ORIGIN AND EVOLUTION OF CHONDRULES BASED ON Na/Al-DISPERSION AND -CONVERGENCE IN YAMATO-74 ORDINARY CHONDRITES

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Abstract: Na/Al convergence has been observed in groundmass of chondrules and matrix from seven Yamato-74 (H, L, 4-5) chondrites. On the other hand, Na/Al dispersion has prevailed in groundmass of chondrules and matrix from Yamato-74191 (L3) chondrite. The results suggest that: (1) Primordial liquid droplets as precursors of the chondrules might be derived from a rapidly cooling solar nebula by fractional condensation. (2) The hot liquid droplets thus formed might be cooled instantaneously by radiation loss in the cooling nebula. (3) The chondrules as quenched products might be subjected to mutual collision during accumulation in the cold nebula. (4) Planetesimals accumulated by the chondrules might be transferred from the place of birth to an unknown hot region in the proto solar nebula.

1. Introduction

During the course of a classification work of Yamato-74 chondritic meteorites (NISHIDA *et al.*, 1979) collected around the Yamato Mountains, East Antarctica (YANAI, 1978), we noticed a peculiar regularity in chemical compositions of "groundmass" in the Yamato-74 ordinary chondrites.

"Groundmass" is defined, here, as a fine-grained portion other than coarsegrained mineral phases (olivine, pyroxenes, Fe-Ni metals and troilite) in matrix of chondritic meteorites. The latter coarse-grained minerals can be defined as "phenocrysts". Similarly, a glassy portion of a single chondrule and coarsegrained olivine and pyroxene embedded in the glassy portion are defined as "groundmass" and "phenocrysts", respectively. The peculiar regularity we noticed is a concomitant phenomenon of Na and Al contents in the groundmass of several Yamato-74 ordinary chondrites. These elements have different chemical properties from each other (Na⁺; 1.02Å, volatile: Al³⁺; 0.53Å, non-volatile), yet the Na-Al correlation can be recognized in the groundmass. We also noticed that a Yamato-74 chondrite does not show the Na-Al correlation.

In this paper, we report the Na-Al correlation and the Na/Al dispersion, along with their cosmochemical implications.

2. Experimental

Approximately 0.05–0.1 g fragments without fusion crust were used to make thin sections for microprobe analysis. The chemical analyses of the sections were carried out with a computer-controlled electron probe X-ray micro-analyzer at Chemical Analysis Center of the University of Tsukuba. The analyzer consists of a JEOL JXA-50A electron probe X-ray micro-analyzer with three channels of spectrometer and an automatic controlled and processing system by ELIONIX. The quantitative analysis of the groundmass was made by a program introducing the corrections to calculate concentrations by a modified version of the Bence-Albee method (1968), under the conditions of accelerating voltage 15 kV, probe diameter $\sim 5 \ \mu m$ and sampling time 20 s. The analytical results fall within the range of 96 to 101%.

3. Results and Discussion

3.1. Na-Al correlation in chondrules and matrix of moderately metamorphosed ordinary chondrites

Fig. 1 shows an example of the Na-Al correlation found in Yamato-74001



Fig. 1. Na-Al correlation in groundmass of a chondrule and matrix in Yamato-74001 (H-5) chondrite (Open circles, matrix; Closed circles, chondrule).

(H4–5) chondrite. Open circles are obtained from groundmass of matrix and closed circles are from groundmass of a single chondrule. The Na contents in the groundmass are increasing regularly with increasing Al contents up to a point of Na₂O:10% and Al₂O₃:21%. There is no difference in the compositional variation between the chondrule and the matrix. The Na-Al correlation has also been observed in bulk compositions of Bjurbole (L4) chondrules (WALTER, 1969).

The Na-Al correlation holds in the groundmass of chondrules and matrix of four H-group chondrites [Y-74001 (H4–5), Y-74193 (H4–5), Y-74498 (H4–5), Y-74507 (H4–5)], and of three L-group chondrites [Y-74053 (L5–6), Y-74454 (L5–6), Y-74603 (L4–5)]. All of the data from the seven Yamato chondrites are plotted in Fig. 2. Open circles correspond to the H group chondrites and closed circles to L-group chondrites. Any appreciable difference cannot be recognized in the Na-Al correlation between H-group and L-group chondrites.

The Na-Al correlation in the fine-grained groundmass of the moderately metamorphosed chondrites might be explained phenomenologically by a twocomponent model. One component is microcrystalline aggregates of Na, Al-rich plagioclase ($<10 \mu$ m) in the groundmass (VAN SCHMUS and WOOD, 1967). The other component is fine-grained Na, Al-poor mineral phases such as olivine and pyroxene crystals in the groundmass. Both of the components in the groundmass make the Na-Al correlation as a mixing line. WALTER (1969) has also stressed an important role of the composition of plagioclase for the Na-Al correlation in the Bjurbole chondrules.

