuFresne and Anders (1962b, 1963) and also by Nagy, et al (Nagy, Meinschein, Hennessy, 1963), who on the basis of other data assume that the matter in carbonaceous chondrites corresponds to the matter in the upper zones of cosmic bodies from which meteorites originated, of asteroidal (Anders) or planetary size (Nagy) respectively. Anders and Nagy, independently of one another, concluded that the matter in carbonaceous chondrites was formed at moderate temperatures in the presence of water in a slightly alkaline medium with an oxidation-reduction potential of approximately -300 mV. They published an equilibrium diagram which was compiled by Garrels (1960) for somewhat similar conditions. This diagram /222

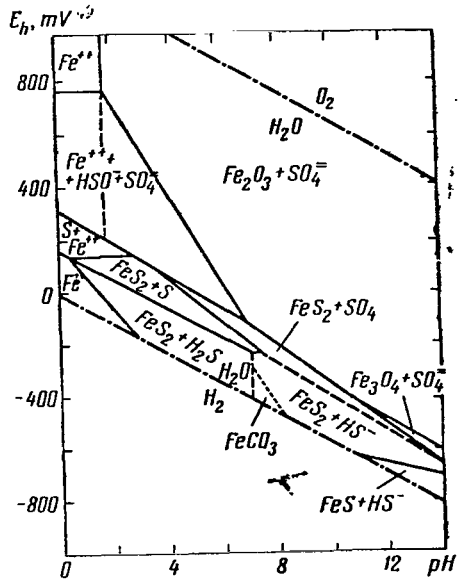


Figure 131. Diagram of the Stability of Some Mineral Components in Carbonaceous Chondrites as a Function of Their Oxidation-Reduction Potential (E_h) and Concentration of Hydrogen Ions pH (the diagram was taken from an article by DuFresne, Anders, 1962b; the base used was a diagram by Garrels, 1960).

can correspond to the mineral composition of carbonaceous chondrites (Figure 131).

Nagy feels that the body from which meteorites originated was of a planetary size, on which a water medium existed which favored biogenous processes. However, this hypothesis is based on the detection of remnants of life ("organized elements") in carbonaceous chondrites and on a certain similarity between the bituminous matter in carbonaceous chondrites and the organic matter in rocks. In actuality, the presence of "organized elements" has not been confirmed and the bituminous matter, as we have seen, has a great similarity to the products of laboratory synthesis from simple initial compounds.

DuFresne and Anders make the assumption that the volatiles were distilled into the upper zones of asteroids during the differentiation of these cosmic bodies. The surface parts of the asteroids were frozen; the matter found in carbonaceous chondrites was formed at some distance from the surface, in the zone where liquid water existed.

The author assumes that the volatiles entered into the upper zones already during the stage of agglomeration of asteroidal bodies. In this process they not only reacted with one another with the formation of organic compounds, but also intensively acted upon the mineral medium of the upper zones of the asteroids. Since the effusion of volatiles transpired through the interchondrule space, they in turn had to exert an effect on the matter within this space. The gases contained compounds in which oxygen was present; there oxygen oxidized the matrix in carbonaceous chondrites. In the surface zones of asteroids, where a concentration of volatiles increased more and more, the oxidation process became increasingly stronger. This may be indicated by the oxidized state of the interchondrule matter in type-III meteorites and the maximum degree of oxidation in type-I carbonaceous chondrites. For example, under the influence of oxygen pyroxenes could yield olivine in a reaction which has been studied by Mueller (1963, 1964):

$$2MgSiO_3 + 2Fe + O_2 \rightarrow Mg_2SiO_4 + Fe_2SiO_4,$$

all reactions with the formation of magnetite transpired there:

$$3FeSiO_3 + {}^1/_2O_2 \rightarrow Fe_3O_4 + 3SiO_2.$$

However, the formation of magnetite is also possible in a different way:

$$2FeS + 2O_2 = Fe_3O_4 + {}^3/_2S_2$$

The latter reaction during the association of pyrite (troilite) and magnetite is possible at temperatures of 600° with P_{S_2} from 1 to 10^{-8} atm (Krauskopf, 1960, 1961).

An increase in the concentration of volatile compounds in the upper part of the asteroidal body could cause the replacement of silicates by minerals of the chlorite-serpentine type. The reactions of serpentinization of olivine at a temperature up to 500° have been discussed by Dir, Khaui and Zeusman (1966). This process in all probability transpired with the participation of water. N. I. Khitarov (1959 and elsewhere) experimentally demonstrated that water can exist in rocks at quite high temperatures and pressures.

The process of substitution of silicates is possible in the direction proposed by Lindgren (1934):

$$4Mg_2SiO_4 + 2CO_2 + 4H_2O = Mg_6(Si_4O_{10})(OH)_8 + 2MgCO_3.$$

V. I. Mikheyev (Stulov, 1960) assumed that the process of substitution by hydrous silicates is possible for a meteorite also without the participation of the water in the reaction: /223

$$3Mg_2SiO_4 + Fe_2SiO_4 + 2CO + 2CH_4 = Mg_6(Si_4O_{10})(OH)_8 + 4C + 2Fe.$$

It is most probable that this process transpired with the participation of many gases; the effect of the gases on the mineral components was not uniform. This can explain the nonequilibrium chemical composition of both the olivine and the chlorite-serpentine in carbonaceous chondrites.

The effect of volatiles on silicates probably led not only to a partial substitution of the olivine in type-II carbonaceous chondrites, but in type-I even to complete substitution. The volatile compound could destroy the chondrules. In actuality, in type-I meteorites the chondrules were completely destroyed. The chondrules in type-II carbonaceous chondrites are small in size, approximately 0.5 mm in diameter in comparison with 2 mm in diameter in ordinary chondrites and here they are "eaten away" not only along the periphery, but in the inner parts as well. This same process probably is responsible for the presence of carbonaceous matter in the chondrules themselves and in individual mineral grains. Under such conditions other "low-temperature" minerals could also be formed in carbonaceous chondrites. In all probability the nonuniform effect of the volatile on the matter in carbonaceous chondrites was also responsible for the nonuniform petrographic structure of these meteorites. These nonuniformities in the structure of carbonaceous chondrites are described by Mueller (1966) as "inclusions".

During the course of this complex interaction of the mineral phases and volatile components there was an isotopic exchange, as is indicated by the results of determination of the isotopic composition of volatile elements in individual components of carbonaceous chondrites.

Thus, the process of formation of organic compounds in carbonaceous chondrites occurred simultaneously with the transformation of all matter in these meteorites. It is most probable that the process began simultaneously with the agglomeration of matter in asteroids. At all stages the transformation of the organic components could be accompanied by the formation of free radicals.

After the fragmentation of the parent bodies the carbonaceous chondrites in open space were later fragmented further; since the matter in these meteorites was rather brittle, they were fragmented more frequently than other meteorites. This is confirmed by data on the radiation age, which is lower for carbonaceous chondrites than for other meteorites.

Origin of Ureilites and the Diamonds Which They Contain

In formulating any model of the parent bodies from which meteorites originated it is very important to study ureilites because for a long time the presence of diamonds in the Novyy Urey meteorite has been known. The diamonds in the Novyy Urey ureilite and the diamond in the Canyon Diablo meteorite were one of the principle arguments in the Urey hypothesis (1956, 1959a) that the "primary" parent body for meteorites was a lunar or somewhat greater size. Urey believes that at the center of such a cosmic body the pressures were sufficiently high for diamond to be formed.

A. Ringwood (1960a) also postulated that the diamonds in the Novyy Urey ureilite were formed within a cosmic body of lunar size. Ringwood noted that in its mineralogical composition this meteorite is similar to chondrites, but differs in its chemical composition. Ringwood feels that the matter in this meteorite was apparently originally of chondritic structure, but was subjected to strong metamorphism. L. G. Kvasha (1958), as a result of a study of the /224 mineral composition and structure of all three ureilites, noted a secondary character of the structure of ureilites and postulated that these meteorites were subjected to a process of heating apparently accompanied by not very rapid cooling.

Lipschutz and Anders (1961b) concluded that the diamonds in the Canyon Diablo iron meteorite were formed during the falling of the meteorite. They postulated that the diamond in the Novyy Urey and Goalpara ureilites were also formed, in all probability, during collision. In addition, Anders (1961) assumed that the diamonds in ureilites could be formed from tiny graphite crystals which were oriented perpendicular to the direction of the impact at the time of a strong collision. If this mechanism of the formation of diamond in ureilites could occur, then, as postulated by Anders, the diamonds in ureilites should be oriented primarily in one predominant direction. This hypothesis was confirmed by Lipschutz (1964b); he demonstrated that the diamonds in the

Goalpara and Novyy Urey ureilites have a predominant orientation.

