PETROLOGICAL STUDY OF Ni-Fe METAL IN SOME ORDINARY CHONDRITES

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Abstract : Mode of occurrence, compositional zoning and bulk chemical composition of metals in the Yamato ordinary chondrites have been studied. Metal grains in the chondrites are discrete taenite and kamacite, and composite grains of the two phases including plessite. They show a remarkable compositional zoning identical to that in iron and stony-iron meteorites. Occasionally Ni-Fe metal coexists with troilite; in such a case, the part in contact with troilite is enriched in Ni. Generally the isopleths of Ni in zoned metal grains are parallel to their outlines, indicating that the compositional zoning was formed *in situ*.

Frequency distribution of bulk chemical composition of metal grains in Yamato-74115 (H5) chondrite shows two distinct peaks at different compositions; one is at the kamacite composition and the other at the taenite composition. Those in Yamato-74354 (L6) and -74190 (L6) chondrites show one distinct peak at the Ni composition intermediate between kamacite and taenite, and two obscured peaks, of which one is at kamacite composition and the other at taenite composition. Based on the Ni-Fe binary phase diagram, it is inferred that metals in H chondrite began to crystallize at least below 700°C and those in L chondrite below 620° C. Probably kamacite began to crystallize apart from the original taenite grains, but changed its composition in equilibrium with taenite down to 550° C in H chondrite and 470° C in L chondrite, having resulted in the formation of the M-shaped zonal pattern. Below this temperature and at least down to 300° C, the two phases were in a partial equilibrium and a remarkable compositional zoning of plessite was formed.

1. Introduction

Ordinary chondrites have extremely homogeneous silicate and oxide minerals in spite of various degrees of recrystallization; some of them contain clearly defined chondrules and their matrix minerals are fine-grained, some contain obscured chondrules and their matrix is mildly recrystallized, and some others contain coarse-grained minerals and their chondrules are hardly defined. However, Ni-Fe metals in them show a remarkable compositional zoning. In this sense, the ordinary chondrites are not wholly equilibrated. In general, at about 900°C which is the equilibrium temperature estimated from olivine and pyroxenes, the diffusion

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coefficients of elements within metals are nearly equal in order of magnitude to that of olivine; the diffusion coefficient of Ni in taenite (8 wt.% Ni) at 1000°C is about 10^{-11} – 10^{-12} in order (MOREN and GOLDSTEIN, 1978) and that of Mg-Fe in olivine (Fa₉₀) at the same temperature is about 10^{-13} (TAKAHASHI, 1979). At lower temperatures, however, the diffusion coefficient of metal is larger than that of silicate minerals, and therefore, the thermal history down to temperature as low as 300°C has been recorded in the chemistry of metal grains. The phase diagram in the Ni-Fe system has been investigated by a number of investigators since OWEN and LIU (1949). GOLDSTEIN and his coworkers have shown the cooling rate of iron and stony-iron meteorites (1965, 1967 and 1972) and emphasized the effect of phosphorus on the Ni-Fe phase relations (1972). On the other hand, WOOD (1967) studied metals in chondrites and estimated the cooling rate on the basis of that of iron meteorites (WOOD, 1964). Few works have been done on the metals of stony meteorites after the study by WooD (1967).

In the present paper, metals in five ordinary chondrites have been examined mainly by means of electron microprobe. Petrographical descriptions of silicate minerals are given in another paper (NAGAHARA, 1979). Three of them belong to L6 chondrite, one LL chondrite and one H5 chondrite.

2. Mode of Occurrence

Both kamacite and taenite are contained in the ordinary chondrites. In iron and stony-iron meteorites, they occur closely together; kamacite was exsolved from taenite, whereas in chondrites they occur as isolated grains. However, their zoning patterns are quite similar. Moreover, plessite, fine intergrowth of the two phases, occurs in both chondrites and stony and stony-iron meteorites. The width of each phase in plessite in chondrites is variable.

In general, taenite grains are more abundant and larger than kamacite grains in the L-type chondrites. Most of them occur in the matrix (parts other than chondrules) and have irregular anhedral shapes. High-Ni taenite is often observed in the outer portion of plessite (Photo 1). In some cases, they coexist with low-Ni kamacite and/or troilite (Photos 2 and 3); in such a case, they show distinct boundaries. Rarely minor troilite globules are contained in taenite grains. Based on the phase relation in the Fe-Ni-S ternary system (KULLERUD, 1963), they have been equilibrated below 400°C.

