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Oxygen isotopic compositions of individual minerals in Antarctic micrometeorites: Further links to carbonaceous chondrites

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Abstract—We report in situ measurements of oxygen isotopic abundances in individual silicate and oxide minerals from 16 Antarctic micrometeorites (AMMs). The oxygen isotopic compositions of 10 olivine and 11 pyroxene grains are enriched in ^{16}O relative to terrestrial minerals, and on an oxygen three-isotope diagram they plot on the low $\delta^{18}\text{O}$ side of the ^{16}O mixing line defined by calcium-aluminum-rich inclusions (CAI) from chondritic meteorites. AMM olivine and pyroxene $\delta^{18}\text{O}$ values range from -9.9% to $+8.0\%$ and $\delta^{17}\text{O}$ ranges from -11.3% to $+5.5\%$, similar to values measured in individual olivine grains and whole chondrules from carbonaceous chondrites. These data indicate that the mineral grains preserve their pre-terrestrial oxygen isotopic compositions, and provide another link between AMMs and carbonaceous chondrites. However, no clear relationship with one single subgroup of carbonaceous chondrite can be established. Based on their textures, crystal chemistries, and oxygen isotopes, some coarse-grained crystalline AMMs could originate from chondrule fragmentation. Whether the remaining mineral grains were formed by igneous or condensation processes is unclear. No clear correlation is observed between isotopic compositions and mineral compositions of AMM olivine grains, suggesting that the FeO- and ^{16}O -enrichment processes are not coupled in a simple way. Nor are any relatively large ^{16}O enrichments measured in any of the olivine grains, however two Mg-Al spinels and a melilite grain are ^{16}O enriched at the level of $\delta^{18}\text{O} \sim \delta^{17}\text{O} \sim -40\%$. The discovery of an ^{16}O -enriched melilite grain in AMMs supports the hypothesis that refractory minerals throughout the solar nebula formed from a relatively uniformly ^{16}O -enriched reservoir. This unique ^{16}O -rich signature of refractory minerals in primitive solar system materials suggests that they either formed from a widespread ^{16}O -rich reservoir in the solar nebula, or that an efficient mechanism (such as bipolar outflows) was acting to spread them from a highly localized ^{16}O -rich region over the early solar nebula. Copyright © 1999 Elsevier Science Ltd

1. INTRODUCTION

The majority of the mass of extraterrestrial matter presently accreted by Earth is comprised of particles in the 50–400 μm size range (Love and Brownlee, 1993). Much of this material is heated during passage through the atmosphere resulting in partial to total melting, but some particles with low enough velocity and/or shallow entry angles survive as unmelted micrometeorites. These particles may be collected from Antarctic blue ice which contains very low levels of terrestrial dust (Maurette et al., 1991, 1994). Many of these Antarctic micrometeorites (AMMs) consist of unequilibrated mixtures of fine-grained anhydrous minerals, hydrated silicates, and carbonaceous matter, and display chemical and mineralogical affinities to CI, CM and CR carbonaceous chondrites (Engrand and Maurette, 1998; Kurat et al., 1994b). However, differences between AMMs and CI/CM/CR chondrites in terms of relative abundances of major silicate minerals, range of mineral chemistries (e.g., fayalite contents of olivine), bulk C/O ratios, and the extremely low abundance of chondrules in AMMs clearly indicate that they sample a different type of extraterrestrial

matter than that represented in conventional meteorite collections (Engrand and Maurette, 1998). This may reflect biases in the macroscopic meteorite collection, such as atmospheric entry selection effects and/or the predominance of samples from a limited number of asteroidal parent bodies (Meibom and Clark, 1999), that are not as prevalent among the micrometeorite population. Thus, it is of considerable interest to understand the intrinsic properties of micrometeorites which may be more representative of small solar system bodies than are common meteorite types (e.g., ordinary chondrites).

Analyses of whole-rock meteorites have demonstrated that oxygen isotopic abundances are heterogeneously distributed on a planetary scale within the inner solar system and have led to the use of oxygen isotopic composition as an essential parameter in the classification of meteorites (Clayton, 1993 and references therein). The distinctive oxygen isotopic compositions of meteorites within a group is thought to reflect their common origin from a singular parent body or, perhaps, a limited number of related asteroids (e.g., in the case of the carbonaceous chondrites). How micrometeorites fit into this classification scheme based on oxygen isotopic composition is not known.

Isotopic studies of micrometeorites are hampered by their small sizes and by potential problems of contamination or isotopic exchange during atmospheric entry and prolonged terrestrial residence times. Some isotopic data exist for hydrogen (Engrand et al., in press), carbon (Wright et al., 1997), and neon (Olinger, 1990; Olinger et al., 1990) in individual AMMs.

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Oxygen isotope compositions have been reported only for suites of cosmic spherules collected from deep sea sediments (Clayton et al., 1986), for 2 olivines from Greenland micrometeorites (Christophe Michel-Levy and Bourot-Denise, 1992), and for refractory minerals (spinel and pyroxene) from 4 AMMs (Greshake et al., 1996; Hoppe et al., 1995; Kurat et al., 1994a). These limited data do not permit definitive comparisons to meteorite compositional groups because either the indigenous oxygen in the micrometeorites has partially exchanged with terrestrial oxygen reservoirs (as is the case for the cosmic deep sea spherules), or because refractory minerals exhibit ^{16}O -rich compositions regardless of meteorite group (e.g., McKeegan et al., 1998b).

We report in situ ion microprobe analyses of the oxygen isotopic compositions of single olivine and pyroxene grains from 15 AMMs, and of refractory minerals from 2 AMMs. Because these minerals are thought to be relict (i.e., they did not form during atmospheric entry), they are expected to preserve their pre-terrestrial oxygen isotopic compositions. The oxygen isotopic analysis of common mafic silicates allows a more direct comparison to similar mineral phases in chondrites for which a relatively large body of data exist (Choi et al., 1997b; Hervig and Steele, 1992; Jones et al., 1998; Leshin et al., 1998; Leshin et al., 1997; Saxton et al., 1998; Sears et al., 1998; Weinbruch et al., 1993). The primary goals of this work are to better constrain the relationships of AMMs to conventional meteorite groups and to attempt to shed light on the origins of anhydrous mineral grains in AMMs. To the extent that AMMs provide a more complete sampling of primitive extraterrestrial materials, these data may also assist in better defining oxygen isotopic reservoirs in the solar nebula.

2. SAMPLES AND METHODS

Individual AMMs containing mineral grains large enough for in situ ion microprobe analysis of their oxygen isotopic compositions were selected from the 50–100 μm and 100–400 μm size fractions of micrometeorites harvested from the blue ice fields of Cap-Prudhomme, Antarctica, (see Maurette et al., 1994 for details regarding sample collection). A total of 10 olivine and 11 pyroxene grains, ranging in size from about 10–30 μm , were identified from among 15 AMMs. One melilite and one spinel grain (~8–10 μm in diameter) were located within an additional micrometeorite, and a small spinel was also documented within one of the olivine and pyroxene-bearing AMMs. All AMMs were mounted in epoxy and polished to expose the sample interiors (see Figs. 1, 2 and 3 for electron micrographs of typical examples of these AMMs).

The major and minor element compositions of all minerals were determined prior to ion microprobe isotopic analysis by using the UCLA CAMECA Camebax microbeam electron microprobe or the Caltech JEOL JXA-733 Superprobe with a sample current of 10 nA at 15 kV.

The oxygen isotopic compositions of the selected minerals were measured in situ within the AMM polished sections by using the UCLA CAMECA ims 1270 ion microprobe. Samples were coated with ~350 Å of gold. The ~0.1–0.4 nA primary Cs^+ beam was shaped into a spot ~8–10 μm in diameter (usually in aperture illumination mode). Charge compensation was achieved using a normal-incidence electron flood gun (Slodzian, 1980). Low energy (~0–30 eV) negative secondary ions were measured at high-mass resolving power ($M/\Delta M$) of about 6500. At this mass resolution, the correction for the contribution of the tail of the $^{16}\text{OH}^-$ peak to the $^{17}\text{O}^-$ signal was typically less than 0.1%. Measurements were made by magnetic peak switching through 25 cycles of counting $^{16}\text{O}^-$ for 3s, $^{17}\text{O}^-$ for 10s and $^{18}\text{O}^-$ for 5s. The secondary $^{16}\text{O}^-$ current was measured in a Faraday cup equipped with a Keithley 642 electrometer, and the $^{17}\text{O}^-$ and $^{18}\text{O}^-$ signals were

pulse-counted on an electron multiplier with a deadtime of typically 20 ns. The analysis conditions typically yielded equivalent count rates of 20–40 million counts per second of $^{16}\text{O}^-$. Measured secondary ion intensities were corrected for background ($^{16}\text{O}^-$), and for deadtime ($^{17}\text{O}^-$ and $^{18}\text{O}^-$). With the count rates employed in these experiments, the total magnitude of these corrections is small (<2%), and thus the additional uncertainties resulting from background and deadtime corrections are negligible (~0.1%).

