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Sodium-metasomatism in chondrules in CO3 chondrites: Relationship to parent body thermal metamorphism

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Abstract–We have studied the mineralogy and petrology of mesostases of 783 type I chondrules in seven CO3 chondrites that range in petrologic subtype from 3.0 to 3.7. Chondrule mesostases in the CO chondrite of subtype 3.0 consist mainly of primary glass and plagioclase, while chondrule mesostases in the CO chondrites of higher subtypes (3.2–3.7) contain various amounts of nepheline in addition to glass and plagioclase. Nepheline has replaced glass and plagioclase, forming fine-grained aggregates and thin parallel lamellar intergrowths with plagioclase. The nephelinization has proceeded preferentially from the outer margins of chondrules toward the inside. Although the degree of nephelinization differs widely among chondrules in each of the metamorphosed chondrites, our modal analyses and bulk chemical analyses of individual mesostases indicate that the amounts of nepheline in chondrules systematically increase with the increasing petrologic subtype of the host chondrites. Nepheline also has a tendency to increase in grain size with increasing petrologic subtype. We conclude that nepheline in chondrules in the CO3 chondrites has formed largely as a result of effects related to heating on the meteorite parent body. We suggest that nepheline initially formed as hydrous nepheline under the presence of aqueous fluids and subsequently was dehydrated after exhaustion of aqueous fluids. The degree of hydrothermal activity must have increased with increasing degree of heating, and thus, chondrules in more thermally metamorphosed chondrites produced larger amounts of nepheline. The results imply that CO3 chondrites have gone through low-grade aqueous alteration and subsequent dehydration at the early stage of heating on the meteorite parent body.

INTRODUCTION

Sodium is highly volatile and mobile in aqueous activity and, thus, serves as a sensitive indicator of secondary processes that chondrites may have experienced. Chondrules and amoeboid-olivine inclusions (AOIs) in Allende and some other CV3 chondrites contain Na-rich nepheline and, less commonly, sodalite (e.g., Ikeda and Kimura 1995; Kimura and Ikeda 1997, 1998; Grossman and Steele 1976). Previous petrographic studies (e.g., Ikeda and Kimura 1995; Kimura and Ikeda 1995) revealed that the feldspathoids in chondrules formed by replacing primary mesostasis glass and plagioclase. However, whether the Na-metasomatism occurred by reaction with the gas in the solar nebula before accretion (e.g., Grossman and Steele 1976; Ikeda and Kimura 1995) or by a reaction that took place on the meteorite parent body (e.g., Krot et al. 1997) has been controversial.

Chondrules and AOIs in CO3 chondrites also contain nepheline (Kurat and Kracher 1980; Jones and Brearley 1994; Kojima et al. 1995), although much less work has been done on it compared to the counterparts in CV3 chondrites. CO3 chondrites are known to have gone through thermal metamorphism on their parent body and can be divided into petrologic subtypes 3.0–3.8 (e.g., Chizmadia et al. 2002). Kojima et al. (1995) studied AOIs in three CO3 chondrites and found that AOIs in more metamorphosed chondrites contain larger amounts of nepheline. Jones (1997a, b) studied chondrules in four CO3 chondrites and found that chondrules in more metamorphosed chondrites tend to show higher degrees of nephelinization, but some chondrules do not. This suggests that nepheline in the chondrules formed in both the solar nebula and the parent body. Despite the work, much still remains to be known regarding the relationship between Na-metasomatism and thermal metamorphism.

Calcium-aluminum-rich inclusions (CAIs) in CV3 (Wark and Lovering 1977; MacPherson and Grossman 1984; Wark 1986) and CO3 chondrites (Kurat 1975; Tomeoka et al. 1992; Russell et al. 1998) also contain nepheline and sodalite, which are clearly out of equilibrium with the primary high-temperature minerals (e.g., Grossman 1972). I-Xe isotopic measurements of sodalite from Allende CAIs indicate that this phase formed 2–6 Myr after the formation of CAIs (Hohenberg et al. 1998). Most previous workers suggested that the feldspathoids formed by reaction of primary phases such as melilite and anorthite with the solar nebula gas and that their formation temperatures are relatively low, probably less than 1000 K (e.g., MacPherson et al. 1981; Wark 1981). However, recent studies showed that CAIs in CO3 chondrites of higher petrologic subtypes tend to show higher degrees of Na-metasomatism (Kojima et al. 1995; Russell et al. 1998; Itoh and Tomeoka 1998) and, thus, suggested that at least part of the metasomatism occurred on the parent body.

If the Na-metasomatism in CAIs indeed occurred as a result of parent body thermal metamorphism, it is expected that chondrules and AOIs in the same chondrites should also have been involved in the Na-metasomatism and that CAIs, AOIs, and chondrules, all together, should indicate synchronized metasomatic effects. This hypothesis has not been verified yet. We have undertaken an extensive survey of chondrules in thin sections of seven CO3 chondrites that span the range of petrologic subtypes (3.0–3.7) using a scanning electron microscope and an electron probe microanalyzer. Here, we present the results of detailed mineralogical and petrographic studies of the chondrules. Our goal was to determine whether nepheline in the chondrules formed in the solar nebula or in the parent body and whether there is a relationship between the nephelinization and thermal metamorphism. We also intended to determine how the nephelinization in chondrules is related to that in CAIs in the CO3 chondrites.

MATERIALS AND METHODS

The samples used for this study are seven polished thin sections of the CO3 chondrites Y-81020 (petrologic type 3.0), Kainsaz (3.2), Y-82050 (3.2), Ornans (3.4), Lancé (3.5), Y-790992 (3.5), and Warrenton (3.7). Thin sections of Kainsaz and Ornans were provided by the Smithsonian Institution (USNM 2486–7 and USNM 1105–2, respectively), those of Lancé and Warrenton by the Natural History Museum, Wien (L3413 and L6617, respectively), and those of Yamato-81020, -82050 and -790992 by the National Institute for Polar Research (Y-81020-61-3, Y-82050-71-2 and Y-790992-95-1, respectively). They were studied using an optical microscope, a scanning electron microscope (SEM) (JEOL JSM-5800) equipped with an energy dispersive X-ray spectrometer (EDS), and an electron probe microanalyzer (EPMA) (JEOL JXA-8900) equipped with wavelength-dispersive X-ray spectrometers (WDS). Data corrections were made by the Phi-Rho-Z method for EDS analysis and by the Bence-Albee method for WDS analysis. EDS analyses were obtained at 15 kV and 0.4 nA, and WDS analyses were obtained at 15 kV and 12 nA. Well-characterized natural and synthetic minerals and glasses were used as standards. For the analysis of each mineral grain, we used a focused electron beam ~2 μm in diameter. For the bulk analysis of chondrule mesostases, we used a defocused electron beam ~5 μm in diameter; this beam size is regarded to be optimum, considering their small sizes and irregular shapes.

To determine modal abundances of altered materials in individual chondrule mesostases, type I chondrules from 200 to 300 μm in diameter were randomly selected from the five chondrites of different subtypes (Y-81020 [subtype 3.0], Kainsaz [3.2], Ornans [3.4], Lancé [3.5], and Warrenton [3.7]); the size range of chondrules subjected to this analysis is narrower than that (100–350 μm) of all the chondrules studied here. By limiting the chondrule size to the narrow range, we expected modal abundances of altered materials to reflect satisfactorily the degree of alteration that their host chondrules have experienced. In the analysis, mesostasis is defined as interstitial materials among phenocrysts of olivine, enstatite, diopside and grains of opaque minerals. Plagioclase occurs in both mesostasis and phenocrysts, but the latter are rare and are often difficult to distinguish from the former, and thus, all plagioclase grains are included in mesostasis. Chondrules with mesostasis areas that are <20% of their host chondrules were not subjected to the analysis. Altered materials are defined as: 1) nepheline and 2) fine-grained aggregates of nepheline with minor variable amounts of glass, plagioclase, Ca-Mg-Fe pyroxene, Fe-rich olivine, and troilite, which are termed nepheline-rich aggregates. The modal analysis was performed by using BSE images. Nepheline and nepheline-rich aggregates appear distinctly darker than primary glass and plagioclase in BSE images and so can be easily distinguished from them. A transparent grid was laid over the BSE images and areas of mesostasis, and altered materials in each chondrule were measured.

PETROGRAPHY AND MINERALOGY

General Petrography of Chondrules

Chondrules in the CO3 chondrites range in diameter from 30 to 700 μm, but 60–70% of the chondrules are in the range between 100 and 350 μm in diameter. To allow comparison of the metasomatic effects between chondrules within each chondrite and among chondrites, randomly selected type I chondrules ranging in diameter from 100 to 350 μm in each thin section of the seven CO3 chondrites (783 chondrules in total) have been studied in detail (Table 1). The metasomatic effects in chondrules involve formation of Na-rich nepheline in mesostasis (Na-metasomatism) and Fe-enrichment in olivine and pyroxene phenocrysts (Fe-metasomatism). Type I chondrules are characterized by low concentrations of Na in mesostasis and Fe in silicate phenocrysts, while type II chondrules are characterized by high concentrations of Na in mesostasis and Fe in silicate phenocrysts. Thus, type II

Table 1. CO3 chondrites and type I chondrules (100–350 μm in diameter) studied.

