STRONTIUM ISOTOPE ANOMALIES IN ALLENDE AND TAGISH LAKE METEORITES: RESULTS FOR SEQUENTIAL ACID LEACHING EXPERIMENTS. Tetsuya Yokoyama[1], Nobuaki Ito[1], Yusuke Fukami[1] and Wataru Okui[1], [1]Department of Earth and Planetary Sciences, Tokyo Institute of Technology, 2-12-1 Ookayama, Meguro-ku, Tokyo, Japan (tetsuya.yoko@geo.titech.ac.jp)

Introduction: Increasing number of highly precise stable isotope data are now available for a variety of trace elements in chondrites and differentiated meteorites. These data point to the existence of nucleosynthetic isotope heterogeneity in the protosolar nebula (e.g., Cr, Ti, Mo, Ru, Sm [1-3]), yet there are conflicting results that support homogeneous isotope distribution in the early solar system (e.g., Zr, Te, Os [4]). The inconsistency would arise from incomplete mixing and/or destruction of some selected presolar materials in the solar nebula. However, the detail about such processes that created isotope heterogeneity is not totally understood. A key issue is to identify main presolar phases for individual elements that contributed to the solar system, in conjunction with their nucleosynthetic origins. Sequential acid leaching of powdered bulk rocks is quite useful for detecting internal isotope anomalies in meteorites, especially for trace elements where concentrations in presolar grains are at ppm level. Previous studies on acid leaching of bulk chondrites revealed the existence of multiple presolar phases from diverse nucleosynthetic sources within single meteorites for Cr, Zr, Mo, Ba and Os [5-7].

In this study, we investigated stable Sr isotopic anomalies in carbonaceous chondrites by applying the sequential acid leaching technique. Strontium has four isotopes produced by the stellar nucleosynthesis of *s*-process (^{86}Sr, ^{87}Sr and ^{88}Sr), *r*-process (^{87}Sr and ^{88}Sr), and *p*-process (^{84}Sr). Radioactive decay of ^{87}Rb ($T_{1/2}$ = 4.8×10^{10} yr) also contributes to the abundance of ^{87}Sr. Stable Sr isotope anomalies in meteorites were first found in some calcium and aluminum rich inclusions (CAIs) from the Allende meteorite (CV3) [8]. The anomalous inclusions which had extremely low ^{84}Sr/^{86}Sr ratios (-0.4‰ relative to terrestrial standard) are now known as the "FUN inclusion" (fractionation and unknown nuclear inclusion). Patchett [9] analyzed Sr isotope compositions in chondrules and CAIs from Allende by applying the double-spike technique, and found that Sr isotopic variations observed in the chondrite components were only the result of mass-dependent fractionation occurred in the solar system. Strontium isotope compositions in presolar SiC fractions separated from the Murchison meteorite (CM2) were measured in [10]. They concluded that SiC grains are enriched in Sr isotopes produced by the s-process in AGB stars. In contrast, Sr isotope compositions in bulk meteorites were precisely determined by using state-of-the-art thermal ionization mass spectrometry (TIMS) [11, 12]. Moynier et al. [12] reported that carbonaceous chondrites, one basaltic achondrite (NWA 2976) and Allende CAIs have ^{84}Sr/^{86}Sr ratios higher than terrestrial samples, ordinary chondrites and HED meteorites at 0.5-1ε level. To further constrain the isotopically anomalous carriers for Sr in chondrites, we performed the sequential acid leaching experiments for bulk rocks of two carbonaceous chondrites, Allende and Tagish Lake (C2 ungrouped), and precisely measured Sr isotopes in the leachates by using TIMS.

Experimental: Approximately 1.6 g and 0.5 g of powdered Allende (Allende-A and Allende-B, respectively) and ~1g of powdered Tagish Lake were subjected to the following sequential leaching procedure as described in [5]:

Step 1) $CH_3COOH + H_2O$, 1 day, 20 °C
Step 2) $HNO_3 + H_2O$, 5 days, 20 °C
Step 3) $HCl + H_2O$, 1 day, 75 °C
Step 4) $HF + HCl + H_2O$, 1 day, 75 °C
Step 5) HF + HCl, 3 days, 150 °C
Step 6) HNO_3 + HF, 15 hours, 120 °C.

The leachates from steps 4-6 were dried and treated with $HClO_4$ in order to suppress insoluble fluoride formation. For Allende, 10-20% of each fraction was removed and used for the determination of trace element concentrations, and the rest was used for high precision Sr isotopic measurement. On the other hand, only Sr isotopic measurements were carried out for Tagish Lake. The Sr in each leachate was purified by passing through 0.3 mL of an extraction chromatographic resin (Sr Spec, Eichrom). This procedure was performed twice, and finally the sample solution was passed through a micro-column filled with 100μl of Amberchrom CG71-C to remove organic matters derived from the Sr Spec resin.

