

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

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Abstract

The alpha-decaying ^{146}Sm nuclide is used for chronology of the Solar System and silicate mantle differentiation in planets. We performed a new determination of ^{146}Sm half-life by measuring $^{146}\text{Sm}/^{147}\text{Sm}$ alpha activity and atom ratios in ^{147}Sm activated via (γ,n) , $(n,2n)$ and $(p,2n\epsilon)$ reactions and obtained a value (68 Myr), smaller than that adopted so far (103 Myr), with important geochemical implications. The experiment required determination of $^{146}\text{Sm}/^{147}\text{Sm}$ ratios by high-energy (6 MeV/u) accelerator mass spectrometry to discriminate ^{146}Sm from isobaric ^{146}Nd 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. $^{146}\text{Sm}^{22+}$, $^{147}\text{Sm}^{22+}$ ions were alternately injected and accelerated with the ATLAS linac by proper scaling of ion source and accelerator components. A tightly-fitted quartz cylindrical liner was inserted in the ECR plasma chamber to reduce contamination from the walls. ^{146}Sm ions were eventually counted in a gas-filled magnet and ^{147}Sm ions either measured as charge current or counted after proper attenuation.

INTRODUCTION

The extinct p -process nuclide ^{146}Sm ($t_{1/2} = 103 \pm 5$ Myr [1,2]) was live in the early Solar System, as established through isotopic anomalies of its α daughter ^{142}Nd , first observed in meteorites [3]. The data have been used to estimate a time interval (≈ 70 Myr) between isolation of the Solar Nebula from the interstellar medium and the start of formation of the Solar System [4]. ^{146}Sm acts also as an important geochronometer for the early silicate differentiation in planetary bodies (meteorite parent bodies, Earth, the Moon and Mars, see [5] for a recent review of the field).

The ^{146}Sm half-life which is a crucial component in these applications has been measured four times with values of ~ 50 Myr [6], 74 ± 15 Myr [7] and 103 ± 5 Myr [1,2]. Considering the range of these values, we have performed a new determination of ^{146}Sm half-life [8] by measuring both the α activity ratio (A_{146}/A_{147}) and the atom ratio (N_{146}/N_{147}) in samples prepared from activated ^{147}S material. The half-life ($t_{1/2}^{146}$) is obtained through the expression

$$t_{1/2}^{146} = \frac{A_{147}}{A_{146}} \times \frac{N_{146}}{N_{147}} \times t_{1/2}^{147},$$

where $t_{1/2}^{147}$ denotes the α -decay half-life of naturally occurring ^{147}Sm ($t_{1/2}^{147} = 107 \pm 0.9$ 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 N_{146}/N_{147} 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 ^{146}Sm discrimination against stable isobar ^{146}Nd .

EXPERIMENTAL SETUP

Samples of ^{146}Sm to be used in the AMS measurements were prepared from three different activations of enriched ^{147}Sm targets ($^{147}\text{Sm}(\gamma,n)^{146}\text{Sm}$, $^{147}\text{Sm}(p,2n\epsilon)^{146}\text{Sm}$ and $^{147}\text{Sm}(n,2n)^{146}\text{Sm}$, see [11,12] for details). Following the measurement of the α activity, the sources were dissolved and quantitatively diluted with high-purity ^{nat}Sm to obtain $^{146}\text{Sm}/^{147}\text{Sm}$ 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} Pa) evaporator and resistively heated to $\sim 1300^\circ\text{C}$; (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 ($\sim 100\text{W}$ at 14 GHz) which ignites and maintains the plasma.

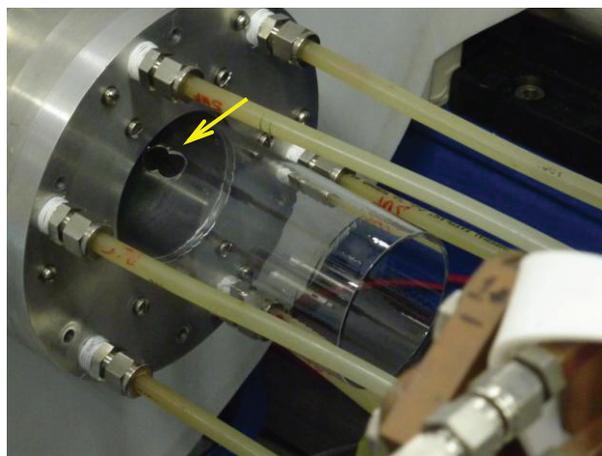


Figure 1: Quartz liner tube is inserted in the ECR chamber, tightly fitted to the chamber cylindrical wall, to reduce contamination from ion source structural materials. An orifice (see arrow) allows the sputter cathode to be radially inserted flush to the walls and sample material to feed the plasma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

