COMBINED STABLE ISOTOPE SIGNATURES IN ALLENDE CAIs: THE NUCLEOSYNTHETIC CONUNDRUM.
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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. Elements such as Ca, Ti, Cr, Sr, Zr, Mo, Te, Ba, Nd, and Sm all 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 materials.

Numerous recent studies of calcium-aluminum-rich inclusions (CAIs) have revealed isotopic signatures that differ from the terrestrial standard for a number of elements. More specifically, studies of CAIs have revealed apparent: *p*-process excesses in Sr [1-3], *r*-process excesses in Zr [4], *r*-process excesses in Mo [5], *r*-process excesses in Ba [6, 7], *r*-process depletions in Nd, and *p*- and *r*-process depletions in Sm [6]. In one particular case where multiple elements were investigated in Allende CAIs, apparent excesses and depletions in the *r*-process isotopes of different elements occur in the exact same samples [6].

This expanding data set continues to demonstrate the presence of non mass-dependent isotopic anomalies in "normal" CAIs for virtually every isotope system investigated. However, understanding how and why these apparent nucleosynthetic anomalies exist (and in some cases contradict one another) requires a comprehensive isotopic study of multiple element systems on the same meteoritic materials. In this study, we present Sr and Mo isotope compositions of eleven CAIs from the Allende meteorite for which we have previously reported Ba, Nd, Sm, and U isotope compositions [6, 8].

Samples and Methods: Aliquots from previously dissolved CAIs of the Allende meteorite (from which U was separated, as described in [8] and Ba, Nd, and Sm were separated, as described in [6]) were investigated for Sr and Mo isotope compositions. This sample set includes fine- and coarse-grained CAIs, including three group II inclusions [8].

Element separation. Sr was separated from the CAI matrix using Eichrom Sr Spec resin and a procedure similar to [1]. Mo was separated from the CAI matrix following a procedure described by [9]. Additional separation of Zr and Ru, which have isobaric interferences with multiple isotopes of Mo, was required and was completed using modified procedures outlined in [5].

Isotopic measurements. Sr isotope measurements were made on the Triton TIMS at LLNL. For each sample and standard analysis, approximately 500 ng of Sr was loaded onto a Re filament using Ta_2O_5 activator. The data were corrected for instrumental mass bias using $^{88}Sr/^{86}Sr$=8.375209 [1]. External reproducibility (2SD) for the $^{84}Sr/^{86}Sr$ ratio based on repeated analyses the terrestrial standards (NBS SRM-987 and BCR-2) is ±0.4 ε.

Mo isotope compositions were measured using a Thermo Neptune MC-ICPMS housed in the Isotope Cosmochemistry and Geochronology Laboratory (ICGL) at ASU. All stable isotopes of Mo ($^{92, 94, 95, 96, 97, 98, 100}$Mo) were measured on faraday cups during a single static run with ^{96}Mo beam intensities of ~1-2×10^{-11} A. The data were corrected for instrumental mass bias using $^{98}Mo/^{96}Mo$=1.453171 [10]. Minor contributions from isobaric inferences of $^{92, 94, 96}$Zr and $^{96, 98, 100}$Ru were corrected by monitoring ^{91}Zr and ^{99}Ru during each run. Repeat analyses of natural and synthetic standard materials were used to establish the precision of the Mo isotope measurements, resulting in an external reproducibility (2SD) of ±0.74 for ε^{92}Mo, ±0.39 for ε^{94}Mo, ±0.36 for ε^{95}Mo, ±0.28 for ε^{97}Mo and ±0.39 for ε^{100}Mo.

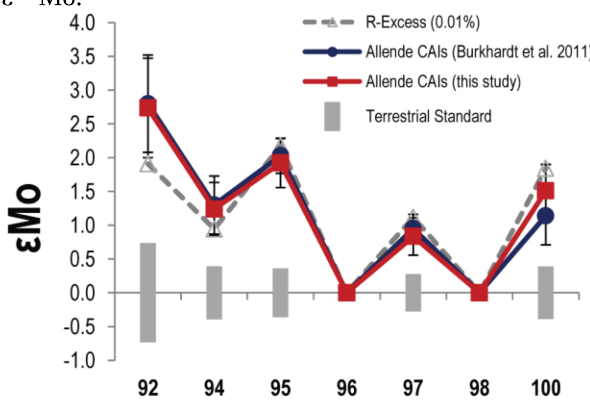

Fig. 1 – A comparison of the Mo isotopic composition of Allende CAIs reported in Burkhardt et al. (2011) (in blue) and this study (red). The gray dashed line represents the expected isotope pattern calculated with an *r*-excess of 0.01%. Reproducibility (2SD) of terrestrial standards for our data set is shown by the solid gray bars.

Results: The eleven CAIs measured in this study have uniform Sr and Mo isotope compositions that are distinct from the bulk silicate Earth (BSE) composition, as represented by terrestrial standards. These

CAIs show an average excess of 1.3±0.5 ε in $^{84}Sr/^{86}Sr$, which is in agreement with previous studies [1-3]. For Mo, CAIs show an average excess of 2.7±0.4 in $\varepsilon^{92}Mo$, 1.2±0.4 in $\varepsilon^{94}Mo$, 1.9±0.2 in $\varepsilon^{95}Mo$, 0.8±0.1 in $\varepsilon^{97}Mo$, and 1.5±0.7 in $\varepsilon^{100}Mo$ (Fig. 1). This isotopic pattern is in excellent agreement with that reported in [5].