High precision Sr isotope measurements were carried out by the thermal ionization mass spectrometry in positive ion mode (Triton *plus* at Tokyo Tech, ThermoFisher Scientific). In addition to individual leachates, we also analyzed a standard (NIST 987) and bulk rocks of Allende and Tagish Lake. Approximately 100-500 ng of Sr was loaded onto single out-gassed W filament (Nilaco, Japan) with a Ta_2O_5 activator slurry. The data were obtained by averaging 100 to 400 ratios with 2σ rejection, acquired by 5 to 20 block measurements (20 scans/block, 16 s integration per scan) in the static multi-collection using five Faraday cups (L2 for ^{84}Sr, L1 for ^{85}Rb, Center for ^{86}Sr, H1 for

^{87}Sr, and H2 for ^{88}Sr). The amplifier rotation system was applied for all analyses. Typical beam intensity for $^{88}Sr^+$ was >10V, and we report only when the average $^{88}Sr^+$ beam intensity exceeded 5V. The Sr isotope ratios were normalized to ^{86}Sr and corrected for mass fractionation by assuming that ^{86}Sr/^{88}Sr = 0.1194

Results: The ^{84}Sr/^{86}Sr ratios for Allende and Tagish Lake leachates are reported in μ^{84}Sr units, which represent relative deviation (parts per million) from the average of NIST 987 (Fig. 1). Because of extremely low Sr^+ beam intensities, we were unable to analyze the leachate 5 of Allende-A and B. The rest of the Allende leachates have positive μ^{84}Sr values (50-100 ppm) that are close to the bulk Allende (75 ppm). It should be noted that the amount of Sr in leachate 5 scarcely contributed (~1%) to the total Sr in bulk Allende, and a mass balance calculation of individual leachates (excluding leachate 5) accounts for the bulk μ^{84}Sr value within analytical uncertainties. For Tagish Lake, the μ^{84}Sr value in leachates 3, 4 and 5 are close to that of NIST 987. Leachates 1 and 2 have slightly positive μ^{84}Sr values (30-40 ppm), but leachate 6 has an extremely large negative anomaly (-326 ppm). Bulk Tagish Lake has a marginally positive μ^{84}Sr value (16 ppm) that is apparently lower than that of bulk Allende.

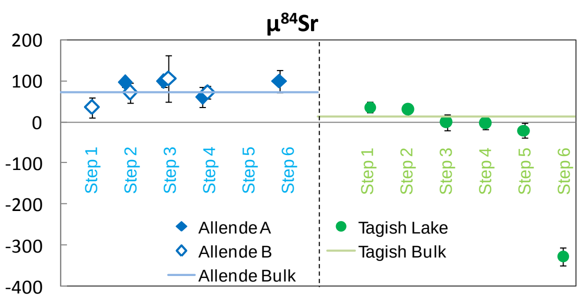

Fig. 1 μ^{84}Sr values in acid leachates and bulk rocks of Allende and Tagish Lake.

Discussion: The positive μ^{84}Sr value represents the existence of materials enriched in Sr synthesized by the r and/or p-process, while the enrichment of s-prosess Sr makes a negative μ^{84}Sr value. Relatively high μ^{84}Sr value in bulk Allende seems intriguing because most of the presolar phases in Allende has been destroyed via thermal metamorphism on the parent body. It was recently found that calcium and aluminum rich inclusion (CAI) from Allende has elevated μ^{84}Sr value (~120 ppm) that is higher than that of bulk Allende and other carbonaceous chondrites [12]. This implies that the Allende matrix has a μ^{84}Sr value lower than that of bulk Allende unless chondrules have extremely low μ^{84}Sr. Assuming that the μ^{84}Sr of Allende matrix is similar to that of a matrix-dominate chondrite such as Tagish Lake, Allende leachate 1 is consist 79% of matrix and 21% of CAI for Sr. Likewise, the contribution of CAI in Allende leachates 2, 3, 4 and 6 are 66%, 84%, 50% and 82%, respectively. Thus, there are multiple phases that have different acid resistance in CAI, which carry μ^{84}Sr values higher than 100 ppm. Interestingly, we observed no FUN-like CAI signature which has a drastically low μ^{84}Sr value (-4200 ppm, [8]).

The μ^{84}Sr values in Tagish Lake leachates suggest that leachates 1 and 2 contain presolar grains rich in r and/or p-process Sr, while leachate 6 contains presolar grains rich in s-process Sr. This means that Tagish Lake has at least two different presolar phases produced in different stellar environment. The easily leachable presolar phase(s) in leachates 1 and 2 is not yet identified, but presumably consists of minerals produced by supernovae. The acid resistant phase in leachate 6 is most likely presolar SiC synthesized in AGB stars.

We obtained contrasting μ^{84}Sr values in acid leachates from Allende and Tagish Lake, because main carriers of isotopically anomalous Sr are different in these meteorites (CAI for Allende and presolar grains for Tagish Lake). On the other hand, Reisberg et al. [5] showed the enrichment of s-process Os in leachates 2, 3, 4 and 6 of Murchison (CM2). The Os result is inconsistent with our Sr result for the same type 2 chondrite, Tagish Lake. We postulate that isotopically anomalous presolar phases for Sr and Os are partly different in carbonaceous chondrites.

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