MELTING OF A YAMATO L3 CHONDORITE (Y-74191) UP TO 30 KBAR

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Abstract: Melting phase relations of a Yamato L3 chondrite (Y-74191) have been studied in the pressure range 6 to 30 kbar. Solidus of the chondrite is defined by eutectic melting of Fe-Ni-S at about 950°C regardless of pressure. At 15 kbar, silicate solidus of the chondrite is about 250°C higher than the metal/ sulfide eutectic solidus and the temperature interval between the two solidi widens as pressure increases. Composition of the metal/sulfide melt changes from FeS-rich to Fe-Ni-rich as temperature increases. Ferrobasaltic liquids containing 18 to 23 wt % FeO and 0.2 wt % sulfur are produced as partial melts over a large pressure and temperature interval. At low temperatures below the silicate solidus, the metal/sulfide melts are small isolated patches dispersed in grain boundaries of silicate minerals. At high temperatures, the metal/sulfide melts become numerous globules surrounded by silicate melts. Segregation of the ferrobasaltic partial melt on top of the residual olivines and the metal/sulfide globules has been observed. Gravitational settling of the dense metal/sulfide globules, however, did not take place under any experimental condition at least up to 10 hr. Based on the partial melting experiments a model for genesis of pallasite meteorite is proposed.

1. Introduction

Terrestrial planets may have formed and evolved from chondritic source materials. There has been proposed a number of theories on the evolution of the terrestrial planets (*e.g.*, UREY, 1952; CAMERON, 1963; RINGWOOD, 1966, 1979) and the source material for the Earth is highly uncertian depending upon the various authors (*e.g.*, C l-type carbonaceous chondrite, RINGWOOD, 1961; H chondrite, MASON, 1966; E chondrite, MIYASHIRO, 1967). In order to study details of the initial igneous processes which might have taken place particularly on the Earth, we have planned a series of high-pressure melting studies using different rock types of meteorites up to about 300 kbar. In this report we note melting behaviors of a L3 chondrite (Y-74191) up to 30 kbar as a reconnaissance of the project.

The L3 chondrite was collected from Antarctica by the National Institute of Polar Research (YANAI *et al.*, 1978). Detailed petrological description on the rock specimen was made by IKEDA and TAKEDA (1979). The rock is composed of equigranular aggregate of spherical chondrules and a small amount of fine-grained matrix. Major constituent minerals are olivine, orthopyroxene, clinopyroxene, Fe-Ni metal and FeS. Clear glass is present in the groundmass of some chondrules. Bulk chemical composition of the chondrite is given in Table 1.

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Table 1. Chemical compositions of the L3 chondrite and its partial melts formed at high pressures.

Y-74191*		Run #	9**	13**	21**	26**
		P kbar	15	15	30	15
		T °C	1400	1650	1600	1400
SiO_2	40.09	SiO_2	49.2	47.8	43.5	45.9
TiO_2	0.24	TiO_2	0.4	0.2	0.2	0.2
Al_2O_3	2.89	Al_2O_3	10.8	4.7	6.1	9.4
FeO	14.68	FeO	18.1	19.4	21.5	23.2
MnO	0.35	MnO	0.5	0.5	0.5	0.3
MgO	24.89	MgO	10.3	22.2	19.4	11.6
CaO	1.79	CaO	6.8	2.7	3.4	4.5
Na_2O	0.97	Na_2O	3.3	0.9	1.9	2.8
K_2O	0.13	K_2O	0.4	0.2	0.3	0.4
Cr_2O_3	0.75	Cr_2O_3	0.7	0.7	0.7	0.7
H_2O-	0.05	NiO	0.1	0.0	0.0	0.1
H_2O+	1.13	S	n.d.	0.2	0.2	n.d.
P_2O_5	0.20	Total	100.6	99.5	97.7	99.1
FeS	5.01					
Fe	5.66	Fo %	77	86	83	70
Ni	0.85	K_D	0.31	0.34	0.34	0.37
Co	0.032	Fo: forsterite mol of coexisting olivine				
Total	99.712	K_D: olivine/liquid Fe-Mg partition coefficient				

* Analyst H. HARAMURA.   ** EPMA analysis.

2. Experimental Procedures

A small chip (about 2 g) of the L3 chondrite was ground in an agate mortar filled with acetone for 30 min.   Because of its ductility, a significant portion of Fe-Ni metal in the chondrite stuck onto the wall of the mortar and had been lost from the starting material.   Final grain size of the starting material was about 10 μm on an average, but coarse Fe-Ni metal and FeS grains up to about 100 μm were present in small amounts. In the present experiments two types of starting materials were prepared; (I) the rock was ground in the mortar and then kept in an oven at 110°C until immediately prior to each experiment; (II) the rock powder (I) was heated in a 1-atmosphere CO_2-H_2 gas mixing furnace at 900°C for 4 hr under oxygen fugacity equivalent to the iron-wüstite buffer.   The starting material-II is believed to have been perfectly dried during this heat treatment.   During the sample preparation, however, most of FeS in the starting material-II was reacted with the ambient gas and formed FeO.   The starting material-II was kept at 110°C in the same manner as that of the starting material-I.

