

NUCLEOSYNTHETIC AND NEUTRON CAPTURE-INDUCED Sm ISOTOPE ANOMALIES IN CHONDRITES.

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Introduction: The use of the short-lived ^{146}Sm - ^{142}Nd radiometric system in constraining early planetary differentiation hinges on the critical assumption that Sm and Nd isotopes were homogeneously distributed in the solar nebula. Recent studies [e.g., 1, 2, 3 and 4] however, point to considerable variations in ^{142}Nd of bulk chondrites that cannot be explained by radiogenic production. The problem is further compounded by the identification of p-process Sm isotope heterogeneity in carbonaceous chondrites [3]. These observations put a question mark on the suitability of chondrites as a reference frame relative to which Sm-Nd isotope variations in planets can be characterized. A heterogeneous distribution of Sm and Nd in the solar nebula could thus seriously compromise the use of the ^{146}Sm - ^{142}Nd chronometer. This study investigates Sm isotope variations in chondritic meteorites and discusses the likely implications on the use of the ^{146}Sm - ^{142}Nd system for geochronology.

Methods: Bulk samples of chondrites were crushed and powdered in an agate mortar and digested using a $\text{HF-HNO}_3\text{-HClO}_4$ mixture on a hotplate at 120°C . After drying down, the samples were dissolved in HCl and a small aliquot spiked using a mixed ^{149}Sm - ^{150}Nd tracer for determining Sm and Nd concentrations. From the unspiked aliquot, rare earth elements (REE) were separated as a group using cation resin. Sm was separated from the other REE using reverse phase chromatography with columns packed with Teflon powder coated with HDEHP. Because of tailing effects in the elution curves, this technique may lead to small amounts of Nd in the Sm cut that can cause isobaric interference during mass spectrometry. Hence, Sm was further purified by eluting with 0.21 M (pH=4.17) α -hydroxy isobutyric acid on cation resin in ammonia form. A final cleanup column containing cation resin was used to remove the α -hydroxy isobutyric acid. Sm dissolved in HCl was loaded on zone-refined rhenium filaments with $1\mu\text{l}$ of 0.04M H_3PO_4 and measured as Sm^+ on a Finnigan Triton thermal ionization mass spectrometer in the static mode. Potential interferences of Nd and Gd on Sm masses were monitored and corrected for during data acquisition. The isotope ratios were corrected for mass-fractionation using $^{147}\text{Sm}/^{152}\text{Sm}=0.56037$. Repeated analyses of the AMES Sm standard during the course of the analyses gives values of 0.114840 ± 22 ppm, 0.420158 ± 7 ppm, 0.516602 ± 13 ppm, 0.275896 ± 14 ppm and 0.851034 ± 8 ppm for the $^{144}\text{Sm}/^{152}\text{Sm}$, $^{148}\text{Sm}/^{152}\text{Sm}$, $^{149}\text{Sm}/^{152}\text{Sm}$,

$^{150}\text{Sm}/^{152}\text{Sm}$ and $^{154}\text{Sm}/^{152}\text{Sm}$ respectively at 2σ confidence.

Results and discussion: The ^{148}Sm and ^{154}Sm isotope composition of all the analyzed chondrites is mostly identical to the terrestrial standard (Fig. 1). Resolvable anomalies are observed only for ^{144}Sm , ^{149}Sm and ^{150}Sm isotopes (Fig. 1). Of these, the ^{149}Sm - ^{150}Sm variations are coupled and can mostly be attributed to neutron capture by ^{149}Sm to produce cosmogenic ^{150}Sm (Fig. 2). However, the carbonaceous chondrite Acfer 186 and ordinary chondrites Sahara 00225 and El Hammami plot below the theoretical neutron capture line. This could be indicative of very minor nucleosynthetic heterogeneity in these isotopes. The neutron capture-induced ^{149}Sm - ^{150}Sm deviations can be used to calculate a neutron fluence of 1.8×10^{14} – 4.5×10^{15} for the chondrites that are similar to those estimated by other studies [3, 5].

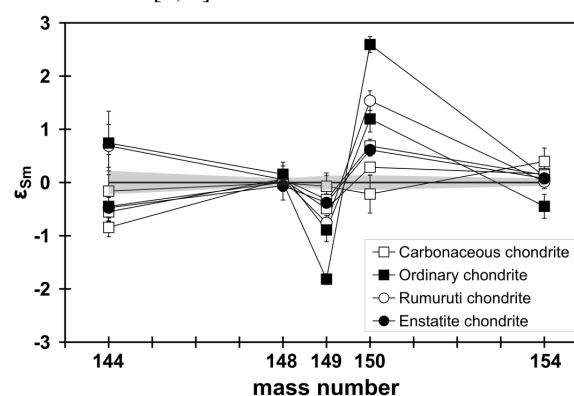


Fig 1. Sm isotope composition of chondrites [$\epsilon_{\text{Sm}} = ((^{x}\text{Sm}/^{152}\text{Sm})_{\text{sample}} / (^{x}\text{Sm}/^{152}\text{Sm})_{\text{standard}} - 1) \times 10^4$]. The grey shaded band denotes the reproducibility of the isotope ratios as determined by repeated analyses of Sm standard solution. Note the presence of resolvable ^{144}Sm , ^{149}Sm and ^{150}Sm anomalies.