In Yamato-74190 (L6) chondrite, which has a strong recrystallized texture and whose plagioclase is changed into maskelynite, fine blebs of taenite are dispersed in a vein, which consists of cryptocrystalline silicate minerals and is black (Photo 4). It seems to have been formed by the shock of impact which transformed plagioclase into maskelynite.

In LL-type chondrite (Yamato-74646), most of metal grains are taenite, and

plessite and kamacite are very rare. They also occur as interstitial and anhedral grains.

In H-type chondrite (Yamato-74115), kamacite and plessite are abundant, and taenite is less.

These features are consistent with the Prior's rule; that is, the Ni content of metal is the highest in LL-type chondrites and lowest in H-type chondrites.

As mentioned above, most of metal grains occur in the matrix, but occasionally occur within chondrules (Photo 5). In the latter case, they show a fused and dispersed appearance. This is particularly well observed in the radial-pyroxene chondrules. Rarely irregular and relatively large taenite has grown over the matrix and chondrules.

3. Pattern of Compositional Zoning

The most common zonal pattern is shown in Fig. 1. Taenite shows a remarkable M-shaped zonal pattern, that is the rim is rich in Ni and the core is rich in Fe. Plessite generally has a Ni-rich rim and a Fe-rich core and the general zonal pattern is the same as that of taenite, and in its central portion many kamacite crystals



Fig. 1. Representative zonal patterns of kamacite, plessite and taenite in Yamato-74115 (H5) chondrite. Kamacite has a slightly Ni-rich core and a Fe-rich rim, whereas plessite generally has a Ni-rich rim and a Fe-rich core with many kamacite grains in its inner part. Taenite has a remarkable zoning with a Ni-rich rim and a Fe-rich core.



Fig. 2. Several different modes of occurrence of metal in Yamato-74354 chondrite. A-D: discrete taenite, E: discrete kamacite, F: simple association of taenite and kamacite, G-N: various associations of taenite and kamacite. Numbers are N: content. Bar represents 10 μ.

are observed. In kamacite, the central portion is enriched in Ni, but has a nearly uniform composition. These three kinds of metal grains contain equal amount of phosphorus, and the difference of their zonal pattern does not depend upon phosphorus.

Fig. 2 shows several different modes of occurrence of metals in Yamato-74354 (L6) chondrite: A–D are isolated single phase taenite and E is an isolated single phase kamacite. As shown in the figure, the composition of the rim is not necessarily uniform but that of the central portion is nearly uniform. F–N show various cases of association of the two phases; F shows simple association of taenite and kamacite, G–I show complex association of the two phases, and J–N show composite grains with the Ni-rich rim and the Fe-rich core with many exsolution lamellae of kamacite. It should be noted that the composition of the two phases near the contact is not uniform even within a single grain. These observations suggest that the stage of formation of compositional zoning of taenite is earlier than that of formation of the internal fine exsolution of kamacite.

The M-shaped zonal pattern of taenite and plessite grains was formed *in situ*. Fig. 3 shows the distribution of Ni in a metal grain which coexists with troilite. It shows a zonal pattern controlled by the outline of the grain; the content



Fig. 3. Ni distribution pattern in taenite grain coexisting with troilite in Yamato-74354 chondrite.



Fig. 4. Ni distribution pattern in the composite grain of taenite and kamacite in Yamato-74362 chondrite.

of Ni is the lowest in the central portion, and increases gradually toward the outer portion. Near the contact with troilite, the Ni content is the highest. This zonal pattern indicates that the metal grains in the chondrites are not the fragments derived from iron meteorite or stony-iron meteorite which already had the Ni-Fe zoning.

Fig. 4 shows the distribution of Ni in a composite grain of taenite and kamacite, having a linear boundary. In the taenite portion, Ni is more abundant and decreases toward the contact with kamacite, whereas in the kamacite portion, Ni is slightly more abundant in the central portion. In this case the zonal pattern of Ni is not clearly concentric as is in the previous case, but the pattern still

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appears to be controlled by the outline of the grain.

