NEGATIVE SECONDARY ION MEASUREMENTS OF ^{54}FE/^{56}FE AND ^{57}FE/^{56}FE IN PRESOLAR SILICATE GRAINS FROM ACFER 094. W. J. Ong, C. Floss, and F. Gyngard. Laboratory for Space Sciences and Physics Department, Washington University, St. Louis, MO, USA (contact email: floss@wustl.edu).

Introduction: Chemical characterization of presolar silicate grains has shown that many have higher than expected Fe contents [1-4]. Although secondary alteration may enhance Fe contents in some presolar silicates [e.g., 5], the pristine nature of most primitive meteorites suggests that much of the Fe is intrinsic to the grains. Fe isotopic measurements can provide confirmation of a primary origin and can help constrain nucleosynthetic processes in evolved stars.

However, presolar silicates are typically identified in meteorite thin sections [1, 2] or densely packed grain size separates [3, 4], and the beam diameter of the O^- primary beam used for Fe isotopic measurements is significantly larger than the sizes of the grains, leading to substantial signal contamination from the surrounding solar grains. Because Fe is a major element in most of these grains, measuring the Fe isotopes with the higher spatial resolution of the Cs^+ primary beam can be a viable alternative, despite the expected lower yields [e.g., 6]. Here we report the results of our NanoSIMS measurements of $^{54}Fe/^{56}Fe$ and $^{57}Fe/^{56}Fe$ ratios in Acfer 094 presolar silicates.

Experimental: Isotopically anomalous grains were initially identified in the NanoSIMS through C and O raster ion imaging [e.g., 1] of a grain size separate (0.1–0.5 µm) of Acfer 094. The grains were subsequently analyzed in the Auger Nanoprobe to determine elemental compositions and to identify suitable candidates for the Fe isotopic analyses.

With a Cs^+ primary ion beam, we measured the Fe isotopes as oxides in grain mode, using two magnetic fields and the following masses: 16 (^{16}O), 68 (^{52}CrO), 70 (^{54}FeO, ^{54}CrO), 73 (^{57}FeO), and 78 (^{62}NiO) in detectors 1–5, followed by 72 (^{56}FeO) and 74 (^{58}FeO, ^{58}NiO) in detectors 2–3. After acquisition of a 10 x 10 µm image of the area of interest, the beam was rastered over individual grains selected from this image. Sizes of the raster boxes varied depending on the grain being measured and ranged from 0.2 to 0.7 µm. The measurements were made at a mass resolution high enough to largely separate $^{56}Fe^{16}OH^-$ from $^{57}Fe^{16}O^-$. As shown in Fig. 1, the contribution from $^{56}Fe^{16}OH^-$ to the $^{57}Fe^{16}O^-$ peak is ~ 1%. Measurements on an FeNi standard show a reproducibility of ~20 ‰ for both $^{57}Fe/^{56}Fe$ and $^{54}Fe/^{56}Fe$. The contribution from the ^{56}FeO hydride peak is greater in the silicate analyses, with similar count rates for both peaks. However, the reproducibility for both ratios is also ~20–30 ‰ for isotopically normal matrix grains in the vicinity of each presolar grain measured, indicating a relatively stable hydride contribution in the different grains. The overall variation for all of the Acfer 094 matrix grains analyzed on the mount is ~40 ‰ for $^{54}Fe/^{56}Fe$ and ~ 80 ‰ for $^{57}Fe/^{56}Fe$ (e.g., Fig. 2).

Corrections were made for ^{54}Cr on ^{54}Fe, based on the measured $^{52}Cr^{16}O^-$, assuming solar Cr isotopic compositions, and were ≤ ~10‰ for all of the grains measured. No data are reported for $^{58}Fe/^{56}Fe$, as the $^{58}Fe^{16}O^-$ signal is dominated by contributions from $^{58}Ni^{16}O^-$ and no meaningful corrections could be made. The data were normalized to the average compositions of the normal matrix grains measured in the vicinity of each presolar grain.

