Chemical Fractionation in the Solar Nebula

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The sequence of condensation of minerals from a cooling gas of solar composition has been calculated from thermodynamic data over the pressure range 10^{-3} to 10^{-5} atm, assuming that complete chemical equilibrium is maintained. The results suggest that the Ca-Al-rich inclusions in Allende and other carbonaceous chondrites are aggregates of the highest temperature condensates. Complete condensation of these elements is followed, $100°$ later, by the onset of the crystallization of nickel-iron, forsterite and enstatite. Transport of Ca-Al-rich refractory condensates from one part of the nebula to another before the condensation of these lower-temperature phases may have been responsible for the refractory element fractionations between the different classes of chondrites and possibly for the inferred refractory element enrichment of the Moon.

The temperature gap between the condensation temperatures of nickel-iron and forsterite increases with increasing total pressure. Because pressure and temperature probably increased with decreasing heliocentric distance in the solar nebula, Mercury may have accreted from a condensate assemblage having a higher metal/silicate ratio than Venus or Earth which may, in turn, have formed from less oxidized material than Mars. Heterogeneous accumulation models in which the Earth accretes in a stratified fashion with refractory condensates in the interior and volatile materials in the surface layers have some geochemical advantages over homogeneous accretion models.

The availability of accurate thermodynamic data, high-quality data on the abundances of the elements in the solar system, and estimates of the initial physical conditions in the solar nebula allows the accurate calculation of the sequence of condensation of minerals during the cooling of the gas cloud. The purpose of this paper is to present the results of these calculations and to review their applicability to some important problems in meteoritics and planetology. For a more detailed review, the reader is referred to Grossman and Larimer (ref. 1).

Method of Calculation

Recent models for the evolution of the solar nebula (ref. 2) suggest that temperatures as high as 2000 K and total pressures between 10^{-3} and 10^{-5} atm prevailed in the inner part of the nebula. At equilibrium under these conditions all the major elements would have been completely in the vapor state. The relative abundances of the elements in the solar system have recently been reviewed by Cameron (ref. 3). It is probable that the abundances of most of the major elements are now known to within a factor of 2 and that trace element abundances are known to within a factor of 10. Because the pressure and composition of the solar nebular vapor are known, the distribution of the elements between gaseous molecules and crystalline phases can be computed as a function of temperature, using equilibrium thermodynamic models.

The general outline of the method used to calculate the sequence of condensation of elements and compounds from a cooling gas of solar composition is given below. The details of the calculations may be found in Grossman (ref. 4).

The system is assumed to contain only the 20 most abundant elements in the solar system, excluding the noble gases. For each element, a mass-balance equation containing terms for the concentrations of all gaseous species containing that element is written. If a chemical reaction is written in which each polyatomic gaseous molecule is formed from its monatomic gaseous constituent elements, its concentration term in the mass-balance equations can be replaced by the product of the equilibrium constant for the reaction and powers of concentrations of the monatomic gaseous component elements. The equation set, containing terms for 300 gaseous species, thus reduces to a system of 20 simultaneous, nonlinear equations in 20 variables—the concentrations of the 20 monatomic gaseous elements. This equation set can be solved easily by a method of successive approximations using a high-speed computer.

The reaction for the decomposition of a crystalline phase into its monatomic gaseous component elements can be used to obtain its condensation temperature. For each of over 100 solid phases of interest, the temperature variation of the equilibrium constant for this reaction is compared to the concentrations of the relevant gaseous species as calculated at different temperatures from the mass-balance equations.

