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PETROCHEMICAL STUDY OF THE YAMATO-691 ENSTATITE CHONDRITE (E3) V: COMPARISON OF MAJOR ELEMENT CHEMISTRIES OF CHONDRULES AND INCLUSIONS IN Y-691 WITH THOSE IN ORDINARY AND CARBONACEOUS CHONDRITES

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Abstract: Major element compositions of chondrules and inclusions in Y-691 are different on the whole from those in ordinary and carbonaceous chondrites; the (CaO+AlO_{1.5})/(CaO+AlO_{1.5}+SiO₂) and MnO/MgO ratios of chondrules in Y-691 are lower than those in ordinary and carbonaceous chondrites, but the $SiO_{2}/(SiO_{2}+MgO)$ and MgO/(MgO+FeO) ratios are higher. These differences suggest that the chondrule precursors of Y-691 were produced by intense fractional condensation in comparison with those of ordinary and carbonaceous chondrites. For Y-691, the Ca/Al ratios of chondrules showing high Na/Al are lower than those of chondrules showing low Na/Al. This indicates that a fraction of Ca component was expelled from the chondrule precursors by reactions with a nebular gas, which also introduced alkalis into the chondrule precursors. In addition to magnesian chondrules, Y-691 includes unusual inclusions and ferroan chondrules. One unusual glassy inclusion, as well as others, are similar in chemical composition to fine-grained CAI's and amoeboid olivine inclusions in carbonaceous chondrites, respectively. The precursors of the unusual inclusions and ferroan barred-Ol-Px chondrules seem to have formed under oxidized conditions, whereas the precursors of magnesian chondrules were produced from a reduced nebular gas. The oxidizing conditions may have been produced by evaporation of chondritic or cometary materials in the solar nebula.

1. Introduction

This study is one of the Y-691 consortium studies, concentrating mainly on comparison of chemical compositions of chondrules in Y-691 with those in chondrites of other chemical groups. The companion papers (IKEDA, 1988a, b, 1989a, b) dealt with bulk chemical compositions of chondrules and inclusions in Y-691 and descriptions and mineral compositions of unusual silicate-inclusions, chondrules, and opaque-mineral nodules in Y-691.

Although major element compositions of chondrules in unequilibrated ordinary chondrites (UOC's) and carbonaceous chondrites of C3 type were already reported and discussed by many authors (e.g., WALTER, 1969; OSBORN et al., 1973, 1974; MCSWEEN, 1977; DODD, 1978a, b; FODOR and KEIL, 1978; IKEDA, 1983a; BISCHOFF and KEIL, 1983), those in unequilibrated enstatite chondrites (LEITCH et al., 1982;

SMITH et al., 1983; BISCHOFF and KEIL, 1983; IKEDA, 1983a; GROSSMAN et al., 1985) have not been sufficiently studied. In this paper, the major element compositions of chondrules and inclusions in Y-691, which were reported by IKEDA (1988a), are compared with those in UOC's and C3's (IKEDA, 1980, 1982, 1983a) in order to elucidate the differences in chemical composition and to discuss their origins.

As is already well known, the major element compositions of whole E chondrites are different from those of O and C chondrites (LARIMER and ANDERS, 1967; WASSON, 1974; SEARS *et al.*, 1982; KALLEMEYN and WASSON, 1986). My major interests in the chemistry of chondrules and inclusions are to answer the following questions: (1) Is there any difference in the chemical compositions of chondrules and inclusions between E, O and C chondrites? (2) Does the difference reflect the bulk chemical compositions of the whole rocks? (3) What was the origin of the difference?

2. Brief Description

The Y-691 chondrite was first classified as an E3 by IKEDA (1979). It consists of chondrules, unusual inclusions, opaque-mineral nodules, mineral fragments (larger than micrometer size), and fine-grained interstitial materials. In this paper, the term "unusual inclusion" is used for objects consisting mainly of silicates, which are dark or opaque in transmitted light (IKEDA, 1988b). "Chondrules" include droplet chondrules and chondrule fragments. Many textural types of chondrules are observed in Y-691 (IKEDA, 1989a). The major type of chondrule is radial-pyroxene (Px), which is also the largest in size. Other common types are porphyritic or granular chondrules. Minor types include barred-olivine(Ol)-Px, spherulitic cryptocrystalline, massive cryptocrystalline, and transparent-SiO₂ chondrules. The detailed mineralogy and petrology of Y-691 were reported by OKADA (1975) and OKADA *et al.* (1975), and detailed description of chondrules and inclusions are given by IKEDA (1988b, 1989a).