Carter and Kennedy (1964) recently again returned to the hypothesis that the diamonds in the Canyon Diablo iron meteorite and in the Novyy Urey ureilite were formed at a high pressure within a large parent body from which meteorites originated. A convincing critique of this hypothesis was given by Anders and Lipschutz (1966a, b). They cited the following proof that the diamonds in ureilites were not formed at high hydrostatic pressure, but under the influence of an impact under cosmic conditions: 1) the structure of ureilites, giving evidence of deformation caused by impact; 2) asterism of silicates; 3) predominant orientation of the diamond; 4) some peculiarities in mineral composition. The peculiarities in structure of the Canyon Diablo iron meteorite, as demonstrated by these authors,(Lipschutz, 1965b, Anders, Lipschutz, 1965; Heymann, Lipschutz, et al, 1965; Heymann, Lipschutz, et al, 1966) give evidence of the formation of diamonds in this meteorite at the time of its impact against the earth.

The origin of the diamonds in the Novyy Urey and Goalpara ureilites was discussed by Hall (1965). In comparing the structure of these meteorites with the structure of the deformed Coconino sandstones from the Arizona meteorite crater, we found that these structures to a certain extent are similar. He postulated that at the time of impact of the Canyon Diablo meteorite pieces of deformed and recrystallized rocks were ejected beyond the sphere of the earth's attraction and transformed into ureilites. In this process diamonds were formed in the secondary meteorites from the carbonaceous matter scattered in the rock at the time of impact of the Canyon Diablo meteorite. In this hypothesis advanced by Hall concerning secondary meteorites, as well as in the hypothesis of V. N. Lodochnikov (1939) on the origin of meteorites as a result of terrestrial volcanic eruption, which is related to Hall's hypothesis, it is difficult to explain the ejection of terrestrial matter into a circumterrestrial orbit.

Most researchers now agree that the diamonds in the Canyon Diablo iron meteorite were formed at the time of impact of this meteorite against the earth, but the diamonds in ureilites were formed at the time of collision of asteroidal bodies in space (Anders, 1965a; Vinogradov, 1965, and others). However, it was unclear from what meteorites ureilites were formed.

The author (Vdovykin, 1964c, 1965a, b; Vinogradov, Vdovykin, 1966) feels that the similarity of certain peculiarities in the composition of ureilites and carbonaceous chondrites, characteristic only for these two types of meteorites is evidence that they are genetically interrelated. Ureilites were formed, in all probability, during the recrystallization of matter of carbonaceous chondrites at the time of collision of asteroidal bodies.

Some of these peculiarities in the composition of ureilites and carbonaceous chondrites have been mentioned above, but they have been clarified with particular completeness during recent years, during which there has been a more detailed study of these meteorites. /22

Ureilites, like carbonaceous chondrites, contain an increased quantity of carbon. Emphasizing the similarity of these meteorites on this basis, A. N. Zavaritskiy and L. G. Kvasha (1952) classify ureilites as carbonaceous achondrites. Ureilites also contain organic matter (Teclu, 1870).

Above we mentioned the similarity in the isotopic composition of some volatile elements in ureilites and carbonaceous chondrites and the difference from other meteorites. According to data published by A. V. Trofimov (1950), with respect to isotopic composition the carbon in the Novyy Urey ureilite is close to the carbon in carbonaceous chondrites; for it the ratio C^{12}/C^{13} = = 90.9. As pointed out by A. P. Vinogradov, et al (1958), in its isotopic composition of total oxygen the Novyy Urey ureilite is similar to carbonaceous chondrites and differs from other meteorites (See Table 54). Taylor, et al (1965) recently determined that the pyroxenes in the Goalpara ureilite, like the pyroxenes in carbonaceous chondrites, have a variable isotopic composition of oxygen (See Table 55). On this basis Taylor combines the Goalpara ureilite and carbonaceous chondrites into a single group. The similarity of ureilites and carbonaceous chondrites is confirmed by the identically high content of primary inert gases (Eberhardt, 1960, 1961; Stauffer, 1961, 1962) (See Table 33).

In their chemical composition ureilites are characterized by a low content of FeS and metallic iron (M. I. D'yakonova, 1964; Wiik, 1966)(See Tables 36 and 37). This is also characteristic for carbonaceous chondrites.

It should be emphasized once again that ureilites have been poorly studied However, the data now available indicate without question that there is a similarity in a number of characteristics of ureilites and carbonaceous chondrites, apparently evidence of their genetic interrelationship.

The principal differences between carbonaceous chondrites and ureilites are in their structure, chondritic and achondritic respectively, and the presence of diamonds in ureilites. The diamonds are in a nonequilibrium state with graphite, and also with the carbonaceous matter. A large part of the latter, in contrast to carbonaceous chondrites, is represented by bituminous components. Data from a luminescent-bituminological analysis and from infrared spectroscopy gives evidence of some difference in the bituminous components in ureilites and carbonaceous chondrites. In ureilites they have a reduced character and are represented by hydrocarbons.

It can be surmised that the hydrocarbon matter in carbonaceous chondrites, particularly aromatic carbon, was recrystallized from graphite to the diamond found in ureilites. However, if this recrystallization occurred under conditions of prolonged high static pressure necessary for the synthesis of diamonds it would be difficult to expect that the bituminous compounds would be preserved at the same time.

Under conditions of brief exposure to high pressure, organic compounds have been observed after the pressure was removed. In this process their molecular weight had been increased and their structure had been disrupted, which was associated with different transformation and deformations. The

genetics of the process has been studied very poorly for the time being, although the importance of studies of the behavior of organic compounds at high pressures had been pointed out as early as 1939 by N. D. Zelinskiy. With respect to this problem experiments have been made by Bridgman (1952), L. F. Vereshchagin (Vereshchagin, Matveyeva, 1952, and others), Yu. N. Ryabinin (1956), M. G. Gonikberg (1960) and other researchers.

It is known that modern methods make it possible to measure dynamic pressures in a region greater than 5 mgbar; however, experiments for the study of the effect of pressure on different materials have been made to a pressure of 1 mgbar, and the results of studies at still lower pressures have been published (Eyring, 1964). /226

At a pressure of several kilobars the polymerization of a number of liquid olefins and aldehydes has been experimentally observed. With shock (impact) pressure in the region of 10 kbar, Koffer, et al, noted that some petroleum hydrocarbons experienced shattering and polymerization. Beyr, et al (1964) investigated the behavior of succinic acid at a pressure up to 50 kbar. In the infrared spectrum of this acid they observed displacements of the adsorbtion bands, evidence of a disruption in structure, as well as a decrease in the intensity of the adsorbtion lines. These authors note changes in the structure of *n*-nitrophenol which was subjected to a pressure up to 160 kbar. For this chemical they also demonstrated a change in the intensity of the adsorbtion lines in the infrared spectrum and a displacement of these lines.

The reduced character of the bituminous compounds in the ureilites, the composition of the hydrocarbon matter and the structure of ureilites as a whole indicate without question that these meteorites were subjected to a shock effect.

Heymann and Lipschutz, et al (Heymann, 1964; Heymann, Lipschutz, et al, 1966) cited convincing proof that the diamonds in the Canyon Diablo iron meteorite were formed during the collision of the meteorite with the earth's surface. They demonstrated that there is a correlation between the initial depth of the samples found in the Canyon Diablo meteorite before its fragmentation (the authors determined this depth from the content of cosmogenic isotopes) and the intensity of the impact which the samples experienced at the time of their falling (the intensity of impact was determined on the basis of a metallographic study). Heymann believes that all samples of this meteorite are remnants of an outer zone with a thickness of several meters which was situated in the rear part of the meteorite. It was found that the meteorite samples collected near the wall of the Canyon Diablo crater were subjected to the greatest impact (about 1 mgbar). In the meteorite they were situated at a depth of 135-180 cm. These samples contain diamonds.

The meteorite samples which were collected on the plain surrounding the crater, with very few exceptions, do not contain diamond. These samples were earlier situated in the meteorite at a depth of 74 cm. During falling of the meteorite they were subjected to a weaker impact (less 0.7 mgbar).

During collision with the earth the meteorite formed a crater 1,265 m in diameter with a depth up to 174 m[1] (Nininger, 1956; Krinov, 1962a). Many computations have shown that the total energy of the meteorite impact was 20 megatons of tnt. This corresponds to a mass of $1.6 \cdot 10^5$ - $1.34 \cdot 10^6$ tons. Coesite was found in the Arizona crater, and recently SiO_2 with a density of 4.3 g/cm^3 (Chao, et al, 1962; Fahey, 1964), which earlier had been synthesized at the Institute of High-Pressure Physics of the Academy of Sciences USSR under laboratory conditions at a pressure of 150 kbar and a temperature of 1500°C (Stishov, Popova, 1961)[2].