San Carlos olivine was used as a standard ($\delta^{18}\text{O} = +5.25\%$; Eiler et al., 1995) to correct for instrumental mass fractionation. The olivine data have been corrected for matrix effects, i.e., the change of the instrumental mass fractionation as a function of the chemical composition of the sample. Based on previous analyses of olivine standards ranging in composition from Fa_9^1 to Fa_{100} the magnitude of matrix effect corrections was determined to be ~+0.5%/amu per increase of Fa_{10} for the analyzed olivine as compared with the standard (San Carlos olivine, $\text{Fa}_{9.2}$). Such corrections for the AMM olivine grains were on the order of 1%, and thus slight inaccuracies in this correction do not affect the interpretation of the data. No matrix effect corrections were made for the other mineral (pyroxene, melilite, spinel) data, because analyses of enstatite, melilite, and spinel standards all show relative variations of instrumental mass fractionation compared to San Carlos olivine of <1%/amu under our experimental conditions. The reported (1σ) uncertainties reflect the internal measurement precision quadratically summed with the standard deviation of analyses on the primary standard during the analytical session (which represents the uncertainty in the instrumental mass fractionation correction and in the relative collection efficiency of the two detectors). Under our experimental conditions, the precision and accuracy of the analysis is typically 1 to 1.5% in both $\delta^{18}\text{O}$ and $\delta^{17}\text{O}$.

3. RESULTS

3.1. Petrography and Mineral Chemistry

The olivine and pyroxene grains studied here occur in two different textural settings: as coarse-grained crystalline AMMs (Fig. 1) and as isolated mineral grains in a fine-grained matrix (Fig. 2). We analyzed olivine and pyroxene grains in 4 coarse-grained crystalline AMMs (# 92-13C-13, -20, -26; # 94-4B-38; Fig. 1), and in 11 fine-grained AMMs which range in texture from unmelted to scoriaceous (see Fig. 2). The minerals are euhedral to rounded and typically range from 5–50 μm in size (Figs. 1 & 2).

Major and minor element abundances in individual olivine and pyroxene grains from 15 AMMs are reported in Table 1. The pyroxene grains analyzed are all Ca-poor and have compositions $\text{Fs}_{1.3-11} \text{Wo}_{0.6-3}$. Olivine grains range in composition between Fa_1 and Fa_{34} . These compositions are comparable to the range previously reported for AMM olivine and pyroxene grains (Beckerling and Bishoff, 1995; Christophe Michel-Levy and Bourot-Denise, 1992; Kurat et al., 1994b; Presper, 1993; Steele, 1992).

Refractory minerals analyzed in this study include two spinel grains and the first melilite mineral discovered in the AMM collection (see Fig. 3). The spinel grains are nearly pure MgAl_2O_4 and the melilite has an intermediate composition, Åk_{22} (Table 1). End-member Mg-Al spinel grains are common in refractory inclusions (CAIs) from carbonaceous chondrites, as are melilite grains with Al-contents comparable with that of the melilite in AMM # 92-13C-23 (MacPherson et al., 1988).

¹ The Fa_x notation is used to represent the fayalite component of the olivine, where $x = 100 \cdot \text{Fe}/(\text{Fe} + \text{Mg})$ with cation concentrations expressed as mole fractions

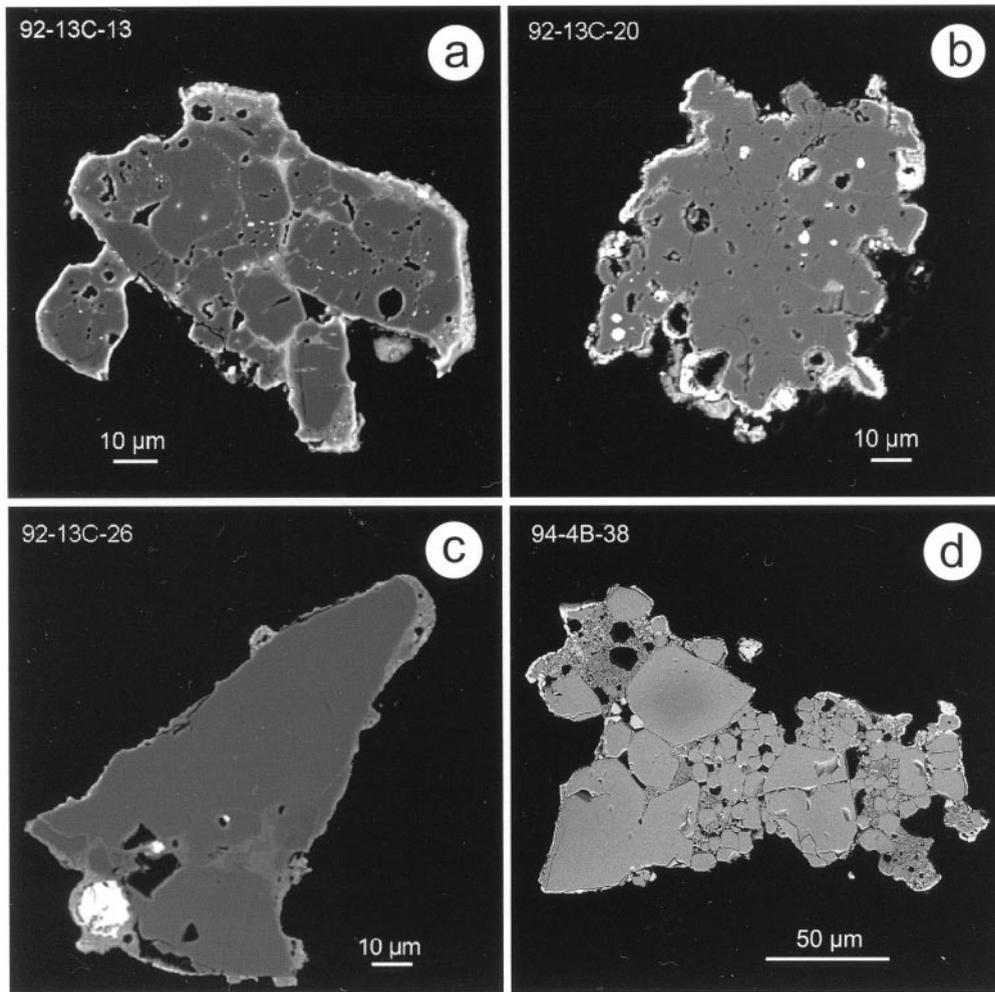


Fig. 1. Backscattered electron micrographs of coarse-grained crystalline Antarctic micrometeorites consisting of assemblages of olivine and pyroxene grains with minor amounts of fine-grained matrix.

3.2. Oxygen Isotopes

The oxygen isotopic compositions of olivine, pyroxene, spinel, and melilite grains from 16 AMMs are reported in Table 2 and shown on a three-isotope diagram in Fig. 4a.

For the 10 olivine grains (Fig. 4b), $\delta^{18}\text{O}$ values range from -8.3‰ to $+8.0\text{‰}$ and $\delta^{17}\text{O}$ from -7.4‰ to $+5.5\text{‰}$. The $\delta^{18}\text{O}$ values for the pyroxenes (Fig. 4b) range from -9.9‰ to $+2.2\text{‰}$, and $\delta^{17}\text{O}$ from -11.3‰ to $+0.4\text{‰}$.

The AMM olivine and pyroxene oxygen isotopic compositions plot parallel to, but slightly above, the CAI line and mostly below the terrestrial fractionation (TF) line in the oxygen three-isotope plot. The displacement from the terrestrial fractionation line, expressed as $\Delta^{17}\text{O}$ values, are typically in the range of -2 to -4‰ for the olivine and pyroxene grains. As a group, the AMM olivine grains have oxygen isotopic compositions which are slightly poorer in ^{16}O than those of the pyroxene grains. No correlation between the oxygen isotopic composition and the apparent size of the host micrometeorite or the mineral grain size is observed.

In 4 AMMs, individual mineral grains were large enough to

permit multiple analyses of the same crystal (1 olivine, in AMM #92-13C-26, and 3 pyroxenes, in AMMs #92-13C-4, -15, -20). No significant ($>2\sigma$) heterogeneity is observed within an individual grain. However, the isotopic compositions of both olivine and pyroxene grains do vary from one AMM to another, and within two AMMs there is slight intergrain heterogeneity between olivine and pyroxene grains of different FeO contents (AMM #94-4b-5) or between olivine grains of different FeO content (AMM #94-4b-38).

