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Evolution of protoplanetary disk inferred from ²⁶Al chronology of individual chondrules

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Abstract–We review the ²⁶Al ages of chondrules in various type 3.0 chondrites. The ²⁶Al ages of chondrules are 1–3 Myr after calcium-aluminum-rich inclusion (CAI) for LL3.0, CO3.0, and Acfer 094 (Ungrouped C 3.0). Available data for chondrules in CR chondrites indicate that many chondrules are relatively younger (\geq 3 Myr), although data from chondrules in CR3.0 are not yet available to confirm their younger ages. The total ranges for the ²⁶Al ages of chondrules in a single chondrite group are more than 0.5–1 Myr. However, most chondrules show relatively narrow range of ages in a single chondrite group (0.2–0.4 Myr, 1 SD), which might be short enough to preserve the group-specific chemical and isotope signatures against radial diffusion of solid in the disk. Distinct oxygen isotope reservoirs might exist in the protoplanetary disk simultaneously, which could be spatially separated.

INTRODUCTION

The timing of chondrule formation has been estimated to be 2-3 million years after the formation of the oldest calcium-aluminum-rich inclusions (CAIs) based on both Pb-Pb absolute ages (e.g., Amelin et al. 2002: Connelly et al. 2008) and the relative chronometers using an extinct nuclide ²⁶Al (Russell et al. 1996; Kita et al. 2000). This time scale is similar to the lifetime of a classical T Tauri (CTT) stars (e.g., Feigelson and Montmerle 1999), during which a circumstellar disk contains dust and gas. The chondrule formed by the transient heating of precursor solids, most likely by the shock wave generated in the protoplanetary disk that was enriched in dust more than 100-1000 times of the solar compositions (e.g., Connolly and Love 1998; Cuzzi and Alexander 2006). Dust regions in which chondrules formed could be dense enough to have a self-gravity, so that a planetesimal might have formed immediately after the chondrule formation periods (Alexander et al. 2008).

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Extensive studies have been made on the ²⁶Al ages of chondrules from different groups of chondrites (e.g., Kita et al. 2005a; Krot et al. 2009; reference therein). Most data are obtained by the internal isochron method using secondary ion mass spectrometer (SIMS), in which initial ²⁶Al/²⁷Al ratios at the time of last melting events are determined from the excess ²⁶Mg in plagioclase (anorthite-albite: CaAl₂Si₂O₈-NaAlSi₃O₈) or glass with high ²⁷Al/²⁴Mg ratios. The relative ages of chondrules can be resolved as short as 0.1-0.2 Myr if the slope of the isochron is determined precisely (e.g., Kurahashi et al. 2008a; Villeneuve et al. 2009). The published ²⁶Al ages of chondrules indicate that there are measurable age differences among chondrules within a single meteorite, at least 0.5 Myr to possibly 1-2 Myr (e.g., Mostefaoui et al. 2002; Kita et al. 2005b; Kurahashi et al. 2008a). An age difference of more than 0.5 Myr is much longer than the time scale of accretion of a planetesimals from a dense dust layer under nonturbulent disk conditions, indicating that the environment of chondrule-forming regions would be at least weakly turbulent (Cuzzi et al. 2008). Thus, the ²⁶Al-²⁶Mg dating of chondrules may provide useful information on the evolution of protoplanetary disk.

THE PRINCIPLE OF ²⁶AI RELATIVE CHRONOLOGY OF CHONDRULES

Kita et al. (2000, 2003, 2005b), Mostefaoui et al. (2002), Kurahashi et al. (2008a), and Ushikubo et al. (2010) established protocols for the ²⁶Al-²⁶Mg dating of chondrules in unequilibrated ordinary and carbonaceous chondrites using IMS-1270/1280 ion microprobes. In these measurements, internal ²⁶Al-²⁶Mg isochrons of individual chondrules were obtained from plagioclase or glass with high ${}^{27}\text{Al}/{}^{24}\text{Mg}$ ratios (≥ 30), which show a few to several [%]₂₆Mg excesses, and olivine and/or pyroxene, which define the origin of the isochron diagram. The Alrich phases are sampled with a 3-5 µm diameter primary beam and secondary Mg ions ($^{24}Mg < 10^5 \text{ cps}$) are detected by Electron Multiplier in ion counting mode, which usually takes 3-8 h per single spot analyses to achieve precision of excess ^{26}Mg as good as 0.5-1% (e.g., Kurahashi et al. 2008a). The Mg-rich minerals are analyzed using multicollection Faraday cup detectors with a precision of ${}^{26}Mg/{}^{24}Mg$ isotope ratios at the levels from 0.05% (15 µm spots using IMS-1280; Ushikubo et al. 2010) to 0.2% (5 µm spots using IMS-1270; Kita et al. 2000).