High-pressure experiments were carried out with a piston-cylinder apparatus at the Institute for Thermal Spring Research.   A 1/2-inch diameter talc-pyrex-graphite-sintered Al_2O_3 assembly (furnace size; 6.0 mm O.D., 3.0 mm I.D. and 31 mm long) was employed with a small graphite capsule (2.4 mm O.D., 1.6 mm I.D. and 3.5 mm long) as a sample container.   The Pt/Pt$_{87}$-Rh$_{13}$ thermocouple was used for runs at temperatures less than 1400° C and the W$_{97}$-Re$_3$/W$_{75}$-Re$_{25}$ thermocouple was used at high temperatures.   No pressure correction was made on emf of the thermocouples.

Table 2. Run details.

Run #	P (kbar)	T (°C)	t (min)	Starting materials	Major phases present
1	8	1200	140	I	ol, opx, cpx, l, m, m/s
2	8	1300	180	I	ol, opx, cpx, l, m, m/s
3	15	900	120	I	ol, opx, cpx, m, s
4	15	1000	180	I	ol, opx, cpx, m, s, m/s
5	15	1100	150	I	ol, opx, cpx, m, s, m/s
6	15	1200	130	I	ol, opx, cpx, l, m, m/s
7	15	1300	120	I	ol, opx, cpx?, l, m, m/s
8	15	1400	20	I	ol, opx, l, m, m/s
9	15	1400	90	I	ol, opx, l, m/s
10	15	1400	600	I	ol, opx, l, m/s
11	15	1500	120	I	ol, opx, l, m/s
12	15	1600	60	I	ol, l, m/s
13	15	1650	200	I	ol, l, m/s
14	22.5	1200	120	I	ol, opx, cpx, m, m/s
15	22.5	1300	200	I	ol, opx, cpx, l, m, m/s
16	22.5	1400	120	I	ol, opx, l, m, m/s
17	30	900	120	I	ol, opx, cpx, m, s
18	30	1000	150	I	ol, opx, cpx, m, m/s
19	30	1400	120	I	ol, opx, cpx?, l, m, m/s
20	30	1500	120	I	ol, opx, l, m/s
21	30	1600	100	I	ol, opx, l, m/s
22	6	1150	90	II	ol, opx, cpx, m, w
23	6	1250	160	II	ol, opx, cpx?, l, m, w
24	15	1200	120	II	ol, opx, cpx, m, w
25	15	1300	120	II	ol, opx, cpx, l, m, w
26	15	1400	140	II	ol, opx, l, m, w
27	25	1300	85	II	ol, opx, cpx, m, w
28	25	1400	120	II	ol, opx, m, w
29	30	1400	120	II	ol, opx, cpx?, m, w

ol; olivine, opx; orthopyroxene, cpx; clinopyroxene, m; Fe-Ni metal, s; FeS, l; quenched silicate liquid, w; wüstite, m/s; metal/sulfide melt.

The high pressure run products were sectioned and polished surfaces of them were examined under a reflected light microscope and a scanning electron microscope (JEM-100C). Back-scattered electron images and semi-quantitative chemical analysis by an energy dispersive spectrometer equipped with the electron microscope were used in order to identify coexisting phases. Chemical analysis of the run products was also made with an electron microprobe X-ray analyzer (EPMA) model JXA-5A.

