ON THE ORIGIN OF EXTRATERRESTRIAL STRATOSPHERIC PARTICLES:

INTERPLANETARY DUST OR METEOR ABLATION DEBRIS?

A Thesis

Presented to

the Faculty of the Department of Geology

San Jose State University

In Partial Fulfillment

of the Requirement for the Degree

Master of Science

By

Frank T. Kyte

August, 1977
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APPROVED FOR THE DEPARTMENT OF GEOLGY

Maxwell B. Blanchard, May 13, 1977

Marshall E. Maddock, May 13, 1977

David W. Andersen, 13 May 1977

APPROVED FOR THE UNIVERSITY GRADUATE COMMITTEE

Grant M. R.
ABSTRACT

The purpose of this thesis is to distinguish meteor ablation debris from unablated interplanetary dust in a collection of extraterrestrial particles collected in the stratosphere using NASA U-2 aircraft. A 62 g sample of the Murchison (C2) meteorite was artificially ablated to characterize ablation debris for comparison with the stratospheric particles. By using proper experimental conditions, artificial ablation debris can be produced that is similar to natural ablation debris.

Analyses of natural fusion crusts, artificial fusion crusts, and artificial ablation debris of the Murchison meteorite have produced the following criteria for recognizing debris ablated by a primitive meteoroid: 1) Melting at high temperatures and rapid cooling results in distinctive textures and morphologies. 2) A silicate "melt" phase will crystallize on cooling, forming olivine, magnetite, and a Ca, Al, Fe, Si-glass matrix. 3) A sulfide-bearing "melt" phase will crystallize on cooling, forming iron oxides (magnetite and/or wustite) and sulfides (pyrrhotite and/or pentlandite). 4) Non-volatile elemental abundances in silicate spherules will approximate those of the meteoroid from which they were derived. 5) More volatile elements (e.g. sulfur) will be depleted. 6) Any angular debris caused by fragmentation (without melting or vaporizing) will produce mafic silicate grains from meteoroid
inclusions and fine-grained aggregates from the matrix.

7) Fine-grained aggregates which fragment from the hydrated, layer-lattice silicate matrix of primitive meteoroids like Murchison can be altered to olivine and magnetite if sufficiently heated.

Nearly all (ninety-five percent) of the stratospheric particles can be described as either ablation debris from a primitive meteoroid, or as very primitive interplanetary dust. Silicate spherules having an olivine-magnetite mineral association and sulfide-bearing spherules having an iron oxide-sulfide mineral association comprise about thirty-five percent of the particles and are identified as ablation debris. Another sixty percent of the stratospheric particles are mafic silicate grains and very fine-grained aggregates which have primitive elemental compositions. These particles are similar to angular debris fragmented from the ablation model, but unablated interplanetary dust must contribute to these groups. If fragmentation produces large quantities of unmelted debris, then a greater proportion of the stratospheric particle collection is debris from meteoroids rather than interplanetary dust.
ACKNOWLEDGEMENTS

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groundwork for this project.

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INTRODUCTION

The Problem

Except for recently collected lunar materials, meteorites are the only samples available for analysis on earth which represent extraterrestrial bodies in this solar system. Stony meteorites are of great scientific interest because of their great age and primitive compositions. Interpretations of meteorite analyses have been used to explain the formational processes of our solar system.

A meteorite is the portion of a meteoroid which survives atmospheric entry and can potentially be recovered on the ground. Meteorites are classified into two groups on the basis of their mode of collection. A fall is a meteorite which was actually seen to fall from the sky and was subsequently recovered. A find is a meteorite which was never seen in flight but was found on the ground and subsequently identified as extraterrestrial.

Unfortunately for meteorite researchers, only a small percentage of extraterrestrial material entering the earth's atmosphere is ever recovered as a meteorite. The ablation caused by friction with the atmosphere at high velocities is a catastrophic process of deceleration and energy release. The high kinetic energy of the meteoroid is converted to heat and light, and during this stage the meteoroid is seen as a fireball or "shooting star". Ablation is an extreme form
of erosion on the meteoroid and its mass rapidly decreases as heat and pressure cause melting, vaporization, and fragmentation. In most cases, the meteoroid is completely destroyed and the surviving unvaporized portion is only fine-grained ablation debris. Occasionally, a small fraction of the original meteoroid survives to reach the ground where it can be recovered as a meteorite. Rarely, however, will its mass exceed 1 kg, and unless it is seen to fall, it is unlikely that it will be recovered.

Meteorite collections cannot be truly representative of the distribution of meteoroids in the solar system for three reasons. 1) Only meteoroid types which have orbits intersecting the earth's orbit can ever be recovered. There are probably meteoroid types which we have never collected. 2) Meteoroids which are friable are much less likely to survive the ablation process and would, therefore, be scarce in meteorite collections. 3) Those meteorites with the greatest resistance to weathering (e.g. iron meteorites) will survive longer on the ground and will, therefore, be overrepresented in collections. Clearly, it is only possible to hypothesize actual abundances of meteoroid types in the solar system.

Some Solutions

Meteorite researchers have employed several techniques to increase our knowledge of meteorites. These efforts have been successful in compiling observational data on fireball phenomena and in collecting more extraterrestrial materials for analysis.
An important contribution to the understanding of the nature of fireballs has been the observational data compiled by the fireball tracking networks. Beginning in 1964, two automated photographic networks (the Prairie Network in the USA, and the All-Sky Network in Czechoslovakia and West Germany) began systematically to photograph bright meteors. They calculated several variables including orbital trajectories, initial and final mass, initial velocities, atmospheric deceleration, and estimated densities (McCroskey and Ceplecha, 1968). These networks were joined in 1971 by the Meteorite Observation and Recovery Project network in western Canada (Halliday, 1973). Unfortunately, in 1974 the Prairie Network closed down after 10 years of operation.

These observational data have shown that although the mass influx at the top of the atmosphere is much greater than previously predicted (McCroskey, 1968), meteorites of recoverable size rarely survive ablation. Estimates of the frequency of occurrence of recoverable meteorites are about 1 yr\(^{-1}\) \(10^{-6}\) km\(^{-2}\) (McCroskey, 1967). On the rare occasions when these photographic networks observed a fireball with a sizeable final mass (> 1 kg) following ablation, systematic searches were made for the residual meteorite. So far, only two meteorites, Lost City and Pribram, have been recovered. Although valuable data on meteorite trajectories and fireball phenomena have been gathered, this approach has not been effective for collecting extraterrestrial materials.

Prairie Network data suggest that fireballs can be
classified into three groups based on their end-heights (end of visible trajectory). Ceplecha and McCroskey (1976) listed Group I fireballs as having the lowest end-heights and the highest densities. Group II fireballs usually have a lower initial velocity and greater total mass, but luminous end heights are at a greater altitude, indicating less resistance to ablation. Group III fireballs begin to ablate at anomalously high altitudes, have very high end-heights, and have orbits beyond Jupiter similar to those of the comets. Frequency of occurrence are approximately equal for groups I and II, and much less (approximately 10 percent of the total) for Group III. Using the Lost City meteorite as a basis for density and ablation coefficients, Ceplecha and McCroskey (1976) suggest that these groups may be composed of chondritic stones (Group I), carbonaceous chondrites (Group II), and more fragile cometary material (Group III).

While the photographic networks recorded the meteor ablation process, other researchers have attempted to collect the fine-grained debris produced by meteor ablation as well as unablated interplanetary dust. With varying degrees of success, materials have been recovered from terrestrial areas of low sedimentation, such as Greenland ice (Langway and Marvin, 1964), deep sea manganese nodules (Finkelman, 1970, 1972), deep sea sediments (Schmidt and Keil, 1966; Millard and Finkelman, 1970), and Paleozoic salt deposits (Mutch, 1966). Others have attempted atmospheric collections with sounding rockets (Farlow et al., 1970; Hemenway and Soberman, 1962), balloons
(Brownlee and Hodge, 1969), and high altitude aircraft (Carr, 1970; Brownlee et al., 1976). Until recently, it has been difficult to distinguish between extraterrestrial particles and terrestrial contaminants. The only criteria for recognizing ablation debris were: 1) spherules and 2) high Ni-Fe content.

