EXTREME $^{26}\text{Mg}$ AND $^{17}\text{O}$ ENRICHMENTS IN AN ORGUEIL CORUNDUM: IDENTIFICATION OF A PRESOLAR OXIDE GRAIN

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ABSTRACT

A corundum ($\text{Al}_2\text{O}_3$) grain from the Orgueil meteorite is greatly enriched in $^{17}\text{O}$ and $^{26}\text{Mg}$. The measured $^{16}\text{O}^{18}\text{O}$ is 1028 ± 11 compared to ($^{16}\text{O}^{18}\text{O}$)$_{\text{SMOW}} = 2610$. This is the largest $^{17}\text{O}$ excess so far observed in any meteoritic material. The $^{26}\text{Mg}$ excess ($^{26}\text{Mg}^*$) is most plausibly due to in situ decay of $^{26}\text{Al}$. The inferred ($^{26}\text{Al}/^{27}\text{Al}$)$_{\text{C}}$ ratio of 8.9 × 10$^{-4}$ is ~18 times larger than the 5 × 10$^{-5}$ value commonly observed in refractory inclusions formed in the solar system. The large $^{17}\text{O}$ excess and high $^{26}\text{Mg}^*/^{27}\text{Al}$ ratio unambiguously identify this corundum as a presolar oxide grain. Enrichments in $^{17}\text{O}$ and $^{26}\text{Al}$ are characteristic of H-burning and point to red giant or AGB stars as likely sources.

Subject headings: circumstellar matter — dust, extinction — nuclear reactions, nucleosynthesis, abundances — stars: AGB and post-AGB — solar system: formation

1. INTRODUCTION

Isotope abundance anomalies and presolar grains in primitive chondritic meteorites show unequivocally that solar system formation did not completely erase the history recorded in material that became the solar system (see Clayton, Hinton, & Davis 1988; Anders & Zinner 1993). Prior to 1992, four types of presolar grains were known in meteorites: diamond, silicon carbide, titanium carbide, and graphite (see Anders and Zinner 1993; Ott 1993). All are carbon rich. The distinctive isotopic compositions, which differ grossly from the solar system composition in ways not explicable by mass-dependent fractionation, radioactive decay, or cosmic-ray interactions, identify these grains as circumstellar condensates. Spectroscopic observations show that ~70% of the dust ejected from stars is oxygen-rich (Gehrz 1989; Greenberg 1989), leading to the expectation that presolar oxides may be preserved. However, their isolation has proved very difficult as the population of oxide grains is overwhelmingly dominated by material derived from Ca-Al-rich inclusions (CAI) formed in the solar system. CI carbonaceous chondrites contain the lowest abundance of high-temperature minerals; anhydrous silicates are depleted by at least a factor of 10 relative to other chondrites, and no CAI have been observed in CI chondrites. To take advantage of this low “background” of refractory solar system materials, we focused our search for presolar oxides on acid residues from the Orgueil CI chondrite and report here the discovery of a presolar oxide grain. Preliminary reports were given by Huss et al. (1992, 1993a).

2. SAMPLE PREPARATION AND ANALYSIS

Acid residues were prepared following procedures developed by Tang & Anders (1988), Stone et al. (1991), and Huss & Lewis (1994). The Mg and O isotopic compositions of spinel, hibonite, and corundum were determined with the PANURGE IMS-3f ion microprobe. Measurements of Mg were made with a 0.5–1 nA $^{16}\text{O}^+$ primary beam defocused to a diameter of ~20 μm, and to increase transmission, a mass resolving power (MRP) of ~1300 was used. Oxygen was measured with a 0.5 nA $^{153}\text{Cs}^+$ primary ion beam defocused to a diameter of ~30 μm. The $^{16}\text{OH}^-$ intensity was never greater than twice the $^{17}\text{O}^-$ signal, and an MRP of ~5000 was adequate to ensure that $^{16}\text{OH}^-$ contribution to the $^{17}\text{O}^-$ signal was <2%. Measured $^{17}\text{O}/^{16}\text{O}$ and $^{18}\text{O}/^{16}\text{O}$ ratios are reported as $\delta^{17}\text{O}_{\text{SMOW}}$ and $\delta^{18}\text{O}_{\text{SMOW}}$, the respective permil deviations from $^{17}\text{O}/^{16}\text{O}_{\text{SMOW}} = 0.003831$ and $^{18}\text{O}/^{16}\text{O}_{\text{SMOW}} = 0.0020052$. Instrumental mass fractionation was determined from the $^{18}\text{O}/^{16}\text{O}$ ratio measured on Burana spinel ($\delta^{17}\text{O}_{\text{SMOW}} = 11.6\%$ and $\delta^{18}\text{O}_{\text{SMOW}} = 22.3\%$; R. N. Clayton and T. K. Mayeda, personal communication), and the measured ratios were corrected for mass fractionation with a power law.

