DISCOVERY OF PRESOLAR SILICATE GRAINS IN THE ACFER 094 CARBONACEOUS CHONDRITE.

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Introduction: Though several presolar oxide phases have been identified in meteorites [1], presolar silicates remained elusive despite astronomical evidence that they are the most abundant grain species around O-rich evolved stars [2], the most likely stellar sources for other presolar oxides found in meteorites. Previous attempts to isolate presolar silicates in meteorites by ion probe analysis [3, 4] were restricted to grains >1μm due to the limiting spatial resolution of the instruments employed. Recently, the NanoSIMS, capable of isotopic measurements on submicron grains, the expected size of presolar silicates, was instrumental in the identification of six presolar silicates in interplanetary dust particles (IDPs) [5]. In addition, the NanoSIMS was used to identify two presolar O-rich grains containing Si in meteorites [6]. The mineralogy of these grains, however, could not be independently confirmed.

In the latter two studies, measurements were made in situ. Here, we use raster ion imaging in the NanoSIMS [7] to perform O isotopic analyses of tightly packed sub-μm size-separated grains from the matrix of Acfer 094, a highly primitive carbonaceous chondrite [8]. Nine anomalous grains were found, and the mineralogy of 5 was determined by X-ray analysis. One grain was found to be enriched in ^{26}Mg, most likely due to the decay of short-lived ^{26}Al.

Experimental: A sample of Acfer 094 was disaggregated by repeated freezing and thawing, followed by ultrasonication. Soluble organics were removed from the fine-grained material with a solution of toluene and methanol. This procedure affords a better sample dispersion by reducing agglomeration. Size separates of matrix grains were produced by centrifugation in 85% isopropanol and 15% water, and grains 0.1-0.5 μm in diameter were dispensed from suspension onto a gold foil for analysis.

To maximize efficiency, measurements were made on densely covered areas of the sample mount with the NanoSIMS ion microprobe. A ~100nm Cs$^+$ primary ion beam was rastered over $20 \times 20 \mu m^2$ areas. High-mass-resolution negative-secondary-ion images of the three O isotopes, ^{28}Si and ^{24}MgO were obtained by simultaneous detection. For each area, between 20-40 consecutive 256×256 pixel images were acquired to produce a final integrated image. The acquisition of one complete image took 3.5-7 hours.

Isotopically anomalous grains were identified in oxygen isotopic ratio images (Fig. 1), where the ratios are given as δ-values or deviations from the average isotopic ratio of the whole image in permil (‰). The analyzed matrix grains are mainly of solar system origin, which allows us to use them as isotopic standards. We divided each image containing a presolar grain candidate into areas comparable in size to that of the candidate grain and determined the oxygen isotopic ratios of these areas. Only grains with isotopic ratios falling well outside the range observed for isotopically "normal" grains were considered to be presolar. The ^{28}Si$^=$ and ^{24}MgO$^-$ images aid in first-order mineralogical identification. Five anomalous grains were relocated in a field emission SEM where secondary electron images and energy dispersive X-ray (EDX) spectra were taken. We succeeded also in measuring the three Mg isotopes, ^{27}Al, and ^{28}Si as positive secondary ions for the anomalous grain shown in Fig. 1. This was done by rastering a ~300nm O$^-$ primary beam over a $10 \times 10 \mu m^2$ area around the grain of interest.
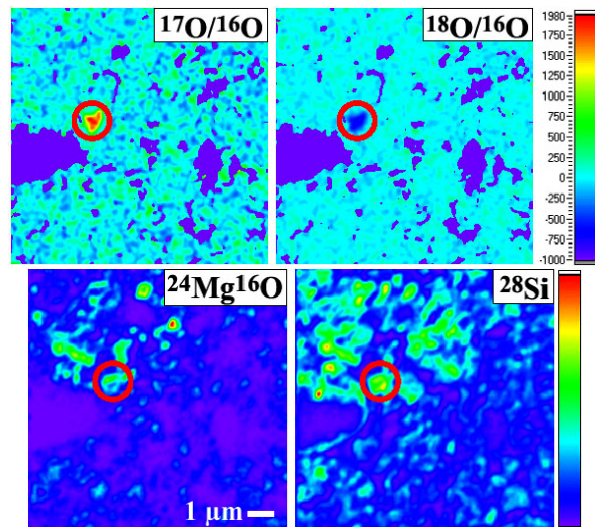

Figure 1. Oxygen isotopic ratio images and negative secondary ion images of ^{24}MgO and ^{28}Si for a $10 \times 10 \mu m^2$ area partially covered with matrix grains from Acfer 094. A ~0.5μm grain (circled) displays a large ^{17}O excess and ^{18}O deficit relative to the surrounding grains, and clearly contains Si and Mg.

Results and discussion: In the 16 different $20 \times 20 \mu m^2$ areas analyzed, a total of 9 anomalous grains with sizes ranging from 0.1μm to 0.6μm were identified. The ^{24}MgO/^{16}O and ^{28}Si/^{16}O ratios for all anomalous grains fall within the range for matrix grains, which are primarily silicates. The oxygen isotopic ratios of the 9 anomalous grains are shown in Fig. 2 along with those of isotopically "normal" grains. Raster ion imaging of densely packed grains generally results in a dilution of anomalies due to contributing signal

from surrounding grains of "normal" isotopic composition. Thus, anomalous isotopic ratios are to be taken as lower limits.