Discussion and Conclusions: The Sr and Mo isotope data presented in this study are in excellent agreement with data reported previously for Allende CAIs [1-3, 5] and further suggest that normal CAIs formed in a region of the solar nebula with a homogeneous isotope composition. The distinct isotopic signatures in CAIs and bulk chondrites (representative of the "bulk" Solar System composition) indicate that CAIs originated from a different reservoir than less refractory phases in chondrites. These reservoirs could be 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.

As discussed previously [5], a scenario with relative excess *r*-process material in the CAI reservoir can be used to explain the Mo isotope pattern of CAIs ([5] and this study). However, this model cannot account for the isotopic compositions of other elements measured in the same CAIs. The same Allende CAIs with apparent *p*-excesses in Sr and *r*-excesses in Mo [this study] also contain apparent *r*-process deficits for Nd and *p*- and *r*-process deficits for Sm [6]. Consequently, an *r*-excess model that fits well for Mo isotopes produces gross mismatches in the isotopes of Nd and Sm (Figs. 2 and 3). Thus, explanation of the isotopic signatures of normal CAIs requires a more complicated scenario beyond simple *r*-process addition.

If addition of *r*-process material is responsible for the distinct isotopic composition of the CAI source reservoir in the solar nebula, a decoupling of the *r*-process must exist, presumably around A≈140 (i.e., elements lighter or heavier than Ba). This has been suggested by [11, 12] and the data reported in [6] appear to support it. In this scenario, input from supernovae is characterized by *r*-process excesses for A<140 and *r*-process deficits for A>140. Alternatively, the isotopic patterns of CAIs might not be representative of the region in which they formed and instead could have been modified by nuclear field shift fractionation [13, 14], and/or magnetic isotope fractionation [15]. Unfortunately, these processes cannot fully account for all of the observed isotopic compositions either.

Thus, no single process (i.e., nucleosynthetic addition/subtraction, nuclear field shift, or magnetic isotope effect) can account for the isotopic patterns seen in CAIs. As a result, a complex scenario of injection to the protoplanetary disk, a decoupling of the *r*-process, or a combination of multiple fractionation processes seems to be required to explain all the data.

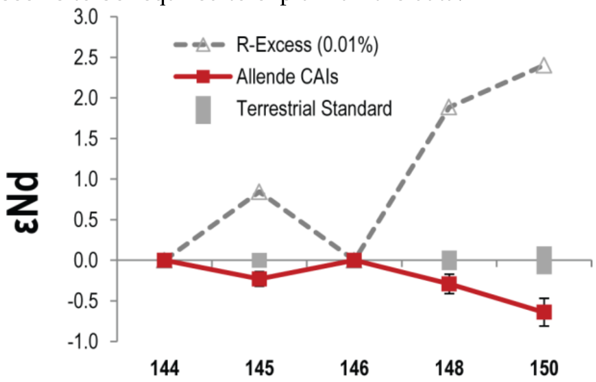

Fig. 2 – Comparison of the Nd isotopic composition of Allende CAIs reported in [6] from the same samples as this study with the predicted isotope pattern of *r*-process excess for Nd. The gray dashed line represents the expected isotope pattern calculated with an *r*-excess of 0.01%. Reproducibility (2SD) of terrestrial standards for this data set is shown by the solid gray bars.

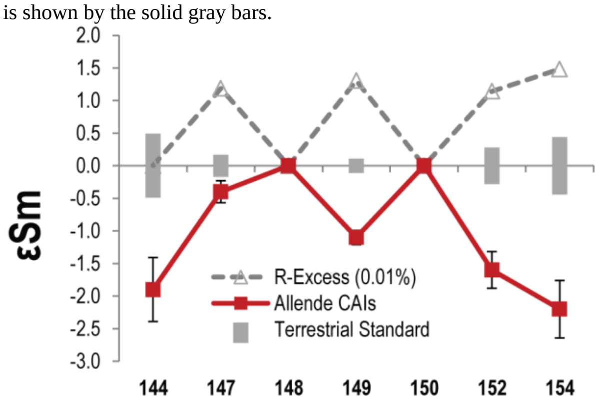

Fig. 3 – Comparison of the Sm isotopic composition of Allende CAIs reported in [6] from the same samples as this study with the predicted isotope pattern of *r*-process excess for Sm. The gray dashed line represents the expected isotope pattern calculated with an *r*-excess of 0.01%. Reproducibility (2SD) of terrestrial standards for this data set is shown by the solid gray bars.

References: [1] Moynier et al. (2011) *LPS XLII*, #1239. [2] Hans et al. (2011) *LPS XLII*, #2672. [3] Patton et al. (2011) *Workshop on the Formation of the First Solids*, #9062. [4] Schönbächler et al. (2011) *Workshop on the Formation of the First Solids*, #9085. [5] Burkhardt et al. (2011) *EPSL*, *312*, 390. [6] Brennecka et al. (2011) *LPS XLII*, #1302. [7] Bermingham et al. (2010) *LPS XLI*, #1735 [8] Brennecka et al. (2010) *Science*, *327*, 449. [9] Duan et al. (2010) *GCA*, *74*, 6655. [10] Lu & Masuda (1994) *IJMS & Ion Processes*, *130*, 65. [11] Qian et al. (1998) *ApJ*. *494*, 285. [12] Nittler & Dauphas (2006) *Meteorites & the Early Solar System II*, p.127. [13] Fujii et al. (2006) EPSL, *247*, 1. [14] Brennecka et al. (2011) *Workshop on the Formation of the First Solids*, #9036. [15] Moynier et al. (2006) *GCA*, *70*, 4287. This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.