

OXYGEN ISOTOPIC COMPOSITION OF THE SUN AND MEAN OXYGEN ISOTOPIC COMPOSITION OF THE PROTOSOLAR SILICATE DUST: EVIDENCE FROM REFRACTORY INCLUSIONS

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ABSTRACT

Preliminary analysis of the oxygen isotopic composition of the solar wind recorded by the *Genesis* spacecraft suggests that the Sun is ¹⁶O-rich compared to most chondrules, fine-grained chondrite matrices, and bulk compositions of chondrites, achondrites, and terrestrial planets ($\Delta^{17}\text{O} = -26.5\text{‰} \pm 5.6\text{‰}$ and $-33\text{‰} \pm 8\text{‰}$ (2σ) versus $\Delta^{17}\text{O} \sim \pm 5\text{‰}$). The inferred ¹⁶O-rich composition of the Sun is similar or slightly lighter than the ¹⁶O-rich compositions of amoeboid olivine aggregates and typical calcium–aluminum-rich inclusions (CAIs) from primitive (unmetamorphosed) chondrites ($\Delta^{17}\text{O} = -24\text{‰} \pm 2\text{‰}$), which are believed to have condensed from and been melted in a gas of approximately solar composition (dust/gas ratio ~ 0.01 by weight) within the first 0.1 Myr of the solar system formation. Based on solar system abundances, 26% of the solar system oxygen must be initially contained in dust and 74% in gas. Because solar oxygen is dominated by the gas component, these observations suggest that oxygen isotopic composition of the solar nebula gas was initially ¹⁶O-rich. Due to significant thermal processing of the protosolar molecular cloud silicate dust (primordial dust) in the solar nebula and its possible isotope exchange with the isotopically evolved solar nebula gas, the mean oxygen isotopic composition of the primordial dust is not known. In CO self-shielding models, it is *assumed* that primordial dust and solar nebula gas had initially identical, ¹⁶O-rich compositions, similar to that of the Sun ($\Delta^{17}\text{O} \sim -25\text{‰}$ or -35‰), and solids subsequently evolved toward the terrestrial value ($\Delta^{17}\text{O} = 0$). However, there is no clear evidence that the oxygen isotopic compositions of the solar system solids evolved in the direction of increasing $\Delta^{17}\text{O}$ with time and no ¹⁶O-rich primordial dust have yet been discovered. Here we argue that the assumption of the CO self-shielding models that primordial dust and solar nebula gas had initially identical ¹⁶O-rich compositions is incorrect. We show that igneous CAIs with highly fractionated oxygen isotopic compositions, fractionation and unidentified nuclear effects (FUN), and fractionation (F) CAIs, have $\Delta^{17}\text{O}$ ranging from -0.5‰ to -24.8‰ . Within an individual FUN or F CAI, oxygen isotopic compositions of spinel, forsterite, and pyroxene define a mass-dependent fractionation trend with a constant $\Delta^{17}\text{O}$ value. The degree of mass-dependent fractionation of these minerals correlates with the sequence of their crystallization from the host CAI melt. These observations and evaporation experiments on CAI-like melts indicate that FUN and F CAIs formed by melting of solid precursors with diverse $\Delta^{17}\text{O}$ values in vacuum (total pressure $< 10^{-6}$ atm). We interpret the observed range of $\Delta^{17}\text{O}$ values among FUN and F CAIs as the result of varying degrees of equilibration between ¹⁶O-poor dust and ¹⁶O-rich nebular gas and suggest the former is characteristic of the primordial dust. The distinctly different oxygen isotopic compositions of the primordial solar nebula dust and gas could have resulted from Galactic chemical evolution or from pollution of the protosolar molecular cloud by a massive star ($> 50\text{ }M_{\odot}$) ejecta. The ¹⁶O-depleted compositions of chondrules, fine-grained matrices, chondrites, and achondrites compared to the Sun's value reflect their formation in the protoplanetary disk regions with enhanced dust/gas ratio (up to $10^5 \times$ solar).

Key words: comets: general – dust, extinction – Earth – ISM: abundances – meteorites, meteors, meteoroids – Sun: fundamental parameters

1. INTRODUCTION

Oxygen has three stable isotopes: ¹⁶O, ¹⁷O, and ¹⁸O. Oxygen isotopic compositions of the solar system materials are typically expressed as permil deviations from the standard mean ocean water (SMOW) value, $\delta^{17}\text{O}$ and $\delta^{18}\text{O}$: $\delta^i\text{O} = ((^i\text{O}/^{16}\text{O})_{\text{sample}} / (^i\text{O}/^{16}\text{O})_{\text{SMOW}} - 1) \times 1000$, where $i = 17$ and 18 . On a three-isotope oxygen diagram, $\delta^{17}\text{O}$ versus $\delta^{18}\text{O}$, all terrestrial samples plot along a line with slope of ~ 0.52 , called the terrestrial fractionation (TF) line, whereas the major components of

primitive (unmetamorphosed) chondrites (calcium–aluminum-rich inclusions (CAIs), amoeboid olivine aggregates (AOAs), chondrules, and fine-grained matrices) plot along a line with a slope of ~ 1 , called the carbonaceous chondrite anhydrous mineral (CCAM) line. Refractory inclusions (CAIs and AOAs) are typically ¹⁶O-enriched relative to fine-grained matrices and most chondrules (e.g., Yurimoto et al. 2008). The degree of ¹⁶O enrichment or depletion of extraterrestrial materials is commonly expressed as deviation from the TF line, $\Delta^{17}\text{O} = \delta^{17}\text{O} - 0.52 \times \delta^{18}\text{O}$.

Although it is generally accepted that the CCAM line resulted from mixing of two isotopically distinct reservoirs, ¹⁶O-rich and ¹⁶O-poor, the nature of these reservoirs is poorly under-

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stood. They may have been (1) primordial and resulted from (i) Galactic chemical evolution (GCE) of solids and gas (Clayton 1988; Timmes et al. 1995; Jacobsen et al. 2007; Meyer 2009) or (ii) recent pollution of the protosolar molecular cloud by a massive star ejecta (Krot et al. 2008a; Gaidos et al. 2009); or (2) originated in the solar nebula as a result of (i) CO self-shielding (Clayton 2002; Yurimoto & Kuramoto 2004; Lyons & Young 2005), (ii) symmetry-based non-mass-dependent chemical fractionation (Thiemens 1999; Marcus 2004), or (iii) other, still poorly understood fractionation processes (e.g., Kimura et al. 2007; Ozima et al. 2007).

Preliminary results reported by McKeegan et al. (2009) and McKeegan et al. (2010) show that oxygen isotopic composition of solar wind recorded by the *Genesis* spacecraft is ^{16}O -enriched relative to SMOW ($\Delta^{17}\text{O} = -26.5\text{‰} \pm 5.6\text{‰}$ and $-33\text{‰} \pm 8\text{‰}$, respectively, 2σ), suggesting that the Sun is ^{16}O -rich. These values are consistent with oxygen isotopic measurements of solar wind implanted into metal grains from lunar soil reported by Hashizume & Chaussidon (2005), but differs from that reported by Ireland et al. (2006): $\Delta^{17}\text{O} < -20\text{‰}$ versus $\Delta^{17}\text{O} \sim 26\text{‰}$, respectively. Based on recent measurements of oxygen isotopic compositions of lunar metal grains, Hashizume & Chaussidon (2009) concluded that there are two extraselenial oxygen isotopic components, ^{16}O -rich and ^{16}O -poor, but only the ^{16}O -rich component represents the solar wind composition. The range in $\Delta^{17}\text{O}$ trapped in lunar metal grains remains poorly understood.