4. Chemical Composition of Metals

As is obvious from the Ni-Fe binary diagram, below 910°C, α -phase, kamacite is exsolved from γ -phase, taenite. To examine the existence of isolated kamacite grains, all the metal grains in a thin section were analyzed with a defocused beam of EPMA. As shown in Fig. 2, kamacite has a nearly constant composition. If a grain contains both kamacite and taenite, the bulk composition of it should be more than that of kamacite only and if isolated kamacite exists, the bulk composition of the grain should be below 7 wt.% Ni. The results are shown in Figs. 5 and 6. In these figures, one square shows the bulk composition of one metal grain. As shown in Fig. 5, Yamato-74115 (H5) chondrite has two distinct peaks; one is at the kamacite composition (Ni 3-8 wt.%) and the other at the taenite composition (Ni 26-31 wt.%). The small peak in the range of Ni 13-18 wt.% indicates the association of the two phases in a single grain. The pattern of association of the two phases is various, such as plessite or coexistence of two phases such as shown in Fig. 4. The peak at the composition of kamacite indicates the existence of isolated kamacite grains. The equilibrium temperature at which taenite of Ni 28 wt.% and kamacite of Ni 6 wt.% can coexist is about 550°C according to the Ni-Fe phase diagram. It is interpreted that the two phases had coexisted in equilibrium with each other along the solvus and that the nucleation site of kamacite was different from the original taenite.

Fig. 6 shows the results on Yamato-74354 (L6), -74362 (L6), -74190 (L6)



Fig. 5. Frequency distribution of bulk chemical composition of metal grains in Yamato-74115 (H5) chondrite. The number in parentheses is that of analyzed grains in one thin section. One square corresponds to one grain, and the effect of size of grain is neglected. Two distinct peaks are remarkable.



Fig. 6. Frequency distribution of bulk chemical composition of metal grains in Yamato-74354 (L6), -74362 (L6), -74190 (L6) and -74646 (LL6) chondrites. Symbols are the same as those in Fig. 5. One distinct peak with intermediate Ni composition and two vague peaks with lower and higher Ni are observed in the upper two samples.

and -74646 (LL6) chondrites. The former two have a broad peak in the range of Ni 10–20 wt.% and small peaks at both the lower Ni (Ni 5–8 wt.%) and the higher Ni (Ni 30–40 wt.%) compositions. Metals with Ni 10–20 wt.% mean the mixture of the two phases. This frequency distribution is clearly different from that for the Yamato-74115 chondrite. It is also obvious that most of the metal grains of these two L6 chondrites are plessite or are composed of various associations of the two phases and a part of them are isolated kamacite grains. This fact indicates that some kamacite grains grew within or near taenite grains. Thus the mode of occurrence of the two phases is different among Yamato-74115 (H5) chondrite and Yamato-74354 and -74362 (L6) chondrites. The equilibrium temperature of taenite of Ni 35 wt.% and kamacite of Ni 7 wt.% is about 480° C according to the Ni-Fe phase diagram.

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In Yamato-74190 (L6) and -74646 (LL6) chondrites, all the metal grains analyzed are high-Ni taenite, however it is not certain whether no kamacite grains exist, because of the scarcity of analyzed grains.

5. Thermal History of Metal Grains

Average H-type chondrites contain metals with 8-12 wt.% Ni and L-type chondrites contain those with 15-20 wt.% Ni (Fig. 7). Equilibrium temperatures estimated from the coexisting olivine and pyroxenes are between 800° C and 950° C in all the above-mentioned chondrites. At such high temperatures, these metals must be single phase taenite.

Metals in H-type chondrites must have been single phase taenite at least down to 700°C. If nucleation started the condition of supercooling, metal must have been single phase down to temperature 700°C. Below this temperature, nucleation of kamacite began apart from the original taenite and the two phases coexisted in equilibrium down to 550°C, keeping their compositions along the solvus. Below 550°C, because of the decrease of the diffusion coefficient, the two phases were in equilibrium only at their contact and the grains were no longer homogeneous and the extremely Ni-rich rim of taenite was formed. Rarely plessite was formed. Judging from the composition of the two phases near the contact, the temperature must have lowered at least to 300°C. Below 300°C, even a partial equilibrium was not attained.



Fig. 7. The Ni-Fe binary equilibrium phase diagram and possible thermal history of metal grains. The patterned areas are the compositional ranges of metals in average H- and L-type chondrites. See text for explanation.

On the other hand, L-type chondrites contained single phase taenite at temperature at least down to 620° C. Below this temperature, kamacite started to crystallize apart from the original taenite grains and the two phases coexisted down to about 470° C, keeping their compositions along the solvus. The volume of kamacite crystallized is much less than that in H-type chondrites, according to the lever rule, as shown in the phase diagram. The M-shaped pattern was then formed. Below 470° C, perhaps due to the lowering of the diffusion coefficient, the two phases were in a partial equilibrium and much kamacite was formed within and/or in contact with taenite, and most of plessite was formed.

