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Oxygen isotopic variations in the outer margins and Wark–Lovering rims of refractory inclusions

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Abstract

Oxygen isotopic variations across the outer margins and Wark–Lovering (WL) rims of a diverse suite of six coarse-grained Types A and B refractory inclusions from both oxidized and reduced CV3 chondrites suggest that CAIs originated from a ^{16}O-rich protosolar gas reservoir and were later exposed to both relatively ^{17,18}O-rich and ^{16}O-rich reservoirs. The O-isotope profiles of CAIs can be explained by changes in the composition of gas near the protoSun or the migration of CAIs through a heterogeneous nebula. Variability within the inclusion interiors appears to have been set prior to WL rim growth. Modeling the isotopic zoning profiles as diffusion gradients between inclusion interiors and edges establishes a range of permissible time–temperature combinations for their exposure in the nebula. At mean temperatures of 1400 K, models that match the isotope gradients in the inclusions yield timescales ranging from 5×10^3 to 3×10^5 years. Assuming CAIs originated with a relatively ^{16}O-rich (protosolar) isotopic composition, differences among the melilite interiors and the isotopic gradients in their margins imply the existence of a number of isotopically distinct reservoirs. Evidence at the edges of some CAIs for subsequent isotopic exchange may relate to the beginning of rim formation. In the WL rim layers surrounding the interiors, spinel is relatively ^{16}O-rich but subtly distinct among different CAIs. Melilite is often relatively ^{16}O-poor, but rare relatively ^{16}O-rich grains also exist. Pyroxene generally exhibits intermediate O-isotope compositions and isotopic zoning. Olivine in both WL and accretionary rims, when present, is isotopically heterogeneous. The extreme isotopic heterogeneity among and within individual WL rim layers and in particular, the observed trends of outward ^{16}O-enrichments, suggest that rims surrounding CAIs contained in CV3 chondrites, like the inclusions themselves, formed from a number of isotopically distinct gas reservoirs. Collectively, these results support numerical protoplanetary disk models in which CAIs were transported between several distinct nebular reservoirs multiple times prior to accretion onto a parent body.

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1. INTRODUCTION

Calcium–aluminum-rich inclusions (CAIs) have refractory mineral assemblages consisting of phases predicted to condense from a hot nebula of solar composition

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(e.g., Grossman, 1972). We now understand that the mineralogy of coarse-grained CAIs is the result of crystallization from melts and that their reset Al–Mg chronologies (MacPherson et al., 2012) and decoupled stable isotope compositions of refractory and more volatile elements (Simon and DePaolo, 2010) likely reflect substantial processing of primordial condensates. Despite evidence for a complicated history, their old ages (Amelin et al., 2002; Bouvier and Wadhwa, 2010; Connelly et al., 2012) attest to the fact that they represent some of the most primitive solar system materials and likely provide our best window into the earliest conditions of the solar protoplanetary disk. After crystallization of their interiors, most inclusions were rimmed by a common mineral layering sequence, implying a fundamental change in their environment. Previous investigations have interpreted these Wark–Lovering (WL) rims (Wark and Lovering, 1977) as products of formation from nebular reservoirs with pressures (Simon et al., 2005), temperatures (Young et al., 2005), and isotopic compositions (Krot et al., 2002; Aleon et al., 2007; Yurimoto et al., 1998; Yoshitake et al., 2005; Simon et al., 2011) distinct from those indicated by the interiors. These environmental changes likely not only produced the mineralogical variability of WL rims (Dyl et al., 2011; Simon et al., 2005), but could also have led to mineralogical (Keller and Buseck, 1991; Metzler et al., 1992) and isotopic (Fagan et al., 2004a; Simon and Young, 2011) changes within the refractory inclusions themselves.