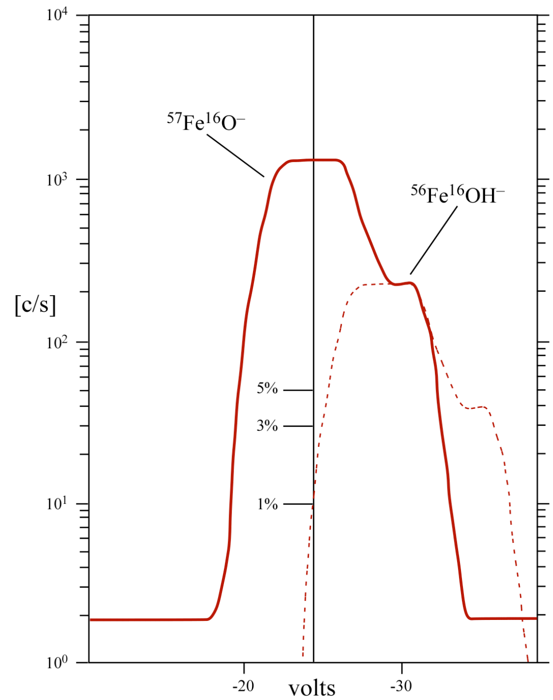

Figure 1. High mass resolution spectrum at mass 73. The dashed line shows an overlay of the $^{57}Fe^{16}O^-$ peak on the $^{56}Fe^{16}OH^-$ shoulder, indicating ~1% contribution of the hydride to the signal.

Results: Seven of the eight presolar grains we measured for Fe show the ^{17}O enrichments and close-to-solar $^{18}O/^{16}O$ ratios typical of group 1 grains [7], while the other grain is enriched in both ^{17}O and ^{18}O and belongs to group 4. One grain is an oxide; the others are silicates. All of the grains are Fe-rich, with Fe contents ranging from 10 to 43 at.%.

Figure 2 shows the Fe isotopic compositions of the grains, along with the compositions of normal matrix grains. Also shown is grain 34C-10, an FeO that also comes from Acfer 094 [8]. Three grains, including the group 4 grain, have normal $^{54}Fe/^{56}Fe$ and $^{57}Fe/^{56}Fe$ ratios. Three other grains, one of which is the oxide grain, have normal $^{54}Fe/^{56}Fe$, but show some evidence of ^{57}Fe depletion. One grain, 16-8, is enriched in both ^{54}Fe and ^{57}Fe ($\delta^{54}Fe = 70 \pm 25$ ‰; $\delta^{57}Fe = 80 \pm 30$ ‰); it is an Fe-rich silicate with Fe+Mg/Si of ~3. Its O isotopes ($^{17}O/^{16}O = 5.5 \pm 0.2 \times 10^{-4}$; $^{18}O/^{16}O = 2.0 \pm 0.04 \times 10^{-3}$) indicate an origin in a low-mass (~1.3 $M_{\odot}$) AGB star of slightly higher than solar metallicity. Finally, grain 2-7 has a normal $^{54}Fe/^{56}Fe$ ratio, but is depleted in ^{57}Fe ($\delta^{57}Fe = -210 \pm 35$ ‰); it is a ferromagnesian silicate compositionally consistent with olivine (Fe+Mg/Si = 19.9 ± 0.3; mg# = 48). It is also a group 1 grain from a low-mass (~1.4 $M_{\odot}$) AGB star of roughly solar metallicity, with $^{17}O/^{16}O = 7.8 \pm 0.2 \times 10^{-4}$ and $^{18}O/^{16}O = 1.9 \pm 0.03 \times 10^{-3}$.

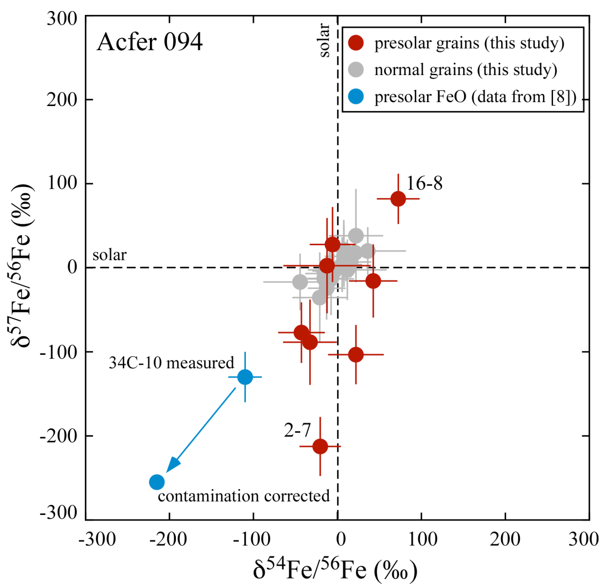

Figure 2. Fe three-isotope plot showing the compositions of presolar and normal matrix grains from Acfer 094. Errors are 1σ.