Here, we adopt a *Genesis* solar wind value, $\Delta^{17}\text{O}$, $-26.5\text{‰} \pm 5.6\text{‰}$, as being representative of the bulk Sun,⁶ based on the evidence from *Genesis*, refractory inclusions from primitive chondrites (see below), and the ^{16}O -rich component in lunar metal grains. We consider that the primordial material from which the solar system formed consisted of gas and dust (we included water with the gas component). Based on solar system elemental abundances (Lodders 2003) and assuming the dust component has the bulk chemical composition of anhydrous CI chondrites, 26% of the solar system oxygen must be initially contained in dust and 74% in gas. The mean oxygen isotopic composition of the primordial dust is not known, but is constrained by mass balance such that gas plus dust must have the composition of the Sun. Since solar oxygen is dominated by the gas component, it is highly likely that the solar nebula gas was initially ^{16}O -rich (it could have been more or less ^{16}O -rich than the Sun, depending on the composition of the dust). However, it is highly unlikely that the solar nebula gas could have had $\Delta^{17}\text{O} = 0$, as this would require the dust to be far more ^{16}O -rich ($\Delta^{17}\text{O} \sim -100\text{‰}$) than any component observed in primitive meteorites.

2. CO SELF-SHIELDING MODELS: ASSUMPTIONS OF THE INITIAL OXYGEN ISOTOPIC COMPOSITION OF THE SOLAR NEBULA GAS AND DUST

In CO self-shielding models (Clayton 2002; Yurimoto & Kuramoto 2004; Lyons & Young 2005), oxygen isotopic compositions of primordial solar nebula dust (molecular cloud dust that avoided thermal processing in the solar nebula) and gas are assumed to have been initially identical ($\Delta^{17}\text{O} \sim -25\text{‰}$). Photodissociation of C^{17}O and C^{18}O in the protosolar molecular cloud (Yurimoto & Kuramoto 2004) or the protoplanetary disk

(Lyons & Young 2005) resulted in the formation of atomic ^{17}O and ^{18}O , which quickly reacted with nebular H_2 and froze out as water ice. Subsequent enrichment in the $\text{H}_2\text{O}/\text{CO}$ ratio in the disk midplane followed by ice evaporation inside the snow line led to the formation of an ^{16}O -poor water-rich gaseous reservoir in the inner solar nebula (Cuzzi & Zahnle 2004). Thermal processing of solids in this reservoir resulted in oxygen isotopic exchange and evolution of solids toward the TF line.

This scenario readily explains oxygen isotopic compositions of silicates in the terrestrial planets and asteroids (e.g., Franchi 2008 and references therein). However, the origin of dominant ^{16}O -poor silicates from chondritic porous interplanetary dust particles (CP IDPs), probably of cometary origin, is problematic (e.g., Aléon et al. 2009). Since oxygen isotopic exchange in solids occurs primarily through thermal processing in the inner solar system (because temperatures are higher and transient heating events are much more effective in regions of high gas and dust density, e.g., Nuth & Johnson 2006), the presence of dominant ^{16}O -poor silicates in CP IDPs requires that thermally processed silicates from the inner solar system were efficiently transported outward through the solar nebula and accreted onto cometary parent bodies at distances beyond ~ 20 AU. Several models for radial transport have been proposed to explain the presence of refractory minerals and crystalline silicates in comets (e.g., Shu et al. 1996, 1997; Bockelée-Morvan et al. 2002; Gail 2001, 2004; Dullemond et al. 2006; Boss 2008; Ciesla 2007, 2009). However, these models are most efficient at transporting materials outward during the very earliest stages of disk evolution, when the rates of mass and angular momentum transport are at their highest. In order for an ^{16}O -poor gaseous reservoir to develop in the inner solar nebula in CO self-shielding models, water ice from the outer nebula must decouple from the ^{16}O -rich gas, drift across the snow line and vaporize, increasing the water abundance several-fold (Cuzzi & Zahnle 2004). Models suggest that timescales of a few hundred thousand to millions of years are required to reach the enhanced water abundance in the inner solar system (Ciesla & Cuzzi 2006). By that time, mass and angular momentum transport would likely have diminished by orders of magnitude, if not stopped altogether (Sicilia-Aguilar et al. 2006), making the efficient outward transport of processed dust grains to the comet formation region extremely difficult.

The inferred relatively long timescales for generating an ^{16}O -poor reservoir in the inner solar nebula due to CO self-shielding (Ciesla & Cuzzi 2006) could also be problematic if ^{16}O -rich and ^{16}O -poor reservoirs co-existed at the time of CAI formation as was concluded by Itoh & Yurimoto (2003) and Aléon et al. (2007). This is because CAI formation appears to have started when the Sun was a young (class 0) or accreting (class I) protostar (e.g., Goswami et al. 2005) and lasted less than 0.1 Myr (Thrane et al. 2006; Jacobsen et al. 2008).

There are two testable predictions of the CO self-shielding models: the mean composition of primordial dust must be ^{16}O -rich and the oxygen isotopic compositions of the solar system solids should evolve with time toward increasing $\Delta^{17}\text{O}$. Neither of these predictions is supported by observations.

1. No primordial ^{16}O -rich dust has yet been discovered (e.g., Aléon et al. 2009). The isotopically anomalous presolar grains typically measured by secondary ion mass spectrometer (SIMS or ion microprobe) are crystalline and very rare (< 0.1 wt%), even in primitive chondrites and CP IDPs. The individual presolar grains have a wide range of oxygen isotopic compositions (e.g., Nguyen & Zinner 2004; Nittler

⁶ Two preliminary values of $\Delta^{17}\text{O}$ of the solar wind reported by McKeegan et al. (2009 and 2010), $-26.5\text{‰} \pm 5.6\text{‰}$ and $-33\text{‰} \pm 8\text{‰}$ (2σ), are not statistically different; they are discussed in Sections 4–6.

et al. 2008), which, however, do not represent the average isotopic composition of the solar system. Most dust in the protosolar molecular cloud was amorphous and may have been well homogenized in the interstellar medium (ISM). The average oxygen isotopic composition of the protosolar molecular cloud dust grains (which are exclusively presolar) is solar, by definition. The question we are addressing in the paper is what was the average $\Delta^{17}\text{O}$ value of this dust.

- The oxygen isotopic compositions of solar system solids appear uncorrelated with their respective times of formation (Krot et al. 2009). Although the oldest solar system solids, CAIs (e.g., Amelin et al. 2002), are generally ^{16}O -rich (Figure 1), chondrules and differentiated asteroids exhibit a wide range of formation ages, which do not correlate with their oxygen isotopic compositions (Clayton 1993; Kita et al. 2005; Halliday & Kleine 2006; Yurimoto et al. 2008; Franchi 2008). Although there is an overlap between the inferred initial $^{26}\text{Al}/^{27}\text{Al}$ ratios (corresponding to their ^{26}Al - ^{26}Mg ages) in chondrules from primitive ordinary and carbonaceous chondrites, the average ^{26}Al - ^{26}Mg ages of chondrules⁷ decrease across the sequence from ordinary chondrites (OC) to CO to CR carbonaceous chondrites, whereas the $\Delta^{17}\text{O}$ values of chondrules increase from CO to CR to OC (Figure 1).

Differentiated asteroids appear to have accreted prior to the formation of chondrules in chondritic asteroids (e.g., Halliday & Kleine 2006), yet differentiated meteorites (achondrites) have ^{16}O -poor compositions, similar to those of chondrites (Clayton 1993; Franchi 2008). Although chondritic and differentiated asteroids accreted at different times, one could assume their components formed at approximately the same times. We note, however, that the young accretion ages of differentiated asteroids (within 0.5–1 Myr after formation of CAIs) inferred from ^{207}Pb - ^{206}Pb and ^{182}Hf - ^{182}W ages of differentiated meteorites (Halliday & Kleine 2006, and references therein) preclude the formation of their components at the same time as chondrules in primitive ordinary and carbonaceous chondrites, which appear to have formed 2–4 Myr after CAIs (e.g., Amelin et al. 2002; Kita et al. 2005; Krot et al. 2009, and references therein).