The Na-Al correlation in the moderately metamorphosed chondrites makes a linear band which attains to sodic plagioclase with compositional range of Ab_{95-70} , as shown in Fig. 2. On the other hand, the strongly metamorphosed



Fig. 2. Na-Al correlation in groundmass of chondrules and matrix in seven Yamato-74 ordinary chondrites [Open circles, Yamato-74001 (H4-5), -74193 (H4-5), -74498 (H4-5) and -74507 (H4-5); Closed circles, Yamato-74053 (L5-6), -74454 (L5-6) and -74603 (L4-5)].

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(type 6) chondrites contain coarse-grained (>20 μ m), well-recrystallized plagioclase with homogeneous compositions (VAN SCHMUS and WOOD, 1976). The compositional range of the plagioclase crystals is Ab_{79,2-83.1} for H6 chondrites and Ab_{83.4-85.0} for L6 chondrites (VAN SCHMUS and RIBBE, 1968). The wide range Ab₇₀₋₉₅ of the type 4–5 groundmass compared with the narrow range Ab₇₉₋₈₅ of the type 6 plagioclase might be explained by difference of degree of metamorphism. The stronger the degree of metamorphism, the narrower the Na-Al correlation band. Thus, Fig. 2 suggests that metamorphism is operative for establishment of the Na-Al correlation.

3.2. Breakdown of Na-Al correlation in chondrules of an unmetamorphosed chondrite

The Na-Al correlation found in the moderately metamorphosed Yamato (H, L, 4–5) chondrites does not hold in an unmetamorphosed Yamato chondrite. Fig. 3 shows the Na and Al contents of glassy groundmass in 18 chondrules of Yamato-74191 (L3) chondrite (open circles). The right edge of a triangle drawn in Fig. 3 indicates compositional variation for idealized plagioclase solid solution and the left edge of the triangle is just drawn to connect the starting point with the pure albite composition. Note that all of the data so far obtained from the chondrules in the unmetamorphosed chondrite fall within the triangle with one exception. The existence of the exceptional point is supported by another unmetamorphosed chondrite (closed circles). The compositions of glasses in Mezö-Madaras (L3) chondrules fall around the exceptional point (KURAT, 1967;



Fig. 3. Na and Al contents in glassy groundmass of 18 individual chondrules in Yamato-74191 (L3) chondrite [Open circles, Yamato-74191 (L3) chondrite; Closed circles, Mezö-Madaras (L3) chondrite after KURAT (1967) and VAN SCHMUS (1967)].

VAN SCHMUS, 1967).

As shown in Fig. 3, the Na_2O/Al_2O_3 ratios in glassy groundmass of the individual chondrules of Yamato-74191 (L3) chondrite disperse from 0 to 0.61. The similar dispersion of the Na/Al ratios has been observed in bulk compositions of Chainpur (LL3) chondrules (WALTER and DODD, 1972). Fig. 3 suggests that liquid droplets as precursor of the chondrules are formed with originally different Na/Al ratios, and that the Na-Al correlation in the metamorphosed chondrites (Fig. 2) is established by secondary process.

Fig. 4 shows two histograms of the Na/Al atomic ratios from the groundmass in chondrules of Yamato-74191 (L3) chondrite (Fig. 4a) and from the groundmass in chondrules and matrix of the seven metamorphosed Yamato-74 (H, L, 4-5) chondrites (Fig. 4b). The difference between them might be explained in terms of material transfer among the primordial chondrules by thermal metamorphism, as previously pointed out by WALTER and DODD (1972).

Originally heterogeneous compositions tend to converge to an averaged composition by thermal metamorphism in a closed system: the wide Na/Al atomic ratios (0–1.4) (Fig. 4a) converge to narrow Na/Al atomic ratios (0.6–0.8) (Fig. 4b). The averaged Na/Al atomic ratio (\sim 0.7) is in good agreement with the cosmic Na/Al ratio (0.71) after CAMERON (1973). Thus, the thermal metamorphism might have operated in their parental bodies after accretion.



Fig. 4. Na/Al dispersion in chondrules of an unmetamorphosed Yamato-74191 (L3) chondrite (a), and Na/Al convergence in chondrule and matrix of seven metamorphosed Yamato-74 (H, L, 4–5) chondrites (b).

3.3. Chondrules and matrix of an unmetamorphosed chondrite

Fig. 5 shows the Na and Al contents in fine-grained opaque matrix in Yamato-74191 (L3) chondrite. Again, the Na-Al correlation is not observed in the unmetamorphosed matrix. It is interesting to note, however, that the Na/Al dispersion of the matrix (Fig. 5) is in good agreement with that of the chondrules (Fig. 3) in the same chondrite. This similarity suggests a close genetic relationship between them.