At a given pressure, the equation set was solved first at the highest temperatures and then at successively lower temperatures, in intervals of 25 or 50°. At each temperature, the solutions were checked against the equilibrium constants for every crystalline phase in order to test for the condensation of solids. When such a phase was found to appear, its exact condensation temperature was calculated, an additional variable was added to the equation set below that temperature to account for its presence, and a new equation was added, imposing the condition that the

new crystalline phase be in complete chemical equilibrium with the vapor. The calculations were continued to lower and lower temperatures where more solid phases were seen to condense and react with the vapor to form new phases. The result of the calculations is a model that describes the progressive condensation of the cooling solar nebula at complete chemical' equilibrium. Calculated condensation temperatures would be in error by ± 20° due to estimated uncertainties in free energies of formation. The year-to-year fluctuations in elemental abundances lead to temperature uncertainties on the order of 10 to 20°.

Results

EARLY CONDENSATES

Figure 1 shows the calculated distribution of Al between crystalline phases and vapor at 10^{-3} atm total pressure. Corundum is the first condensate containing a significant fraction of any of the major elements. It condenses at 1742 K and consumes more than 95 percent of the Al before it begins to react with the vapor to form melilite at 1608 K.
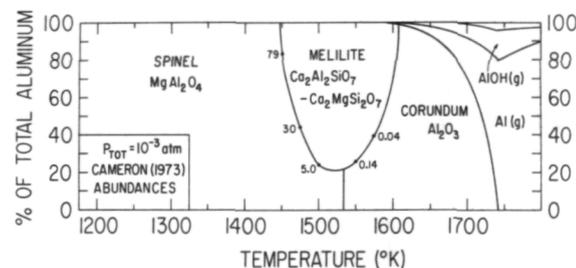

Figure 1.—*The calculated distribution of Al between crystalline phases and vapor in the solar nebula. Corundum condenses at 1742 K and begins to react with the vapor of 1608 K to form gehlenite-rich melilite. Excess corundum reacts to form spinel at 1533 K. Between 1500 and 1450 K, melilite reacts with the vapor to produce more spinel and increase its akermanite content (mole % indicated by numbers on the melilite curve). Melilite reacts completely with the vapor at 1442 K to form diopside and more spinel. Al is virtually completely condensed at 1600 K.*
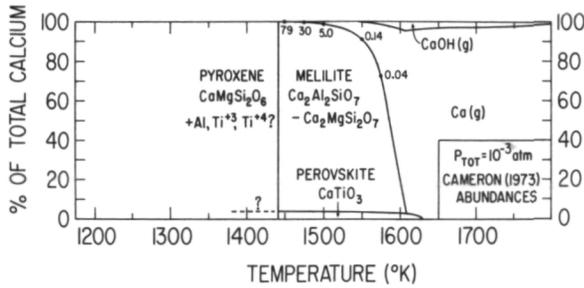
Figure 2.—*The calculated distribution of Ca between crystalline phases and vapor in the solar nebula. Perovskite condenses at 1632 K and melilite at 1608 K. Melilite reacts completely with the vapor to form spinel and diopside at 1422 K. Numbers on the melilite curve give the mole % akermanite in solid solution in the melilite. Ti is totally condensed at 1600 K and Ca is 90-percent condensed at 1550 K.*

The amount of melilite increases with decreasing temperature, forming at the expense of corundum. The remaining corundum reacts with gaseous Mg at 1533 K to form spinel. Melilite is a solid solution of gehlenite, $Ca_2Al_2SiO_7$, and akermanite, $Ca_2MgSi_2O_7$. The first melilite to form is virtually pure gehlenite. At 1500 K it still contains only 5 mole percent akermanite, but by 1450 K this concentration must reach 79 mole percent, according to ideal solid solution theory. Consequently, the melilite expels Al to form more coexisting spinel below 1500 K. The numbers on the melilite curve in figure 1 indicate the mole percent of akermanite in solid solution. At 1442 K, the melilite reacts completely with the vapor to form diopside and more spinel.