In contrast to the Canyon Diablo meteorite, ureilites could not have been recrystallized at the time of collision with the earth. They weigh several kilograms. Their velocity during falling apparently did not exceed several hundred meters per second. During the falling of the Novyy Urey meteorite only a shallow hole was formed in plowed land (half the diameter of the meteorite sample; Yerofeyev, Lachinov, 1888). It should be noted that all /227 three ureilites fell at different times and at different places but have a completely identical structure and mineral composition, particularly the Novyy Urey and Dyalpur ureilites. This indicates that the recrystallization of the matter in the meteorites did not occur during falling to the earth, but had already occurred in space. Finally, if the diamonds in the ureilites were formed at the time of these meteorites with the earth, one would expect the presence of diamonds in many other meteorites containing carbon, including in carbonaceous chondrites and in the Kainsaz meteorite. The latter, however, contains only graphite and carbonaceous matter, although its weight is about 200 kg (Krinov 1963a).

All this makes it possible to conclude that the diamonds in ureilites were formed during impact while the meteorite was still in space. Since ureilites, in contrast to all other meteorites, are similar in many respects to carbonaceous chondrites, but are highly recrystallized, this makes it possible to regard carbonaceous chondrites as the primary matter from which ureilites were formed. This conclusion is logical to draw from the model of general development of carbonaceous chondrites proposed above, since the matter in the latter form the outer zones of asteroidal bodies or a single asteroidal body.

It is obvious that if carbonaceous chondrites represented independent formations, whose matter was the primary substance for other meteorites, especially ordinary chondrites, then at the time of impact of any other solid

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[1] It might be mentioned in passing that in the Soviet Union there are two groups of meteorite craters: on Saaremaa Island in Estonia (Kaali) and the Sikhote-Alin group. In the Transcaspian region, on the Mangyshlak Peninsula there is a group of depressions and among them, one of the largest depressions with interior drainage in the world, the Karagiye depression. Its area is about 1,200 km, the relative elevation of the slopes is greater than 200 m and the absolute elevation of the floor is 132 m. However, the formation of the Transcaspian depression was the work of weathering processes (Vdovykin, 1964b).

[2] Recently DeCarli and Milton (1965), as well as A. A. Deribas, et al (1966), synthesized super dense SiO_2 by impact.

body at cosmic velocities (about 30 km/sec) against the matter of carbonaceous chondrites, which has a low density, the latter would simply be fragmented, dispersed or evaporated. In this case pressure would not cause the synthesis of diamonds, for which high compression conditions are required. This could happen if in the asteroidal body a matrix of denser matter was situated under the matter making up carbonaceous chondrites. The density of underlying matter in the asteroidal body could be the same as the density of the matter in the other impacting asteroidal bodies. The collision of two bodies probably would lead in the last analysis to fragmentation of both of them. The strongest compression of matter at the time of collision should be at the point of impact in the surface zone, that is, in the zone consisting of the matter from which carbonaceous chondrites were formed.

The theory of strong impact (with cosmic velocity) was developed by L. I. Sedov (1966), K. P. Stanyukovich (1960, 1966, and elsewhere), G. I. Pokrovskiy (1966a, b) and other researchers on the basis of the law of similarity for strong explosions formulated by L. D. Landau. Many examples of the effect of strong shock waves on various substances have been given in the corresponding literature.

The author made great efforts to estimate the magnitude of the energy accompanying collision of asteroidal bodies which led to their destruction. The probable qualitative picture of a collision could be as follows.

Directly in the zone of impact, with conversion of the kinetic energy into thermal energy, the matter is completely destroyed and evaporated. Since the amplitude of the shock wave changes, beyond its front there could be zones of residual deformations, zones of strong destruction of matter (its recrystallization), moderate and partial destruction (formation of breccia, formation of veins, etc.) and a zone of elastic deformations.

Naturally, the compression wave was propagated not only into the depth of the asteroid, into its denser matter; the wave front also affected the surface zones consisting of the less dense matter from which carbonaceous chondrites were formed.

The matter in carbonaceous chondrites as a whole experienced complex physicochemical transformations[1]. In this process the mineral grains and chondrules present in the more mobile carbonaceous matter must have been combined and recrystallized. The process of compaction of matter was accompanied by some change in the mineral and chemical composition[2]. For example, in the /228

[1] If the matter present in the deeper zones of the particular asteroid, subjected to a collision, can be related by analogy with rocks to "brittle" or "elastic" substances, carbonaceous chondrites obviously combine the properties of both "brittle" and, in part, "plastic" bodies. The latter was caused by the presence of carbonaceous matter.

[2] The conversion of orthopyroxenes, which are observed in carbonaceous chondrites into clinopyroxenes, characteristic for ureilites, has been confirmed by an experimental study by Sclar, et al (1964), who used pressures from 20 to 130 kbar in their experiments.

compaction process the hydroxyl group must have been set free from the chlorite-serpentine minerals. Magnetite probably could be reduced to ferronickel, which was facilitated by the presence of carbon. The ferronickel grains were spatially reformed, forming margins along individual grains of olivine or in the case of a somewhat lesser stress at the time of compression could also remain in a finely distributed state. The oxygen set free during reduction probably reacted with other components (for example, with elementary sulfur). According to data published by L. F. Vereshchagin, sulfur is oxidized at a pressure of 30 kbar in the presence of even minute admixtures of oxygen.

This possible process of transformation of matter is indicated by the structure and composition of ureilites, in which oriented large grains of silicates, and sometimes fragmented chondrules are scattered. In a roentgenometric study the silicates in ureilites reveal a clearly expressed asterism, which it is assumed is the result of an impact. Anders and Lipschutz (1966a) observed asterism for dunite subjected to an impact pressure of 600 kbar. For olivine, which was subjected to other factors (thermal, mechanical and chemical processing), Anders and Lipschutz did not observe asterism.

The carbonaceous matter was redistributed at the time of collision. Its composition and structure must also have been modified. It underwent a stage of complex transformations, during which there was apparently a destruction of the organic compounds, subsequent formation of different organic substances, loss of some part of the volatiles from the reaction zones and a strong growth of the carbon lattices in the macromolecules of aromatic carbon. The growth of the carbon lattices in the carbonaceous matter, as pointed out by many researchers (for example, Bresler, et al, 1952, 1965, and elsewhere), occurs as a result of the removal of volatiles when there is a temperature increase, which was inevitable at the time of shock compression. The polymerization of macromolecules, probably transpiring with the participation of macroradicals which formed in this process (Bresler, Yerusalimskiy,1965), could result in the formation of graphite particles. In the graphite the carbon lattices, as is well known, are situated at a distance of 3.41 Å (Figure 132a). Strong shock compressions must have caused a further compaction, reorientation with the formation of a diamond structure, in which C-C distance was 1.54 Å (Figure 132b), that is, there was a polymorphic transformation of the forming graphite into diamond in a solid phase. Naturally, the microcrystals of diamond must have been formed most readily from those graphite particles in which the c axis was situated along the direction of compression.

One can scarcely expect a significant isotopic fractionation of carbon in this last stage in the transformation of carbonaceous matter (graphite-diamond)[1]. Hoering (1961), for example, demonstrated that in its isotopic composition of carbon a synthetic diamond differs from the initial graphite /229
by $0.3^0/_{00}$; this exceeds by only a factor of two the measurement of air.

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[1] This can be confirmed by recent studies of the isotopic composition of the carbon in graphite and diamond in meteorites (See Table 51).

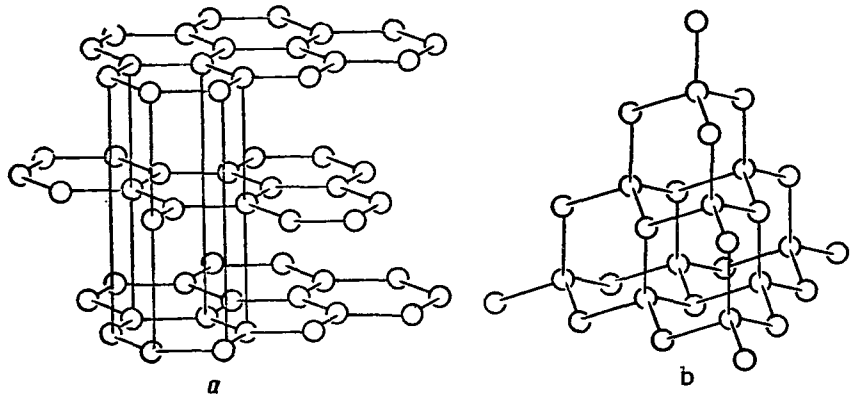

Figure 132. Structure of Graphite (a) and Diamond(b) (Bragg, Claringbull, 1965).
In graphite the distance between C-C atoms in layers is 1.42 Å; the distance between layers is 3.41 Å. In diamond the distance between C-C atoms is 1.54 Å.

