

STRONTIUM ISOTOPE HETEROGENEITY WITHIN SINGLE CAI FROM ALLENDE.

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Introduction: Calcium and aluminum rich inclusions (CAIs) in chondrites are the first condensates in the solar system. Most of CAIs possess a variety of nucleosynthetic isotope anomalies for heavy elements.

Trinquier et al. found a correlation between ⁴⁶Ti and ⁵⁰Ti for bulk meteorites and CAIs [1]. They suggested that the enrichment of ⁴⁶Ti and ⁵⁰Ti in CAIs relative to bulk chondrites were caused by thermal processing of pristine molecular cloud material. Brennecka et al. measured Ba, Nd and Sm isotopic compositions in a CAI, and reported that *r*-process excess was shown only for Ba isotopes [2]. They explained the inconsistency was the result of an injection of a small amount of H-event supernova material. In contrast, Moynier et al. found elevated ⁸⁴Sr/⁸⁶Sr values in Allende CAI, which implies excess of *p*-process or *r*-process nuclides in the CAIs [3]. They concluded that the anomaly caused by injection of *r*-process nuclides by comparing the results of Ba isotope analysis in [2].

These studies pointed out that the extent of isotope anomaly is nearly constant over different CAI grains, suggesting that CAIs have formed in a specific region within a limited period of time and redistributed in the solar system. The observed isotope anomalies in CAIs were most likely caused by the injection of refractory materials via single supernova near the proto sun.

However, most of these studies analyzed "bulk" CAIs without detailed mineral description. CAIs are composed of various mineral combinations which are classified as Type A, B and C. Such difference may reflect the environment and timing of individual CAI formation. It should be noted that the variation of mineral assemblage exists even within a single CAI grain. Therefore, it is important to determine the isotopic compositions not only of different types of CAIs, but also of multiple spots within a single CAI grain.

In this study, we performed *in-situ* Sr isotope measurement for a single CAI grain with a detailed mineral description using SEM-EDS and TIMS, coupled with a micro milling sample collection method. We discuss the relationship between mineralogy and Sr isotopic composition to elucidate the origin of nucleosynthetic isotope anomalies observed in CAIs.

Samples: A specimen of Allende meteorite (~2 cm) was sliced into two fragments. One of the fragments was mounted in resin with a diameter of 1/2 inch. Mineralogical compositions of CAI were determined

using two Scanning electron microscopes (SEM) at the Univ. of Tokyo (JSM-5310, JEOL) and Tokyo Tech (Hitachi 3400). In this study, one cm-size fluffy type A CAI was analyzed (Fig.1). We separated this CAI into three regions that consist of melilite (AL-1; Fig.2), melilite-spinel-anorthite (AL-2; Fig.3), and melilite-spinel-anorthite-hibonite (AL-3; Fig.4). AL-1 is composed of melilite. AL-2 and AL-3 were composed mainly of melilite and some spinel surrounded by anorthite. Hibonite exists only in AL-3 region. In addition, Åkermanite values are different between AL-2 (Åk=5) and AL-3 (Åk=5-20).

Experimental: For Sr isotope analysis, we used the counterpart of the fragment that was used in the SEM analysis. The CAI was sampled using a micro milling system (Geomill 326, Izumo, Japan). Approximately 1-2 mg of CAI was sampled to collect sufficient amount of Sr (~100 ng) for TIMS measurement. The CAI powder recovered was decomposed in a pressure digestion system (Digestec DAB-2, Berghof, Germany) with HF and HNO₃ (205°C for 32 hours). The Sr was purified by passing through 0.3 mL of an extraction chromatographic resin (Sr Spec, Eichrom).

High precision Sr isotope measurements were carried out by TIMS (Triton *plus* at Tokyo Tech, ThermoFisher). Approximately 100-500 ng of Sr was loaded onto single out-gassed W filament (Nilaco, Japan) with a Ta₂O₅ activator slurry. The data were obtained by averaging 400 ratios with 2σ rejection. The Sr isotope ratios were normalized to ⁸⁶Sr and corrected for mass fractionation by assuming ⁸⁶Sr/⁸⁸Sr = 0.1194.

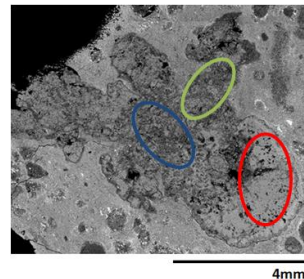


Fig.1 BSE image of Allende CAI overall. Red circle is AL-1, green circle is AL-2 and blue circle is AL-3.

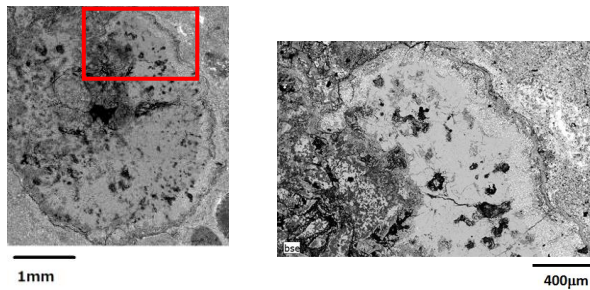


Fig.2 (left) BSE image of AL-1. (right) Detail BSE image of AL-1. Mostly composed of melilite, and fine-grained perovskite exist along the rim (white grain).

