

# A link between oxygen, calcium and titanium isotopes in $^{26}\text{Al}$ -poor hibonite-rich CAIs from Murchison and implications for the heterogeneity of dust reservoirs in the solar nebula

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## Abstract

PLACs (platy hibonite crystals) and related hibonite-rich calcium-, aluminum-rich inclusions (CAIs; hereafter collectively referred to as PLAC-like CAIs) have the largest nucleosynthetic isotope anomalies of all materials believed to have formed in the solar system. Most PLAC-like CAIs have low inferred initial  $^{26}\text{Al}/^{27}\text{Al}$  ratios and could have formed prior to injection or widespread distribution of  $^{26}\text{Al}$  in the solar nebula. In this study, we report  $^{26}\text{Al}$ – $^{26}\text{Mg}$  systematics combined with oxygen, calcium, and titanium isotopic compositions for a large number of newly separated PLAC-like CAIs from the Murchison CM2 chondrite (32 CAIs studied for oxygen, 26 of these also for  $^{26}\text{Al}$ – $^{26}\text{Mg}$ , calcium and titanium). Our results confirm (1) the large range of nucleosynthetic anomalies in  $^{50}\text{Ti}$  and  $^{48}\text{Ca}$  (our data range from  $-70\text{‰}$  to  $+170\text{‰}$  and  $-60\text{‰}$  to  $+80\text{‰}$ , respectively), (2) the substantial range of  $\Delta^{17}\text{O}$  values ( $-28\text{‰}$  to  $-17\text{‰}$ , with  $\Delta^{17}\text{O} = \delta^{17}\text{O} - 0.52 \times \delta^{18}\text{O}$ ), and (3) general  $^{26}\text{Al}$ -depletion in PLAC-like CAIs.

The multielement approach reveals a relationship between  $\Delta^{17}\text{O}$  and the degree of variability in  $^{50}\text{Ti}$  and  $^{48}\text{Ca}$ : PLAC-like CAIs with the highest  $\Delta^{17}\text{O}$  ( $\sim -17\text{‰}$ ) show large positive and negative  $^{50}\text{Ti}$  and  $^{48}\text{Ca}$  anomalies, while those with the lowest  $\Delta^{17}\text{O}$  ( $\sim -28\text{‰}$ ) have small to no anomalies in  $^{50}\text{Ti}$  and  $^{48}\text{Ca}$ . These observations could suggest a physical link between anomalous  $^{48}\text{Ca}$  and  $^{50}\text{Ti}$  carriers and an  $^{16}\text{O}$ -poor reservoir. We suggest that the solar nebula was isotopically heterogeneous shortly after collapse of the protosolar molecular cloud, and that the primordial dust reservoir, in which anomalous carrier phases were heterogeneously distributed, was  $^{16}\text{O}$ -poor ( $\Delta^{17}\text{O} \geq -17\text{‰}$ ) relative to the primordial gaseous ( $\text{CO} + \text{H}_2\text{O}$ )

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reservoir ( $\Delta^{17}\text{O} < -35\text{‰}$ ). However, other models such as CO self-shielding in the protoplanetary disk are also considered to explain the link between oxygen and calcium and titanium isotopes in PLAC-like CAIs.

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## 1. INTRODUCTION

Oxygen is unique among the chemical elements in the solar system in that significant fractions were present in solid and gaseous states over a large range of temperatures in the solar nebula. Meteoritic materials show mass-independent variations in oxygen isotopes that are of extraordinary magnitude for a major rock-forming element. For example, many calcium-, aluminum-rich inclusions (CAIs), the oldest dated materials that formed in the solar system (e.g., Connelly et al., 2012), are enriched in  $^{16}\text{O}$  by  $\sim 5\%$  compared to terrestrial planets and asteroids (Clayton et al., 1973; MacPherson et al., 2008; Makide et al., 2009; Ushikubo et al., 2011). The degree of enrichment/depletion in  $^{16}\text{O}$  is commonly expressed using the  $\Delta^{17}\text{O}$  value, ( $\Delta^{17}\text{O} = \delta^{17}\text{O} - 0.52 \times \delta^{18}\text{O}$ , with  $\delta$ -values being defined as  $\delta^i\text{O} = [(^i\text{O}/^{16}\text{O})_{\text{unknown}} / (^i\text{O}/^{16}\text{O})_{\text{VSMOW}} - 1] \times 1000$ , for isotope  $i$  and where VSMOW corresponds to Vienna standard mean ocean water; Baertschi, 1976), which allows for comparison of  $^{16}\text{O}$ -enrichment even for materials with mass-dependent isotope fractionation in oxygen. Analyses of solar wind collected by NASA's Genesis mission confirmed a prediction (Clayton, 2002) that the Sun, like most CAIs, is enriched in  $^{16}\text{O}$  relative to the Earth and terrestrial planets (McKeegan et al., 2011). As the Sun contains most of the mass of the solar system and is thus representative of its average composition, it is not CAIs but terrestrial planets that are highly aberrant in their oxygen isotopic composition. Several models have been proposed to explain this discrepancy, which are either based on inheritance of distinct nucleosynthetic oxygen isotope reservoirs from the molecular cloud core (Krot et al., 2010) or isotopically selective gas phase reactions. The latter include self-shielding during photodissociation of CO molecules followed by water ice/CO gas fractionation and thermal processing of silicate dust in a gaseous reservoir enriched in  $^{17}\text{O}$  and  $^{18}\text{O}$  (Clayton, 2002; Yurimoto and Kuramoto, 2004; Lyons and Young, 2005), as well as symmetry-based reactions (Thiemens, 2006).