In order to better understand the ablation process and to develop better criteria for the identification of meteor ablation debris, a systematic study of ablation products was initiated (Blanchard, 1969). Blanchard (1972, 1973), Cunningham (1973; Blanchard and Cunningham, 1974), and Davis (1976) artificially ablated carefully analyzed samples of iron oxides, olivine, and Fe and Ni-Fe materials, respectively. They performed analyses on the artificial fusion crusts and ablation debris and compared the results with fusion crusts of natural meteorites. From these analyses they developed the following criteria for recognizing ablation debris: 1) Melting at high temperatures and rapid cooling produced melt features and disequilibrium mineral assemblages. 2) Distinctive textures were produced by intergrown dissimilar phases. 3) Volatile elements were vaporized. 4) Less volatile elements underwent fractionation and concentration in some phases according to their affinity for oxygen.

This Study

The purpose of this thesis is to distinguish meteor ablation debris from unaltered interplanetary dust in the
particles collected in the stratosphere (Brownlee et al., 1976). To date, over 250 particles have been collected which may be extraterrestrial. Some of these have been identified preliminarily as ablation debris on the basis of the criteria summarized by Davis (1976). Some other particles (in particular some abundant spherules composed of Fe, S, and Ni) are quite difficult to explain using the aforementioned criteria. However, the majority of these particles have elemental abundances and textures similar to the most primitive carbonaceous chondrites.

To identify the ablation debris in this collection, X-ray diffraction analyses were performed on selected extraterrestrial particles to determine their mineralogy, and a 62 g sample of the Murchison meteorite, a carbonaceous chondrite, was artificially ablated so the characteristics of the artificial debris could be compared to the stratospheric particles. This study is based on experiments that demonstrate that ablation debris from the Murchison meteorite, produced in an artificial environment, is similar to the debris produced naturally by the meteorite.
Since this study is concerned with primitive meteorites (and especially the carbonaceous chondrites), a short review of the characteristics, classification, and significance of these meteorites is appropriate.

What is a Primitive Meteorite?

The group of meteorites called the chondrites comprise some of the most primitive materials in the solar system. Their most significant property is that their relative abundances of non-volatile elements (e.g. Ca, Mg, Fe, Al), when normalized to Si, are quite similar to those estimated to be in the sun. Because of this similarity, the chondrites are considered to be undifferentiated material derived from the solar nebula. That is to say, they condensed directly out of the cooling solar nebula which formed our sun and have not been subjected to any processes of differentiation (e.g. sedimentation, magmatism) since their formation.

Chondrite Classification

The name "chondrite" is derived from the fact that these meteorites typically contain chondrules. Chondrules are spheroidal objects containing high temperature minerals or glass. They range from a few tenths to a few millimeters in size. Chondrules often account for more than 70 percent of the mass of the meteorite, and their average chemical
composition is typically the same as the matrix in which they are embedded. In some of the very primitive carbonaceous chondrites, however, chondrules are not present and the classification of these meteorites is based on elemental abundances rather than the presence of chondrules.

The chondrites are divided into three main groups: enstatite, ordinary, and carbonaceous. This division is based mainly on chemical compositions, but further subdivision incorporates physical and petrographic features as well. Atomic ratios for the refractory elements Ca, Mg, and Al relative to Si show a definite separation into these three main groups. The enstatite chondrites have the lowest concentrations of Ca, Mg, and Al, and the carbonaceous chondrites have the highest concentrations of these elements relative to Si. Values for these atom ratios (Wasson, 1974) are as follows:

<table>
<thead>
<tr>
<th>Group</th>
<th>(Ca/Si) x 10^2</th>
<th>(Mg/Si) x 10^2</th>
<th>(Al/Si) x 10^2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Enstatite</td>
<td>3.7 - 3.9</td>
<td>74 - 86</td>
<td>5.0 - 5.6</td>
</tr>
<tr>
<td>Ordinary</td>
<td>4.7 - 5.3</td>
<td>88 - 96</td>
<td>6.2 - 7.4</td>
</tr>
<tr>
<td>Carbonaceous</td>
<td>&gt;6.3</td>
<td>&gt;101</td>
<td>&gt;8.3</td>
</tr>
</tbody>
</table>

The abundance of Fe relative to Si and its distribution between oxidized and reduced phases is also quite diagnostic (Fig. 1). In the enstatite chondrites, the abundance of Fe relative to Si is quite variable but the oxidation state of Fe is invariably reduced, with Fe occurring only in the metallic or sulfide (FeS) phase. In ordinary chondrites, three distinct closely related types based on Fe abundances are recognized:
Figure 1. This figure shows the distribution of Fe between "reduced" metal and troilite and the more oxidized phases in the chondrites. The enstatite (E) chondrites contain virtually no oxidized Fe. The ordinary chondrites (H, L, and LL) may represent a fractionation trend as indicated by the curve. The carbonaceous chondrites (CI, CM, CV, and CO) contain little or no reduced Fe. The letters in the figure correspond with individual meteorites (reproduced from Wasson, 1974).
H, L, and LL. The H type have the highest abundance of Fe and are richer in the reduced phase than the other ordinary types. The L type are intermediate, and the LL's are lowest in total Fe but are the richest in the oxidized phase. The carbonaceous chondrites invariably have a high amount of total Fe but are richest in the oxidized phase. Except for Kainsaz and Renazzo, the reduced phase of metallic Fe is present only in small amounts or is absent.

Wiik (1956) originally subdivided the carbonaceous chondrites into three types based on their content of C and H$_2$O. These are termed Cl, C2, and C3 in this paper. The mean values for weight percent combined H$_2$O and C are as follows:

<table>
<thead>
<tr>
<th></th>
<th>Cl</th>
<th>C2</th>
<th>C3</th>
</tr>
</thead>
<tbody>
<tr>
<td>C</td>
<td>3.54</td>
<td>2.46</td>
<td>0.46</td>
</tr>
<tr>
<td>H$_2$O</td>
<td>20.08</td>
<td>13.35</td>
<td>0.99</td>
</tr>
</tbody>
</table>

Mason (1963), realizing that carbon in the form of graphite and diamond exists in other meteorite groups, adopted the following working definition:

"The carbonaceous chondrites are a group of stony meteorites characterized by the presence of an appreciable amount of carbonaceous material other than free carbon (diamond and graphite)."

He purposely avoided defining quantitatively the "appreciable" amount.

More recent classification schemes for the carbonaceous chondrites have utilized chemical data, physical properties, mineralogical composition, and petrography. Density is a very
useful discriminant. Density ranges for Wiik's three groups are: C1, 2.20-2.42; C2, 2.57-2.92; C3, 3.40-3.78 g/cm³ (Renazzo is an exception due to its content of 12 percent Ni-Fe).

C1 meteorites contain no chondrules and consist largely of poorly crystallized, hydrated, Mg-Fe, layer-lattice silicates. C2 meteorites contain variable amounts of chondrules and grains of Fe-poor olivine and pyroxene in a fine-grained matrix of chlorite or serpentine. They contain little or no free metal except for Kainsaz (1-2 percent) and Renazzo. In C3 meteorites the matrix is largely fine-grained Fe-rich olivine (Fa₄₀-₈₀), and they tend to be richer in chondrules than the C2's; metal content is usually low, generally 0-6 percent. Van Schmus and Wood (1967) have further subdivided Wiik's C3 group into groups C3 and C4, the latter type being characterized by larger "spongey" chondrules with a higher ratio of matrix to chondrules.

Origins

The abundance of the non-volatile elements in chondrites closely resembles that of the sun. For this reason, chondrites are considered to be undifferentiated condensates derived from the cooling solar nebula. In carbonaceous chondrites, the similarities to solar abundances are even more striking when elements which are volatile (e.g. F, S, Zn, Cl, Br, Pb) are considered. Chemical analyses have shown that there is an enrichment of these volatiles in the carbonaceous chondrites
relative to the ordinary chondrites, and volatile abundances in the Cl chondrites closely approximate solar abundances. Except for the most volatile elements (H, C, O, N, Hg, and the noble gasses), the major elemental abundances of Cl chondrites match spectroscopically estimated solar abundances so well that trace element analyses on these meteorites are currently used to define solar (and essentially cosmic) abundances.

Chemical variations in the chondrites are generally attributed to different temperatures at which they are thought to have condensed and aggregated. For instance, there is a well defined mineralogical break between the groundmasses of C2 and C3 chondrites. C3 meteorites have a groundmass of Fe-rich olivine, while the groundmass of C2 meteorites is composed of an Fe-rich hydrated, layer-lattice silicate similar to serpentine or chlorite. Mason (1971) has pointed out that "Pure magnesium serpentine decomposes to olivine at 500°C and that substitution of iron for magnesium lowers the temperature of decomposition." He, therefore, postulated a thermal boundary of 300° to 500°C between the regions of aggregation for these two types. DuFresne and Anders (1961) have found a strained glass in Mighei, a C2, which anneals at temperatures below 300°C, indicating that this C2 has not been raised to this temperature since it aggregated. Mason (1971) further points out that Cl meteorites are more fine grained and have a less well crystallized groundmass than C2 meteorites. Because C2 meteorites possess high temperature inclusions,
(e.g. olivine, pyroxene), they represent aggregates from a transitional temperature region where higher temperature minerals are drifting in from hotter regions to form inclusions in a coarser matrix.