3. RESULTS

3.1. Magnesium Isotopes

The Mg isotopic compositions of 24 spinel (MgAl$_2$O$_4$), six hibonite (CaAl$_2$O$_4$), and 30 corundum (Al$_2$O$_3$) grains were determined. Most spinels contain isotopically normal Mg. Three spinels exhibit evidence of mass-dependent isotope fractionation ($F_{\text{Mg}}$); the range in $F_{\text{Mg}}$ is within that observed for minerals in CAI. All hibonites have $F_{\text{Mg}}$ ≈ 0 and clearly resolved $^{26}\text{Mg}$ excesses ($^{26}\text{Mg}^*$). All the Orgueil corundum grains also have $F_{\text{Mg}}$ ≈ 0 (i.e., normal $^{25}\text{Mg}/^{26}\text{Mg}$), and nine of 30 grains have well resolved $^{26}\text{Mg}^*$. Eight of these nine grains with $^{26}\text{Mg}^*$ have $^{26}\text{Mg}^*/^{27}\text{Al}$ between ~1 × 10$^{-6}$ and 5 × 10$^{-5}$. Only one grain, corundum B, has a $^{26}\text{Mg}^*/^{27}\text{Al}$ ratio distinct from the canonical solar system value, $^{26}\text{Mg}^*/^{27}\text{Al} = 5 × 10^{-5}$ (cf. Wasserburg 1985). Two analyses of grain B give large excesses of $^{26}\text{Mg}$ = 11,530 ± 182% and $^{26}\text{Mg}$ = 14,566 ± 356%, corresponding to $^{25}\text{Al}/^{24}\text{Mg}$ ratios of ~12.5 and ~15.5 times the terrestrial value. The $^{25}\text{Al}/^{24}\text{Mg}$ ratios are, respectively, 1840 ± 90 and 2360 ± 120. The extreme enrichment of $^{26}\text{Mg}$ and the normal $^{25}\text{Al}/^{24}\text{Mg}$ ratio most plausibly reflect the in situ decay of $^{26}\text{Al}$ (t = 1.05 × 10$^6$ yr). On an $^{26}\text{Al}/^{24}\text{Mg}$ evolution diagram (Fig. 1), the data from grain B, together with normal Mg at $^{25}\text{Al}/^{24}\text{Mg} = 0$, define a line with slope corresponding to $^{26}\text{Mg}^*/^{27}\text{Al} = (8.9 ± 0.1) × 10^{-4}$. Similar $^{26}\text{Mg}^*/^{27}\text{Al}$ ratios had
previously been observed only in SiC and spherulitic graphite grains whose circumstellar origin is unambiguous (Zinner et al. 1991). These data led Huss et al. (1992) to infer that Orgueil corundum B might be an interstellar oxide grain.

3.2. Oxygen Isotopes

Most oxide grains in the Orgueil residues are enriched in $^{16}\text{O}$ and have oxygen isotope compositions similar to other refractory solar system materials (Clayton et al. 1977). Eight of nine Orgueil spinels, all six hibonites, and 19 of 29 corundums show enrichments in $^{16}\text{O}$ and have oxygen compositions plotting close to the CAI mixing line on an oxygen 3-isotope plot (Fig. 2). Compared to spinels from Allende or corundum from Murchison (Virag et al. 1991), Orgueil corundums exhibit an unusually large range in O composition with $^{17}\text{O}$ values between $-67\%$ and $-55\%$, and corresponding $^{18}\text{O}$ values between $-63\%$ and $-58\%$. This large range notwithstanding, these grains have compositions readily understandable in terms of mixing and mass-dependent fractionation processes in the solar nebula. However, we note that the source of the $^{16}\text{O}$ excesses in refractory solar system material has not been identified (Clayton et al. 1977, 1988). In contrast to the other grains, the oxygen isotope composition of corundum B (Fig. 2) is extremely enriched in $^{17}\text{O}$ with $^{17}\text{O} = 1538\%_{\circ} + 26\%_{\circ}$ ($^{16}\text{O}/^{17}\text{O} = 1028 \pm 11$). Surprisingly, the $^{18}\text{O}$ abundance is normal, $^{18}\text{O} = -6\%_{\circ} \pm 20\%_{\circ}$ ($^{16}\text{O}/^{18}\text{O} = 502 \pm 10$). The $^{17}\text{O}$ enrichment of corundum B contrasts with the $^{18}\text{O}$ enrichment in refractory solar system materials, indicating that this type of Al$_2$O$_3$ grain did not supply $^{16}\text{O}$ to the solar nebula.