3. Chemical Compositions of Chondrules and Inclusions

IKEDA (1988a) measured the major element compositions of chondrules and inclusions by using a defocussed beam (about 30-50 microns across) of an electronprobe microanalyzer (EPMA). A broad beam was bombarded on each chondrule studied, avoiding the the area of opaque phases such as metals and sulfides in order to measure the major element compositions of the silicate-oxide portion. The same procedure was repeated for different portions of a chondrule to cover the entire area of the chondrule except for the opaque minerals. However, very fine-grained opaque phases (smaller than a few microns) such as Fe-metal and troilite are sometimes present. Therefore, small fractions of the measured FeO contents might be "Fe" of Fe-metal and/or troilite. Unusual inclusions in Y-691 generally do not contain grains of metal and sulfide larger than ten microns across (IKEDA, 1988b), indicating that the measured chemical compositions are near representative of the bulk compositions. However, chondrules in Y-691 often include grains of opaque phases, larger than ten They are mainly Fe-metal and troilite; other microns across (IKEDA, 1989a). sulfides such as niningerite and oldhamite are rare in Y-691 chondrules (IKEDA, 1989a). Therefore, the measured Fe contents do not represent the total Fe contents of the chondrules. Except for Fe, the contents of other elements obtained by IKEDA (1988a) are near representative of the bulk contents of chondrules with a few exceptions where chondrules include niningerite or oldhamite in fairly large amounts.

According to GROSSMAN *et al.* (1985), chondrules in Qingzhen (E3) include metals and sulfides in fairly large amounts; the most common opaque phases are troilite and oldhamite, which are found in every chondrule, and niningerite and kamacite occur in many chondrules. The elements, Na, Al, Si, and K are almost completely lithophile in the chondrules, whereas Ca, Mg, Ti, Cr, and Mn are variably distributed between silicates and sulfides in chondrules. This situation is different for Y-691 chondrules. Niningerite and oldhamite are not common in Y-691 chondrules, and the amounts are negligible for most chondrules; therefore, Ca, Mg, and Mn are mostly lithophile. Cr and Fe are distributed between silicate and opaque phases in Y-691 chondrules and are partly lithophile and partly chalcophile or siderophile.

IKEDA (1983a) classified chondrules in unequilibrated chondrites into three chemical types, calcic plagioclase (CP), intermediate plagioclase (IP), and sodic plagioclase (SP), on the basis of the atomic ratios of Al/(Al+Na+K) of the chondrules. The boundaries of this ratio between CP and IP and between IP and SP are 0.95 and 0.65, respectively. The SP type has the highest (Na+K)/Al and includes a normative sodic plagioclase component, and is the most abundant in all chondrites. IP type, which contains a normative intermediate plagioclase component, is subordinate. CP type, which has low (Na+K)/Al and includes a normative calcic plagioclase component, is rare. This classification is used in this paper.

4. Comparison of Chondrule Composition

Y-691 chondrules which contain more than 1 wt% of $(Na_2O+K_2O+Al_2O_3)$ are plotted in Fig. 1. Those which contain less than 1 wt% are not shown because of large errors in measurement. As shown in Fig. 1, the most abundant type in Y-691 is SP, and the CP type is rare. The overall distribution of Y-691 chondrule types (Fig. 1) is nearly the same as that of chondrule types in most UOC's (IKEDA, 1983a). Figure 2 shows the $(CaO+AlO_{1.5})/(CaO+AlO_{1.5}+SiO_2)$ ratio vs. SiO₂/(SiO₂+







Fig. 2. Chemical compositions (in molar ratio of the oxides) of chondrules (open circles) in Y-691 (a), ALH-76004 (LL3) and Y-74191 (L3) (b), and ALH-77003 (CO3) (c). In (a), open squares are unusual inclusions, and solid triangles are the Y-691 whole rock composition with and without the CaS component (SHIMA and SHIMA, 1975). Solid circles in (a) and (b) are an average composition of ordinary chondrites (HARAMURA et al., 1983). In (c) are shown two compositional ranges of fine-grained CAI's (fCAI) and amoeboid olivine inclusions (AOI) in ALH-77003 (IKEDA, 1982). Equilibrium condensation trend is shown by solid squares and dashed line (see the caption of Fig. 7).



Fig. 3. Chemical compositions (in molar ratio of the oxides) of chondrules of IP type (solid circles) and SP type (open circles) in Y-691 (a), ALH-76004 (LL3) (b), and ALH-77003 (CO3) and Y-790992 (CO3) (c). Open stars are solar system elementary abundances (CAMERON, 1973).

MgO) ratio of chondrules and inclusions in Y-691, in two UOC's and in one C3. The $SiO_2/(SiO_2+MgO)$ ratio ranges from 0.5 to 0.6 for most Y-691 chondrules, and from 0.4 to 0.6 uniformly for most C3 chondrules. In UOC's, they are intermediate between the former two. Therefore, the $SiO_2/(SiO_2+MgO)$ ratio of chondrules is highest in Y-691 and lowest in C3. On the other hand, the $(CaO+AlO_{1.5})/(CaO+AlO_{1.5}+SiO_2)$ ratio of Y-691 chondrules ranges from 0.0 to 0.1 and is lowest. C3 chondrules range from 0.1 to 0.2 and are highest, while that of UOC chondrules ranges from 0.05 to 0.15 and is intermediate.