Urey (1952) was the first to suggest that volatiles might serve as cosmothermometers to determine accretion temperatures. Anders (1971) listed 31 volatile elements which appear to be fractionated in chondrites. The simplest fractionation pattern is shown by the carbonaceous chondrites (Fig. 2). To account for these trends, Anders postulated that carbonaceous chondrites are a mixture of two components: a low temperature, volatile-rich component (the matrix) and a high temperature, volatile-free component (the inclusions). He suggested that the low temperature component comprises 100, 55, and 32 percent for CI, C2, and C3 meteorites respectively. This matches well with the relative percentages of matrix in these three types. More recent analyses (Krahenbuhl et al., 1973; Case et al., 1973) support this.

It appears likely that the chondrites (and especially the carbonaceous chondrites) must represent some of the most primitive materials in the solar system. The most primitive of these (CI) appear to be composed almost entirely of fine-grained, low temperature condensates which formed in equilibrium with the cooling solar nebula. The less primitive types (C2 and higher) contain increasingly greater amounts of high temperature condensates in a matrix containing progressively smaller amounts of low temperature minerals.
Figure 2. Volatile elemental abundances for the C2, C3, and ordinary chondrites relative to the CI chondrites show a depletion trend which probably represents variations in temperatures of condensation and aggregation for these different meteorites (from Anders, 1971).
Table 1

Chemical analyses of the Murchison (C2) meteorite by Fuchs et al. (1973) and Jarosewich (cited in Fuchs et al., 1973). The elements listed are those studied in this investigation.

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<tr>
<th></th>
<th>Fuchs et al.</th>
<th>Jarosewich</th>
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<tr>
<td>SiO₂</td>
<td>27.22</td>
<td>29.07</td>
</tr>
<tr>
<td>TiO₂</td>
<td>0.099</td>
<td>0.13</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>2.05</td>
<td>2.15</td>
</tr>
<tr>
<td>MgO</td>
<td>18.90</td>
<td>19.94</td>
</tr>
<tr>
<td>Fe (total)</td>
<td>20.44</td>
<td>22.13</td>
</tr>
<tr>
<td>MnO</td>
<td>0.22</td>
<td>0.20</td>
</tr>
<tr>
<td>CaO</td>
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<td>1.89</td>
</tr>
<tr>
<td>Na₂O</td>
<td>0.57</td>
<td>0.24</td>
</tr>
<tr>
<td>K₂O</td>
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<td>0.04</td>
</tr>
<tr>
<td>P₂O₅</td>
<td>0.23</td>
<td>0.23</td>
</tr>
<tr>
<td>Cr₂O₃</td>
<td>0.402</td>
<td>0.48</td>
</tr>
<tr>
<td>NiO</td>
<td>1.73</td>
<td>1.75</td>
</tr>
<tr>
<td>H₂O (total)</td>
<td>12.06</td>
<td>10.09</td>
</tr>
<tr>
<td>S (total)</td>
<td>3.248</td>
<td>3.00</td>
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</table>
The Murchison (C2) Meteorite

The Murchison (C2) meteorite is a carbonaceous chondrite containing a mixture of high and low temperature condensates. The chemistry and mineralogy of this meteorite have been thoroughly analyzed (see Table 1) by Fuchs et al. (1973). Murchison is mainly composed of a fine-grained, hydrated, layer-lattice silicate (Fe-rich, Al-poor chamosite) with inclusions of olivine and Ca-poor pyroxene (Fig. 3). There is very little free troilite, metal, or magnetite. Most of the sulfur, and much of the iron, is in a poorly characterized non-crystalline Fe-S-O phase in the matrix. The olivine to pyroxene ratio is 2:1. These two minerals are typically Fe-poor with a range of 1-5 percent mole fraction of Fe, although more Fe-rich grains do exist.

Chondrite Abundances

The ordinary chondrites are aptly named in that they represent 78 percent of all known falls (Wasson, 1974) for a total of over 600 (problems with classification and cataloging do not permit an exact figure). The H, L, and LL types account for 32, 39, and 7 percent, respectively, of these falls. In contrast, only 36 carbonaceous chondrites thus far have been collected, and all but 3 are confirmed falls. Ordinary chondrite finds number well over 400. These relative abundances most likely do not represent the true extraterrestrial abundances of these meteor types. The high volatile concentration and friable nature of the carbonaceous chondrites suggest
Figure 3. A thin section of the Murchison meteorite viewed in transmitted light. The visible crystals are inclusions of olivine and pyroxene which are in a black, very fine-grained, hydrated, layer-lattice silicate matrix. The size of the largest inclusion is about 1 mm.
a low probability of surviving ablation during atmospheric entry and weathering on the ground. This would greatly reduce the quantities of observed falls and recovered finds for this group. Statistics for the ordinary chondrites, however, should not be appreciably enhanced relative to the non-chondritic meteorites, such as the irons, by processes of ablation and weathering. The high fall frequency of the chondrites must imply a relatively high abundance of these meteorites in the solar system. Prairie Network data (Ceplecha and McCroskey, 1976), which suggest that the majority of meteors entering the atmosphere are similar to the chondrites, agree with fall and find statistics.
SAMPLE COLLECTION AND PRODUCTION

Artificial Ablation of the Murchison Meteorite

A 406 g specimen (#Me2684) of the Murchison meteorite was broken by hand and trimmed with a diamond impregnated copper wire to provide a 62 g sample for ablation. The sample was ablated using a plasma beam of ionized air, argon, and electrons (Fig. 4) in Ames' 0.5 in (1.3 cm) constricted-arc jet facility (Fig. 5a, 5b). Experimental conditions simulated a 30 cm diameter meteoroid travelling 12 km/sec at an altitude of 70 km. It was not possible, however, to achieve a high enough stagnation pressure in the gas cap (Shepard, et al 1967) of the experimental model to simulate pressures at this altitude. Actual impact pressures experienced by the model simulated those at an altitude well above 100 km. It should be emphasized that this experiment primarily simulated heating rates, not pressures. Lower impact pressures had a significant effect on the experiment. First, the partial pressure of oxygen was lower than in the real environment at this altitude, so oxidation was not as pronounced. Second, lower pressures with correct heating rates favored melting and vaporization rather than fragmentation in the ablation process. Because the sample was rather fragile, higher impact pressures would have destroyed it faster, producing an even larger quantity of fragmented debris.

Ablation debris was collected from the chamber for
Figure 4. Schematic drawing of a constricted arc supersonic jet (after Shepard et al., 1967)
Figure 5a. The cylindrical chamber of the constricted-arc jet facility can be seen in the background of this photograph. The experimental model is mounted on the prop which can be seen (arrow) between the author (left) and the facility engineers. During the ablation the model can be viewed through the porthole on the right.

ORIGINAL PAGE IS OF POOR QUALITY
Figure 5b. This photograph, taken during the artificial ablation, shows the model bathed in a plasma beam of ionized air, argon, and electrons.
analysis. Water cooled copper plates were placed directly behind and below the ablating model. Chrome plated trays were placed along the sides and rear of the chamber. Besides the debris collected on the plates and trays, debris was picked off the floor and walls of the chamber. The experiment continued only until the copper sample holder started to ablate. In this way, an artificial meteorite fusion crust was also obtained.

Stratospheric Particle Collection

Since March, 1974, stratospheric particles have been collected by NASA U-2 aircraft using an inertial impaction device (Ferry and Lem 1974) designed for collecting submicrometer aerosols. Minor modifications were incorporated to expose larger surface areas and collect larger particles (up to 50 μm). Fourteen of these particles were analyzed in this investigation. The characteristics of these particles were compared with the artificially created ablation debris and the natural fusion crust on the Murchison meteorite. The stratospheric particle collection is maintained by D.E. Brownlee and D. Tomandl at the Astronomy Department of the University of Washington. They perform the initial analyses using a scanning electron microscope to select the particles and screen out likely contaminants. Electron micrographs and X-ray energy dispersive elemental analyses are used to categorize the particles.
METHODS OF ANALYSIS

Optical Microscopy

Petrographic thin sections and polished sections were made from the original sample (including the fusion crust) and the ablated model. This allowed comparison between the original sample and the ablated model. Most features, however, were too small to be resolved optically, so microscopy was primarily used to select samples and to locate areas of interest for other analyses.

