

ISOTOPIC STUDIES OF STRONTIUM, BARIUM, CERIUM, NEODYMIUM, SAMARIUM, AND GADOLINIUM IN EUCRITES. H. Hidaka¹, K. Sera¹ and S. Yoneda², ¹Department of Earth and Planetary Systems Science, Hiroshima University, Higashi-Hiroshima 739-8526, Japan (hidaka@hiroshima-u.ac.jp); ²Department of Science and Engineering, National Museum of Nature and Science, Tsukuba 305-0005, Japan (s-yoneda@kahaku.go.jp).

Introduction: Most eucrites have basaltic composition and represent lava flows or shallow intrusions from a differentiated parent body, possibly asteroid 4 Vesta. Chronological information of eucrites provides important constraints of the evolutionary history of the eucrite parent body (EPB). Although many isotopic studies have been conducted by several kinds of geochronometers using decay systems with long-lived and short-lived radioisotopes to understand the processes of accretion, core formation, mantle differentiation, and the formation of primary basalts on the EPB [1-4], the exact time-scales for eucrites petrogenesis are still unclear. Complex post-crystallization events such as impact heating and remelting may have reset the isotopic systems in eucrites to some degree, showing in inconsistent results from several kinds of geochronometers.

The final goal of this study is better understanding of the early evolution processes of the solar planets from the precise isotopic variations of multi-elements including radiogenic and neutron-captured isotopes of differentiated meteorites. Because Sr, Ba, Ce, and Nd include radiogenic isotopes, ⁸⁷Sr, ¹³⁵Ba, ¹³⁸Ce, ¹⁴²Nd, and ¹⁴³Nd, respectively, the isotopic measurements of these elements can be used for the chronometrical application. The isotopic shifts of ¹⁵⁰Sm/¹⁴⁹Sm and ¹⁵⁸Gd/¹⁵⁷Gd have been used to characterize the neutron capture records of meteorites, because ¹⁴⁹Sm and ¹⁵⁷Gd isotopes are very sensitive to thermal neutron arising cosmic-ray irradiation. In this study, systematic isotopic analyses of Sr, Ba, Ce, Nd, Sm and Gd were performed on eucrites.

Samples and Experiments: Establishment of the experimental techniques for multi-element isotopic studies on a single material is required. Eight eucrites, Juvinas, Millbillillie, Stannern, DaG 380, DaG 391, DaG 411, DaG 443, and DaG 480, were used in this study. 0.5-0.9 g of each powdered sample was decomposed by treatment with HF-HClO₄ with heating. Then, the samples were taken to dryness and redissolved in 10 mL of 2M HCl. The solution was divided into two portions: the main portion for isotopic measurements by TIMS and the rest for the determination of elemental abundances of Sr, Ba, and REE by ICP-MS.

A variety of geological reference materials (JA-1, JB-1a, JG-1, JGb-1) and chemical reagents (standard solutions produced from SPEX CertiPrep for Ba, Ce, Sm, and Gd; JNdi-1 for Nd; NIST SRM-987 for Sr) were also analyzed as terrestrial standard samples.

Each solution for the isotopic study was loaded onto a cation exchange resin packed column (Bio-Rad AG50WX8, 200-400 mesh, H⁺ form, 50 mm length × 4.0 mm diameter). The column was washed with 2 M HCl for Sr elution. Next, the column was washed with 2 M HNO₃ for Ba elution. Finally, the column was washed with 6M HCl for REE elution. The solution was then evaporated to dryness and redissolved in a drop of 0.1 M HCl. The first Sr fraction was purified using a quartz column packed with Sr-specific resin (Eichrom, Sr resin, particle size of 100-150 μm, 100 mm length × 2.5 mm diameter). The REE fraction was loaded onto a second column packed with lanthanide-specific resin (Eichrom, LN resin, particle size of 100-150 μm, 100 mm length × 2.5 mm diameter) to separate Ce, Nd, Sm and Gd using 0.15, 0.25, 0.35 and 0.5 M HCl, respectively.

