

EVIDENCE FOR THE SURVIVAL OF A *p*-PROCESS ANOMALY CARRIER IN FINE-GRAINED CAIS FROM ALLENDE. F. L. H. Tissot^{1,2}, B. L. A. Charlier³, H. Vollstaedt⁴, N. Dauphas⁵, C. J. N. Wilson³, R. T. C. Marquez², ¹EAPS Department, MIT, Cambridge, MA 02139, USA, ²The Isotoparium, Division of Geological and Planetary Sciences, California Institute of Technology, Pasadena, CA 91125, USA, tissot@caltech.edu, ³School of Geography, Environment and Earth Sciences, Victoria University of Wellington, Wellington 6140, New Zealand, ⁴Thermo Fisher Scientific, Bremen, Germany, ⁵Origins Laboratory, Department of the Geophysical Sciences and Enrico Fermi Institute, The University of Chicago, 5734 South Ellis Avenue, Chicago, IL 60637, USA.

Introduction: The existence of Sr nucleosynthetic anomalies in CAIs and their host meteorites, relative to Earth, is well documented [1-7]. Typically on the order of a few tens to hundreds of ppm (relative to NBS987 after internal normalization to $^{86}\text{Sr}/^{88}\text{Sr}=0.1194$), the origin of these anomalies remain a matter of debate. Indeed, it is unclear if the apparent ^{84}Sr -excesses seen in internally normalized data result from variations in the *p*-process nuclide ^{84}Sr (e.g., [5-7]), or reflect (through internal normalization) an *r*-process anomaly in ^{88}Sr (e.g., [2-4]).

The uncertainty surrounding the nature of the ^{84}Sr anomalies has implications beyond unraveling the pre-solar heritage of the solar system. Most importantly, it affects dating of volatile element depletion in the early solar system. Indeed, given that Rb being is more volatile than Sr (T_{c50} of 800 versus 1464 K, respectively [8]), one can constrain the timing of the evaporation/condensation event(s) that lead to the depletions in moderately volatile elements observed in planetary bodies [1, 9-10], assuming (i) that the initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of solar system materials is known and homogeneous, and (ii) that $^{87}\text{Sr}/^{86}\text{Sr}$ variations are entirely due to ^{87}Rb decay. These assumptions break down if nucleosynthetic anomalies present in the nascent solar system were not completely homogenized. Indeed, $^{87}\text{Sr}/^{86}\text{Sr}$ values are calculated using an internal normalization scheme to a fixed $^{86}\text{Sr}/^{88}\text{Sr}$ ratio, which is only warranted if the nucleosynthetic anomalies in CAIs and bulk solar system materials reside in the *p*-process isotope ^{84}Sr . If, however, the anomalies lie on ^{86}Sr (*s*-deficit) or ^{88}Sr (*r*-excess), the normalization will shift the initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio, and variations in initial $^{87}\text{Sr}/^{86}\text{Sr}$ would no longer reflect temporal differences, but instead the compounded effect of radiogenic ingrowth and nebular isotopic heterogeneity.

These outstanding questions can be answered, provided that one finds materials that (i) formed early in the history of the solar system, and (ii) were minimally affected by radiogenic ingrowth of ^{87}Sr . As the oldest materials formed in the solar system, CAIs are the prime target for such investigations. Yet, finding CAIs with pristine, low Rb/Sr ratios is non-trivial because Rb (and to a lesser extent Sr) are fluid-mobile, and the Rb-Sr systematics in CAIs is often significantly disturbed during aqueous alteration (e.g., [3, 7, 11]).

In order to isolate phases with low Rb/Sr ratios and constrain the nature of the carrier of ^{84}Sr anomalies, we subjected to a step-leaching dissolution protocol a suite of nine fine-grained (fg-)CAIs from Allende [12]. Fg-CAIs were chosen for their relatively primitive characteristics: they display condensate-like features (petrographic and group II REE patterns), have never been melted, and their chemical compositions match those predicted by equilibrium thermodynamic calculation for partial condensation from a gas of solar composition gas (e.g., [13-14]). Because in-situ methods would provide insufficient precision, the isolation of pristine components (*i.e.*, with low primary Rb/Sr ratios) was performed chemically via step-leaching.

Methods: After visual inspection of slabs of the Allende CV chondrite, CAIs with fine-grained morphologies were extracted using clean steel dental tools [15]. Given the limited mass of the samples (13 to 158 mg), and to ensure sufficient precision even on the smallest sample fractions (<10% of total Sr in the CAI), $10^{13} \Omega$ feedback resistors were used on TIMS instruments as these permitted high-precision Sr measurements on load sizes as low as 150 pg. Each leachate fraction was split into three aliquots to quantify (i) nucleosynthetic anomalies through internal normalization on unspiked material, (ii) stable ^{86}Sr and ^{88}Sr relationships using double spike methodologies, and (iii) ^{87}Rb abundances by high-sensitivity ICP-MS measurements to assess the contribution of ^{87}Rb -decay on $^{87}\text{Sr}/^{86}\text{Sr}$ variations and estimate initial $^{87}\text{Sr}/^{86}\text{Sr}$ values using Rb-Sr isochrons. At each stage of the step-leaching process, the residues were weighed to estimate the amounts of components dissolved, and their chemical and Sr-isotopic composition. Two instruments were used in this work: the $10^{11} \Omega$ equipped Triton TIMS at Victoria University of Wellington (VUW), which was used for large Sr loads (100-1000 ng), and the $10^{13} \Omega$ equipped Triton XT at the Thermo-Fisher factory in Bremen, which was used for Sr loads below 100 ng.