3. ANALYTICAL PROCEDURES

Oxygen-isotope compositions of most refractory inclusions discussed in this paper were measured in situ with the University of Hawaii Cameca ims-1280 ion microprobe. Two analytical procedures were used for these measurements. In the first procedure, a ~ 1.8 nA Cs^+ primary ion beam was focused to a diameter of ~ 7 – 10 μm and rastered over 10×10 μm^2 area on the sample for data collection. Secondary ions of $^{16}\text{O}^-$, $^{17}\text{O}^-$, and $^{18}\text{O}^-$ were measured simultaneously in multicollection mode with the magnetic field controlled by a nuclear magnetic resonance probe. $^{16}\text{O}^-$ and $^{18}\text{O}^-$ were measured by multicollector Faraday cups (FCs) with low mass-resolving power (MRP ~ 2000), while $^{17}\text{O}^-$ was measured using the axial monocollector electron multiplier (EM) in pulse counting mode with MRP ~ 5600 , sufficient to separate the interfering $^{16}\text{OH}^-$ signal. In the second procedure, to reduce the beam size, ~ 200 pA Cs^+ primary ion beam was focused to a diameter of

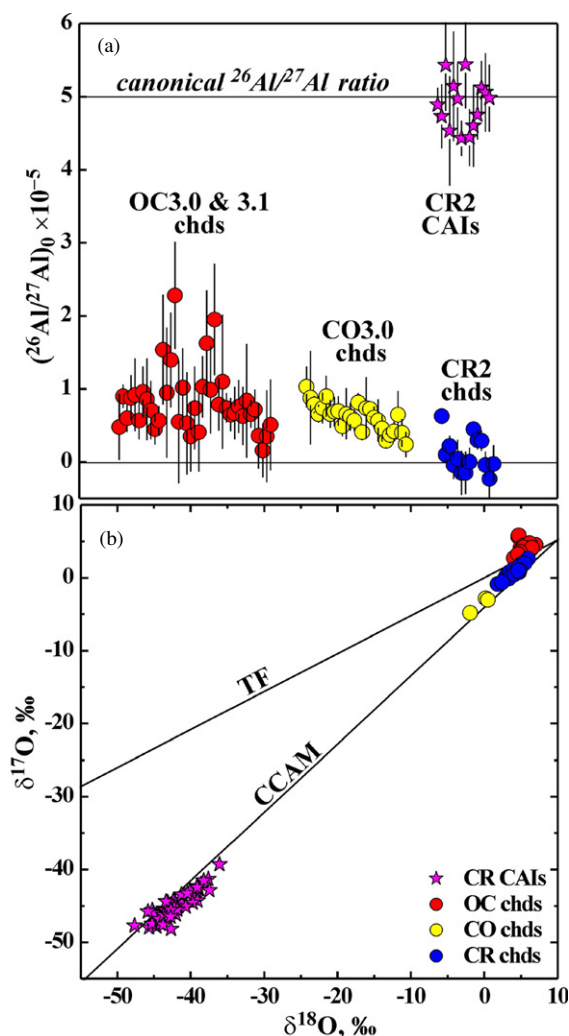


Figure 1. Initial $^{26}\text{Al}/^{27}\text{Al}$ ratios (a) and three-isotope oxygen diagram (b) of CAIs from CR chondrites and bulk chondrules (chds) from ordinary (petrologic types 3.0 and 3.1), CR2, and CO3.0 carbonaceous chondrites (data from Clayton 1993; Kita et al. 2000; Huss et al. 2001; Mostefaoui et al. 2002; Kunihiro et al. 2004; Rudraswami & Goswami 2007; Nagashima et al. 2008a; Kurahashi et al. 2008; Makide et al. 2009a). Carbonaceous chondrite anhydrous mineral (CCAM) line, the terrestrial fractionation (TF) line, and the canonical $^{26}\text{Al}/^{27}\text{Al}$ ratio are shown for reference. Although the oldest solar system solids, CAIs, are ^{16}O -enriched relative to chondrules, there is no correlation between chondrule ages and their oxygen-isotope compositions.

~ 5 μm and rastered over $\sim 5 \times 5$ μm^2 on the sample. Three oxygen isotopes were measured by the combination of multicollection mode and peak-jumping. $^{16}\text{O}^-$ and $^{17}\text{O}^-$ were measured simultaneously using the multicollector FC and the monocollector EM, respectively. Subsequently, ^{18}O was measured with the monocollector EM by peak-jumping. Mass resolving power for $^{16}\text{O}^-$ and for $^{17}\text{O}^-$ and $^{18}\text{O}^-$ were set to ~ 2000 and ~ 5600 , respectively. The instrumental mass fractionation was corrected for each session using San Carlos olivine, Eagle Station pallisite olivine, Miyakejima anorthite, synthetic enstatite, augite, and Burma spinel standards. Because of the difficulty of precisely determining relative efficiencies among FC and EM detectors, measured $\Delta^{17}\text{O}$ values on terrestrial standards showed systematic shifts (typically $\pm 1\text{‰}$) from the terrestrial value ($\Delta^{17}\text{O} = 0$). The mean of $\Delta^{17}\text{O}$ on standard measurements was used to correct systematic shift from the terrestrial value. The reported uncertainties include both the internal measurement

⁷ Since ^{26}Al is generally considered to have been the major heating source of asteroids, a degree of thermal metamorphism experienced by chondritic asteroids is expected to correlate with ^{26}Al - ^{26}Mg ages of their chondrules. Indeed, the degree of thermal metamorphism decreases from OC to CO to CR chondrites (e.g., Weisberg et al. 2006).

precision of an individual analysis and the external reproducibility for standard measurements during a given analytical session. For the first procedure, the point-to-point reproducibility (external reproducibility) on the multiple analyses of the standards was 0.5‰ – 1‰ (2σ) for both $\delta^{17}\text{O}$ and $\delta^{18}\text{O}$. For the second procedure, the external reproducibility was 1‰ – 1.5‰ (2σ) for both $\delta^{17}\text{O}$ and $\delta^{18}\text{O}$.

4. OXYGEN ISOTOPIC COMPOSITIONS OF AOAs AND CAIs: CONFIRMATION OF ^{16}O -RICH COMPOSITION OF THE SUN

AOAs and CAIs appear to have formed in a gas of approximately solar composition (gas/dust ratio of ~ 0.01 by mass) as condensates from vaporized nebular material and/or remelted condensates (Grossman et al. 2002, 2008; Krot et al. 2004; Sugiura et al. 2009), possibly within the first 0.1 Myr of solar system formation (Thrane et al. 2006; Jacobsen et al. 2008). The oxygen isotopic compositions of AOAs and CAIs likely represent the composition of the Sun and provide a means to test the solar wind value recorded by the *Genesis* spacecraft. We briefly summarize in situ oxygen isotopic measurements of CAIs and AOAs from CR and CV chondrites recently acquired with the University of Hawaii Cameca ims-1280 ion microprobe (MacPherson et al. 2008; Makide et al. 2009a; Krot et al. 2010; Nagashima et al. 2010; Petaev et al. 2010).

Mineralogically pristine CAIs and AOAs from unmetamorphosed CR chondrites are isotopically uniform and have an average $\Delta^{17}\text{O}$ value of $-23.3\text{‰} \pm 1.9\text{‰}$ (2σ ; Figures 2a and 2c). Oxygen isotopic heterogeneity is observed only in rare relict CAIs associated with ferromagnesian chondrule-like silicates (olivine and low-Ca pyroxene) suggesting these CAIs experienced remelting during chondrule formation (Makide et al. 2009a). In relict CAIs, spinel (MgAl_2O_4) typically escaped melting and retained its original, ^{16}O -rich composition ($\Delta^{17}\text{O} \sim -24\text{‰}$), whereas pyroxene and anorthite experienced melting and are ^{16}O -depleted ($\Delta^{17}\text{O}$ up to $\sim -5\text{‰}$). This distribution of oxygen isotopes suggests isotope exchange between relict CAI, host chondrule melt, and an external gaseous reservoir.