This complex step-by-step, but actually rapid process of the transformation of carbonaceous matter at the time of impact apparently probably also can explain the nonequilibrium composition of carbonaceous matter in ureilites. The formation of diamond in this process was accelerated by the catalytic effect of the metallic phase, that is, the latter not only experienced the effect of the carbonaceous matter, but itself exerted an effect upon it. Grains of kamacite and troilite also subsequently remained in part in these reaction sectors. This can be concluded from their presence in the polymineral aggregates of graphite and diamond in ureilites.

The genetic interrelationship between the carbonaceous matter in ureilites and carbonaceous chondrites can be confirmed by the presence of some quantity of nitrogen in the bituminous matter and in the diamonds in ureilites. It is assumed that nitrogen enters into the diamond lattice at the time of diamond crystallization, not later (Milledge, Meyer, 1962). However, in the process of transformation of carbonaceous matter during the considered shock compression, a large part of the volatile elements, including hydrogen, oxygen and sulfur [1], as well as the alkali elements, must have been transported from the reaction zones. Since the matter in carbonaceous chondrites was situated in the surface zone of a fragmenting asteroidal body, these volatiles were transported in the form of simple gaseous compounds along the most weakened sectors, that is, compression fissures. After compression and fragmentation of the asteroidal bodies there was a rapid cooling of the matter and therefore traces of removal of the volatiles must have been retained. This process, in all probability can explain the macroscopically easily visible pores and voids which in ureilites extend in general in a single direction.

[1] The absence of free and other forms of "volatile" sulfur in ureilites has been discussed by Mueller (1966) (See Table 49). Mueller also feels that this was also possibly caused by impact.

The model of formation of diamond in ureilites during the recrystallization of the carbonaceous matter in carbonaceous chondrites at the time of collision between asteroids which has been proposed by the author can be confirmed by many modern studies of the synthesis of diamond.

In synthesizing diamonds it is common to use a method developed by Bundy, et al (1955) and Liander (1955). The diamond is obtained by the recrystallization of carbon at a pressure of 55-90 kbar and at a temperature of 1500-2500°, but the crystallization of diamond occurs here from a supersaturated solution of carbon in a metal melt. /230

Figure 133. Crystals of Synthetic Diamond (The photograph was provided to the author by V. P. Butuzov, 1967).

In 1961 DeCarli and Jamieson (1961) synthesized very small black particles of diamonds (<10 μ) from graphite with an imperfect structure by means of a directed shock wave created in an explosion. The compression pressure was approximately 300 kbar. The behavior of graphite during shock compression was studied by Alder and Christian (1961). During a time of approximately 1 microsecond they created a pressure between 300 and 600 kbar at a temperature of 1000-2000°. Bundy (1964) brought about a polymorphic transformation of graphite into diamond in the solid phase in a static apparatus in which the pressure attained 130 kbar at a temperature of 3500°. The mechanism of polymorphic transformation of graphite into diamond has been studied by L. F. Vereshchagin, et al (1965).

At the present time synthetic diamonds are also obtained under a static pressure up to 100 kbar with the participation of catalysts, iron, nickel, and others (Bundy, 1963a, b; 1965; Bundy, Strong, 1965). It was found that aromatic carbon, less perfect in its crystal structure, gives better results in the synthesis of diamonds than graphite with a perfect crystal structure (Bundy, 1963b). Moreover, Eversoul (Bezrukov, et al, 1966) was granted a patent for

obtaining diamonds in the thermal decomposition of compounds containing carbon (CO, CH_4) at temperatures 600-1600° and pressures from 0.1 mm Hg to several kilobars. In this process the synthethic diamond was built up on diamond nuclei. V. A. Bezrukov, et al (1966) reported the successful synthesis of diamonds with low parameters (Figure 133).

Figure 134 shows a diagram of the phase state of graphite and diamonds which was constructed by Bundy (1963b) on the basis of thermal dynamic computations and experimental data published by many authors. The diagram has been supplemented by data published by V. A. Bezrukov, et al (1966). On the basis of the phase diagram and other comparisons it can be postulated that the diamond in ureilites were formed in the range of pressures studied by DiCarli and Alder, that is, about 300-600 kbar. Lipschutz (1964c) believes that the diamonds in the Novyy Urey and Dyalpur ureilites were formed at a pressure of approximately 300 kbar, caused by impact; however, the diamonds in the Goalpara meteorite were formed at a pressure of about 700 kbar.

Thus, the structure of ureilites, the characteristics of the composition /231
of their matter as a whole and the composition of the carbonaceous matter are evidence that ureilites in all probability were formed from carbonaceous chondrites at the time of collision of the parent bodies from which meteorites originated. The matter in carbonaceous chondrites was the substance which formed the upper zones of these parent bodies or parent body. At the time of collision between asteroids this matter was subjected to the strongest effect of the shock waves, a compression wave. The carbonaceous matter was redistributed and the aromatic carbon underwent a transition through the stage of graphitization into the structure of diamond. The bituminous matter experienced a series of complex transformations. The silicate phase was recrystallized; at the same time there was some change in the chemical composition and the metallic phase was reduced. The process of the formation of diamond was accompanied by the elimination of some part of the volatile components.

Farther along the surface zone of the asteroid, where the shock waves had a lesser amplitude, it could cause brecciation of the meteorite and exert other effects.

L. G. Kvasha (1963) noted sectors with a crystalline structure in the Migei carbonaceous chondrite. Milton and DeCarli (1963) experimentally demonstrated that at the time of impact maskelynite, a glass of plagioclase composition could be formed in meteorites. They observed the formation of maskelynite from plagioclase at a pressure of 250-300 kbar. The transformation occurred in the solid state. Fredriksson, et al also demonstrated that at the time of impact there can be formation of veinlets in chondrites, and other phenomena can also occur (Fredriksson, DeCarli, et al, 1963, 1964; Fredriksson, DeCarli, 1964).

To be sure, the presence of breccia and veins in meteorites could be caused not only by the collision of asteroidal bodies from which the meteorites originated, but also by the subsequent collision of the fragments of these parent bodies in space.

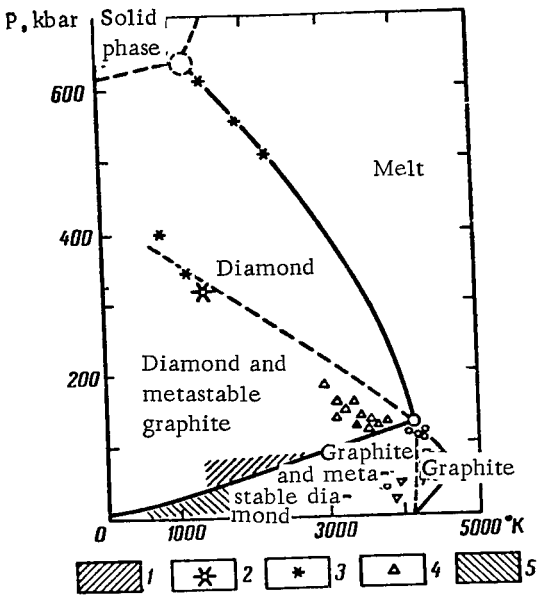

Figure 134. Diagram of the Phase State of Carbon (graphite-diamond). Experimental data: 1 - Bundy, et al, 1955; Liander, 1955; 2 - DeCarli, Jamieson, 1961; 3 - Adler, Christian, 1961; 4 - Bundy, 1963; 5 - Bezrukov, et al, 1966.

What is the probability of such destructive collisions between asteroids? Anders (1963a, b; 1965b) and B. Yu. Levin (1965) cite computations by Keyper which show that the probability of collision between asteroids is high: for the 10 largest asteroids the probability of collision of any of them during a period of $3 \cdot 10^9$ years of 0.1. According to data obtained Piotrovskiy (Anders, 1963a), the mean lifetime between collisions for a typical asteroid is 10^8-10^9 years. Urey (1963), on the basis of a study made by Öpik, also believes that the possibility of collisions between bodies moving in intercepting orbits is high. For example, Urey points out: "the probability of collisions between a meteorite moving near the earth's orbit and the earth is 1 collision in 25 million years". /232 However, it must be remembered that these and other computations apply to the present-day distribution of asteroids, whose recorded number at present is more than 1,700 (Anders, 1965). Obviously, prior to the fragmentation of the parent bodies the number of asteroids was far less and the probability of their collision was accordingly smaller. However, ureilites and the diamonds present in them, in the author's opinion, are probably the result of collision between primary parent bodies and not collisions between smaller fragments.