Meteorite	Petrologic type[a]	No. of Chondrules
Yamato-81020	3.0	80
Kainsaz	3.2	173
Yamato-82050	3.2	73
Ornans	3.4	157
Lancé	3.5	117
Yamato-790992	3.5	46
Warrenton	3.7	137

[a]After Chizmadia et al. (2002), except Y-790992. We tentatively assign subtype 3.5 to Y-790992.

chondrules were considered to be unsuitable for our purpose and were not subjected to study. Textural types of the chondrules that we studied include porphyritic olivine-pyroxene, porphyritic olivine, porphyritic pyroxene, and barred olivine (classification after Rubin [1989]). Other minor types of chondrules were not subjected to study. These types of chondrules consist primarily of phenocrysts of olivine and/or enstatite and mesostasis. Minor amounts of diopside and plagioclase also occur as phenocrysts. Most chondrules contain opaque nodules (1–30 μm in diameter) that consist of Fe-Ni metal, troilite, and occasionally magnetite and Ca-phosphate.

Before going to further descriptions, we note here the classification schemes for the petrologic subtypes of the CO3 chondrites. The previous classification is mainly based on the FeO content and Fe-Mg zoning in olivines and pyroxenes in chondrules (Scott and Jones 1990; Kojima et al. 1995) and peak thermoluminescence temperatures of whole meteorite samples (Keck and Sears 1987). The classification differs slightly among the authors. Recently Chizmadia et al. (2002) found that amoiboid-olivine inclusions (AOIs) are sensitive indicators of thermal metamorphism of the host chondrites, because they are fine-grained and have high surface-area/volume ratios, and refined the classification based on the characteristics of AOIs as well as the results of the previous workers. In this paper, we use this new classification for the CO3 chondrites (Table 1).

Olivines in chondrules in CO3 chondrites show systematic compositional changes (Fe-metasomatism) due to thermal metamorphism (e.g., Scott and Jones 1990). Olivines in the Y-81020 CO3.0 chondrite are very forsteritic ($Fa_{<3}$) and show no Fe-Mg zoning (Figs. 1a–1d). In the chondrites of higher petrologic subtypes (3.2–3.7), however, forsteritic olivine is increasingly converted to Fe-rich olivine with an increasing degree of metamorphism. Iron is enriched along the edges and cracks of olivine crystals, showing zoning of magnesian cores with ferroan rims. In chondrules in the relatively highly metamorphosed Lancé (subtype 3.5), Y-790992 (3.5), and Warrenton (3.7), most olivine grains >10 μm in diameter exhibit strong Fe-Mg zoning, and those <10 μm in diameter are homogeneously Fe-rich in the range of $Fa_{35–45}$ because Fe diffusion has reached the grain centers. In heavily altered chondrules, in particular, even enstatite grains are partially enriched in Fe and replaced by Fe-rich olivine. Those Fe-enrichments in olivine and pyroxene are regarded to have resulted mainly from solid-state diffusion of Fe from surrounding Fe-rich matrix and also from Fe-Ni metal and troilite in chondrules.

Chondrule Mesostases: Variations with Increasing Petrologic Subtype

Yamato-81020 (Petrologic Type 3.0)

Mesostases of chondrules in this chondrite consist mainly of glass and plagioclase (Figs. 1a–1d). Plagioclase fills interstices among phenocrysts and also occurs as laths (5–20 μm in width and 50–150 μm in length) or blocky crystals (5–50 μm in size) in glass. Both glass and plagioclase are smooth on the surfaces and are homogeneous in composition. Glass has a composition close to anorthitic plagioclase with 0.5–3.1 wt% Na_2O, 15–20 wt% CaO, 0.6–5.5 wt% MgO, and 0.2–1.1 wt% FeO (Table 2). Plagioclase is very anorthitic ($An_{85–92}$) with <1.3 wt% MgO, <1.2 wt% FeO, and <0.1 wt% K_2O (Table 2). Glass commonly includes dense arrays of parallel, thin lath-shaped quenched crystallites (0.5–3 μm in width) of Ca-Mg-rich pyroxene, mostly diopside; this feature can be seen in Fig. 3b. Almost all the chondrules show no traces of alteration (Fig. 2). Even chondrules with mesostases that are in direct contact with the matrix show no alteration (Figs. 1b and 1d). These characteristics, together with no Fe-enrichment in olivine and pyroxene phenocrysts, indicate that the chondrules remain almost intact of alteration and represent primary mineralogy and texture of igneous origin. The texture and mineralogy are common to all the type I chondrules in other CO3 chondrites described below except secondary alteration features.

Kainsaz and Yamato-82050 (Petrologic Type 3.2)

In Kainsaz, ~38% of the chondrules show no alteration in their mesostases and have texture, mineralogy, and composition similar to those in Y-81020 described above (Fig. 2). The rest (~62%) show minor degrees of alteration in their mesostases. In altered chondrules in both chondrites, glass has been partially enriched in Na_2O (up to 10 wt%) and replaced by fine-grained, porous aggregates of nepheline, occasionally with minor amounts of diopside, which are the remnants of quenched crystallites (Figs. 3a and 3b). Plagioclase has been replaced by characteristic thin parallel lamellae of nepheline (typically 0.5–2 μm in width) (Figs. 4a and 4b); this occurrence of nepheline has previously been reported from some specific (plagioclase-rich) type chondrules in CO3 chondrites (Kurat and Kracher 1980; Jones 1997a) and from chondrules in some CV3 chondrites (Kimura and Ikeda 1997). Alteration of both glass and plagioclase occurs preferentially from the chondrule edge

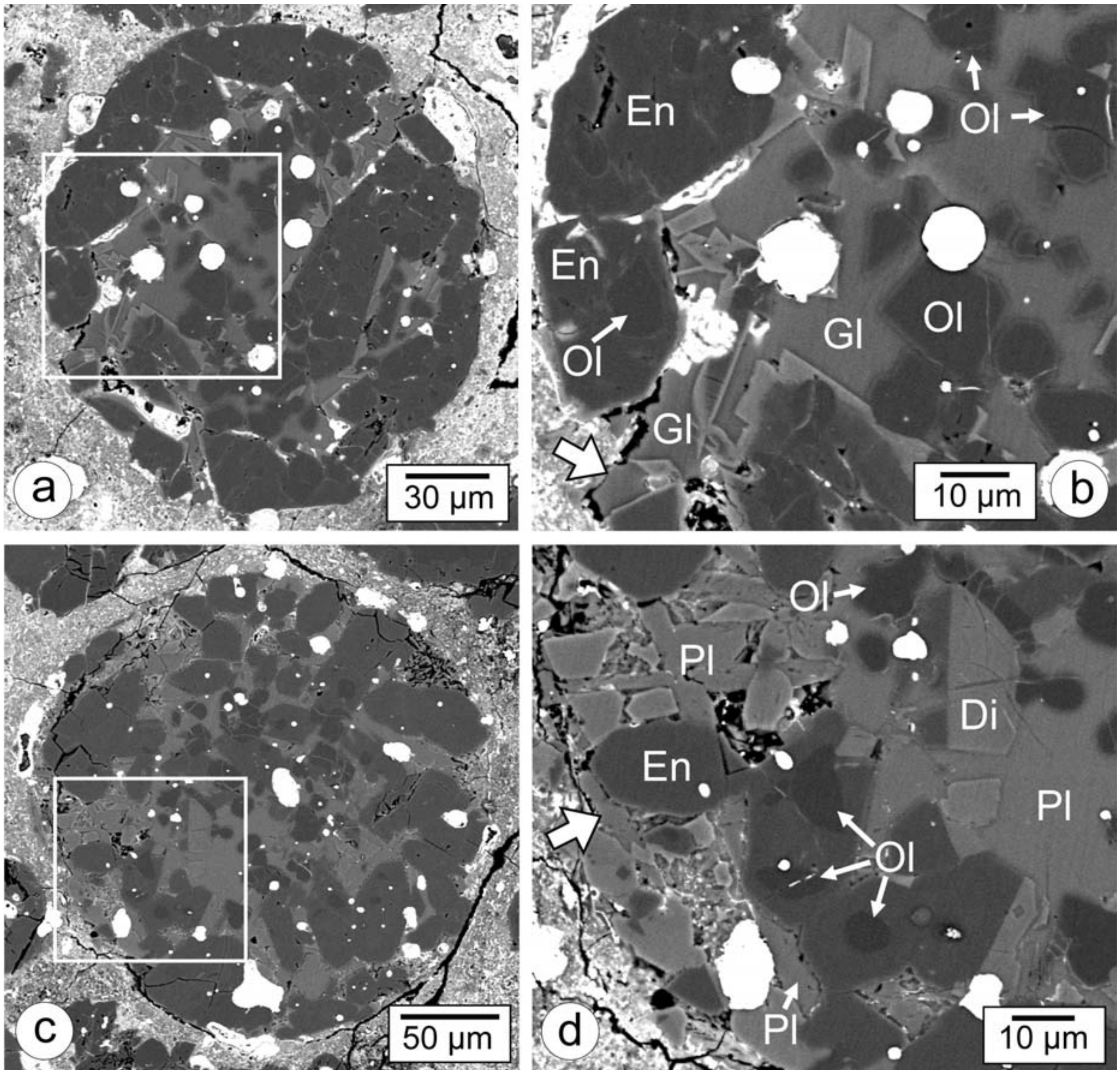


Fig. 1. Backscattered electron images of type I chondrules in Y-81020 (petrologic subtype 3.0): a) chondrule whose mesostasis consists of glass (Gl); b) image of boxed area in (a); c) chondrule with mesostasis that consists of plagioclase (Pl); d) image of boxed area in (c). The mesostases of both chondrules have smooth surfaces and show no traces of nephelinization. Indicated by large arrows in (b) (lower left) and (d) (center left) are portions of the mesostases that are in direct contact with the matrices. Also, note that phenocrysts of olivine (Ol) and enstatite (En) show no Fe-Mg zoning. Bright round grains are Fe-(Ni) metal and/or troilite. Di = diopside.