3. Experimental Results

3.1. Solidus

In the sulfur-bearing starting material-I, solidus is defined by eutectic melting of Fe-Ni metal and FeS. The eutectic melting temperature is between 900° and 1000°C and is insensitive to pressure (Fe-Ni-S solidus in Fig. 1). This result is consistent with melting experiments in the system Fe-Ni-S (Kullerud, 1963; Usselman, 1975). The
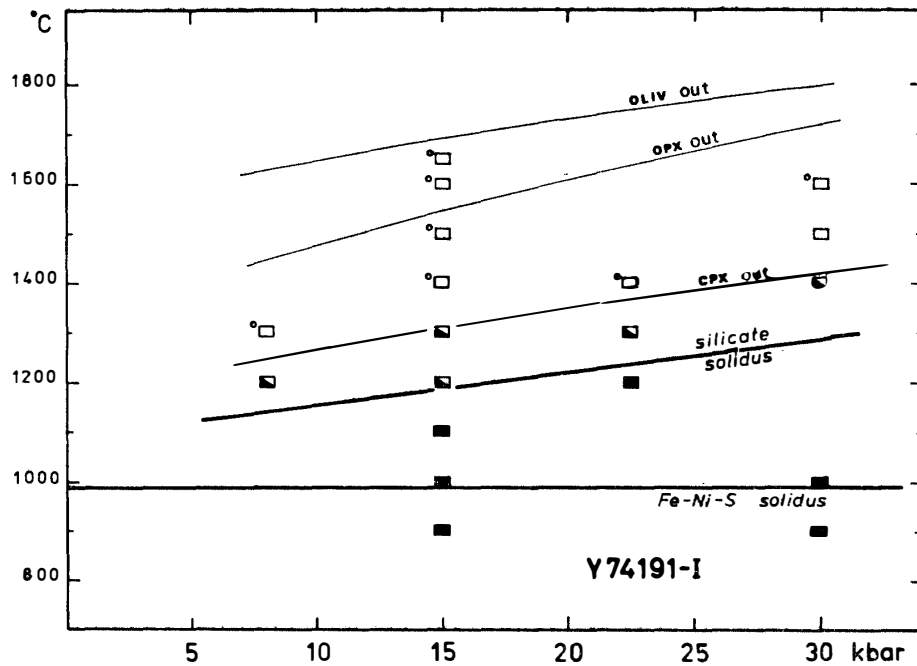
Fig. 1. Melting phase relations of the starting material-I. Solid squares represent experimental charges without quenched silicate melts, half-solid squares are those with quenched silicate melts only in some limited portions of the charges (cf. Fig. 3C), and open squares represent those with quenched silicate melts throughout the charges. Open circles denote the metal/sulfide globules (see Fig. 3D). The Fe-Ni-S solidus is drawn in accordance with the experimental data by USSELMAN (1975).
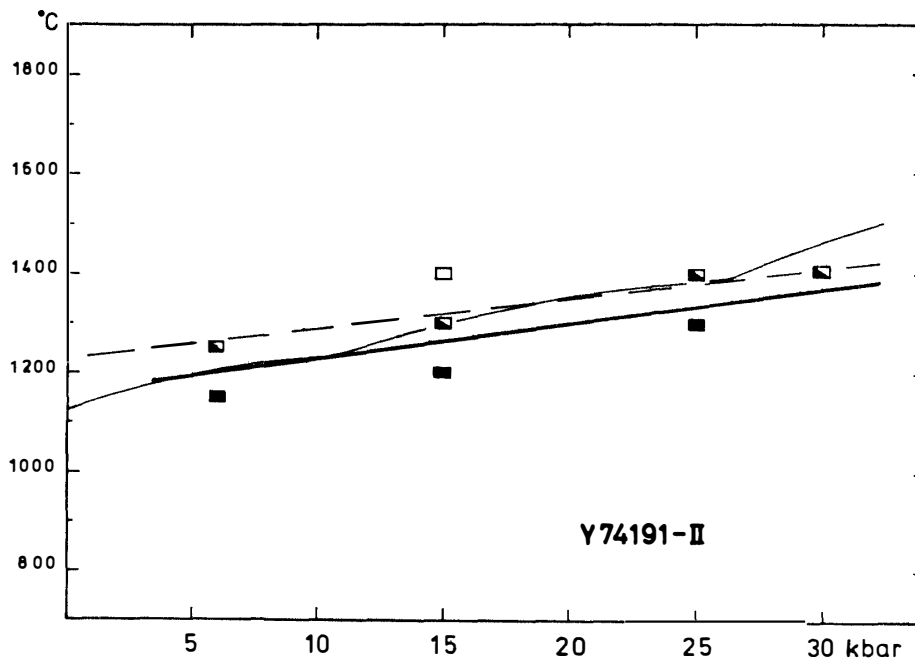

Fig. 2. Dry and sulfur-free solidus for the starting material-II is shown in the bold-solid line. Dashed line denotes the dry solidus for Allende C3 chondrite (SEITZ and KUSHIRO, 1974) and the light-solid line represents dry solidus of a magnesian peridotite (TAKAHASHI and KUSHIRO, 1983).

first appearance of silicate melt in the starting material-I (silicate solidus in Fig. 1) is at temperatures 100° to 300°C higher than the Fe-Ni-S solidus. The dT/dP slope of the silicate solidus has a positive value, hence the melting interval between the two solidi widens as pressure increases.

In the dried starting material-II which is free from sulfur as well as water, solidus is defined by melting of silicates (Fig. 2). The dry solidus of the L chondrite is at temperatures about 100° C higher than the silicate solidus of the starting material-I but show similar dT/dP slope (cf. Fig. 2 with Fig. 1). Effect of a small amount of H_2O which might be present in the starting material-I is a possible explanation for lowering of the solidus by about 100° C. The fact that the silicate melts produced from the starting material-I contain a small amount of sulfur (see below), however, could be another reason responsible for the discrepancy in the silicate solidi. Partial melts formed near the silicate solidus of the starting material-I are distributed in close vicinity to large Fe-Ni-S melt patches (Fig. 3C). It is suggested that the small amount of sulfur dissolved in the silicate melts lowered the dry solidus by about 100°C.