The presolar silicates located in this study span all 4 presolar oxide groups [9], and their oxygen isotopic ratios fall within the range previously seen for presolar oxides. The majority of the presolar silicates derived from low mass RGB and AGB stars. Due to current limited statistics, it is impossible to make any comparisons between circumstellar silicates from meteorites and those from IDPs, nor between different types of presolar oxide grains.
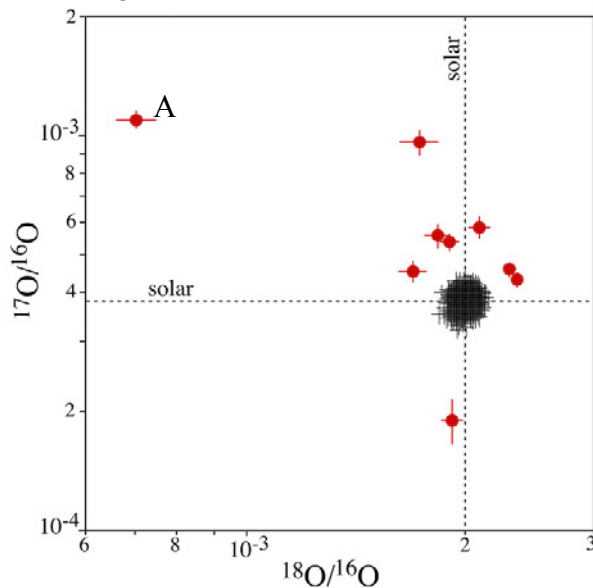

Figure 2. Oxygen isotopic ratios of presolar silicate grains from Acfer 094 (red). Grain A is shown in Fig. 1. Also shown are the isotopic ratios of grains of "normal" isotopic composition (black). The dashed lines indicate solar isotopic ratios.

Silicate grains afford for isotopic analysis of not only O, but Mg and Si as well. Mg isotopic analysis of grain A found it to be enriched in ^{26}Mg by 119 ± 5‰. This enrichment is attributed to the in situ decay of ^{26}Al and implies an initial ^{26}Al/^{27}Al of ~0.067. The O isotopic ratios for this particular grain suggest an origin in a low-mass ABG star. Furthermore, the large ^{26}Al/^{27}Al ratio, which is higher than any found in presolar oxide grains, indicates very deep mixing (cool bottom processing) in the parent star during the thermally pulsing phase [10].

The mineralogy of 5 presolar silicates was determined from EDX spectra (Fig. 3). Three of the grains are olivines and two are pyroxenes. Astronomical observations identified amorphous silicates, and crystalline forsterite, enstatite, and diopside as the major silicate species around young and evolved O-rich stars [2]. While it is predicted that circumstellar silicates are Mg-rich,

4 of the presolar silicates found in this study are Fe-rich. This, however, could be attributed to subsequent oxidation of forsteritic and enstatitic grains.
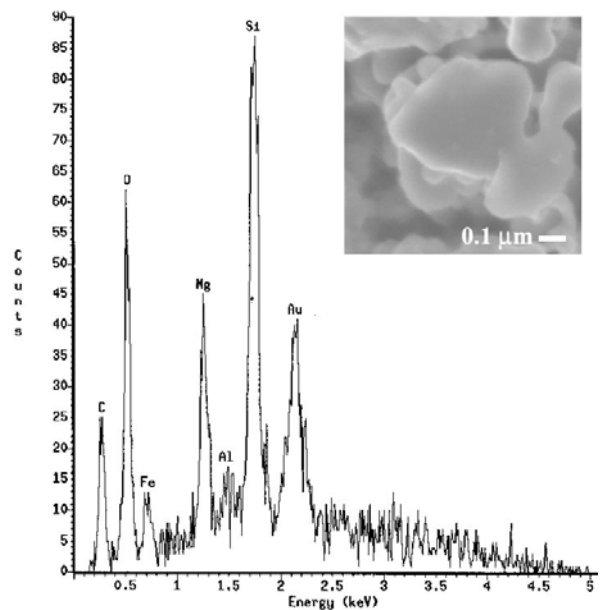

Figure 3. EDX spectrum of presolar grain A, a pyroxene, obtained with an electron beam of 5keV. A high-magnification image of this grain was also obtained. The Au line is due to the Au foil, and the C line is due to contamination in the SEM.

The abundance of presolar silicate grains in Acfer 094 is calculated to be ~25ppm (~40ppm relative to the matrix). This abundance, which should be taken as a lower limit, is higher than that of other presolar phases in meteorites, excluding possibly nanodiamonds. In contrast, the abundance of presolar silicates in IDPs is ~5500ppm [2]. This conveys the highly primitive state of IDPs relative to meteorites. The sample chosen for this study, Acfer 094, is one of the most primitive meteorites. Future studies conducted on different meteorite types will hopefully allow us to understand the processes that potentially destroy presolar silicates, and conditions in different solar system environments.

References: [1] Zinner, E. (1998) *Ann. Rev. Earth Planet. Sci. 26*, 147-188. [2] Waters, L. B. F. M. *et al.* (1996) *A&A 315*, L361-L364. [3] Messenger, S. and Bernatowicz, T. J. (2000) *M&PS 35*, A109. [4] Alexander, C. M. O'D. *et al.* (2001) *LPS XXXII* Abstract #2191. [5] Messenger, S. *et al.* (2003) *Science 300*, 105-108. [6] Mostefaoui, S. *et al.* (2003) *M&PS 38*, A99. [7] Nguyen, A. *et al.* (2003) *Publ. Astron. Soc. Australia 20*, 382-388. [8] Newton, J. *et al.* (1995) *Meteoritics 30*, 47-56. [9] Nittler L. R. *et al.* (1997) *ApJ 483*, 475-495. [10] Nollett, K. M. *et al.* (2003) *ApJ 582*, 1036-1058.