A single analyzed melilite (Åk_{22}) grain exhibits a large ^{16}O enrichment with $\delta^{18}\text{O} = -42.2\text{‰}$ and $\delta^{17}\text{O} = -43.8\text{‰}$. Spinel grains, of nearly pure MgAl_2O_4 , were analyzed in 2 AMMs. The analysis of spinel in AMM # 94-4b-5 yielded only a moderate ^{16}O enrichment ($\delta^{18}\text{O} = -21\text{‰}$ and $\delta^{17}\text{O} = -27\text{‰}$) probably due to its small size ($<10\text{ }\mu\text{m}$) which resulted in partial overlap of the ion beam onto adjacent fine-grained matrix material. In particle # 92-13C-23, multiple analyses of the spinel yielded a range of ^{16}O -enrichments between approximately -40‰ and -48‰ with an average composition of $\delta^{18}\text{O} = -42\text{‰}$ and $\delta^{17}\text{O} = -43\text{‰}$. It is likely that the range of values observed in the spinel from #92-13C-23 also

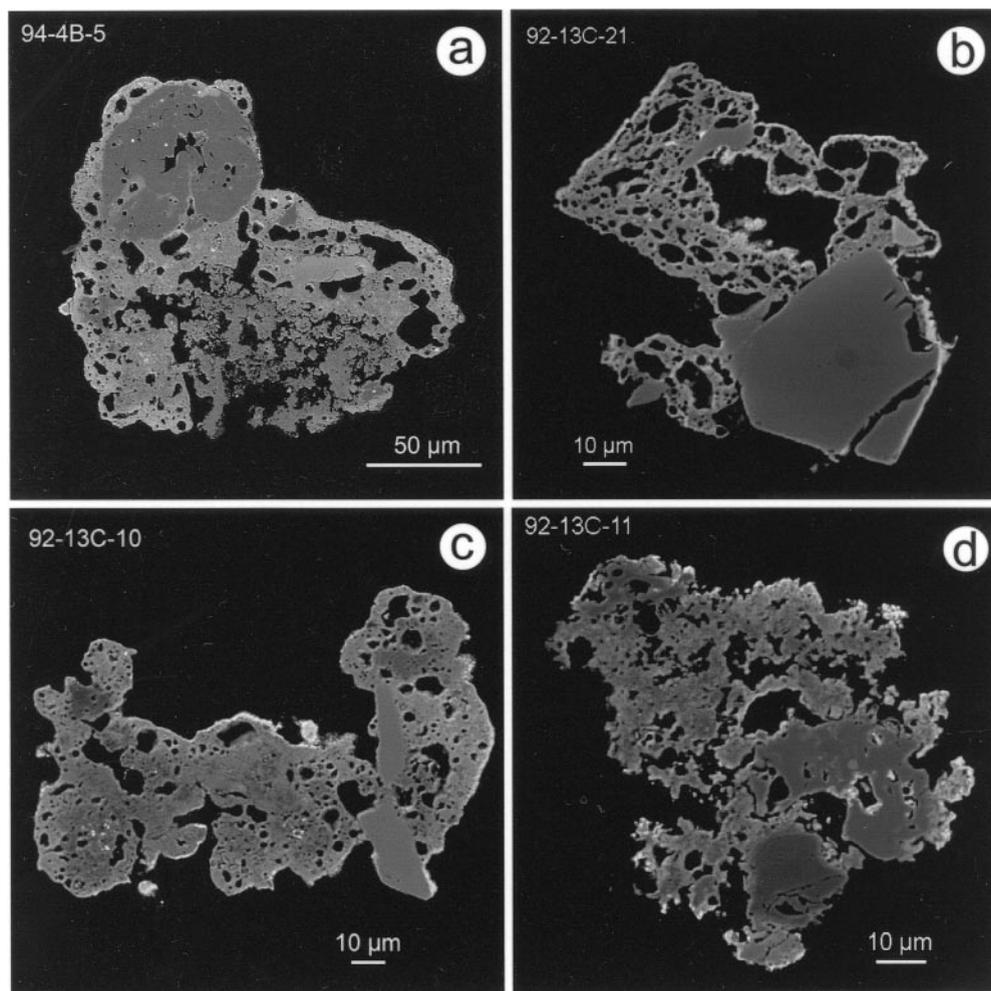


Fig. 2. Backscattered electron micrographs of Antarctic micrometeorites containing individual minerals of olivine (a,b,c) and pyroxene (a,d). The mineral grains are embedded in unmelted to partially-melted (scoriaceous) matrix. The rounded crystalline structure in Fig. 2a consists of an assemblage of coarse Mg-rich olivine and pyroxene grains. An additional FeO-rich olivine grain (Fa_{32}), that is located within the fine-grained matrix of this AMM, was analyzed for oxygen isotopic composition. See Engrand and Maurette (1998) and Kurat et al. (1994b) for the definition of the textural classification of micrometeorites.

reflects minor ion beam mixing rather than true heterogeneity within this single spinel grain.

4. DISCUSSION

We have analyzed the oxygen isotopic compositions of individual mineral grains from 16 AMMs. Based on their textures and chemical compositions, these minerals have been interpreted to be relict and unaltered by atmospheric entry heating. The observed isotopic homogeneity within single AMM mineral grains, as well as the lack of a trend toward isotopically heavy (i.e., ^{18}O and ^{17}O -enriched) oxygen as observed in clearly melted cosmic spheres (Engrand et al., 1998), argues strongly against mass fractionation or isotopic exchange with terrestrial oxygen during atmospheric entry. Thus, these data represent the pre-terrestrial oxygen isotopic composition of the grains and allow a direct comparison to data for similar phases in chondrites.

4.1. Olivine and Pyroxene Data: Comparison to Meteorites and Interplanetary Dust Particles

Chondritic meteorites are unequilibrated with respect to oxygen isotopic composition on size scales ranging from whole-rock (Clayton, 1993) to discrete, mm-sized nebular constituents such as chondrules and inclusions (e.g., Clayton and Mayeda, 1983; Clayton et al., 1991; Clayton et al., 1977) to, in some cases, μm scale, sub-mineralic domains (e.g., McKeegan et al., 1996). The ordinary chondrites are characterized by having whole-rock oxygen isotopic compositions that plot above the terrestrial mass-dependent fractionation line (TF line), while almost all carbonaceous chondrites are more ^{16}O -enriched than ordinary chondrite or terrestrial samples, and thus plot below the TF line (Clayton, 1993 and references therein). The main exception to this general rule are the highly aqueously altered CI chondrites, which have bulk isotopic compositions which plot close to but slightly above the TF line

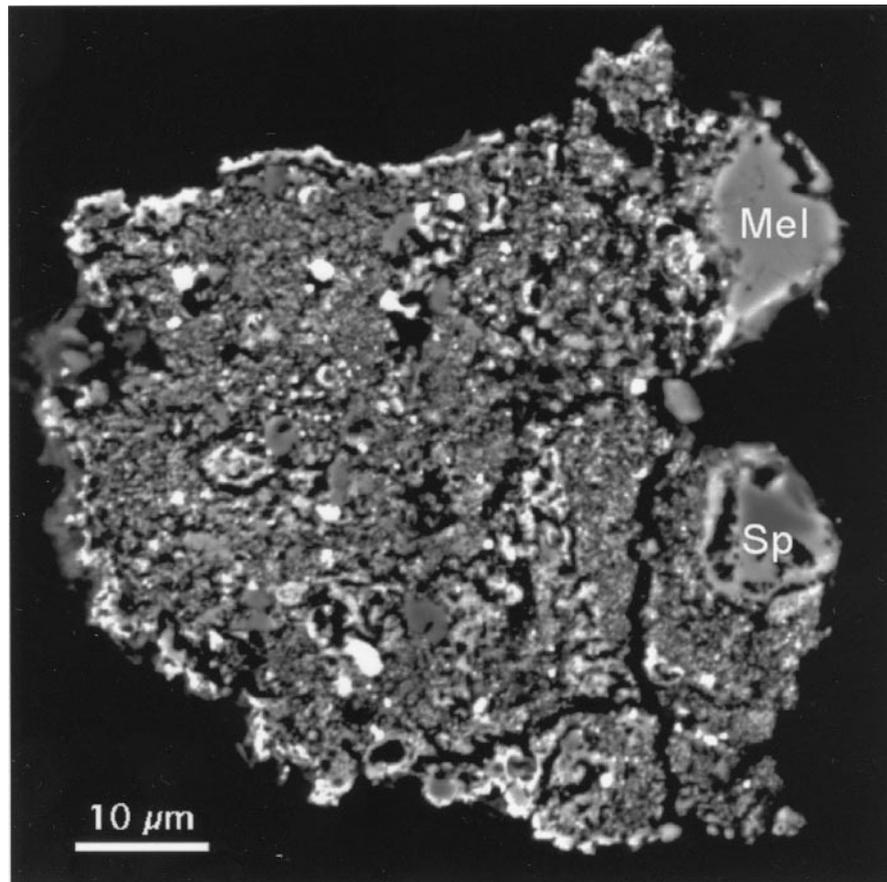


Fig. 3. Backscattered electron micrographs of a fine-grained Antarctic micrometeorite containing a spinel and a mellilite (Åk_{22}) grain.