Melting behaviors of Allende C3 carbonaceous chondrite have been studied by SEITZ and KUSHIRO (1974). The dry solidus for the C3 chondrite is shown in Fig. 2 for comparison. The dry solidus for a magnesian terrestrial peridotite (TAKAHASHI and KUSHIRO, 1983) is also shown in Fig. 2. Compared with the solidus of the magnesian peridotite, dry solidi of the chondrites have smaller dT/dP gradients at least up to 30 kbar and are significantly lower in temperatures than the former at pressures above 25 kbar. The difference in solidus temperatures between the peridotite and the chondrites may probably be due to the difference in the $Fe/(Mg+Fe)$ ratio of constituent minerals. Because chondrites contain much greater amount of Na_2O than the terrestrial peridotite, clinopyroxenes in the chondrites contain larger jadeite component than those in the latter at high pressures where plagioclase is no longer stable. This may also be part of the reason why solidi of the chondrites are lower than the peridotite especially at high pressures.

3.2. Chemical composition of partial melts

Chemical compositions of silicate partial melts were determined with EPMA. Because of overgrowth effect of adjacent olivine and pyroxenes during quenching, glasses in grain boundaries are chemically modified from equilibrium compositions (JAQUES and GREEN, 1979; also see Fig. 1 of TAKAHASHI and KUSHIRO, 1983). Therefore, the EPMA analysis of quenched melts was made only where large pods of silicate melts are segregated. Some quenched silicate melts are occupied with dendritic quench crystals (Fig. 4B). A broad electron beam (about 10 μm in diameter) was employed for the glass analysis and a number of analyses from different beam spots were averaged. The results of the EPMA analysis are listed in Table 1.

The partial melts contain 18 to 23 wt % FeO, 10 to 22 wt % MgO and 43 to 49 wt % SiO_2. They may be called ferrobasalt to ferropicrite. Compared with terrestrial olivine tholeiites and picrites, they are depleted in MgO, CaO, Al_2O_3 and TiO_2 but enriched in FeO, Na_2O, Cr_2O_3 and MnO. Compared with lunar ferrobasalts, they are depleted in SiO_2, CaO, Al_2O_3 and TiO_2 but enriched in MgO, Na_2O, K_2O and Cr_2O_3. The compositions of the partial melts resemble that of howardite which is a mechan-

ical mixture of Ca-poor achondrites and Ca-rich basaltic achondrites.

About 0.2 wt % of sulfur was found to dissolve in the partial melts at 15 kbar and 1650°C and 30 kbar and 1600°C, respectively. According to WENDLANDT (1982), the maximum solubility of sulfur in a basaltic melt at a given pressure is a function of the FeO content in the melt. About 0.4 wt % S is expected for a ferrobasalt melt containing 20 wt % FeO and coexisting with pure FeS. Since the sulfide phase in our experiments is an Fe-Ni-S melt and the starting material contains approximately equal amounts of FeS and Fe-Ni (Table 1), our liquid analyses are not inconsistent with WENDLANDT's experiments. Silicate partial melts formed after extensive partial melting of L chondrite may contain approximately 0.2 wt % S or 0.5 wt % FeS. The partial melts formed near the solidus of the same chondrite may contain larger amounts of sulfur (0.3 to 0.4 wt % S) because coexisting Fe-Ni-S melts are more enriched in FeS component at those temperatures (*cf.* Fig. 3B with 3D). Partial melts of reduced chondrites (E and H chondrites) may contain smaller amount of sulfur than those of oxidized chondrites (LL and C condrites) because the (Fe-Ni metal)/FeS ratio is higher in the former.

3.3. Melting textures of Fe-Ni-S

The L3 chondrite (Y-74191) is a mixture of metal, sulfide and silicates. Upon melting of the chondrite, the three substances form two immiscible liquid systems (namely, metal/sulfide melt and silicate melt) which are very different in physical properties as well as chemical compositions (*i.e.*, density, viscosity, surface energy, diffusions, etc.). Melting textures of the chondrite are interesting in that how those immiscible liquids interact.

Due to the low viscosity and the high chemical diffusion rate, the metal/sulfide melt is unquenchable in our experiments (where the quenching rate is about 400°C/s) but produced a characteristic intergrowth of Fe-Ni metal and FeS (see Figs. 3B and 3D). Different from the silicate melts, the metal/sulfide melts do not wet the grain boundary of olivine and pyroxenes, thereby they occur as small isolated patches dispersed among grain boundaries of silicate minerals or as inclusions in them.

At temperatures slightly above the Fe-Ni-S solidus (see Table 1), a small amount of FeS-rich metal/sulfide melt was produced. Both Fe-Ni metal and FeS grains were found from the same run product (Run #5; 15 kbar, 1100°C) together with the metal/sulfide melt (see Figs. 3A and 3B). Because the bulk chemical composition of the chondrite (Y-74191) is enriched in Fe-Ni and depleted in FeS compared with the eutectic melt composition in the system Fe-Ni-S (USSELMAN, 1975), the stable phase at the above condition must be Fe-Ni if the metal/sulfide melts were homogeneous within the experimental charge buffered to a low oxygen fugacity. Presence of FeS relict adjacent to Fe-Ni metal grains indicates that chemical reactions did not take place between metals and sulfides which are isolated by silicate minerals.