Mg contents in glass or plagioclase in chondrules would be zoned at micrometer-scale due to igneous fractionation (e.g., Kurahashi et al. 2008a), resulting in variable ²⁷Al/²⁴Mg ratios among these phases. If ²⁶Al/²⁷Al ratios within a chondrule were homogeneous at the time of solidification, the correlated ²⁶Mg excesses with variable ²⁷Al/²⁴Mg ratios may be observed as a result of in situ decay of ²⁶Al to ²⁶Mg (halflife = 7.05×10^5 yr; Norris et al. 1983). The slope of the isochron corresponds to the initial ²⁶Al/²⁷Al ratio at the time of last melting of the chondrule. An example of ²⁶Al-²⁶Mg isotope data of a chondrule is shown in Fig. 1 (data from Ushikubo et al. 2010). By assuming the homogeneous distribution of ²⁶Al in the solar system and the solar system initial ${}^{26}\text{Al}/{}^{27}\text{Al}$ ratio of 5×10^{-5} according to the canonical initial ratio of CAIs (MacPherson et al. 1995), the relative ²⁶Al age (Δt) of individual chondrules is calculated from the equation below.

$$\Delta t \,(\mathrm{Myr}) = ln \left[\frac{({}^{26}\mathrm{Al}/{}^{27}\mathrm{Al})_{\mathrm{CAI}}}{({}^{26}\mathrm{Al}/{}^{27}\mathrm{Al})_{\mathrm{Chondrule}}} \right] \times \frac{0.705}{ln(2)}$$
(1)

Recent re-evaluation of bulk CAI isochron indicates that the initial ${}^{26}\text{Al}/{}^{27}\text{Al}$ ratio of the solar system is $(5.23 \pm 0.13) \times 10^{-5}$ (Jacobsen et al. 2008). Similar initial ${}^{26}\text{Al}/{}^{27}\text{Al}$ ratios were estimated from the internal isochron of pristine unmelted CAIs (MacPherson et al. 2010a, 2010b). The small difference from conventional canonical value of 5×10^{-5} is at the level of 5%, which

(b) 5 4 (d) (26AI/27AI)0=(7.6±1.8)×10-6 3 δ²⁶Mg 2 1 plagioclase 0 olivine, pyroxene -1 10 30 40 50 0 20 60 27 Al/24 Ma

Fig. 1. An example of 26 Al- 26 Mg dating of chondrules (after Ushikubo et al. 2010). a) Backscattered electron (BSE) image of a type I chondrule G73 in Acfer 094. A scale bar is 100 µm. BSE images of ion probe pits with high intensity beam on olivine (b) and with low intensity beam on plagioclase (c). Scale bars are 20 µm. d) The internal isochron of chondrule G73. Error bars for plagioclase analyses are 2 SE of the mean of 300 cycles of Mg isotope analysis in a single spot.

changes Δt only by +0.05 Myr and is insignificant compared to typical uncertainties of the relative ages of chondrules (≥ 0.2 Myr). Therefore, relative ages shown in this article are calculated by applying ($^{26}\text{Al}/^{27}\text{Al})_{CAI} =$ 5.0×10^{-5} to Equation 1, which are consistent with most literature data. We should note that ^{26}Al relative chronology of meteorites often used half-life of ^{26}Al to be 7.2×10^5 or 7.3×10^5 yr. These values are slightly longer than the value of Norris et al. (1983), which is widely used for cosmogenic nuclide studies. Nishiizumi (2004) noted the average of three direct half-life measurements to be $(7.08 \pm 0.17) \times 10^5$ yr, which is very close to the value of Norris et al. (1983). Therefore, we applied the value of 7.05×10^5 yr in the above equation. The Δt from Equation 1 decreases by 3.5% compared to those using the half-life of 7.3×10^5 yr, equivalent to 0.1 Myr reduction of age for chondrules with $\Delta t \sim 3$ Myr.

As shown in Fig. 1, the ranges of ${}^{27}\text{Al}/{}^{24}\text{Mg}$ ratios in glass and plagioclase in a single chondrule are relatively limited, so that slopes of the isochrons are often calculated from a "two-point line" connecting data from Mg-rich phases (olivine/pyroxene) at the origin and the cluster of data from Al-rich phases (plagioclase/glass) with ²⁶Mg excesses. This is potentially dangerous if Mg isotope re-equilibration was not achieved throughout the chondrule during the last melting event. We usually analyze Al-rich phases with ${}^{27}\text{Al}/{}^{24}\text{Mg}$ ratios higher than 30 that typically show the excess ${}^{26}\text{Mg}$ more than 1%, so that the slope of the isochron is not sensitive to the initial $^{26}Mg/^{24}Mg$ ratios at 0.1% levels. We also restrict chondrule samples in type 3 chondrites to petrologic subtypes 3.0 (e.g., Kita et al. 2000; Kurahashi et al. 2008a: Ushikubo et al. 2010), which are least altered or thermally metamorphosed in parent bodies. To focus our investigation and discussions on the primary formation ages of chondrules, we review the ${}^{26}\text{Al}{}^{-26}\text{Mg}$ data on chondrules from type 3.0 chondrites.