Most CAIs from metamorphosed CV chondrites (for metamorphic classification of CV chondrites, see Bonal et al. 2006) have heterogeneous oxygen isotopic compositions (Figures 2b and 2c). Spinel, forsterite (Mg_2SiO_4), and pyroxene appear to have preserved their initial oxygen isotopic compositions and have an average $\Delta^{17}\text{O}$ of $-23.6\text{‰} \pm 1.0\text{‰}$ (2σ), similar to CR CAIs and AOAs. In contrast, melilite (a solid solution of $\text{Ca}_2\text{Al}_2\text{SiO}_7$ and $\text{Ca}_2\text{MgSi}_2\text{O}_7$) and anorthite ($\text{CaAl}_2\text{Si}_2\text{O}_8$) are ^{16}O -depleted, suggesting sub-solidus isotopic exchange with an external ^{16}O -poor reservoir (Ryerson & McKeegan 1994; MacPherson et al. 2008; Yurimoto et al. 2008).

The $\Delta^{17}\text{O}$ value of the Sun, $\Delta^{17}\text{O} \sim -24\text{‰} \pm 2\text{‰}$, inferred from CR AOAs and CAIs, which avoided remelting during chondrule formation, and from CV CAI minerals, which avoided postcrystallization isotope exchange, is in good agreement with the solar wind value reported by McKeegan et al. (2009). It is slightly heavier than the solar wind value reported by McKeegan et al. (2010). The inferred $\Delta^{17}\text{O}$ value of the Sun is consistent with the solar wind value measured in metal grains from lunar soil by Hashizume & Chaussidon (2005); it is, however, inconsistent with the high $\Delta^{17}\text{O}$ value measured in metal grains from lunar soil reported by Ireland et al. (2006) and Hashizume & Chaussidon (2009).

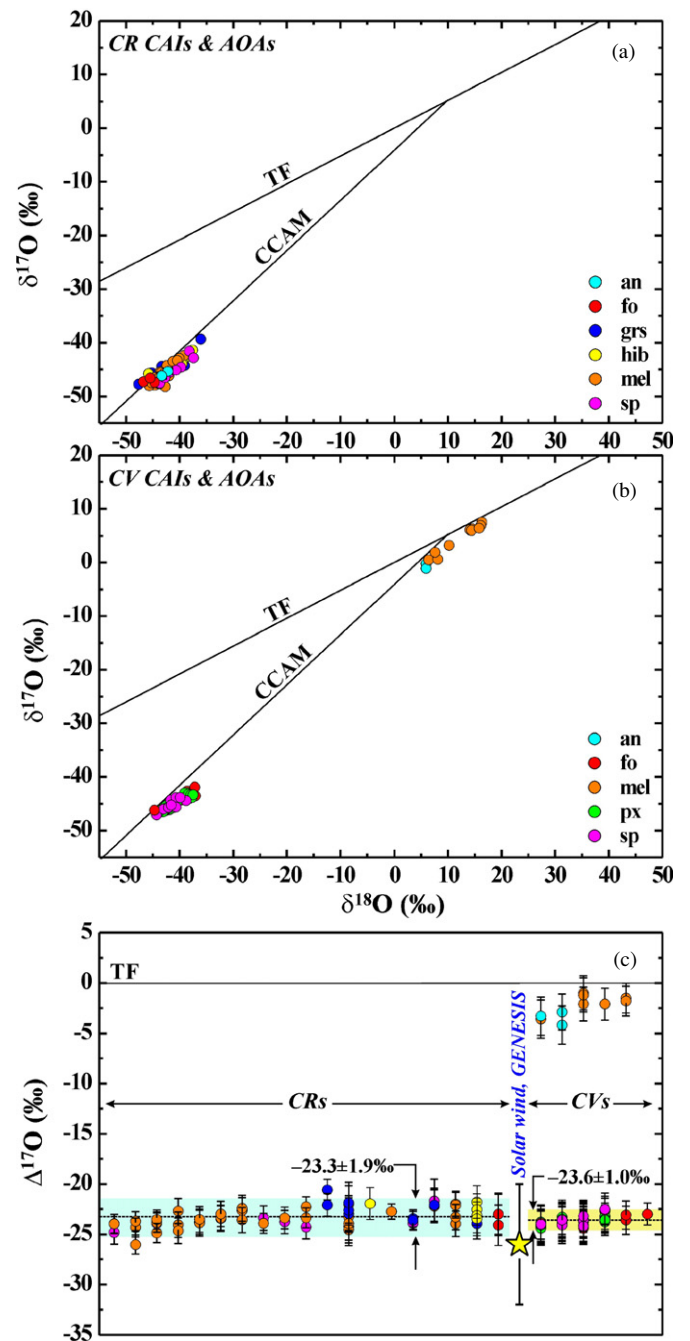


Figure 2. Three-isotope oxygen diagrams (a, b) and $\Delta^{17}\text{O}$ values (c) of CAIs and AOAs from unmetamorphosed CR (a) and metamorphosed CV (b) chondrites (data from Makide et al. 2009a and MacPherson et al. 2008, respectively). Abbreviations hereafter: an = anorthite; fo = forsterite; grs = grossite; mel = melilite; px = pyroxene; sp = spinel. The CR CAIs and AOAs are mineralogically pristine, isotopically uniform, and have an average $\Delta^{17}\text{O}$ value of $-23.3\text{‰} \pm 1.9\text{‰}$ (2σ), similar to the solar wind value reported by McKeegan et al. (2009; $-26.5\text{‰} \pm 5.6\text{‰}$, 2σ); it is slightly heavier than the value reported by McKeegan et al. (2010; $-33\text{‰} \pm 8\text{‰}$, 2σ). The CV CAIs contain abundant secondary minerals and are typically isotopically heterogeneous, suggesting postcrystallization isotope exchange. Forsterite, pyroxene, and spinel in CV CAIs appear to have retained original oxygen isotopic compositions ($\Delta^{17}\text{O} = -23.6\text{‰} \pm 1.0\text{‰}$, 2σ), which are similar to those of CR CAIs and the *Genesis* solar wind value of $-26.5\text{‰} \pm 5.6\text{‰}$.

5. ANOMALOUSLY ^{16}O -RICH CHONDRULE AND CAIs FROM METAL-RICH CHONDRITES

Kobayashi et al. (2003) described a unique chondrule in the Acfer 214 CH chondrite with a remarkably ^{16}O -rich compo-

sition ($\Delta^{17}\text{O} = -37\text{‰} \pm 4\text{‰}$, 2σ). In addition, Krot et al. (2008a) and Gounelle et al. (2009) reported two grossite-rich (CaAl_4O_7) CAIs from the CH/CB-like chondrite Isheyevo with similarly large ^{16}O enrichments ($\Delta^{17}\text{O} \sim -35\text{‰}$). Assuming the oxygen isotopic compositions of dust and gas in the solar system were initially both ^{16}O -rich and dust evolved toward heavier composition with time (Clayton 2002; Yurimoto & Kuramoto 2004; Lyons & Young 2005), Gounelle et al. (2009) suggested that the extreme $\Delta^{17}\text{O}$ value was representative of the initial composition of the solar nebula, and, hence, the Sun. However, thermodynamic calculations show that grossite-rich CAIs did not form in a gas of solar composition (Ebel 2006) and, therefore, the oxygen isotopic compositions of these objects should not be used to infer the oxygen isotopic composition of the Sun. In addition, this suggestion is difficult to reconcile with the narrow range of oxygen isotopic compositions of AOAs and CAIs from primitive chondrites (Figure 2c), which lie much closer to the preliminary *Genesis* solar wind value reported by McKeegan et al. (2009) than the Isheyevo grossite-rich CAIs and the solar wind value reported by McKeegan et al. (2010). Finally, oxygen isotopic compositions of micron-sized solar corundum (Al_2O_3) grains ($\Delta^{17}\text{O} = -22.7\text{‰} \pm 8.5\text{‰}$, 2σ), which may represent the earliest nebular condensates (Makide et al. 2009b), do not support the extremely ^{16}O -rich composition of the Sun.