Elemental and isotopic release patterns: Each CAI sample was subjected to a five-step leaching protocol of increasing acid strength and temperature similar to those in [6]. Of the nine CAIs analyzed, only one had no material remaining after the fourth leaching step. In all cases, the first three leach steps (L1 to L3) contained

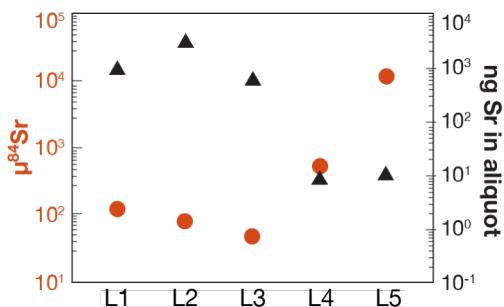


Fig. 1: Sr ng (filled triangles) and $\epsilon^{84}\text{Sr}$ (filled circles) in successive leachates for one of the CAIs studied here (7 of the 9 CAIs show similar patterns).

>80% of the total Sr budget and the amount of Sr released in each step typically decreased by 2-3 orders of magnitude across the total experiment (Fig. 1). To test the efficiency of the step-leaching protocol and accuracy of the gravimetric accounting for Rb and Sr yields, a separate fraction of powder from each CAI was dissolved in bulk (using a Parr bomb) to ensure total dissolution, and analyzed in the same way as the leach aliquots. Excellent agreement was observed between the Sr isotopic characteristics for the bulk samples versus the gravimetrically summed leach fractions.

p-process anomalies: All leachates and final residue show positive $\mu^{84}\text{Sr}$ values. The first three leaching steps (80 to 99% total Sr) have elevated $\mu^{84}\text{Sr}$ values that are within the range of published values observed in bulk single CAIs (~ 100-200 ppm, Fig. 1). In contrast, the final leach (L4) and residue digest (L5) show $\mu^{84}\text{Sr}$ anomalies reaching per-mille to percent levels (Fig. 2), and the fractions most depleted in Sr also display the largest anomalies.

If the positive $\mu^{84}\text{Sr}$ values measured here were the result of *r*-process excesses in ^{88}Sr propagating to ^{84}Sr as an internal normalization (to $^{86}\text{Sr}/^{88}\text{Sr} = 0.1194$) artifact, this would also result in apparent negative ^{87}Sr shifts of a magnitude half those of ^{84}Sr (*i.e.*, the samples should appear less radiogenic). Despite the large $\mu^{84}\text{Sr}$ values in fractions L4 and L5, no negative ^{87}Sr anomalies are seen in those fractions (Fig. 2), which provides the strongest evidence to date that these anomalies are true (and not only apparent) ^{84}Sr -excesses, due to the presence of *p*-process carriers in the early solar nebula and incorporated into fg-CAIs.

Our data are consistent with the work by Pravdivtseva et al. [16] which, based on a step-wise pyrolysis study of noble-gas isotopic signatures in fg-CAIs from Allende, argued that distinct presolar carriers are present in fg-CAIs. Pravdivtseva et al. proposed that the carriers were presolar SiC (<0.2 μm in size). Although our data do not permit identification of the specific carrier(s) of the ^{84}Sr anomalies, SiC grains are extremely unlikely candidates, as the few SiC grains that have

been measured for Sr isotopes ($n=39$) display either solar composition or negative anomalies (down to $-90.6 \pm 2.0\%$) [17-19]. In fact, no presolar grains, to date, have been found with Sr *p*-process signatures. We present progress on a search for the carrier of the ^{84}Sr anomalies in a companion abstract (by Marquez et al.).

The fact that Sr anomalies lie in the *p*-process isotope means that careful re-evaluation of the assumptions and hypotheses around the concept of the timescales of volatile depletion in the early solar system based on Rb-Sr systematics are necessary [3, 9-10].

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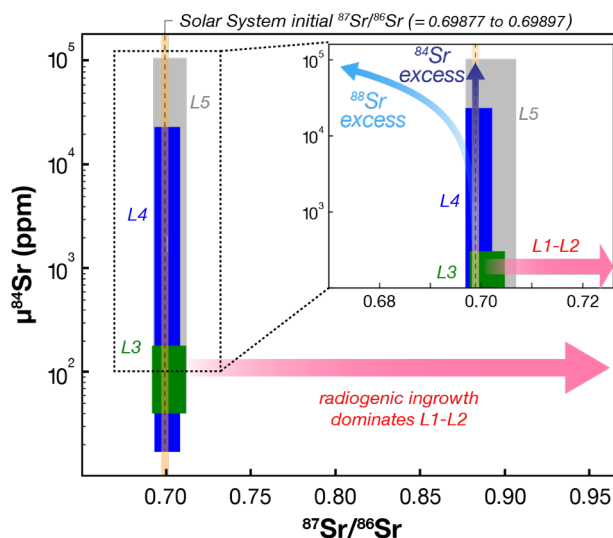


Fig. 2: Anomalies ($\mu^{84}\text{Sr}$) vs $^{87}\text{Sr}/^{86}\text{Sr}$, both from internally normalized data. Arrows show expected relationships for radiogenic ingrowth, ^{84}Sr -excess, and correlation arising from internal normalization to $^{88}\text{Sr}/^{86}\text{Sr}$ in the presence of *r*-process anomalies. Color areas show regions populated by data from L1 to L5.