$^{146}\text{Sm}^{22+}$ and $^{147}\text{Sm}^{22+}$ ions, alternately selected by magnetic analysis, are bunched and accelerated to a final energy of 6 MeV/u in the superconducting linear accelerator. Acceleration of neighboring isotopes such as ^{146}Sm and ^{147}Sm , both in 22+ charge state, requires adjustment of resonator field amplitudes and ion-optical elements by scaling a master tune according to the m/q ratios of the atomic mass and charge state of the ions, in order to conserve an exact velocity profile. The

accelerator system acts then as a high-energy accelerator mass spectrometer and its abundance sensitivity (defined as the abundance ratio of two resolvable neighboring isotopes) before the detection system was shown [10] to be $\sim 5 \times 10^{-12}$ for Pb isotopes, separating in the present experiment ^{146}Sm from ^{147}Sm . The reliability of the m/q scaling procedure to transport alternately $^{146}\text{Sm}^{22+}$ or $^{147}\text{Sm}^{22+}$ from the ECRIS to the detection setup was checked by measuring the ratio of charge currents for $^{147}\text{Sm}^{22+}$ and $^{152}\text{Sm}^{22,23+}$. The measured $^{152}\text{Sm}/^{147}\text{Sm}$ (1.8 ± 0.2), after correcting when necessary for the 23+/22+ population ratio (measured experimentally), is consistent with the natural abundance ratio 1.78 (Fig. 2)

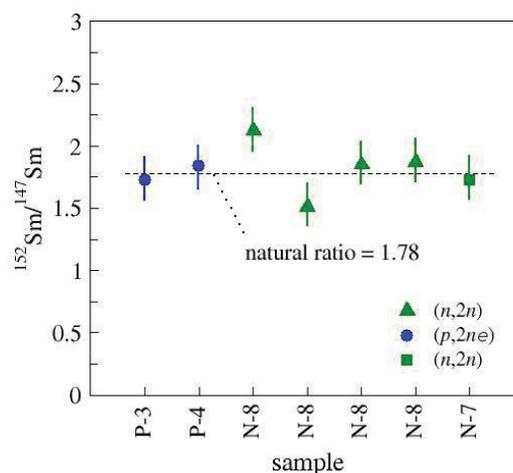


Figure 2: Stable $^{152}\text{Sm}/^{147}\text{Sm}$ isotopic ratio measured by scaling the ECR and ATLAS elements, based on the mass-to-charge ratio of the isotopes. The symbols P-x and N-x on the x-axis represent samples prepared from the proton-activated and neutron-activated ^{147}Sm targets, respectively. The measured ratio is consistent with the natural abundance ratio of 1.78.

Isobaric separation of ^{146}Sm from residual ^{146}Nd ($(m/q)/\delta(m/q) \sim 2.4 \times 10^6$), 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}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}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.

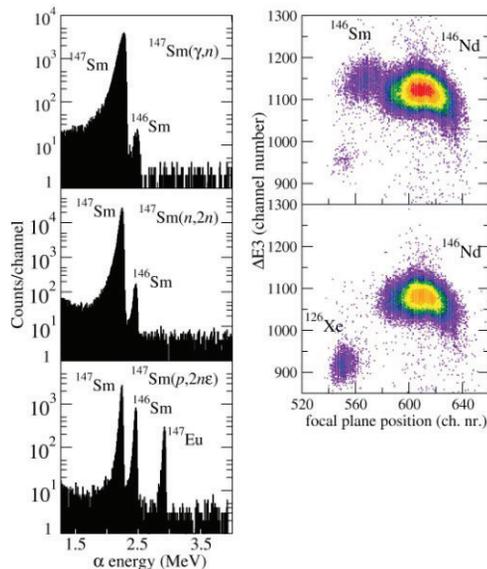


Figure 3: (left panel) Alpha energy spectra measured for (top to bottom) the gamma, neutron and proton activated samples, determining their (A_{146}/A_{147}) activity ratios; (right panel) Ion identification spectra of differential energy loss versus position along the focal plane of the gas-filled magnet. The groups corresponding to ^{146}Sm and stable isobaric ^{146}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 α -activity, using the presently adopted half-lives for ^{146}Sm (103 My) and ^{147}Sm (107 Gy). The double ratio is equal to the ratio of

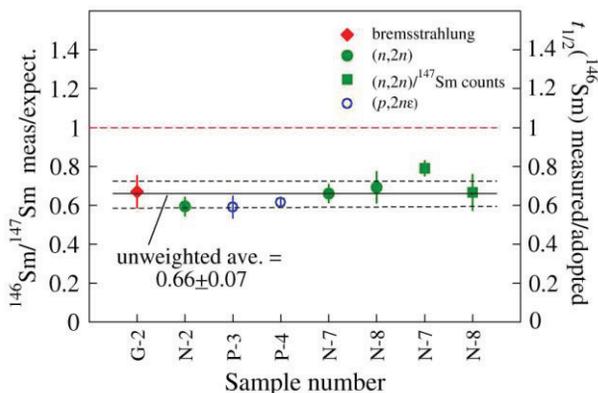


Figure 4: Double ratios of N_{146}/N_{147} atom ratios measured by AMS to those expected from the α activity ratios of the different samples. G-x, N-x and P-x represent the gamma, neutron and proton activated samples. The double ratios are equal to the ratios of the determined half-life for each sample to that adopted in the literature.

the determined ^{146}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}Sm - ^{142}Nd systematics (see [8] for details).

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