

STABLE ISOTOPIC FRACTIONATIONS OF 8 REES IN GROUP II CAIS AND INSIGHTS INTO EARLY SOLAR SYSTEM EVOLUTION. J.Y. Hu¹, N. Dauphas¹, F.L.H. Tissot^{1,2}, A.M. Davis¹, F. Ciesla¹, R. Yokochi¹ and T.J. Ireland¹, ¹Origins Laboratory, Department of the Geophysical Sciences and Enrico Fermi Institute, The University of Chicago, Chicago IL, ²The Isotoparium, Division of Geological and Planetary Sciences, Caltech, Pasadena, CA.

Introduction: The chemical evolution of the Solar System and its material is mainly controlled by early nebular and subsequent planetary accretion processes. The latter overprinted the primary signatures of nebular processes in planetary materials, making it difficult to decipher the evolution of the Solar Nebula.

Calcium-aluminum-rich inclusions (CAIs) are the first solids that condensed out of the solar nebula, providing a snapshot of the earliest stage of the Solar System [1-3]. Near 40% of CAIs have a highly fractionated REE pattern, characterized by a uniform enrichment in moderately REEs (Tm and La-Sm), and depletions in both the most volatile REEs (Eu and Yb) and the most refractory REEs (Gd-Lu except Tm and Yb) [4-6]. This so-called group II pattern is most commonly found in fine-grained CAIs, the most abundant type of CAIs in most chondrite groups [6]. This suggests that intense nebula-stage thermal processing might have also affected the building blocks of planets.

The depletion of both the most volatile and refractory REEs in the group II pattern was originally thought to represent a snapshot of the condensation sequence: in the first stage, the nebular gas with chondritic REE composition partially condensed and formed an ultrarefractory mineral phase that sequestered part of the most refractory REEs, leading to HREE depletions in the gas; in the second stage, the gas residue was isolated from the ultrarefractory phase and further condensed to form group II CAIs, during which REEs were quantitatively condensed except for the most volatile REEs, leading to depletions of the most volatile REEs [1-3]. In this framework, the most refractory REEs are quantitatively condensed in the second stage and their isotopes are only likely to be fractionated during partial condensation in the first stage. Since CAIs formed at extremely high temperature, little equilibrium stable isotopic fractionation is expected and any substantial isotopic fractionation should be induced by kinetic effects. One would therefore expect that the most refractory REEs (e.g., Gd, Dy, Er) would either be isotopically unfractionated or enriched in heavy isotopes as a result of partial condensation in stage 1. The quantitatively condensed moderately volatile REEs are expected to show no significant stable isotope fractionation, and the most volatile REEs to be enriched in light isotopes as a result of partial condensation.

Here we test this snapshot scenario by analyzing the stable isotopic fractionation of 8 REEs (Ce, Nd, Sm, Eu, Gd, Dy, Er and Yb) in 5 well characterized group II CAIs. Our study reveals the complex thermal histories of group II CAIs, inconsistent with the canonical snapshot scenario, and points to the existence of a widespread, intense

heating event in the earliest stages of solid formation in the Solar System.

Methodology: The CAIs analyzed were selected from a group of well characterized samples from the Allende chondrite. The sampling and digestion procedures are described in detail in [7] and only briefly summarized here. The CAIs were picked from a few Allende slabs using stainless-steel dental tools and ground in an agate mortar. Sample powders were digested in using a mixture of HF:HNO₃ and HCl:HNO₃ acid attacks on hot plates. After dissolution, 80% of the sample was taken and processed through 2 Eichrom UTEVA[®] columns for U extraction. The matrix cuts that contained all REEs were recovered and recombined. REEs in the matrix cuts were extracted using a prepacked TODGA column [8]. The extracted REEs were then loaded onto a FluoroPolymer Liquid Chromatography system (FPLC) developed at the University of Chicago for separation of all multi-isotopic REEs from one another [9,10]. After separation, the REEs were measured using an Apex Omega desolvating nebulizer and a Thermo Scientific MC-ICPMS upgraded to Neptune Plus specifications [11]. To ensure that no artificial fractionation was introduced during the chemistry, a geostandard BCR-2 that was also used in U chemistry was purified and analyzed along with the CAIs. Two different portions of the same CAI were also processed separately to test the reproducibility.

Results: The typical uncertainties of REE isotopic analyses were ± 0.05 ‰/amu. The total procedural blank yielded negligible amount of REEs compared to those in samples. The isotopic compositions of all 8 REEs in BCR-2 are consistent with earlier measurements using both sample-standard bracketing and the double-spike method [11]. The two replicates of one CAI also yielded identical results within errors.