(Clayton and Mayeda, 1984; Rowe et al., 1994). Recent ion microprobe studies have demonstrated that oxygen isotopic heterogeneities are even more extensive than previously indicated, further complicating this picture. For example, ^{16}O -enriched material has been identified in ordinary chondrites (McKeegan et al., 1998b; Russell et al., 1997; Saxton et al., 1995; Saxton et al., 1996). However, these are minor components of ordinary chondrites (such as refractory inclusions) and the majority of mafic silicates in these meteorites, e.g., the olivine and pyroxene in chondrules, are characterized by oxygen isotopic compositions that lie above the TF line by a few permil (Ash et al., 1998; Clayton and Mayeda, 1983; Clayton et al., 1991). In addition, other ion microprobe data have confirmed that the anhydrous precursor (olivine and pyroxene) minerals in CI carbonaceous chondrites are ^{16}O -enriched and are likely to have been derived from the breakup of chondrules (Leshin et al., 1997). Thus, although some mixing has undoubtedly occurred, to first order the oxygen isotope data for whole-rock chondrites and their chondrules indicate that the nebular accretion regions of the ordinary and carbonaceous chondrites were distinct with respect to oxygen isotopic composition. It is therefore plausible that the oxygen isotopic signatures of the same minerals from AMMs could provide a distinctive link to one of these classes of meteorites.

Our data on oxygen isotopes in AMM olivine and pyroxene grains plot below the TF line with $\Delta^{17}\text{O}$ values typically in the

range of -2 to -4% . Such compositions are distinct from those of olivines measured as single mineral grains (Choi et al., 1998; Sears et al., 1998) or from whole chondrule data (Clayton et al., 1991) in ordinary chondrites, but are compatible with those measured in anhydrous silicate minerals from carbonaceous meteorites (Choi et al., 1997b; Clayton, 1993 and references therein; Clayton and Mayeda, 1983; Clayton et al., 1991; Hervig and Steele, 1992; Jones et al., 1998; Leshin et al., 1998; Leshin et al., 1997; Weinbruch et al., 1993). Thus, these data provide a further link between AMMs and carbonaceous chondrites. However, within the carbonaceous chondrites significant overlap exists in isotopic compositions reported for individual silicate (mostly olivine) mineral grains (CV: Choi et al., 1997b; Hervig and Steele, 1992; Leshin et al., 1998; Weinbruch et al., 1993), (CR: Leshin et al., 1998), (CO: Jones et al., 1998), (CI: Leshin et al., 1997). In addition, our data demonstrate that there is considerable spread within the AMM population (Fig. 4a) which overlaps much of the range of data reported from carbonaceous chondrite chondrules (Fig. 4c). Thus, oxygen isotope data of olivine and pyroxene minerals are not sufficiently diagnostic at the individual grain level to discriminate amongst different groups of carbonaceous chondrites, and our data cannot assign a definite affiliation of AMMs with a single class of carbonaceous chondrite.

In chondritic meteorites, coarse-grained olivine and pyroxene minerals occur predominantly in chondrules and CAIs, or,

Table 1. Electron microprobe analyses of AMM olivine, pyroxene, spinel and melilite grains analyzed for their oxygen isotopic composition.

Sample designation	AMM size fraction (μm)	Mineral	Mineral composition	MgO	Al ₂ O ₃	SiO ₂	CaO	TiO ₂	Cr ₂ O ₃	MnO	FeO	NiO	Total
92-13C-4	50-100	Px	Fs _{1.3} Wo _{1.1}	38.5	1.28	60.3	0.58	n.d.	0.68	0.14	0.8	0.01	102.31
92-13C-5	50-100	Px	Fs _{2.3} Wo _{1.2}	38.4	1.10	59.1	0.64	n.d.	0.50	0.06	1.6	0.03	101.38
92-13C-10	50-100	Ol	Fa _{33.8}	32.0	0.05	37.4	0.17	n.d.	0.51	0.50	29.1	0.16	99.94
92-13C-11	50-100	Px	Fs _{2.0} Wo _{0.8}	38.9	0.96	60.2	0.44	n.d.	0.65	0.09	1.4	0.02	102.62
92-13C-11	50-100	Px	Fs _{8.2} Wo _{2.9}	34.0	1.84	57.5	1.52	n.d.	0.98	0.52	5.0	0.08	101.46
92-13C-13	50-100	Ol	Fa _{2.2}	54.4	0.21	43.7	0.20	n.d.	0.61	0.33	2.2	0.11	101.90
92-13C-13	50-100	Px	Fs _{1.6} Wo _{1.1}	37.8	0.85	59.9	0.59	n.d.	0.81	0.00	1.1	0.05	101.20
92-13C-15	50-100	Px	Fs _{5.3} Wo _{0.9}	37.2	0.43	59.1	0.47	n.d.	1.24	0.32	3.4	0.00	102.12
92-13C-20	50-100	Px	Fs _{3.5} Wo _{1.2}	37.9	0.68	59.5	0.68	n.d.	0.75	0.20	2.3	0.07	102.02
92-13C-21	50-100	Ol	Fa _{28.7}	35.8	0.05	38.0	0.19	n.d.	0.53	0.31	25.6	0.07	100.54
92-13C-23	50-100	Mel	Åk ₂₂	3.6	28.33	26.3	40.5	n.d.	0.03	0.02	0.25	0.00	99.02
92-13C-23	50-100	Sp	Sp	28.8	70.27	0.3	0.52	n.d.	0.38	0.00	0.20	0.08	100.50
92-13C-26	50-100	Ol	Fa _{3.8}	53.8	0.02	42.6	0.21	n.d.	0.62	0.29	3.8	0.12	101.53
92-13C-33	50-100	Px	Fs _{4.4} Wo _{1.1}	35.9	0.91	57.7	0.58	n.d.	0.62	0.11	2.9	0.00	98.81
92-13C-35	50-100	Px	Fs _{1.3} Wo _{0.6}	39.7	0.37	60.7	0.36	n.d.	0.49	0.07	0.9	0.00	102.52
94-4b-2	100-400	Ol	Fa _{2.6}	54.7	0.09	42.2	0.13	0.02	0.47	0.29	2.6	0.00	100.46
94-4b-5	100-400	Ol	Fa _{1.1}	54.8	0.02	41.7	0.21	0.02	0.48	0.20	1.1	0.01	98.59
94-4b-5	100-400	Ol	Fa _{32.4}	33.0	0.02	37.2	0.25	0.00	0.43	0.29	28.1	0.00	99.38
94-4b-5	100-400	Px	Fs _{2.6} Wo _{1.2}	37.1	0.75	57.3	0.64	0.12	0.57	0.17	1.6	0.03	98.28
94-4b-5	100-400	Sp	Sp ₉₉	27.6	68.09	0.5	0.05	0.11	0.11	0.00	1.23	0.01	97.68
94-4b-32	100-400	Ol	Fa _{8.9}	45.5	0.01	39.9	0.18	0.03	0.92	4.51	7.9	0.04	99.00
94-4b-32	100-400	Px	Fs _{10.9} Wo _{0.8}	33.6	0.29	55.9	0.41	0.03	0.79	0.42	6.9	0.00	98.38
94-4b-38	100-400	Ol	Fa _{32.2}	33.0	0.02	36.4	0.24	0.00	0.40	0.77	27.9	0.00	98.75
94-4b-38	100-400	Ol	Fa _{22.6}	39.2	0.01	37.6	0.19	0.00	0.42	0.29	20.4	0.06	98.19

n.d.: not determined.

Note: Na₂O and K₂O were analyzed but omitted in this table because they were always found to be below detection limit.

to a lesser degree, within rims surrounding chondrules and CAIs and as isolated grains in matrix. In AMMs, on the other hand, chondrules are known to be rare (Engrand and Maurette, 1998; Walter et al., 1995). Although most of the olivine and pyroxene analyzed here are petrographically “isolated” grains, the possibility that they represent fragments of disrupted chondrules or refractory inclusions should be considered.

Recent oxygen isotopic analyses of meteoritic olivine grains petrographically associated with CAIs, occurring either within olivine-rich refractory inclusions in CM Murchison (Hiyagon and Hashimoto, 1999) or in the fine-grained accretionary rims of CAIs from CV meteorites (Hiyagon, 1998; McKeegan et al., 1998a), showed large enrichments in ¹⁶O to approximately the same level as the most ¹⁶O-enriched refractory minerals in CAIs ($\delta^{18}\text{O} \approx \delta^{17}\text{O} \approx -40$ to -50‰). Although forsteritic, these olivines are not known to be especially chemically distinctive. So far, such isotopically anomalous olivine grains have not been found as isolated crystals (in matrix) or within chondrules. We have not found any such highly ¹⁶O-enriched olivines in our selection of AMM grains, including particle #94-4B-5, which contains spinel in addition to olivine.