X-ray Diffraction

X-ray diffraction was used to determine the mineralogy of the stratospheric particles and the artificial ablation debris. The stratospheric particles range in size from 6 to 30 μm and the artificial debris analyzed ranged in size from 25 to 200 μm. Exposure times to Cr Kα radiation in a 57.3 mm diameter Debye-Scherrer diffraction camera ranged from 4 to 21 days, for the stratospheric particles, and from 4 hours to 5 days, for the artificial debris. Kodak, No-Screen medical X-ray film, was used. The cameras were purged with helium to reduce the background level of the film caused by air scatter.

All particles were mounted on thin quartz glass rods with Apiezon H grease. Numerous exposures were made using clean glass rods coated with Apiezon H grease. Exposure times
ranged from 4 to 5 days. The glass rods commonly produced a diffuse X-ray diffraction pattern in a zone at about 4.5 Å. Minute crystallites in the Apiezon H grease also occasionally produced diffraction lines. These lines correspond to the unidentified lines reported by Davis (1976). Crystallite-free Apiezon H grease was used and rarely caused contamination in the X-ray diffraction patterns. When contamination did occur it was eliminated by washing and remounting the particle.

**Bulk Analysis**

Bulk analyses using X-ray fluorescence were performed on original meteorite samples which did not contain fusion crust and on artificial debris, (e.g. spherules) which had been molten. These analyses were performed under contract by Gary G. Cunningham, Geology Department, University of Oregon. The samples were prepared by fusion with a flux of lithium borate. Lanthanum oxide was added to the flux as a heavy absorber to decrease matrix effects. There were two drawbacks to this technique. First, the fusion process tended to vaporize some volatile components, notably S, Na, and H₂O. Second, there was an interfering peak in the flux which made it impossible to analyze quantitatively for S. All that could be said for this element was that it was either present in appreciable amounts (a few percent) or it was not detectable.

**Electron Probe Microanalysis**

Several hundred artificial ablation debris particles were mounted on sapphire (Al₂O₃) substrates for elemental analysis
in a Materials Analysis Corporation, Model 400, electron microprobe. Analyses were performed using a KEVEX, Model 5500, energy dispersive X-ray detector having a 152 eV resolution. Data were treated qualitatively and used to characterize typical elemental abundances of the artificial ablation debris. This procedure was useful in screening out possible contaminants. It should be noted that this method can only detect Na at concentrations greater than several weight percent and cannot detect any elements with atomic numbers lower than Na (most importantly, O). Although this procedure is qualitative (or at best semiquantitative) it allows a rapid analysis of numerous particles, and relative weight percents for major and minor elements can be estimated.

An attempt was made to select the particles so that common types of debris could be identified. A glass rod, coated with Apiezon H grease, was usually rolled in collected debris of a specific size range (usually <100 μm) until coated with particles. The grease was dissolved from the particles with toluene and they were recovered on a Millipore filter. Then every particle was mounted for electron microprobe analysis. Usually spheres and angular debris were mounted on separate substrates. Some of these samples were later used for X-ray diffraction and scanning electron microscope analyses.

**Scanning Electron Microscopy**

Scanning electron microscope (SEM) analyses were essential to the project. All analyses were performed with a Joelco
JSM-3 SEM, equipped with a KEVEX energy dispersive X-ray detector. Textures in the fusion crusts and the artificial ablation debris are typically so fine-grained that they can only be resolved at very high magnifications. Elemental abundances of the fine-grained mineral intergrowths could only be analyzed with the 70 Å diameter beam of the SEM. SEM analyses were performed on: individual spherules, angular debris, polished sections of spherules, and polished sections of the original and artificial fusion crusts.

Sample Selection

In sampling the artificial ablation debris, every possible precaution was taken to exclude contaminants from the reported analyses. Particles were considered contaminants unless their elemental abundances closely matched the original meteorite or unless they could otherwise be demonstrated to have been created by the artificial ablation experiment.
RESULTS

**Murchison Fusion Crusts**

The original and artificial fusion crusts are quite similar when viewed in reflected light. Both are characterized by two zones (Fig. 6a, 6b, 7a, 7b) which suggests the amount of heating experienced. The outer zone (zone 1) is glassy and is typically discontinuous along the fusion crust. This zone was originally molten material that solidified after ablation and formed glassy, lobate protuberances, often connected by a thinner glassy layer. The inner zone (zone 2) is characterized by a high degree of alteration due to heating. Melting, however, apparently has not occurred to any great extent and some silicate grains (e.g. olivine) are sometimes unaltered. This zone is commonly transected by cracks and elongate holes, perpendicular to the surface of the crust, along which vaporizing volatiles probably have escaped. In both the natural and artificial crusts, molten troilite has invaded smaller cracks and spaces in zone 2.

There are some differences between the two fusion crusts. The original fusion crust typically is a little more than 400 μm thick and the artificial crust is slightly thinner, ranging from about 200 μm at the center of the ablated face to more than 300 μm thick near the sides. In the original fusion crust zone 1 ranges from 0 to 125 μm thick and zone 2 is about 300 μm thick. In the artificial crust, zone 1 is 0 to 80 μm
Figure 6a. Photomicrograph of a polished section of the original fusion crust of the Murchison meteorite. (1) Zone 1 is glassy and has lobate protuberances connected by a thinner glassy layer. (2) Zone 2 is composed of thermally altered, unmelted silicates. The original meteorite (3) remains unaltered.
Figure 6b. Photomicrograph of the original Murchison fusion crust showing (1) troilite (light colored) invading cracks in zone 2 and (2) magnetite (light colored) in zone 1.
Figure 7a. Photomicrograph of a portion of a polished section of the artificial fusion crust on the Murchison. There is a glassy zone 1 (1), a thermally altered zone 2 (2), and the unaltered original meteorite (3).
Figure 7b. Photomicrograph of the artificial fusion crust showing (1) troilite (light colored) intrusion in zone 2. Note the absence of magnetite in zone 1 (2).
thick and zone 2 is 150 to 250 µm thick. In the original fusion crust, zone 1 contains numerous micrometer-sized, equant grains of magnetite. However, this mineral is notably absent in zone 1 of the artificial crust. In transmitted light, zone 1 of the original crust is opaque, but this zone of the artificial crust is not. In the latter case, minute, birefringent crystallites can be distinguished. Also, invasion of cracks by troilite is more developed in zone 2 of the artificial crust than in the original.

SEM analyses of the fusion crusts were concentrated on zone 1 because this is where spherules would be produced. High magnifications of zone 1 in the original fusion crust showed that there are actually 3 mineral components. The most obvious of these is the equant micrometer sized grains of magnetite. More difficult to see, but present throughout much of zone 1, are two silicate phases here interpreted to be olivine and glass (Fig. 8). One of these is an Fe, Mg silicate with subhedral to euhedral grains up to 10 µm in size. These have an olivine composition estimated at approximately Fo70 at the center, but they are zoned with a higher Fe content at their rims. These crystals are in a matrix thought to be a silicate glass, usually less than 2 µm in width, which is, therefore quite difficult to analyze. Most attempts at analysis on this matrix included partial analysis of adjacent crystallites. Typical composition for the matrix appears to be an Fe-rich silicate with Ca and Al, and almost no Mg. The magnetite grains were observed to be exclusively surrounded by the matrix.
Figure 8. A SEM image of a portion of zone 1 of the original fusion crust, showing the olivine-glass-magnetite association. The dark grains with bright rims are zoned olivine and the smaller white grains are magnetite. They are in a glass matrix. The large, dark grain on the left is part of a large forsterite grain, bordering the two zones of the fusion crust.
Figure 9. SEM image of zone 1 of the artificial fusion crust shows zoned olivine crystals in a glass matrix.
material. Although magnetite is absent in the artificial fusion crust (Fig. 9), the two silicate phases are present and were easier to recognize than in the original meteorite. They are more developed in the artificial crust, especially in flanges along the side of the experimental model. SEM analyses were also performed on a polished section of Allende (C3) fusion crust and the same three components were observed in zone 1 of this meteorite.

Ablation Debris

Ablation debris has been divided into two groups based on morphology. Spheroidal particles which solidified from molten droplets were classified as spherules. Angular particles which were never completely melted were classified as angular debris.

Spherules. The spherules range from 1 mm to less than 1 μm in diameter. The spherules are typically gray-black with a slightly shiny luster. Crystallites are too fine grained to be resolved in a stereomicroscope. Hundreds of spherules were analyzed with the electron microprobe. Since the electron beam penetrated only a small part of the total spherule volume, these analyses must be considered as qualitative only.

