

THE RISE AND FALL OF IRON-60.

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Introduction: Iron-60 is a uniquely important short-lived radionuclide because it cannot be produced by particle irradiation, and thus, high initial ⁶⁰Fe abundances inferred in early-formed Solar System objects must be attributed to injection from a stellar source. Estimates of the initial ⁶⁰Fe/⁵⁶Fe ratio range over a factor of 60 from 1×10^{-8} as determined by bulk measurements [1] to $\sim 6 \times 10^{-7}$ from in-situ measurements of high Fe/Ni phases [2,3]. The reasons for this significant discrepancy between high-precision bulk analyses and in-situ analyses of high Fe/Ni phases are not clear. It is possible that SIMS (secondary ion mass spectrometry) analyses are biased by unrecognized, low-level interferences on ⁶⁰Ni. Resonance ionization mass spectrometry (RIMS) can be used to overcome these problems.

Methods: To investigate this, we used the UCLA IMS 1290 ion microprobe to analyze Orgueil carbonate grains that exhibit very high Fe/Ni ratios ($>10^6$) and are known from ⁵³Mn-⁵³Cr systematics to have formed within ~ 5 Myr of CAIs [4]. The analyses were made at high mass resolving power (MRP >6000) to separate known molecular ion interferences. It is notable, however, that there is no accessible MRP that can resolve the isobars ⁵⁸Fe and ⁶⁴Zn from corresponding Ni isotopes. Because ⁶¹Ni is a very low abundance isotope, poor counting statistics preclude precise correction for mass fractionation, and therefore, SIMS analyses effectively rely on only one isotope ratio, ⁶⁰Ni/⁶²Ni, as a measure of the potential abundance of radiogenic ⁶⁰Ni*.

To test the accuracy of the SIMS Ni isotope measurements, we analyzed the same carbonate grains with RIMS using CHILI, the CHicago Instrument for Laser Ionization [5]. CHILI was configured to ionize Ni resonantly using a four-color scheme. Iron was ionized nonresonantly by the Ni lasers. By not using a Fe RIMS scheme, the Fe sensitivity was diminished relative to the Ni sensitivity, which resulted in Fe and Ni signals being of roughly equal intensity during analysis of the high-Fe/Ni samples.

Results: Our preliminary SIMS investigations found two Orgueil breunnerite grains that show apparent ⁶⁰Ni*. Correlations with Fe/Ni are generally not well defined, but the data imply a relatively high ⁶⁰Fe/⁵⁶Fe ratio, in the range of $\sim 10^{-7}$, at the time of carbonate formation.

The same grains with apparent ⁶⁰Ni* were analyzed by CHILI. Because of the very high Fe/Ni ratios in the carbonates, Fe peaks are present in most mass spectra, although at low enough levels to permit accurate peak-stripping of ⁵⁸Fe. This correction was smaller than 5‰ (of the total mass 58 peak), except in two analysis spots where it was 8 and 12‰. The data obtained on the Orgueil carbonates reveal correlations between ⁶²Ni/⁵⁸Ni and ⁶⁰Ni/⁵⁸Ni that are strictly mass-dependent. The range in mass fractionation is very large, ~ 25 to 30‰/u favoring the light isotopes, whereas the range of mass fractionation effects in terrestrial siderite and San Carlos olivine standards is -2.5 to $+2.5$ ‰/u. The large mass fractionation effects in Orgueil carbonates are made evident only by the ability to measure ⁵⁸Ni with CHILI, even in phases with very high Fe/Ni ratios. In SIMS analyses, where mass fractionation of Ni isotopes cannot be independently determined with good precision, isotopically light Ni, with high ⁶⁰Ni/⁶²Ni ratio, can be misinterpreted as addition of radiogenic ⁶⁰Ni*. Our RIMS measurements conclusively demonstrate the occurrence of mass-fractionated Ni in meteorite samples, which is large enough that elevated ⁶⁰Ni/⁶²Ni ratios could be incorrectly attributed to ⁶⁰Fe decay if this mass fractionation is not accurately accounted for. We are not yet able to place a limit on the early Solar System ⁶⁰Fe/⁵⁶Fe ratio because we cannot reliably measure Fe/Ni ratios with the current RIMS analyses, but it is clear that no excess ⁶⁰Ni* has been detected in old high-Fe/Ni carbonates from Orgueil.

References: [1] Tang H. and Dauphas N. (2015) *The Astrophysical Journal* 802:22 (9pp). [2] Mishra R. and Chaussidon M. (2014) *Earth and Planetary Science Letters* 398:90–100. [3] Telus M. et al. (2012) *Meteoritics & Planetary Science* 47:2013–2030. [4] Steele R. et al. (2017) *Geochim. Cosmochim. Acta* 201: 245-259. [5] Stephan T. et al. (2016) *International Journal of Mass Spectrometry* 407:1–15. LLNL-ABS-730725