BARIUM, NEODYMIUM, & SAMARIUM ISOTOPIC COMPOSITION OF CAIs: NUCLEOSYNTHETIC ANOMALIES?

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Introduction: Non mass-dependent variations in the stable, non-radiogenic isotope compositions of refractory inclusions are commonly interpreted as nucleosynthetic signatures resulting from distinct inputs of materials produced as a result of p-, s-, and r-processes. Ba, Nd, and Sm each have multiple stable isotopes with variable inputs from p-, s-, and r-process nucleosynthesis, making these elements ideal for an integrated study of nucleosynthetic signatures in meteoritic material.

Previously reported data from eleven calcium-aluminum-rich inclusions (CAIs) of the Allende meteorite show apparent r-process *excesses* in isotopes of Ba and apparent p- and r-process *deficits* in isotopes of Nd and Sm in the same samples [1]. These data also suggest that CAIs formed from an isotopically uniform reservoir that was distinct from that from which the less refractory chondritic phases were formed. Data from [1] are shown in Figs. 1-3 (black lines and symbols). The details regarding these data, including analytical methods, and comparisons to previously reported results are discussed in [1]. Here we discuss one possible explanation for these, and other, reported isotope compositions of CAIs.

Methods: The existence of apparent r-process excesses in one isotope system (Ba) recorded along with apparent r-process deficits in other isotope systems (Nd and Sm) in the same CAIs requires a plausible explanation. This work expands on work by [2, 3] exploring the possibility that isotope anomalies in CAIs may instead be due to mass-independent isotope fractionation induced by nuclear field shift.

Experimental studies for multiple isotopes systems have confirmed the existence of mass-independent fractionation as predicted by the nuclear field shift effect [3, refs therein], and these fractionation patterns can be predicted based on differences in the mean-squared nuclear charge radii between isotopes. The interaction of CAIs with the nebular gas during the early stages of condensation is one possible location in which nuclear field shift fractionation could have occurred in these samples [3]. The predicted isotope patterns for multiple elements based on the nuclear field shift are calculated here and compared to measured results. Mass-independent isotope effects are calculated in ε units (parts per 10^4) using equation 1, as follows (for full derivation see [2]):

$$\varepsilon_{mi} = \left(\delta\langle r^2 \rangle_{m1,mi} - \frac{m_2(m_i - m_1)}{m_i(m_2 - m_1)} \times \delta\langle r^2 \rangle_{m1,m2} \right) \times a$$

In this equation, m_1 and m_2 are the atomic masses of the nuclides chosen for internal normalization, and m_i represents the atomic mass of a nuclide indexed with the variable "i". Differences in mean-squared nuclear charge radii, $\delta\langle r^2 \rangle$, of each isotope pair are taken from [4-6]. The variable "a" is an adjustable parameter proportional to the inverse of the temperature T (i.e., 1/T) and represents the overall extent of mass-independent fractionation [2].

Results: Data for the stable, non-radiogenic isotope compositions of Ba, Nd and Sm from [1] and of Mo [7] for Allende CAIs are shown in Figs. 1-4 (black lines and symbols), along with isotope fractionation patterns predicted by the nuclear field shift effect at an optimized value of "a" for each element (red lines and symbols). The uncertainties on the data shown in Figs. 1-4 are 2SD of the population of measured CAIs. Uncertainties on the calculated isotope compositions represent the ranges obtained by using the range of nuclear charge radii values published in the literature [4-6].
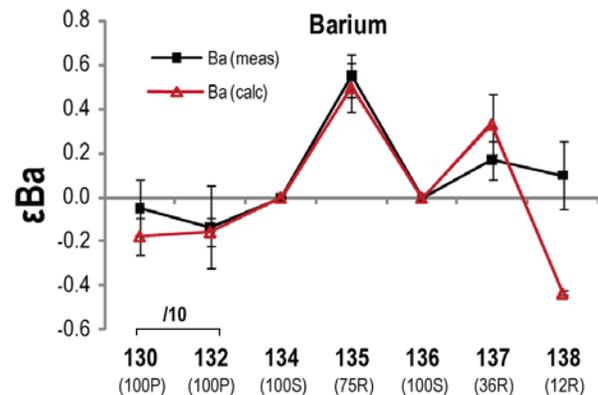