The isotopic variations of volatiles to moderately volatile REEs (Eu, Yb and LREEs) range from -1 to +1 ‰/amu with averages near 0 while the refractory REEs (HREEs except Yb) show significant negative stable isotopic fractionations ranging from 0 to -3 ‰/amu with averages approximately at -2 ‰/amu (Fig. 1). For comparison, all REEs in a group V CAI (i.e., unfractionated REE pattern) show fractionations within 0.5 ‰/amu. Isotopic variations of Eu were found to be linearly correlated with those of Sr in the group II CAIs from -1.3 to +0.8 ‰/amu for Sr and from -1.1 to +0.8 ‰/amu for Eu, with a slope of 0.8 [12].

Discussion: According to the canonical snapshot scenario, the most refractory REEs were expected to be either isotopically unfractionated or enriched in heavy isotopes. On the contrary, our analyses reveal that they are

significantly enriched in light isotopes. The remarkably light isotopic compositions of the most refractory REEs in the group II CAIs are in contrast with the general lack of stable isotopic fractionation for the most volatile REEs despite the fact that both the most refractory and volatile REEs were significantly depleted in the REE abundance pattern. The relatively moderate stable isotopic fractionations suggest that the most volatile REEs were partially lost in near equilibrium conditions in contrast to the far-from-equilibrium process that has fractionated the most refractory REEs.

Here we propose that the canonical partial condensation scenario is incorrect and more complex thermal processing must have been involved. The formation of group II CAIs can be explained by a two-stage process involving CAI evaporation followed by recondensation [13]. In the first stage, precursor with chondritic REE composition was subjected to fast evaporation. The most volatile and moderately refractory REEs completely vaporized into the gaseous phase and hence displayed little stable isotopic fractionations. The most refractory REEs were only partially vaporized, leading to depletion in abundance and significant negative isotopic fractionations in the gas. In the second stage, the gas was physically isolated from the solid residue and then condensed to form Type II CAIs under near equilibrium conditions. The

most refractory REEs and the moderately refractory REEs completely condensed at this stage and inherited the abundance pattern and isotope signatures from the gas. The most volatile REEs only partially condensed and hence were depleted in the CAIs. The limited isotopic fractionations observed in the most volatile REEs indicated that the cooling that induced the condensation must be slow enough to maintain a near equilibrium state. Group II REE patterns therefore do not represent a snapshot in the condensation sequence but rather a signature of complex thermal processing of pre-existing materials, which will be discussed at the time of the conference.

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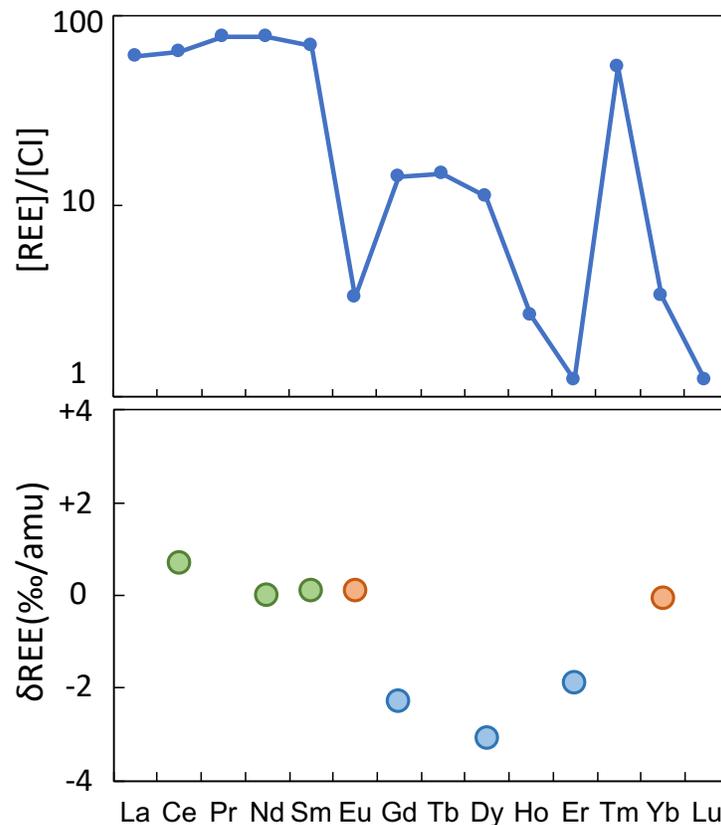


Fig. 1. REE patterns and isotopic compositions of 8 REEs of the group II CAI “AL3S5”. The isotopic fractionations of the REEs (‰/amu) are normalized to the geostandard BCR-2.