where they are in direct contact with the matrix (Figs. 3 and 4). However, there are also chondrules, although rare, with mesostases that are in direct contact with the matrix but show no alteration (Figs. 5a and 5b). The degree of alteration along the chondrule edge differs even within individual chondrules. For example, in the chondrule shown in Fig. 3a, the mesostasis is exposed directly to the matrix along the nearly entire chondrule edge, however, alteration occurs only in limited portions indicated by arrows. Olivine phenocrysts in almost all chondrules, regardless of containing nepheline, show Fe-Mg zoning; the degree of zoning is indistinguishable between chondrules with nepheline and without nepheline.

Ornans, Lancé, and Yamato-790992 (Petrologic Type 3.4–3.5)

In Ornans and Lancé, mesostases of >90% of the chondrules have been replaced by nepheline or nepheline-rich aggregates to various extents (Fig. 2). In ~22% of the chondrules, altered materials comprise >25 vol% of individual mesostases. In Ornans, we found a chondrule with mesostasis that has been almost completely replaced by fine-grained aggregates of nepheline (Fig. 6). In all the three chondrites, glass tends to be more enriched in Na_2O than in the CO3.2 chondrites described above (Table 2) and has been partially replaced by aggregates of nepheline grains (<5 µm in size) that tend to be larger in size than those in the CO3.2 chondrites (Figs. 7a and 7b). Na-enrichment in glass appears

Table 2. Selected electron microprobe analyses of glass and plagioclase in chondrules in the CO3 chondrites (wt%).[a]

	Glass					Plagioclase				
	Y	K	O	L	W	Y	K	O	L	W
	1	2	3	4	5	1	2	3	4	5
SiO_2	54.4	49.1	58.4	55.0	56.7	46.8	46.3	46.3	47.1	46.4
TiO_2	0.77	0.38	0.07	0.06	0.19	n.d.	n.d.	0.11	n.d.	n.d.
Al_2O_3	19.9	23.2	24.1	27.4	22.6	32.4	33.5	32.5	32.6	33.4
Cr_2O_3	0.29	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
FeO	0.20	2.21	1.74	0.83	1.78	0.95	0.66	0.87	0.42	0.60
NiO	n.d.	n.d.	n.d.	n.d.	0.06	n.d.	n.d.	0.21	n.d.	n.d.
MnO	n.d.	0.14	n.d.	0.04	n.d.	n.d.	n.d.	0.17	0.08	n.d.
MgO	3.82	7.11	1.10	0.25	3.85	0.41	0.62	0.32	0.63	0.41
CaO	19.2	16.5	7.76	9.94	6.68	18.6	17.6	17.5	17.2	16.4
Na_2O	1.13	2.14	7.69	5.97	8.30	1.57	1.70	2.44	2.36	2.44
K_2O	n.d.	0.04	0.11	n.d.	0.39	n.d.	0.04	0.17	0.11	0.19
Total	99.7	100.8	101.0	99.5	100.6	100.7	100.4	100.6	100.5	99.8

[a]Y = Yamato-81020; K = Kainsaz; O = Ornans, L = Lancé; W = Warrenton; n.d. = not determined.

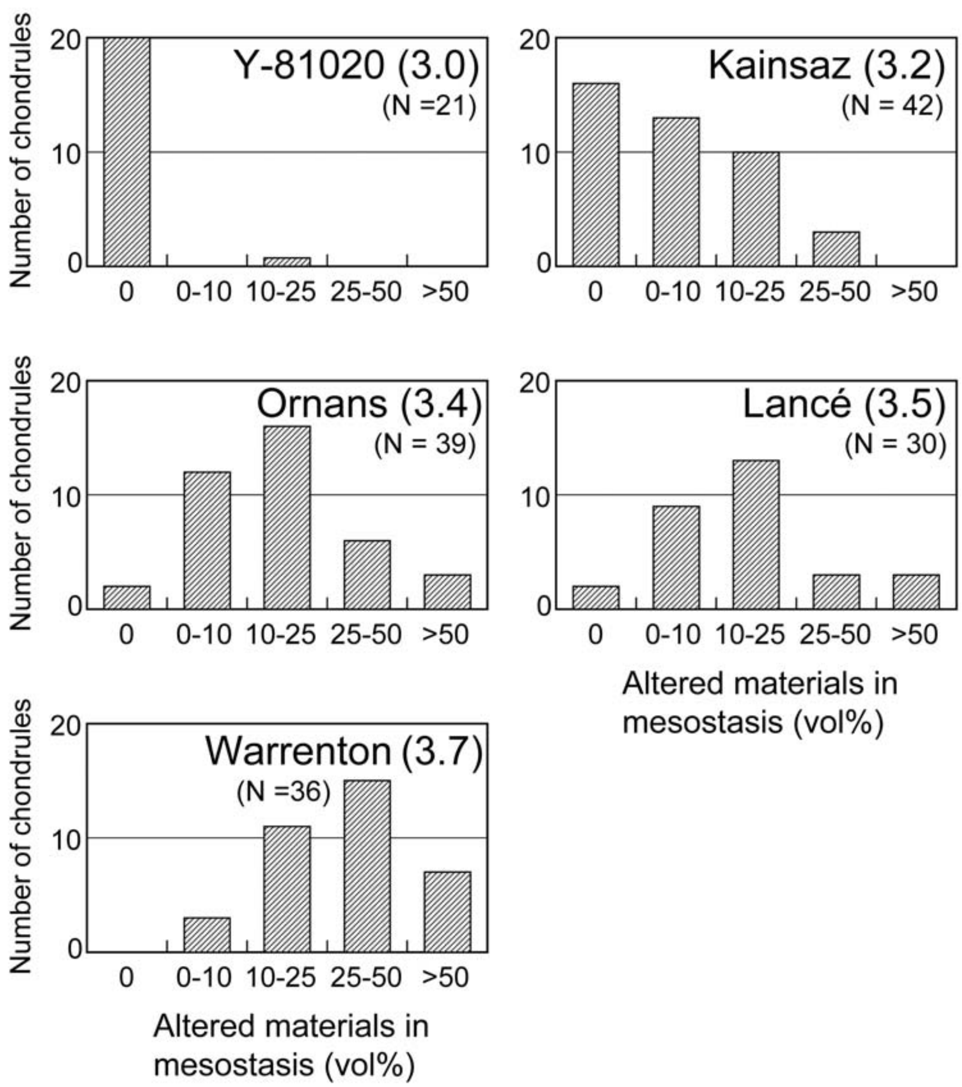


Fig. 2. Modal abundances of altered materials (nepheline and nepheline-rich aggregates) in individual mesostases of type I chondrules (200–300 μm in diameter) in the five CO3 chondrites of different petrologic subtypes (3.0–3.7). The chondrules were randomly selected. N = number of chondrules. See the Materials and Methods section for details of the analysis.

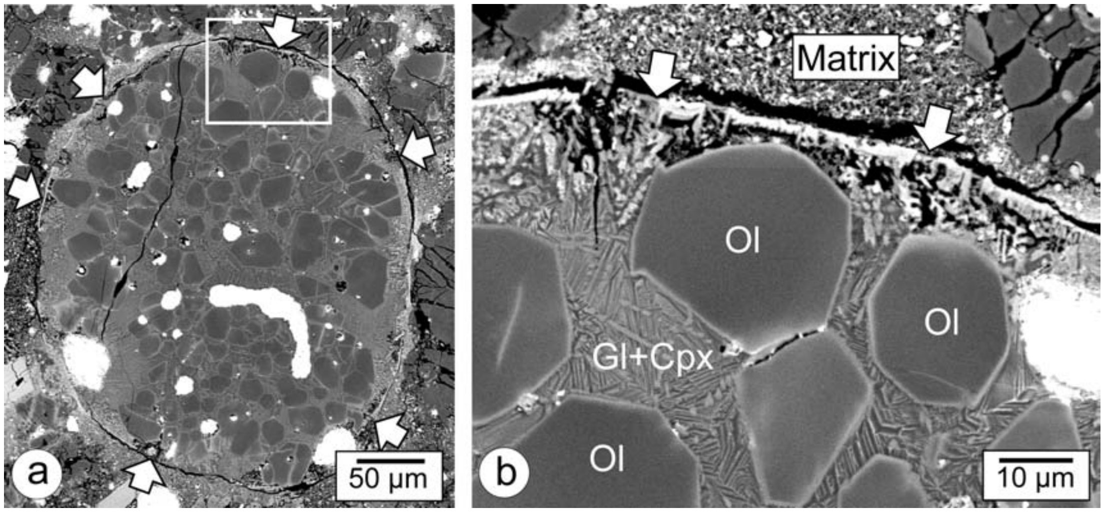


Fig. 3. a) Backscattered electron image of a chondrule in Kainsaz (subtype 3.2) with mesostasis that is in direct contact with the matrix along the nearly entire chondrule edge. Portions of the mesostasis that have been replaced by nepheline-rich aggregates are indicated by arrows. The rest of the chondrule edge shows no replacement; b) image of boxed area in (a) showing that the mesostasis, which consists of glass (Gl) and quenched crystallites of Ca-rich pyroxene (Cpx, lighter grayish narrow plates), has been replaced by fine-grained nepheline-rich aggregates (indicated by arrows) preferentially from the chondrule edge to the inside. Ol = olivine.