The distribution of Ca between vapor and solids at 10^{-3} atm is shown in figure 2. Perovskite is the first Ca-bearing phase to condense, appearing at 1632 K. Because the Ti/Ca atomic ratio is only 0.03, however, even when all the Ti is consumed by perovskite at 1600 K, only 3 percent of the Ca is condensed. The major sink for Ca at high temperature is melilite. More than 90 percent of the total Ca is condensed at 1550 K. At 1442 K, the total reaction of melilite to spinel and pyroxene is evident. The calculations refer to a pyroxene of diopside composition,

$CaMgSi_2O_6$, although there is some evidence from meteorites that this pyroxene might be rich in Al, Ti^{4+}, and Ti^{3+}. Thermodynamic data are lacking, however, for pyroxenes of these compositions.

The early condensates are thus dominated by minerals rich in Al and Ti, which are totally condensed at 1600 K, and Ca, which is 90-percent condensed at 1550 K.

CONDENSATION OF THE MOST ABUNDANT NONVOLATILE ELEMENTS

In distinct contrast to Ca, Al, and Ti, the most abundant metallic elements, Fe, Mg, and Si, are virtually uncondensed above 1550 K, and significant fractions of them begin to condense only below 1460 K (Fe) and 1430 K (Mg, Si). Figure 3 illustrates the distribution of Mg between crystalline phases and vapor at 10^{-3} atm. The condensation curve for metallic nickel-iron is also shown. The bulk of the Mg begins to condense at 1430 K as forsterite, which later reacts with the vapor to form enstatite at 1355 K. Iron begins to condense at 1460 K and is 46-percent condensed when forsterite appears. Figure 4 shows that significant condensation of Si also does not begin until forsterite formation.

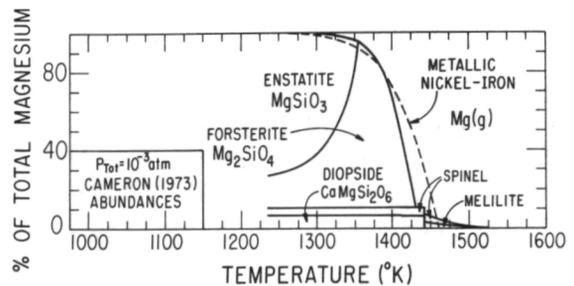This difference in the condensation behav-

Figure 3.—*The calculated distribution of Mg between crystalline phases and vapor in the solar nebula. Less than 10 percent of the total Mg is condensed above 1450 K. Forsterite appears at 1430 K and begins to react with the vapor at 1355 K to form enstatite. The condensation curve for iron as an iron-nickel alloy is shown for reference. Fe and Mg are more than 90-percent condensed at 1350 K.*
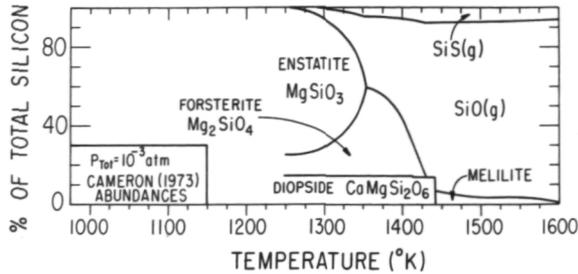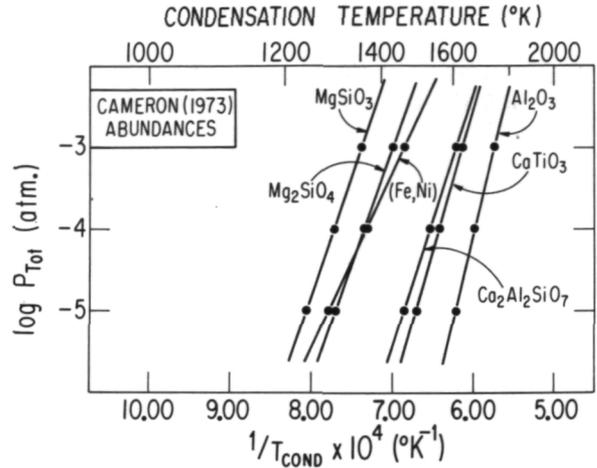
Figure 4.—*The calculated distribution of Si between crystalline phases and vapor in the solar nebula. Less than 10 percent of the total Si is condensed above 1450 K. Si is more than 90-percent condensed at 1300 K.*