At temperatures above the clinopyroxene liquidus in Fig. 1, most of the metal/sulfide melts in the experimental charges are surrounded by silicate melts. Due to a large surface energy relative to that of a silicate melt the metal/sulfide melts become globules of various sizes (Fig. 3D). It is noted that the metal/sulfide globules do not show coarsening effect in a time study at 15 kbar and 1400°C up to 10 hr (Run # 8–10).
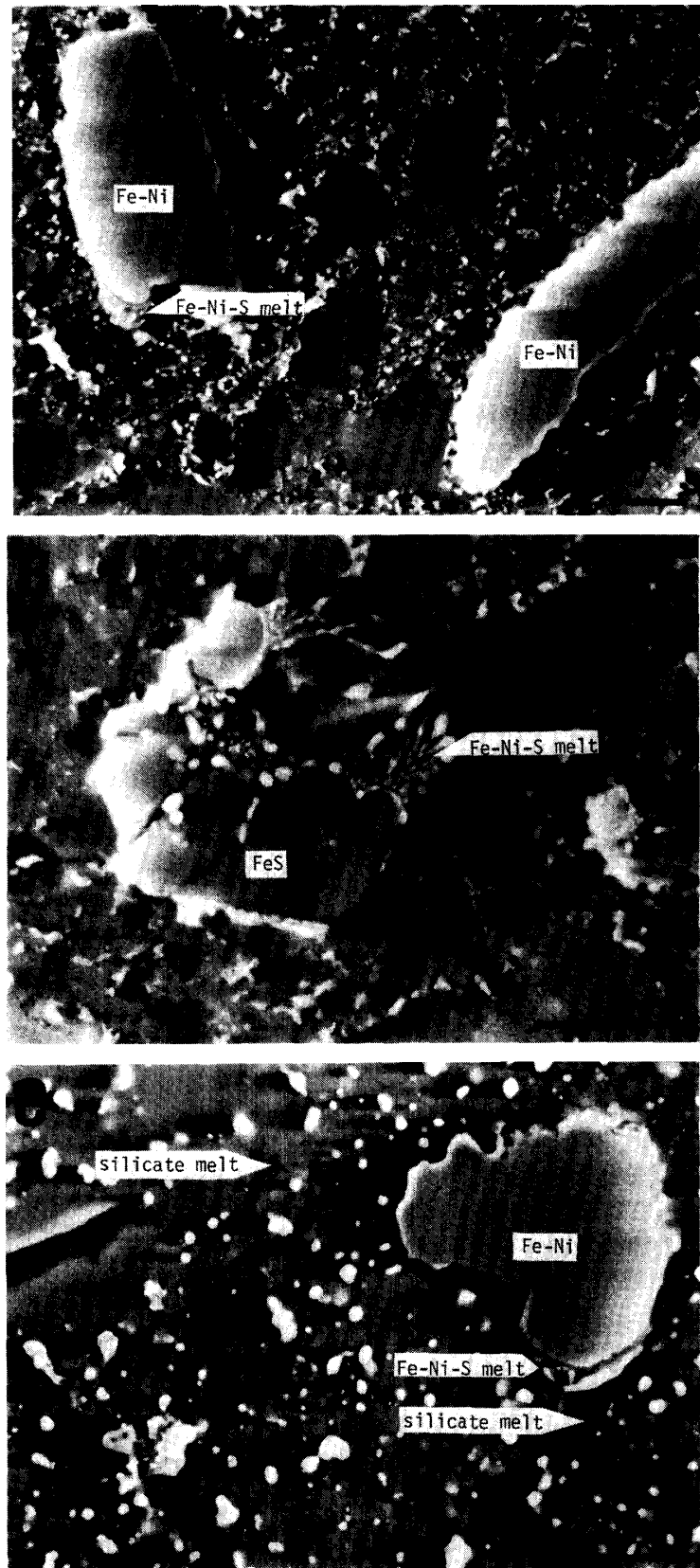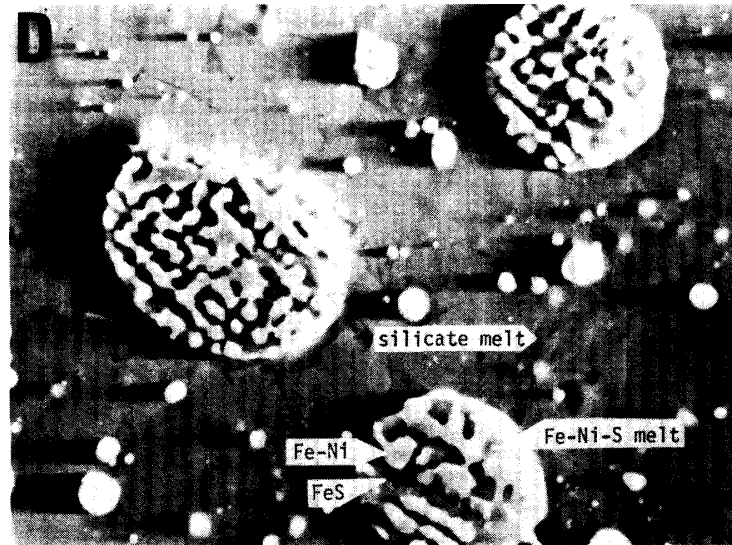
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Fig. 3.