Most chondrule olivine and pyroxene grains result from igneous crystallization from the chondrule melt, with the exception of relict minerals that may be in chemical and isotopic disequilibrium with phenocrysts. However, the origin of the isolated olivine grains in meteorites is controversial. An origin from direct nebular condensation for some of the grains (particularly the forsterite enriched in refractory elements such as Ca, Ti, Al; Steele, 1986) is supported by several authors (Olsen and Grossman, 1978; Steele, 1986, 1988, 1989, 1992; Weinbruch et al., 1993, 1997). Other researchers favor an origin of isolated grains as fragments of disaggregated chondrules

(Desnoyers, 1980; Jones, 1992; Jones and Danielson, 1997; McSween, 1977; Richardson and McSween, 1978). This latter viewpoint is mostly based on textural arguments and chemical characteristics of the olivines, in particular their CaO vs. FeO content (Beckerling and Bishoff, 1995; McSween, 1985; Scott and Taylor, 1983). Recent ion microprobe analyses have demonstrated that oxygen isotopic signatures of isolated olivine grains, even the most refractory forsteritic olivine that might be of nebular condensate origin, are not necessarily distinct from some chondrule olivine (Leshin et al., 1998).

Whether the olivine and pyroxene grains in AMMs formed by igneous crystallization (in chondrule melt) or as direct nebular condensates remains an open question. Five of the AMMs we have examined (#92-13C-13, #92-13C-20, #92-13C-26, #94-4B-5, #94-4B-38) contain coarse-grained aggregates of olivine and/or pyroxene crystals (Figs. 1 & 2a). The textures of these aggregates are consistent with origin of the minerals within a chondrule and their chemistry is plausibly compatible with crystallization of the grains from a single silicate melt composition. With a single exception (AMM #94-4B-38), the olivine and pyroxene minerals in these AMMs within a given mineral aggregate are in equilibrium with regard to their oxygen isotopic composition at the 2σ level (Table 2), consistent with (but not requiring) formation by crystallization from a single chondrule melt. The twelve other mineral grains we analyzed (see Figs. 2b,c,d) are petrographically similar to isolated minerals in carbonaceous chondrites in that they occur as single mineral grains within a fine-grained matrix, although the olivines in AMMs tend to be of smaller size than those studied in carbonaceous chondrites (Steele, 1988). The oxygen isotopic compositions of these grains are also compatible with those measured in carbonaceous chondrite whole chondrules

Table 2. Oxygen isotopic compositions of individual minerals in AMMs.

Sample designation	Mineral	Mineral composition	$\delta^{18}\text{O} \pm 1\sigma$ (‰) [†]	$\delta^{17}\text{O} \pm 1\sigma$ (‰) [†]	$\Delta^{17}\text{O}$ (‰) ^{†,‡}
92-13C-4	Px	Fs _{1.3} Wo _{1.1}	-1.5 ± 1.2	-3.6 ± 1.0	-2.8
92-13C-4	Px	Fs _{1.3} Wo _{1.1}	0.8 ± 1.3	-4.0 ± 1.0	-4.4
92-13C-5	Px	Fs _{2.3} Wo _{1.2}	-4.5 ± 1.6	-7.0 ± 0.8	-4.7
92-13C-10	Ol	Fa _{33.8}	4.3 ± 1.3	0.6 ± 0.8	-1.6
92-13C-11	Px	Fs _{2.0} Wo _{0.8}	-6.0 ± 1.1	-5.3 ± 0.7	-2.2
92-13C-11	Px	Fs _{8.2} Wo _{2.9}	-3.8 ± 1.3	-2.6 ± 0.8	-0.6
92-13C-13	Ol	Fa _{2.2}	-1.3 ± 1.3	-2.8 ± 1.0	-2.1
92-13C-13	Px	Fs _{1.6} Wo _{1.1}	2.2 ± 1.3	-0.4 ± 0.9	-1.5
92-13C-15	Px	Fs _{5.3} Wo _{0.9}	2.1 ± 1.2	0.0 ± 1.0	-1.1
92-13C-15	Px	Fs _{5.3} Wo _{0.9}	-2.0 ± 1.3	-2.9 ± 1.0	-1.9
92-13C-20	Px	Fs _{3.5} Wo _{1.2}	-5.4 ± 1.4	-7.4 ± 1.4	-4.6
92-13C-20	Px	Fs _{3.5} Wo _{1.2}	-5.5 ± 1.2	-6.9 ± 1.0	-4.0
92-13C-21	Ol	Fa _{28.7}	1.0 ± 1.5	-2.1 ± 1.3	-2.6
92-13C-23	Mel	Äk ₂₂	-39.8 ± 1.3	-41.7 ± 1.1	-21.0
92-13C-23	Mel	Äk ₂₂	-44.6 ± 1.3	-45.1 ± 0.9	-21.9
92-13C-23	Sp	Sp ₁₀₀	-40.3 ± 1.4	-41.7 ± 1.1	-20.7
92-13C-23	Sp	Sp ₁₀₀	-48.4 ± 2.4	-47.2 ± 1.6	-22.0
92-13C-23	Sp	Sp ₁₀₀	-42.1 ± 1.6	-41.2 ± 1.2	-19.3
92-13C-26	Ol	Fa _{3.8}	1.9 ± 1.5	-1.4 ± 1.5	-2.4
92-13C-26	Ol	Fa _{3.8}	1.7 ± 1.8	-1.9 ± 1.4	-2.8
92-13C-33	Px	Fs _{4.4} Wo _{1.1}	0.6 ± 1.1	-3.2 ± 1.0	-3.5
92-13C-35	Px	Fs _{1.3} Wo _{0.6}	-3.4 ± 1.5	-6.9 ± 0.9	-5.1
94-4b-2	Ol	Fa _{2.6}	-4.6 ± 1.3	-6.1 ± 1.8	-3.7
94-4b-5	Ol	Fa _{1.1}	-8.3 ± 1.3	-7.4 ± 1.4	-3.1
94-4b-5	Ol	Fa _{32.4}	-3.6 ± 1.5	-6.5 ± 1.6	-4.6
94-4b-5	Px	Fs _{2.6} Wo _{1.2}	-9.9 ± 1.5	-11.3 ± 1.4	-6.2
94-4b-5	Sp	Sp ₉₉ + matrix	-21.1 ± 1.3	-26.8 ± 1.4	-15.8
94-4b-32	Ol	Fa _{8.9}	4.8 ± 1.3	3.4 ± 1.5	0.9
94-4b-32	Px	Fs _{10.9} Wo _{0.8}	1.3 ± 1.3	0.4 ± 1.3	-0.3
94-4b-38	Ol	Fa _{32.2}	8.0 ± 1.2	5.5 ± 1.4	1.3
94-4b-38	Ol	Fa _{22.6}	1.7 ± 1.2	-0.1 ± 1.4	-1.0

[†] Relative to SMOW

[‡] $\Delta^{17}\text{O} = \delta^{17}\text{O} - 0.52 \times \delta^{18}\text{O}$

(Clayton and Mayeda, 1983; Clayton et al., 1977; McSween, 1985; Rowe et al., 1994; Rubin et al., 1990; Weisberg et al., 1993). However, oxygen isotopic composition by itself may not be very diagnostic of mode of origin for either individual olivine grains in carbonaceous meteorites or for AMM olivines.

On the CaO vs. FeO diagram (Fig. 5), the AMM olivine grains analyzed in this study plot within the fields defined by carbonaceous chondrite olivines, including those from both type I and type II chondrules. Additionally, their oxygen isotopic compositions are compatible with those measured in carbonaceous chondrite whole chondrules (Clayton and Mayeda, 1983; Clayton et al., 1977; McSween, 1985; Rowe et al., 1994; Rubin et al., 1990; Weisberg et al., 1993). Thus, it is not possible based on our chemical and isotopic data to rule out a chondrule origin for the AMM olivine and pyroxene grains.

In meteorite olivine grains, the degree of ¹⁶O enrichment has often been reported to be broadly negatively correlated with the FeO content of olivine grains (Hervig and Steele, 1992; Leshin et al., 1997; Saxton et al., 1995; Weinbruch et al., 1993). Likewise, an inverse correlation between bulk chondrule ¹⁶O-enrichment and FeO content has also been observed (Clayton and Mayeda, 1983; McSween, 1985; Rubin et al., 1990). Various hypotheses have been advanced to account for this apparent correlation. McSween (1985) suggested that a greater degree of isotopic exchange could occur between ¹⁶O-enriched chondrule precursors and an ¹⁶O-depleted nebular gas during

more prolonged melting of the more FeO-rich chondrules, while Rubin et al. (1990) invoked mixing of two solid components with distinctive ¹⁶O and FeO contents to form chondrule precursors. Weinbruch et al. (1993) interpreted a correlation of ¹⁶O-enrichment with FeO content between the cores and rims of two isolated Allende olivine grains as reflecting condensation processes in the solar nebula.

Although the most forsteritic AMM olivine grain (#94-4B-5) shows the greatest ¹⁶O-enrichment, there is no overall correlation of FeO-content and oxygen isotopic composition in our AMM olivines (Fig. 6). In fact, this observation agrees with the most recent ion microprobe studies of the oxygen isotopic compositions of individual minerals in carbonaceous chondrites (Jones et al., 1998; Leshin et al., 1998) which demonstrate that this correlation breaks down upon detailed examination. Thus, the data indicate that the processes responsible for the FeO-content and oxygen isotopic compositions of meteoritic (and AMM) olivine grains are not linked in a straightforward manner.

