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# Oxygen isotopes in calcium–aluminum-rich inclusions from enstatite chondrites: new evidence for a single CAI source in the solar nebula

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#### Abstract

Calcium–aluminum-rich inclusions (CAIs) from enstatite chondrites have large <sup>16</sup>O excesses, similar to CAIs in carbonaceous and ordinary chondrites, and are also similar in morphology, mineralogy and Al–Mg isotopic systematics. These similarities provide new evidence that most CAIs might have formed in a single, restricted nebular locale and were then distributed unevenly throughout the various chondrite accretion regions. © 2000 Elsevier Science B.V. All rights reserved.

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## 1. Introduction

Calcium–aluminum-rich inclusions (CAIs) from primitive chondritic meteorites were among the first-formed solids in the solar nebula  $\sim 4.6$  Ga, and therefore bear important physical and chemical information about the earliest stages of solar system evolution. CAIs are common constituents in most varieties of carbonaceous (C) chondrite, but are rare in ordinary (O) and enstatite (E) chondrites [1]. Among their many unusual properties, CAIs from C-chondrites are markedly en-

\* Corresponding author. Tel.: +1-202-357-2260; Fax: +1-202-357-2476; E-mail: glenn@volcano.si.edu riched in <sup>16</sup>O relative to other meteoritic compo-

nents [2,3]. A recent study [4] showed that two

CAIs from O-chondrites share similar distinctive oxygen isotopic features to those from C-chondrites. The oxygen isotopic compositions of meteoritic components are generally assumed to reflect isotopic variations among the different locations within the meteorite-forming regions of the solar nebula. For example, the chondrules from C- and O-chondrites have different oxygen isotopic compositions and may have formed in different nebular environments [3]. Therefore, the observation that C- and O-chondrite CAIs share a similar oxygen isotopic composition suggests the intriguing possibility [4] that CAIs from all chondrite varieties formed in a single restricted region of the solar nebula and were then distributed rather unevenly throughout the various chondrite accre-

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tion regions. One test of this model is to study CAIs from other chondrite varieties.

E-chondrites are highly reduced rocks whose mineral assemblages and bulk oxygen isotopic compositions are so distinct from those of other chondrite types that they likely formed under very different redox conditions and in a different region of the solar nebula from C- and O-chondrites [5]. Recent reports [6-8] have demonstrated, however, that CAIs from E-chondrites contain such reduced minerals only as late-stage overprints, not as primary phases. In all other respects these CAIs are similar to those from C- and O-chondrites, including morphology, texture, primary mineralogy and mineral chemistry, and Al-Mg isotopic systematics. These observations suggest that E-chondrite CAIs either formed during an earlier and more oxidized evolutionary stage in the E-chondrite accretion locale or else were introduced from an outside source, i.e. possibly from the postulated single CAI-forming region. To discriminate between these two possibilities and thereby test the idea of a single CAI-forming region, we have carried out oxygen isotopic analvses of CAIs and an Al-rich chondrule from E-chondrites and explored the implications of these data for CAI formation and oxygen isotopic distributions in the early solar nebula. Preliminary results of this study were reported in an abstract [9].

### 2. Analytical techniques

We analyzed 10 CAIs and one Al-rich chondrule from three unequilibrated E-chondrites (EET 87746, GRO 95517 and Sahara 97072) for their oxygen isotopic compositions. The CAIs were located via systematic X-ray area mapping of entire thin sections for Na, Mg, Al, Si, K, Ca, Fe and Ti, using a JEOL-840A scanning electron microscope (SEM) and a Kevex energy-dispersive detector system. Details of the analytical methods used for measuring mineral chemistry are given in [8].

Oxygen isotopic compositions were measured with the UCLA ims 1270 ion microprobe using analytical techniques similar to those previously described [4]. Because the CAIs from E-chondrites are small and some of them were extensively sputtered in previous ion probe studies [8], a primary Cs<sup>+</sup> beam of ~10  $\mu$ m in diameter was used for all the analyses. Subsequent SEM examination showed that the majority of ion probe craters were targeted on single mineral phases. In some areas, however, it was not possible to avoid analyzing mixtures of different minerals even at ~10  $\mu$ m resolving scale. The proportions of different minerals of those sputtered areas were obtained from digital phase analysis of their backscattered electron images (Table 1).

