EXTREME UNCORRELATED ^{54}CR, ^{17}O, AND ^{18}O ENRICHMENTS IN SUB-MICRON ORGUEIL GRAINS.

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Introduction: Cr isotopic anomalies, most notably enrichments in ^{54}Cr, are widely distributed in inner Solar System materials [1-6]. These variations have been attributed to nucleosynthetic effects, possibly carried by as-yet-unidentified presolar grains. We recently reported the discovery of sub-micron ^{54}Cr-rich (by up to 300 ‰) oxide grains in an Orgueil acid residue [7]. Continued searches have identified additional grains with more extreme ^{54}Cr enrichments, reaching at least as high as 1500 ‰. O isotopic measurements on the same mount revealed numerous presolar oxide grains, some with extreme O anomalies, but no resolved anomalies for ^{54}Cr-rich grains. Auger spectroscopic measurements are in progress to characterize the mineralogy of the isotopically anomalous grains.

Methods: A CsF/HCl Orgueil residue [7] was ashed in an O-plasma to remove C and remaining grains dispersed onto a gold substrate. SEM examination of the mount shows that the sample consists mostly of sub-micron Cr-rich oxide grains (most likely Cr spinels with varying amounts of Mg, Fe and Al). Isotopic measurements were made with the Carnegie NanoSIMS 50L ion microprobe in imaging mode. The density of grains on the mount is very high, allowing 100s to 1000s of grains to be analyzed in each image, but with the drawback that isotopic compositions of single grains can be strongly influenced by dilution of signals from surrounding material, particularly for the Cr measurements. O isotopes were first measured with a ~100 nm Cs^+ beam rastered over 20×20 μm areas and simultaneous collection of negative secondary ions of ^{16,17,18}O, ^{28}Si, Al^{16}O and ^{52}Cr^{16}O. Counting times resulted in typical uncertainties of 2-10% for O isotopic ratios for sub-micron oxide grains, sufficient to identify presolar grains. Cr isotopes were subsequently measured for many of the same areas with a 300-700 nm O^- beam and simultaneous collection of positive secondary ions of ^{28}Si, ^{48}Ti ^{50}Cr+^{50}Ti, ^{52}Cr, ^{53}Cr, ^{54}Cr+^{54}Fe, and ^{56}Fe. Isotopic ratio images were generated and isotopic ratios for individual regions determined through image processing; data for individual grains were normalized to the average composition in an image. ^{54}Cr and ^{50}Cr were corrected for interference from Fe and Ti, respectively, with the assumption of normal isotopic compositions for these elements.

Results: O isotopic imaging of ~100 areas revealed at least 160 presolar oxide grains (Fig. 1), making up <0.1% of the cumulative analyzed grain area. Most are <400 nm in size and appear to be Al-rich. However, the true abundance is certainly higher by at least a factor of a few, since many presolar grains are surely missed due to the high density of the mount [8]. The grains span similar ranges of O isotopic ratios to previously observed in presolar oxides [9] and silicates [8]. However, few grains with extreme ^{18}O depletions are seen, likely due to dilution by surrounding normal material. Three grains with extreme ^{17}O enrichments (^{17}O/^{16}O ~1%) and two with the highest ^{18}O enrichments yet seen for presolar oxides were also found.

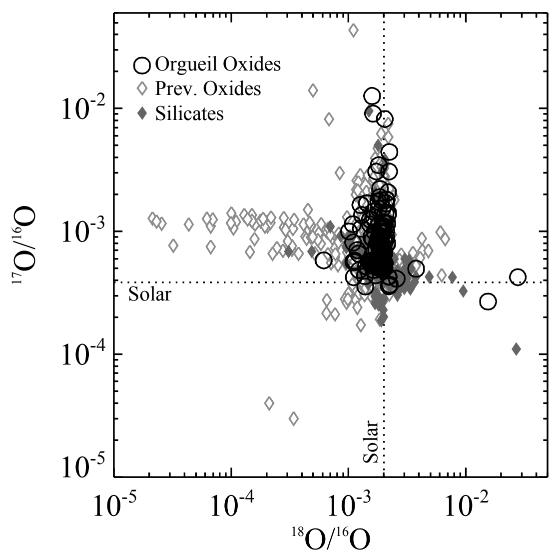

Figure 1: O isotopic ratios of newly identified presolar grains from an Orgueil residue and previously reported oxide and silicate grains.

Cr isotopic imaging of the same areas revealed three grains with resolvable enrichments of ^{54}Cr, with δ^{54}Cr values ranging from 200 to 1500‰. All three contain O with isotopic ratios that are within error of normal. δ^{54}Cr and δ^{53}Cr ratios are plotted in Fig. 2 for the 13 ^{54}Cr-rich grains we have identified to date, together with isotopically normal grains from the same ion images. We found no significant anomalies in ^{50}Cr or ^{53}Cr, with the exception of a slight ^{53}Cr deficit in grain 7-5 (δ^{53}Cr = −69±13 ‰).