Fig. 3. Back-scattered electron images of experimental run products. Scale bars represent 10 µm. (A and B, Run #5, 15 kbar, 1100°C; C, Run #7, 15 kbar, 1300°C; D, Run #9, 15 kbar, 1400°C). At 1100°C, both Fe-Ni and FeS solids are found to coexist with Fe-Ni-S melt (see A and B). The metal (white), sulfide (light-gray), and the metal/sulfide eutectic melt (fine-intergrowth of metal and sulfide) occur as small isolated patches dispersed in silicate minerals (dark-gray). At temperatures slightly above the silicate solidus (see C), the silicate melts are found only in the vicinity of the metal/sulfide melts. At temperatures more than 100°C above the silicate solidus (see D), the metal/sulfide melts are surrounded by the silicate melt and become globules of various sizes. Judging from the areal ratios of Fe-Ni metal to FeS in the quench-intergrowth textures of the metal/sulfide melts, chemical composition of the metal/sulfide melts change from FeS-rich (A and B) to Fe-Ni metal-rich (D). Chemical heterogeneity among the metal/sulfide melt globules exists, however, even at high temperatures (see D).

It is also to be noted that chemical compositions of adjacent metal/sulfide melt globules (which are estimated from the areal ratios of FeS to Fe-Ni in the quench intergrowth) are significantly different (see Fig. 3D for example). These observations suggest that chemical reactions among the metal/sulfide globules by virtue of chemical diffusions in silicate melts are sluggish. Sizes of the metal/sulfide globules may represent original grain sizes of the metal and sulfide which are in contact with each other in the starting material.

3.4. Gravitational separation of silicate melt and metal/sulfide melt

Segregation of silicate melt on top of residual phases was observed in an experimental charge which has run at 15 kbar and 1600°C (Run #12, see Fig. 4A). Segregation of silicate melt was observed in some other run products (Run #9, 13, 21, 26). Because of large temperature gradients across the experimental charges, however, the silicate melts in those run products were segregated at various portions of the sample container (top, bottom and wall). Run #12 was made at the University of Tokyo using a specially designed 3/4-inch diameter tapered-furnace assembly (KUSHIRO, 1976).
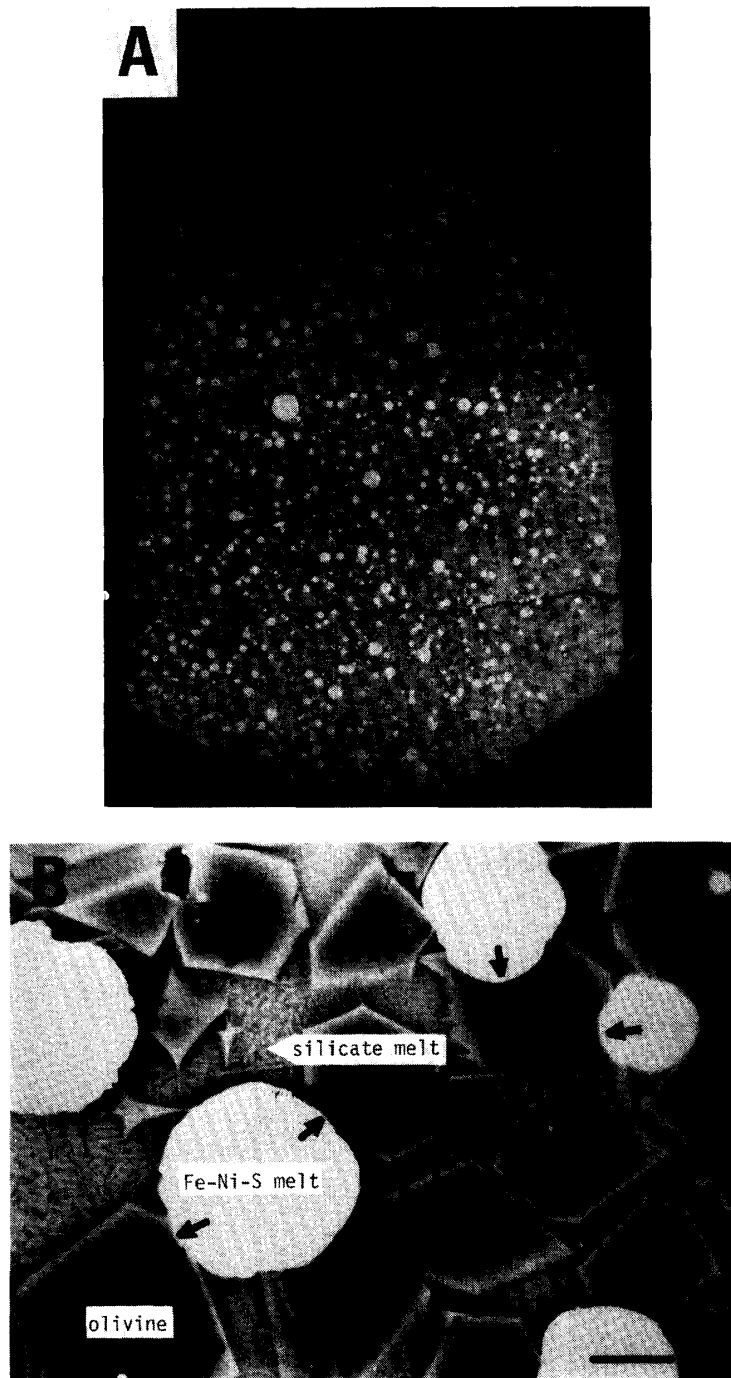
Fig. 4.   Back-scattered electron images of a run product (Run #12, 15 kbar, 1600°C) showing gravitational segregation of silicate melt on top of residual phases. (A, a whole view of the experimental charge, width of the photo is 1.7 mm; B, an enlarged photo of a residual part, scale bar represents 10 μm). Quenched silicate melts both at the top of the charge and in the interstitial part of the residues are filled with dendritic quench crystals. The metal/sulfied globules are fixed onto the surface of olivine crystals (see arrows in B). Compare Fig. 4A with Fig. 5B.