The bulk solar system, in terms of oxygen, consisted of 26% dust and 74% gas. If the bulk Sun has $\Delta^{17}\text{O} \sim -26\text{‰}$ (McKeegan et al. 2009) and primordial dust had $\Delta^{17}\text{O} \sim 0\text{‰}$, simple mass balance calculation implies the nebular gas had $\Delta^{17}\text{O} \sim -35\text{‰}$, the same value found for the Acfer 214 chondrule (Kobayashi et al. 2003) and the Isheyevo grossite-rich CAIs (Krot et al. 2008a; Gounelle et al. 2009). This raises the possibility that the primordial dust was ^{16}O -poor and the anomalously ^{16}O -rich chondrule from Acfer 214 and grossite-rich CAIs from Isheyevo equilibrated with an ^{16}O -rich nebular gas in a dust-poor environment. If, however, the bulk Sun has $\Delta^{17}\text{O} \sim -35\text{‰}$, as reported by McKeegan et al. (2010), it would mean that only the anomalously ^{16}O -rich CAIs and chondrule formed in a gas of solar composition, whereas refractory inclusions with $\Delta^{17}\text{O} \sim -25\text{‰}$ formed from a gas of non-solar composition (with enhanced dust/gas ratio). If this is the case, one could estimate oxygen isotopic compositions of dust and gas, if dust/gas ratio in the CAI-forming region can be independently inferred (e.g., based on the abundance of Fe^{2+} and Cr^{2+} in olivine of AOAs and thermodynamic calculations).

6. CAIs WITH HIGHLY FRACTIONATED OXYGEN ISOTOPIC COMPOSITIONS: CONSTRAINTS ON COMPOSITIONS OF THE PRIMORDIAL DUST

The majority of igneous CAIs show small mass-dependent fractionation effects in oxygen isotopes ($<5\text{‰}/\text{amu}$; Figure 2) consistent with melting and evaporation during transient heating events at relatively high ambient pressure ($>10^{-5}$ bar) when fast evaporation, back-reaction and exchange with nebular gas minimized mass fractionation (Richter et al. 2006). Much larger mass-dependent fractionation effects in oxygen isotopes ($>10\text{‰}$ – $15\text{‰}/\text{amu}$) are found in rare fractionation and unidentified nuclear effects (FUN) and fractionation (F) CAIs (Wasserburg et al. 1977; Clayton et al. 1984; Ireland et al. 1992; Davis et al. 2000; Ushikubo et al. 2004; Thrane et al. 2008; Krot et al. 2008b; Hiyagon & Hashimoto 2008; Nagashima et al. 2008b; Liu et al. 2009; Makide et al. 2009a; Rout et al. 2009). The common presence of large stable isotope anomalies

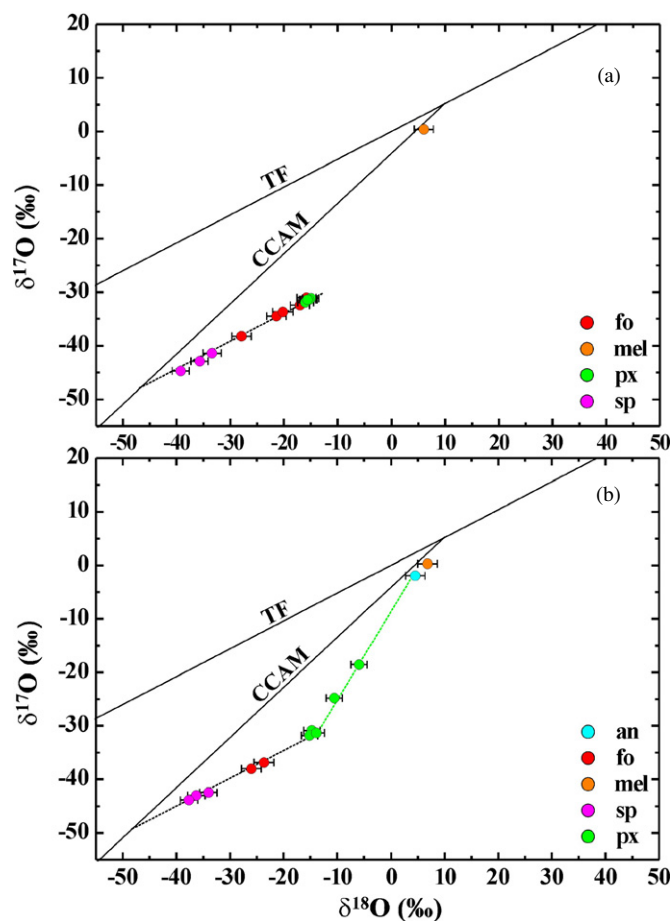


Figure 3. Three-isotope oxygen diagrams of FUN CAIs CG14 (a) and DH8 (b) from the Allende (CV) chondrite. an = anorthite; fo = forsterite; mel = mellilite; px = pyroxene; sp = spinel. Spinel, pyroxene, and forsterite have highly fractionated compositions and plot along mass-dependent fractionation lines (indicated as black dashed lines) parallel to the TF line; a degree of mass-dependent fractionation correlates with mineral crystallization sequence: spinel \rightarrow forsterite \rightarrow pyroxene. Melilite in CG14 and DH8, anorthite and some pyroxene grains in DH8 are ^{16}O -depleted and define a trend towards the TF line (indicated as green dashed line), suggesting subsequent isotope exchange with an ^{16}O -poor reservoir.

in many elements in FUN CAIs (e.g., Wasserburg et al. 1977; Lee 1988; Loss et al. 1994) suggests that their precursors escaped complete evaporation-recondensation in the solar nebula. Therefore, FUN CAIs may have retained the oxygen isotopic signature of the primordial dust. Here, we show that hibonite ($\text{CaAl}_{12}\text{O}_{19}$), spinel, forsterite, and pyroxene grains in FUN and F CAIs (hereafter F(UN) CAIs) preserved oxygen isotopic signatures of their precursors, and, possibly of the primordial dust.

In situ oxygen isotopic measurements of F(UN) CAIs (Ireland et al. 1992; Davis et al. 2000; Ushikubo et al. 2004; Thrane et al. 2008; Krot et al. 2008b; Hiyagon & Hashimoto 2008; Liu et al. 2009; Makide et al. 2009a; Rout et al. 2009) and evaporation experiments on melts of CAI-like compositions (Mendybaev et al. 2009) suggest that F(UN) CAIs experienced melting and evaporation in vacuum ($\leq 10^{-6}$ bar), resulted in large mass-dependent fractionation effects for oxygen, magnesium, and silicon isotopes. Within an individual F(UN) CAI, the degree of mass-dependent fractionation of oxygen isotopes for spinel, pyroxene, and forsterite correlates with the sequence of crystallization of these minerals (Figure 3a). On a three-isotope oxygen diagram, the compositions of these minerals plot along mass-dependent fractionation lines parallel to the TF

line; $\Delta^{17}\text{O}$ remains constant for each CAI but can vary from one inclusion to the next. We infer the $\Delta^{17}\text{O}$ value of a F(UN) CAI reflects the oxygen isotopic composition of its precursor prior to evaporation. The fact that oxygen isotopic compositions of spinel, pyroxene, and forsterite in many F(UN) CAIs lie along mass-dependent fractionation lines implies oxygen isotopic exchange with an external reservoir did not occur during or after melting and evaporation.