Two types of spherules were observed (Table 2), evidently reflecting the presence of two compositionally different liquid phases. One "melt" phase is characterized by a chemical composition similar to the original meteorite (Table 1) with the exception that sulfur is present only in trace amounts, or is
Table 2

Qualitative elemental abundances and mineralogy of the artificial ablation debris. The white sections of the histogram bars represent the approximate amount of variation of an element.

<table>
<thead>
<tr>
<th>TYPES OF ABLATION DEBRIS</th>
<th>ELEMENTAL ABUNDANCES</th>
<th>MINERALOGY</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Mg  Al  Si  S  Ca  Cr  Fe  Ni</td>
<td></td>
</tr>
</tbody>
</table>

**SPHERULES**

- **Silicate Phase**
  - Olivine with accessory magnetite

- **Sulfide Phase**
  - Magnetite and/or wustite and pentlandite and/or pyrrhotite

**ANGULAR DEBRIS**

- **Fine-Grained**
  - Olivine and magnetite with some enstatite and pyrrhotite (rare)

- **Mafic silicate**
  - Olivine and enstatite
absent. This phase is high in Mg, Si, and Fe, low in Ca, and Al, and contains traces of Cr, and Ni. The other melt phase, a sulfide-bearing phase, is characterized by high amounts of Fe with variable amounts of S and Ni. Nearly all of the spherules analyzed were composed of the silicate phase, but at least 20 percent contained inclusions of the sulfide "melt" phase. These inclusions are typically spheroidal but also occur as irregular patches and are up to 50 μm in diameter. Only a few percent of the spherules are composed entirely of the sulfide "melt" phase.

The spherules are typically composed of two mineral associations (Table 2). The most common mineral association is characterized by the presence of olivine (approximately Fo70) with, or without, magnetite. The olivine lines are somewhat spotty but those of the magnetite are smooth, even without sample rotation. A mixture of very fine-grained magnetite with coarser olivine grains is required to produce lines of this type. Unfortunately, at this olivine composition one of the strongest olivine lines (2.533 Å) overlaps with the strongest magnetite line (2.532 Å). Therefore, although diagnostic magnetite lines were not always observed, it is believed to be a common accessory mineral. A typical diffraction X-ray pattern for this mineral association is listed in Table 3.

The other mineral association is characterized by the presence of Fe-oxides and sulfides. Minerals identified are magnetite, wustite, pyrrhotite, and pentlandite. In some spherules all four of these phases (Table 4) were identified,
Table 3

Typical X-ray diffraction pattern of a silicate spherule showing the olivine-magnetite mineral association.

<table>
<thead>
<tr>
<th>SILICATE SPHERULE</th>
<th>OLIVINE*</th>
<th>MAGNETITE</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>2(Mg_{0.64}Fe_{0.36})SiO_4</td>
<td>Fe_3O_4</td>
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<td>ASTM# 7-159</td>
<td>ASTM# 19-629</td>
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<th>I/I_o</th>
<th>d(Å)</th>
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<tr>
<td>15</td>
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<tr>
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<td>3.920</td>
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<td>3.749</td>
<td>10</td>
<td>3.744</td>
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<tr>
<td>60</td>
<td>3.519</td>
<td>30</td>
<td>3.516</td>
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</tr>
<tr>
<td>15</td>
<td>3.023</td>
<td>10</td>
<td>3.030</td>
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<tr>
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<td>2.967</td>
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<td>80</td>
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<td>100</td>
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<tr>
<td>60</td>
<td>1.763</td>
<td>50</td>
<td>1.761</td>
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*plus other olivine lines

| *Weak olivine lines have been omitted for clarity. |
Table 4

Comparison of the X-ray diffraction pattern of an artificial sulfide spherule with the patterns for pentlandite, pyrrhotite, wustite, and magnetite. Unit cell dimensions of pentlandite and wustite in the spherule are calculated to be 10.136±0.019Å and 4.27±0.005Å, respectively. The match for magnetite is not good and the spherule may contain a ferrous trevorite.

<table>
<thead>
<tr>
<th>SULFIDE SPHERULE</th>
<th>PENTLANDITE</th>
<th>PYRRHOTITE</th>
<th>WUSTITE</th>
<th>MAGNETITE</th>
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</thead>
<tbody>
<tr>
<td>(Fe, Ni)₈S₉</td>
<td>Fe₈S₈</td>
<td>Fe₁₋ₓS₀</td>
<td>Fe₃O₄</td>
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<td>#22-1120</td>
<td>#6-615</td>
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<table>
<thead>
<tr>
<th>I/Iₒ d(Å)</th>
<th>I/Iₒ d(Å)</th>
<th>I/Iₒ D(Å)</th>
<th>I/Iₒ d(Å)</th>
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<tbody>
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<td>10 3.582  5 3.55</td>
<td>100 3.056  80 3.03</td>
<td>20 2.985</td>
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</tr>
<tr>
<td>30 3.056  80 3.03</td>
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<td>100 2.532</td>
</tr>
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<td>100 2.532</td>
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<tr>
<td>70 2.058</td>
<td>100 2.064</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>70 1.950  50 1.931</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>100 1.791 100 1.775</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>10 1.723</td>
<td>40 1.720</td>
<td></td>
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</tr>
<tr>
<td>20 1.509</td>
<td>60 1.523</td>
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<td></td>
</tr>
<tr>
<td>70 1.322  20 1.307</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>20 1.289</td>
<td>25 1.299</td>
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</table>
although, in some cases any one sulfide and/or oxide was minor or absent. Relative intensities of the X-ray diffraction lines for these four minerals were observed to be quite variable. Pentlandite cell dimensions are large and indicate an Fe/Ni atomic ratio greater than 2.

The olivine-magnetite assemblage was produced by the silicate "melt" phase. Both assemblages were observed in only one diffraction pattern, suggesting that the silicate spherules containing inclusions of the sulfide "melt" phase produce patterns for only olivine and magnetite. The sulfide inclusions are common but are usually too small to be identified by X-ray diffraction.

Surface textures observed in the SEM were found to be quite variable and there is no particular texture which could be described as typical. A few of the more common textures are illustrated in Figures 10, 11, and 12. Only one interior texture was observed in SEM images of polished silicate spherules. This texture is similar to that found in zone 1 of the fusion crust. The three phases, magnetite, olivine, and a Ca, Al, Fe-silicate matrix were observed (Fig. 13). Since X-ray diffraction revealed only olivine and magnetite, this third phase is believed to be glass. In addition, inclusions of the sulfide "melt" phase were commonly observed in polished sections.

Angular Debris. Two types of angular debris have been analyzed. The first is typically black, fine grained, and fairly dense and non-porous. The larger size fraction of this
Figure 10. SEM image of a silicate spherule with pyramid-shaped crystals (olivine?) on the surface.
Figure 11. SEM image of a silicate spherule with a complex, fine-grained texture.
Figure 12. A silicate spherule with a smooth texture and a sulfide melt phase inclusion (center of spherule).
Figure 13. SEM image of a portion of a polished section of an artificial silicate spherule showing the olivine-magnetite-glass texture. Subhedral to euhedral crystals of olivine are in a glass matrix that contains small crystallites which are believed to be magnetite. A small inclusion of the sulfide melt phase is visible in the lower left corner of the photograph.
debris commonly exhibited a thin layer of fusion crust on one or more sides. In the smaller size fractions (<150 μm) the debris frequently exhibited no partial melting. Some angular debris is quite large (up to 5 mm), indicating that the model had fragmented well back from the fusion crust. These particles are similar to the original meteorite in elemental abundances, except for sulfur which was present in moderate to trace amounts (Table 2). Their elemental abundances and fine-grained textures indicate that they were originally part of the fine-grained matrix of the meteorite. The other type of angular debris consists of single angular grains of mafic silicates high in Si and Mg and usually low in Fe. These single grains are transparent with a reddish tinge and have either fractured, angular edges, or rounded edges which suggest partial melting. The mafic silicate grains produce very spotty X-ray patterns, but olivine and enstatite have been identified. The fine-grained angular debris typically contains Fe-poor olivine with magnetite. Enstatite is also common in these particles and pyrrhotite is rare. SEM micrographs of the two types of angular debris particles are shown in Figures 14, 15, and 16.