The oxygen isotope data for olivine and pyroxene from AMMs also overlap the data available for whole chondritic interplanetary dust particles (IDPs) collected from the stratosphere (McKeegan, 1987a; McKeegan, 1987b; Stadermann, 1990). However, a valid comparison is not readily made since the IDPs were measured by crushing the entire particle onto a gold foil. Recent analyses of single silicate mineral grains in 2

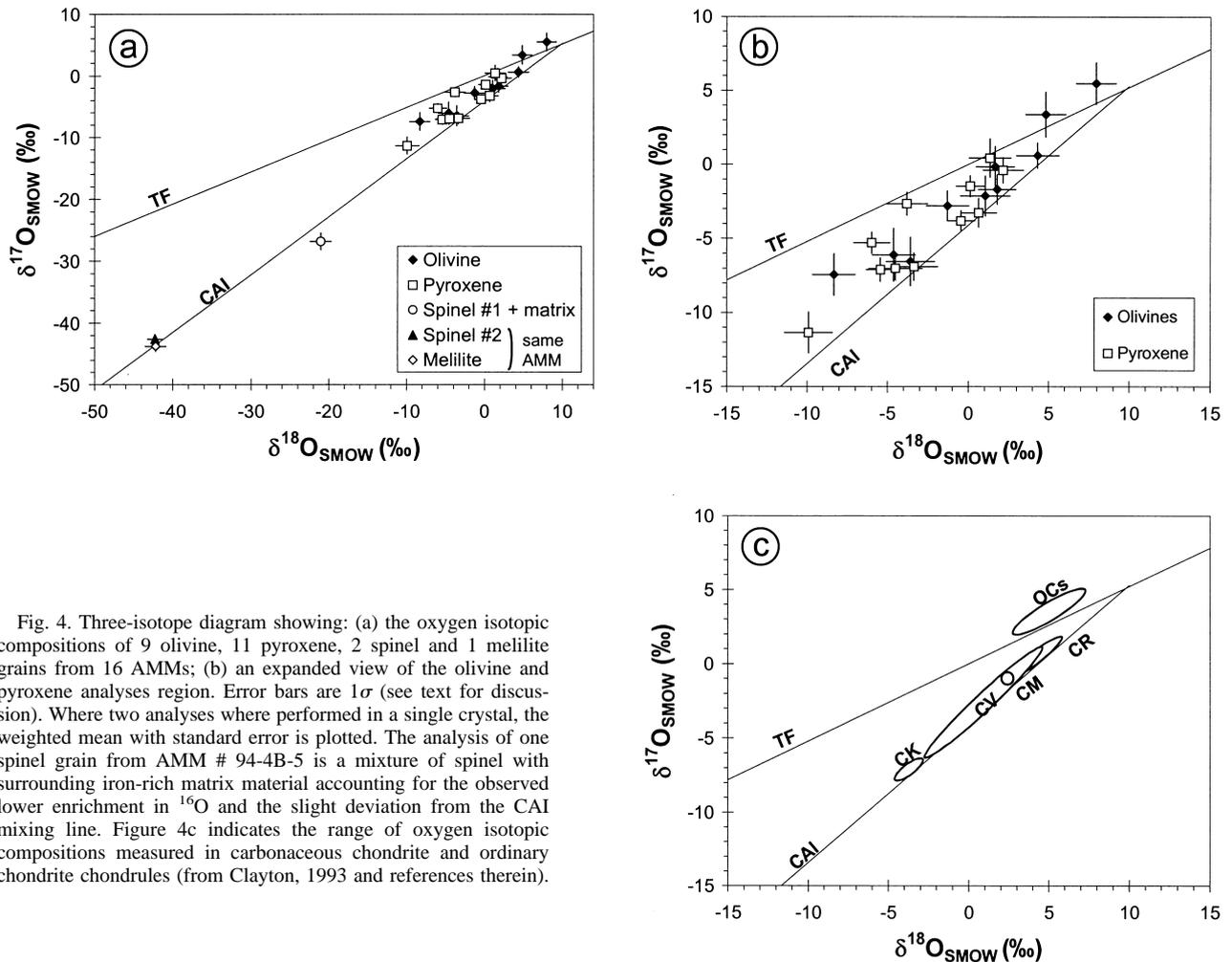


Fig. 4. Three-isotope diagram showing: (a) the oxygen isotopic compositions of 9 olivine, 11 pyroxene, 2 spinel and 1 melilite grains from 16 AMMs; (b) an expanded view of the olivine and pyroxene analyses region. Error bars are 1σ (see text for discussion). Where two analyses were performed in a single crystal, the weighted mean with standard error is plotted. The analysis of one spinel grain from AMM # 94-4B-5 is a mixture of spinel with surrounding iron-rich matrix material accounting for the observed lower enrichment in ^{16}O and the slight deviation from the CAI mixing line. Figure 4c indicates the range of oxygen isotopic compositions measured in carbonaceous chondrite and ordinary chondrite chondrules (from Clayton, 1993 and references therein).

IDPs (Engrand et al., 1999) are compatible with the AMM mafic silicate data presented here.

4.2. Refractory Mineral Data: Comparison to Meteorites and Interplanetary Dust Particles

In chondritic meteorites, with the exception of the recently discovered olivine discussed above, large ^{16}O -excesses are exclusively associated with the refractory oxide and Ti-rich pyroxene (fassaite) minerals found in CAIs. The relative abundance of AMMs that bear refractory mineral phases is at most a few percent (Engrand and Maurette, 1998; Greshake et al., 1996; Kurat et al., 1994b), hence these materials have not been very extensively studied compared to their counterparts in carbonaceous chondrites. The overall mineralogy of refractory phases in AMMs appears to be similar to that for refractory inclusions in CM chondrites (see Greshake et al., 1996; Kurat et al., 1994b). As in CM chondrites, melilite may be relatively rare as only one melilite grain has been identified in AMMs thus far (Engrand et al., 1997, this study).

Previous oxygen isotopic data on 3 spinel-bearing AMMs showed variable ^{16}O -excesses that are broadly consistent with the CAI mixing line (Greshake et al., 1996; Hoppe et al., 1995;

Kurat et al., 1994a). Our oxygen isotopic data are in general agreement with these measurements, but for the first time convincingly show that refractory minerals from AMMs fall on the CAI line (Fig. 4a). Although in one particle the small grain size prevented a clean analysis of only refractory material (AMM #94-4B-5), in the other particle examined (AMM #92-13C-23) it is clear that both spinel and melilite are enriched in ^{16}O at the level of $\delta^{18}\text{O} \sim -40\text{‰}$, as is the case for most spinel grains in CAIs from carbonaceous chondrites (Clayton, 1993 and references therein; McKeegan et al., 1996). The isotopic composition of AMM #92-13C-23 is strikingly similar to that measured in a melilite-spinel-hibonite bearing inclusion from the unequilibrated ordinary chondrite Semarkona (McKeegan et al., 1998b).

The AMM data are also in good agreement with measurements of "bulk" refractory stratospheric IDPs which have oxygen isotopic compositions on the CAI line, with enrichments in ^{16}O up to $\delta^{18}\text{O} \approx \delta^{17}\text{O} \approx -40\text{‰}$ (McKeegan, 1987a, 1987b; Stadermann, 1990, 1991). The refractory mineral assemblages identified in IDPs (Christoffersen and Buseck, 1986; Tomeoka and Buseck, 1985; Zolensky, 1987) consist mainly of spinel, perovskite, fassaite, hibonite, melilite, corundum, diop-