to be correlated with Ca depletion (Table 2). Diopside crystallites in glass are commonly enriched in FeO but remain unaltered. Plagioclase has been replaced by nepheline along grain edges and also by nepheline lamellae that tend to be thicker and higher in density than those in the CO3.2 chondrites (Figs. 7c, 7d, and 8). In the three chondrites, all the chondrules with mesostases that are in direct contact with matrix show alteration except one in Ornans, and they have been replaced by nepheline preferentially from the surface toward the center of each chondrule (Figs. 7a–7d and 8b).

Warrenton (Petrologic Type 3.7)

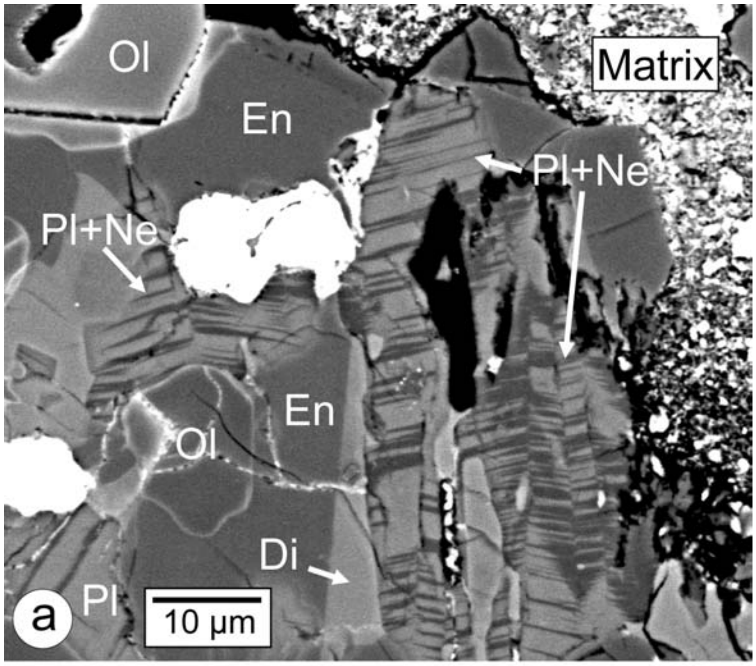
Chondrules in this highly metamorphosed CO3 chondrite show the highest degree of alteration of the chondrules studied (Fig. 2). All the chondrules studied contain various amounts of nepheline, and in ~61% of the chondrules, altered materials comprise >25 vol% of individual mesostases (Fig. 2). Nepheline grains >5 µm in size are much more abundant than in the other chondrites described above, and the largest nepheline grain (18 × 25 µm in size) of those studied has been found in this chondrite (Fig. 9). Coarse-grained nepheline has a smooth, homogeneous appearance and is free of other phases. In nepheline-rich aggregates, troilite particles (<3 µm in diameter) are more common than in the other chondrites. In heavily altered chondrules, alteration has converted diopside crystallites in glass to hedenbergite and even to nepheline (Figs. 10a and 10b). Nepheline is commonly intermixed with secondary Fe-rich olivine produced by FeO enrichment of enstatite (Figs. 11a–11c). Opaque nodules have been highly corroded, forming complex mixtures of nepheline, troilite, metal, and Fe-rich olivine in their peripheries (Fig. 11c). Although nepheline has formed extensively throughout mesostases in most of altered chondrules, some chondrules retain unreplaced plagioclase in their interiors (Fig. 11d).

Chemical Compositions of Nepheline

Nepheline is commonly fine-grained and intermixed with small grains of various minerals, and thus, it is difficult to obtain high-quality analysis data even by a focused electron beam. The analyses shown in Table 3 were obtained from relatively coarse-grained nepheline (>5 µm in size) that is considered to be the least contaminated by nearby minerals. The analyses are similar to those of nepheline in AOIs, CAIs (Grossman and Steele 1976; Hashimoto and Grossman 1987), and chondrules (Kimura and Ikeda 1995) in Allende in that they are slightly excess in Si and deficient in Na and Al relative to the ideal nepheline stoichiometry. However, the CaO (<2.0 wt%) and K_2O (<2.0 wt%) contents of nepheline in the CO3 chondrites are lower and variable compared to the nepheline in Allende (1.6–5.2 wt% CaO and 1.6–2.4 wt% K_2O; from Grossman and Steele 1976). These characteristics are rather similar to those of nepheline in CAIs in a CO3 chondrite (Tomeoka et al. 1992). Although sodalite is commonly associated with nepheline in AOIs, CAIs, and chondrules in Allende, it is rare in chondrules in the CO3 chondrites. Only in nepheline-rich aggregates in Warrenton chondrules, minor amounts of Cl are occasionally detected, which suggests that minor amounts of sodalite are intermixed with nepheline.

Defocused-Beam Analysis of Chondrule Mesostases

Because nepheline generally forms porous, submicron-to-micron-scale intergrowths with plagioclase, glass,



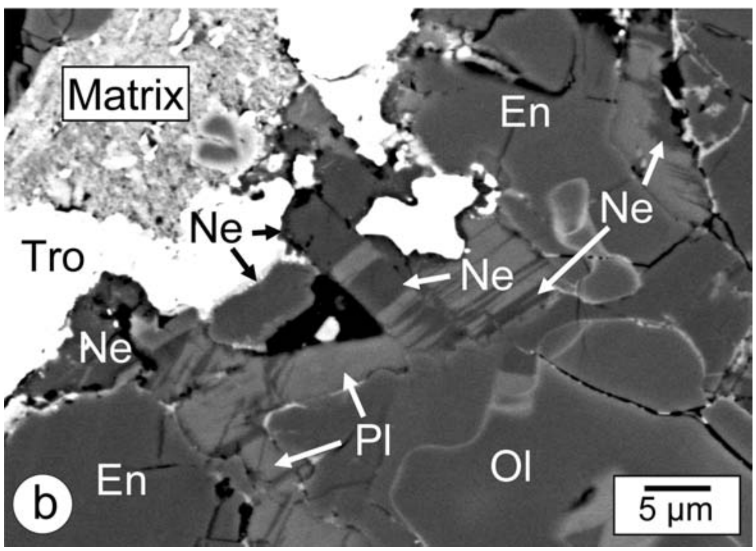


Fig. 4. Backscattered electron images of two different portions of a chondrule in Kainsaz in which the plagioclase mesostasis is in direct contact with the matrix: a) plagioclase (Pl) has been replaced by parallel thin lamellae of nepheline (Ne), which appear dark relative to plagioclase; b) relatively thick (3–6 μm) nepheline lamellae form in plagioclase near the matrix. Ol = olivine. En = enstatite. Di = diopside. Tro = troilite.

diopside, and various secondary minerals, it is difficult to quantify the abundance of nepheline in individual chondrules by the modal analyses. However, in the case of chondrules having plagioclase mesostases, it is possible to estimate the relative proportions of nepheline and plagioclase in individual chondrule mesostases from their bulk compositions. This is because byproducts of nephelinization of plagioclase are relatively minor in abundance, and thus, nephelinized plagioclase can be regarded roughly as mixtures of plagioclase and nepheline. To obtain bulk compositions of individual plagioclase mesostases, we performed defocused-beam analysis using an ~5 μm-diameter beam.

