

Mg AND U ISOTOPIC SYSTEMATICS IN ALLENDE CAIs: IMPLICATIONS FOR THE ORIGIN OF URANIUM ISOTOPIC VARIATION IN REFRACTORY INCLUSIONS. P. Mane¹, G. A. Brennecka², S. J. Romaniello¹ and M. Wadhwa¹, ¹School of Earth and Space Exploration, Arizona State University, Tempe, AZ 85287. (E-mail: Prajka.Mane@asu.edu). ²Lawrence Livermore National Laboratory, Livermore, CA 94550.

Introduction: Calcium-aluminum-rich inclusions (CAIs) are considered to be the first formed solids in the Solar System [1]. Determining absolute ages of CAIs using long-lived radiochronometers such as Pb-Pb, and relative ages using short-lived radiochronometers such as ²⁶Al-²⁶Mg, is important for establishing the time sequence of early Solar System events. The long lived Pb-Pb chronometer has been utilized extensively for precise absolute dating of early Solar System objects (e.g., [2-4]). In earlier studies, this method assumed a constant ratio of ²³⁸U/²³⁵U (=137.88). However, recent studies have shown that the U isotope composition varies in early Solar System materials, particularly in CAIs [5-7]. In Allende CAIs, the U isotopic variation was attributed to the decay of short lived ²⁴⁷Cm to ²³⁵U (t_{1/2} ~15.6 Ma) based on the observed correlation of the ²³⁸U/²³⁵U ratios with the Th/U or Nd/U ratios (since Nd and Th are anticipated to have similar geochemical behavior to Cm during nebular fractionation processes) [5].

Other possibilities, such as the presence of nucleosynthetic anomalies or isotopic fractionation (during evaporation/condensation or secondary alteration processes) in the early Solar System are also plausible explanations for the variation in U isotopes in CAIs [4, 6, 7]. However, because U has only two primordial isotopes, it is difficult to distinguish between these processes. To evaluate whether isotopic fractionation during the thermal processing of CAIs is a primary driver for U isotopic variations in the CAIs, we analyzed the Mg isotopic composition of 11 refractory inclusions from the Allende CV3 carbonaceous chondrite that were previously measured for U isotopes [5]. If isotopic fractionation due to thermal evaporation or condensation was the primary cause of the U isotope variation in these CAIs, this may be expected to result in a correlation of the ²³⁸U/²³⁵U and ²⁵Mg/²⁴Mg ratios in these samples, thereby providing clues to the origin of U isotopic variation in CAIs.

Analytical Methods: The Mg isotopic compositions of 11 Allende CAIs (for which REE abundances and U isotope compositions were previously reported [5]) have been investigated here. This sample set included both coarse- and fine-grained inclusions. The detailed analytical protocols for our Mg isotope analyses have been described in [9].

A ~5% aliquot of each dissolved CAI sample (equivalent to ~6-16 µg Mg) was passed through a cation exchange column (packed with AG x8 200-400

mesh resin), and Mg was eluted in 1N HNO₃. This column procedure was repeated 3 times for each sample to assure complete separation of Mg from Al, Ca, and other interfering cations, and ensured recovery of >98% of the Mg for each sample. Allende whole rock (WR) and the terrestrial basalt standard BCR-2 were also processed through the same column chemistry procedure and analyzed along with the CAIs to verify the accuracy and precision of our analytical protocols.

Magnesium isotopic analysis was performed on the Thermo-Finnigan Neptune multicollector inductively coupled plasma mass spectrometer (MC-ICP-MS) in the Isotope Cosmochemistry and Geochronology Lab (ICGL) at Arizona State University (ASU). The purified Mg samples were introduced into the mass spectrometer with a flow rate of 100 µl/min using an ESI APEX[®] dissolving nebulizer system. The mass spectrometer was operated in medium resolution mode. The instrumental mass bias was corrected using the sample-standard bracketing method, where DSM-3 was used as a bracketing standard. Each measurement cycle consisted of 20 integrations of 8 seconds each. Each measurement was repeated 3 times. The errors reported here are 2 × standard deviation of the 3 repeat measurements of every sample.

Table 1: Mg isotopic compositions of 11 CAIs analyzed here. U isotope composition are from [5], and are corrected for the updated SRM950a standard value (²³⁸U/²³⁵U = 137.837 [10]).