Bulk Analyses

Data for bulk analyses of both the original meteorite and the artificial spherules are listed in Table 5. The data represent an average of two analyses. The figure for "loss" on the original meteorite represents H₂O and S which were not deter-
Figure 14. SEM image of a fine-grained angular debris particle.
Figure 15. SEM image of a fine-grained angular debris particle which has a fusion crust on the left side.
Figure 16. SEM image of an angular grain of olivine produced by the artificial ablation.
Table 5

Bulk analyses (in weight percent) of the original meteorite and artificial ablation spherules (G. G. Cunningham, analyst).

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<thead>
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<th></th>
<th>ORIGINAL METEORITE</th>
<th>ABLATION SPHERULES</th>
</tr>
</thead>
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<td>RECALCULATED ANALYSIS</td>
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</tr>
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<td>2.71</td>
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<tr>
<td>MgO</td>
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<tr>
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<td>31.02</td>
<td>36.87</td>
</tr>
<tr>
<td>MnO</td>
<td>0.24</td>
<td>0.29</td>
</tr>
<tr>
<td>CaO</td>
<td>1.89</td>
<td>2.25</td>
</tr>
<tr>
<td>Na₂O</td>
<td>0.18</td>
<td>0.21</td>
</tr>
<tr>
<td>K₂O</td>
<td>0.08</td>
<td>0.09</td>
</tr>
<tr>
<td>P₂O₅</td>
<td>0.23</td>
<td>0.27</td>
</tr>
<tr>
<td>LOSS</td>
<td>13.42</td>
<td>0.00</td>
</tr>
<tr>
<td>TOTAL</td>
<td>97.57</td>
<td>100.00</td>
</tr>
</tbody>
</table>

Cr 0.380 0.458
Ni 0.754 0.446
S detected (a few percent) not detected (<1 percent)
mined in this analysis. Data are presented as corrected and recalculated analyses. The corrected analysis is derived from data after matrix corrections, and the analysis has been recalculated to 100 percent, and does not include the percent loss in the recalculated analysis. S was not treated quantitatively (see METHODS OF ANALYSIS). Ni and Cr were analyzed separately as trace elements.

**Stratospheric Particles**

Mineralogical analyses have been performed on 14 selected particles from the U-2 stratospheric collection. Diffraction data for two of these particles were provided for interpretation by D.E. Brownlee, and they are used in this report. Table 6 is a list of these particles and describes their mineralogy.

The particles are divided into two groups on the basis of the presence of a silicate phase. The silicates can be subdivided into 7 Å layer-lattice silicates and mafic silicates (olivine and pyroxene). The mafic silicate-bearing particles may also contain pyrrhotite and/or magnetite. The nonsilicate particles are subdivided into a sulfide-bearing group and a metal-bearing group. The sulfide-bearing particles contain a sulfide (pyrrhotite and/or pentlandite) and an oxide (magnetite and/or wustite). The metal bearing particles contain taenite associated with magnetite and wustite.
Table 6

Results of X-ray diffraction analyses on 14 stratospheric particles. Minerals are listed in an estimated order of decreasing abundance. The minerals listed in parentheses are identified from very weak patterns or from single lines.

<table>
<thead>
<tr>
<th>TYPE</th>
<th>NUMBER</th>
<th>SIZE (µm)</th>
<th>MINERALOGY</th>
</tr>
</thead>
<tbody>
<tr>
<td>SILICATE</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>XP 3</td>
<td>16</td>
<td>7Å layer-lattice silicate</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>(Serpentine?)</td>
</tr>
<tr>
<td></td>
<td>XP25</td>
<td>14</td>
<td>olivine</td>
</tr>
<tr>
<td></td>
<td>XP11</td>
<td>20</td>
<td>olivine (Fa &gt; 40), magnetite</td>
</tr>
<tr>
<td></td>
<td>XP20</td>
<td>25</td>
<td>clinoenstatite, magnetite</td>
</tr>
<tr>
<td></td>
<td>XP10</td>
<td>35</td>
<td>olivine, enstatite, pyrrhotite</td>
</tr>
<tr>
<td></td>
<td>XP 4</td>
<td>30</td>
<td>olivine, pyrrhotite, magnetite</td>
</tr>
<tr>
<td></td>
<td>XP19</td>
<td>14</td>
<td>olivine, pyrrhotite (weak pattern)</td>
</tr>
<tr>
<td>NONSILICATE</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sulfide</td>
<td>XP22</td>
<td>9</td>
<td>pyrrhotite, magnetite</td>
</tr>
<tr>
<td>Bearing</td>
<td>XP26</td>
<td>8</td>
<td>pyrrhotite, magnetite</td>
</tr>
<tr>
<td></td>
<td>XP27</td>
<td>10</td>
<td>pentlandite (Fe-rich), magnetite</td>
</tr>
<tr>
<td></td>
<td>XP28</td>
<td>5</td>
<td>pyrrhotite, magnetite, (wustite?)</td>
</tr>
<tr>
<td></td>
<td>XP21</td>
<td>14</td>
<td>magentite, pyrrhotite, wustite, (pentlandite)</td>
</tr>
<tr>
<td>Metal</td>
<td>XP14</td>
<td>10</td>
<td>taenite, wustite, magnetite</td>
</tr>
<tr>
<td>Bearing</td>
<td>XP29</td>
<td>7</td>
<td>taenite, magnetite, (wustite)</td>
</tr>
</tbody>
</table>
Principle Differences Between the Original and Artificial Crusts

The principle differences between the original and artificial fusion crusts from the Murchison meteorite are the following: 1) The artificial fusion crust is somewhat thinner than the original. This is true for both zones of fusion crust. 2) Magnetite is absent in zone 1 of the artificial crust, but is an important constituent of zone 1 of the original crust. 3) Olivine crystals are larger and better developed in zone 1 of the artificial crust. 4) Intrusion of troilite into cracks in zone 2 is more pronounced in the artificial crust. 5) Zone 1 of the artificial crust is not opaque as it is in the original crust. This is the result of the absence of magnetite and the presence of larger olivine crystals in the artificial fusion crust.

Experimental conditions of the artificial ablation are responsible for these differences. Meteor ablation in the atmosphere is a dynamic process in which conditions (e.g. atmospheric pressure, velocity) are continually changing. The artificial ablation, on the other hand, used a set of conditions which varied little during the experiment. Also, as noted previously (see Artificial Ablation of the Murchison Meteorite), gas cap pressures (and thus P_{O_2}) were lower than in the real environment. One other factor is the
original fusion crust formed at the end of the ablation of this meteorite. Higher pressures and temperatures (at low altitudes) which formed the original fusion crust are probably responsible for the formation of magnetite in zone 1 and the thicker crust. Differences in cooling rates may have affected olivine crystal growth (zone 1) and troilite intrusion (zone 2). These differences are not great and it seems reasonable to assume that debris collected in the experiment should resemble debris created by the actual ablation of the Murchison meteorite.

**Origin of the Artificial Ablation Mineral Assemblages**

Four distinct mineral assemblages have been identified in the artificial ablation debris: 1) olivine-magnetite-glass, 2) iron oxide (magnetite and/or wustite)-sulfide (pyrrhotite and/or pentlandite), 3) olivine-magnetite with accessories enstatite and (rare) pyrrhotite, and 4) unmelted olivine and pyroxene grains. These different mineral assemblages represent the effects of heating and fragmentation experienced by the ablation model. The olivine-magnetite-glass assemblage demonstrates a genetic relationship between the silicate spherules and zone 1 of the fusion crust. The olivine-magnetite assemblage with accessories enstatite and (rare) pyrrhotite of the fine-grained angular debris originated in zone 2 of the fusion crust. Unmelted grains of mafic silicates represent individual particles separated from the ablation model by fragmentation without having been extremely altered by heating. The relationship of the iron oxide-
sulfide assemblage to the ablation model is not so clear, but one would expect it to have originated in zone 2 of the fusion crust. The sulfides are not likely to survive the high temperatures of zone 1.

The olivine-magnetite-glass assemblage may be typical of fusion crusts of the chondrites and the silicate spherules they produce. In zone 1, complete melting occurs and silicate droplets will fly off. As this liquid cools, olivine crystallizes rapidly in a disequilibrium environment, with increasing Fe content toward the crystal rims as the spherule cools and Mg is used up. There can be little substitution of Mg by cations other than Fe (Deer et al., 1963). Ca, Al and the remaining Fe are forced into the residual silicate melt. In a more oxidizing environment, much of this Fe would form magnetite. In this study, the olivine-magnetite-glass assemblage was found in both Murchison (C2) and Allende (C3). It has also been described by Brownlee et al. (1975) in Orgueil (C1), and by Ramdohr (1967) in Warrenton (C3). A similar assemblage has been described in zone 1 of Kunashak, an L type ordinary chondrite (Yudin, 1955) in which magnetite, wustite, enstatite, olivine, and glass have been observed. Future studies may document similar assemblages in other chondrites.