Bulk chemical analyses of individual plagioclase mesostases of randomly selected chondrules are plotted in terms of (Na + K) versus total cations based on O = 24 (filled circles) in Fig. 12. In these diagrams (see the diagram at the lower right in Fig. 12 for reference), the lower-left corner (Na + K = 0 and total cations = 15; point A in the reference diagram) corresponds to anorthite ($Ca_3Al_6Si_6O_{24}$, Ca-rich end member of plagioclase), the point at Na + K = 3 on the vertical axis (point B in the reference diagram) corresponds to albite

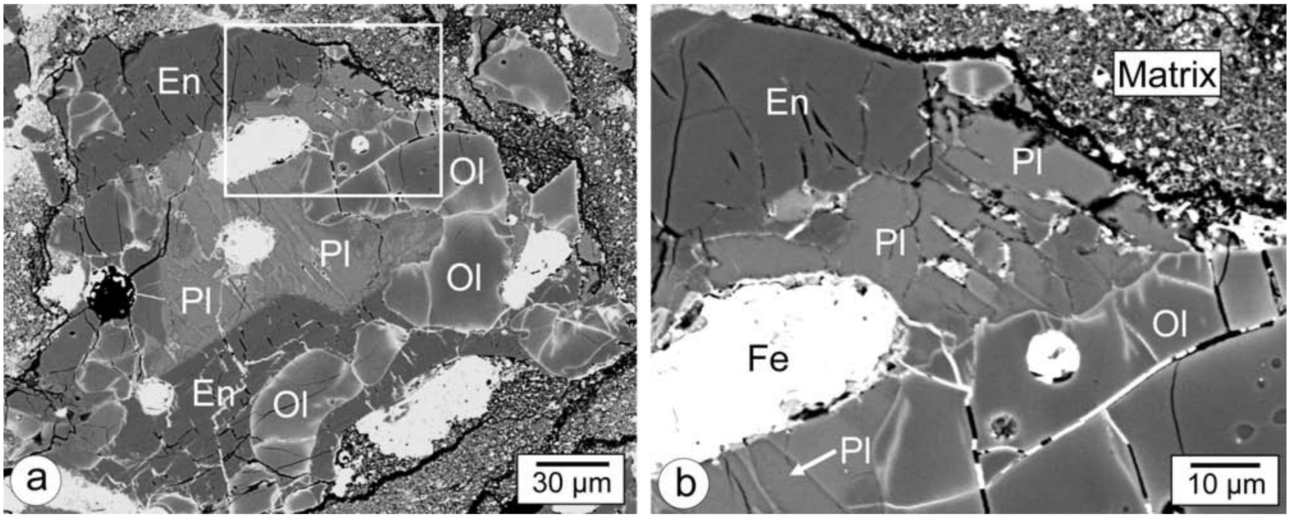


Fig. 5. a) Backscattered electron image of a chondrule in Kainsaz with plagioclase (Pl) mesostasis that shows no nephelinization; b) image of boxed area in (a) showing that the plagioclase mesostasis is in direct contact with the matrix but shows no nephelinization. The olivine (Ol) phenocrysts exhibit Fe-Mg zoning. Also, note that this chondrule is very irregular in external shape, suggesting that it has gone through fragmentation. En = enstatite. Fe = Fe metal.

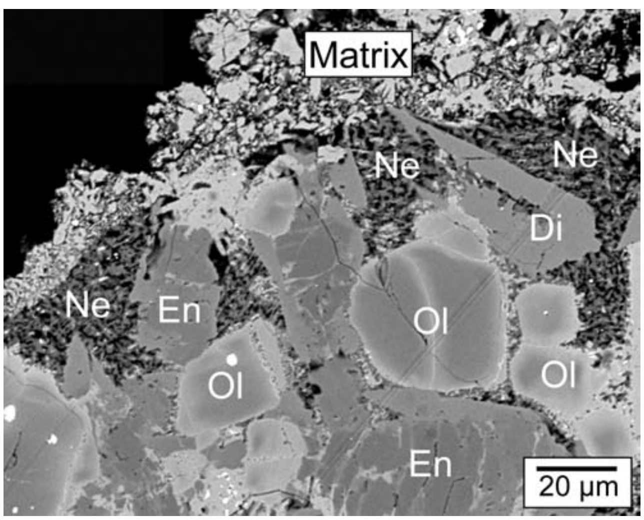


Fig. 6. Backscattered electron image of a portion of a chondrule in Ornans (subtype 3.4) with mesostasis that has been almost completely replaced by fine-grained aggregates of nepheline (Ne). The olivine (Ol) phenocrysts in this chondrule show particularly strong Fe-Mg zoning. En = enstatite. Di = diopside.

($Na_3Al_3Si_9O_{24}$, Na-rich end member of plagioclase), and the upper-right corner (Na + K = 6 and total cations = 18; point C in the reference diagram) corresponds to nepheline ($Na_6Al_6Si_6O_{24}$). Thus, if individual mesostases are mixtures of plagioclase and nepheline, the analyses should plot within triangle ABC, and the relative molar proportions of anorthite, albite, and nepheline components can be given by the plotted points.

In the diagram of Y-81020, most analyses are concentrated closely near A, which indicates that the mesostases consist almost entirely of anorthitic plagioclase. In Kainsaz, analyses are also concentrated near A but shift slightly upward and extend along line AC toward C, which indicates that plagioclase is relatively high in albite-component and has been weakly replaced by nepheline. In Y-82050, analyses lie tightly along line AC and extend more widely from point A toward C than those in Kainsaz, which indicates that plagioclase has also been weakly replaced by nepheline, but the degree of nephelinization is higher than in Kainsaz. In Ornans, Lancé, Y-790992, and Warrenton, analyses spread tightly and widely along line AC, and more analyses in Warrenton lie closer to C, which indicates that the degree of nephelinization of plagioclase varies widely in these chondrites and the degree in Warrenton is higher than those in the other chondrites. The results of these analyses are consistent with our observations that plagioclase mesostases in the CO3.0–3.7 chondrules consist mainly of mixtures of anorthitic plagioclase and nepheline, and as the petrologic subtype increases, the proportion of nepheline in those mesostases increases, while the proportion of plagioclase decreases concordantly.

DISCUSSION

Location and Timing of Nephelinization

Our study has revealed that type I chondrules in the CO3.0 chondrite (Y-81020) consist of primary minerals of igneous origin with little traces of alteration, while type I chondrules in the CO chondrites of higher subtypes (3.2–3.7) contain various amounts of nepheline as a secondary alteration product. Although the degree of nephelinization differs in a wide range among chondrules within each of the CO3.2–3.7 chondrites, our modal analyses and bulk chemical analyses of chondrule mesostases indicate that there is an

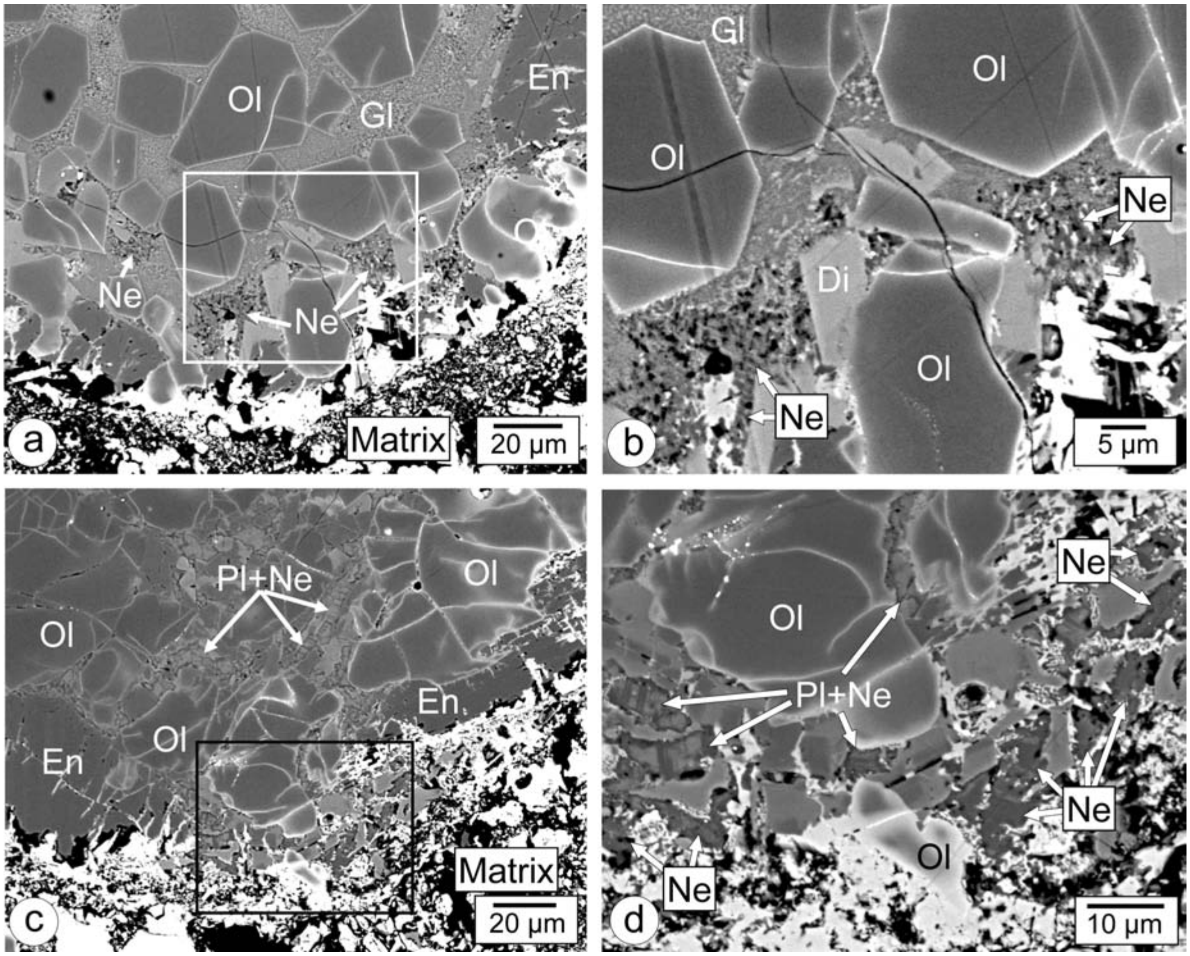


Fig. 7. a) Backscattered electron image of a portion of a chondrule in Ornans with glass (Gl) mesostasis that has been replaced by nepheline (Ne)-rich aggregates preferentially along a 20–40 µm-thick zone from the chondrule edge; b) image of boxed area in (a) showing formation of micron-size nepheline grains (dark) in the mesostasis; c) portion of a chondrule with plagioclase (Pl) mesostasis that has been replaced by nepheline preferentially from the chondrule edge; d) image of boxed area in (c) showing formation of relatively coarse grains (3–8 µm in size) of nepheline. Ol = olivine. En = enstatite.