#### 3. Sample descriptions

Four of the CAIs, E4631-1<sup>1</sup> (melilite-spinel), E4640-1 (hibonite-spinel), S721-2 (spinel-pyroxene) and E4631-3 (hibonite-pyroxene microspherule), were previously described in [6,8]. The other six CAIs include one corundum-spinel (G1724-1), two spinel-rich (E4631-9, S721-12) and three hibonite-rich (E4642-2, E4642-4, S721-5) inclusions. The three hibonite-rich inclusions (all  $< 50 \ \mu m$ ) were analyzed previously for Al-Mg isotopic systematics [6,8]. They consist of stubby hibonite plates ( $\sim 10 \times 20$  µm), surrounded by spinel (Fig. 1AA) or sodalite. The two spinel-rich inclusions (E4631-9, S721-12; Fig. 1B) have spinel cores that are rimmed by pyroxene and/or sodalite. G1724-1 ( $\sim$  30×20 µm; Fig. 1C) consists of densely intergrown corundum and spinel, with abundant troilite decorating the corundum-spinel interstices, and a partial exterior rim of pyroxene.

The Al-rich chondrule E4642-1 ( $\sim 100 \ \mu m$  diameter; Fig. 1D) consists of spinel microphenocrysts (up to  $\sim 20 \times 50 \ \mu m$ ) and feathery aluminous diopside in a mesostasis that in some places is clear glass (CaO < 2%, Na<sub>2</sub>O ~ 10–

<sup>&</sup>lt;sup>1</sup> Inclusions are labeled as follows: the first three characters represent the first letter and last two numbers from the meteorite name, the next two numbers are the individual thin section number and the last number is the specific object identification number: e.g. E4631-1 is CAI #1 from EET 87746 PTS#31, S721-5 is CAI#5 from Sahara 97072 PTS#1, and G1724-1 is CAI#1 from GRO95517 PTS#24.

Table 1

Oxygen isotopic compositions of CAIs and an Al-rich chondrule from E-chondrites and approximate proportions of minerals analyzed in each ion probe spot