Figure 5.—*The pressure variation of the condensation temperatures of some important minerals. All condensation temperatures increase with pressure. The condensation sequence is the same for all the lithophile elements from 10^{-3} to 10^{-5} atm. Nickel-iron condenses at higher temperatures than forsterite for total pressure greater than $\sim 5.5 \times 10^{-5}$ atm. The reverse is true at lower pressures.*

ior of Ca, Al, and Ti on one hand and Fe, Mg, and Si on the other may have led to chemical fractionations in the solar nebula. If physical conditions existed during the condensation of the nebula which allowed the efficient separation of dust from gas and the concentration of dust relative to the gas in some regions, then some bodies in the inner solar system may have accreted from materials enriched or depleted in Ca, Al, and Ti, relative to Mg, Fe, and Si, compared to the solar system abundances.

The first metal alloy to condense from a gas of solar composition at 10^{-3} atm should contain 15.1 mole percent Ni, 0.76 percent Co, 0.35 percent Cr, and 83.8 percent Fe. If the metal continues to equilibrate with the vapor as the temperature falls, its composition should be 6.0 percent Ni, 0.28 percent Co, 1.00 percent Cr, and 92.7 percent Fe at 1375 K.

The variation with pressure of the condensation temperatures of some of the phases discussed above is shown in figure 5. The sequence of condensation and reaction is the same from 10^{-3} to 10^{-5} atm for all the litho-

phile elements, with the entire sequence moving downward in temperature by about 70° for each factor of 10 decrease in total pressure. Metallic nickel-iron, however, has a lower condensation temperature than forsterite below $\sim 5.5 \times 10^{-5}$ atm and condenses above forsterite at higher pressure. The temperature gap between the condensation points of metal and forsterite increases with increasing pressure, suggesting that metal/silicate fractionation during condensation increased in importance toward the center of the nebula where the pressure was highest.

SOME LOW-TEMPERATURE CONDENSATION REACTIONS

As Larimer (ref. 5) pointed out, Na and K are relatively low-temperature condensates, appearing below 1250 K at 10^{-3} atm, perhaps in the form of alkali feldspar. The high-temperature magnesium silicates remain FeO-free until the temperature falls below 800 K, when the nebula becomes more oxidizing by

Table 1.—*Chemical Compositions of Allende White Inclusions Compared to Calculated Condensate Compositions at* $P_{tot} = 10^{-3}$ *atm.*

	Condensate 1475 K	Coarse-Grained Inclusion [1]	Condensate 1450 K	Fine-Grained Inclusion [2]	Condensate 1440 K
CaO	32.31	26.76	27.23	21.6	18.86
Al$_2$O$_3$	34.81	31.61	29.22	26.6	20.21
TiO$_2$	1.77	0.99	1.49	1.3	1.02
MgO	9.39	10.82	16.98	13.1	21.03
SiO$_2$	21.71	29.79	25.09	33.7	38.87
Total	99.99	99.97 [3]	100.01	96.3 [4]	99.99

NOTES: (1) Type a chondrule NMNH 3529 (ref. 8).
(2) Single aggregate NMNH 3510 (ref. 8).
(3) Also contains 0.37 percent FeO and 0.11 percent Na$_2$O.
(4) Also contains 0.1 percent Cr$_2$O$_3$, 2.3 percent FeO, and 1.1 percent Na$_2$O.

the reaction of CO with H$_2$ to produce CH$_4$ and H$_2$O. Sulphur condenses when gaseous H$_2$S reacts with excess metallic nickel-iron to form troilite at 700 K. Magnetite becomes stable at 405 K, and the hydration of ferro-magnesian silicates takes place at approximately 350 K.