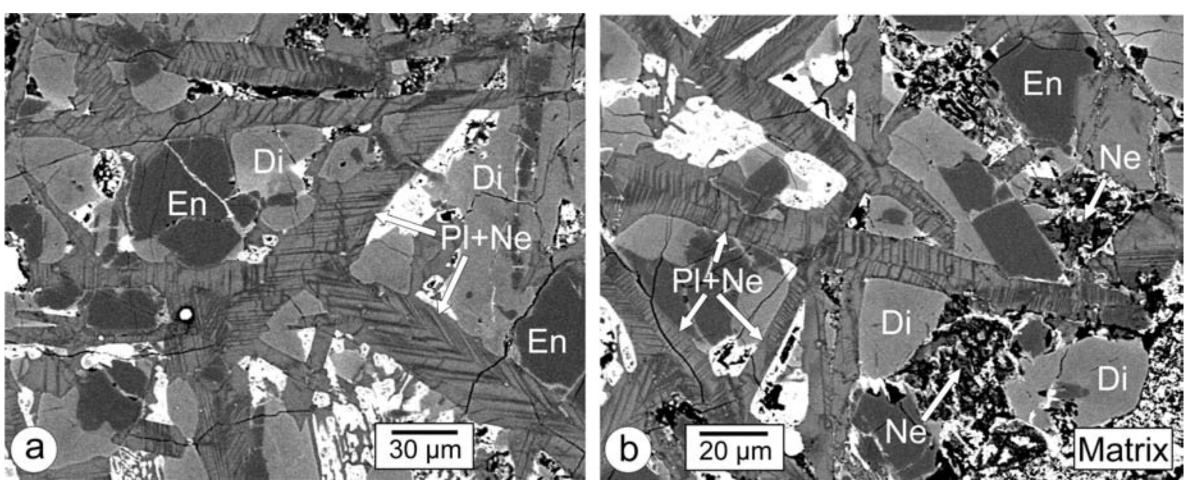


Fig. 8. a) Backscattered electron image of an inner portion of a plagioclase-pyroxene-rich chondrule in Ornans. Nepheline (Ne) lamellae (dark) form at high density throughout plagioclase (Pl) laths; b) outer portion of the chondrule showing that, in addition to plagioclase, the mesostasis near the matrix (lower-right) has also been replaced by nepheline-rich aggregates (indicated by arrows). En = enstatite. Di = diopside.

Table 3. Selected electron microprobe analyses of nepheline in chondrules in the CO3 chondrites (wt %).[a]

	Nepheline						
	K	O	O	L	L	W	W
	1	2	3	4	5	6	7
SiO_2	48.4	43.0	45.3	41.6	44.3	43.5	43.5
TiO_2	0.04	n.d.	0.08	0.04	n.d.	n.d.	0.15
Al_2O_3	34.7	34.1	34.0	35.4	34.2	36.3	36.6
Cr_2O_3	0.04	0.06	0.09	n.d.	n.d.	n.d.	n.d.
FeO	0.78	0.81	1.25	0.30	0.65	0.10	0.29
NiO	n.d.	n.d.	0.09	n.d.	0.05	0.06	n.d.
MnO	n.d.	0.03	n.d.	n.d.	n.d.	n.d.	n.d.
MgO	0.20	0.24	0.48	n.d.	0.49	n.d.	0.18
CaO	0.69	1.02	1.34	0.09	1.51	0.13	0.53
Na_2O	15.1	18.5	17.5	21.9	17.6	20.1	19.5
K_2O	0.04	2.03	0.72	n.d.	1.58	0.04	0.10
Total	100.0	99.8	100.9	99.3	100.4	100.2	100.9

[a]Y = Yamato-81020; K = Kainsaz; O = Ornans; L = Lancé; W = Warrenton; n.d. = not determined.

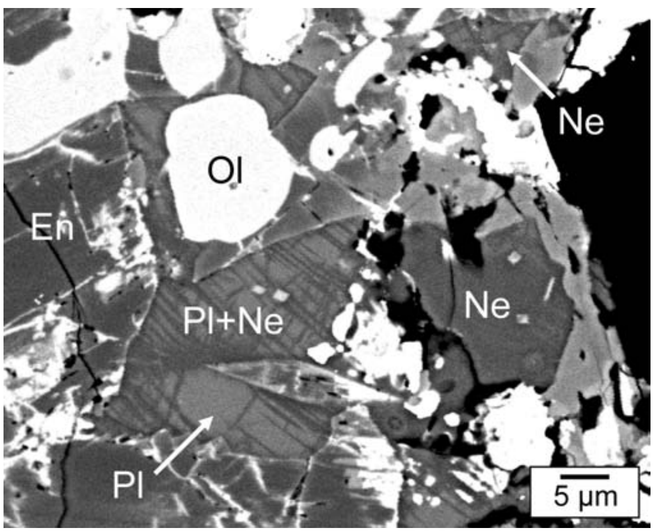


Fig. 9. Backscattered electron image of the largest nepheline (Ne) grain (18 × 25 µm in size, center right) found in the present study, from a chondrule in Warrenton (subtype 3.7). Ol = olivine. En = enstatite. Pl = plagioclase.

apparent systematic tendency for the proportions of nepheline in individual mesostases to increase with petrologic subtype of the host chondrites (Figs. 2 and 12). Nepheline also shows an apparent tendency to increase in grain size with increasing petrologic subtype. From these observations and analyses, we conclude that nepheline in chondrules in the CO3 chondrites formed largely as a result of effects related to heating that occurred on the meteorite parent body.

Some previous workers argued that the coexistence of nepheline-bearing chondrules and nepheline-free chondrules in the same chondrite indicates a nebular process for the formation of nepheline (e.g., Jones 1997b; Kimura and Ikeda 1995); that is, some chondrules had formed nepheline by reaction with a Na-rich gas in the solar nebula, and later, during accretion, they were assembled together with chondrules that had not experienced such alteration reaction. However, our study has revealed that the wide range in degree of nephelinization among chondrules is a common characteristic of all the metamorphosed CO3 chondrites. All the chondrites except Warrenton include chondrules with mesostases that show no nephelinization (Fig. 2), and some of them contain glass or plagioclase that is in direct contact with matrix (e.g., Figs. 5a and 5b). We also found that there are often marked differences in the degree of nephelinization among locations of mesostasis that are in direct contact with matrix within individual chondrules (e.g., Figs. 3a and 3b). We suggest that the different degrees of nephelinization among chondrules within individual chondrites and also within individual chondrules resulted from heterogeneous distribution of alteration effects as discussed below.

In the case of chondrules containing unaltered mesostases that are directly exposed to matrix, in particular, we also suggest the possibility that they resulted from fragmentation during brecciation after nephelinization ceased at its early stage. We found that most chondrules with mesostases that are in direct contact with matrix exhibit texture and external shapes suggestive of fragmentation (e.g., see Fig. 5). Although lithic clasts have rarely been reported from CO3 chondrites, fragments of chondrules, AOIs, and CAIs are abundant in those chondrites (also see McSween [1977] and Bunch and Rajan [1988]). Our recent study of dark inclusions in CO3 chondrites also revealed evidence that brecciation occurred actively on the CO parent body (Itoh and Tomeoka 2003).

Nephelinization by Aqueous Alteration and Dehydration

The next question to be addressed is how the nephelinization in chondrules proceeded with heating. To answer to this question, we first need to consider from which Na in nepheline was supplied. Type I chondrules are characterized by low concentrations of Na and high

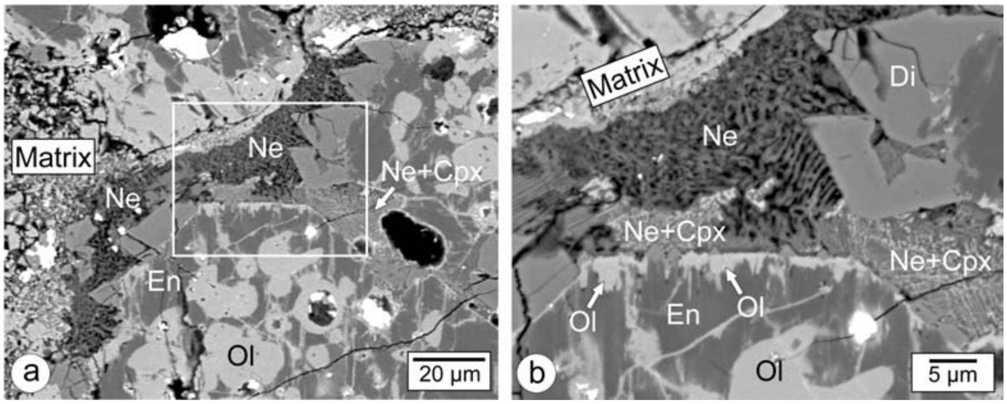


Fig. 10. a) Backscattered electron image of a portion of a chondrule in Warrenton showing formation of nepheline (Ne, dark) along the chondrule edge; b) image of boxed area in (a) showing that nepheline has replaced mesostasis glass and Ca-rich pyroxene crystallites (Cpx, lighter grayish needle-shaped grains). Glass has been largely replaced by nepheline, while Ca-rich pyroxene crystallites, especially those located inside of the chondrule, remain unaltered. Fe-metasomatism is also intense in this chondrule, so that most olivine (Ol) phenocrysts are homogeneously Fe-rich, because Fe diffusion has reached the grain centers, and even enstatite (En) phenocrysts have been replaced by Fe-rich olivine along their edges.