The olivine-magnetite-enstatite-pyrrhotite (rare) assemblage of the fine-grained angular debris originated in zone 2 of the fusion crust. The pyrrhotite is not common, evidently because the sulfide-rich "melt" segregated in zone 2 (Fig. 7). When the fine-grained particles ablated off the
model, they must have separated from the sulfide veins. The olivine and magnetite formed from the original chamosite matrix by alteration due to heating. Fuchs et al. (1973) heated matrix material of Murchison up to 450°C and formed olivine and magnetite from chamosite. In specimens they heated in which the poorly characterized Fe-S-O phase was present, they also formed FeS, and this process is probably the source of the sulfide-rich veins.

The unmelted mafic silicate grains and the fine-grained angular debris demonstrate that fragmentation of the ablation model allows individual fragments of matrix and inclusions from the meteoroid to escape the ablating model. These grains fragment without being completely melted. In the artificial model, fragments need only to escape the arc jet plasma to avoid melting and vaporization. However, in the real environment, this may not be possible because fragments that become free of the meteoroid must still undergo deceleration and will be further heated and ablated. This experiment has demonstrated that unmelted particles can survive meteor ablation in the laboratory environment. If this is possible in a natural environment it is most likely to occur at very high altitudes at the onset of ablation.

The iron oxide (magnetite and/or wustite)-sulfide (pyrrhotite and/or pentlandite) assemblage is the most difficult to explain. It is definitely a product of the experiment. It is commonly found as inclusions within the artificial silicate spherules. The sulfide "melt" phase must
be related to the sulfide veins in zone 2 of the fusion crust in the experimental model. It is necessary to envision a process which will allow this assemblage to form. Since the sulfides are not found in zone 1 of the fusion crust, they could not have survived the high temperatures which formed the silicate "melt". Rapid melting and cooling must be essential to the formation of this assemblage. This process must also include sulfide exposure in zone 2 of the fusion crust and the formation of angular debris which requires fragmentation. It is possible that when the experimental model fragmented, exposing zone 2 of the fusion crust, droplets were produced which sprayed out of the sulfide veins to produce the sulfur rich spherules. Silicate spherules with sulfide inclusions could have formed in two ways: 1) Combinations of silicates and sulfides from zone 2 may have fragmented away, melted rapidly, and cooled rapidly to produce this mixture. 2) Liquid droplets of both the silicate and sulfide "melt" phases may have combined in flight after leaving the model.

**Interpretation of the Bulk Analyses**

Bulk analyses of the original meteorite and the spherules can be used to interpret general trends in the ablation process. The bulk analysis values for the original meteorite in this study agree with the published values (Table 1) of Fuchs et al. (1973) and Jarosewich (cited in Fuchs et al., 1973). There is no major change in the relative proportions
of Mg, Al, Ca, and Fe relative to Si in the spherules when compared to the original meteorite. However, there is a definite trend of depletion for the volatiles: H$_2$O, S, and Na. There are some discrepancies, such as enrichment of Cr and depletion of Ni in the spherules. If these relationships were to hold true in the natural environment, then one would expect that spherules produced by the chondrites would have the same non-volatile composition as their parent meteoroids.

**Mineralogy of the Stratospheric Particles and the Artificial Ablation Debris**

It is possible to correlate the mineralogical assemblages found in the artificial ablation debris with similar assemblages in the stratospheric particles. The mineral groups found in the artificial debris all occur in the stratospheric particles. While no layer-lattice silicates were observed in the artificial debris, chamosite does occur in the original Murchison matrix (Fuchs et al., 1973). These hydrated silicates are not expected to survive the heating in zone 2 of the fusion crust. The absence of the taenite-magnetite-wustite assemblage in the artificial debris suggests that ablation of a non-metal-bearing carbonaceous chondrite does not produce this assemblage of minerals.
CONCLUSIONS

Similarities of the Artificial Debris and the Stratospheric Particles

Correlations between the mineral assemblages of the stratospheric particles and the artificial ablation debris match very well with particle groups classified by Brownlee et al. (1976) using textural features and elemental abundances. Brownlee et al., have described four groups of particles classified as chondritic, mafic silicate, FSN, and Fe-Ni. Two types of chondritic particles comprise over 60 percent of the 250 stratospheric particles collected, and they closely approximate the elemental abundances of the chondrites for the elements Mg, Al, Si, Ca, and Fe. Ninety percent of the chondritic particles, termed chondritic aggregates, are very fine-grained aggregates, high in S. Six have been analyzed for C, yielding values from 2.2 to 15 weight percent. The mineralogy described for these particles includes a layer-lattice silicate, olivine, pyrrhotite, and magnetite. The other 10 percent of the chondritic particles lack S and are typically spherules or rounded with "melt" textures. Because of the lack of volatiles and the "melt" textures these are termed chondritic ablation particles. The mafic silicates are single crystals or aggregates of crystals commonly with adhering chondritic materials. These have been identified as olivine and pyroxene with some magnetite and pyrrhotite.
The FSN particles are composed of Fe, S, and Ni. Some of these are angular and form platelets or subhedral and euhedral crystals, commonly associated with chondritic aggregate material. However, the majority of these particles are spherules. Both are typified by the presence of iron oxides and sulfides. The presence of wustite in some FSN spherules indicates an ablation origin (Davis, 1976). FSN particles comprise thirty percent of the collection but are typically very small (<10 μm) and represent only a few percent of the total mass of the collection. The fourth group, classified as Fe-Ni is represented by only 7 particles which are typically spheroidal. Taenite, magnetite, and wustite have been identified in this group.

The four groups of artificial ablation debris produced in this study have textural, elemental, and mineralogical similarities to the members of Brownlee et al's., three largest groups. (Table 7). The stratospheric chondritic aggregates are very similar to the artificial fine-grained angular debris. Although a layer-lattice silicate has not been identified in the artificial debris, these particles have similar mineralogies. In addition, they have similar elemental abundances and fine-grained textures. However, most stratospheric chondritic aggregates are much more porous and more fine-grained than the artificial debris. The stratospheric chondritic ablation particles correlate very well with the artificial silicate spherules. Both contain olivine and magnetite, have "melt"
Table 7

Comparison between the stratospheric particles and the artificial ablation debris. The classification used for the stratospheric particles is that of Brownlee et al. (1976). The particle numbers listed for the stratospheric particles are those used in Table 6.

<table>
<thead>
<tr>
<th>STRATOSPHERIC PARTICLES</th>
<th>ARTIFICIAL ABLATION DEBRIS</th>
</tr>
</thead>
<tbody>
<tr>
<td>GROUP NUMBER</td>
<td>MINERALOGY</td>
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<tr>
<td>CHONDRICTIC</td>
<td></td>
</tr>
<tr>
<td>Aggregates XP 3</td>
<td>7 A layer-layer lattice silicate</td>
</tr>
<tr>
<td>XP 4</td>
<td>olivine, pyrrhotite (magnetite)</td>
</tr>
<tr>
<td>XP19</td>
<td></td>
</tr>
<tr>
<td>Ablation XP11</td>
<td>olivine, (magnetite)</td>
</tr>
<tr>
<td>XP10</td>
<td>magnesiochromite, pyroxene</td>
</tr>
<tr>
<td>MAFIC SILICATES</td>
<td></td>
</tr>
<tr>
<td>XP22</td>
<td>pyrrhotite, magnetite</td>
</tr>
<tr>
<td>XP21</td>
<td>magnetite and/or wustite</td>
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<tr>
<td>XP27</td>
<td>pyrrhotite and/or magnetite</td>
</tr>
<tr>
<td>XP28</td>
<td>pentlandite</td>
</tr>
<tr>
<td>Fe-Ni XP14</td>
<td>taenite, magnetite.</td>
</tr>
<tr>
<td>XP29</td>
<td>wustite</td>
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</table>
textures, and have similar elemental abundances. Prior to this investigation, D. E. Brownlee (personal communication), discovered an inclusion in a stratospheric chondritic ablation spherule which was rich in Fe and Ni, with some S. This is similar to the sulfide inclusions observed in the artificial silicate spherules. Equant, micrometer-sized magnetite crystals were also observed, but glass and olivine were not. X-ray diffraction was not used to identify the mineralogy. The stratospheric mafic silicates resemble the angular olivine and pyroxene in the artificial debris. The stratospheric FSN particles have a similar mineralogy and composition to the sulfide "melt" spherules in the artificial debris. However, the angular FSN particles have no textural equivalent in the artificial debris and only the FSN spherules correlate in this respect.