Figure 3 shows the CaO/(CaO+AlO_{1.5}) ratio of SP and IP chondrules in Y-691 with the data of UOC and C3 chondrules. The ratios of SP and IP chondrules in UOC and C3 chondrites are similar to each other and almost the same as that of solar system elemental abundances. On the other hand, the CaO/(CaO+AlO_{1.5}) ratio of SP chondrules in Y-691 is lower than that of solar system abundances although that of IP chondrules in Y-691 is nearly equal to the latter.

The MnO/MgO wt% ratios of IP and SP chondrules in an UOC are lower and higher, respectively, than that of solar system abundances (Fig. 4). On the other hand, those of both IP and SP chondrules in Y-691 are lower than that of solar system abundances.

The FeO and Cr_2O_3 contents of chondrules and inclusions in Y-691 are plotted in Fig. 5 along with the compositional ranges of chondrules and amoeboid olivine inclusions (AOI's, hereafter) in C3 chondrites. Most chondrules in Y-691 are plotted in the magnesian portion of the compositional range of C3 chondrules, although some chondrules contain fairly large amounts of FeO and Cr_2O_3 (Fig. 5). For example,



Fig. 4. MnO and MgO wt% of IP (solid circles) and SP (open circles) chondrules in Y-691 (a) and Y-74191 (L3) (b). A star in (a) is the Y-691 whole rock composition (SHIMA and SHIMA 1975).



Fig. 5. Cr_2O_3 and FeO wt% of chondrules and inclusions in Y-691. Unusual inclusions and barred-Ol-Px chondrules are shown by open squares and solid circles, respectively. Compositional ranges of chondrules (CHOND) in Y-790992 (CO3) (IKEDA, 1983a) and amoeboid olivine inclusions (AOI) in Y-790992 and ALH-77003 (CO3) (IKEDA, 1982) are shown for reference. Inset shows three hypothetical condensation trends under reduced (a), moderately reduced (b) and oxidized (c) conditions (see text).





a radial-Px chondrule (No. 154) contains 10.7 wt% FeO and 1.6 wt% Cr_2O_3 (IKEDA, 1988a). Most of the unusual inclusions in Y-691 are plotted in the ferroan range of the C3 chondrule range or in the range of AOI's. Barred-Ol-Px chondrules seem to be intermediate between the magnesian chondrules and the unusual inclusions.

The Al/(Al+Na+K) ratio of chondrules which contain more than 1 wt% of $(Na_2O+K_2O+Al_2O_3)$ is plotted against the MgO/(MgO+FeO) ratio (mg value, hereafter) in Fig. 6. This shows that most radial-Px chondrules and porphyritic or granular chondrules are magnesian and their mg values are higher than 0.95, with a few exceptions. The Al/(Al+Na+K) ratio of most radial-Px chondrules is low in comparison with other textural types of chondrules. Most radial-Px chondrules belong to the SP type, and their Al/(Al+Na+K) ratio clusters around the region lower than 0.57 of solar system abundances. Porphyritic or granular chondrules

show a wide range in Al/(Al+Na+K) and are distributed among the SP and IP types in nearly equal amounts (Fig. 6). In contrast, barred-Ol-Px chondrules differ in chemical composition from radial-Px and porphyritic or granular types. They are higher in Al/(Al+Na+K), belonging to CP or IP types, and are slightly lower in mg values, on the whole, than radial-Px and porphyritic or granular types. Spherulitic and massive cryptocrystalline chondrules are similar to one another in composition and show a wide compositional variation (Fig. 6), covering the whole range of other textural types of chondrules. Unusual inclusions in Y-691 display a wide range in mg value, from 0.99 to 0.83 (Fig. 6). The mg value of an unusual inclusion (No. 172) is extremely high (mg=0.99), and it contains a large amount of alkalis (Na₂O=3 wt%, K₂O=0.4 wt%) (IKEDA, 1988a).

The chemical compositions of the various textural types of Y-691 chondrules are plotted separately in an $AlO_{1.5}$ -SiO₂-(MgO+FeO) ternary diagram (Fig. 7). Radial-Px chondrules are distributed near the tie line connecting pyroxene with albite, and most are plotted within the triangle of Px-Ab-SiO₂. Porphyritic or granular chondrules are slightly poorer in silica than radial-Px chondrules, and most are plotted within the Ol-Px-Ab triangle of Fig. 7. The Al contents of porphyritic or granular chondrules are nearly the same as, or slightly higher than, those of radial-Px chondrules. Barred-Ol-Px chondrules are poorer in Si and richer in Al than radial-Px,



Fig. 7. Chemical compositions (in molar ratio of the oxides) of Y-691 chondrules; radial-Px (R), porphyritic or granular (P), cryptocrystalline (C), barred-Ol-Px (B), and unsual types (U). In the left triangle, compositional ranges of fine-grained CAI's (fCAI) and amoeboid olivine inclusions (AOI) in Allende (IKEDA, 1983b) are shown, and inclusion No. 172 in Y-691 plots in the fCAI range and other four inclusions plot in the AOI range. Inclusion No. 169 is rich in silica (near g in the figure U), and inclusion No. 300 is depleted in Al (plotted on the edge between Ol and Px). The dash line is the metal-free equilibrium condensation trend (GROSSMAN and LARIMER, 1974), and the temperatures and condensing phases at each point are as follows; a (1650 K, corundum), b (about 1600 K, corundum and melilite), c (1500 K, melilite and spinel), d (1450 K, spinel and melilite), g (1250 K, enstatite, forsterite, anorthite and diopside), h (in Fig. 6; about 1000 K, enstatite, forsterite, diopside and sodic plagioclase). A fractional condensation trend g-k may be produced by fractionation of high temperature condensates corresponding to f.