Spot	δ <sup>18</sup> O <sup>a</sup> (‰)	δ <sup>17</sup> Ο (%)	Δ <sup>17</sup> O <sup>b</sup> (‰)	Mineralogy (vol%)
E4631-1 #1, b	$-44.3 \pm 1.8$	$-45.6 \pm 1.3$	$-46.4 \pm 1.4$	
E4631-1 #2	$-42.6 \pm 2.5$	$-42.1 \pm 1.2$	$-44.4 \pm 1.6$	Mel (80), Sp (20)
E4631-1 Sp	$-47.8 \pm 1.8$	$-47.4 \pm 1.2$	$-50.5 \pm 1.3$	Sp (95), Mel, Pv
E4631-3 Pyx, a	$-41.6 \pm 1.5$	$-47.5 \pm 1.4$	$-43.2 \pm 1.4$	Pyx (100)
E4631-3 Pyx, b	$-43.1 \pm 1.2$	$-48.5 \pm 1.2$	$-45.0 \pm 1.2$	
E4631-3 Hb	$-44.3 \pm 1.8$	$-51.3 \pm 1.2$	$-46.4 \pm 1.4$	Hib (99), Pv
E4631-9 #1	$-48.1 \pm 1.5$	$-49.2 \pm 1.1$	$-51.0 \pm 1.2$	Sp (80), Pyx (20)
E4640-1 #1	$-47.8 \pm 1.3$	$-49.1 \pm 1.2$	$-50.6 \pm 1.2$	Sp (66), Hib (21), Tr (13)
E4640-1 #2	$-42.8 \pm 1.3$	$-44.0 \pm 1.1$	$-44.6 \pm 1.1$	Sp (47), Hib (36), Tr (16)
E4642-1 Sp, a	$-2.7 \pm 1.4$	$-3.5 \pm 0.9$	$-2.1 \pm 1.0$	Sp (100)
E4642-1 Sp, b	$-1.1 \pm 1.4$	$-1.9 \pm 1.2$	$-1.4 \pm 1.3$	
E4642-1 Pyx	$0.9 \pm 1.4$	$-1.9 \pm 1.0$	$-2.4 \pm 1.1$	Pyx (100)
E4642-1 Gls	$-2.3 \pm 1.3$	$-2.1 \pm 1.1$	$-0.9 \pm 1.2$	Gls (100)
E4642-2 Hb	$-39.6 \pm 1.5$	$-41.5 \pm 1.4$	$-40.8 \pm 1.4$	Hib (99), Pv
E4642-5 Hb	$-49.1 \pm 1.6$	$-53.2 \pm 1.5$	$-52.1 \pm 1.6$	Hib (99), Pv
G1724-1 #1	$-34.8 \pm 1.3$	$-34.9 \pm 1.1$	$-35.0 \pm 1.1$	Cor (66), Sp (31), Tr (4)
G1724-1 #2	$-34.5 \pm 1.2$	$-35.1 \pm 1.0$	$-34.7 \pm 1.0$	Cor (75), Sp (15), Tr (10)
S72A-2 Sp	$-40.7 \pm 1.6$	$-41.8 \pm 1.3$	$-42.1 \pm 1.3$	Sp (97), Tr
S72A-5 Hb	$-45.8 \pm 1.7$	$-45.0 \pm 1.3$	$-48.2 \pm 1.3$	Hib (99), Tr
S72A-12 Sp	$-48.5 \pm 1.5$	$-49.1 \pm 1.2$	$-51.5 \pm 1.2$	Sp (99), Tr

Abbreviations: Mel, melilite; Sp, spinel; Pyx, pyroxene; Hib, hibonite; Cor, corundum; Pv, perovskite; Gls, glass; Tr, troilite. 10 errors.

<sup>a</sup>Oxygen isotopic compositions are expressed as  $\delta$ -values, the deviation in % relative to a standard composition (standard mean ocean water, SMOW):  $\delta^{m}O = [({}^{m}O/{}^{16}O)_{sample}/(({}^{m}O/{}^{16}O)_{sMOW}-1] \times 1000$ , where m = 17 or 18.

<sup>b</sup>The deviation of the oxygen isotopic composition from the TF line is expressed by  $\Delta^{17}$ O, where  $\Delta^{17}$ O= $\delta^{17}$ O-0.52 $\delta^{18}$ O.

11%) and elsewhere is glass+microcrystalline py-roxene dendrites.

#### 4. Results and discussion

All of our E-chondrite CAI data show large <sup>16</sup>O-enrichments (Table 1), with  $\delta^{18}$ O and  $\delta^{17}$ O values reaching -49 and -53‰, respectively (Fig. 2). Most of the data plot in a region characteristic of the maximum <sup>16</sup>O-enrichments observed in CAIs from C-chondrites, and all plot far from the distinct and restricted region occupied by whole-rock E-chondrites and their ferromagnesian chondrules [3,10,11]. Except for phases in the microspherule E4631-3, individual CAI minerals plot within 2 $\sigma$  error of the C-chondrite anhydrous mineral mixing line (CCAM; [12]). These <sup>16</sup>O-enriched compositions are also consis-

tent with the 'primitive' mixing line obtained by Young and Russell [13]. The Al-rich chondrule, E4621-1, has a relatively <sup>16</sup>O-depleted composition unlike the CAIs; its spinel, pyroxene and glass all plot (Fig. 2) in a restricted area on the three-isotope diagram, between the CCAM and the terrestrial fractionation (TF) lines and mostly on the Young and Russell line [13]. Fagan et al. [14] observed spinel from one E-chondrite CAI that also plots near the Young–Russell line at <sup>16</sup>O-depleted compositions similar to those in this chondrule, but unlike any other CAIs that they [14] or we studied.