Interpretation

CHONDRITES

Refractory Aggregates in the Carbonaceous Chondrites

Irregularly shaped, white aggregates are found in the C2 and C3 carbonaceous chondrites. They are enriched in Ca, Al, and Ti and depleted in Mg, Fe, and Si relative to the solar system abundances. This suggests that they may be related to the highest temperature condensates from the solar nebula (refs. 1, 4, 6, and 7). In table 1, the bulk chemical compositions of two of these inclusions from the Allende meteorite are compared to calculated equilibrium condensate compositions, excluding metallic Fe, in the temperature range 1475 to 1440 K at 10^{-3} atm total pressure. The quantitative similarities reinforce the suggestion that the inclusions may be samples of the early condensates. The small quantities of alkali metals and FeO may have entered the inclusions as a result of metamorphic reactions inside the parent body, long after condensation was over.

The common mineral assemblage in the Ca-rich inclusions consists of perovskite, spinel, and melilite, which usually contains about 25 mole percent akermanite (refs. 8 through 11). These are precisely the same phases which are predicted to be in equilibrium with the solar nebular gas in the temperature range 1500 to 1450 K at 10^{-3} atm. Sometimes these aggregates are surrounded by narrow rims of diopside which may signify the onset of the reaction of melilite with the vapor to form diopside and spinel at 1442 K. Other types of Ca-rich inclusions are found which contain coarse crystals of pyroxene rich in Al$_2$O$_3$, TiO$_2$, and Ti$_2$O$_3$. This mineral might be a condensate or, alternatively, it may be a crystallization product of a liquid formed when a condensate aggregate was later melted.

Although they have very low partial pressures in a gas of solar composition, some trace elements form such stable, refractory compounds that they can condense in the same temperature range as melilite, spinel, and perovskite (ref. 12). Table 2 shows the approximate condensation temperatures of a

Table 2.—*Condensation Temperatures of Refractory Trace Elements Compared to Those of the Major High-Temperature Minerals*

Crystalline Phase	Condensation Temperature (°K)		Enrichment Factor[1] in Allende Inclusions
	10^{-3} atm	10^{-4} atm	
Os	1925	1840	15
W	1885	1798	13
ZrO_2	1840	1789	10
Re	1839	1759	21
Corundum (Al_2O_3)	1742	1671	
HfO_2	1719	1652	10
Y_2O_3	1719	1646	21
Sc_2O_3	1715	1644	22.9 [2]
Mo	1684	1603	10
Perovskite ($CaTiO_3$)	1632	1557	
RE_2[3]O_3 (in solution)	1632	1557	22.5 [3,4]
Ir	1629	1555	24.1 [2]
Ru	1614	1541	12
Gehlenite ($Ca_2Al_2SiO_7$)	1608	1528	
V_2O_3	1534	1458	
Spinel ($MgAl_2O_4$)	1533	1451	
Ta_2O_5	1499	1452	18
ThO_2	1496	1429	
Diopside ($CaMgSi_2O_6$)	1442	1375	

NOTES: (1) Enrichment factor = concentration in Allende inclusion/concentration in Type I carbonaceous chondrites.

(2) RE = rare earth element. The rare earths can condense in solid solution in perovskite.

(3) Data from Grossman (ref. 12) who analyzed 16 inclusions. All other enrichment factors are based on one Allende inclusion, analyzed by Wänke et al. (ref. 13).

(4) Average of La, Sm, Eu, Yb

number of these elements compared to the Ca-Al-rich minerals. In the final column of this table are the results of trace element analyses of some Allende inclusions. These show that almost all of these elements are enriched in the inclusions by factors of 10 to 25 relative to Cl chondrites. Thus, in spite of the vast differences in geochemical behavior between elements as dissimilar in chemical properties from one another as the rare earths and the platinum metals, for example, these elements are all concentrated into the same mineral aggregates. The only feature common to all of these elements is that they condense from a gas of solar composition over the same temperature range as the major phases in the inclusions. They may have nucleated upon the major minerals or may have gone into solution in some of them during condensation. Alternatively, the trace elements may have become associated with th Ca-Al-rich phases by serving as the condensation nuclei for them. The trace element characteristics of the Allende inclusions are a very powerful argument in support of a condensation origin.

