NANOSIMS OXYGEN- AND SULFUR-ISOTOPE IMAGING OF PRIMITIVE SOLAR SYSTEM MATERIALS.
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Introduction: Primitive meteorites and interplanetary dust particles (IDPs) contain nm- to µm-sized presolar dust grains that formed in the winds of evolved stars or in the ejecta of supernova and nova explosions [1-3]. Silicates are the major constituent of O-rich dust around young stars and in outflows from evolved red giant stars [4]. Although the first presolar minerals, namely diamond and SiC, were identified already in 1987 in carbonaceous meteorites, presolar silicates were discovered only recently, first in IDPs and later also in primitive meteorites [5-8]. The identification of presolar silicates was based on the application of improved measurement techniques and the invention of the NanoSIMS ion microprobe with its superior lateral resolution (<100 nm) and capability for the search of in-situ presolar dust in slices of IDPs and meteorites has played a key role in this respect.

In a previous study we reported the discovery of abundant in-situ presolar silicate and spinel grains in the matrix of the Acfer 094 meteorite [8]. The finding of these grains was based on O-isotope mapping of several matrix areas with the NanoSIMS ion microprobe at MPI for Chemistry. In an attempt to further characterize presolar minerals in IDPs and meteorites we report here results from an O- and S-isotope imaging survey of the Acfer 094 meteorite and of two IDPs.

Experimental: The O-isotopic measurements were performed on a polished thin section of Acfer 094 and on 11 microtome sections of IDPs U2071J2 and U2071C9 using the NanoSIMS at MPI for Chemistry. For the O-isotopic measurements in Acfer 094 we selected 40 different matrix areas. A focussed Cs^+ ion beam (<100 nm) of ~0.5 pA was rastered over the selected areas, each 9×9 µm^2 in size. Negative secondary ions of the three O isotopes, ^{28}Si, and ^{27}Al^{16}O were simultaneously measured in multi-collection and 256×256 pixel image sequences (total integration time of ~30 minutes to ~1 hour per image set) were acquired. Microtome sections of the two non-cluster IDPs, 9×5 µm^2 (U2071J2) and 12×8 µm^2 (U2071C9) in size, were prepared. These particles are part of a comprehensive consortium study of particles from collection surface U2071 [9,10]. The O-isotope imaging was done on four (U2071J2) and seven (U2071C9) microtome sections using similar analytical conditions as for Acfer 094, except that raster sizes varied from 8×8 to 14×14 µm^2 and that ^{27}Al^{16}O was not included in the measurements.

The S-isotopic measurements were done on three areas in the matrix of Acfer 094 which were studied for O-isotopic compositions before. A focused Cs^+ ion beam of ~0.5 pA was rastered over the three areas, 8x8 to 10×10 µm^2 in size. Negative secondary ions of ^{16}O, ^{32}S, ^{33}S, ^{34}S, and ^{36}S were simultaneously measured in multi-collection and 256×256 pixel image sequences (total integration time of ~1 to ~1.5 hours per image set) were acquired.

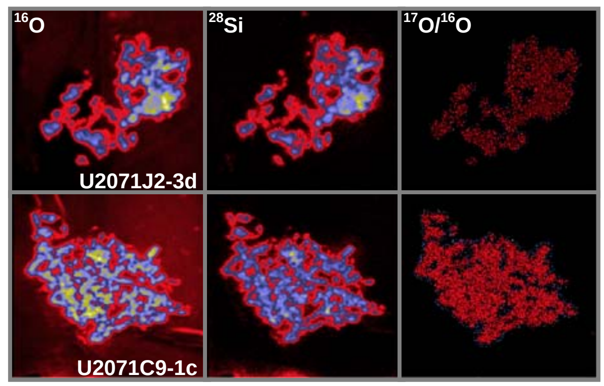

Figure 1. ^{16}O, ^{28}Si, and ^{17}O/^{16}O ion images made on microtome sections of IDPs U2071J2 and U2071C9. Field of view is 10×10 µm^2 (U2071J2) and 12×12 µm^2 (U2071C9).

Results: *O-isotope mapping of IDPs.* Automatic particle recognition revealed 793 objects in the 11 microtome sections, 150 to 1200 nm in size, representing a total area of 225 µm^2. All of these objects have normal ^{17}O/^{16}O and ^{18}O/^{16}O ratios (Fig. 1) within the analytical uncertainties of typically 13 % (^{17}O/^{16}O) and 6 % (^{18}O/^{16}O), respectively, i.e., no presolar grains were identified.

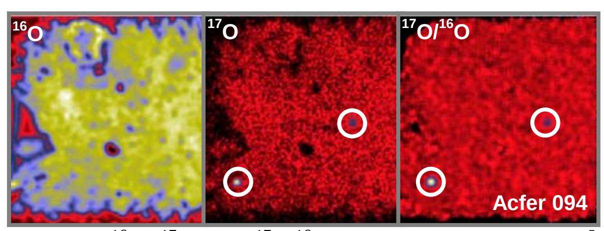

Figure 2. ^{16}O, ^{17}O, and ^{17}O/^{16}O ion images of a 9×9 µm^2 area in the matrix of Acfer 094. Two presolar silicate grains are encircled.