OVERVIEW OF ²⁶AI AGES OF CHONDRULES

The inferred initial ²⁶Al/²⁷Al ratios of chondrules have been obtained from L, LL, CO, CV, CR, and Acfer 094 with most values range from 1.0×10^{-5} to 5×10^{-6} , corresponding to the relative age from 1.7 to 2.4 Myr after CAIs (e.g., Kita et al. 2000; Mostefaoui et al. 2002; Kunihiro et al. 2004; Rudraswami and Goswami 2007; Kurahashi et al. 2008a; Rudraswami et al. 2008; Hutcheon et al. 2009: Ushikubo et al. 2010). The approximately 2 Myr time difference between CAIs and chondrules is generally consistent with the differences in their Pb-Pb ages (Amelin et al. 2002; Connelly et al. 2008), although more investigations are needed for their ²³⁸U/²³⁵U ratios since the realization of uranium isotope heterogeneity in the early solar system (Brennecka et al. 2010). Kita et al. (2000) and Huss et al. (2001) indicate that the ²⁶Al-²⁶Mg systems of chondrules in mildly metamorphosed ordinary chondrites (subtypes ≥ 3.4) were disturbed. Examples of disturbed isochrons from mildly metamorphosed chondrites are reported by Kita et al. (2004a, 2004b) that are obtained from glass or plagioclase data with ²⁷Al/²⁴Mg ratios higher than 100, which are relatively uncommon in chondrules from type 3.0 chondrites. In the Ningqiang ungrouped carbonaceous chondrite, plagioclase-olivine inclusions (one type of chondrule) show disturbed Al-Mg isochron from plagioclase data with the ²⁷Al/²⁴Mg ratios of 100-300 (Kita et al. 2004a). In Yamato (Y)-82038 (H3.2), a glassy type IA chondrule shows high ²⁷Al/²⁴Mg ratios of 90-140 with only a limited amount of ²⁶Mg excess (Kita et al. 2004b). Huss et al. (2001) showed absence of ²⁶Mg excesses from glass and plagioclase in chondrules from Chainpur (LL3.4) with high ²⁷Al/²⁴Mg ratios up to 600. In type 3.0 chondrites, the ${}^{27}\text{Al}/{}^{24}\text{Mg}$ ratios of glass higher than 100 are only observed when mesostasis contain abundant high-Ca pyroxene microcrystallites, which might have formed as quench crystals during rapid cooling of melt (example is shown in Kita et al. 2000 in their fig. 2). The 27 Al/ 24 Mg ratios of plagioclase are typically < 100 in chondrules from type 3.0 chondrites, which are significantly lower than those in CAIs and basaltic achondrites, possibly due to higher bulk MgO contents and higher temperature of crystallization in chondrules. Therefore, recent data focus more on chondrules in type "3.0" chondrites (e.g., Kurahashi et al. 2008a; Villeneuve et al. 2009; Ushikubo et al. 2010). In Fig. 2, inferred initial ²⁶Al/²⁷Al ratios of chondrules in the literature are compiled for type 3.0 chondrites, Semarkona (LL3.0; Hutcheon and Hutchison 1989; Kita et al. 2000; Rudraswami et al. 2008; Villeneuve et al. 2009), Y-81020 (CO3.0; Kunihiro et al. 2004; Kurahashi et al. 2008a) and Acfer 094 (Ungrouped C; Hutcheon et al. 2009; Ushikubo et al. 2010). We note that the ²⁶Al/²⁷Al ratios of three plagioclase-bearing chondrules in Semarkona reported by Kita et al. (2000) are reduced by 20% compared to those originally reported due to re-evaluation of the Mg contents in plagioclase standard (see Kurahashi et al. 2008a). In addition, recent data from primitive CR2 chondrites (Nagashima et al. 2007, 2008; Kurahashi et al. 2008b; Hutcheon et al. 2009) are shown in the same figure together.

Most chondrule data in Fig. 2 show relative ages between 1 and 3 Myr after CAIs. No data indicate the relative age within 1 Myr from CAI formation, in contrast to bulk model Al-Mg ages of Allende chondrules as old as CAIs by Bizzarro et al. (2004). As discussed in Kita et al. (2005a), the relative ages estimated from bulk Al-Mg data with low ²⁷Al/²⁴Mg ratios (~ 0.1) may not provide meaningful values. There seem to be differences in the peak ²⁶Al/²⁷Al ratios among different group of chondrites; approximately 7×10^{-6} (~2 Myr after CAIs) for LL and CO, approximately 6×10^{-6} (~2.3 Myr after CAIs) for Acfer 094, and $\leq 3 \times 10^{-6}$ (≥ 3 Myr after CAIs) for CR2. In Table 1, the average and a standard deviation (1 SD) of ²⁶Al ages of chondrules in individual meteorites are shown. For chondrules in Semarkona (LL3.0), the ²⁶Al ages of Mg-rich (type I) chondrules and FeO-rich (type