Many CAIs seem to have formed in a uniform oxygen isotopic reservoir that was slightly depleted in  $^{16}\text{O}$  compared to the solar  $\Delta^{17}\text{O}$  value of  $-28.4 \pm 3.6\text{‰}$  ( $\pm 2\sigma$ ; McKeegan et al., 2011). For example, CAIs from CR chondrites and the primitive ungrouped chondrite Acfer 094 commonly show a  $\Delta^{17}\text{O}$  value of approximately  $-23\text{‰}$  (Makide et al., 2009; Ushikubo et al., 2011). Many CV CAIs likely formed in a similar reservoir, as indicated by the common  $\Delta^{17}\text{O}$  value of  $\sim -23\text{‰}$  in spinel, hibonite, fassaite (Al, Ti-diopside), and forsterite grains (e.g., MacPherson et al., 2008). More  $^{16}\text{O}$ -poor compositions in

melilite and anorthite in the same CV CAIs are commonly interpreted as subsequent mineral-dependent exchange with or remelting in a  $^{16}\text{O}$ -poor reservoir (Krot et al., 2009, and references therein). However,  $\Delta^{17}\text{O}$  variations in some CV and CO CAIs have also been attributed to fluctuations in the isotopic composition of the CAI formation region (e.g., Simon et al., 2011; Kawasaki et al., 2012; Park et al., 2012). Additional important exceptions from the uniform CAI value are several grossite-rich CAIs and a chondrule from CH carbonaceous chondrites that are  $^{16}\text{O}$ -enriched relative to the Sun's value ( $\Delta^{17}\text{O}$  as low as  $-37\text{‰}$ ; Kobayashi et al., 2003; Krot et al., 2008, 2015; Gounelle et al., 2009).

In addition to oxygen isotopes, CAIs provide important information about the presence of various nucleosynthetic components in the solar nebula. For example, most CV CAIs incorporated the short-lived (half-life of  $\sim 0.7$  Ma) radionuclide  $^{26}\text{Al}$  at a uniform  $^{26}\text{Al}/^{27}\text{Al}$  ratio of  $\sim 5.2 \times 10^{-5}$ , called the canonical ratio (Jacobsen et al., 2008; MacPherson et al., 2012). In addition, CAIs show small-scale anomalies in stable isotopes like  $^{50}\text{Ti}$ ,  $^{96}\text{Zr}$ , and  $^{92}\text{Mo}$ , typically within  $\sim 1\text{‰}$  of the terrestrial values (e.g., Schönbachler et al., 2003; Trinquier et al., 2009; Burkhardt et al., 2011; Akram et al., 2013). In the context of this study, we define “regular CAIs” as those having (1) a uniform  $\Delta^{17}\text{O}$  of about  $-23\text{‰}$ , (2) a canonical  $^{26}\text{Al}/^{27}\text{Al}$  ratio, and (3) small-scale nucleosynthetic anomalies.

When compared to regular CAIs, FUN (fractionated and unidentified nuclear effects) CAIs and PLACs (platy hibonite crystals) are isotopically distinct. In particular, both FUN CAIs and PLACs have lower inferred  $^{26}\text{Al}/^{27}\text{Al}$  ratios than regular CAIs, and some have evidence for formation in the absence of  $^{26}\text{Al}$  (e.g., Wasserburg et al., 1977; Ireland, 1990; Liu et al., 2009; Park et al., 2013). This  $^{26}\text{Al}$  deficiency is commonly attributed to an early formation prior to arrival of  $^{26}\text{Al}$  rather than a late formation after  $^{26}\text{Al}$  decay, because FUN CAIs and PLACs preserve larger nucleosynthetic anomalies than regular CAIs (Wood, 1998; Sahijpal and Goswami, 1998). In PLACs, nucleosynthetic anomalies can be up to two orders of magnitude greater than in CV CAIs (e.g., PLACs have a  $\delta^{50}\text{Ti}$  range of  $\sim 300\text{‰}$ ; Hinton et al., 1987; Ireland, 1990; Liu et al., 2009); in FUN CAIs, the range of anomalies is intermediate between PLACs and regular CV CAIs (e.g., FUN CAIs have a  $\delta^{50}\text{Ti}$  range of  $\sim 35\text{‰}$ ; Krot et al., 2014; Park et al., 2014). The  $\Delta^{17}\text{O}$  values of these  $^{26}\text{Al}$ -poor, anomalous CAIs also span a wide range from  $\sim -28\text{‰}$  to  $\sim 0\text{‰}$  (Liu et al., 2009; Krot et al., 2008, 2010, 2014, 2015), indicating oxygen isotopic heterogeneity in the earliest stages of solar system history.

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