O-isotope mapping of Acfer 094. Based on the O-isotopic compositions seven presolar grains were identified (Fig. 2). These grains have ^{17}O/^{16}O ratios between 1× and 16× solar and $^{18}O/^{16}O$ ratios ranging from 0.9× to 1.4× solar (Fig. 3). The $^{17}O/^{16}O$ ratio of 16× solar is among the highest ratios found so far for presolar O-rich dust [11]. From the Si/O and AlO/O ratios measured by NanoSIMS six of these grains appear to be silicates and one spinel. A more detailed mineralogy determination by SEM/EDX is in progress. Grain sizes vary between 200 and 350 nm.

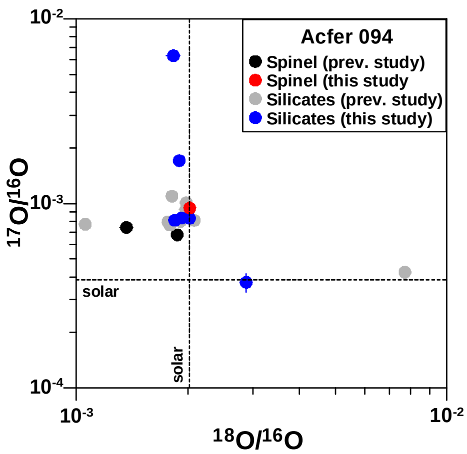

Figure 3. O-isotopic ratios of presolar spinel and silicates in the Acfer 094 meteorite. Previous study: [8].

S-isotope mapping of Acfer 094. Automatic particle recognition identified 402 S-rich grains (Fig. 4). All of these grains have normal $^{33}S/^{32}S$ and $^{34}S/^{32}S$ ratios within the uncertainties of typically 30 ‰ and 13 ‰, respectively. In $^{36}S/^{32}S$, however, two grains, about 150 nm in size, exhibit a strong depletion in ^{36}S with $\delta^{36}S$ of -793±104 ‰ and -816±130 ‰. A determination of their mineralogy is in progress.

Discussion: *IDPs U2071J2 and U2071C9.* The analyzed volume represents only several percent of the total material of these two IDPs and the non-finding of presolar grains is still consistent with an abundance of several 100 ppm of presolar grains in these two IDPs as observed for other IDPs [12]. We will continue our search for presolar grains in microtome sections of IDPs in the near future.

O-rich presolar dust in Acfer 094. The O-isotopic signatures of four of the presolar silicate grains and of the presolar spinel grain are consistent with an origin in 1.5-2 $M_\odot$ RGB/AGB stars of close-to-solar metallicity [13]. Although its $^{17}O/^{16}O$ ratio is outside the range of predicted O-isotopic ratios in RGB/AGB stars [13] the presolar silicate grain with the largest ^{17}O enrichment might have formed in an RGB/AGB star as well. The silicate grain with the high $^{18}O/^{16}O$ ratio could have formed in an AGB star or a supernovae [11,14]. However, the Si- and Fe-isotopic ratios of a silicate grain with similar O-isotopic signature [8] seems to favor the AGB star origin. Combining our new data with those from [8] we obtain matrix-normalized abundances of 130 ppm for presolar silicates and of 50 ppm for presolar spinel.

S-rich (presolar?) dust in Acfer 094. The errors given above for the $\delta^{36}S$ values of the two ^{36}S-depleted grains were calculated from counting statistics, i.e., are based essentially on the actually observed counts for ^{36}S. However, the distribution of $\delta^{36}S$ values for a population of grains with normal $^{36}S/^{32}S$ ratios is expected to have a standard deviation of

$$1000\Big/\sqrt{\left(^{36}S/^{32}S\right)_{normal}\cdot cts\left(^{32}S\right)},$$

which is larger than the calculated errors. Taking this into account the observed low $\delta^{36}S$ values are only 2.7 and 3.5σ away from normal, i.e., they may just represent statistical outliers. A final positive identification of grains with low ^{36}S thus requires the finding of larger grains that permit to detect a larger number of ^{36}S ions.

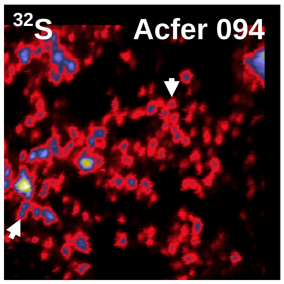

Figure 4. ^{32}S ion image of a 8×8 μm^2 area in the matrix of Acfer 094. The arrows indicate two grains with strong depletion in ^{36}S.

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