The results of study of organic compounds and other forms of carbonaceous matter in carbonaceous chondrites and ureilites make it possible to give an overall consideration to the characteristics of formation of these meteorites. In their composition carbonaceous chondrites in all probability correspond to the matter present in the upper zones of an asteroidal body, modified by secondary processes; during collision of the asteroidal body with another body ureilites were formed from the matter of carbonaceous chondrites. All three types of carbonaceous chondrites are interrelated with respect to a regular change in the content of volatile elements and other peculiarities in the structure of these meteorites. This gives basis for assuming that carbonaceous chondrites, at least most of them, constitute the matter present in the upper zones of one asteroidal body which apparently was the parent body from which they originated. During the agglomeration of this parent body its matter contained a great quantity of highly volatile components.[1]

[1] M. Studier, et al (Studier, Hayatsu, Anders, 1965a-c) feel that organic substances could be formed in the solar system even prior to the agglomeration of the parent bodies from which meteorites originated.

The origin of meteorites of other types (including enstatic chondrites), except for carbonaceous chondrites and ureilites, is not considered by the author. Possibly, beneath the matter from which carbonaceous chondrites were formed in the parent body there was matter from which ordinary chondrites or achondrites were formed. However, if we take into account the process of evolution of the matter in carbonaceous chondrites described above, it is more probable to assume that the volatiles, rising to the outer zone of the asteroidal body, would cause a rather strong modification of the matter present in the intermediate zones of this body. However, a study of the carbonaceous matter in meteorites which possibly formed the intermediate zone will scarcely be of particular assistance in solving this problem. This matter was subjected to high temperatures and pressures and the organic compounds must have been carbonized because of this. Therefore, organic compounds possibly are encountered extremely rarely in small quantities (traces) in other meteorites. The carbon which they contain is usually represented by graphite. However, in the process of rising to the outer zones the highly volatile components could exert some effect on the mineral medium of the intermediate zone. This could result, for example, in a nonequilibrium chemical composition of the silicates, disruption of the isotopic composition of their oxygen, etc.

In this respect it can be very well worth noting that Taylor, et al (1965) found that the isotopic composition of the oxygen in pyroxenes extracted from the Bencubbin mesosiderite, which contained carbon, had very great variations. This distinguished the Bencubbin mesosiderite from the other meteorites which they investigated and indicated a closer resemblance to carbonaceous chondrites and the Goalpara ureilite. Taylor believes that the Bencubbin mesosiderite is genetically somehow related to the Goalpara ureilite. However, it /23
is more probable that in actuality its matter originated in the inner zones of the asteroidal body from which carbonaceous chondrites originated.

In explaining the origin of carbonaceous chondrites and ureilites the author based his study on the idea formulated by A. P. Vinogradov that the chondrules in chondrites predated the latter.

This priority of chondrules has also been accepted by other researchers (Suess, 1949; Levin and Slonimskiy, 1957), but the formation of the chondrules has been explained differently. Wood (1963a) believes that the primary matter from which asteroids and the planets of the solar system were formed consisted of reduced chondrules and oxidized dust. This was matter of the type found in the Rennazo carbonaceous chondrite. However, the oxidation of the matrix of this meteorite was in all probability a secondary process.

To be sure, the priority of chondrules does not mean that carbonaceous chondrites represent the matter found in ordinary chondrites with secondary modification under the influence of volatiles. The matter found in ordinary chondrites is possibly a product of the process of some redistribution and fractionation of matter possibly occurring even prior to the formation of the asteroidal bodies from which meteorites originated. This may be indicated by some differences in the chemical composition of chondrites and the ratio of some elements which it is customary to assume under terrestrial conditions

Table 57

Total Content of Iron and Fe/Mg and Fe/Si Ratios in Chondrites

Group	Chondrites	Fe_{tot}%**	Fe/Mg	Fe/Si
—	Enstatic			
HH	Carbonaceous	28,52	0,97	0,81
H	Type-I: Aláis	27,00	0,82	0,90
	Orgueil	28,48	0,87	0,91
	Type-II: Migei	26,18	0,79	0,82
	Staroye Boriskino	27,35	0,87	0,86
	Type III: Groznaya	26,65	0,81	0,83
	Felix	26,17	0,78	0,82
H	Olivine-bronzite	26,32	0,83	0,79
L	Olivine-hypersthene	21,85	0,64	0,59

*HH - group with extremely iron content (Wiik, 1956), H - high iron content, L - group with low iron content (Urey, Craig, 1953).

**For the carbonaceous chondrites the analyses were made by Wiik (1956) and Mason (1963d); for the other chondrites the table gives average data presented in publications by Mason (1963a, b).

characterize processes of differentiation of matter under conditions of somewhat increased temperatures and low pressures.

A. A. Yavnel' (1963b) examined for meteorites such a pair of elements as iron-silicon; he demonstrated that on the basis of the ratio of different forms of iron chondrites can be rarely classified into groups; each group is characterized by its own typical Fe/Si ratio. In a study of the differentiation of magmas under terrestrial conditions A. A. Yaroshevskiy (1965) takes into account the iron-magnesium ratio. The ratio of both pairs for the products of differentiation of magmas increased upwards in the cross section.

Table 57 gives the ratios of these pairs of elements and the total iron content in chondrites.

These data may indicate some thermal process which exerted an effect during the formation of the matter found in chondrites. There are at least two possible explanations: 1) the fractionation occurred even prior to the formation of the asteroidal bodies (some spatial fractionation of matter of solar composition); however, here we encounter a number of cosmogonic diffi- /234
culties; 2) the fractionation occurred within the parent asteroidal bodies from which meteorites originated, but the ascent of matter in this case occurred in the form of droplets through a rather dense "cold" medium; this process is not known to us under terrestrial conditions.

In any case, the character of the distribution of organic compounds and other components containing volatiles in carbonaceous chondrites definitely indicates that the volatiles entered the outer zones of the asteroidal bodies (or body), exerting an effect on the "high-temperature" mineral matter on chondritic structure already existing there. This evidences the priority of chondrules, but is not evidence of their formation of process of differentiation of the parent asteroidal body.

It can be concluded that carbonaceous chondrites cannot be regarded as the primary matter for ordinary chondrites, like ordinary chondrites are considered the primary matter for carbonaceous chondrites. Both are products of some fractionation of matter. However, in mineral composition the two, if we exclude products of secondary transformation in carbonaceous chondrites, in the first approximation are rather close. Therefore, the mineral composition of ordinary chondrites is closer to the initial composition of matter for all chondrites. The chemical composition of carbonaceous and ordinary chondrites in general is also similar, if we exclude the volatile elements in carbonaceous chondrites. However, volatile elements entered into the primary matter from which asteroidal bodies were formed. Accordingly, in a study of the composition of the primary matter we must not exclude the volatiles. Type-I carbonaceous chondrites (of the Orgueil type), most enriched with volatiles as a result of the specific nature of the development of their matter, correspond better than other meteorites to the composition of the primary matter from which the asteroids, the parent bodies for meteorites, were formed. It must be emphasized very strongly that in chemical composition they are closer than all other meteorites to the primary matter, but they do not constitute this primary matter.

The carbonaceous matter in iron meteorites in turn underwent a stage of complex transformations. It contains graphite and clistonite, carbides (primary cohenite Fe_3C and rarely moissanite SiC), diamonds and carbonaceous matter. The latter contains an insignificant quantity of organic compounds. Obviously, on the basis of the presence of traces of organic compounds it is difficult to draw conclusions concerning the origin of iron meteorites because the organic compounds which they contain in all probability have a secondary character. Their carbon apparently passed through the cohenite stage. More reliable data on the origin of iron meteorites can be obtained by a study of the conditions of formation of cohenite; these have been discussed in many works (Ringwood, 1960b, 1965; Ringwood, Seabrook, 1962; Lipschutz, Anders, 1964; Olsen, 1964; Lovering, 1964; Brett, 1966)[1]. The author of this book did not make a special study of cohenite.

At an early stage the cohenite apparently could be formed in a reaction between carbon and iron or in a reaction between iron and methane. With further cooling and crystallization of ferronickel there probably was a redistribution of matter causing diffusion (Khauffe, 1963) which resulted in desegregation of compounds containing carbon in the form of isolated nodules.