4. DISCUSSION

The oxygen isotope composition and the $^{26}\text{Mg}^{*}/^{27}\text{Al}$ ratio of corundum B are extraordinary and unambiguously identify this grain as a circumstellar condensate of a star other than the Sun. This identification is based on the fact that the Al$_2$O$_3$ grain is comprised totally of exotic (non-solar) O and Al with a $^{26}\text{Mg}^{*}/^{27}\text{Al}$ ratio far above the estimated solar system value. The isotopic characteristics of corundum B are similar to those expected in envelopes of AGB stars. Corundum B and a Murchison corundum grain (83-5) with similar characteristics (Nittler et al. 1993) are the first identified oxygen-rich stellar grains and bring to five the types of presolar grains identified as carriers of $^{26}\text{Al}$.

4.1. Stellar Sources

The characteristic signatures identifying corundum grains from the circumstellar objects are large excesses in $^{17}\text{O}$ and $^{26}\text{Mg}^{*}$. The two grains have similar isotopic compositions; grain B has $^{16}\text{O}/^{17}\text{O} = 1028 \pm 11$, $^{16}\text{O}/^{18}\text{O} = 502 \pm 10$, and $^{26}\text{Mg}^{*}/^{27}\text{Al} = (8.9 \pm 0.1) \times 10^{-4}$; corundum 83-5 has $^{16}\text{O}/^{17}\text{O} = 1260 \pm 36$, $^{16}\text{O}/^{18}\text{O} = 660 \pm 21$, and $^{26}\text{Mg}^{*}/^{27}\text{Al} = 8.7 \times 10^{-4}$ (Nittler et al. 1993). The stellar source(s) of the Al$_2$O$_3$ must thus satisfy the following criteria: (1) $^{16}\text{O}/^{17}\text{O} \leq 1000$; (2) $^{16}\text{O}/^{18}\text{O}$ near the solar value ($\sim 500$), (3) $^{26}\text{Al}/^{27}\text{Al}$ ratio $\geq 10^{-3}$, and (4) a circumstellar environment suitable for condensation of refractory oxides.

The $^{26}\text{Mg}^{*}/^{27}\text{Al}$ ratio in corundum B is $\sim 18$ times higher than the $5 \times 10^{-5}$ characteristic of CAI (Wasserburg 1985). Only SiC and graphite grains, whose C and/or Si isotopic compositions suggest they formed in stellar atmospheres, have $^{26}\text{Mg}^{*}/^{27}\text{Al}$ as high as reported here (Zinner et al. 1991; Huss, Hutcheon, & Wasserburg 1993b). The requirement of a high $^{26}\text{Al}/^{27}\text{Al}$ does little to constrain the stellar source since $^{26}\text{Al}$ production occurs in a variety of stellar sites (cf. Forestini, Paulus, & Arnold 1991; Wasserburg et al. 1994). The need for coproduction of $^{26}\text{Al}$ and $^{17}\text{O}$, keeping $^{16}\text{O}/^{18}\text{O} \sim 500$, strongly favors hydrostatic hydrogen burning via the CNO

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cycle (Wannier 1985) as the dominant nucleosynthesis process recorded in the corundum.

Spectroscopic observations of red giant and AGB stars reveal large, but variable enrichments in \(^{17}O\) with \(^{16}O/^{18}O\) ratios near the solar value (Harris et al. 1987; Wannier & Sahai 1987; Harris, Lambert, & Smith 1988; Kahane et al. 1992). In these stars, \(H\)-burning produces \(^{17}O\) and \(^{26}Al\). The stellar atmosphere becomes enriched in \(^{17}O\) and \(^{26}Al\) when the convective envelope extends inward to "dredge up" material that has experienced hydrogen burning during the preceding main-sequence evolution. The initial dredge-up events enhance the surface abundance of \(^{13}C\), \(^{14}N\) and \(^{17}O\), while producing little change in the \(^{16}O\) and \(^{18}O\) abundance. Subsequent mixing events penetrate the \(H\)-burning zone dredging up \(^{16}O\)-enriched material that dilutes the \(^{17}O\) and \(^{18}O\) in the stellar envelope (Harris et al. 1988) and produces atmospheric oxygen isotope compositions similar to Orgueil corundum B and Murchison corundum 83-5 (Fig. 3).