Fig. 5. Na/Al dispersion in matrix of unmetamorphosed Yamato-74191 (L3) chondrite.

A straightforward interpretation for the similarity is that the matrix might be derived from the chondrules as abrasion products by mutual collision during accumulation.

If chondrules are formed from a pre-existent dust material (matrix) by remelting processes such as lightning discharge (WHIPPLE, 1966) and high velocity impacts (WHIPPLE, 1972), a discordance of the Na/Al dispersion can be expected between the chondrules and the matrix. A high temperature ($\sim 2000^{\circ}$ C) heating experiment on Allende (C3) chondrite by NOTSU *et al.* (1978) revealed that Na volatilizes out within a few minutes, while Al remains in the melted residue for a long time. Therefore, the Na/Al ratio of the melted residue (chondrules) decreases, while the Na/Al ratio of the dust material (matrix) escaped from the reheating events increases with addition of Na as re-condensates.

Thus, we prefer the formation process of matrix from chondrules, rather than of chondrules from a pre-existent dust material. Inevitably, we lean toward a direct condensation model for origin of chondrules as liquid droplets from a solar nebula, originally proposed by WOOD (1963) and later improved by BLANDER and KATZ (1967). However, we do not agree with any equilibrium condensation model for the formation of the chondrules because of the Na/Al dispersion.

Vapor fractionation might be invoked as an interpretation of the Na/Al dispersion in the primordial chondrules. WALTER and DODD (1972) have claimed

this mechanism, based on an inverse correlation between SiO_2 and refractory oxides found in Chainpur (LL3) chondrules. Indeed, Si is more volatile than refractory elements such as Al, Ca and Ti at high temperature (NOTSU *et al.*, 1978). However, the inverse correlation is not considered as an evidence for vapor fractionation because of the presence of Na in the chondrules. Na volatilizes out completely before Si begins to volatilize (NOTSU *et al.*, 1978).

The Na/Al dispersion in the primordial chondrules might have been established originally by fractional condensation from a rapidly cooling solar nebula.

3.4. Crystal fractionation within original liquid droplets

Glassy groundmass of several chondrules in Yamato-74191 (L3) chondrite, sometimes, provides a clear Na-Al correlation. Two examples are shown in Fig. 6. Open circles are obtained from a porphyritic olivine chondrule and closed circles are from a porphyritic pyroxene chondrule.



Fig. 6. Na-Al correlation in glassy groundmass in two individual chondrules of Yamato-74191 (L3) chondrite (Open circles, porphyritic olivine chondrule; Closed circles, porphyritic pyroxene chondrule).

The Na-Al correlation within the individual chondrules might be explained, not by the plagioclase mixing model, but by crystal fractionation of olivine and pyroxene in the original liquid droplets. Partition coefficients of Na and Al between the ferromagnesian minerals and a melt are less than 0.5 (MATSUI *et al.*, 1977). Therefore, Na and Al do not enter into the crystallizing minerals and both are enriched in the residual liquid. The Na-Al correlation within the individual chondrules might be established by quenching of the minerals, preserving the heterogeneity of the residual liquids^{*}.

Relative proportion of crystallized minerals to residual liquid is reflected in the Na and Al contents in glass of unmetamorphosed chondrules. The less the amounts of residual liquid, the higher the Na and Al contents in the residual

^{*} Thus, a disequilibrium condensation model is much more plausible than an equilibrium condensation model for the formation of the chondrules.

liquid. The exceptional points sited outside the triangle in Fig. 3 might be derived from residual liquids by crystal fractionation in liquid droplets with originally high Na and Al contents.

4. Origin and Evolution of Chondrules

A scenario for origin and evolution of the chondrules, based on the Na/Aldispersion and -convergence, is summarized as follows:

(1) Primordial liquid droplets as precursor of the chondrules might be derived directly from a rapidly cooling solar nebula by fractional condensation. Thus, bulk compositions of the individual chondrules show low Na and Al contents with the Na/Al dispersion.

(2) The hot liquid droplets thus formed might be cooled instantaneously by radiation loss in the cooling nebula. Crystal growth in the liquid droplets might start instantaneously and might stop suddenly. Thus, chemical compositions of the glassy groundmass in the individual chondrules show heterogeneous high Na and Al contents, preserving the original Na/Al dispersion.

(3) The chondrules as quenched products might be subjected to mutual collision during accretion in a cold environment. The matrix now found in the unmetamorphosed chondrite might be derived from the chondrules as abrasion products. Thus, the Na/Al dispersion of the chondrules is in good agreement with that of the matrix.

(4) Planetesimals accumulated with the chondrules and the matrix might be transferred from the place of birth to a hot region in the proto solar nebula. Some of the planetesimals might be long-residents and others might be short-, or non-residents in the unknown hot place. Thus, the Na/Al-convergence and -dispersion are separately observed in the ordinary chondrites.

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