porphyritic and granular types. Spherulitic and massive cryptocrystalline chondrules cover the compositional range of all other textural types also in Fig. 7. Several unusual inclusions are also shown in Fig. 7. One (No. 172), exceptionally high in Al, plots in the compositional range of fine-grained CAI's in C3 chondrites. Four other unusual inclusions are on the tie line connecting olivine with anorthite, and are in the compositional range of AOI's in C3's.

5. Discussion

The chemical compositions of chondrules are controlled by three factors; (I) chemical compositions of chondrule precursors, (II) evaporation loss during heating events that formed the chondrule melt droplets, and (III) reactions of the chondrules either in the nebula or in the parent body, after their solidification.

IKEDA (1983a) showed that the silicates of most chondrule precursors consisted mainly of olivine, pyroxene and plagioclase and that most of Na and K were preserved durring the heating events which formed the chondrule melts. This means that the second factor (II) did not play an important role in the modification of the major element content of chondrules.

On the other hand, the third factor (III) sometimes plays an important role in the equilibrated O chondrites and in the C2 and C3 chondrites. The chemical compositions of chondrules in the equilibrated O chondrites were modified during metamorphism in the parent body. In some C3 chondrites, ferrous- and alkali-zoning is observed in chondrule groundmasses. This was produced by reactions with an oxidized nebular gas after the solidification of some chondrules (IKEDA, 1982). Other C chondrites include some chondrules which have been subjected to subsolidus alteration to varing degrees. They usually contain the hydrous minerals found in these chondrules (IKEDA, 1983c). However, alkali-zoned chondrules or hydrous-mineralbearing chondrules are not known in the UOC and unequilibrated E chondrites, including Y-691. In Y-691, FeO-bearing olivine in most of unusual inclusions and barred-Ol-Px chondrules has suffered subsolidus reduction by reactions with a reduced nebular gas which has decomposed the ferroan olivine (IKEDA, 1988b, 1989a). In addition, fassaitic pyroxene and nepheline in some FeO-bearing unusual inclusions were changed to albite by the introduction of Na₂O and/or SiO₂ from nebular gases (IKEDA, 1988b). These indicate that FeO-bearing inclusions and barred-Ol-Px chondrules in Y-691 have experienced chemical modifications after their solidification. However, the main change in chemical composition for these FeO-bearing objects in Y-691 seems to be restricted to oxygen, alkalis, and silica (IKEDA, 1988b, 1989a). The silica content which was introduced into FeO-bearing inclusions and chondrules seems to be subordinate in comparison with their original silica content (IKEDA, 1988b, 1989a). Therefore, the third factor (III) may not be important for most magnesian chondrules in Y-691 and even for the FeO-bearing objects, except for oxygen and alkalis.

The discussion above supports factor one (I), that the major element composition of chondrule precursors is essentially the same as those of chondrules and inclusions in Y-691, except for oxygen and alkalis in the FeO-bearing objects. The

following discussion is based on that factor I is the most important.

5.1. Precursors of chondrules and inclusions

Figure 1 indicates that Y-691 chondrules are plotted within the solubility limit of the orthoclase component of plagioclase at a temperature of about 1200 K, suggesting that the precursors of Y-691 chondrules contained the Al, Na, and K contents found in the plagioclases.

Figure 7 indicates that the precursors of most radial-Px chondrules consisted mainly of low-Ca pyroxene with small amounts of sodic plagioclase and silica minerals. The precursors of some radial-Px chondrules, however, included a small amount of olivine instead of silica. Precursors of most porphyritic or granular chondrules consisted mainly of pyroxene with minor amounts of olivine and plagioclase. Finally, the precursors of barred-Ol-Px chondrules included abundant olivine in addition to pyroxene and plagioclase.

An unusual inclusion (No. 172) has a quenched texture, with pyroxene laths in glass, and its chemical composition (Fig. 7) suggests that the precursor was formed as a high temperature condensate in a manner similar to that of the fine-grained CAI's. After condensation, it lost its original texture of a fine-grained CAI probably by impact, to form the silicate melt which crystallized as glassy material including small quenched laths of pyroxene (IKEDA, 1988b). Four other unusual inclusions in Y-691 are similar in chemical composition to AOI's in C3 chondrites (Fig. 7). Ferroan olivine of matrices and AOI's in C3 chondrites are considered to have formed as high temperature condensates under oxidized conditions (PALME and FEGLEY, 1987), and the precursors of FeO-bearing unusual inclusions in Y-691 may have formed under similar oxidized conditions. The unusual inclusions show igneous textures (IKEDA, 1988b), but the original amoeboid olivine texture has disappeared probably by later impact events, to form the melts which resulted in aggregates consisting mainly of ferroan olivine and fassaitic pyroxene. After recrystallization they reacted with an oxidized gas at low temperatures to produce ferroan augite and plagioclase from the fassaitic pyroxenes. Finally, the inclusions were driven into a reduced nebular gas, and the ferroan olivine decomposed, by reaction with the reduced gas, to produce enstatite and Fe-metal in the inclusions (IKEDA, 1988b). Another inclusion (No. 167) is rich in silica, and similar in chemical composition to barred-Ol-Px chondrules (Fig. 7). It was originally a barred-Ol-Px chondrule which suffered later reduction by a reduced nebular gas (IKEDA, 1988b). An inclusion (No. 300) is depleted in Al (Fig. 7), indicating that the precursor consisted mainly of olivine and pyroxene with a negligible amount of plagioclase.