The stratospheric Fe-Ni particles do not correlate with any groups identified in the artificial debris. They are, however, similar in mineralogy and texture to spherules produced by Davis (1976) during Fe and Ni-Fe artificial ablation experiments and to those found in deep sea sediments (Schmidt and Keil, 1966; Millard and Finkelman, 1970) and manganese nodules (Finkelman 1970, 1972). The presence of wustite in these particles indicates an ablation origin from a metal-bearing meteorite (e.g. iron, ordinary chondrite).

**Ablation Debris in the Stratosphere**

Identification of ablation debris in the stratospheric particles is now confirmed for some groups. The obvious
Ablation debris in the stratospheric particles are classified as chondritic ablation, FSN (spherules), and Fe-Ni. The chondritic ablation particles are identified by their "melt" texture, the presence of olivine and magnetite, and in one case, by the spheroidal inclusion of an Fe, Ni, S phase and magnetite grains. The FSN and Fe-Ni spherules can be considered the products of ablation solely by the presence of wustite (Davis, 1976). The production of spherules in the artificial ablation experiment which are very similar to the FSN's supports an ablation origin. Table 8 lists the diffraction pattern for an FSN spherule, and comparison with the pattern for an artificial sulfide spherule of similar composition (Table 4) illustrates the similarity.

In other particle groups, the data remains ambiguous. These groups are the chondritic aggregates (which comprise half of the stratospheric collection), mafic silicates, and angular FSN's. Unablated interplanetary dust, which probably exists in the stratosphere, could contribute to any of these groups. On the other hand, if fragmentation produces very many unmelted grains, some particles of these groups could also be formed by ablation. McCroskey (1967) has cited evidence that many fireballs undergo gross fragmentation. Also, even at the low impact pressures of the artificial ablation, the quantity of debris produced by fragmentation of Murchison was comparable to that produced by melting effects. The mafic silicates, with their high melting points and density, are the most likely to survive after fragmentation of a meteoroid.
Table 8
Comparison between the X-ray diffraction pattern for stratospheric FSN particle #XP21 (10 day exposure) and the patterns for pyrrhotite, magnetite, wustite, and pentlandite. Despite the two missing lines (in parentheses) for magnetite and pentlandite this is believed to be a good match. The pyrrhotite values approach those for pure troilite.

<table>
<thead>
<tr>
<th>FSN SPHERULE</th>
<th>PYRRHOTITE</th>
<th>MAGNETITE</th>
<th>WUSTITE</th>
<th>PENTLANDITE</th>
</tr>
</thead>
<tbody>
<tr>
<td>ASTM#20-535</td>
<td>#19-629</td>
<td>#6-615</td>
<td>#8-90</td>
<td></td>
</tr>
<tr>
<td>I/Io d(Å)</td>
<td>I/Io d(Å)</td>
<td>I/Io d(Å)</td>
<td>I/Io d(Å)</td>
<td>I/Io d(Å)</td>
</tr>
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<td>8   4.85</td>
<td>80  3.03</td>
<td></td>
<td></td>
</tr>
<tr>
<td>5   3.058</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>50  2.993</td>
<td>40   2.98</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>20  2.902</td>
<td>(30  2.967)</td>
<td>40   2.90</td>
<td></td>
<td></td>
</tr>
<tr>
<td>50  2.656</td>
<td>60   2.65</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
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<td></td>
</tr>
<tr>
<td>10  2.475</td>
<td></td>
<td>80  2.486</td>
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</tr>
<tr>
<td>10  2.424</td>
<td>8    2.424</td>
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<td></td>
</tr>
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<td>10  2.141</td>
<td></td>
<td>100  2.153</td>
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</tr>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>100 2.081</td>
<td>100   2.07</td>
<td>(50  1.931)</td>
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<td></td>
</tr>
<tr>
<td>5   1.779</td>
<td></td>
<td>100  1.775</td>
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</tr>
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<td>20    1.72</td>
<td></td>
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<td></td>
</tr>
<tr>
<td>20  1.616</td>
<td>5     1.61</td>
<td>30     1.616</td>
<td></td>
<td></td>
</tr>
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<td>60     1.523</td>
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</tr>
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<td>40    1.485</td>
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<td></td>
</tr>
<tr>
<td>20  1.448</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>20  1.329</td>
<td>5     1.33</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>10  1.303</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Original page is of poor quality
or as individual dust-sized particles. The angular FSN grains are usually intimately associated with the chondritic aggregates so the two groups must have a common origin. The olivine-magnetite-pyrrhotite association in the chondritic aggregates is similar to that observed in the artificial fine-grained angular debris.

Criteria for recognition of ablation debris in the stratosphere which has been produced by primitive meteoroids can be summarized in the following way: 1) Melting at high temperatures and rapid cooling will result in melt features (spherules) and distinctive textures. 2) Two "melt" phases, silicate- and sulfide-rich, will be produced and can occur separately or together. 3) The silicate phase formed from a chondrite will be composed of olivine and magnetite in a Ca, Al, Fe, Si glass matrix. 4) The sulfide phase will form a combination of iron oxides (magnetite and/or wustite) and sulfides (pyrrhotite and/or pentlandite). 5) Non-volatile elemental abundances for the silicate spherules will approximate those of the meteoroid from which they formed. 6) More volatile elements will be depleted. 7) If angular debris produced by fragmentation survives without melting or vaporizing, mafic silicate grains and fine-grained aggregates will be produced from meteoroid inclusions and matrix, respectively. 8) Fine-grained aggregates which fragment from the layer-lattice silicate matrix of a primitive meteoroid can be altered to olivine and magnetite if sufficiently heated.
Unablated interplanetary dust must exist in the stratosphere. Kornblum (1969) has demonstrated that small particles (<50 μm) travelling at low velocities (<15 km/sec) experience little or no loss of mass due to vaporization during atmospheric entry if they have a low entry angle. The smaller a particle is, the more easily it is decelerated at very high altitudes and the less it is affected by heating. Such particles will be collected in the stratosphere by the U-2 and the problem arises in distinguishing them from meteor ablation debris.

The stratospheric particle groups which must contain unablated dust are those classified as chondritic aggregates, angular FSN's, and mafic silicates. Although the 7Å layer-lattice silicate in one chondritic aggregate has not been altered by heating, the olivine-pyrrhotite-magnetite assemblage evidently resulted from heating of a more primitive mineral composition. Only the C1 and C2 chondrites have textures as fine grained as the chondritic aggregates, but their matrix is typically composed of a layer-lattice silicate. No known meteorite type has a matrix this fine grained with an olivine-pyrrhotite-magnetite composition. Therefore, these particles must have been heated to temperatures of at least a few hundred degrees C, but not high enough to vaporize the sulfur. The angular FSN grains have not been affected by heating so the temperatures cannot have been too high. If these two groups can survive atmospheric entry, the mafic silicates, with higher
melting points and density would also be expected to survive as dust-sized particles.

These particle groups, therefore, must contain unablated dust, but there is presently no criterion for distinguishing these from angular ablation debris. The fact that FSN spherules can be formed by ablation and retain a high proportion of sulfur indicates that particles can survive without being excessively heated. In addition, ablation debris which fragments at extremely high altitudes when atmospheric entry begins would fragment into the same environment as that entered by interplanetary dust, and the fragments might decelerate without significant additional heating.

Source of the Stratospheric Particles

Collection of extraterrestrial particles in the stratosphere can provide much new information for meteorite scientists regarding the composition of interplanetary dust and the vast majority of the mass of meteoroids which can never be collected on the ground. Some groups of stratospheric particles can be identified as ablation debris produced by primitive meteoroids. Other stratospheric particle groups contain unablated interplanetary dust, but as yet these cannot be distinguished from angular debris produced by gross fragmentation of primitive meteoroids. Whether or not such debris can actually be produced is not certain and could be demonstrated by a collection from a fireball wake.

The most significant contribution of the analyses
comparing the stratospheric and artificial ablation articles is that the majority of the stratospheric particles originated from either primitive meteoroids, or from very primitive interplanetary dust. The few Fe-Ni particles must be derived from a metal-bearing meteoroid, but the ordinary chondrites can be a source for these particles. The chondritic, FSN, and mafic silicate stratospheric particle groups all resemble the artificial debris produced from the Murchison meteorite, and those particles which may be interplanetary dust have very primitive compositions and textures.
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