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# A long duration of the <sup>16</sup>O-rich reservoir in the solar nebula, as recorded in fine-grained refractory inclusions from the least metamorphosed carbonaceous chondrites

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#### Abstract

Oxygen isotope ratios and corresponding <sup>26</sup>Al-<sup>26</sup>Mg isotope systematics of refractory inclusions from the least metamorphosed carbonaceous chondrites, Acfer 094 (C-ungrouped 3.00) and Yamato 81020 (CO3.05), were measured with an ion microprobe. Most of the samples are fine-grained refractory inclusions which are considered as condensates from high temperature solar nebular gas. The refractory inclusions consistently exhibit <sup>16</sup>O-enriched signatures among their interior phases (spinel, melilite, and high-Ca pyroxene), as well as phases within their rim structures (spinel, high-Ca pyroxene, and adjacent anorthite). This observation indicates that aggregated refractory condensates and the formation of rim structures occurred in the same <sup>16</sup>O-rich environment. Evidence for mass-dependent isotopic fractionation in oxygen and magnesium, which would indicate a later flash heating process, was not observed in rims. All oxygen isotope data from fine-grained CAIs are distributed between the Carbonaceous Chondrite Anhydrous Mineral (CCAM) line and the Primitive Chondrule Mineral (PCM) regression line based on oxygen isotope data from the Acfer 094 chondrules. The inferred initial  $^{26}$ Al/ $^{27}$ Al ratios, ( $^{26}$ Al/ $^{27}$ Al)<sub>0</sub>, of spinel-melilite-rich CAIs are  $(4.08 \pm 0.75) \times 10^{-5}$  to  $(5.05 \pm 0.18) \times 10^{-5}$  (errors are  $2\sigma$ ), which are slightly lower than the canonical value of  $5.25 \times 10^{-5}$ . As there is no petrologic evidence for re-melting after condensation, the lower  $(^{26}\text{Al})^{27}\text{Al})_0$ values of these CAIs indicate either they formed up to  $\sim$ 0.3 Ma after canonical CAIs or they formed before  $^{26}$ AI was homogeneously distributed in the solar nebula. A pyroxene-anorthite-rich CAI, G92, has an  $^{16}\text{O}$ -rich signature like other CAIs but also has an order-of-magnitude less  $^{26}\text{Mg}$ -excess in anorthite, corresponding to a  $(^{26}\text{Al}/^{27}\text{Al})_0$  of  $(5.21 \pm 0.54) \times 10^{-6}$ . As there is no evidence for a later Mg isotopic disturbance, G92 anorthite is interpreted to have formed by interaction with <sup>16</sup>O-rich nebular gas at 2–3 Ma after CAI formation. With the observation that <sup>16</sup>O-rich refractory inclusions, relatively <sup>16</sup>O-poor chondrules, and extremely <sup>16</sup>O-poor cosmic symplectites within Acfer 094 all plot on the PCM line, it suggests that <sup>16</sup>O-rich nebular gas and extremely <sup>16</sup>O-poor primordial volatiles represent mass-independent fractionated endmembers in the early solar system and that the PCM line represents a mixing line of these two endmembers. © 2016 Elsevier Ltd. All rights reserved.

Keywords: Oxygen isotope; Al-Mg systematics; Refractory inclusion; Solar nebula; SIMS; Acfer 094

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#### 1. INTRODUCTION

In the early Solar System, evidence for an <sup>16</sup>O-rich isotopic signature that fractionated independent of mass  $(\Delta^{17}O \equiv \delta^{17}O - 0.52 \times \delta^{18}O \sim -24\%)$  has been found in Ca-, Al-rich inclusions (CAIs) and amoeboid olivine aggregates (AOAs) (Clayton et al., 1977; Hiyagon and Hashimoto, 1999; Krot et al., 2002, 2010a). As refractory inclusions (CAIs and AOAs) are commonly interpreted to be the first solar system condensates (Grossman, 1972; Grossman et al., 2008), their oxygen isotope ratios are inferred to reflect the primordial characteristics of the earliest refractory dust forming environment in the solar nebula. In contrast, oxygen isotope ratios of chondrules that formed ~2 million years (Ma) after CAIs (Kita and Ushikubo, 2012; Kita et al., 2013; Ushikubo et al., 2013) have a smaller mass-independent oxygen isotopic anomaly relative to CAIs, indicating a later oxygen isotope environment that was relatively <sup>16</sup>O-poor (e.g., Clayton et al., 1983; Clayton, 2003; Connolly and Huss, 2010; Kita et al., 2010; Krot et al., 2010b; Rudraswami et al., 2011; Weisberg et al., 2011; Nakashima et al., 2012; Ushikubo et al., 2012; Schrader et al., 2013, 2014; Tenner et al., 2013, 2015).