The spatial resolution of the Cr isotopic images is slightly lower than the typical size of grains in the Orgueil residue. Thus, the Cr measurements are much more severely affected by isotope dilution than are the O ones and the measured δ^{54}Cr values are lower limits.

Moreover, the true abundance of ^{54}Cr-rich grains in the Orgueil residue is certainly much larger than implied by our results (<0.002 area%).

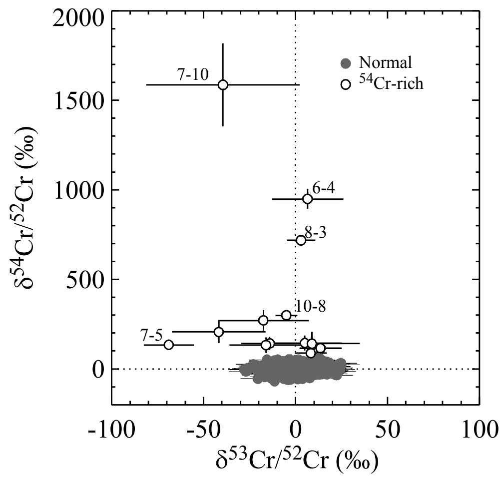

Figure 2: Cr 3-isotope plot for ^{54}Cr rich oxide grains (1σ errors) and similarly sized grains identified in the same SIMS images, indicating the range of compositions beyond which anomalous grains can be distinguished.

The small sizes of the anomalous grains precludes SEM-EDS as a useful method for determining their chemical compositions, but Auger spectroscopy is a viable alternative [10]. We have performed preliminary Auger measurements on a few of the most extreme ^{54}Cr-rich grains, but the analyses have been hampered by instrumental problems and prior contamination by hydrocarbon deposition in the SEM. Nonetheless, the initial data confirm that the grains are indeed Cr-bearing oxides as indicated by the SIMS results. The most likely carrier grain corresponding to the ^{54}Cr-hotspot 6-4 appears to contain Ti as well, as indicated by both NanoSIMS and Auger data.

Discussion: Despite their small size, the extreme ^{54}Cr enrichments of the grains observed here indicate that they could be an important contributor to ^{54}Cr variations in bulk chondrites. The moderate ^{53}Cr depletion observed in ^{54}Cr-rich grain 7-5 suggests that at least part of the anti-correlation between ^{53}Cr and ^{54}Cr seen in bulk meteorite leaching experiments can be attributed to similar grains. However, the specific origin of the Cr anomalies observed in the grains, without correlated effects in other isotopes, especially those of O, is a major puzzle. ^{54}Cr and other neutron-rich nuclei, such as ^{50}Ti, are believed to largely form in neutron-rich environments in both Type Ia and Type II supernovae (SN) [11, 12]. However, if the ^{54}Cr-rich oxide grains we have observed were indeed pure SN condensates, highly anomalous O isotopes would be expected, as observed in known presolar SN grains [13]. The lack of correlated O anomalies thus argues against a direct SN origin of the Orgueil grains and suggests that the grains are analogous to refractory inclusions and CM hibonites with Solar System O isotopes, but nuclear anomalies in other elements [14, 15]. Such anomalies are likely the result of `chemical memory' effects. One possibility is original formation as Cr-bearing metal grains in supernova ejecta, followed by oxidation either in the interstellar medium or solar nebula to form the observed oxide grains.

Alternatively, the ^{54}Cr enrichments might not be nucleosynthetic in origin. Qin et al. [6] have suggested that the correlated ^{54}Cr and ^{53}Cr anomalies could be spallation products formed by cosmic-ray irradiation in Fe-rich grains in the Solar System, or perhaps the ISM. To explain the lack of ^{53}Cr excesses in leachates of acid residues with large ^{54}Cr excesses, they postulated that prior to significant decay of the ^{53}Mn, the Fe-rich grains were altered in the parent bodies and the ^{54}Cr-rich Cr and the ^{53}Mn-rich Mn partitioned into separate phases. A similar process would be necessary to explain the grains reported here.

The majority of presolar oxide grains are well-explained as originating in low-mass AGB stars where mixing of H-burning ashes has increased the surface $^{17}O/^{16}O$ ratios [9]. However, such mixing cannot explain $^{17}O/^{16}O$ ratios higher than $\sim 4 \times 10^{-3}$ so grains with higher ratios, including 3 identified here (Fig. 1) require another origin. Both classical novae and mass-transfer in binary systems have been suggested as possible progenitors for such grains. The extreme ^{18}O enrichments observed in two Orgueil grains (Fig. 1) almost certainly require an origin in the pre-SN He-shell of massive stars. Additional isotopic measurements of the extreme ^{17}O- and ^{18}O-rich grains should help better constrain their origins.

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