All of the primary minerals within the individual E-chondrite CAIs we analyzed show similar levels of <sup>16</sup>O enrichment. The inclusion E4631-1 especially is noteworthy in this respect, because its melilite has comparably large <sup>16</sup>O-excesses to spinel in the same CAI and to hibonite and spinel



Fig. 1. Back-scattered electron images of E-chondrite objects; details are given in the text. (A) Hibonite-spinel inclusion E4642-2; (B) spinel-pyroxene inclusion S721-12; (C) corundum-rich inclusion G1724-1; (D) Al-rich chondrule E4642-1; the dark glass in the lower half of the chondrule is free of enclosed microcrystals, whereas the lighter 'glass' in the upper half is densely packed with tiny dendrites of pyroxene that are not resolved in this photo. Mineral abbreviations are explained in Table 1 except: Sod, sodalite.

in other CAIs. In contrast, the melilite in wellstudied CV3 chondrite CAIs is <sup>16</sup>O-poor relative to spinel, hibonite and pyroxene [3,12]. <sup>16</sup>O-rich melilite has been observed previously in CAIs from O- [4] and some C- (e.g. [15,16]) chondrites, and in a micrometeorite [17]. The isotopic nearhomogeneity of primary minerals in our E-chondrite CAIs and other cited examples (even, rarely, in CV3 CAIs; [16]) indicates that most CAIs from all chondrite varieties were originally <sup>16</sup>O-rich. The CV3 inclusions owe their isotopic heterogeneity to partial secondary exchange [12] and, indeed, it is they rather than the O- or E-chondrite CAIs that may be distinctive in this regard. The isotopic similarity of minerals in each E-chondrite CAI means they preserve approximately the original bulk isotopic composition of the host inclusion. The fact that these bulk compositions do not extend to values more negative than  $\delta^{18}O \approx \delta^{17}O \approx -50\%$  poses a problem for a nu-



Fig. 2. Oxygen isotopic compositions of individual minerals from CAIs and an Al-rich chondrule in E-chondrites. The data for hibonite–pyroxene microspherule E4631-3 and Al-rich chondrule E4642-1 are outlined to facilitate discussion. TF = terrestrial fractionation line; CCAM = C-chondrite anhydrous mixing line. Data for E-chondrite ferromagnesian chondrules are from [10]. Error bars are  $1\sigma$ .

cleosynthetic origin of the <sup>16</sup>O-enriched signature in CAIs, because admixture of an <sup>16</sup>O-rich component should have produced a wide range of compositions (e.g. [18]). In contrast, some massindependent gas-phase fractionation processes [19] can produce such a limited range of <sup>16</sup>O-enrichment, although incorporation of such signatures into condensed phases remains a problem [18].

The isotopic composition of the corundum in inclusion G1724-1 is unexpected. Corundum is calculated to be one of the most refractory of all phases [20], yet it is rare in CAIs. Given its chemical and crystallographic similarities to hibonite, one might reasonably expect corundum to preserve a <sup>16</sup>O-rich signature like hibonite. Yet, the corundum in G1724-1 contains relatively heavier oxygen ( $\delta^{17}O \approx \delta^{18}O \approx -35\%$ ) than phases in other E-chondrite CAIs. Whether this is due to formation of the corundum by, for example, a secondary solid-state process or to later remelting of the CAI in a <sup>16</sup>O-depleted environment is unclear, but clearly G1724-1 sampled an oxygen reservoir that was isotopically heavier than that originally sampled by most CAI hibonite and spinel.