5.2. Chondrule compositions and whole rock composition

Chondrules are the main constituents of most chondrites, except for CI and CM chondrites, and the average major element compositions of chondrules should be similar to the whole rock compositions except for the siderophile and chalcophile elements.

The Al/(Al+Na+K) ratio of whole rock Y-691 is about 0.506 (SHIMA and SHIMA, 1975), although Y-691 chondrules range from 0.48 to 0.98 (Figs. 1 and 6). The

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largest and most abundant chondrule type in Y-691 is radial-Px, and the ratio of Al/ (Al+Na+K) in these chondrules is mostly from 0.51 to 0.53 (Fig. 6). Therefore, the average ratio of all chondrules seems to be slightly higher than that of the whole rock. The reason for this discrepancy is that whole rock Y-691 includes alkali- or Al-bearing phases outside chondrules. They occur in opaque-mineral nodules and are mainly albite, caswellsilverite, roedderite and djerfisherite (OKADA, 1975; EL GORESY et al., 1988; IKEDA, 1989b). Albite in opaque-mineral nodules is almost pure NaSi₈AlO₃, and the other alkali-bearing phases are nearly free from Al, except for minor Al in roedderite (EL GORESY et al., 1988; IKEDA, 1989b). Therefore, the Al/ (Al+Na+K) ratio for alkali-bearing phases other than chondrules ranges from 0.0 to 0.5. The existence of the alkali-bearing phases other than chondrules explains the discrepancy in Al/(Al+Na+K) between the whole rock and the chondrule average. This discrepancy suggests that fractional condensation of silicates prevailed during the formation of the chondrule precursors of Y-691. After the formation and separation of the chondrule precursors from the nebular gas, the alkalis remaining as nebular gas components reacted with Fe-metal and/or sulfide, instead of Al-bearing silicates, to produce alkali-bearing sulfides such as caswellsilverite and djerfisherite (IKEDA, 1989b).

The CaO/(CaO+AlO_{1.5}) ratio for whole rock Y-691 is 0.46, which coincides with that of IP chondrules in Y-691 (Fig. 3a). However, SP chondrules have a lower ratio than that of the whole rock. For Y-691, the Al content of SP chondrules is nearly the same as, or slightly lower than, that of IP chondrules on the whole (IKEDA, 1988a). The low CaO/(CaO+AlO_{1.5}) ratio of SP chondrules in Y-691 is due to the depletion of Ca in the SP chondrules. This depletion may be explained by the following idea: during condensation of the precursors of SP chondrules in a nebular gas, oldhamite was produced, and then separated, to form opaque-mineral nodules with Fe-metal and other sulfides (IKEDA, 1989b) so that the precursors of SP chondrules were depleted in Ca prior to the formation of these chondrules. If so, the condensation and fractionation of oldhamite should have taken place at the same time that condensation of alkalis formed the alkali-rich and Ca-poor precursors of SP chondrules in Y-691.

The $(CaO+Al_{1.5})/(CaO+Al_{1.5}+SiO_2)$ ratio for whole rock Y-691 is 0.082 (SHIMA and SHIMA, 1975), whereas the ratio of most chondrules in Y-691 ranges uniformly 0.0 to 0.1 (Fig. 2). This discrepancy may be partly due to the depletion of Ca by the formation of oldhamite outside of the chondrules in Y-691.

The MnO/MgO wt% ratio of whole rock Y-691 is about 0.013 (SHIMA and SHIMA 1975), which is too high to coincide with those of chondrules (Fig. 4). The main Mn-bearing phases in Y-691 outside of chondrules are niningerite and sphalerite (EL GORESY *et al.*, 1988; IKEDA, 1989b). Niningerite is modally more abundant than sphalerite and contains 3 to 4 wt% Mn. In Y-691 most niningerite occurs outside of chondrules, although minor niningerites are present rarely inside chondrules (IKEDA, 1989b). These facts suggest that the depletion of MnO in Y-691 chondrules (Fig. 4) may be due to the formation and separation of MnS components in niningerites, and the precursors of IP and SP chondrules were depleted in Mn prior to the formation of the chondrules.

