

SYSTEMATIC P-PROCESS ISOTOPIC EXCESSES OF SR, BA, CE AND SM OBSERVED IN THE CHEMICAL SEPARATES OF THE KAPOETA METEORITE. H. Hidaka¹ 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: A number of proton-rich isotopes of naturally occurring stable nuclides cannot be synthesized by neutron capture reactions as r- and s-process nucleosynthetic paths. P-process isotopes might be synthesized in different supernovae with varying amounts. The presently most favored production mechanism for the p-process isotopes heavier than Fe is photodisintegration. It is generally known that neutron-deficient nuclei can be produced mainly by three processes, γ -process ((γ, n) , (γ, p) and/or (γ, α) reactions) in stellar plasma, rp-process (rapid proton capture reaction) in X-ray burst and vp-process in strong neutrino fluxes environments in core collapse supernovae.

Some brecciated meteorites contain high amounts of solar gas component, suggesting the implantation by solar cosmic rays (SCR) released from an ancient Sun before compaction of fragmental materials on the parent asteroidal bodies. The idea that solar system materials were irradiated by SCR around the early Sun has long been discussed from isotopic evidence of spallogenic nuclides [1], and been recently renewed from cosmochemical and astronomical viewpoints. The former is shown by isotopic evidence for the existence of ¹⁰Be in refractory inclusions of primitive meteorites [2-6], because Be is destroyed rather than produced in most stellar events. The latter is from observations of strong X-ray emissions in young stars that are accompanied by intense fluxes of accelerated particles [7, 8].

In this study, Sr, Ba, Ce, Nd and Sm isotopic compositions of the sequential acid-leachates from the Kapoeta meteorite (howardite) were determined to find systematic and correlated variations of their p-process isotopic abundances.

Samples and experiments: The Kapoeta meteorite is a regolith breccia meteorite, and is known as a highly gas-rich meteorite that experienced early irradiation by cosmic rays, but its irradiation history is complicated and still arguable with regard to evidence for possible contribution of early solar irradiation. Our previous study on neutron capture history of Kapoeta suggests the existence of preirradiation materials migrating into the meteorite parent body [9]. The existence of space-weathered rims in several regolith grains in the Kapoeta meteorite is resulted from the irradiation of cosmic-ray on the regolith parent body [10].

Sequential acid-leaching treatments were carried out to obtain chemically different phases from the single samples. About 0.5 g of powdered sample was leached using 5 mL of 0.5 mM HNO₃ + 0.002 mM HF, 2 M HCl, and aqua regia, successively. Finally, the residue was decomposed by treatment with HF-HClO₄ with heating. This procedure was modified from the previous methods [11, 12]. In particular, the first leaching step of the procedures is effective to dissolve very surficial material of the fine grains in the depth within 1.5 μ m [11]. The four leaching fractions are designated L1, L2, L3, and L4, respectively.

The four leachates (L1 to L4) were taken to dryness and redissolved in 1 mL of 2M HCl. The solutions were 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.

As the first step, 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 \times 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 μ m, 100 mm length \times 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 μ m, 100 mm length \times 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 Triton thermal ionization mass spectrometer equipped with nine Faraday cup collectors was used for the isotopic measurements of Sr, Ba, Ce, Nd, Sm, and Gd. The purpose of Gd isotopic measurement is to cross-check the neutron capture effect of the samples in combination with Sm isotopic data.

Sr was measured on single Re filament with Ta₂O₅ activator. Instrumental isotopic mass fractionation was corrected by the exponential law using $^{88}\text{Sr}/^{86}\text{Sr}=8.375209$ as a normalizing factor.

Ba, Ce, Nd, Sm and Gd were measured on double Re filaments. For their isotopic measurements, $^{134}\text{Ba}/^{136}\text{Ba}=0.307776$, $^{140}\text{Ce}/^{142}\text{Ce}=7.941$, $^{146}\text{Nd}/^{144}\text{Nd}=0.7219$, $^{147}\text{Sm}/^{152}\text{Sm}=0.56081$, $^{156}\text{Gd}/^{160}\text{Gd}=0.9361$ were used to correct for instrumental mass fractionation using the exponential law. When the isotopic depletions of ^{155}Gd were found in the analyses, the Gd isotopic data sets were normalized by $(^{155}\text{Gd}+^{156}\text{Gd})/^{160}\text{Gd}=1.61290$ [9,12]. 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 to determine the elemental abundances of Sr, Ba, and REE.

Results and Discussion: Significantly large isotopic excesses of ^{84}Sr , ^{130}Ba , ^{132}Ba , ^{136}Ce , ^{138}Ce and ^{144}Sm were observed particularly in the first leaching fraction L1. As one of the isotopic data in this study, the isotopic deviation patterns of Sm in the chemical separates from Kapoeta are shown in Fig. 1. The first leaching fraction L1 shows a significantly large excess

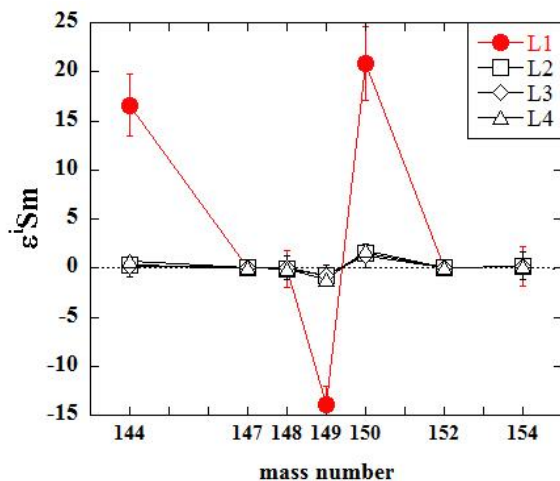


Figure 1. Isotopic deviation patterns of Sm obtained from the chemical separates (L1-L4) of Kapoeta. The isotopic data are expressed in ϵ units after normalization to $^{147}\text{Sm}/^{152}\text{Sm}$. The standard material used in this study is a chemical reagent as a standard solution for ICP-MS analysis produced from SPEX CertiPrep.

of ^{144}Sm ($\epsilon=+16.6 \pm 3.1$), while the excess of the other fractions L2, L3 and L4 are variable and much less ($0 < \epsilon < +0.75$) than those in L1. Previous studies on ^{144}Sm isotope display a large excess ($\epsilon=+15 \pm 5$) in FUN inclusion C-1 of Allende [13] and small deficits ($\epsilon=-1.18 \pm 0.38$) in bulk carbonaceous chondrites [14] caused by heterogeneous distribution of p-process isotopes in the solar nebula. However, a different reason is required to explain the excess of p-process isotopes in this study. Interestingly, a large isotopic shift of $^{150}\text{Sm}/^{149}\text{Sm}$ due to neutron capture reactions was also found in the fraction L1, corresponding to the neutron fluence of $2.6 \times 10^{16} \text{ n cm}^{-2}$, which is more than 10 times higher than those in the other fractions. The large difference of neutron capture records of L1 from the other three suggests the existence of preirradiation materials migrating into the Kapoeta parent body.

Systematic isotopic data in this study reveal the enrichment of several p-process isotopes of the fraction L1 leached probably from a very surficial part of regolith grains of Kapoeta, suggesting the production of p-process isotopes resulted from the interaction of SCR during the regolith process of the Kapoeta parent body. The result provides isotopic evidence for an activity of the early Sun.

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