Fig. 1 – The average Ba isotopic composition of Allende CAIs [1] (solid black line and black symbols). The amount of ^{138}Ba ingrowth estimated from the decay of ^{138}La (as discussed in [8]) has been subtracted. The calculated isotope composition based on the nuclear field shift effect is also shown (solid red line and red symbols). The best-fit to the data for Ba is found using $a = -15$ in eq. 1. In this and following figures, the main nucleosynthetic process and the percentage proportion that it represents, is shown in parenthesis below each isotope.
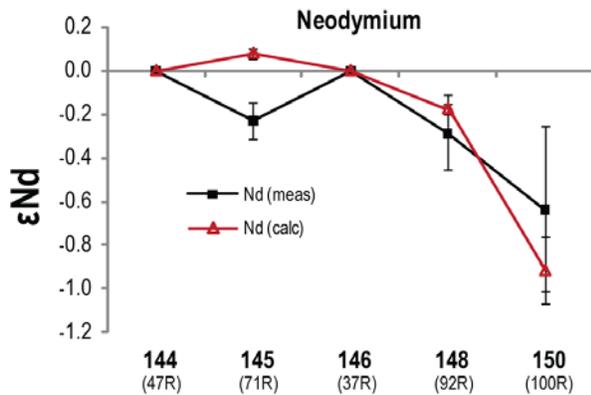
Fig. 2 – The average Nd isotopic composition of Allende CAIs [1] (solid black line and black symbols). The calculated isotope composition based on the nuclear field shift effect is also shown (solid red line and red symbols) The best-fit to the data for Nd is found using $a = -5$ in eq. 1.
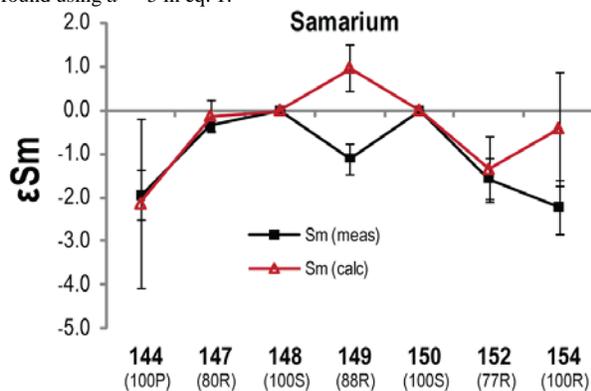

Fig. 3 – The average Sm isotopic composition of Allende CAIs [1] (solid black line and black symbols). The calculated isotope composition based on the nuclear field shift effect is also shown (solid red line and red symbols). The best-fit to the data for Sm is found using $a = -13$ in eq. 1.
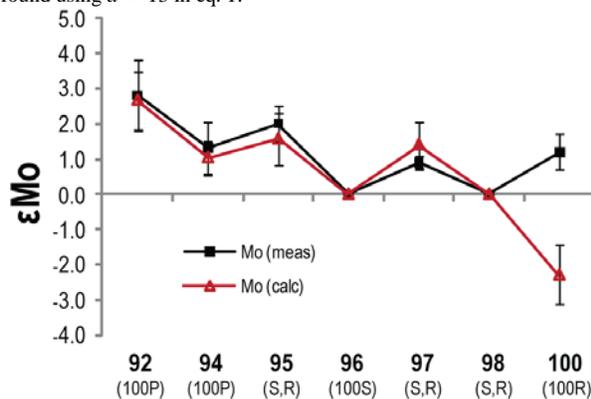

Fig. 4 – The average Mo isotopic composition of Allende CAIs [7] (solid black line and black symbols). The calculated isotope composition based on the nuclear field shift effect is also shown (solid red line and red symbols). The best-fit to the data for Mo is found using $a = -23$ in eq. 1.

Discussion and Conclusions: With the exception of a few isotopes, the measured isotopic compositions

for many elements in Allende CAIs can be explained by fractionation resulting from nuclear field effects. The mismatches for a few isotopes, however, preclude this being the sole cause for the isotopic anomalies in CAIs. For instance, the [135]Ba and [137]Ba peaks in the measured data are reproduced fairly well by the calculated isotope compositions by optimizing "*a*", but fractionation caused by nuclear field shift effects predicts a large negative anomaly in [138]Ba not present in the CAI data. Similarly, large differences exist between the measured and predicted compositions of [145]Nd, as well as [149]Sm and [154]Sm. The measured Mo isotope pattern of Allende CAIs [7] is similar to the predicted pattern (Fig. 4), with the exception of [100]Mo, which is in disagreement by approximately 3 ε units. The measured Te isotope composition [9] can be reproduced within error for all reported isotopes [10] (not shown).

While the measured isotopic compositions are not entirely consistent with the calculated mass-independent fractionations from the nuclear field shift, the overall agreement across multiple isotope systems strongly suggests that this mechanism may play a significant role in explaining some, if not all, of the compositions of stable, non-radiogenic isotopes in CAIs.

The differences in the isotopic compositions of a variety of elements in CAIs on the one hand, and bulk chondrites on the other, may suggest that CAIs and the less refractory phases in chondritic meteorites formed from two distinct reservoirs. If this was the case, it is unclear if these reservoirs were separated by 1) space (i.e., different regions of the protoplanetary disk with distinct compositions), 2) time (i.e., a temporal evolution in the isotopic composition of the disk), or 3) a combination of the above. However, if nuclear field shift fractionation in the early Solar System was responsible for the composition of stable, non-radiogenic isotopes in CAIs, isotopically distinct reservoirs would no longer be required to explain the isotopic differences between CAIs and bulk chondrites.

References**:** [1] Brennecka et al. (2011) *LPSC*, #1302. [2] Fujii et al. (2006) *EPSL*, 247, 1. [3] Moynier et al. (2009) *MAPS*, 44, 1735. [4] Heilig & Steudel (1974) *Atom Data Nucl Data*, 14, 613. [5] Aufmuth et al. (1984) *Atom Data Nucl Data*, 37, 455. [6] Angeli (2004) *Atom Data Nucl Data*, 87, 185. [7] Burkhardt et al. (2011) *LPSC*, #2554. [8] Brennecka et al. (2010) *MetSoc Abs.* #5318. [9] Fehr et al. (2009) *MAPS*, 44, 971. [10] Moynier et al. (2009) *MAPS*, 44, 1735.