The sample was contained in the same graphite capsule and then encased in a sealed Pt-tube of 3 mm O.D. The Pt-tube was located in the center of the furnace assembly surrounded by powdered Pyrex-glass. The experimental charge of Run #12, therefore, is considered to have run under a hydrostatic condition and the expected temperature gradient across the charge is 1° to 2°C/mm.

In Run #12, about 30 vol % of the partial melt (total volume of silicate melt is about 50 vol %) was segregated on top of the residues within 1 hr (Fig. 4A). The residues consist of olivine (about 40 vol %), metal/sulfide globules (about 30 vol %) and interstitial silicate melt (about 30 vol %) (Fig. 4B). It is important to note that gravitational settling of the metal/sulfide globules was not observed in the experimental charge in spite of the very high degree of partial melting.

No sings of gravitational settling was detected under any experimental condition at least up to 10 hr for the metal/sulfide liquids, even though they are distinctly heavier than silicates. Because the sizes of the metal/sulfide globules are not large, the sinking velocity of them through partially molten silicate may be negligibly small. Furthermore, the metal/sulfide globules are fixed onto the surface of silicate minerals (see Fig. 4B) and are set in the three-dimensional network of silicate residues. The sinking velocity of the metal/sulfide globules, therefore, may be much smaller than the value calculated from the Stokes' equation.

4. Discussion

4.1. A model for genesis of pallasite

Pallasite is a stony-iron meteorite composed dominantly of olivine and Fe-Ni metal (MASON, 1962). Olivines are large in grain size (1 to 10 mm across) and are euhedral to ovoidal in shape. Average volume fraction of olivine in pallasite is about 65 vol % and is close to the volume of close-packed equidimensional matters (BUSECK, 1977). Based on its textural resemblance to magmatic olivine cumulates of terrestrial origin, BUSECK (1977) proposed a cumulate hypothesis for the origin of pallasite. However, the following problems have not been solved thoroughly (MATSUI *et al.*, 1980): 1) Why olivine, Fe-Ni metal and FeS which are very different in density reside together? 2) Under what conditions and by what mechanisms these three minerals have separated from the rest of the components in the primordial material? The present melting experiments provide useful information for understanding the origin of this unique stony-iron meteorite.

Olivine is the liquidus phase of the L chondrite at all pressures studied (Fig. 1). In a small parent body of pallasite (probably less than 500 km in radius according to WOOD, 1978 and MATSUI *et al.*, 1980), therefore, olivine is possible to form as the only residual silicate mineral after extensive partial melting. Metal/sulfide melt with or without Fe-Ni metal may have coexisted with the silicate melt together with the residual olivine at temperatures 1400° to 1600° C. As demonstrated in Run #12, the silicate partial melt may have segregated from the residues and migrated upwards. Gravitational separation of the metal/sulfide melts from the olivine residues, however, may have been disturbed as discussed previously. Upon cooling, iron/sulfide melts may crystallized as Fe-Ni metal and FeS. Olivines, however, remained essentially unchang-