4.2. Relative Abundances of Interstellar Grains

The inventory (by weight) of carbon-rich interstellar grains in Orgueil is 1450 ppm diamond, 14 ppm SiC (all sizes), and \(\sim 6\) ppm graphite (Huss & Lewis 1994); abundances in Murchison are roughly 50\% of those in Orgueil, reflecting the presence of chondrules and CAI in Murchison (e.g., Amari et al. 1993). The abundance of corundum \(>1\) \(\mu\)m in our Orgueil separates is approximately 1/10 that of SiC (5 ppm > 1 \(\mu\)m), or \(\sim 0.5\) ppm \(^{26}Al\) in the 1–8 \(\mu\)m size range. Based on the identification of one interstellar \(^{26}Al\)O\(_3\) grain out of 60 grains examined, we estimate an abundance of presolar corundum larger than \(1\) \(\mu\)m of 0.01 ppm. The disparity in the relative abundances of C-rich and O-rich presolar grains may reflect (1) oxide grains were preferentially destroyed in nature or in the laboratory; (2) oxide grains are typically very small (<0.1 \(\mu\)m) and have escaped detection; or (3) corundum grains were not efficiently produced in the stellar sources compared to graphite and SiC.

 Destruction of interstellar dust grains occurs primarily via sputtering by high-velocity ions and vaporization in grain-grain collisions. Preferential destruction of \(^{26}Al\)O\(_3\) grains would require either a much higher sputtering rate for \(^{26}Al\)O\(_3\) relative to graphite and SiC or a higher rate of chemical reaction in environments experienced by interstellar dust. Given available data, there is no reason to expect \(^{26}Al\)O\(_3\) to sputter more rapidly than SiC. Grains may also be destroyed by chemical reactions in the ISM or solar nebula, but both environments are typically O-rich and likely to be more hostile to SiC and graphite than corundum. Destruction of grains during preparation of the separates cannot be completely excluded, but if the size distributions of \(^{26}Al\)O\(_3\) and SiC are similar, preferential destruction of \(^{26}Al\)O\(_3\) is unlikely.

The C/Al ratio for the Orgueil presolar grain population is \(\sim 5000\) times the solar C/Al ratio. This value is determined by the diamond abundance and the Al content of SiC; corundum is much too rare to affect the total Al concentration. Diamonds, with a nearly solar \(^{12}C/^{13}C\) ratio, must originate from a different stellar source than the majority of the graphite and SiC grains (Anders & Zinner 1993; Ott 1993). If we consider only the carbon in graphite and SiC, the discrepancy between the C/Al ratio in separated presolar dust and the solar value is reduced to a factor of \(\sim 1.7\). This value underestimates the true C/Al ratio in the stellar source since most C in stellar environments is in the form of CO, while all of the Al should be condensed into grains. We note that the fraction of total C condensed as graphite is \(C_{\text{graphite}}/C \approx (C/O - 1)/(C/O)\) for C/O > 1. The ratio of total available Al to graphite is thus \(Al/C_{\text{graphite}} \approx (Al/O)_{\text{gr}}/(C/O - 1) \approx 4 \times 10^{-3}/(C/O - 1)\).

Red giant and AGB stars produce dust in both the O-rich and C-rich phases of their evolution (Gehrz 1989) and are plausible sources of both the SiC and graphite grains found in meteorites (Gallino et al. 1990; Ott 1993). The abundances of several short-lived nuclei found can also be accounted for by contamination of the nascent solar system with \(\sim 1\%\) of net AGB ejecta (Cameron 1993; Wasserburg et al. 1994). Corundum is a stable phase in both O-rich and C-rich stellar atmospheres but there are major differences in the temperature and pressure ranges of \(^{26}Al\)O\(_3\) stability (Lattimer, Schramm, & Grossman 1978). In an O-rich gas of solar composition \(^{26}Al\)O\(_3\) is the most refractory oxide with a condensation temperature of 1740 K. Under the reducing conditions (C/O > 1) required for SiC and graphite formation, \(^{26}Al\)O\(_3\) remains the most refractory oxide but Al condenses primarily as AlN; \(^{26}Al\)O\(_3\) forms by later reaction of AlN with the circumstellar gas below 1200 K (Sharp & Wasserburg 1993; Lodders & Fegley 1993). At the low pressures in an expanding stellar wind (0.1 dyne cm\(^{-2}\)), formation of \(^{26}Al\)O\(_3\) by reaction of AlN is depressed to 900 K. The high concentrations of Al and N (up to 10\%) found in SiC (Huss et al. 1993b; Hoppe et al. 1994) indicate that a substantial portion of these elements condense at much higher temperatures in solid solution in graphite and SiC. The formation and growth of \(^{26}Al\)O\(_3\) grains in an AGB envelope with a C/O ratio of > 1, but otherwise solar abundances, could be severely inhibited if most of the Al is contained within graphite and SiC and thus would be unavailable to react with the gas. The approximately solar C/Al ratio characteristic of the presolar SiC grains suggests that this scenario is a plausible explanation for the low abundance of interstellar \(^{26}Al\)O\(_3\).
4.3. Corundum, SiC, and Graphite as Carriers of $^{26}$Al