A thermal ionization mass spectrometer (Triton Plus) equipped with nine Faraday cup collectors was used for the isotopic measurements of Sr, Ba, Ce, Nd, Sm, and Gd. Sr was measured on single Re filament with Ta₂O₅ activator. Instrumental isotopic mass fractionation was corrected by the exponential law using ⁸⁸Sr/⁸⁶Sr= 8.375209 as a normalizing factor. Ba, Ce, Nd, Sm and Gd were measured on double Re filaments. For their isotopic measurements, ¹³⁴Ba/¹³⁶Ba=0.307776, ¹⁴⁰Ce/¹⁴²Ce=7.941, ¹⁴⁶Nd/¹⁴⁴Nd=0.7219, ¹⁴⁷Sm/¹⁵²Sm=0.56081, ¹⁵⁶Gd/¹⁶⁰Gd=0.9361 were used to correct for instrumental mass fractionation using the exponential law. When the isotopic depletions of ¹⁵⁵Gd were found in the analyses, the Gd isotopic data sets were normalized by (¹⁵⁵Gd+¹⁵⁶Gd)/¹⁶⁰Gd=1.61290. Cerium was measured as oxide ions (CeO⁺), while Ba, Nd, Sm and Gd were measured as mono-atomic ions (M⁺).

Another minor aliquot of each sample solution was once evaporated to dryness, and redissolved using 5 mL of 0.5 M HNO₃. A 0.05 g quantity of a 10 ppb indium solution was added to the individual sample solutions as an internal standard element to optimize the analytical conditions for REE measurements. An ICP-MS (Agilent 7500cx) was used for determination of the elemental abundances of Sr, Ba, and REE.

Results and Discussion: ⁸⁷Sr includes radiogenic component from the decay of ⁸⁷Rb, and its isotopic variation correlates with Rb/Sr elemental ratios. In order to estimate the initial ⁸⁷Sr/⁸⁶Sr of the EPB, namely basaltic achondrite best initial (BABI) [5, 6], from our

Rb-Sr data, precise determination of Rb/Sr elemental ratios by isotope dilution method were also performed together with the Sr isotopic works. The fit of the data points from three non-desert eucrites to a straight line is consistent with the eucritic isochron given by previous study [5, 6], while the data points from five desert eucrites are randomly deviated from the eucritic isochron probably due to terrestrial contamination. We are now remeasuring the Rb-Sr data set of five DaG eucrites after washing of the bulk samples to remove the possible contaminants of terrestrial materials.

On the other hand, the data points from 8 noncumulate eucrites including desert eucrites lie on a single isochron of ^{147}Sm - ^{143}Nd isotopic systematic with a slope of 4.567 Ga. The isotopic deviations of ^{142}Nd of eucrites show slightly negative to zero values relative to terrestrial standard materials ($\epsilon^{142}\text{Nd}=-0.2$ to 0), and no positive values which are observed in typical cumulate eucrites having high Sm/Nd elemental ratios. These results are consistent with previous study [7].

Ba has seven stable isotopes consisting of p-, s-, and r-process nucleosynthetic origin. Most previous Ba isotopic studies of meteorites focused on the variation of r- and s-process nucleosynthetic components due to additional inputs in the early solar system. ^{135}Ba and ^{137}Ba isotopes are sensitive to s- and r-process variations, and often have deficits and/or excesses in chemical separates in carbonaceous chondrites due to the existence of presolar grains [8]. However, Ba isotopic deviation patterns of eucrites show much flatter than those in carbonaceous chondrites, and little evidence for additional nucleosynthetic components and radiogenic ^{135}Ba decayed from extinct ^{135}Cs . The results conclude that the isotopic information of the additional nucleosynthetic components disappears during the evolution process of eucrites.

Since ^{138}Ce isotopic excesses of eucrites correlate with their La/Ce elemental ratios, these excesses are identified to be decay product from ^{138}La . Our data are consistent with the results from previous study on the EPB evolution of Ce isotope [9], showing the La-Ce isochron of 4.567 Ga with the initial Ce isotopic ratio of $^{138}\text{Ce}/^{142}\text{Ce}=0.0225321$.

Sm isotopic compositions of the eucrites show the depletions of ^{149}Sm and excesses of ^{150}Sm caused by neutron capture reactions due to cosmic rays irradiation. These Sm isotopic shifts correspond to the neutron fluences ranging from 3.2 to 6.1×10^{15} n cm⁻². The estimated neutron fluences are almost consistent with their cosmic-ray exposure ages, suggesting no strong evidence of initial cosmic-ray irradiation on the surface of EPB. The isotopic shifts of Sm are caused by the cosmic-ray irradiation after release from the EPB. The

variation of ^{157}Gd - ^{158}Gd isotopic shifts also show the consistency with that of ^{149}Sm - ^{150}Sm .

Systematic isotopic data obtained in this study provide a hint to understand the evolution processes of differentiated meteorites. We are now applying this technique for the analyses of cumulate eucrites and diogenites.

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