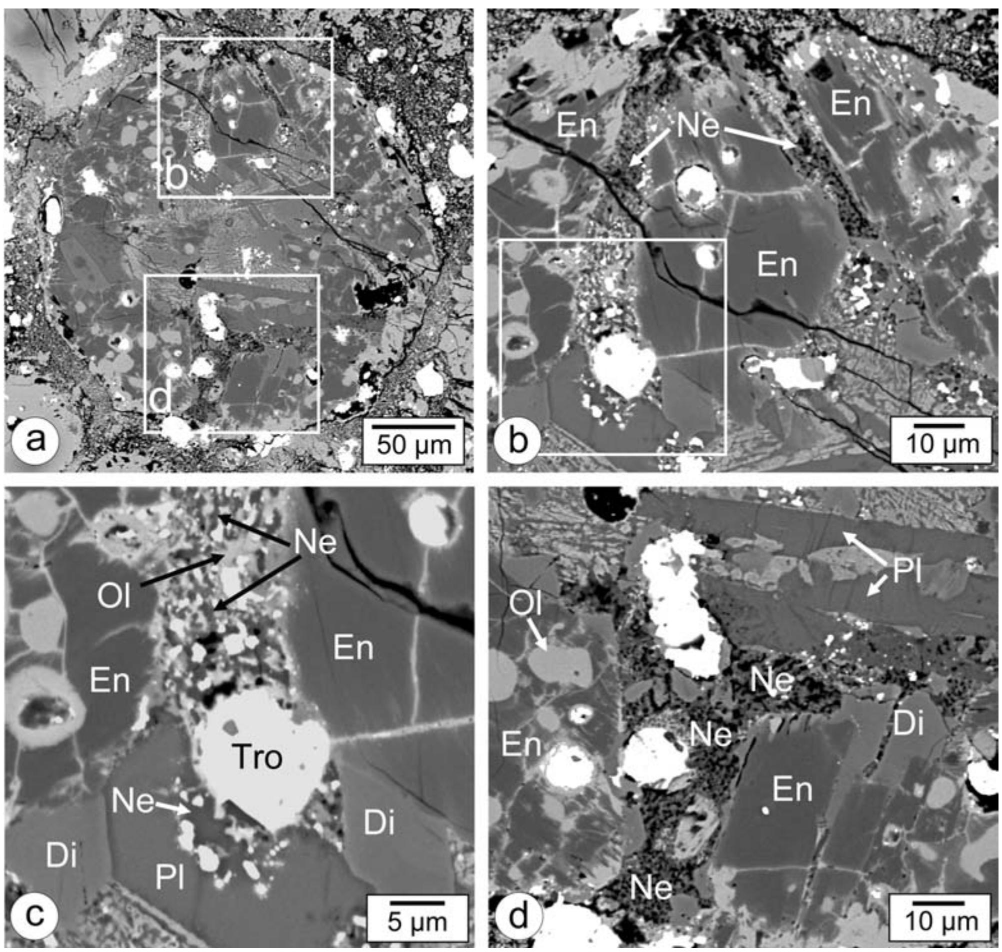


Fig. 11. a) Backscattered electron image of a chondrule in Warrenton with mesostasis (glass + Ca-rich pyroxene crystallites) that has been replaced by nepheline preferentially from the chondrule edge; b) image of boxed area "b" in (a) showing formation of nepheline (Ne) by replacing the mesostasis; c) image of boxed area in (b) showing that troilite (Tro, bright) particles and Fe-rich olivine (Ol, lighter gray) are intermixed with nepheline (dark). Note that a plagioclase (Pl) grain (bottom) has been partly replaced by a mixture of nepheline and troilite; d) image of boxed area "d" in (a) showing formation of nepheline by replacing the mesostasis. Note that plagioclase laths (top) located near the center of the chondrule remain unaltered despite the extensive formation of nepheline in the outer portion of the chondrule.

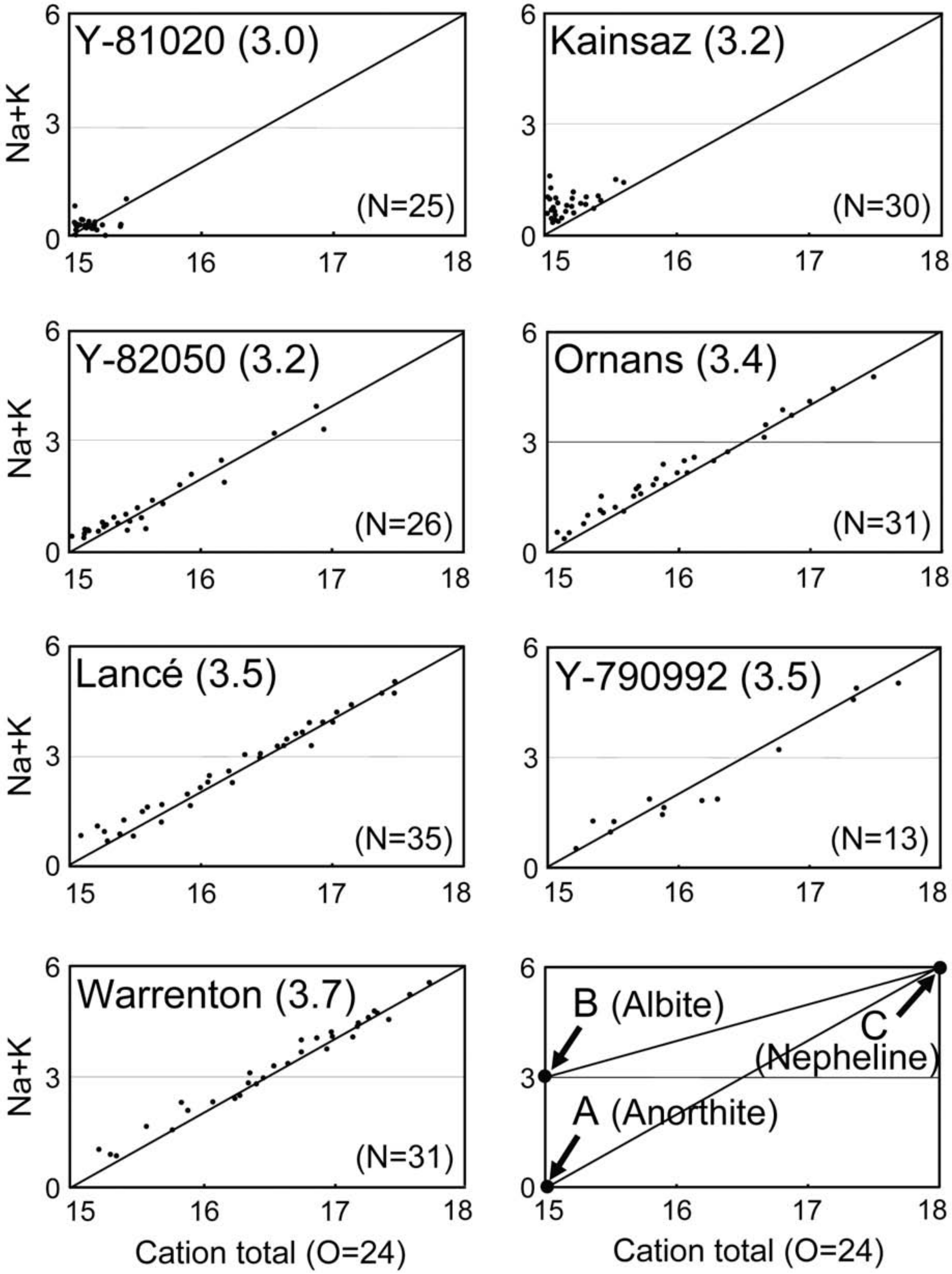


Fig. 12. Bulk chemical analyses of individual plagioclase mesostases of chondrules in the seven CO3 chondrites (petrologic type 3.0–3.7) plotted in terms of (Na + K) versus total cations based on O = 24. The chondrules were randomly selected. Each analysis is an average of 3 to 7 defocused (5 μm in diameter) beam analyses of each chondrule mesostasis. In these diagrams (see the diagram at the lower right for reference), the lower left corner (A in the reference diagram), the point at Na + K = 3 on the vertical axis (B), and the upper right corner (C) correspond to anorthite ($Ca_3Al_6Si_6O_{24}$), albite ($Na_3Al_3Si_9O_{24}$), and nepheline ($Na_6Al_6Si_6O_{24}$), respectively. N = number of analyses.

concentrations of Ca relative to surrounding matrix, which indicates that Na could not be supplied internally from chondrules. Second, we point out textural evidence that nepheline in all the metamorphosed CO3 chondrites shows a tendency to form preferentially from the outer margins of chondrules toward the inside. These compositional and textural characteristics indicate that Na was supplied from outside of chondrules through matrix, and at the same time, Ca leached out from chondrules. Sodium and calcium are known to be highly mobile in aqueous activity (McSween and Richardson 1977). From these results and considerations, we suggest that the Na-Ca exchange reaction probably occurred under the presence of aqueous fluids.