Discussion: Relatively few presolar silicate (or oxide) grains have been measured for their Fe isotopic compositions and in most cases only $^{54}Fe/^{56}Fe$ ratios have been reported. The majority are group 1 grains and have solar $^{54}Fe/^{56}Fe$ [6, 9], as do most of the grains from this study. This is consistent with expectations that $^{54}Fe/^{56}Fe$ ratios in low-mass AGB stars don't change substantially from the initial values of the parent stars; most mainstream SiC grains also have solar $^{54}Fe/^{56}Fe$ ratios [10]. The $^{54}Fe/^{56}Fe$ ratios of silicates from ~solar metallicity AGB stars are, therefore, not diagnostic for distinguishing between primary and secondary Fe in the grains. However, the anomalous compositions of grains 2-7 and 16-8, along with the marginal ^{57}Fe depletions in three other grains, indicate a primary origin for at least some of the Fe in these presolar silicates.

Grain 16-8 has higher than solar $^{54}Fe/^{56}Fe$ and $^{57}Fe/^{56}Fe$ ratios. Two mainstream SiC grains studied by [10] show similar enrichments in these isotopes. Grains forming in low-mass AGB stars are expected to show s-process enrichments in ^{57}Fe, along with minor depletions in $^{54}Fe/^{56}Fe$ [10]. The ^{57}Fe excess in grain 16-8 is consistent with model predictions for stars of ~solar metallicity [10]. The ^{54}Fe excess is more problematic. One possibility is suggested by model predictions of excesses in ^{54}Cr for low-mass AGB stars [11]. If our assumption of solar Cr isotopic compositions is incorrect, the excess in ^{54}Fe could be due to under-correction of the ^{54}Cr contribution. However, we don't see similar ^{54}Fe excesses in the other group 1 grains we analyzed, suggesting that this explanation is unlikely. Instead, as suggested by [10] for mainstream SiC grains with ^{54}Fe and ^{57}Fe enrichments, the excess may be explained by galactic evolution of the $^{54}Fe/^{56}Fe$ ratio. Enrichments in ^{54}Fe have been observed in two other presolar silicates. One is a group 4 grain with a probable supernova origin [8, 9]. In the other grain [6], no correction was made for ^{54}Cr, leaving the magnitude of any excess in doubt.

Depletions in ^{57}Fe are not easily explained. Grain 2-7 has a probable AGB origin, based on its O isotopic composition, but shows a large deficit in ^{57}Fe. Grain 34C-10 (Fig. 2) is also depleted in ^{57}Fe, as well as ^{54}Fe; its O isotopic composition shows a small excess in ^{18}O [8]. These authors noted that the Fe isotopic composition of this grain could not be easily explained by either a supernova or AGB origin. Similarly, [10] were unable to satisfactorily explain the ^{57}Fe depletions of several mainstream SiC and X grains.

Conclusions: Our results show that both $^{54}Fe/^{56}Fe$ and $^{57}Fe/^{56}Fe$ ratios can be successfully measured as negative secondary ions of Fe oxide. The Fe isotopic compositions of some of the O-anomalous grains studied here, as well as those of some presolar SiC grains, remain difficult to understand in terms of current models of stellar nucleosynthesis.

References: [1] Floss and Stadermann (2009) *GCA* 73, 2415. [2] Vollmer et al. (2009) *GCA* 73, 7127. [3] Bose et al. (2010) *ApJ* 714, 1624. [4] Nguyen et al. (2010) *ApJ* 719, 166. [5] Floss and Stadermann (2012) *MAPS*, submitted. [6] Vollmer and Hoppe (2010) *LPSC XLI*, #1200. [7] Nittler et al. (2008) *ApJ* 682, 1450. [8] Floss et al. (2008) *ApJ* 672, 1266. [9] Mostefaoui and Hoppe (2004) *ApJ* 613, L149. [10] Marhas et al. (2008) *ApJ* 689, 622. [11] Zinner et al. (2005) *GCA* 69, 4149.

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