It is well known that whole rock compositions differ between E, O, and C chondrite groups (WASSON, 1974; LARIMER and ANDERS, 1967; KALLEMEYN and WASSON, 1981). The atomic ratios of Al/Si, Ca/Si, and Mg/Si for the E chondrite group are the lowest, and those for the C chondrite group are the highest. Whole chondrule compositions in Y-691 are depleted in refractory lithophiles (Ca and Al) and Mg in comparison to chondrules in C3 chondrites (Fig. 2). Chondrules in UOC's are intermediate between chondrules of Y-691 and the C3 chondrites. This indicates that the differences in major refractory lithophile element compositions between E, O, and C chondrite groups correspond to that of the chondrules. The depletion of the refractory lithophiles in chondrules of E and O chondrites may be due to fractional condensation which took place in nebular gases to form the chondrule precursors which were depleted in high temperature condensates. The degree of fractional condensation for E chondrites was more intense than for O chondrites.

WARK (1983) showed, by calculation, that the whole rock composition of E chondrites can be produced by the subtraction of CAI and other high temperature components from CI starting material. BISCHOFF *et al.* (1985) found a small amount of high temperature condensates including perovskite, hibonite and spinel in unequilibrated E chondrites. In addition to these, the existence of unusual inclusions in Y-691 which are equivalent in chemical composition to fine-grained CAI's and AOI's supports the concept that the precursors of Y-691 chondrules were produced by intense fractional condensation.

5.3. Fractional condensation trends

In Figs. 2, 6, and 7, an equilibrium condensation trend is shown for reference. This trend was calculated using the data of GROSSMAN and LARIMER (1974), on a metal-free basis, and shows the chemical composition of metal-free materials condensed in equilibrium from a nebular gas having solar system abundances under a total gas pressure of 10⁻³ atm. The equilibrium condensation trend from a to g in Figs. 6 and 7 corresponds to condensates from high to low temperatures. At low temperatures, 400–600 K, metallic Fe is oxidized to FeO which forms fayalite and ferrosilite components in olivine and pyroxene at the points from g to i in Figs. 6 and 7. Alkalis condense at temperatures from 1200 to 800 K as albite or orthoclase components in plagioclase prior to the introduction of FeO into the condensates (GROSSMAN and LARIMER, 1974; FEGLEY and LEWIS, 1980; SAXENA and ERIKSSON, 1983), as shown by the trend g-h in Fig. 6. However, if fractionation of high temperature components such as forsterite and fassaite takes place prior to the condensation of FeO, the fractional condensation trend should be from h to j in Fig. 6 or from g to k in Fig. 7, because of the fractionation of Si-poor and Na-free high temperature condensates.

Major amounts of Ca, Al, Si and Mg in the nebular gas condense at temperatures higher than 1200 K mainly as olivine, pyroxene and plagioclase (GROSSMAN and LARIMER, 1974). Therefore, the composition of the condensates at 1200 K equals that of solar system abundances. As shown in Fig. 2, C3 chondrules correspond, as a whole, to condensates at temperatures of about 1200 K. On the other hand, the $(CaO+AlO_{1.5})/(CaO+AlO_{1.5}+SiO_2)$ ratio of Y-691 chondrules is lower, as a whole, than that in C3 chondrites (Fig. 2). This can be explained by fractional condensation: The precursors of Y-691 chondrules formed by separation of high temperature components from a gas having solar system abundances.

The low ratios of Al/(Al+Na+K) for most magnesian radial-Px chondrules (Fig. 6a) may be explained by a hypothesis that the precursors of most magnesian radial-Px chondrules were formed from fractional condensates as shown by the trend h-j shown in Fig. 6a. Considering their silica-rich nature (Fig. 7) and the occurrence of transparent-SiO₂ chondrules in Y-691 (IKEDA, 1989b), it is expected that a silica mineral should condense from a nebular gas as a component of precursors of the silica-rich chondrules in Y-691. The condensation of a silica mineral is also explained by fractional condensation with separation of high temperature condensates such as forsterite and fassaite, under reducing conditions, shown by the trend g-k in Fig. 7. The reduced condition prevents oxidation of metals to form FeO which could become fayalite and ferrosilite components by consuming SiO₂ components.

Porphyritic or granular chondrules in Y-691 show continuity in chemical composition with magnesian radial-Px chondrules, as shown in Figs. 6 and 7. Their precursors may form in a similar manner to those of magnesian radial-Px chondrules, although the former include more high temperature components than the latter.

Barred-Ol-Px chondrules plot on the condensation trend between f and g in Fig. 7, indicating that their precursors were higher temperature condensates in comparison to those of magnesian radial-Px chondrules and porphyritic or granular chondrules.

5.4. Oxidation and reduction conditions

Recently, ubiquitous occurrences of FeO-bearing objects in E3 were reported (RAMBALDI *et al.*, 1984; NAGAHARA, 1985; PRINZ *et al.*, 1985; LUSBY *et al.*, 1987; IKEDA, 1988b, 1989a), and multi-stage origins for E chondrites were discussed, in terms of oxidation and reduction conditions.