The coarse-grained igneous textures of F(UN) CAIs and evaporation experiments suggest the F(UN) CAI precursors were heated to maximum temperatures of less than 1900 °C; heating lasted 1–20 hr and the CAI melts cooled at rates of several degrees per hour (Mendybaev et al. 2009). The inferred thermal history of F(UN) CAIs resembles that of typical coarse-grained igneous CAIs suggesting a common formation mechanism (Richter et al. 2002, 2006; Mendybaev et al. 2009). Although transient heating events are also invoked for the origin of chondrules (e.g., Desch & Connolly 2002), chondrule formation produced no resolvable evidence for mass-dependent fractionation and is believed to have occurred in regions with very high dust/gas ratio ($>10^5 \times$ solar; e.g., Cuzzi & Alexander 2006; Alexander et al. 2008).

Two FUN CAIs from CV chondrites, KT-1 from North West Africa 779 (Thrane et al. 2008) and DH8 from Allende (Krot et al. 2008b), experienced incomplete remelting and partial isotope exchange with an ^{16}O -depleted nebular gas recorded by some pyroxene grains (Figure 3b; see also Figure 4 in Thrane et al. 2008). In addition, melilite and anorthite in all F(UN) CAIs from CV chondrites are ^{16}O -depleted (Figures 3a and 3b) (Davis et al. 2000; Thrane et al. 2008; Krot et al. 2008b; Hiyagon & Hashimoto 2008), suggesting sub-solidus isotopic exchange with an ^{16}O -poor external reservoir. Oxygen isotopic exchange associated with these processes shifted oxygen isotopic compositions of melilite, anorthite and some pyroxene grains off the F(UN) CAI fractionation lines (Figure 3). As a result, these grains cannot be used to infer oxygen isotopic compositions of the F(UN) CAI precursors prior to melt evaporation events.

Figure 4 shows the oxygen isotopic compositions of spinel, hibonite, pyroxene, and forsterite in F(UN) CAIs measured with high precision using ion microprobes. In contrast to AOAs and CAIs having similar oxygen isotopic compositions ($\Delta^{17}\text{O} \sim -24\text{‰} \pm 2\text{‰}$; Figure 2c), F(UN) CAIs show a large range of $\Delta^{17}\text{O}$, from -0.5‰ to -24.8‰ (Figure 4), indicating a large range of oxygen isotopic compositions of the precursors. This range may reflect initial variations in the oxygen isotopic compositions of F(UN) CAI precursors, if they formed during single-stage event (Lee & Sheng 2001), or varying degrees of isotopic exchange of F(UN) CAI precursors and nebular gas, if they experienced multiple melting events in the solar nebula prior to melt evaporation in vacuum. The second scenario appears more plausible, because it requires only two isotopically distinct reservoirs, ^{16}O -rich and ^{16}O -poor. We suggest that F(UN) CAIs experienced at least two melting events: melting of solid precursors and isotope exchange of melts with the nebular gas, resulting in the observed range of $\Delta^{17}\text{O}$ values (indicated by red circles along the CCAM line in Figure 4b) and additional melting and evaporation generating the mass-dependent fractionation arrays parallel to the TF line (indicated by red dashed lines in Figure 4b).

We interpret the observed range of $\Delta^{17}\text{O}$ values among F(UN) CAIs as the result of varying degrees of equilibration between ^{16}O -poor dust and ^{16}O -rich nebular gas and suggest the former

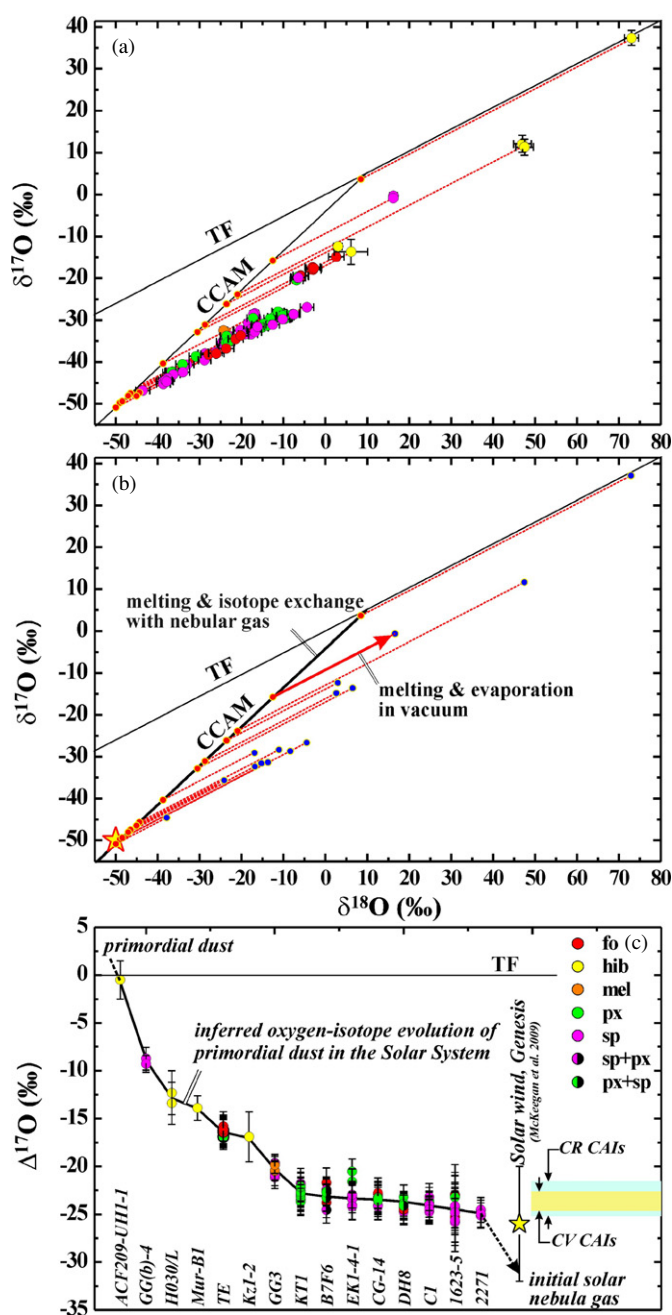


Figure 4. Three-isotope oxygen diagrams (a, b) and $\Delta^{17}\text{O}$ values (c) of F(UN) CAIs (data from Davis et al. 2000; Ushikubo et al. 2004; Krot et al. 2008b; Nagashima et al. 2008b; Liu et al. 2009; Makide et al. 2009a; Rout et al. 2009; this study). Mass-dependent fractionation lines for F(UN) CAIs (red dashed lines) are extended to CCAM line, along which their precursors may have originally plot (red-yellow circles in “a” and “b”). Two-stage evolution of oxygen isotopic compositions of F(UN) CAIs is inferred: melting and isotope exchange with nebular gas generated the observed range of $\Delta^{17}\text{O}$ values, and melting and evaporation in vacuum produced mass-dependent fractionation trends. We suggest the observed range of $\Delta^{17}\text{O}$ among F(UN) CAIs is the result of varying degrees of isotope exchange between ^{16}O -poor dust ($\Delta^{17}\text{O} \sim 0$) and ^{16}O -rich nebular gas ($\Delta^{17}\text{O} < -25\text{‰}$) prior to melt evaporation in vacuum, and the ^{16}O -poor dust is characteristic of the primordial dust.

is characteristic of the primordial dust. The distinct oxygen isotopic compositions of the primordial solar nebula dust and gas could have resulted from GCE (Meyer 2009) or be a result of pollution of the protosolar molecular cloud by massive star ($>50\text{☉}$) ejecta (Gaidos et al. 2009). CO self-shielding may have

played only a minor role in oxygen isotopic evolution of the solar system dust.