[1] The cohenite in the Canyon Diablo and Odessa iron meteorites was investigated by Lipschutz, et al (Brown, Lipschutz, 1965; Lipschutz, Jaeger, 1966).

In addition to carbides, these nodules contained troilite, schreibersite and other minerals. This crystallization evidently transpired much like the crystallization of carboniferous alloys (Zubarev, 1956, 1957).

With the cooling of the matter in iron meteorites to a temperature of approximately 600° the unstable structure of cohenite breaks down with the release of carbon[1] in the form of graphite[2]. Under these same conditions there is formation of molecular hydrogen, which is capable of reacting with the carbon in cohenite, again with the formation of methane, in the well known reaction of $Fe_3C + 2H_2 \rightarrow 3\ Fe + CH_4$. In turn, methane can serve as the basis for a number of organic compounds. Accordingly, here there could be a synthesis of hydrocarbons and the destruction of carbonaceous compounds. /235

Thus, the carbonaceous matter in meteorites underwent a complex path of development, during which simple initial compounds were transformed into high-molecular matter. These transformations were determined by the physicochemical conditions of the environment in which it existed, that is, by the general development of the asteroidal bodies from which the meteorites originated.

The most probable energy sources in the complex process of synthesis of organic compounds and the transformation of carbonaceous substances in meteorites were not only solar ultraviolet radiation and cosmic radiation, but also the energy released during the decay of radioactive elements. Also of some importance were temperature and pressure, and in individual cases, also dynamic pressure, compression accompanying the collision of the parent bodies from which meteorites originated. The process of transformation of carbonaceous matter in meteorites was accompanied by a catalytic effect of the mineral medium in which it was distributed. At different stages this process could be complicated not by the formation of compounds with a more complex structure, but on the contrary, by the destruction of organic molecules. Therefore, meteorites contain a series of carbonaceous compounds ranging from carbonaceous matter and the organic compounds constituting part of this matter in carbonaceous chondrites, graphite, characteristic for iron meteorites, pallasites and ordinary chondrites to diamonds, forming in ureilites at the time of the destruction of asteroids during their collision, and in the Canyon Diablo iron meteorite at the time of collision of this meteorite against the earth.

The distribution of carbonaceous matter in meteorites of different types, the composition and structure of this matter make it possible to represent the conditions of formation of meteorite matter as a whole. In this book the author has purposely not considered other fields for which the results of study of the carbonaceous matter in meteorites are of importance.

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[1] Isotopic exchange possibly occurred at the same time. This can be concluded, in particular, from the different isotopic compositions if the carbon in cohenite and graphite in the Canyon Diablo octahedrite, as was observed by Craig (1953) (See Figure 127).

[2] The formation of spherulites of graphite under somewhat similar conditions was recently examined by A. A. Baranov and K. P. Bunin (1963), I. Ye. Bolotov, (1965), E. N. Pogrebnoy, K. M. Zhak (1966) and others.

Obviously, the results of study of carbonaceous matter and the meteorites which contain it are of importance for clarifying problems concerning the origin of life, the origin of petroleum and gas, various conditions for the crystallization of diamonds, the evolution of terrestrial matter as a whole, in particular, for example, not only the biosphere, but also the hydrosphere and atmosphere, and possibly the evolution of all planets of the earth group. It is known that in the model of zone smelting of the matter from which the earth's crust is formed, proposed by A. P. Vinogradov and successfully developed by him, an allowance is made not only for melting, but also for degassing, the escape of volatiles. According to A. P. Vinogradov (1964a), degassing resulted in a concentration of highly volatile elements (C, H, N, O and others) in the outer layers of the earth. A somewhat similar process apparently also transpired during the development of the parent bodies from which meteorites originated, in which volatile elements enriched the outer layers.

It has become clear that in the course of its evolution the earth to a /236
considerable degree lost its light inert gases but retained H_2O, C, N and other gases (Table 58). Miller and Urey (1964) feel that already during the early stages in the earth's formation carbon could not enter into such simple compounds as CH_4, which also would be lost. Probably, as they postulate, carbon could be found in carbides, carbonates, graphite or even in more or less complex organic compounds.

Accepting the proposed model of development of the matter found in carbonaceous chondrites, it is obvious that the surface zones of the earth, even in an early stage of its development, must not have contained carbides, but instead complex organic compounds, probably the same ones found in carbonaceous chondrites.

Table 58

Degree of Concentration of Volatiles in Comparison with Xenon under Terrestrial Conditions (Miller, Urey, 1964)

Highly Volatile Components	Content on Earth, mole/cm^3	Degree of Concentration in Comparison with Xenon	Part Remaining on Earth After Loss of Highly Volatile Components
Ne	$6,5\cdot10^{-4}$	$0,9\cdot10^{-4}$	$0,9\cdot10^{-11}$
$Ar^{36}+Ar^{38}$	$1,2\cdot10^{-3}$	$2,6\cdot10^{-2}$	$2,6\cdot10^{-9}$
Kr	$3,5\cdot10^{-5}$	1,0	$1,0\cdot10^{-7}$
Xe	$2,8\cdot10^{-6}$	1,0	$1,0\cdot10^{-7}$
N	54	16	$1,6\cdot10^{-6}$
C	350	2100	$2,0\cdot10^{-5}$
H_2O	15000	3200	$3,0\cdot10^{-4}$

However, Hochstim (1963) postulates that under terrestrial conditions organic compounds were synthesized as a result of exposure to a shock wave at the time of falling of large meteorites at an ultrahigh velocity during their passage through the atmosphere or during the interaction between meteorite matter and water. Miller and Urey (1964) feel that this energy contribution was very small.

There is also no basis for assuming that the organic compounds on the basis of which living systems subsequently develop were carried to the earth from space, as postulated by Oró (1961a). In his opinion, during the first two billion years of the earth's existence up to 10^8-10^{12} tons of cometary material fell upon the earth at the time of collisions with comets; part of this material consisted of carbonaceous compounds. However, the contribution of these compounds to the total quantity of terrestrial carbonaceous matter was very small (Vdovykin, 1963d; Miller, Urey, 1964). On the basis of some similarity between the organic compounds in carbonaceous chondrites and terrestrial organic matter, as well as on the basis of computations of the probable number of fallings of these meteorites, Briggs (1962a, 1963a) recently postulated that the organic compounds in meteorites can be of considerable importance in the overall balance of terrestrial organic matter (Uspenskiy, 1956; Ronov, 1958). Obviously, this contribution is very small.

The escape of volatiles and the synthesis of organic compounds in the surface zones could probably occur on other planets in the solar system as well.

The literature contains repeated hypotheses to the effect that carbonaceous matter can be present in the surface layers of the moon. A communication by N. A. Kozyrev (1959) can serve as an indirect confirmation of this; /237
in observing volcanic eruptions from the central peak of Alphonsus crater, with a study of spectroscopic data, he detected the presence of C_2 and complex multiatomic molecules among the gases. These observations were confirmed in 1963 at the Lowell Observatory (Arizona) in observations of Aristarchus crater. Urey (1961) feels that molecular carbon C_2, which is not found under terrestrial conditions, could be formed on the moon in a vacuum during the eruption and subsequent decomposition of acetylene. However, Urey feels that the acetylene is formed in the lunar interior under the influence of water on CaC_2 in the well known reaction. However, it is difficult to postulate high calcium carbide concentrations.

Another explanation of the presence of molecular C_2 in the products of a lunar eruption is possible if we assume the existence of other carbonaceous compounds in the surface layers of the moon. It is known that during heating to high temperatures complex organic compounds decompose with the formation of free radicals, as well as molecular carbon and hydrogen. Experiments with the heating of graphite in the gas phase show that the products contain C_2, C_3 and C_5 molecules in addition to carbon atoms.

To be sure, it will be sounder to draw conclusions concerning the presence of organic compounds in the matter present at the lunar surface after it has been studied in the laboratory.

SUMMARY

Only a few years ago only very unclear ideas prevailed concerning the /238 carbonaceous matter in meteorites, particularly concerning the organic compounds present. A thorough study of this matter using various up-to-date research methods has made it possible to obtain data concerning its distribution in meteorites, chemical composition and structural peculiarities. Moreover, these data make it possible to visualize the conditions under which meteorites containing carbon were formed.