^{148}Sm and ^{154}Sm are produced exclusively by the s- and r-process respectively. The lack of any significant heterogeneity in ^{148}Sm and ^{154}Sm within chondrites and terrestrial samples therefore indicate that the products of s- and r-process Sm nucleosynthesis were uniformly distributed in the solar nebula. In contrast, ^{144}Sm in chondrites show large variations some of which are distinct from terrestrial values. The carbonaceous chondrite Acfer 186 and Allende have well resolved ^{144}Sm deficits of -84 ± 18 and -55 ± 27 ppm respectively. Dar Al Gani 412, another carbonaceous chondrite, also

shows negative ^{144}Sm deviations (-16 ± 69 ppm), albeit poorly resolved. In contrast, bulk samples of ordinary chondrites show both deficits and excesses of ^{144}Sm . Within this group, El Hammami shows a well resolved -45 ± 28 ppm negative deviation while Bruderheim has an excess ^{144}Sm of 74 ± 60 ppm. The Enstatite chondrite, Sahara 00225 shows a clearly resolved negative anomaly of -47 ± 24 ppm while the Rumuruti chondrite North West Africa has a 69 ± 40 ppm ^{144}Sm excess.

Since ^{144}Sm is produced exclusively by p-process nucleosynthesis, variations in ^{144}Sm in bulk chondrite samples clearly imply that the solar nebula was heterogeneous with respect to the distribution of p-process Sm isotopes. This is in agreement with a similar inference was made by Andreasen and Sharma [3 and 4]. However, in contrast to these earlier studies [3, 4], it is evident that not only the carbonaceous but also enstatite and some ordinary chondrites have ^{144}Sm deficits. Also noteworthy is the fact that some ordinary and Rumuruti chondrites have a complementary ^{144}Sm excess.

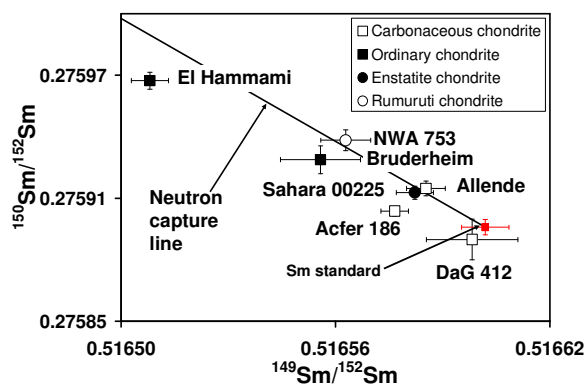


Fig. 2. A plot of $^{150}\text{Sm}/^{152}\text{Sm}$ vs. $^{149}\text{Sm}/^{152}\text{Sm}$ showing that the variations in these isotope ratios are mostly coupled and can be explained as neutron capture by ^{149}Sm to produce ^{150}Sm .

A heterogeneous distribution of ^{144}Sm in chondrites inevitably implies that the now extinct ^{146}Sm , another p-process nuclide, must also have been non-uniformly distributed in the solar nebula. A similar feature is observed for Nd isotopes in chondrites. Recent high precision data [1, 2, 3] indicates the absence of any significant heterogeneity in s- and r-process Nd nuclides but shows considerable variations in ^{142}Nd without the correlated variation in ^{143}Nd or the Sm/Nd. This is an indication that the ^{142}Nd variations may be of nucleosynthetic origin [2, 3]. Since ^{142}Nd has a p-process contribution, this would imply that p-process Nd isotopes were also heterogeneously distributed in the initial solar system. The possibility that both the radioactive parent (i.e., ^{146}Sm) and the radiogenic daughter (i.e.,

^{142}Nd) may have been non-uniformly distributed in the solar nebula could seriously compromise the use of this radiometric system in understanding early planetary processes. Under such conditions, the ^{146}Sm - ^{142}Nd system can be meaningfully used for deciphering early planetary differentiation only if the ^{146}Sm and ^{142}Nd abundances of bulk planetary bodies can be precisely constrained. This gives rise to the corollary that using chondrites as a ^{142}Nd reference relative to which early silicate differentiation on planets is constrained may not be correct.

Conclusions: Except for differences due to the capture of neutrons by ^{149}Sm to produce cosmogenic ^{150}Sm , isotopes produced by s- and r process Sm nucleosynthesis do not show any significant variations within different chondrite groups and are identical to terrestrial values. This implies that s- and r-process Sm isotopes were homogeneously distributed in the solar nebula. In contrast, p-process Sm isotopes were heterogeneously distributed. Coupled with the fact that p-process Nd isotopes may also have been heterogeneously distributed in the solar nebula, this poses a serious challenge for the use of the ^{146}Sm - ^{142}Nd chronometer in understanding silicate differentiation processes in early planetary material.

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