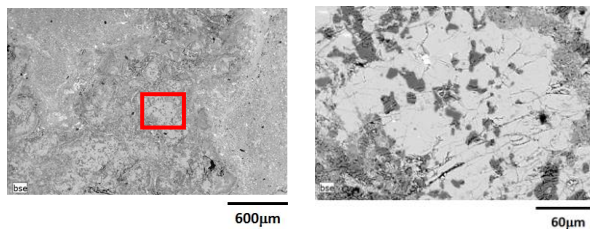


Fig.3 (left) BSE image of AL-2. (right) red area of left image. Melilite (light gray area) and a few spinel (black area) surrounded by anorthite (dark gray area).

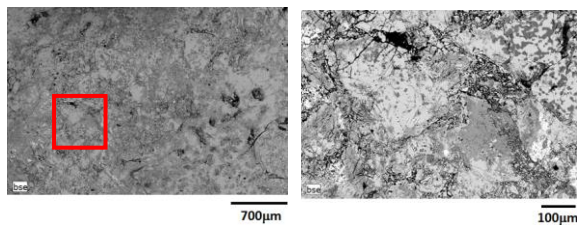


Fig.4 (left) BSE image of AL-3. (right) Red area of left image. Mineral composition is very similar to AL-2, but hibonite exist and have higher Åkermanite value.

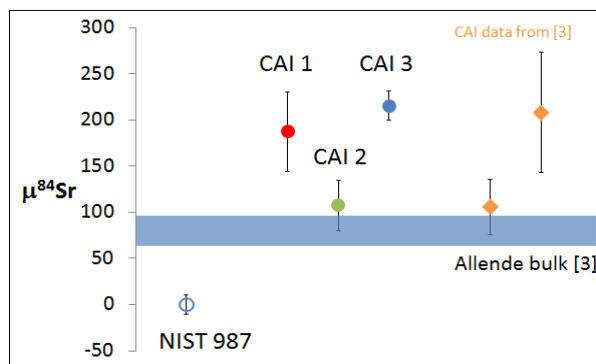


Fig.5 Results of Sr isotope analysis and previous data from [3] plotted by μ value. All CAIs have obvious anomalies from terrestrial standard. AL-1 consisted with both AL-2 and AL-3 within error ranges. AL-2 and AL-3 have different isotopic compositions over error ranges from each other.

Result: The results for Sr isotope measurement are shown in Fig. 5 together with previously published bulk CAI data. The $^{84}\text{Sr}/^{86}\text{Sr}$ values for AL-1, AL-2 and AL-3 were 187 ± 42.9 ppm, 107 ± 27.2 ppm and 215 ± 15.9 ppm relative to the terrestrial standard (NIST 987), respectively. Interestingly, $^{84}\text{Sr}/^{86}\text{Sr}$ values of AL-1 and AL-3 match one bulk CAI which showed the highest $^{84}\text{Sr}/^{86}\text{Sr}$ values in [2], whereas AL-2 has slightly lower $^{84}\text{Sr}/^{86}\text{Sr}$ value which is consistent with another bulk CAI reported in [2]. Of note, two bulk CAIs separated from Efremovka meteorite show $^{84}\text{Sr}/^{86}\text{Sr}$ values similar to AL-2 (111 and 123 ppm) [4].

Discussion: We found nucleosynthetic Sr isotope anomaly that heterogeneously distributed within a single CAI grain. The result suggests that Sr isotopic composition was heterogeneous in the CAI formation region, although the possibility of isotopic shift via parent body processing should be taken into account.

Because the $^{84}\text{Sr}/^{86}\text{Sr}$ ratio of bulk Allende is lower than CAIs, thermal event(s) on the Allende parent body should lower the $^{84}\text{Sr}/^{86}\text{Sr}$ ratio of CAI. Thus, the $^{84}\text{Sr}/^{86}\text{Sr}$ values of AL-1 and AL-3 would be much closer to the “original” Sr isotopic composition of this CAI compared to AL-2. However, AL-3 has lower Al/Mg ratio than AL-2, suggesting that AL-2 preserved more primitive isotopic information than AL-3. In addition, Sr concentration of CAI is approximately ten times higher than matrix and chondrule, which makes it difficult to produce 100 ppm shift of the $^{84}\text{Sr}/^{86}\text{Sr}$ ratio within a single CAI.

We propose that Sr isotopic heterogeneity within a single CAI was caused by a nebula process. It is likely that Sr isotopic composition of the CAI formation region has changed through time due to the injection of *p*-process or *r*-process enriched nuclides derived from a nearby supernova. Thermal processing in the CAI formation region was suggested in [1], and this event may have caused partial modification of Sr isotopic composition. Such event was thought to occur before the formation of the majority of CAIs, thus AL-2 would preserve more primitive chemical information than other CAIs. To discuss the timing and extent of Sr isotope modification within a single CAI, we will perform Mg isotope analysis using SIMS to determine Al-Mg chronology for this CAI.

References: [1] Trinquier et al. (2009) *Science*, 324, 374-376. [2] Brennecka et al. (2011) *LPSC Abstract #1302* [3] Moynier et al. (2012) *ApJ*, 758, 45. [4] Paton et al. (2013) *ApJL*, 763, L40.