Nucleosynthesis theory distinguishes primary, ^{16}O , and secondary, ^{17}O and ^{18}O , isotopes (e.g., Clayton 1988). Standard GCE models show that the ISM enrichment of a primary isotope, such as ^{16}O , grows approximately linearly with time, while those of secondary isotopes, such as ^{17}O and ^{18}O , grow approximately quadratically (Clayton & Pantelaki 1986). As a result, the ISM mass fraction ratios $^{17}\text{O}/^{16}\text{O}$ and $^{18}\text{O}/^{16}\text{O}$ increase roughly linearly with time. If ISM dust is older on average than the ISM gas and isotope exchange was incomplete, GCE is expected to produce ^{16}O -enriched solid and ^{16}O -depleted gaseous reservoirs which on a three-isotope oxygen diagram would lie along a \sim slope-1 line (Timmes et al. 1995; Jacobsen et al. 2007). On the other hand, if ISM dust is dominated by recent stellar ejecta, which are substantially enriched in secondary isotopes relative to the average ISM gas, GCE should produce ^{16}O -depleted solid and ^{16}O -enriched gaseous reservoirs (Meyer 2009). The latter appears to be consistent with a correlation of $\Delta^{17}\text{O}$ and $\varepsilon^{54}\text{Cr}$ in bulk carbonaceous chondrites (Yin et al. 2009).

Our interpretation is consistent with the inferred ^{16}O -rich composition of the Sun (Hashizume & Chaussidon 2005; McKeegan et al. 2009, 2010), the ^{16}O -rich composition of the nebular gas of solar composition from which AOAs, most CAI precursors, and solar corundum grains condensed (Itoh et al. 2004; Makide et al. 2009a, 2009b), apparent absence of the ^{16}O -rich primordial dust (Aléon et al. 2009), and the wide range of oxygen isotopic compositions of presolar grains (Nguyen & Zinner 2004; Nittler et al. 2008). This scenario may also explain the co-existence of ^{16}O -rich and ^{16}O -poor reservoirs since the earliest stages of the solar system evolution (Itoh & Yurimoto 2003; Aléon et al. 2007) and the lack of correlation between oxygen isotopic compositions and formation ages of solar system solids (Krot et al. 2009 and references therein). (1) In nebular regions with solar composition, 26% of oxygen is tied up in dust and 74% in gas. If primordial dust were ^{16}O -poor, nebular regions with enhanced dust/gas ratio relative to solar would be dominated by oxygen in solids. The existence of a dusty disk since the beginning of the solar system formation may explain the coexistence of ^{16}O -rich and ^{16}O -poor reservoirs at the time of CAI formation (Itoh & Yurimoto 2003; Aléon et al. 2007). (2) It is generally believed that chondrules and a significant portion of fine-grained matrix materials formed during high-temperature ($\sim 1400^\circ\text{C}$ – 1600°C) transient heating events, possibly shockwaves, in solar nebula regions with enhanced (up to $10^5\times$ solar) dust/gas ratio (e.g., Desch & Connolly 2002; Cuzzi & Alexander 2006; Alexander et al. 2008). This may explain ^{16}O -poor compositions of most chondrules, fine-grained matrix materials, chondrites, achondrites, and terrestrial planets (Ozima et al. 2009).

We offer several testable predictions: (1) the least thermally processed dust in the solar system, amorphous dust in CP IDPs and Kuiper Belt Objects (e.g., McKeegan et al. 2006; Messenger et al. 2009), should have ^{16}O -poor bulk composition; (2) ^{16}O -rich solids in this dust are expected to be very rare, crystalline and related to CAIs and AOAs; (3) because chondrules formed in dust-rich nebular regions dominated by ^{16}O -poor dust, uniformly ^{16}O -rich chondrules are expected to be extremely rare.

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REFERENCES

- Aléon, J., El Goresy, A., & Zinner, E. 2007, *Earth Planet. Sci. Lett.*, **263**, 114
- Aléon, J., Engrand, C., Leshin, L. A., & McKeegan, K. D. 2009, *Geochim. Cosmochim. Acta*, **73**, 4558
- Alexander, C. M. O'D., Grossman, J. N., Ebel, D. S., & Ciesla, F. J. 2008, *Science*, **320**, 1617
- Amelin, Y., Krot, A. N., Hutcheon, I. D., & Ulyanov, A. A. 2002, *Science*, **297**, 1678
- Bockelée-Morvan, D., Gautier, D., Hersant, F., Huré, J.-M., & Robert, F. 2002, *A&A*, **384**, 1107
- Bonal, L., Quirico, E., Bourot-Denise, M., & Montagnac, G. 2006, *Geochim. Cosmochim. Acta*, **70**, 1849
- Boss, A. P. 2008, *Earth Planet. Sci. Lett.*, **268**, 102
- Ciesla, F. J. 2007, *Science*, **318**, 613
- Ciesla, F. J. 2009, *Icarus*, **200**, 655
- Ciesla, F. J., & Cuzzi, J. N. 2006, *Icarus*, **181**, 178
- Clayton, D. D. 1988, *ApJ*, **334**, 191
- Clayton, D. D., & Pantelaki, I. 1986, *ApJ*, **307**, 441
- Clayton, R. N. 1993, *Ann. Rev. Earth Planet. Sci.*, **21**, 115
- Clayton, R. N. 2002, *Nature*, **415**, 860
- Clayton, R. N., et al. 1984, *Geochim. Cosmochim. Acta*, **48**, 535
- Cuzzi, J. N., & Alexander, C. M. O'D. 2006, *Nature*, **441**, 483
- Cuzzi, J. N., & Zahnle, K. J. 2004, *ApJ*, **614**, 490
- Davis, A. M., McKeegan, K. D., & MacPherson, G. J. 2000, *Meteorit. Planet. Sci.*, **35**, 47
- Desch, S. J., & Connolly, H. C., Jr. 2002, *Meteorit. Planet. Sci.*, **37**, 183
- Dullemond, C. P., Apai, D., & Walch, S. 2006, *ApJ*, **640**, L67
- Ebel, D. S. 2006, in *Meteorites and the Early Solar System II*, ed. D. Lauretta & H. Y. McSween, Jr. (Tucson, AZ: Univ. of Arizona Press), 253
- Franchi, I. A. 2008, *Rev. Mineral. Geochem.*, **68**, 345
- Gaidos, E., Krot, A. N., & Huss, G. R. 2009, *ApJ*, **705**, L163
- Gail, H.-P. 2001, *A&A*, **378**, 192
- Gail, H.-P. 2004, *A&A*, **413**, 571
- Goswami, J. N., Marhas, K. K., Chaussidon, M., Gounelle, M., & Meyer, B. S. 2005, in *ASP Conf. Ser. 341, Chondrites and the Protoplanetary Disk*, ed. A. N. Krot, E. R. D. Scott, & B. Reipurth (San Francisco, CA: ASP), 485
- Gounelle, M., Krot, A. N., Nagashima, K., & Kearsley, A. 2009, *ApJ*, **698**, L18
- Grossman, L., Ebel, D. S., & Simon, S. B. 2002, *Geochim. Cosmochim. Acta*, **66**, 145
- Grossman, L., et al. 2008, *Geochim. Cosmochim. Acta*, **72**, 3001
- Halliday, A. N., & Kleine, T. 2006, in *Meteorites and the Early Solar System II*, ed. D. S. Lauretta & H. Y. McSween Jr. (Tucson, AZ: Univ. of Arizona Press), 775
- Hashizume, K., & Chaussidon, M. 2005, *Nature*, **434**, 619
- Hashizume, K., & Chaussidon, M. 2009, *Geochim. Cosmochim. Acta*, **73**, 3038
- Hiyagon, H., & Hashimoto, A. 2008, *Meteorit. Planet. Sci.*, **43**, 5128
- Huss, G. R., MacPherson, G. J., Wasserburg, G. J., Russell, S. S., & Srinivasan, G. 2001, *Meteorit. Planet. Sci.*, **36**, 975
- Ireland, T. R., Holden, P., Norman, M. K., & Clarke, J. 2006, *Nature*, **440**, 776
- Ireland, T. R., Zinner, E. K., Fahey, A. J., & Esat, T. M. 1992, *Geochim. Cosmochim. Acta*, **56**, 2503
- Itoh, S., Kojima, H., & Yurimoto, H. 2004, *Geochim. Cosmochim. Acta*, **68**, 183
- Itoh, S., & Yurimoto, H. 2003, *Nature*, **423**, 728
- Jacobsen, S. B., Yin, G. Z., & Petaev, M. I. 2007, *Lunar Planet. Sci.*, **38**, 1804
- Jacobsen, S. B., et al. 2008, *Earth Planet. Sci. Lett.*, **272**, 353
- Kimura, Y., Nuth, J. A., Chakraborty, S., & Thiemens, M. H. 2007, *Meteorit. Planet. Sci.*, **42**, 1429
- Kita, N. T., Nagahara, H., Togashi, S., & Morishita, Y. 2000, *Geochim. Cosmochim. Acta*, **64**, 3913
- Kita, N. T., et al. 2005, in *ASP Conf. Ser. 341, Chondrites and the Protoplanetary Disk*, ed. A. N. Krot, E. R. D. Scott, & B. Reipurth (San Francisco, CA: ASP), 558
- Kobayashi, S., Imai, H., & Yurimoto, H. 2003, *Geochem. J.*, **37**, 663
- Krot, A. N., et al. 2004, *Chemie der Erde*, **64**, 185