CO3 chondrites contain few hydrous minerals compared to the CI, CM, and hydrated CV chondrites, such as Mokoia, Bali and Kaba, which commonly has been taken as evidence against the major role played by aqueous alteration in the history of CO3 chondrites. However, there is growing evidence that the CO3 chondrites have gone through considerable aqueous alteration. Some CO3 chondrites contain minor amounts of hydrous phyllosilicates in their matrices and chondrules (Kerridge 1964; Ikeda 1983; Keller and Buseck 1990; Brearley 1993). From petrographic studies of amoeboid olivine inclusions (AOIs) in many CO3 chondrites, Rubin (1998) and Chizmadia et al. (2002) suggested that aqueous alteration is responsible for many petrologic and compositional features of AOIs including the rim formation and the conversion of forsteritic olivine to fayalitic olivine. We suggest that the rarity of hydrous phases in the CO3 chondrites does not necessarily indicate the lack of aqueous alteration in their history but, rather, resulted from dehydration that occurred subsequent to aqueous alteration.

Itoh and Tomeoka (2003) revealed that CO3 chondrites contain abundant dark inclusions (DIs), although they are smaller in size than those in CV3 chondrites. The DIs in the CO3 chondrites were found to be lithic clasts similar in mineralogy and chemistry to their host chondrites and that show evidence for aqueous alteration and subsequent dehydration, like DIs in CV3 chondrites (e.g., Kojima et al. 1993; Kojima and Tomeoka 1996; Krot et al. 1997, 1998; Ohnishi and Tomeoka 2002). This implies that the CO parent body once contained aqueous fluids and that there was a region (or regions) in the parent body in which aqueous alteration and subsequent dehydration took place. We envisage that the alteration that produced nepheline in CO3 chondrules occurred under the presence of small amounts of aqueous fluids. Thus, fluids did not infiltrate the parent body extensively, and the aqueous alteration ceased at the early stage of heating, probably due to exhaustion of water. Owing to continuous heating, hydrous minerals were subsequently dehydrated. Therefore, the resultant effects of aqueous alteration were spatially very limited and heterogeneous on small scales (perhaps on the order of 0.1–10 mm).

The idea that nepheline, rather than phyllosilicates, formed by aqueous alteration and subsequent dehydration is supported by the results of hydrothermal experiments conducted by Nomura and Miyamoto (1998). They hydrothermally altered various constituent minerals in CAIs and found that no phyllosilicates form from typical primary minerals in CAIs at any pH conditions and 200 °C, however, hydrous nepheline ($NaAlSiO_4 \cdot 1/2H_2O$) forms from melilite at highly alkaline conditions. They also found that nepheline forms by heating the hydrous nepheline at 700 °C for 24 hr. From these experimental results, we infer that nepheline in the CO3 chondrules formed initially as hydrous nepheline during aqueous alteration at relatively low temperatures and alkaline conditions, and the hydrous nepheline was subsequently dehydrated to nepheline. It is probable that hydrous nepheline in the CO3 chondrules largely formed at the initial stage of heating, during which aqueous fluids were available. The degree of hydrothermal activity must have increased with increasing degree of heating, and thus, the more thermally metamorphosed chondrites produced larger amounts of hydrous nepheline and eventually nepheline.

Fe-enrichment in olivines and pyroxenes in chondrules (Fe-metasomatism) is the major phenomenon that occurred as a result of thermal metamorphism of the host meteorites, as mentioned above. Thus, an important question is how the Fe-metasomatism is related to the nephelinization. Our study revealed that there is an overall systematic correlation between the degrees of nephelinization and Fe-metasomatism among chondrites. However, among chondrules within each chondrite, the degrees of nephelinization and Fe-metasomatism are not always well-correlated. For example, in weakly metamorphosed Kainsaz and Y-82050, there are many nepheline-free chondrules with olivine phenocrysts that are enriched in Fe similarly to those in nepheline-bearing chondrules. The lack of correlation within each chondrite suggests that the processes of nephelinization and Fe-metasomatism were not completely overlapped. We infer that the formation of nepheline was completed at the initial stage of heating during which aqueous fluids were available, and the major part of Fe-metasomatism occurred subsequently without the aid of aqueous fluids. Chizmadia et al. (2002) ascribed the observed variations of Fe enrichment in olivines in AOIs to a combination of aqueous alteration and thermal diffusion of Fe and Mg. They suggested that the initial formation of Fe-rich olivine occurred through aqueous alteration, and subsequently, as the degree of thermal metamorphism increased, thermal diffusion became prevailing. This two-stage alteration model appears to be consistent with our interpretation.

We propose the following model for the formation of nepheline in chondrules in the CO3 chondrites. In a portion (or portions) of the CO parent body where a small amount of water or ice existed, as thermal metamorphism took place, hydrothermal activity started, and Na was mobilized and transported from matrix to chondrules through aqueous fluids and reacted with glass and plagioclase in mesostases to form hydrous nepheline. We are currently uncertain whether there was a specific reservoir of Na in the parent body. Increasing amounts of hydrous nepheline were produced with the increasing degree of hydrothermal activity, which was probably correlated with the degree of heating. A minor degree of Fe-enrichment in olivine in chondrules also occurred in this hydrothermal process. The extent of nephelinization reactions probably differed locally on small scales in the parent body due to heterogeneous distribution of aqueous solutions or availability of Na. Shortly, continuous heating must have resulted in exhaustion of water, and the aqueous alteration ceased. Subsequently, as the degree of heating increased, hydrous nepheline was dehydrated to nepheline. After completion of dehydration, the heating mainly controlled the metamorphism with thermal diffusive exchange of atoms without the aid of fluids.

Comparison to CAIs in CO3 chondrites

Previous studies showed that nepheline and sodalite in CAIs in CO3 chondrites have been produced mainly by replacing melilite, anorthite, fassaite, and spinel (Tomeoka et al. 1992; Kojima et al. 1995; Russell et al. 1998). The CAIs tend to increase in bulk Fe content with an increasing degree of Na-metasomatism; spinel and high-Ca pyroxene have been converted to hercynite and hedenbergite, respectively; perovskite has been converted to ilmenite; troilite particles have been produced in nepheline. Among these secondary minerals, hedenbergite and troilite are commonly associated with nephelinization in the CO3 chondrules. There is also a general tendency for the degrees of Na- and Fe-metasomatism in the CAIs to increase with increasing petrologic subtype of the host chondrites (Kojima et al. 1995; Russell et al. 1998; Itoh and Tomeoka 1998). These trends of mineralogical and compositional changes are consistent with those observed in the chondrules presently studied. Taking into consideration that chondrules in the CO3 chondrites have been subjected to such a significant degree of parent body nephelinization, we infer that CAIs in the chondrites could not escape the metasomatic effects and the major part of nepheline and sodalite in them formed on the parent body.

CONCLUSIONS

Our study has revealed that chondrules in the unmetamorphosed CO3.0 chondrite contain almost no nepheline, while those in the metamorphosed CO3.2–3.7 chondrites contain various amounts of nepheline as a secondary alteration product of mesostasis glass and plagioclase. Our modal analyses, bulk chemical analyses, and mineralogical observations of chondrule mesostases in those chondrites show that there is an apparent systematic tendency for the degree of nephelinization in the chondrules to increase with increasing petrologic subtype of the host chondrites. We conclude that nepheline in chondrules in the CO3 chondrites has formed largely as a result of effects related to heating on the meteorite parent body. We suggest that nepheline in the chondrules initially has formed as hydrous nepheline through aqueous alteration that occurred at the early stage of heating, and hydrous nepheline was subsequently dehydrated. The hydrothermal activity presumably became increasingly active with an increasing degree of heating, and thus, more thermally metamorphosed CO3 chondrites produced larger amounts of nepheline. The aqueous alteration probably occurred under the presence of relatively small amounts of aqueous fluids, and thus, aqueous fluids did not infiltrate the meteorite parent body extensively, and the alteration ceased shortly after heating, due to exhaustion of fluids. Consequently, the resultant effects of alteration were spatially very limited and heterogeneous on small scales. This implies that CO3 chondrites have gone through a three-stage alteration process on their parent bodies: 1) low-grade aqueous alteration; 2) thermal dehydration; and 3) thermal diffusive exchange of atoms without the aid of fluids.

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