Fig. 2. Compilation of the initial ²⁶Al/²⁷Al ratios inferred from internal isochron regressions of ²⁶Al-²⁶Mg data for chondrules from primitive chondrites. Error bars shown are 2 SE from isochron regression. Data from type 3.0 chondrites are shown except for those in CR chondrites. Al-rich, type I, and type II chondrules are shown as diamonds, circles, and squares, respectively. Open larger symbols are those from Kita et al. (2000), Kurahashi et al. (2008a), and Ushikubo et al. (2010) using either IMS-1270 at Geological Survey of Japan (GSJ) or IMS-1280 at WiscSIMS that uses 3–5 µm small spots with long counting time. Data from other SIMS laboratories are shown as smaller filled symbols. Data sources: Hutcheon and Hutchison (1989), Kita et al. (2000), Rudraswami et al. (2008), and Villeneuve et al. (2009) for Semarkona (LL3.0); Kunihiro et al. (2004) and Kurahashi et al. (2008a) for Y-81020 (CO3.0); Hutcheon et al. (2009), and Ushikubo et al. (2010) for Acfer 094; Nagashima et al. (2007, 2008), Kurahashi et al. (2008b), and Hutcheon et al. (2009) for CR chondrites. More than 60% of chondrules in CR chondrites do not show resolvable excess ²⁶Mg and at least 10 chondrules are younger than 3 Myr as indicated by the arrow.

Table 1. The All age distribution of chondrules.						
Data set	GSJ/Wisc ^b	1 SD	п	All ^c	1 SD	п
Semakona all	2.03	0.24	5	1.98	0.41	25
Semarkona type I	2.0	_	1	2.02	0.39	5
Semarkona type II	2.03	0.28	4	1.97	0.42	20
Y-81020 all	2.10	0.30	22	2.19	0.36	27
Y-81020 Al-rich	2.19	0.51	2			
Y-81020 type I	2.02	0.23	14			
Y-81020 type II	2.26	0.36	6	2.41	0.38	11
Acfer 094 all	2.11	0.24	10	2.12	0.37	12
Acfer 094 Al-rich	2.16	_	1	1.74	0.45	3
Acfer 094 type I	2.25	0.25	9			

Table 1. The ²⁶Al age distribution of chondrules^a

^aThe ²⁶Al ages are calculated by using the half-life of 7.05×10^5 yr and the initial solar system 26 Al/ 27 Al = 5.0×10^{-5} .

^bData sources: Kita et al. (2000), Kurahashi et al. (2008a), and Ushikubo et al. (2010).

^cAll data shown in Fig. 2.

II) chondrules are indistinguishable. For chondrules in Y-81020 (CO3.0), the ²⁶Al ages of Al-rich, type I and type II chondrules are statistically indistinguishable (see Table 1). Yet, there is an indication that type II chondrules in CO3.0 (2.4 ± 0.4 Myr, 1 SD) are slightly younger than type I chondrules in the same meteorite (2.0 ± 0.2 Myr, 1 SD).

In CR chondrites, many chondrules show the initial ${}^{26}\text{Al}/{}^{27}\text{Al}$ ratios that are lower than 3×10^{-6} (>3 Myr

after CAIs) and some do not show resolvable ²⁶Mg excess (Nagashima et al. 2007, 2008; Kurahashi et al. 2008b; Hutcheon et al. 2009). If the Al-Mg system in these chondrules was not disturbed since their last melting events, formation of some chondrules in CR chondrites postdates chondrules in LL, CO, and Acfer 094 by more than 1–2 Myr. However, many CR chondrites experienced aqueous alteration to some extent, and the effects on the Al-Mg system are not well



Fig. 3. The time scale of disk evolution and chronology of meteoritic materials. The disk time scale is from infrared spectroscopy of young star (after Feigelson and Montmerle 1999). Class I stage corresponds to protostars with time scales approximately 10^5 yr. The accretion rate of circumstellar materials remained relatively high and the outflows from the central star were active. Class II stage corresponds to classical T Tauri (CTT) stars with the time scale of $(0.5-3) \times 10^6$ yr. Circumstellar disk with dust and gas might have existed around the central star. Class III stage corresponds to weak-lined T Tauri (WTT) stars with the time scale of approximately 10^7 yr. Circumstellar disk disappeared due to planetary growth. The ²⁶Al ages of CAIs and chondrules match the time scales of class I and class II, respectively. Chondrules in CB_b might form during class III stage by impact between planetesimals (Krot et al. 2005).

understood. At least the Al-Mg data of some of chondrules studied previously are considered to be pristine based on chemical composition of plagioclase (Kurahashi et al. 2008b). There are two CR3.0 chondrites (Queen Alexandra Range 99177 and Meteorite Hills 00426) that did not experience significant aqueous and thermal metamorphism (Abreu and Brearley 2010). None of data shown in Fig. 2 are from these pristine CR3.0 chondrites. Data from these meteorites would test relatively late formation ages of chondrules in CR chondrites.