Since some CAIs exhibit evidence for later re-heating processes after their initial formation, including significant differences in  $\Delta^{17}$ O values between primary phases and secondary phases (Yurimoto et al., 1998; Hsu et al., 2000; Fagan et al., 2007; Ushikubo et al., 2007; Krot et al., 2008; MacPherson et al., 2012; Kawasaki et al., 2015), they are expected to record temporal changes of oxygen isotope environments within the solar nebula. In addition, detailed petrologic and isotopic studies of fine-textured Wark-Lovering rims surrounding coarse-grained CAIs, which consists of (1) a spinel layer with minor perovskite and hibonite (inner part); (2) a melilite/anorthite/secondary altered phase layer (middle part); and (3) a Ti-Al-bearing diopside layer (outer part) (Wark and Lovering, 1977), suggest that variability in oxygen isotope ratios existed when the Wark and Lovering rims formed (e.g., Yoshitake et al., 2005; Simon et al., 2011, 2016). Impartantly, however, Bodénan et al. (2014) observed that such an oxygen isotopic variability is not recognized among Wark-Lovering rims from pristine carbonaceous chondrite CAIs. This suggests that the oxygen isotopic variability found in the aforementioned studies, from which refractory inclusions from higher petrologic type chondrites were investigated, could be due to later isotopic disturbances in chondritic parent bodies. If oxygen isotopic variability (16O-rich and <sup>16</sup>O-poor environments) existed in the early solar system when coarse-grained CAIs and Wark-Lovering rim formed, it is expected that evidence for oxygen isotopic variability would also be recorded within fine-grained CAIs and AOAs (Grossman and Ganapathy, 1976; Grossman and Steele, 1976).

Although coarse-grained CAIs are advantageous for in situ high-precision isotope analyses, they experienced re-melting after aggregation of precursor dust condensates and they commonly have positively fractionated Mg and Si isotope ratios. As positive isotopic fractionation in Mg and Si provides evidence for significant evaporative loss

by re-melting, primary oxygen isotope ratios of such coarse-grained CAIs are probably modified (e.g., Wang et al., 2001; Alexander, 2004). In contrast, the texture of fine-grained CAIs and AOAs (e.g., small grain sizes of major phases and complex nodules) indicates that they formed by aggregation of primary condensates from early solar nebular gas. As such, fine-grained CAIs and AOAs are perhaps the best candidates for recording pristine oxygen isotope ratios of early solar system condensates. However, the petrology and isotopic compositions of such fine-scale materials are particularly susceptible to metamorphism while on chondritic parent bodies. Thus, in order to establish that isotope signatures of fine-grained refractory inclusions are indeed primordial nebular signatures, selection of pristine CAIs and AOAs that properly rule out evidence for parent body metamorphism is critically

In this study, we measured oxygen isotope ratios and <sup>26</sup>Al-<sup>26</sup>Mg systematics of fine-grained CAIs and AOAs from Acfer 094 (C-ungrouped 3.00) and Yamato-81020 (Y-81020, CO3.05). These refractory inclusions could have recorded pristine isotope characteristics because Acfer 094 and Y-81020 are two of the least metamorphosed carbonaceous chondrites (Grossman and Brearley, 2005; Kimura et al., 2008). For example, Acfer 094 chondrules preserve intrinsic oxygen isotope ratios without any indication of a later oxygen isotopic disturbance, even in mesostasis glass that is highly sensitive to fluid assisted parent body metamorphism (Ushikubo et al., 2012). In addition, chondrules from both Acfer 094 and Y-81020 have <sup>26</sup>Al-<sup>26</sup>Mg isotope systematics showing no evidence for a later disturbance (Kunihiro et al., 2004; Kurahashi et al., 2008; Hutcheon et al., 2009; Ushikubo et al., 2013). Thus, oxygen isotope ratios and <sup>26</sup>Al-<sup>26</sup>Mg isotope systematics of fine-grained refractory inclusions from the same meteorites should dependably elucidate the evolution of oxygen isotope ratios in the solar nebula, recording their formation and interaction with ambient gas before accreting to the chondritic parent body.

### 2. SAMPLES

Ten refractory inclusions, including five spinel-meliliterich CAIs (G5, G16, G49, G104, and Y81020-E-8), one pyroxene-anorthite-rich CAI (G92), and four AOAs (G17, G28, G44, and G58) were selected for investigation by secondary ion mass spectrometry (SIMS) (Figs. 1–3). Y81020-E-8 is from Y-81020 and the others are from Acfer 094. The classification of Acfer 094 refractory inclusions by Krot et al. (2004a) is applied in this study.

Three of the five spinel-melilite-rich CAIs (G5, G16, and G49) have a fine-grained, complex nodular texture. CAI G5 is an irregular-shaped aggregate of multiple nodules (Fig. 1a). It consists of gehlenitic melilite (Åk<sub>6</sub>) and spinel, along with anorthite and diopside rims. Some nodules do not contain melilite. CAI G16 is a large CAI (ca.  $300~\mu m \times 500~\mu m$  in size, Fig. 1b) consisting of multiple nodules. The larger nodules consist of melilite (Åk<sub>8–10</sub>) and sub-micron spinel grains surrounded by a diopside rim (Fig. 1f). Regarding the smaller nodules, spinel is rare,

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