In this book the author has made the first attempt, on the basis of his own and other studies, to examine thoroughly the results of studies of carbonaceous matter, organic compounds, diamond and graphite in meteorites of rare types. It has been demonstrated that the organic compounds in meteorites have an extraterrestrial abiogenous nature. Carbonaceous chondrites, containing organic compounds, in all probability correspond to the matter existing in the outer zones of the parent bodies from which meteorites originated or a single parent body of asteroidal size. Ureilites, containing diamonds, obviously were formed from carbonaceous chondrites during the collision of the parent bodies from which meteorites were formed.

Organic compounds are present in a number of meteorites. Their traces are detected in some ordinary chondrites, ureilites, and also in the carbonaceous nodules of coarse-structured octahedrites. However, carbonaceous chondrites are particularly enriched with organic compounds: they contain up to 5-7% organic matter.

A study of the morphology of organic inclusions in carbonaceous chondrites and their relationship to the mineral components, made using a polarization microscope (in sections) and under electron and ultraviolet microscopes has demonstrated that the organic compounds in carbonaceous chondrites are distributed primarily in the interchondrule space. The bituminous compounds extracted by means of solvents are natural in meteorites and are not the result of chemical transformation of the solvents themselves.

The study of the morphology of microparticles which have been described by some authors as remnants of extraterrestrial life forms ("organized elements"), made for a number of meteorites, has shown that these microparticles are unrelated to the organic compounds of carbonaceous chondrites. They constitute various formations and are primarily mineral particles, chlorite-serpentine minerals, sometimes having an envelope consisting of carbonaceous matter, and also troilite, magnetite, in some cases limonite, etc. In individual cases the "organized elements" are the result of contamination under terrestrial conditions.

The study of the chemical composition of the organic matter in meteorites has been made for two fractions of carbonaceous matter: bituminous compounds and high-molecular organic matter in carbonaceous chondrites. In the study of the bituminous matter various authors have made bituminological studies and

elementary microanalysis; it has been studied by spectroscopic methods in the infrared, ultraviolet and visible regions and has been studied by the chromatographic and mass spectrometer methods, etc.

During extraction by means of organic solvents, without heating of the latter, up to 0.5% bituminous matte been extracted from carbonaceous chondrites, and in the case of extra in a Soxhlet apparatus, up to 1% of the weight of the meteorite. In comp son with the bituminous matter in rocks, the bituminous compounds in car aceous chondrites are characterized by a high degree of oxidation and they contain increased quantities of oxygen as well as sulfur and nitrogen and chlorine. On the other hand, the bituminous matter in ureilites has a more reduced character, represented by hydrocarbons in the paraffin series. /239

The bituminous matter in carbonaceous chondrites has a complex composition. It contains hydrocarbons of the C_{15}-C_{29} series--*n*-paraffins and cycloalkanes, as well as aromatic hydrocarbons of the benzene series and polynucleate aromatic hydrocarbons. Among the latter it has been possible to identify phenanthrenes, pyrenes, chrysenes and other hydrocarbons, as well as individual hydrocarbons such as anthracene, 3,4-benzopyrene, and others. In a number of carbonaceous chondrites it has been possible to identify aromatic acids, and in the Orgueil meteorite there are fatty acids of the type $C_nH_{2n}O_2$, among which there is a predominance of acids with an even number of carbon atoms in the chain. In this same meteorite there are traces of organic compounds containing sulfur, that is, sulfides and heterocyclic compounds of thiophenes. Some carbonaceous chondrites contain amino acids in carbohydrates, as well as such nitrogenous cyclic compounds as melamine, ammeline, adenine and guanine.

Most of the carbonaceous matter in carbonaceous chondrites extracted from meteorites after being freed from the mineral components by processing with acids is primarily high-molecular (polymeric) organic matter with a condensed aromatic structure. It contains free organic radicals, possibly formed in meteorites under the influence of cosmic irradiation.

In contrast to carbonaceous chondrites, the carbonaceous matter in the three ureilites is represented primarily by diamond and graphite. Aggregates of diamond and graphite, with a black color, irregular configuration and up to 0.3-0.9 mm in diameter, have a rather uniform distribution in ureilites, being found among large recrystallized silicate grains. In polycrystalline and polymineral aggregates containing diamonds there is kamacite, troilite and chromite in the form of very small grains. The monomineral fraction of diamond extracted from the Novyy Urey meteorite by fusing the pulverized matter of this ureilite with sodium peroxide consists of light-colored particles of diamonds; each tiny particle consists of micromonocrystal grains measuring <1 μ. The diamond in ureilites is characterized by inhomogeneities in the crystal structure, caused at least in part by admixtures of donor nitrogen.

In iron meteorites, coarse-structured octahedrites, the carbonaceous matter is represented primarily by graphite, which is frequently encountered in such meteorites in nodular form. In its structure this graphite does not differ from terrestrial graphite. X-ray studies of the clistonite in octahedrites has not revealed structural differences between this mineral and the graphite in iron meteorites.

In this book it has been shown that the organic compounds in meteorites are an integral part of the carbonaceous matter in meteorites. They are natural in meteorites and cannot be the result of contamination under terrestrial conditions. On the other hand, the organic compounds in meteorites have some similarity to terrestrial organic matter. All the organic compounds identified in carbonaceous chondrites are also known under terrestrial conditions. The distribution of the principal classes of organic compounds in meteorites is somewhat similar to the distribution in various geological objects. However, there are differences within each class. /240

Some peculiarities in the chemical composition of the organic compounds in meteorites, the isotopic composition of their carbon and hydrogen, as well as the nature of the distribution of these compounds in carbonaceous chondrites (the presence of carbonaceous matter not only in the interchondrule space, but in the composition of the chondrules) give evidence of an extraterrestrial radiogenic nature of the organic components in meteorites. This is confirmed by experimental studies on the synthesis of organic compounds.

The general similarity between organic compound in meteorites, the organic compounds formed by abiogenous synthesis from simple initial substances, and terrestrial organic compounds of biogenous nature, as well as the similarity in the structure of the diamond and graphite in meteorites and similar terrestrial minerals, indicate a similar nature of the transformation of carbonaceous compounds regardless of the place where they were transformed (under terrestrial conditions or in the parent bodies from which meteorites originated) and regardless of the origin of the organic compounds.(abiogenous or biogenous). This indicates, incidently, that biogenous evolution under terrestrial conditions must be regarded as part of the overall process of transformation of carbonaceous compounds in nature.

The carbonaceous matter in meteorites, which is most sensitive of all the other meteorite components to changes in its environment, can serve as a singular indicator of this environment, that is, of the peculiarities of evolution of the matter in meteorites as a whole. The distribution of the organic compounds in carbonaceous chondrites, the regular change in the composition and structure of the carbonaceous matter in these meteorites parallel with the changes in the mineral composition of carbonaceous chondrites, in the content of volatile elements and their isotopic composition, all these things indicate that carbonaceous chondrites in all probability correspond to the matter existing in the outer zones of asteroidal bodies from which meteorites originated. The matter in carbonaceous chondrites was subjected to the influence of volatile components. This confirms the idea advanced by A. P. Vinogradov that the chondrules found in chondrites predate the chondrites.

Some characteristics in the structure of ureilites and carbonaceous chondrites and the peculiarities of their carbonaceous matter give evidence of genetic interrelationships between these types of meteorites. It is most probable that ureilites were formed from the matter of carbonaceous chondrites at the time of collision between asteroidal bodies in space. At the time of collision the carbonaceous matter in the carbonaceous chondrite was redistributed and recrystallized through graphite to the structure of diamond. This can be confirmed by the synthesis of diamonds under pressure. The collision caused a recrystallization of the matter in meteorites as a whole and a loss of some of the volatile components. The diamond in the Canyon Diablo iron meteorite was probably formed at the time of impact of the meteorite against the earth.

Naturally, some aspects of the origin of meteorites containing carbon that have been discussed in this book are debatable and require further experimental confirmation. It is also necessary to continue the study of the carbonaceous matter in meteorites and this should be done at the molecular level. This will make it possible to detect other differences between it and terrestrial matter of biogenous nature and can cast light on many aspects of the cosmic history of meteorites. It should be emphasized very strongly that taking into account the possibility of terrestrial contamination, the organic matter in meteorites must be studied at the working level with highly purified substances during the course of future investigations.