The Pb-Pb age of chondrules in metal rich CB_b chondrite Gujba is precisely determined to be 4562.7 ± 0.5 Myr (Krot et al. 2005). By comparing the Pb-Pb ages of Gujba chondrules to that of type B CAI from Northwest Africa 2364 (CV3) with the Pb-Pb age of 4568.7 \pm 0.3 Myr (Bouvier and Wadhwa 2010), the Gujba chondrules formed 6.0 ± 0.6 Myr after type B CAI formation. We should note that the absolute Pb-Pb ages of meteorites would be in error by more than a million years depending on their uranium isotope ratios (²³⁸U/²³⁵U; Brennecka et al. 2010). However, Yamashita et al. (2010) also obtained the Mn-Cr age of Gujba to be 4563.7 ± 1.2 and 4563.5 ± 1.1 Myr using angrites D'Orbigny and Lewis Cliff 86010 as time anchors, respectively, which are consistent with the Pb-Pb age within errors. No ²⁶Al-²⁶Mg data are available from CB_b



Fig. 4. Radial diffusion model (Cuzzi et al. 2010). Materials originally located at 2 and 4 AU might be retained within approximately 0.2 Myr, while they will be fully mixed after 1 Myr.

chondrites using SIMS. It may not be possible to resolve small ²⁶Mg excess in chondrule mesostasis for such young samples. The young formation age is interpreted as a result of vapor-melt plume produced by impact between planetesimals (Krot et al. 2005), so that their formation process may be very different from formation of other chondrules.

The relative ages of CAIs and chondrules inferred from their initial ${}^{26}\text{AL}/{}^{27}\text{Al}$ ratios are consistent with the time scale of CTT stars, in which dust layers form in the protoplanetary disk (Fig. 3). Except for cases like chondrules in CB_b chondrites, chondrules in primitive chondrites might record the processes in the protoplanetary disk. The small differences in peak of ${}^{26}\text{Al}$ ages among chondrules in different chondrite groups may indicate variation in the timing of chondrule forming events in different region of the disk, such as at different heliocentric distances. Planetesimal growth would have started subsequently to the chondrule formation (Alexander et al. 2008).

VARIATION WITHIN SINGLE CHONDRITE GROUPS

The variations of ²⁶Al ages among chondrules in a single chondrite group are reported to be more than 0.5–1 Myr from all groups of chondrites (e.g., Mostefaoui et al. 2002; Rudraswami and Goswami 2007; Rudraswami et al. 2008; Hutcheon et al. 2009; Villeneuve et al. 2009). According to radial diffusion model of turbulent disk (Cuzzi et al. 2010), solid materials within asteroidal belt regions (2–4 AU) would be fully mixed after 1 Myr

(Fig. 4). However, chondrules in different groups of chondrites show group-specific chemical and isotopic characters, such as limited range of bulk major element compositions (Grossman 1988) and oxygen three-isotope ratios (Clayton 1993). Occurrence of chondrules with different formation ages as long as 1 Myr seems to contradict the preservation of chondrules with similar chemical properties that accreted to a single parent asteroid. Therefore, it has been argued that relative ²⁶Al ages observed among chondrules in a single chondrite group resulted from disturbance of the ²⁶Al-²⁶Mg systems in their parent body.

Based on Mg self-diffusion experiments in anorthite (1200–1400 °C), LaTourrette and Wasserburg (1998) addressed that ²⁶Mg excesses in anorthite would be effectively preserved below 450 °C in the asteroidal bodies, which was likely the case for type 3.0 chondrites. By contrast, Mg diffusion in albitic plagioclase might be orders of magnitude faster than that in anorthite, as expected from enhanced Sr diffusion in albitic plagioclase than anorthite (Cherniak and Watson 1994; Giletti and Casserly 1994). The results of Mg tracer diffusion experiment in albitic glass (800-1020 °C: Roselieb and Jambon 2002) indicate that diffusion rate of Mg in albitic glass is nearly approximately 1000 times faster than that of anorthite crystal at the same temperatures. By extrapolating Mg diffusion data of albitic glass to lower temperatures, similar to what was done by LaTourrette and Wasserburg (1998), Mg diffusion would be effectively slow only below 300 °C (extrapolated diffusion rate is $D = 10^{-26} \text{ m}^2 \text{ s}^{-1}$, which is equivalent to that of anorthite at 450 °C from the experiments of LaTourrette and Wasserburg 1998). Thus, the closure of Al-Mg system could correlate with susceptibility of Mg diffusional exchange of measured Alrich phases at the metamorphic temperatures between 300 and 450 °C. Because the temperature of metamorphism for Y-81020 (CO3.05) is not well constrained and could be anywhere below 550 °C (see discussion by Kurahashi et al. 2008a), we cannot fully refute the possibility that some type II chondrules containing more Na-rich plagioclase were partially disturbed in their Al-Mg system while those of type I chondrules containing anorthite were intact. We may need more diffusion experiments in a wide range of compositions and at lower temperatures to better address the behavior of Al-Mg system in chondrules. In addition, some of type 3 chondrites experienced aqueous alterations that replaced anorthite by nepheline and sodalite (e.g., Krot et al. 1995), which would result in loss of ²⁶Mg excesses, although Mg diffusion could have been slow. Thus, detailed microscopic inspection of glass and plagioclase in chondrules is also very important for selecting chondrules that had minimal secondary processes in the asteroidal environments (e.g., Kurahashi et al. 2008a).



Fig. 5. The ²⁶Al-²⁶Mg isochron diagrams of chondrules in Semarkona (after Kita et al. 2000). a) Type II chondrule CH4 containing albitic glass. Correlated ²⁶Mg excesses are observed from multiple analyses in glass. b) Type I chondrule CH3 containing anorthite. The ²⁷Al/²⁴Mg ratios of anorthite do not spread significantly. Due to re-evaluation of Mg contents in the plagioclase standard (see Kurahashi et al. 2008), inferred ²⁶Al/²⁷Al ratios of CH3 were revised to be $(7.0 \pm 2.5) \times 10^{-6}$, which is approximately 20% lower than the value originally reported in Kita et al. (2000). Error bars indicate 2 SE of individual analyses.