The nebular gas having solar system abundances condenses Fe and Cr as metals by cooling, and then they are oxidized in a low temperature range to form FeO and Cr_2O_3 components in silicates or oxides. If all Cr and a part of the Fe were oxidized, the FeO and Cr_2O_3 contents of low-temperature silicate-oxide condensates would increase in the manner schematically indicated by arrow (b) in the inset of Fig. 5. On the other hand, FeO and Cr_2O_3 contents in silicate-oxide condensates would not increase under extremely reduced conditions, because most Fe and Cr reside as metals and sulfides even in low temperature condensates, resulting in arrow (a) in the inset of Fig. 5. If condensation takes place under intensely oxidized conditions, Fe would condense directly as FeO even in a high temperature range (PALME and FEGLEY, 1987), and Cr_2O_3 would be diluted by the large amounts of FeO in silicate-oxide condensates, resulting in arrow (c) of the inset of Fig. 5.

As shown in Fig. 5, most chondrules in Y-691 are poor in FeO with a moderate amount of Cr_2O_3 , suggesting that most chondrules in Y-691 were produced from precursors which formed under reduced conditions where most Fe and a fraction of the Cr were metallic or sulfide components.

Barred-Ol-Px chondrules include moderate amounts of FeO and Cr_2O_3 , and their mg values are lower than those of the other textural types of chondrules in spite

of the fact that their chemical compositions correspond to higher temperature condensates (Fig. 7). The low mg values of barred-Ol-Px chondrules suggest that their precursors condensed from nebular gas under more oxidized conditions in comparison with the reduced conditions for the other textural types of chondrules.

Although unusual inclusion (No. 172) is nearly free of FeO and Cr_2O_3 , most other unusual inclusions have high contents of FeO, similar to those of AOI's in C3 chondrites (Fig. 5). As shown in Figs. 2 and 7, the precursors correspond to a condensate at temperatures higher than those of barred-OI-Px chondrules, although the former shows lower mg values than the latter. The lower mg values of the inclusions suggest that the condition to produce the precursors was more oxidized than those for the precursors of barred-OI-Px chondrules.

JOHNSON (1986) presented a new equation showing H_2/H_2O molecular ratio of a nebular gas which coexists with olivine, low-Ca pyroxene and Fe-metal. On the assumption that the Fe/(Fe+Mg) atomic ratio of olivines in the chondrule precursors is similar to that of the bulk chondrule, we can obtain the H_2/H_2O ratios of the nebular gases which condensed the precursors of various chondrules.

Magnesian radial-Px chondrules, the main type in Y-691, show representative mg values of about 0.97–0.98 (Fig. 6) and they include small amounts of Fe-metal and sometimes normative magnesian olivine. Their Al/(Al+Na+K) atomic ratio is about 0.51–0.53, on average, suggesting that the precursors of magnesian radial-Px chondrules formed during and/or after the condensation of alkalis. This means that the formation temperatures of their precursors were lower than 1200 K, because the condensation of alkalis takes place mainly in the temperature range from 1200 K to 800 K (GROSSMAN and LARIMER, 1974; FEGLEY and LEWIS, 1980; SAXENA and ERIKSSON, 1983). Thus, the H₂/H₂O molecular ratios of a nebular gas, which condensed the precursors of magnesian radial-Px chondrules with mg=0.98, are 39 at the temperature of 1200 K, 79 at 1000 K, 226 at 800 K, 1320 at 600 K, and 44800 at 400 K.

Barred-Ol-Px chondrules range in mg value from 0.85 to 0.98 although they are poor in alkalis in comparison with magnesian radial-Px chondrules. The mg value of barred-Ol-Px chondrule No. 264 is about 0.94, and the Al/(Al+Na+K) ratio is extremely high (0.97), suggesting that the formation temperature of the precursor may be higher than 800 K. The above-stated equation gives the H₂/H₂O ratio of 13 at 1200 K, 26 at 1000 K, and 75 at 800 K.

An unusual inclusion (No. 152) includes ferroan olivine, fassaitic to pigeonitic pyroxene, and intermediate plagioclase. According to IKEDA (1988b), the inclusion consisted originally of ferroan olivine and ferroan fassaite, and after the solidification, the inclusion reacted with an oxidized gas to introduce alkalis into the inclusion. This means that the precursors of the inclusion contained FeO components prior to the condensation of alkalis. Considering that alkalis condense in the temperature range from 1200 to 800 K under a total gas pressure of 10^{-4} atm in spite of the redox state (LATTIMER *et al.*, 1978) and that FeO can condense at temperatures higher than the temperature range of alkali condensation from an extremely oxidized gas which has a H_2/H_2O molecular ratio smaller than 10 (PALME and FEGLEY, 1987), the precursor of inclusion No. 152 may have formed from an oxidized gas with an H_2/H_2O ratio

smaller than 10 at temperatures higher than 800 K.

