Fe ISOTOPIC COMPOSITION OF PRESOLAR SiC MAINSTREAM GRAINS. C. E. Tripa¹,², M. J. Pellin², M. R. Savina², A. M. Davis¹³, R. S. Lewis¹, and R. N. Clayton¹³⁴, ¹Enrico Fermi Institute, University of Chicago, 5640 S. Ellis Ave., Chicago, IL 60637 (tripa@anl.gov), ²Materials Science Division, Argonne National Laboratory, 9700 S. Cass Ave., CHM 200, D101-121, Argonne, IL 60439 (pellin@anl.gov, msavina@anl.gov), ³Department of Geophysical Sciences (a-davis@uchicago.edu, r-lewis@uchicago.edu), ⁴Department of Chemistry, University of Chicago (r-clayton@uchicago.edu).

Introduction: The consensus is that mainstream presolar SiC grains originated in low-mass asymptotic giant branch (AGB) stars [1-4], which are the main source for s-process isotopes [5]. However, iron is predominantly formed by high temperature nuclear statistical equilibrium processes and 28Si burning, perhaps in type Ia and II supernovae [6]. An AGB star can only s-process the Fe isotopic composition inherited from previous galactic evolution, and the expected signature is an enrichment in the heavier stable iron isotopes, 57Fe and 58Fe. Up until now, no Fe isotopic composition in SiC grains has been reported because of isobaric interferences from 2Si, 30SiC2, 54Cr, and 58Ni associated with traditional mass-spectrometric methods for trace analysis of solid materials, such as secondary ion mass spectrometry (SIMS) and thermal ionization mass spectrometry. Here, we report the first measurements of Fe isotopes in presolar SiC grains made by time-of-flight resonance ionization mass spectrometry (TOF-RIMS), a technique which eliminates the unwanted isobaric interferences without sacrificing the sensitivity required for trace analysis [1-4].

Experimental: The SiC grains analyzed here were isolated from the Murchison CM chondrite and they all belonged to the 2.1-4.5 µm (KJG) size fraction [7]. The grains were pressed onto a soft pure gold mount, and mapped and characterized by scanning electron microscopy, energy dispersive X-ray microanalysis, and SIMS. Iron isotope measurements were made on the CHARISMA instrument at Argonne National Laboratory, which has been described elsewhere [8-10]. Material from each grain was removed by laser ablation inside an ultra-high vacuum chamber, and the gas-phase Fe was selectively photoionized through resonant electronic transitions with a pair of tunable Ti:sapphire lasers. The resonance scheme used here, a3D4 (ground state) → w5F3 (44,243.7 cm⁻¹) → unassigned autoionizing state (65,950.1 cm⁻¹) above the Fe(I) ionization potential (63,737 cm⁻¹), was a two-photon two-color scheme, 226.021 nm vac and 460.694 nm vac, respectively. The Fe photoions were then extracted into a TOF mass spectrometer and detected with microchannel plates.

Results: A typical Fe TOF-RIMS mass spectrum from a mainstream SiC grain is shown in Fig. 1 (top).

![Fe Photoions from a Mainstream Grain](image)

While the four Fe isotope peaks (54, 56, 57, and 58 amu) are well resolved, very low counts of grouped noise can be seen at non-integer masses, particularly at ~55.5 and ~57.5 amu. Additional experiments, done by blocking either the desorption laser beam or either of the two photoionization laser beams, revealed that the background noise consists primarily of C4Hn hydrocarbon fragments originating from the surface of the sample mount, non-resonantly ionized by the 226.021 nm (UV) laser beam grazing the surface. The hydrocarbons are part of the residual gas background inside the ultrahigh vacuum chamber, continuously adsorbing on the gold mount, and are not intrinsically present in the SiC grains. Figure 1 (bottom) shows the background associated with the mass spectrum shown in

![Background in the Absence of λ=460.694 nm](image)

Fig. 1. Top: Fe resonance ionization mass spectrum from a single SiC mainstream grain, including all the background contributions. Bottom: The nature and magnitude of the background obtained by suppressing the second color ionization beam, 460.694 nm.
the upper panel. The background was produced by laser desorption, followed by non-resonant photoionization with the UV laser. The background proves that: (1) under the desorption laser intensity required to detect Fe in the SiC grains, no Si or SiC is detected, and (2) non-resonant Fe ionization is negligibly small. Subtracting the background contribution from the Fe mass spectra shifted the results by less than 30‰ in δ54Fe, 47‰ in δ56Fe, and 99‰ in δ57Fe. The errors introduced by background subtraction were propagated into the final isotope compositions, but the precision of the measurement, which depends primarily on the total number of counts, is essentially unaffected by subtraction of such small backgrounds.

The Fe isotopic composition in thirteen SiC mainstream grains was measured as described above, and the results are shown in Fig. 2 as δ values in three-isotope plots. All the δ values are normalized to 56Fe according to the formula: δFe[‰] = [(Fe/56Fe)grain / (Fe/56Fe)sample - 1] × 1000. Here, (Fe/56Fe)sample are isotope ratios measured on a piece of stainless steel. For comparison, Fig. 2 also includes theoretical predictions for s-process nucleosynthesis of Fe of 3 solar mass, solar metallicity AGB stars reported from Ref. 11. Compared to solar, these grains show a 50% to 200% depletion in 54Fe, much larger than what is expected from theory (5 to 10‰). 57Fe and 56Fe do not reach the level of enhancement predicted by theory, although there seems to be a weak linear correlation between these two isotopes.

**Discussion:** The solar isotopic distribution of iron isotopes is 5.8% 54Fe, 91.7% 56Fe, 2.2% 57Fe, and 0.3% 58Fe, and the corresponding thermal neutron absorption (n,γ) cross sections are 2.3, 2.6, 2.5, and 1.3 barns, respectively [12]. Iron-54 is the only isotope which is outside the s-process path and cannot be produced by this mechanism in an AGB star. Iron-56 can, however, be synthesized from both nuclides along the valley of stability and 54Fe consumption through the 54Fe → 55Fe(2.7 yr) → 56Fe → 56Mn(2.6 h) → 56Fe sequence. During the lifetime of 55Fe, a small fraction of this isotope may change directly into 56Fe by a second n-capture. For the majority of the mainstream grains measured here, 54Fe is indeed depleted, and 57Fe and 56Fe are enriched, compared to solar abundance, just as is qualitatively expected from the values of the n-capture cross sections. The disagreement rests in the magnitude of the expected δ-value deviations compared to the best available predictions for s-process nucleosynthesis in AGB stars. The simulations shown in Fig. 2 assume AGB stars with solar composition. A possible constraint on the simulation would be to consider a different initial Fe isotope distribution, one in which 54Fe, 57Fe, and 56Fe are all depleted compared to the solar distribution. The basis for this assumption would be the galactic chemical evolution, in which the 54Fe/56Fe would be lower in the early universe than it is today. Thus, the δ54Fe value could be used to estimate the age of mainstream SiC grains.

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**References:**