Some of the least metamorphosed ordinary chondrites, like Semarkona (LL3.0), might not experienced metamorphic temperature more than 300 °C (Alexander et al. 1989), so that ²⁶Mg excess in albitic plagioclase and glass would be preserved in chondrules. Semarkona is known as the least metamorphosed ordinary chondrite (Grossman and Brearley 2005; Kimura et al. 2008), although the meteorite also shows evidences of aqueous alteration (e.g., Alexander et al. 1989; Grossman et al. 2000, 2002). Kita et al. (2010) argued that the oxygen isotope ratios in the glassy mesostasis of chondrule from Semarkona likely partially exchanged with aqueous fluid within the parent body. The oxygen isotope exchange between silicate glass and fluid at low temperature could be faster than Mg diffusion in glass, because the rate of oxygen isotope exchange is enhanced by the orders of magnitude due to fast diffusion of neutral water molecules in silicate structure (e.g., Cole and Chakraborty 2001). However,



Fig. 6. Comparison of oldest and youngest type I chondrules in type 3.0 carbonaceous chondrites. a) Y-81020 (Kurahashi et al. 2008a). b) Acfer 094 (Ushikubo et al. 2010). Error bars indicate 2 SE of individual analyses. The slopes of the isochrones are determined by anorthitic plagioclase data, which are unlikely disturbed by parent body processes. Therefore, both meteorites contain chondrules that formed nearly 1 Myr apart.

there are no diffusion data for oxygen and magnesium isotope at low temperature and in the presence of aqueous fluid that can be used to compare the behavior of oxygen and magnesium isotope in chondrule glass. Therefore, the effect of aqueous alteration on Al-Mg isotope system is difficult to evaluate. For five chondrules in Semarkona that were reported for ²⁶Al dating by Kita et al. (2000), there is no obvious correlation between phases in chondrules with albitic glass show the relative ages of 1.8 ± 0.2 Myr (CH4; Fig. 5a) and 2.1 ± 0.3 Myr (CH36) that are determined from correlated ²⁶Mg excess with ²⁷Al/²⁴Mg ratios.

These data are not younger than two anorthite-bearing chondrules CH3 (IAB; Fig. 5b) and CH23 (IIAB) with the ²⁶Al ages of 2.0 (-0.3/+0.5) and 1.9 (-0.4/+ 0.8) Myr, respectively. Therefore, we consider that the ²⁶Al-²⁶Mg systems in chondrules from type 3.0 chondrites were preserved since their formation, but not affected by metamorphism in the parent body.

Data obtained from anorthite in type I chondrules in pristine chondrites, such as Y-81020 (CO3.0) and Acfer 094 (Ungrouped C) with minimal thermal metamorphism and aqueous alteration, are most likely to be preserved because of slow Mg diffusion in anorthite (LaTourrette and Wasserburg 1998). In Fig. 6, data from two anorthite-bearing type I chondrules with the highest and lowest inferred initial ²⁶Al/²⁷Al ratios are compared for both Y-81020 and Acfer 094. We use data sets from Kurahashi et al. (2008a) and Ushikubo et al. (2010) for Y-81020 and Acfer 094, respectively, with high precision isotope analyses ($\leq 1\%$) from small spots (3–5 µm) that took 3–8 h for one analysis. In Y-81020, the ²⁶Al ages of chondrules Y10 and Y01 are 1.6 (-0.2/+0.3) and 2.4 ± 0.3 Myr, respectively. The difference between two ages $\Delta t(Y10 - Y01)$ is calculated to be 0.8 (+0.4/-0.5) Myr. In Acfer 094, the ²⁶Al ages of G39 and G95 are 1.7 ± 0.2 and 2.4 (-0.3/+0.4) Myr, respectively. The difference between two ages $\Delta t(G39 - G95)$ is calculated to be 0.6 (+0.3/-0.5) Myr. Thus, both carbonaceous chondrite parent bodies do contain chondrules that last melted at different times, to possibly nearly a million years apart as is the case with Semarkona (LL3.0) (e.g., Kita et al. 2000; Villeneuve et al. 2009).

Although there are resolvable time differences between some chondrules in a single meteorite, type I chondrule data from Y-81020 and Acfer 094 show a tight cluster within each meteorite. As shown in Table 1, type I chondrules from Y-81020 (Kurahashi et al. 2008a) and Acfer 094 (Ushikubo et al. 2010) show mean ages of 2.02 ± 0.23 Myr (1 SD, n = 14) and 2.25 ± 0.25 Myr (1 SD, n = 9), respectively. Therefore, the ²⁶Al ages of most type I chondrules in a single meteorite are indistinguishable and they formed within a narrow time scale (i.e., < 0.5 Myr). The 1 SD of chondrule formation age distribution is equivalent to the time scale of migration in the radial diffusion model of Cuzzi et al. (2010) shown in Fig. 4. For diffusion time scale of 0.2 Myr, the model predicts that majority of solid materials originally located at 2 and 4 AU are hardly mixed with each other (Fig. 4). Thus, time scale of chondrule-forming events inferred from the ²⁶Al-²⁶Mg dating of chondrules seems to be very reasonable in preserving their group-specific chemical and isotope signatures.