- Krot, A. N., et al. 2008a, *ApJ*, 672, 713
- Krot, A. N., et al. 2008b, *Lunar Planet Sci.*, 39, 2162
- Krot, A. N., et al. 2009, *Geochim. Cosmochim. Acta*, 73, 4963
- Krot, A. N., et al. 2010, *Lunar Planet. Sci.*, 41, 1406
- Kunihiro, T., Rubin, A. E., McKeegan, K. D., & Wasson, J. T. 2004, *Geochim. Cosmochim. Acta*, 68, 2947
- Kurahashi, E., Kita, N. T., Nagahara, H., & Morishita, Y. 2008, *Geochim. Cosmochim. Acta*, 72, 3865
- Lee, T. 1988, in *Meteorites and the Early Solar System*, ed. J. F. Kerridge & M. S. Matthews (Tucson, AZ: Univ. of Arizona Press), 1063
- Lee, T., & Sheng, J. J. 2001, *Meteorit. Planet. Sci. A*, 36, 111
- Liu, M.-C., et al. 2009, *Geochim. Cosmochim. Acta*, 73, 5051
- Lodders, K. 2003, *ApJ*, 591, 1220
- Loss, R. D., Lugmair, G. W., Davis, A. M., & MacPherson, G. J. 1994, *ApJ*, 436, L193
- Lyons, J. R., & Young, E. D. 2005, *Nature*, 435, 317
- MacPherson, G. J., Nagashima, K., Bullock, E. S., & Krot, A. N. 2008, *Lunar Planet Sci.*, 39, 2039
- Makide, K., et al. 2009b, *Geochim. Cosmochim. Acta*, 73, 5018
- Makide, K., Nagashima, K., Krot, A. N., & Huss, G. R. 2009a, *ApJ*, 706, 142
- Marcus, R. A. 2004, *J. Chem. Phys.*, 121, 8201
- McKeegan, K. D., et al. 2006, *Science*, 314, 1724
- McKeegan, K. D., et al. 2009, *Lunar Planet. Sci.*, 40, 2494
- McKeegan, K. D., et al. 2010, *Lunar Planet. Sci.*, 41, 2589
- Mendybaev, R. A., Richter, F. M., Georg, R. B., & Davis, A. M. 2009, *Lunar Planet. Sci.*, 40, 2461
- Messenger, S., Keller, L. P., Nakamura-Messenger, K., & Nguyen, A. N. 2009, *Meteorit. Planet. Sci. A*, 44, 141
- Meyer, B. S. 2009, *Meteorit. Planet. Sci. A*, 44, 141
- Mostefaoui, S., Kita, N. T., Togashi, S., Tachibana, S., Nagahara, H., & Morishita, Y. 2002, *Meteorit. Planet. Sci.*, 37, 421
- Nagashima, K., Krot, A. N., & Huss, G. R. 2008a, *Lunar Planet Sci.*, 39, 2224
- Nagashima, K., Krot, A. N., & Huss, G. R. 2008b, *Meteorit. Planet. Sci. A*, 43, 108
- Nagashima, K., Krot, A. N., Huss, G. R., & Yurimoto, H. 2010, *Lunar Planet. Sci.*, 41, #2255
- Nguyen, A., & Zinner, E. 2004, *Science*, 303, 1496
- Nittler, L. R., et al. 2008, *ApJ*, 682, 1450
- Nuth, J. A., & Johnson, N. M. 2006, *Icarus*, 180, 243
- Ozima, M., Podosek, F. A., Higuchi, T., Yin, Q.-Z., & Yamada, A. 2007, *Icarus*, 186, 582
- Ozima, M., Yamada, A., Abe, O., Nanbu, S., & Kasai, Y. 2009, *Meteorit. Planet. Sci. A*, 44, 164
- Petaev, M. I., Nagashima, K., Krot, A. N., & Jacobsen, S. J. 2010, *Lunar Planet. Sci.*, 41, 1818
- Richter, F. M., Davis, A. M., Ebel, D. S., & Hashimoto, A. 2002, *Geochim. Cosmochim. Acta*, 66, 521
- Richter, F. M., Mendybaev, R. A., & Davis, A. M. 2006, *Meteorit. Planet. Sci.*, 41, 83
- Rout, S. S., et al. 2009, *Geochim. Cosmochim. Acta*, 73, 4264
- Rudraswami, N. G., & Goswami, J. N. 2007, *Earth Planet. Sci. Lett.*, 257, 231
- Ryerson, F. J., & McKeegan, K. D. 1994, *Geochim. Cosmochim. Acta*, 58, 3713
- Shu, F. H., Shang, H., Glassgold, A. E., & Lee, T. 1997, *Science*, 277, 1475
- Shu, F. H., Shang, H., & Lee, T. 1996, *Science*, 271, 1545
- Sicilia-Aguilar, A., et al. 2006, *ApJ*, 638, 897
- Sugiura, N., Petaev, M. I., Kimura, M., Miyazaki, A., & Hiyagon, H. 2009, *Meteorit. Planet. Sci.*, 44, 559
- Thiemens, M. H. 1999, *Science*, 283, 341
- Thrane, K., Bizzarro, M., & Baker, J. A. 2006, *ApJ*, 646, L159
- Thrane, K., Nagashima, K., Krot, A. N., & Bizzarro, M. 2008, *ApJ*, 680, L141
- Timmes, F. X., Woosley, S. E., & Weaver, T. A. 1995, *ApJS*, 98, 617
- Ushikubo, T., Hiyagon, H., & Sugiura, N. 2004, *Earth Planet. Sci. Lett.*, 254, 115
- Wasserburg, G. J., Lee, T., & Papanastassiou, D. A. 1977, *Geophys. Res. Lett.*, 4, 299
- Weisberg, M. K., McCoy, T. J., & Krot, A. N. 2006, in *Meteorites and The Early Solar System II*, ed. D. Lauretta & H. Y. McSween, Jr. (Tucson, AZ: Univ. of Arizona Press), 19
- Yin, Q.-Z., Yamashita, K., Yamakawa, A., Tanaka, R., Nakamura, E., Krot, A. N., & Meyer, B. S. 2009, *Meteorit. Planet. Sci. A*, 44, 224
- Yurimoto, H., & Kuramoto, K. 2004, *Science*, 305, 1763
- Yurimoto, H., et al. 2008, *Rev. Mineral. Geochem.*, 68, 141