Gray et al. (ref. 14) reported the lowest Sr^{87}/Sr^{86} ratio (0.69877) ever measured in meteorites from a Rb-poor, Ca-rich inclusion in Allende. Such inclusions are thus the most primitive objects yet sampled in the solar system.

High-Temperature Metal and Forsterite Condensates in Carbonaceous Chondrites

Fuchs et al. (ref. 15) and Grossman and Olsen (ref. 16) described single euhedral crystals and loose aggregates of crystals of nearly pure forsterite from Murchison and other C2 chondrites. Their textures suggest that they may be direct condensates from the solar nebula. In that case, their compositions would indicate equilibration with the nebular gas at temperatures in excess of 900 K. Inside the crystals are tiny grains of metal with compositions in the range 3.8 to 8.9 mole percent Ni, 0.16 to 0.70 percent Co, 0.17 to 1.07 percent Cr, and up to 5.7 percent P. The Ni, Co, and Cr concentrations suggest that most of these grains could have equilibrated with a gas of solar composition in the temperature range 1430 to 1350 K at 10^{-3} atm. The phosphorous may have entered the grains when schreibersite formed as a result of the reaction of gaseous P_2 with the metal at 1420 K. The absence of grains with higher Ni contents than are observed may indicate that early, high-Ni metal was able to equilibrate before being isolated from the vapor by the growth of forsterite around it. This implies a time lag between the condensation of metal and that of forsterite, such as would be the case at pressures greater than $\sim 5.5 \times 10^{-5}$ atm.

Fractionation of Refractory Condensates

Larimer and Anders (ref. 6) showed that the ordinary and enstatite chondrites are depleted in the refractory lithophile elements by mean factors of 0.69 and 0.50, respectively, relative to the C1 chondrites. These include Ca, Al, Ti, Hf, Zr, Sc, Th, Y, and rare earths, many of the elements listed in table 2. In addition, the Vigarano Type C3 chondrites apear to be enriched in Ca, Al, and Ti by about 28 percent relative to C1 chondrites (ref. 17). Apparently these elements were transported together, as a group, from one part of the nebula to another prior to the accretion of the different classes of chondrites. The observed fractionations could have been produced by the addition or removal of only 1 to 2 percent of the total condensable matter in the form of high-temperature condensate aggregates, such as the Allende inclusions, to or from a gas of solar composition.

PLANETS

There is now considerable evidence in the chondritic meteorites that chemical fractionations accompanied the condensation of the solar nebula. This suggests that it may be more reasonable to picture planetary-sized objects as being composed of different proportions of the condensate components found within chondrites than to think of them as having accreted from different mixtures of the various meteorite types.

The Moon

Numerous analytical studies have shown that the lunar surface rocks are enriched in the refractory lithophile elements by factors of 5 to 100 and strongly depleted in the volatile transition metals and alkali metals relative to the solar system abundances (refs. 13, 18, and 19). These data have prompted a number of investigators to propose that this applies to the Moon as a whole and that the Moon accreted from a higher-than-chondritic proportion of the high-temperature condensates. The Allende inclusions represent approximately 4.5 wt% of the total condensable mater. Ganapathy and Anders (ref. 20) suggested that the Moon contains 24 percent early condensate; Wänke et al. (ref. 21) proposed 60 percent, and Anderson (ref. 22), in an extreme version of these models, suggested that the Moon is virtually 100 percent high-temperature condensate.