5

EVOLUTION OF OXYGEN ISOTOPE RESERVOIRS

As shown in Fig. 2, Kurahashi et al. (2008a) argued that the ²⁶Al ages of chondrules from Y-81020 (CO3.0) are similar to those from LL3.0–3.1 chondrites (Kita et al. 2000, 2005b; Mostefaoui et al. 2002), although oxygen isotope ratios in these chondrules are very different (e.g., Clayton 1993). They concluded that chondrules in LL and CO chondrites formed at the same period, while the LL and CO chondrite-forming regions could be spatially separated with distinct oxygen isotope reservoirs. This is the first clear evidence indicating that asteroidal regions within the protoplanetary disk were heterogeneous in oxygen isotope ratios.

Libourel and Krot (2007) suggested that olivine grains in type I chondrules were relict fragments of differentiated planetesimals that survived against melting of type I chondrules in the solar nebula. Subsequently, Chaussidon et al. (2008) reported systematic variation in the oxygen three-isotope ratios of olivine, low-Ca pyroxene, and glass in the individual chondrules from CV and CR chondrites. They concluded that oxygen three-isotope ratios in olivine and low-Ca pyroxene generally follow the equations $\delta^{18}O_{\text{pyroxene}} = 2/3 \times \delta^{18}O_{\text{olivine}} + \delta^{18}O_{\text{gas}}$ and $\delta^{17}O_{\text{pyroxene}} = 2/3 \times \delta^{17}O_{\text{olivine}} + \delta^{17}O_{\text{gas}}$, in which $\delta^{18}O_{\text{gas}}$ and $\delta^{17}O_{\text{gas}}$ represent oxygen isotope ratios of SiO gas in the solar nebula. In their more recent paper (Libourel and Chaussidon 2011), they suggested that there were several broken planetesimals that provided precursor Mg-rich olivine grains with specific oxygen isotope ratios in localized disk regions. [Correction made here after online publication.]

Recently, Ushikubo et al. (2009, 2011) obtained high precision ($\leq 1_{00}^{\circ}$) SIMS oxygen three-isotope analyses of minerals and glass in chondrules from Acfer 094. In contrast to data by Chaussidon et al. (2008), most olivine grains in type I chondrules are in oxygen isotope equilibrium with low-Ca pyroxene phenocrysts, high Ca-pyroxene, anorthite, and glass in the chondrule mesostasis. The majority of chondrules are internally homogeneous in oxygen isotopes with rare occurrence of relict olivine grains with distinct isotope ratios. Similar results showing oxygen isotope equilibrium between olivine and low-Ca pyroxene in type I chondrules have been obtained from chondrules in Y-81020 (CO3.0; Tenner et al. 2011). Thus, our high precision oxygen three-isotope analyses of chondrules indicate that Mgrich olivine grains in type I chondrules are not necessarily "relict" grains, rather most of olivine phenocrysts would be crystallized from a chondruleforming melt with homogeneous oxygen isotope ratios.

Ushikubo et al. (2011) further found that the average $\Delta^{17}O$ (= $\delta^{17}O - 0.52 \times \delta^{18}O$) values of individual



ratios of chondrules in Acter 094, ungrouped C chondrite (after Ushikubo et al. 2010). a) Oxygen three-isotope ratios. Error bars indicate 2 SE of individual analyses. Chondrules in Acter 094 show at least two major oxygen isotope reservoirs; at $\Delta^{17}O \sim -5\%_{00}, \sim -2\%_{00}$, and possibly $\sim 0\%_{00}$ as a third component. b) The initial ²⁶Al/²⁷Al ratios. Error bars shown are 2 SE from isochron regression. The majority of chondrules formed at the same time regardless of their oxygen isotope ratios, with the initial ²⁶Al/²⁷Al ratios of approximately 5×10^{-6} that corresponds to approximately 2.3 Myr after CAI formation. Oxygen isotope ratios of G76 and G18 are not shown in Fig. 7a, but they belong to $-5\%_{00}$ group (Ushikubo et al. 2011).

chondrules in Acfer 094 show two main peaks at -5% and -2%, and possibly a third one at approximately 0% (Fig. 7), which may represent that the local oxygen isotope reservoirs existed in the disk. Libourel and Chaussidon (2011) also found peaks in Δ^{17} O values among olivine in type I chondrules that are specific to chondrite groups, which may be consistent with the observation in Acfer 094 by Ushikubo et al. (2011). As mentioned earlier, Libourel and Chaussidon (2011) interpreted the oxygen isotope signatures of Mg-rich olivine grains to represent several planetesimals that had been broken into pieces and spread in the protoplanetary disk, because they assumed that

olivine grains were mainly "relict." Although new oxygen isotope data by Ushikubo et al. (2011) alone may not fully refute the planetary origin for the olivine in type I chondrules, Whattam et al. (2008) also argued against the planetary origin suggested by Libourel and Krot (2007) mainly from grain size of olivine being much smaller than what would be expected from differentiated bodies.

