QUANTITATIVE DETERMINATION OF 146Sm/147Sm RATIOS BY ACCELERATOR MASS SPECTROMETRY WITH AN ECR ION SOURCE AND LINEAR ACCELERATION FOR 146Sm HALF-LIFE MEASUREMENT


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Abstract

The alpha-decaying 146Sm nuclide is used for chronology of the Solar System and silicate mantle differentiation in planets. We performed a new determination of 146Sm half-life by measuring 146Sm/147Sm alpha activity and atom ratios in 147Sm activated via (γ,n), (n,2n) and (p,2nα) reactions and obtained a value (68 Myr), smaller than that adopted so far (103 Myr), with important geochemical implications. The experiment required determination of 146Sm/147Sm ratios by high-energy (6 MeV/u) accelerator mass spectrometry to discriminate 146Sm from isobaric 146Nd contaminant. Activated Sm targets were dissolved, chemically purified and reconverted to metallic Sm. Sputter cathodes, made by pressing the Sm metal into high-purity Al holders, were used to feed the Argonne Electron Cyclotron Resonance (ECR) ion source. 146Sm22+ and 147Sm22+ ions were alternately injected and accelerated with the ATLAS linac by proper scaling of ion currents. The high charge states produced in the ECR ion source allow us to accelerate Sm ions at energies of about 6 MeV/u with the ATLAS superconducting linear accelerator for unambiguous ion identification and also eliminates any stable molecules in the injected beam, two basic properties of AMS material. The half-life (t1/2) is obtained through the expression

\[ t_{1/2}^{146} = \frac{A_{147} \times N_{146}}{A_{146} \times N_{147}} \times t_{1/2}^{147} \]

where t147 denotes the α-decay half-life of naturally occurring 147Sm (t147 = 107 ± 0.9 Gyr, [9]). The ratio measurement eliminates most systematic α-activity uncertainties due to detector efficiency and geometrical acceptance. We focus in this contribution on the use of the ECR ion source at the ATLAS facility for accelerator mass spectrometry (AMS) in order to measure N146/N147 atom ratios. The high charge states produced in the ECR ion source allow us to accelerate Sm ions at energies of about 6 MeV/u with the ATLAS superconducting linear accelerator for unambiguous ion identification and also eliminates any stable molecules in the injected beam, two basic properties of AMS [10]. Ion identification was considered important in this experiment because of the need of 146Sm discrimination against stable isobar 146Nd.

EXPERIMENTAL SETUP

Samples of 146Sm to be used in the AMS measurements were prepared from three different activations of enriched 146Sm targets (147Sm(γ,n)146Sm, 147Sm(p,2nα)146Sm and 147Sm(n,2n)146Sm, see [11,12] for details). Following the measurement of the α activity, the sources were dissolved and quantitatively diluted with high-purity natSm to obtain 146Sm/147Sm ratios in the range 10\(^{-7}\)-10\(^{-9}\).
The solutions were purified of Nd impurities by repeated liquid-chromatography steps using a lanthanide-specific resin (Ln resin, manufactured by Eichrom Ltd.). The samples were eventually reduced to high-purity Sm metal, observed to have higher ionization yields in the Electron Cyclotron Resonance (ECR) ion source. This was performed by the following steps for each individual sample: (i) Sm hydroxide was precipitated with an ammonia solution and evaporated to dryness; (ii) ignition to oxide in a quartz crucible at 600°C; (iii) the oxide was thoroughly mixed with freshly filed Zr powder and pressed to 3mm diameter pellets; (iv) the pellets were introduced in a tightly fitting cylindrical tube made of 0.1 mm thick Ta having a 1 mm diameter orifice; (v) the sealed tube was then placed in a high-vacuum (10^{-5} \text{Pa}) evaporator and resistively heated to ~1300°C; (vi) reduced and evaporated metallic Sm was collected by distillation on a water-cooled Cu collector placed just above the 1-mm orifice. The Sm sample was eventually pressed in a holder made of high-purity Al to be used as sputter cathode in the ECR ion source. In order to reduce background of parasitic ions, the chamber walls were lined with a closely fitted cylinder made of quartz (Fig. 1), transparent to the microwave radiation (~100W at 14 GHz) which ignites and maintains the plasma.

Figure 1: Quartz liner tube is inserted in the ECR chamber, tightly fitted to the chamber cylindrical wall, to reduce contamination from ion source structural materials. An orifice (see arrow) allows the sputter cathode to be radially inserted flush to the walls and sample material to feed the plasma in the quartz tube.

Isobaric separation of $^{146}\text{Sm}$ from residual $^{146}\text{Nd}$, beyond the separation power of the accelerator, is achieved in a gas-filled magnetic spectrograph (GFM). The ions are physically separated in the GFM owing to their (Z-dependent) mean charge state in the gas and are unambiguously identified by measurement of their position and differential energy loss in a focal-plane detector (see Fig. 3 and [12] for details). The AMS determination of the atom ratio ($^{146}\text{Sm}/^{147}\text{Sm}$ in this case) is normally performed in our setup by alternate measurements of the $^{146}\text{Sm}$ (accelerated as $^{146}\text{Sm}^{22+}$) count rate in the focal plane detector and the charge current of the stable isotope $^{147}\text{Sm}^{22+}$. The $^{147}\text{Sm}$ beam current is measured in an electron-suppressed Faraday cup positioned in front of the spectrograph. The atom ratio was also measured as the ratio of $^{146}\text{Sm}^{22+}$ counts in the focal-plane detector (see Fig. 3) to $^{147}\text{Sm}^{22+}$ counts in the same detector, after quantitative attenuation of the $^{147}\text{Sm}^{22+}$ beam [13], avoiding systematic effects of ion-transmission efficiency between the Faraday-cup and the detector.
Figure 3: (left panel) Alpha energy spectra measured for (top to bottom) the gamma, neutron and proton activated samples, determining their \( \frac{A_{146}}{A_{147}} \) activity ratios; (right panel) Ion identification spectra of differential energy loss versus position along the focal plane of the gas-filled magnet. The groups corresponding to \(^{146}\text{Sm}\) and stable isobaric \(^{146}\text{Nd}\) (from chemical impurities) for the \( n \)-activated sample are indicated. The lower spectrum corresponds to an unactivated Sm sample.

RESULTS AND DISCUSSION

The results for the different samples are shown in Fig. 4 as the double ratio of \( N_{146}/N_{147} \) measured by AMS to the same ratio as expected from the \( \alpha \)-activity, using the presently adopted half-lives for \(^{146}\text{Sm} \) (103 My) and \(^{147}\text{Sm} \) (107 Gy). The double ratio is equal to the ratio of the determined \(^{146}\text{Sm} \) half-life for each sample to the value adopted in the literature (103 My), with an overall unweighted mean and standard deviation of 0.66 ± 0.07. The mean value of the measured half-life is 68 ± 7 My, substantially (34%) shorter than the literature value. This lower value implies a shorter time interval between (1) isolation of the Solar Nebula from the Interstellar Medium and formation of the Solar System, and (2) formation of the Solar System and planetary silicate differentiation events in the early Solar System, as inferred from \(^{146}\text{Sm}^{142}\text{Nd} \) systematics (see [8] for details).

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