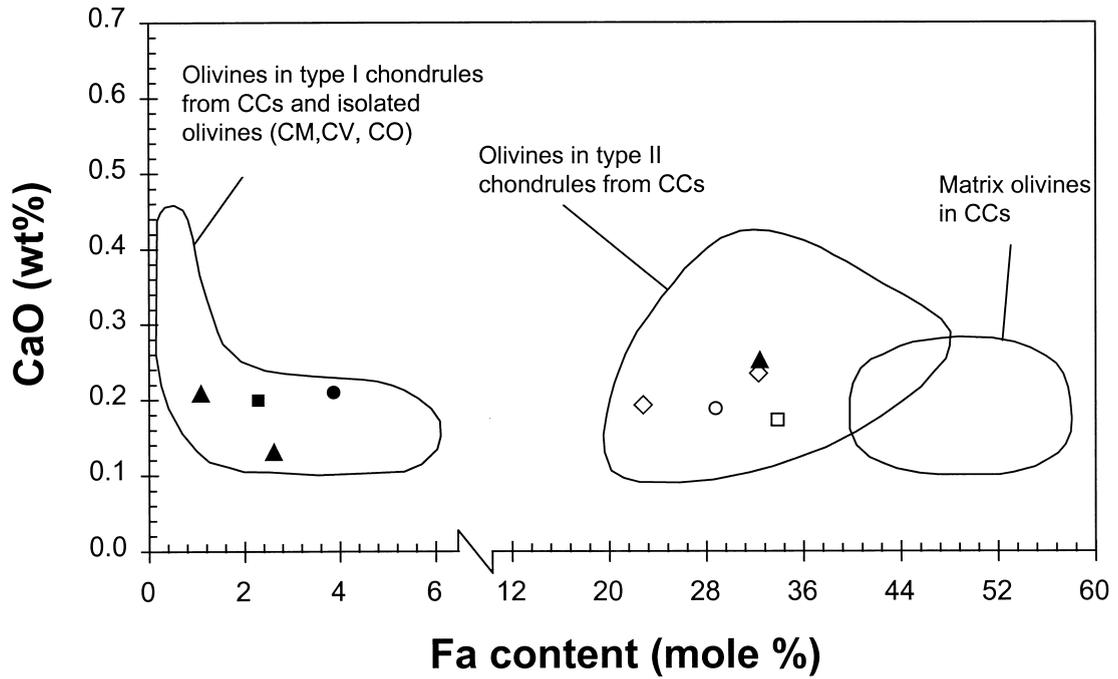


Fig. 5. Weight percent CaO vs. FeO measured in olivines from carbonaceous and ordinary chondrites (adapted from Fig. 4 from Beckerling and Bishoff, 1995), and in the AMM olivines analyzed in this study. Identical symbols are used for olivine grains within a single micrometeorite.

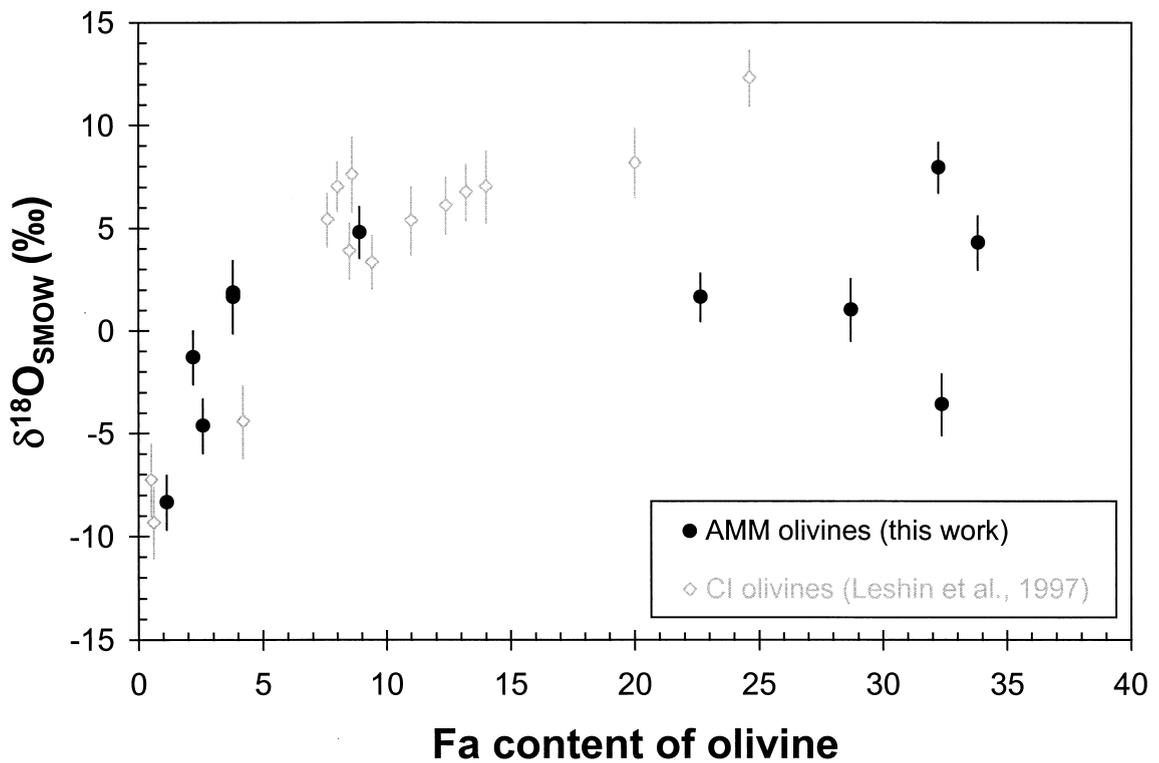


Fig. 6. $\delta^{18}\text{O}$ as a function of the fayalite (Fa) content of the olivine. The lack of a definite correlation is consistent with other recent ion microprobe studies of olivine from carbonaceous chondrites. See text for discussion.

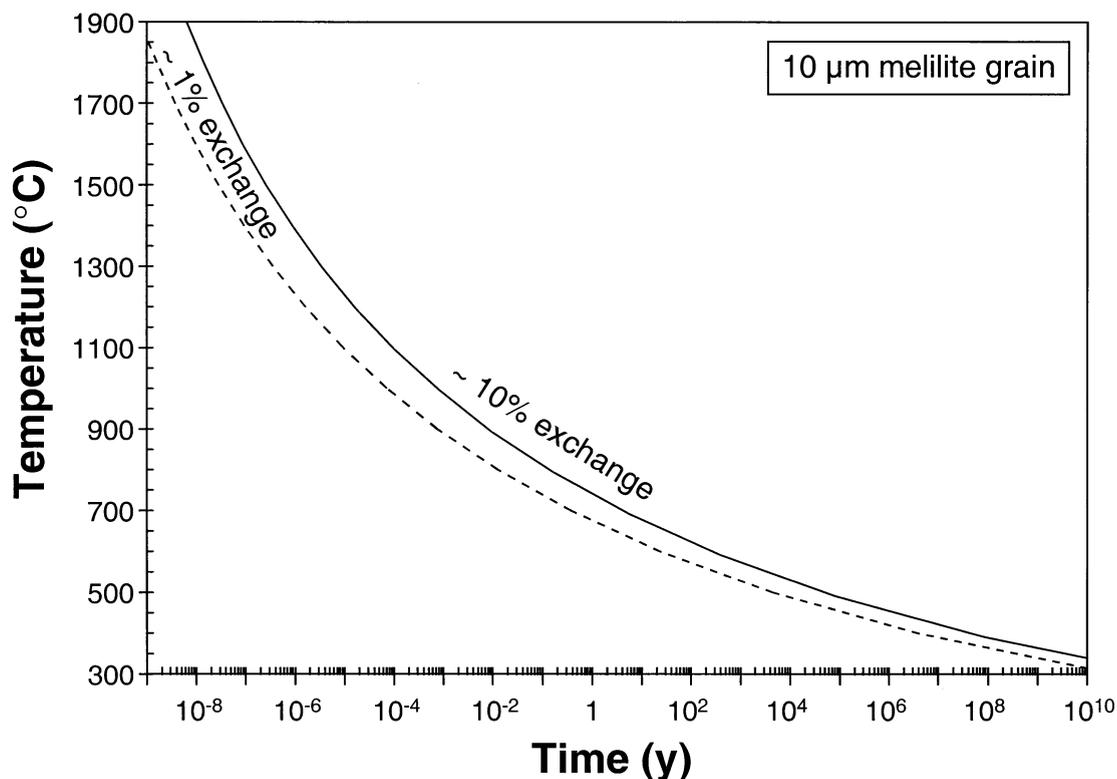


Fig. 7. Time-temperature diagram for a gas-solid exchange diffusion scenario constrained by the isotopic composition of the AMM melilite grain (see text). For a given temperature, the curves represent the time necessary to exchange 10% and 1%, respectively, of the oxygen by gas-solid reaction and diffusion in a 10 μm diameter melilite crystal.

side and anorthite (see Greshake et al., 1996 for a review). The refractory minerals in these particles differ from those in most carbonaceous chondrite CAIs by their sub-micron grain size (e.g., Zolensky, 1987), whereas the typical grain size of AMM CAIs is on the order of microns. Although the data are limited, refractory IDPs also seem to exhibit a wider mineral variety than AMM CAIs. However, from the viewpoint of oxygen isotopes, micrometeorite refractory minerals bear a striking similarity to meteoritic CAIs and refractory IDPs.

A possible exception to this isotopic similarity is provided by the observation of ^{16}O -rich melilite in AMM #92-13C-23. Although recent ion microprobe analyses have uncovered isotopically anomalous (^{16}O -enriched) melilite in CAIs from both unequilibrated ordinary chondrites (McKeegan et al., 1998b) and carbonaceous chondrites (Hiyagon, 1998; Ito et al., 1998; Kim et al., 1998; Kimura et al., 1993), most melilite in CAIs from carbonaceous chondrites has close to a “normal” oxygen isotopic composition (Clayton et al., 1977). The most commonly accepted interpretation to explain the isotopic heterogeneity between melilite and ^{16}O -enriched spinel and fassaite in (type B) carbonaceous chondrite CAIs invokes post-crystallization solid-gas isotopic exchange between initially ^{16}O -enriched refractory minerals and ^{16}O -depleted nebular gas (Clayton and Mayeda, 1977). In this model, the extent of isotopic exchange depends on the rate of oxygen self-diffusion in each mineral phase and on the thermal history of the CAI.