Coordinated oxygen isotope analyses and ²⁶Al dating of the same chondrules would help us to understand the relationship between two isotope reservoirs, whether they existed at the same time or they evolved from one to the other. Currently, only a limited number of chondrules have both high precision ${}^{26}Al$ ages and oxygen three-isotope data. The initial ${}^{26}Al/{}^{27}Al$ ratios of 10 type I chondrules analyzed by Ushikubo et al. (2010) include those with Δ^{17} O values of $-5\%_{00}$, $-2\%_{00}$, and $0\%_{00}$, as well as one chondrule with internally heterogeneous oxygen isotope ratio (fig. 2a in Ushikubo et al. 2009). As shown in Fig. 7b, there is no obvious correlation between the $^{26}\text{Al}/^{27}\text{Al}$ ratios and $\Delta^{17}\text{O}$ values, indicating that chondrule formation occurs at the same time in different isotope reservoirs. The parent asteroid of Acfer 094 might have collected solid materials from these multiple isotope reservoirs. Tenner et al. (2011) obtained preliminary oxygen three-isotope analyses of chondrules in Y-81020 (CO3.0) that were dated by Kurahashi et al. (2008a) for 26 Al ages. The results show bimodal Δ^{17} O values at $-5^{\circ}_{\circ 00}$ and -2% without obvious correlation with their ²⁶Al ages, similar to the case observed in Acfer 094.

Oxygen three-isotope systematics of dated chondrules would be important to evaluate the amount of mixing in the different regions of the disk by radial diffusion., For example, a large olivine fragment (G39) from Acfer 094 shows the oldest ²⁶Al age, while its Δ^{17} O value of approximately 0% is a relatively minor group compared to others with Δ^{17} O value of -5% and -2% (Ushikubo et al. 2010). One potential interpretation is that this older chondrule fragment was derived from other regions with distinct oxygen isotope reservoirs, such as from ordinary chondrite regions. In other instances, Tenner et al. (2011) reported the Δ^{17} O value of youngest type I barred olivine (BO) chondrule Y01 from Y-81020 to be -3.2%, which is intermediate between the two major reservoirs. Considering that chondrules formed in these two distinct isotope reservoirs and accreted in a single parent asteroid for CO chondrites, materials from two isotope reservoirs would have been migrated and mixed at a late stage of chondrule formation in the disk. In terms of oxygen three-isotopes, the BO chondrule Y01 could represent the youngest generation of chondrules in the local disk regions immediately before the parent asteroid formation. These arguments for the oldest and youngest type I chondrules in Acfer 094 and Y-81020, respectively, are too speculative at this moment because they are based on only a small number of data. Nonetheless, these data underscore the importance of high precision isotope data presented in this review.

Radial transportation mechanisms in the protoplanetary disk are also important in understanding the origin of refractory silicates in the Stardust mission returned samples (comet 81P/Wild 2), which are similar to CAIs and chondrules in primitive meteorites (Zolensky et al. 2006; Nakamura et al. 2008). These high temperature silicates are not expected to form in the outer solar system under the conditions of current disk models, so that they were probably transported from the inner solar nebula (Ciesla 2007). Nakashima et al. (2011a) indicated that most of coarser grained ($\geq 5 \,\mu m$) olivine and low-Ca pyroxene particles from Wild 2 that were analyzed for oxygen three-isotope ratios show Δ^{17} O value of approximately -2%, one of two major oxygen isotope reservoirs of chondrules in Acfer 094. Ushikubo et al. (2011) pointed out that the isotope reservoirs with Δ^{17} O value of approximately -2% could be widely spread in asteroidal regions, because $\Delta^{17}O = -2^{\circ}_{/00}$ is frequently observed in chondrules in CR, CH, and CB chondrites (e.g., Krot et al. 2006a, 2006b, 2010; Nakashima et al. 2011b). Thus, further systematic investigations on the combined high quality ²⁶Al age and oxygen three-isotope studies in chondrules from all types of chondrite classes are promising for better understanding of the nature of oxygen isotope reservoirs in the protoplanetary disk.

CONCLUSIONS

We reviewed the ²⁶Al ages of chondrules in various types of the least metamorphosed chondrites. The ²⁶Al ages of chondrules are 1–3 Myr after CAI for LL3.0, CO3.0, and Acfer 094. Available data for CR include the relative ages approximately 2 Myr, although more than half of chondrules are \geq 3 Myr. Systematically younger ages of chondrules in CR need to be confirmed by analyses of chondrules in CR3.0. The total range of ²⁶Al ages of chondrules in a single chondrite is at least 0.5– 1 Myr. However, most chondrules show relatively narrow range of ages (0.2–0.4 Myr, 1 SD), consistent with preserving the group-specific chemical and isotope signatures. Distinct oxygen isotope reservoirs might exist in the protoplanetary disk simultaneously, which could be spatially separated.

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