

INSIGHTS INTO CAI FORMATION FROM NICKEL ISOTOPIC SIGNATURES

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Introduction: Information about the earliest stages of our stellar system can be derived from Ca-Al-rich inclusions (CAIs), which represent the first solids that condensed in the cooling solar nebula [1-2]. Despite extensive investigations of CAIs, significant questions regarding their formation remain unanswered. Nucleosynthetic isotope signatures, which serve as distinct “fingerprints” of genetic heritage, can be used to place important constraints on the CAI formation region(s).

Earlier work has shown that normal (non-FUN) CAIs of various types, mineralogies, REE patterns, and host chondrites exhibit broadly uniform nucleosynthetic isotope anomalies for lithophile elements such as Ti, Ba, Sr, Zr, Hf, Cr, and REEs [3], hinting at formation in a largely isotopically homogeneous reservoir. In contrast, substantial isotopic variability indicative of different nucleosynthetic characters exists between fine- and coarse-grained inclusions in the refractory siderophile elements Mo [4-5] and W [6], complicating the previous interpretation. It is unknown whether the variability in these two minor elements results from additional carriers or if this is evidence of multiple CAI forming events that were separated by time and/or space.

As a major element with five isotopes and similarly siderophile behavior, nickel (Ni) can be used to shed light on the variations seen for Mo and W. Previous cosmochemical studies that reported Ni isotopic compositions of CAIs had limited sample sets and comparatively low precision at that time [7] or were unable to report data for the low-abundance, neutron-rich isotope ⁶⁴Ni [8], which suffers from severe isobaric interferences from Zn. In this work, we investigate full Ni isotopic compositions of a multitude of CAIs in an attempt to better understand CAI provenance.

Samples and Methods: A broad set of >25 CAI samples from different host carbonaceous chondrites and of different petrologic types were selected for this study. Being part of an integrated study to obtain information about multiple elements [6,9-11], this sample set had previously undergone ion-exchange chemistry for extraction of other elements. To ensure that the previous chemical processing did not affect our data, so-called ‘nochem’ fractions of some samples—which had not seen any prior chemistry—were measured alongside this sample set for comparison. Ni was separated from the sample matrix with a newly developed ion-exchange procedure employing both cation and anion exchange resins and reagents including acetone-HCl and acetic acid-HCl based on a procedure from [12]. To verify the accuracy of our method we processed terrestrial basalt standards. Isotope measurements were performed using the ThermoScientific Neptune *Plus* MC-ICPMS at the University of Münster. Samples and standards were internally normalized to ⁶¹Ni/⁵⁸Ni = 0.016744 [13] to correct for mass bias during measurement.

Results and Discussion: The analyzed terrestrial basalt standards show Ni isotopic compositions within uncertainty to our bracketing standard SRM986 ($\epsilon^i\text{Ni}=0$). The isotopic compositions of ‘nochem’ aliquots are in agreement with previously processed fractions of the same inclusions. The CAI samples analyzed in this study show significant variations in Ni isotopic compositions, ranging from -0.3 to 0.8 in $\epsilon^{60}\text{Ni}$, from -0.4 to 1.4 in $\epsilon^{62}\text{Ni}$ and from -1.2 to 3.3 in $\epsilon^{64}\text{Ni}$. In contrast to Mo and W, the Ni nucleosynthetic anomalies of fine-grained CAIs are generally smaller than those of coarse-grained inclusions. Furthermore, the isotopic variations observed in Ni are of much smaller magnitudes than those seen in Mo and W and do not show evidence for distinct nucleosynthetic characters, as seen for Mo.

The observed variations in Ni isotopes are correlated with one another and form a fairly continuous range. We interpret this range to be a mixing line between the indigenous Ni isotopic composition of CAIs with larger $\epsilon^i\text{Ni}$ excesses, and a second component with slight deficits in all three Ni isotopes relative to terrestrial standards. This interpretation is strengthened by a correlation of the nucleosynthetic isotope data with mass-dependent fractionations for the same sample set reported in [14]. Since the mixing line does not intersect $\epsilon^i\text{Ni}=0$, it is not a result of mixing CAIs with terrestrial material. Instead, the variation could represent evidence for (1) at least two CAI reservoirs with distinct (both mass-dependent and mass-independent) Ni isotopic compositions that were later mixed, or (2) alteration on the parent body that lead to different levels of partial equilibration between the original Ni isotopic composition of CAIs and Ni of the later-formed host meteorite. This possibility is bolstered by the fact that the mixing line intersects the Ni isotopic signature of bulk CV chondrites as defined by measurements of Allende and Leoville from [15].

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