ed except that their euhedral crystal surfaces have been rounded due to successive sub-solidus annealing effect (OHTANI, 1983). Thus, a cumulate composed of metal, sulfide and olivine is expected to have formed in the core of a small planet after extensive partial melting of its chondritic source material (Fig. 5).
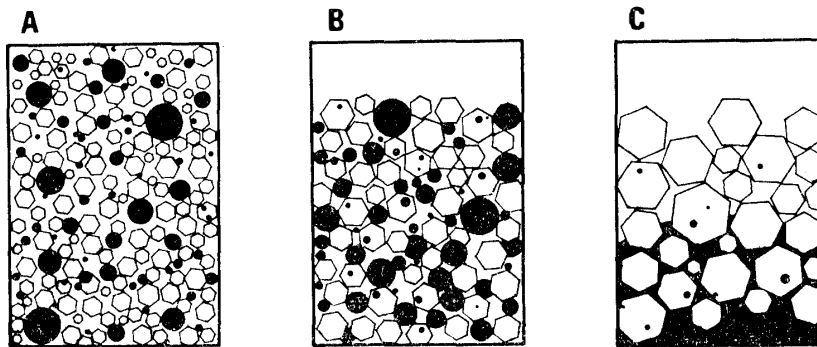

Fig. 5. Gravitational separation of silicate melt (white), olivine residue (hexagons) and metal/sulfide melt (black) in a hypothetical experimental time study. A; partial melting starts at a temperature considerably higher than the silicate solidus, yielding ferrobasaltic silicate melts, metal/sulfide melt globules and residual olivines. B; the silicate melt segregates on top of the residual phases but the metal/sulfide melt globules do not separate from the residual olivines. C; segregation of the silicate melt has completed and a pallasite-like cumulate is formed at the bottom. The experimental charge of Run #12 (Fig. 4A, B) corresponds to stage B.

The $Fe/(Mg+Fe)$ ratio of pallasite olivine is in the range 0.10 and 0.20 and the value between 0.12 and 0.13 is most frequent (BUSECK, 1977). Judging from the present results, a very high degree of partial melting (70 to 90 vol %) is necessary to form pallasite from the L chondrite provided that graphite buffered oxidation state of the materials in nature as well as in the present experiments. Pallasites with magnesian olivines might have been formed by partial melting of H chondrite (RINGWOOD, 1961). Further experimental melting studies are necessary examining various chondrite compositions in order to specify conditions of pallasite formation.

4.2. Some implications on the early history of the Earth

There are a number of reasons suggesting that the Earth has experienced a very high temperature stage in its early history. If the Earth was formed from chondritic source material (homogeneous accretion), formation of the Earth's core must be the biggest event in its early history. Currently it has been accepted that the core formation was initiated by gravitational settling of a molten Fe-Ni (and probably with S) layer formed at some depth of the primitive earth (ELSASSER, 1963).

The present experiments, however, demonstrated that the metal/sulfide eutectic melts form small isolated patches dispersed in silicate matrix and do not interconnect each other. Under such circumstances, segregation of the metal/sulfide melts is not promoted. To form a large-scaled metal/sulfide melt layer as postulated by ELSASSER (1963), therefore, it would take much longer time than he expected.
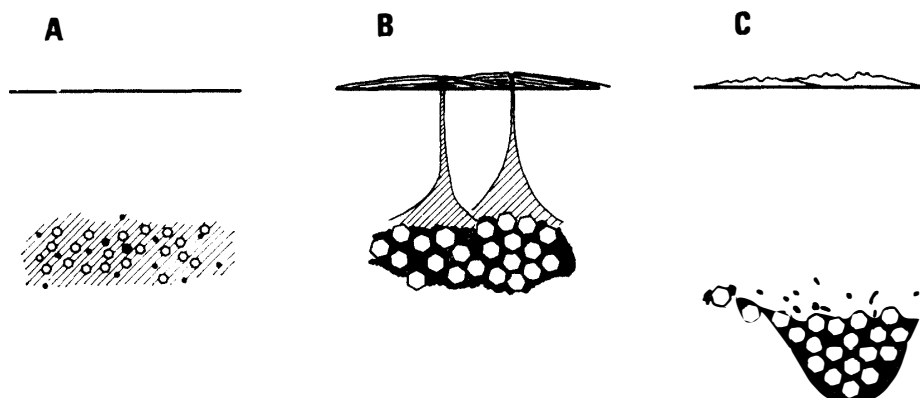
Fig. 6.   A model for partial melting process in the primitive Earth.   A; partial melting of the initially homogeneous chondritic Earth starts at some depth.   B; ferro-basaltic silicate melt segregates from the pallasite-like residue and extrudes onto the Earth's surface.   C; the Fe-Ni-rich dense residue subsides down to the center of the primitive Earth and begins to form the core.

Alternatively, if the time constant for the metal/sulfide segregation was large enough and the temperature of the primitive Earth exceeded the silicate solidus of the source material, segregation of the silicate melts may have taken place within a short period as demonstrated in Run #12.   After the silicate melt segregation, a batch of metal/sulfide-rich residues would have been formed at some depths and the dense residuals then have subsided down to the center of the primitive Earth (Fig. 6).   Formation of the Earth's core might have been initiated by such a mechanism.

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