Although it is not yet known how representative it is, the discovery of an isotopically anomalous melilite grain in an

AMM supports the hypothesis that refractory minerals throughout the solar nebula formed from a relatively uniformly ^{16}O -enriched reservoir (Clayton, 1993; McKeegan et al., 1998b). Assuming that both the spinel and the melilite grains start with the same initial oxygen isotopic composition, the observed isotopic homogeneity between melilite and spinel in AMM #92-13C-23 implies a low degree of exchange with nebular gas and thus provides some constraints regarding the thermal history experienced by this AMM since the time of its formation. Considering analytical uncertainties, the maximum degree of isotopic exchange that could be permitted by the data would be a change of the $\delta^{18}\text{O}$ value by $\sim 5\%$ which is equivalent to $\sim 10\%$ exchange with a gas reservoir of assumed nebular composition (Clayton, 1993; Rowe et al., 1994). If the diffusion length-scale can be approximated by the grain size ($\sim 10 \mu\text{m}$ in diameter) and the oxygen self-diffusion coefficient for this melilite ($\text{\AA}^2\text{s}^{-1}$) can be interpolated from the experiments of Yurimoto et al. (1989, see their Fig. 6), then $\sim 10\%$ isotopic exchange can occur in a matter of minutes at high temperatures ($>1200^\circ\text{C}$) but could not be achieved in the lifetime of the solar system for temperatures below 300°C (see Fig. 7).

4.3. Possible Implications for Oxygen Isotopic Reservoirs in the Solar Nebula

Because micrometeorites constitute the majority of the mass flux currently being accreted by the Earth, they may provide a more representative sampling of extraterrestrial materials than

conventional meteorite collections. Therefore, their oxygen isotope compositions could give important insights into the distribution of oxygen isotopic reservoirs in the solar nebula. If the micrometeorite population is mainly derived from asteroids, our data imply that carbonaceous chondrite material is dominant in the asteroid belt, as opposed to the ordinary chondrites which constitute $\sim 80\%$ of meteorites. This is in agreement with conclusions reached by Meibom and Clark (1999) based mostly on spectroscopic observations of asteroids. One important limitation, however, is that the origins of individual micrometeorites, in the sense of locale within the solar system, are not well constrained. Unlike chondrites, which certainly originate in the asteroid belt (Wetherill and Chapman, 1988), some fraction of micrometeorites are probably samples of comets. However, the proportion of cometary particles is uncertain (Levasseur-Regourd et al., 1991; Liou et al., 1995 and references therein; Liou and Zook, 1996, 1997; Liou et al., 1996) and the source of any individual particle—such as those measured here—is a matter of speculation. Current knowledge about the oxygen isotopic compositions of comets is insufficient to provide much useful information regarding possible differences in oxygen isotope reservoirs between inner and outer solar system materials. In the water vapor of comet Halley, Eberhardt et al. (1995) and Balsiger et al. (1995) quoted values of $\delta^{18}\text{O} = +12 \pm 74\%$ and $-38 \pm 62\%$, respectively. Within their substantial uncertainties, these values overlap with all terrestrial and meteoritic compositions, including those for AMMs and those inferred for volatile species (e.g., Choi et al., 1997a; Clayton and Mayeda, 1984; Leshin et al., 1997). Note also that this isotopic value, measured in water vapor, might not be representative of that of the cometary silicate grains, for which no direct measurements are available as yet. Despite the uncertainties regarding micrometeorite sources, the most straightforward interpretation of the data presented here is that the oxygen isotopic compositions of the majority of the unequilibrated anhydrous silicates in the solar system are similar to those of carbonaceous chondrites and not ordinary chondrites.

The refractory minerals from CAIs in both carbonaceous and ordinary chondrites as well as from refractory IDPs and AMMs exhibit a nearly constant maximum ^{16}O -enrichment of $\sim +50\%$. This indicates that an ^{16}O -enriched reservoir of refractory dust must have been ubiquitous in the solar nebula or alternatively requires a mechanism for scattering refractory materials, originally formed in a restricted nebular locale, over widely variable heliocentric distances (McKeegan et al., 1998b). One such mechanism could be the bipolar outflows (x-winds) discussed by Shu et al. (1996). Recent studies (Hiyagon, 1998; Ito et al., 1998; Kim et al., 1998; Kimura et al., 1993; McKeegan et al., 1998b, this study) have demonstrated that ^{16}O -enriched melilite exists, and may be more common in small, fine-grained refractory mineral assemblages than in coarse-grained (primarily type-B) CAIs where ^{16}O -enriched melilite appears to be rare. Either these refractory objects, which were incorporated into parent bodies that probably were assembled at different heliocentric distances, experienced distinct thermal histories or they were exposed to different isotopic environments. To what extent a possible correlation of petrographic occurrence and isotopic composition of melilite provides evidence for production of refractory minerals in a

highly localized region of the solar nebula, as in the Shu et al. (1996) model, remains an open question that can be addressed by further analyses of refractory minerals in primitive extraterrestrial materials. The answer ultimately has important implications for understanding the origin of the premier isotopic anomaly in the solar system, whether as a manifestation of the chemical memory of interstellar dust (Clayton, 1982; Clayton et al., 1973), or as a product of non-equilibrium chemistry in the solar nebula (Thiemens, 1996; Thiemens and Heidenreich, 1983).

5. CONCLUSIONS

The oxygen isotopic compositions of the individual minerals in AMMs studied here do not show evidence for exchange with terrestrial oxygen upon atmospheric entry. This is consistent with the unmelted texture and pristine chemistry of the minerals. Thus, the oxygen isotopic data can be directly compared to data for similar phases in meteorites.

The oxygen isotopic compositions of olivine and pyroxene grains in AMMs overlap those observed in carbonaceous chondrites. Thus, this work documents an additional important similarity between AMMs and primitive carbonaceous meteorites, adding to the chemical, mineralogical and isotopic similarities previously discussed by Kurat et al. (1994b), Kurat (1998) and Engrand and Maurette (1998).

The petrographic, chemical, and isotopic data presented here do not permit a unique interpretation of the origin of the AMM mineral grains studied. Whole chondrules are extremely rare in the AMM collection, and thus a chondrule origin is not definitively indicated by the petrographic occurrence of the AMM minerals studied. The chemistry and oxygen isotopic compositions of at least some AMM mineral grains, however, are consistent with an origin from the igneous crystallization and subsequent breakage of chondrules, based upon comparison to the available data in chondritic meteorites. Further studies of AMMs, as well as of isolated minerals in chondrites, are necessary to define evidence for igneous versus condensation origin of isolated minerals which are not petrographically associated with chondrules.

We did not observe any extreme ^{16}O enrichments in our selection of olivine grains, such as has been recently reported in carbonaceous chondrite olivines associated with refractory inclusions (Hiyagon, 1998; Hiyagon and Hashimoto, 1999; McKeegan et al., 1998a). Thus the olivine grains studied here probably did not originate in a similar way to the olivine within or rimming refractory inclusions in carbonaceous chondrites.

In agreement with other recent studies (Jones et al., 1998; Leshin et al., 1998), we do not observe a correlation between the ^{16}O -enrichment of the olivine grains and their FeO content, suggesting that the processes responsible for the FeO and ^{16}O abundances are not related in a straightforward way.

The AMM spinel and melilite grains in this study are enriched in ^{16}O at the level of $\delta^{17}\text{O} \approx \delta^{18}\text{O} \approx -40\%$, similar to refractory minerals in carbonaceous chondrites (Clayton, 1993 and references therein), in bulk refractory IDPs (Greshake et al., 1996; McKeegan, 1987b; Stadermann, 1990), and from a previous study of AMMs (Greshake et al., 1996; Kurat et al., 1994a). The discovery of an isotopically pristine AMM melilite

grain (enriched in ^{16}O at the same level as spinel) supports the model of formation of refractory minerals from an isotopically homogeneous ^{16}O -rich reservoir and subsequent gas-solid isotopic exchange with ^{16}O -depleted nebular gas to yield the mineralogically-dependent oxygen isotopic compositions thus far observed in most carbonaceous chondrite CAIs (e.g., Clayton, 1993 and references therein). ^{16}O -enriched melilite has now been documented in carbonaceous chondrites (Hiyagon, 1998; Ito et al., 1998; Kim et al., 1998; Kimura et al., 1993), ordinary chondrites (McKeegan et al., 1998b), and micrometeorites (this study), suggesting that there could be abundant examples of CAI minerals that escaped the secondary exchange process discussed above and testifying to the variety of environments and/or processes which must have affected materials in the early solar system.

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