<i>Sample</i>	$\delta^{25}\text{Mg}$	<i>2SD</i>	$\delta^{238}\text{U}$	<i>2SD</i>
Allende	-0.15	0.09	-0.45	0.11
BRC-2	-0.05	0.07	-0.27	0.19
CAI 164	3.32	0.09	-0.30	0.12
CAI 165	4.13	0.06	-0.33	0.11
CAI 166	-0.50	0.05	-3.42	0.28
CAI 167	-0.99	0.05	-1.76	0.30
CAI 168	4.80	0.06	0.04	0.11
CAI 170	3.54	0.06	-0.51	0.28
CAI 171	3.04	0.04	-0.56	0.22
CAI 172	4.02	0.06	-0.31	0.28
CAI 173	-0.54	0.05	-0.61	0.28
CAI 174	4.35	0.07	-0.23	0.11
CAI 175	-1.28	0.09	-1.45	0.22

Results and Discussion: The measured ²⁵Mg/²⁴Mg ratios (expressed relative to the DSM3 standard in parts per mil, or δ²⁵Mg) of 11 Allende CAIs, Allende WR, and BCR-2 are reported in Table 1. Also presented in Table 1 are the previously reported U isotope

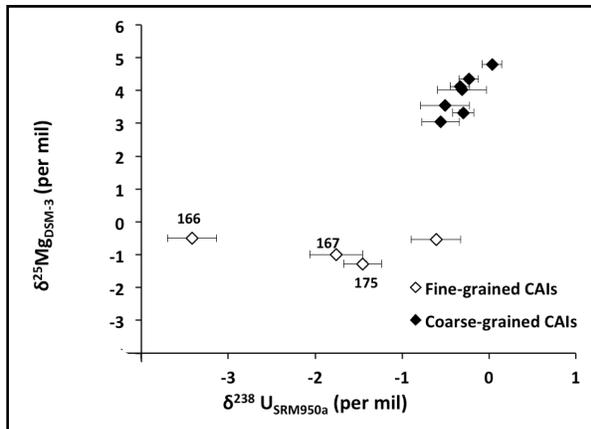


Figure 1: $\delta^{25}\text{Mg}$ and $\delta^{238}\text{U}$ of the 11 CAIs from Allende. The open symbols represent fine-grained CAIs whereas the filled symbols represent coarse-grained CAIs. Group II CAIs have been identified with their numbers next to them. The U isotope composition is from [5].

compositions of each of these samples from [5] (but corrected for the updated U isotope composition of the SRM950a standard [10]). The $\delta^{25}\text{Mg}$ values for the 11 Allende CAIs show considerable variation. Figure 1 shows the U isotope compositions of these CAIs versus their $\delta^{25}\text{Mg}$ values.

It has recently been suggested that isotopic fractionation during CAI-forming processes may have contributed to some, if not all, of the U isotope variation in CAIs [5, 7]. As can be seen in Fig. 1, coarse-grained Allende CAIs (164, 165, 168, 170, 171, 172 and 174), which have undergone melting and thermal processing, show a possible weak correlation between $\delta^{238}\text{U}$ and $\delta^{25}\text{Mg}$. This may suggest that isotopic fractionation during thermal processing of these samples may have indeed contributed to the (relatively small) U isotope variation in this subset of samples.

On the other hand, fine-grained Allende CAIs (166, 167, 173, and 175), which did not undergo melting, are characterized by relatively light Mg isotope compositions (i.e., $\delta^{25}\text{Mg}$ values between -0.5 and -1.5‰). These CAIs show a much larger range of U isotopic compositions than the coarse-grained CAIs that do not correlate with $\delta^{25}\text{Mg}$, suggesting that the U isotope variation in these samples may be dominated either by ^{247}Cm decay or by the presence of nucleosynthetic anomalies. Previous work on the same sample set for the isotopic systems of Sr, Mo, Ba, Nd, Sm, Gd, and Dy show evidence for a CAI source reservoir in the solar nebula that was homogenous (at the level of precision of these analyses), yet distinct from the terrestrial composition [11, 12], thus suggesting that large isotopic variation among “normal” (i.e., non-FUN) CAIs due to nucleosynthetic anomalies in U may be unlikely.

It is notable that three of the fine-grained Allende CAIs (166, 167 and 175) are also classified as group II inclusions (highlighted with their designated numbers in Figure 1). The previously reported correlation of U isotopes with Th/U and Nd/U in Allende CAIs [5] is in fact dominated by the compositions of these group II inclusions and is additionally suggestive of ^{247}Cm decay as being the dominant cause of the U isotope variation in these fine-grained CAIs.

Another possible process that could cause relatively large variations (>1‰) in the U isotope composition without systematically affecting the Mg isotope composition in these Allende fine-grained inclusions may be secondary alteration under different redox environments. Uranium is a redox sensitive element which shows significant isotopic fractionation effects under variable redox conditions in terrestrial systems [e.g., 12, 13]. In contrast, Mg is not redox sensitive, and different redox conditions would not affect its isotopic composition. Further detailed investigations of redox indicators in fine-grained inclusions along with their U isotope compositions will be required to rigorously evaluate this possibility.

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