The difference in the mode of occurrence of metals between H-type chondrites and L-type chondrites is considered to be due to that the original bulk composition of metals is richer in Ni in the L-type chondrites than in H-type chondrites, so that in H-type chondrites kamacite recrystallized at higher temperature and its amount is greater than in L-type chondrites. It is not certain, however, why the two types had been equilibrated down to different temperatures. Possible explanations of this difference are: 1) the thermal history of L-type chondrite and H-type chondrite was different, 2) the compositional difference caused the difference in the temperature of equilibration, 3) the diffusion coefficient is different between the two types (in general, the greater the Ni content in Ni-Fe metal, the smaller the diffusion coefficient (MOREN and GOLDSTEIN, 1978)). Of course, more samples should be examined to know if these differences can be generalized to that of chemical groups.

6. Conclusion

The remarkable compositional zoning observed in metal grains in ordinary chondrites was formed *in situ*. Metal grains crystallized were initially single phase taenite at temperature above 700°C in H-type chondrites and 620°C in L-type chondrites. Then kamacite started to crystallize apart from the original taenite grains. These two phases coexisted in equilibrium at temperature down to 550°C in H chondrite and 470°C in L chondrite and the M-shaped zonal pattern was formed. Disequilibration of the two phases began and the grains became heterogeneous below these temperatures, and the remarkable zoning of taenite in H chondrite and plessite in L chondrite was formed at least down to 300°C.

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References

- GOLDSTEIN, J. I. (1967): Cooling rates of 27 iron and stony-iron meteorites. Geochim. Cosmochim. Acta, **31**, 1001–1023.
- GOLDSTEIN, J. I. and DOAN, A. S., Jr. (1972): The effect of phosphorous on the formation of the Widmanstätten pattern in iron meteorites. Geochim. Cosmochim. Acta, 36, 51-69.
- GOLDSTEIN, J. I. and OGILVIE, R. E. (1965): The growth of the Widmanstätten pattern in metallic meteorites. Geochim. Cosmochim. Acta, 29, 893-920.
- KULLERUD, G. (1963): The Fe-Ni-S system. Carnegie Inst. Washington, Yearb., 62, 175-189.
- MOREN, A. E. and GOLDSTEIN, J. I. (1978): Cooling rate variations of group IVA iron meteorites. Earth Planet. Sci. Lett., 40, 151-161.
- NAGAHARA, H. (1979): Petrological studies on Yamato-74354, -74190, -74362, -74646 and -74115 chondrites. Mem. Natl Inst. Polar Res., Spec. Issue, **15**, 77–109.
- OWEN, E. A. and LIU, T. H. (1949): Further X-ray study of the equilibrium diagram of the iron-nickel system. J. Iron Steel Inst., London, 163, 132–137.
- TAKAHASHI, E. (1979): Thermal history of lherzolite xenoliths. To be submitted to Geochim. Cosmochim. Acta.
- Wood, J. A. (1964): The cooling rates and parent planets of several iron meteorites. Icarus, 3, 429-459.
- WOOD, J. A. (1967): Chondrites: Their metallic minerals, thermal histories, and parent planets. Icarus, 6, 1-49.

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Photo 1. Photomicrograph of plessite in Yamato-74115 (H5) chondrite. Inner part is fine intergrowth of kamacite and taenite. Clear taenite outlines the outer portion. Reflected light. Scale bar is 0.5 mm.

Photo 2. Photomicrograph showing taenite (light white) and associated troilite (dark white) in Yamato-74354 (L6) chondrite. Reflected light. Scale bar is 0.25 mm.

Photo 3. Photomicrograph showing association of taenite (light white) and troilite (dark white) in Yamato-74354 (L6) chondrite. Ni distribution of taenite grain is shown in Fig. 3. Reflected light. Scale bar is 0.5 mm.



Photo 4. Photomicrograph of dispersed taenite grains in the fused vein of cryptocrystalline minerals in Yamato-74190 (L6) chondrite. Reflected light. Scale bar is 0.5 mm.

Photo 5. Photomicrograph of dispersed kamacite grains within a radial-pyroxene chondrule in Yamato-74115 (H5) chondrite. Reflected light. Scale bar is 0.5 mm.

Photo 6. Photomicrograph of Ni-Fe metal and troilite (opaque) surrounding the granular olivine chondrule in Yamato-74354 (L6) chondrite. Transmitted light. Scale bar is 0.5 mm.