Compared to the terrestrial planets, many CAIs exhibit an ^{16}O-rich composition ($\delta^{18}O \approx \delta^{17}O \leqslant -40‰$) that likely reflects the O-isotopic composition of protosolar gas in the nebula (McKeegan et al., 2011). Yet, it has been known for decades that individual CAIs have heterogeneous O-isotopic compositions, in some cases spanning the range between protosolar to planetary reservoirs (Clayton et al., 1977). One can postulate that CAIs condensed, melted, crystallized, and were rimmed in the relatively ^{16}O-rich protosolar reservoir, and that any internal O-isotopic heterogeneity resulted from mineralogically controlled isotope exchange with a planetary reservoir ($\delta^{18}O \sim 0‰$) on the chondrite parent body, e.g., Wasson et al. (2001). Alternatively, the O-isotopic heterogeneity in CAIs can be explained by formation from both relatively ^{16}O-rich and ^{16}O-poor reservoirs of nebular gas (Clayton et al., 1977; Yurimoto et al., 1998; Krot et al., 2002; Yoshitake et al., 2005; Aleon et al., 2007; Simon et al., 2011). This might occur if igneous CAIs formed through admixing melanges of isotopically heterogeneous material and/or through transport, exposure, and exchange with distinct nebular reservoirs. As of yet the locations and identities of the reservoirs, the formation processes involved in recording the isotopic shifts, and the timing of isotopic exchange are not well-constrained.

In a prior study (Simon et al., 2011), secondary ion mass spectrometry (SIMS) measurements of the outer margin and WL rim of the Allende compact Type A CAI A37 revealed large O-isotope heterogeneity across the outermost ~70 μm of the inclusion and showed >25‰ variations in $\Delta^{17}O$ (departure from the terrestrial mass fractionation line; $\Delta^{17}O = \delta^{17}O - 0.52 \times \delta^{18}O$) within the ~100 μm-thick WL rim. Those workers proposed that the variations reflected either transport of the inclusion between distinct nebular reservoirs inherited from the parental molecular cloud, or a time-varying oxygen isotopic composition of a region of the nebula, such as the relatively ^{16}O-poor gas predicted for the outer margins of the disk by photochemical self-shielding models (Lyons and Young, 2005). Although some rims surrounding CAIs exhibit extreme O-isotopic heterogeneity, others are isotopically homogeneous (Matzel et al., 2013; Bodénan et al., 2014). It is noteworthy that some rims consist of only a single pyroxene layer (e.g., Bodénan et al., 2014), while others are far more complex, such as the WL rim surrounding Leoville CAI L6 (this study) that consists of at least 9 distinct mineral layers. In order to address this discrepancy, studies such as ours that involve inclusions with more complex rims and textural evidence spanning a range of formation processes are needed to evaluate solar protoplanetary disk evolution models, such as those of Cuzzi et al. (2003), Boss (2004), and Ciesla (2007).

The nature of chondritic host rocks and the potentially disparate formation histories of their constituent components create challenges to interpretation of the O-isotopic records of individual CAIs. In particular, parent body alteration may produce O-isotope heterogeneity. To document O-isotopic heterogeneity to test models for the evolution of the protoplanetary disk, this work presents the results of coordinated high-spatial resolution oxygen and petrologic investigations of CAIs with a range of mineralogical compositions and alteration histories from the Allende, Efremovka, and Leoville CV3 chondrites.

2. METHODS

2.1. Oxygen isotopic zoning profiles by NanoSIMS

Oxygen isotope measurements were performed using the NanoSIMS 50 at Lawrence Livermore National Laboratory (LLNL), following the method developed by Simon et al. (2011), during ~5 weeks of analysis over an ~3-year period (Table 1, Appendix I). Measurements were performed by sputtering the polished surfaces of CAIs in carbon-coated thin sections and epoxy embedded thick sections with a 16 keV, ~12 pA Cs^+ beam focused to a ~100 nm spot size and rastered over 2 × 2 μm regions. Negative secondary ions of $^{16}O^-$, $^{17}O^-$, $^{18}O^-$, and $^{28}Si^-$ (or $^{30}Si^-$) were collected in a Faraday cup and three separate electron multipliers, respectively. Each measurement comprises $(3–6) \times 10^5$ counts of $^{17}O^-$ and $(1.5–3.0) \times 10^6$ counts of $^{18}O^-$ collected during a 368-s analysis time. The data were corrected for detector dead time. Measured uncertainties reflect the standard deviation of isotope ratio data acquired over 300 rastered measurement cycles and follow a Poisson distribution. Uncertainties reported in Table 1 reflect a combination of counting statistics and either 2 standard errors (se), or 2 standard deviations (sd) of the average value of the measured mineral standards used for sample-standard bracketing (Appendices I and II). A mass resolving power of ~7000 was used to resolve $^{17}O^-$ from $^{16}OH^-$. Monitoring the $Si^-/^{16}O^-$ ratio (in addition to pre- and post-analysis

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