Concentrations of $^{26}$Al and other short-lived nuclides in the early solar system require injections of freshly synthesized material within a few million years of the formation of the first solids (cf. Wasserburg 1985). The $^{26}$Al will be accompanied by $^{27}$Al from the same source. For transit times near zero, the $^{26}$Al/$^{27}$Al production ratio, $P_{26}/P_{27} = 10^{-3}$, is equal to the $^{26}$Mg*/$^{27}$Al ratio in the carrier grains. For $P_{26}/P_{27} = 10^{-3}$, the fraction of the solar system $^{27}$Al inventory from the $^{26}$Al stellar source is $X_{27}^A = 5 \times 10^{-2}$, and for $P_{26}/P_{27} = 1$, $X_{27}^A = 5 \times 10^{-2}$. For an $^{26}$Al source to contribute only a small fraction of the total Al inventory, carrier grains derived from that source must have $^{26}$Al/$^{27}$Al ratios approaching unity. If the $^{26}$Al source for the solar system was an AGB star with $C/O \sim 1$ and $^{26}$Al/$^{27}$Al = $10^{-3}$ (but otherwise solar abundances), then $\sim 17\%$ of the total solar system C would have arrived with the Al, unless substantial gas-dust fractionation occurs. Since AGB stars have $^{12}$C/$^{13}$C ratios between $-0.1\%$ and $5 \times 10^{-3}$, the fractions of the total solar system C would greatly perturb the $^{12}$C/$^{13}$C ratio of the protosolar cloud. While we infer corundum B to originate in a red giant or AGB stellar environment, its $^{26}$Mg*/$^{27}$Al is far too low to be compatible with the AGB injection model of Wasserburg et al. (1994) for supplying $^{26}$Al/27Al$_{\odot}$ = $5 \times 10^{-3}$.

Presolar graphite grains have an inferred average $^{26}$Al/27Al$_{\odot}$ = $5 \times 10^{-2}$ (Amari et al. 1993) and contain $\sim 0.1\%$ Al by weight. If this graphite supplied the $^{26}$Al to give a solar system $^{26}$Al/27Al$_{\odot}$, it would also have supplied $0.1\%$ of $^{27}$Al and roughly $25\%$ of the carbon in bulk Orgueil. This is $\sim 2500$ times greater than the observed abundance of presolar graphite. SiC is characterized by a mean $^{26}$Al/$^{27}$Al$_{\odot}$ of $\sim 1 \times 10^{-3}$ and contains $\sim 1\%$ Al. If SiC were the $^{26}$Al carrier, $\sim 42\%$ of the solar system Si must have arrived as SiC, requiring $\sim 10^{4}$ times more SiC than is observed in Orgueil. We conclude that the identified presolar grains cannot have transported sufficient $^{26}$Al to produce an average solar system $^{26}$Al/$^{27}$Al$_{\odot}$ ratio of $5 \times 10^{-5}$. It appears likely that the $^{26}$Al-carrier has not survived due to chemical reactions in the nebula, in the host meteorite, or in the laboratory.

5. SUMMARY

In the Orgueil meteorite we have found a preserved presolar Al$_2$O$_3$ grain composed of oxygen greatly enriched in $^{18}$O, with normal $^{16}$O/$^{18}$O, and containing a large $^{26}$Mg* excess from the decay of $^{26}$Al (initial $^{26}$Al/$^{27}$Al = $8.9 \times 10^{-4}$). This rare grain is the first presolar oxide identified in meteorites. The enrichments in $^{17}$O and $^{26}$Al point to H-burning and an origin as a circumstellar condensate of a red giant or AGB star. Neither this corundum grain, nor any of the other four types of presolar grains thus far identified in meteorites, have the characteristics to be the source of the $^{18}$O or $^{26}$Al enrichments found in refractory solar system materials.

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REFERENCES


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