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LIST OF METEORITES MENTIONED IN BOOK

No.	Meteorite	Transliteration of Name of Meteorite	Meteorite Type	Place of Falling	Date of Falling or Year of Find
			USSR Meteorites		
Unnumbered	Арус	Aroos	See Yardymlinskiy		
1	Брагин	Bragin	Pallasite	Belorussian SSR	1807—1952
2	Бургавли	Burgavli	Coarse-structured octahedrite	RSFSR*	1941
3	Грозная	Groznaya	Carbonaceous chondrite	RSFSR	28.VI 1861
4	Ефремовка	Efremovka	Chondrite containing carbon	Kazakh SSR	1962
5	Индарх	Indarch, Indarkh	Enstatic chondrite	Azerbaydzhan SSR	7.IV 1891
6	Каинсаз	Kainsaz	Chondrite containing carbon	RSFSR	13.IX 1937
7	Крымка	Krymka	Chondrite	Ukranian SSR	21.I 1946
8	Кульп	Kul'p	Crystalline chondrite	Azerbaydzhan SSR	29.III 1906
9	Кунашак	Kunashak	Crystalline chondrite	RSFSR	11.VI 1949
10	Ликсна	Liksna	Dark chondrite	Latvian SSR	12.VII 1820
11	Маныч	Manych	Unmodified chondrite	RSFSR	21.X 1951
12	Мигеи	Mighei, Migei	Carbonaceous chondrite	Ukranian SSR	18.VI 1889
13	Никольское	Nikol'skoe	Granular chondrite	RSFSR	6.III 1954
14	Новый Урей	Novo Urei, Novyi Urei	Ureilite	RSFSR	4.IX 1886
15	Пилиствере	Pilistvere, Pillistfer	Enstatic chondrite Unmodified	Estonian SSR RSFSR	8.VIII 1863
16	Саратов	Saratov	Chondrite, dark variety	RSFSR	6.X 1918
17	Севрюково	Sevryukovo, Sevriukovo	Octahedrite, very coarse-structured	RSFSR	11.V 1874
18	Сихотэ-Алинский	Sikhote-Alin	Carbonaceous chondrite	RSFSR	12.II 1947
19	Старое Борискино	Staroye Boriskino, Staroe Boriskino			20.IV 1930
20	Чувашские Киссы	Chuvashskie Kissy	Crystalline chondrite, dark	RSFSR	1899
21	Ярдымлинский (синоним Арус)	Yardymlinsky Iardymlinskii	Coarse-structured octahedrite	Azerbaydzhan SSR	24.XI 1959

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No.	Meteorite	Transliteration of Name of Meteorite	Meteorite Type	Place of Falling	Date of Falling or Year of Find
			Meteorites of Other Countries		
22	Эби	Abee	Enstatic chondrite	Canada	9.VI 1952
23	Алэ	Alais	Carbonaceous chondrite	France	15.III 1806
24	Ал Райс	Al Rais	Carbonaceous chondrite	Arabia	10.XII 1957
25	Бали	Bali	Carbonaceous chondrite	Cameroons	22.XII 1907
26	Бэллс	Bells	Carbonaceous chondrite	United States	9.IX 1961
27	Бенкаббин	Bencubbin	Mesosiderite	Australia	1930
28	Бьюрболе	Bjurböle	Chondrite, slightly modified	Finland	12.III 1899
29	Брэйтшейд	Breitscheid	Chondrite	West Germany	11.VIII 1956
30	Брудерхейм	Bruderheim	Gray chondrite	Canada	4.III 1960
31	Каньон Дьябло	Canyon Diablo	Coarse-structured octahedrite	United States	1891
32	Каркот	Carcote	Crystalline chondrite	Chile	Known before 1888
33	Чейнпур	Chainpur	Chondrite containing carbon	India	9.I 1907
34	Колд Бокке-велд	Cold Bokkeveld	Carbonaceous chondrite	South Africa	13.X 1838
35	Коллесчиполи	Collescipoli	Unmodified chondrite	Italy	3.II 1890
36	Косбис Крикк	Cosby's Creek	Coarse-structured chondrite	United States	Known before 1837
37	Крэнбоурн	Cranbourne	Coarse-structured chondrite	Australia	1854
38	Кресцент	Crescent	Carbonaceous chondrite	United States	17.VIII 1936
39	Диалпур	Dyalpur	Ureilite	India	8.V 1872
40	Эракот	Erakot	Carbonaceous chondrite	India	22.VI 1940
41	Эсеби	Essebi	Carbonaceous chondrite	Congo, capital Kinshasa	28.VII 1957
42	Фармингтон	Farmington	Black chondrite	United States	25.VI 1890
43	Феликс	Felix	Carbonaceous chondrite	United States	15.V 1900
44	Форест Сити	Forest City	Light gray chondrite	United States	2.V 1890
45	Гхубара	Ghubara	Black chondrite	South Arabia	1954
46	Джилгойн	Gilgoin	Crystalline chondrite, dark	Australia	1889

LIST OF METEORITES MENTIONED IN BOOK

No.	Meteorite	Transliteration of Name of Meteorite	Meteorite Type	Place of Falling	Date of Falling or Year of Find
47	Гоалпара	Goalpara	Ureilite	India	1868
48	Харипура	Haripura	Carbonaceous chondrite	India	17.I 1921
49	Хессле	Hessle	Unmodified chondrite	Sweden	1.I 1869
50	Хоба	Hoba	Ataxite rich in nickel	Southwest Africa	1920
51	Холбрук	Holbrook	Crystalline chondrite, dark	United States	19.VII 1912
52	Хоумстед	Homestead	Crystalline chondrite, dark	United States	12.II 1875
53	Хвиттис	Hvittis	Enstatic chondrite	Finland	21.X 1901
54	Ивуна	Ivuna	Carbonaceous chondrite	Tanzania	16.XII 1938
55	Йах дех Кот Лалу	Jajh deh Kot Lalu	Enstatic chondrite	Pakistan	2.V 1926
56	Каба	Kaba	Carbonaceous chondrite	Hungary	15.IV 1857
57	Карунда	Karoonda	Chondrite containing carbon	South Australia	25.XI 1930
58	Ла Лейнд	La Lande	Crystalline chondrite, dark	United States	1933
59	Лансе	Lancé	Carbonaceous chondrite	France	23.VII 1872
60	Магура	Magura	Coarse-structured octahedrite	Czechoslovakia	1840
61	Мезё — Мадараш	Mezö-Madaras	Unmodified chondrite	Rumania	4.IX 1852
62	Мокойа	Mokoia	Carbonaceous chondrite	New Zealand	26.XI 1908
63	Муррей	Murray	Carbonaceous chondrite	United States	20.IX 1950
64	Навапали	Nawapali	Carbonaceous chondrite	India	6.VI 1890
65	Ногойа	Nogoya	Carbonaceous chondrite	Argentina	30.VI 1879
66	Одесса	Odessa	Coarse-structured octahedrite	United States	1921—1922
67	Оргей	Orgueil	Carbonaceous chondrite	France	14.V 1864
68	Орнан	Ornans	Carbonaceous chondrite	France	11.VII 1868
69	Пис Ривер	Peace River	Gray chondrite	Canada	31.III 1963
70	Петц	Peetz	Black chondrite	United States	1937
71	Поллен	Pollen	Carbonaceous chondrite	Northern Norway	6.IV 1942

LIST OF METEORITES MENTIONED IN BOOK

No.	Meteorite	Transliteration of Name of Meteorite	Meteorite Type	Place of Falling	Date of Falling or Year of Find
72	Пржибрам	Přibram	Octahedite, very coarse-structured	Czechoslovakia	7.IV 1959
73	Ренаццо	Renazzo	Carbonaceous chondrite	Rumania	15.I 1824
74	Ревелстоук	Revelstoke	Carbonaceous chondrite	Canada	31.III 1965
75	Ричардтон	Richardton	Gray chondrite	United States	30.VI 1918
76	Рич Маунтин	Rich Mountain	Veined chondrite	United States	30.VI 1903
77	Сен-Совер	Saint Sauveur	Enstatic chondrite	France	10.VII 1914
78	Санта-Круц	Santa Cruz	Carbonaceous chondrite	New Zealand	3.IX 1939
79	Смитвилл	Smithville	Coarse-structured octahedrite	United States	1840
80	Спрингуотер	Springwater	Pallasite	Canada	1931
81	Сен-Капрэ	St. Caprais	Carbonaceous chondrite	France	28.I 1883
82	Сен-Маркс	St. Marks	Enstatic chondrite	South Africa	3.I 1903
83	Тайбан	Taiban	Black chondrite	New Zealand	1934
84	Толука	Toluca	Medium-structured octahedrite	New Zealand	Known before 1776
85	Тонк	Tonk	Carbonaceous chondrite	India	22.I 1911
86	Вигарано	Vigarano	Carbonaceous chondrite	Rumania	22.I 1910
87	Ваконда	Waconda	Gray chondrite	United States	1873
88	Уоррентон	Warrenton	Carbonaceous chondrite	United States	3.I 1877
89	Йоундегин	Youndegin	Coarse-structured octahedrite	Australia	1884

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