IRON AND NICKEL ISOTOPE MEASUREMENTS ON SIC X GRAINS WITH CHILI.

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Introduction: X grains, characterized by large depletions in the heavy Si isotopes, are the most abundant supernova (SN) derived presolar SiC type (e.g., [1]). Despite the efforts to study their isotope composition to better understand SN nucleosynthesis, so far only [2] reported the Fe- and Ni isotope compositions of such grains. This is because of the combination of the challenges posed by low Fe concentrations, small sample volumes, and the limited mass resolution of available mass spectrometers. Here we report the results of Fe and Ni isotope measurements of presolar SiC grains of type X, which we obtained using a novel instrument, CHILI [3], which mitigates the above analytical difficulties using resonance ionization.

Experimental: The X grains selected for Fe and Ni isotope measurements were found in SiC grain separates of the Murchison meteorite using C and Si isotope mapping with the NanoSIMS 50 of the Max Planck Institute for Chemistry [4,5]. Isotope mapping was performed on four mounts of two grain separates, KJD [6] (one mount) and Mur2012B ([7]; three mounts). After isotope mapping, X grains were documented by secondary electron imaging, to select grains for analysis with CHILI, based on their size and their proximity to other grains. CHILI measurements were performed at the University of Chicago, following the procedure reported by [3]. The energy of the desorption laser (2–3 µJ per pulse) was adjusted to achieve constant ion count rates in the time-of-flight mass spectrometer. The spot size of the desorption laser and thus the lateral resolution of our Fe-Ni isotope measurements was about 1 µm.

Results and Discussion: We measured the Fe and Ni isotope composition of 19 SiC grains of type X. Only nine grains had anomalous Fe or Ni isotope compositions at the 3σ confidence level, when compared to our artificial SiC reference material of (presumed) terrestrial isotope composition. We found two grains with negative δ^{54}Fe and five with negative δ^{57}Fe values, but no ^{58}Fe anomalies ($\delta^{x}Fe = 1000 \times [(^{x}Fe/^{56}Fe)_{grain}/(^{x}Fe/^{56}Fe)_{reference} - 1]$). Six grains had anomalous Ni isotope composition. Two grains had positive and another two grains had negative δ^{60}Ni ($\delta^{x}Ni = 1000 \times [(^{x}Ni/^{58}Ni)_{grain}/(^{x}Ni/^{58}Ni)_{reference} - 1]$). From the latter two grains, one showed negative δ^{61}Ni, δ^{62}Ni, and δ^{64}Ni as well, which indicates an overall enrichment in the reference isotope ^{58}Ni, but the apparent depletion in ^{64}Ni (>4 σ) is significantly larger than the depletion in ^{60}Ni, so this grain had a complex Ni isotope enrichment/depletion pattern. From the six Ni-anomalous grains, one had terrestrial ^{60}Ni/^{58}Ni but positive δ^{61}Ni, δ^{62}Ni, and δ^{64}Ni values. The anomalous Ni was hosted by a certain region within the otherwise isotopically normal grain. This region showed the largest anomalies in our entire dataset (δ^{61}Ni = 593 ± 58 ‰; δ^{62}Ni = 175 ± 29 ‰; δ^{64}Ni = 422 ± 61 ‰; errors are 1σ). Finally, one Ni-anomalous grain was depleted in ^{62}Ni and ^{64}Ni but showed no ^{60}Ni and ^{61}Ni anomalies. This was the only grain which had significant isotope anomalies in Ni as well as in Fe (its δ^{57}Fe was negative).

Grains with Fe and/or Ni isotope anomalies are rarer among our X grains than found previously by [2], and the observed isotope anomalies of our grains are overall smaller than those reported by the same authors. We believe that this is partly owing to contamination of our sample mount by Fe- and Ni-bearing phase(s), possibly at very small scales. Contamination is indicated by the high inferred Fe concentrations (median Fe/Si = 740×10^{-6}) and high Fe/Ni ratio (median Fe/Ni =3.7) of our X grains relative to those observed by [2] (214 and 0.35, respectively).

Because of the contamination discussed above, a quantitative reproduction of the observed *isotope anomalies* using SN models is of limited use. Instead, we tried to reproduce the observed *isotope patterns* using the SN models of [8], but so far, we have not been able to find models that predict the observed isotope patterns by condensation of material from single SN zones or their mixture, if the grains' C and Si isotope compositions are maintained and a C/O ratio of the condensing gas of >1 is assumed. This suggests element fractionation in the condensing gas, which removed some of the Fe and Ni from the expanding gas prior to SiC condensation.

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