Condensation calculations suggest, however, and the Allende inclusions confirm, that the early condensate is rich in refractory siderophile metals such as Ir, Re, Os, and Ru (ref. 12). On the other hand, the lunar

rocks are depleted in these elements relative to chondrites by factors of 10^2 to 10^4. If these models of lunar composition are correct, the entire Moon should be enriched to a similar degree in these elements as it appears to be in the case of the refractory lithophiles. Their absence from the surface rocks would seem to imply that they have been concentrated into the lunar interior by a very efficient process such as partitioning into an Fe or Fe-S melt which sank under the influence of gravity.

Every meteoritic condensate aggregate whose oxygen isotopic composition has been measured is found to contain a 1- to 3-percent enrichment in O^{16} compared to "normal" solar system oxygen. Clayton et al. (ref. 23) attributed this effect to the survival of pre-existing interstellar grains during the early high-temperature stage of the nebula and their incorporation into the forming condensate crystals. Grossman et al. (ref. 24) reported that the lunar samples show no evidence of this anomaly. Furthermore, there is no known meteoritic material which can be mixed with the Allende inclusions to yield the inferred oxygen isotopic composition of the Moon. The major cosmochemical argument against a refractory condensate-rich Moon appears to be that the required early condensates having normal isotopic composition exist only in theory and have not been observed in meteorites.

The Terrestrial Planets

From the densities of the terrestrial planets, Urey (ref. 25) estimated that the Fe/Si ratio of Mercury is three times that of the Earth and five times that of Mars. This progressive decrease in planetary metal/silicate ratio with increasing heliocentric distance can be understood, at least qualitatively, in terms of condensation models. Hoyle and Wickramasinghe (ref. 26) and Cameron and Pine (ref. 2) presented physical models of the solar nebula in which the pressure and temperature in the median plane are highest in the center and decrease with increasing

heliocentric distance. In addition, we have seen that the temperature gap between the condensation temperatures of metal and forsterite increases with pressure. Mercury probably accreted from materials which condensed at a higher pressure than Venus or Earth. Therefore, it might be expected to possess a higher metal/silicate ratio if the nebular gases were dissipated at a time when metal was totally condensed and forsterite only partially condensed. At the same time, both metal and silicates may have been totally condensed in the region of the nebula where Venus and Earth formed because the pressure and temperature were considerably lower there. Mars may have formed largely from volatile-rich condensates containing a high ratio of oxidized iron to metallic iron. Grossman (ref. 4) and Lewis (ref. 27) have both pointed out the similarity between the pressure-temperature gradients required in this model and those actually predicted in hydrodynamic treatments.

The Earth: Homogeneous Versus Heterogeneous Accretion

Ringwood (ref. 28) proposed that the Earth was built homogeneously from material having the composition of C1 chondrites, and that the core formed when accretional heating reduced oxidized iron to the metallic state and melted it, allowing it to sink to the center of the Earth. Ringwood noted some features of terrestrial geochemistry which could not be explained adequately by this model. Among these are the highly oxidized nature and the high siderophile element content of the Earth's crust and upper mantle. Had the upper part of the Earth ever been in chemical contact with a sinking metallic melt, it should be very reduced and stripped of its siderophile elements.

In order to overcome these difficulties, Turekian and Clark (ref. 29) and Clark et al. (ref. 30) proposed that the Earth accreted heterogeneously; that is, that the Earth began accreting from material of one composition and finished with material of another

composition. Specifically, the Earth is pictured as having accreted in a stratified fashion from materials in the order of their condensation from the nebula. Thus, part of the metal core may have formed before lower-temperature magnesium silicates began to condense. A volatile-rich oxidized crust may have been the final layer to accumulate (ref. 31). Any siderophile elements present in this crust would remain in the upper part of the Earth since, in this model, the metal in the core was never in contact with the surface layers. This heterogeneous accumulation model makes use of the high-temperature reducing atmosphere of the solar nebula in order to produce the metallic core and avoids the difficulty of having to lose an enormous primitive atmosphere of CO which is a formidable obstacle to the in-situ reduction process envisioned by Ringwood.

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