Hibonite and pyroxene in the microspherule E4631-3 lie slightly off to the right (high <sup>18</sup>O side) of the CCAM line (Fig. 2). This deviation is unlikely to be due to an instrument artifact. It might in principle be explained as the result of mass fractionation but, unlike some other hibonite-rich inclusions such as HAL [21], the Mg isotopic composition of this microspherule [6,8] gives only a hint of mass fractionation ( $F_{Mg} = -5.3$ -0.2% /amu) that favors the light, not heavy, isotopes. Thus, it seems unlikely that the oxygen isotopic composition in E4631-3 is mass fractionated as a result of a process such as Rayleigh distillation. Rather, the unusual morphology, mineralogy and isotopic composition of E4631-3 reinforces the idea that this and similar hibonitepyroxene spherules from other chondrite types [22-24] define a distinct petrologic and isotopic class of CAIs.

Some mineralogic features of the E-chondrite CAIs, such as abundant Ti-rich troilite [8], are related to the highly reduced mineralogy of host E-chondrites and seem to set these CAIs apart from those in C- and O-chondrites. However, such minerals occur only as a late-stage overprint [6-8] on a primary CAI mineral assemblage that is in other respects like that in O- and C-chondrite CAIs. The E-chondrite CAIs formed earlier and under more oxidizing conditions than did the superimposed reduced minerals, suggesting the possibility that the CAIs may not have originated in the E-chondrite formation region at all. The oxygen isotope data support this latter idea. Bulk E-chondrites have oxygen isotope compositions that plot close to the TF line [11,25] and their constituent ferromagnesian chondrules fall in a very restricted range along a line of slope  $0.7 \pm 0.1$  on a three-isotope diagram [10] (Fig. 2). Taken at face value, an extrapolation of this short line does not intersect the field of E-chondrite CAIs presented in this report; it does however pass through the data for Al-chondrule E4621-1. Therefore, the E-chondrite CAIs may not represent the <sup>16</sup>O-rich end-member of the Echondrite mixing line. Neither can these CAIs be linked to E-chondrite whole rocks or their ferromagnesian chondrules simply through the CCAM or Young and Russell lines [12,13], as the bulk Echondrite data do not plot on either of those lines. Similar to the relationship between Mgand Al-rich chondrules in O-chondrites [26], the E-chondrite chondrules define a mixing line different from that in C-chondrites and unrelated to the E-chondrite CAIs. The CAIs and chondrules in E-chondrites originated in separate isotopic reservoirs from one another.

The CAIs from E-chondrites do share a common isotopic signature with, and are probably genetically related to, the CAIs from C- and O-chondrites (this study and [4,9,14]) even though the bulk C-, O- and E-chondrites have distinct oxygen isotopic compositions [3]. If oxygen isotopic compositions are indicative in some fashion of the nebular regions in which various bodies formed, our data are consistent with a model (e.g. [27]) in which all CAIs formed in the same nebular locale. However, the observed CAI size-sorting among the diverse chondrite groups (but see [27]) and, especially, the extensive partial isotopic exchange experienced mainly by CV3 CAIs, are unresolved issues with this model.

## 5. Conclusions

The oxygen isotope data presented in this study indicate that CAIs from E-chondrites have large <sup>16</sup>O excesses, similar to their counterparts from C- and O-chondrites. The diverse minerals within individual CAIs are isotopically similar to one another, and thus preserve CAI bulk compositions that were not more extreme than  $\delta^{18}$ O  $\approx \delta^{17}$ O  $\approx -50 \%$ . If those compositions originated through simple mixing of an <sup>16</sup>O-rich dust component into the CAI precursor material, that component must itself have had  $\delta^{18}O$  and  $\delta^{17}O$ not far from -50 to -60% in order to easily explain the limited range of original CAI bulk compositions (e.g. [18]). Alternatively, a process such as mass-independent fractionation [19] may be responsible for the <sup>16</sup>O-rich signature.

The oxygen isotopic compositions, as well as morphology, mineralogy and Al–Mg isotopic systematics, indicate that CAIs from E-chondrites are probably not indigenous to the highly reducing E-chondrite accretion region. This observation supports the idea that most CAIs from a variety of different chondrite classes formed in a common nebular region and were later distributed (and size-sorted) unequally throughout the various chondrite accretion regions [4,27].

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