The H/O atomic ratio for solar system abundances is 1480 (CAMERON, 1973), and the main O-bearing gas species is CO at high temperatures and H_2O at low temperatures (GROSSMAN and LAIMER, 1974), indicating that the H_2/H_2O ratios of the solar gas are larger than about 740 in all temperature ranges. The H_2/H_2O molecular ratios estimated for barred-Ol-Px chondrule (No. 264) and unusual inclusion (No. 152) are very low, suggesting that their precursors condensed from oxidized gases with H/O ratios much lower than that for solar system abundances. The low H_2/H_2O gases might be produced by evaporation of ice and/or rocks, namely chondritic or cometary materials (IKEDA and KIMURA, 1985). On the other hand, the precursors of magnesian radial-Px chondrules could condense from a reduced gas similar to the solar system abundances in a low temperature range or from extremely reduced gases of much higher H/O ratios.

5.5. A scenario

It is a most important fact for the origin of unequilibrated E chondrites that the chondrules were depleted in refractory lithophile elements and that the redox states of the precursors of chondrules and inclusions varied. Thus, a scenario for their origin must take into account the depletion of refractory components in addition to the redox states.

In order to explain the depletion of refractory lithophile elements in chondrule precursors and the various redox states of nebular gases, a hypothesis is proposed here with the assumption that various redox states are controlled by the H/O atomic ratios of nebular gases. Consider the case where many comets (ice and rocks), with a bulk chemical composition similar to that of solar system abundances (except for lowtemperature gas components such as H₂ and He) sedimented at the ecliptic plane of the solar nebula, as shown in Fig. 8A. Then, evaporation took place in the solar nebula by heating events which may have been caused by extrasolar explosions. A low-H/O gas which was enriched in ice and rock components, in comparison with H_2 and He components, was produced along the ecliptic plane where evaporation residues could remain (Fig. 8B), although the gas far from the ecliptic plane was depleted in those components. Diffusion of the low-H/O gas took place from the ecliptic plane in a direction perpendicular to the plane (Fig. 8C). With decreasing temperature in the nebula, refractory lithophile components condensed near the ecliptic plane under highly oxidized conditions. The oxidized gas, which fractionated the high temperature condensates to various degrees, diffused and expanded into the surrounding reduced gas rich in H_2 and He components, where fractional condensation took place in the low temperature range. With a further decrease of temperature, low temperature condensates having low- and high-mg values formed from the oxidized gas along the ecliptic plane and from the reduced gas far from the plane, respectively (Fig. 8D).

The condensates along the ecliptic plane may have been rich in high temperature components and included abundant FeO, resulting in refractory lithophile-rich and low-mg materials. They may have included various kinds of materials similar to CAI's and AOI's in C3 chondrites. On the other hand, the condensates which formed



far from the ecliptic plane may have been poor in high temperature components and FeO, resulting in refractory lithophile-poor and high-mg materials. They may be the precursors of magnesian chondrules in Y-691. Si-bearing Fe metals and sulfides such as CaS and MgS in Y-691 may have formed under the reduced conditions.

Either after, or at the same time as, the condensation, many planetesimals formed and collided with each other to produce many silicate-melt droplets (KIEFFER, 1975) which crystallized as chondrules in the cool nebular gases. Unusual inclusions were produced as fragments of melted fine-grained CAI's or melted AOI's near the ecliptic plane. After the formation of chondrules, FeO-rich inclusions which formed under oxidized conditions were expelled into the reduced gas and mixed by a turbulent motion with magnesian chondrules and opaque-mineral nodules, where ferroan olivine in the FeO-rich inclusions and barred-OI-Px chondrules decomposed by reaction with the reduced gases. After the mixing, accretion took place to form the parent body of the Y-691 enstatite chondrite.

6. Conclusion

1) Bulk chemical compositions of chondrules in E, O, and C chondrite groups differ from one another in the contents of refractory lithophile elements, and the differences correspond to that of the whole rocks.

2) The low Ca and Mn contents of SP chondrules in Y-691, in comparison with IP chondrules, may be due to the separation of oldhamite and niningerite from the precursors of SP chondrules.

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3) The main chondrule types in Y-691, magnesian radial-Px chondrules and magnesian porphyritic to granular chondrules, are richer in SiO_2 and poorer in FeO and refractory lithophile elements than those in UOC's and C3 chondrites. They were produced by fractional condensation from a reduced nebular gas.

4) An unusual inclusion (No. 172) has a chemical composition similar to that of fine-grained CAI's in C3 chondrites and might have been produced originally as a high temperature condensate. Most other unusual inclusions have compositions similar to those of AOI's in C3 chondrites and may have been produced as high temperature condensates under oxidized conditions. Barred-Ol-Px chondrules may have been produced from precursors which were intermediate-temperature condensates, under moderately oxidized conditions.

5) Unusual inclusions and barred-Ol-Px chondrules may have been expelled into a highly reduced gas, where the decomposition of ferroan olivines took place by the reaction with the reduced gas.

6) After heating events which took place in the solar nebula, the region near the ecliptic plane became low in H/O where some unusual inclusions similar to finegrained CAI's and AOI's in C3 chondrites were produced, and the region far from the ecliptic plane in perpendicular direction may have had high H/O ratios where most components of unequilibrated E chondrites, such as magnesian chondrules and opaque mineral nodules, were produced.

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