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Shorter half-life of *p*-process ^{146}Sm measured and ^{146}Sm/^{142}Nd chronology of the Solar System

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The short-lived nuclide ^{146}Sm, synthesized in stellar events by the *p*-process and now extinct in the Solar System, serves as both an astrophysical and geochemical chronometer through measurements of isotopic anomalies of its α-decay daughter ^{142}Nd. Using artificially produced ^{146}Sm via ^{147}Sm(γ,*n*), ^{147}Sm(*n*,2*n*) and ^{147}Sm(*p*,*n*ε) reactions, we performed a new measurement of the ^{146}Sm half-life by measuring the

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^{146}Sm/^{147}Sm alpha activity with a Si surface barrier detector and the ^{146}Sm/^{147}Sm atom ratio with accelerator mass spectrometry (AMS). Our result, $t^{146}_{1/2}$ = 68 ± 7 (1σ) million years (My), is significantly shorter than the adopted value (103 ± 5 My). The shorter ^{146}Sm half-life value implies a higher initial Solar System ratio, (^{146}Sm/^{144}Sm)$_0$ = 0.0094 ± 0.0005 (2σ), than the recently derived value 0.0085 ± 0.0007(2σ). The time interval between isolation of the Solar Nebula from the interstellar medium and formation of the first solids, is reduced by a factor of ~2.5 to 20 from previous estimates. Early planetary mantle differentiation processes on Earth, the Moon and Mars dated by ^{146}Sm-^{142}Nd converge to a shorter time span, due to the combined effect of the new ^{146}Sm half-life and (^{146}Sm/^{144}Sm)$_0$ values.

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1. Introduction

The *p*-process nuclide ^{146}Sm ($t_{1/2}$= 103 ± 5 My [1,2]), now extinct, was live in the early Solar System as established through isotopic anomalies of its α daughter ^{142}Nd, first observed in meteorites [3]. The initial ^{146}Sm/^{144}Sm abundance (see also [4] in these proceedings), as that of other short-lived nuclides ($t_{1/2} \leq 100$ My) present in the early Solar System, has been used to study the time evolution of nucleosynthesis products in the Interstellar Medium until the start of formation of the Solar System [5,6]. Samarium-146 acts also as an important geochronometer for the early silicate mantle differentiation in planetary bodies (meteorite parent bodies, Earth, the Moon and Mars, see [7] for a recent review of the field). The ^{146}Sm half-life has been measured four times with values of ~50 My [8], 74 ± 15 My [9] and 103 ± 5 My [1,2]. Considering the range of these values and its importance for Solar System evolution and chronology, we have performed a new determination of ^{146}Sm half-life, leading to a shorter value of 68 ± 7 My [10]. We describe below the measurement and implications of this value.

2. Experimental method and results

We have performed a new determination of ^{146}Sm half-life [10] by measuring both the ^{146}Sm/^{147}Sm α activity ratio (A_{146}/A_{147}) and the atom ratio (N_{146}/N_{147}) in activated ^{147}Sm samples. The ^{146}Sm half-life ($t_{1/2}^{146}$) is obtained through the expression

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

where $t_{1/2}^{147} = 107 \pm 0.9$ Gy [11] is the α-decay half-life of naturally occurring ^{147}Sm. The measurement of ratios eliminates some of the systematic uncertainties in α-activity (detector efficiency and geometrical acceptance) and in determination of absolute number of atoms. Samples of ^{146}Sm were prepared from three different activations of enriched ^{147}Sm targets (^{147}Sm(γ,*n*)^{146}Sm, ^{147}Sm(*p*,2*nε*)^{146}Sm and ^{147}Sm(*n*,2*n*)^{146}Sm, see [12,13] for details). The α activities were measured with a surface barrier detector. Accelerator mass spectrometry (AMS) was used to measure the atom ratios because of the need to discriminate ^{146}Sm from stable isobaric ^{146}Nd impurities. Following the measurement, the sources were dissolved and quantitatively diluted with high-purity ^{nat}Sm to obtain ^{146}Sm/^{147}Sm atom ratios in the range 10^{-7}-10^{-9}. The AMS measurements were performed at the Argonne ECR-ATLAS facility by accelerating highly-charged ions ($^{146}Sm^{22+}$) produced in the Electron Cyclotron Resonance (ECR) source to an energy of 6 MeV/u and detected using the Enge gas-filled spectrograph for ion identification (see [13] for details). Fig. 1 shows the alpha energy and ion identification spectra. The double ratios of the ^{146}Sm/^{147}Sm isotopic abundances measured by using AMS to those derived from the A_{147}/A_{146} activity ratios using the adopted ^{146}Sm half-life (103 My) are plotted in fig.2 for the independently activated samples; the double ratio is equivalent to the ratio of the measured half-life (Eq. (1)) to the adopted ^{146}Sm half-life. From fig.2, our determination of this half-life is 68 ± 7 My, significantly shorter than the adopted value.

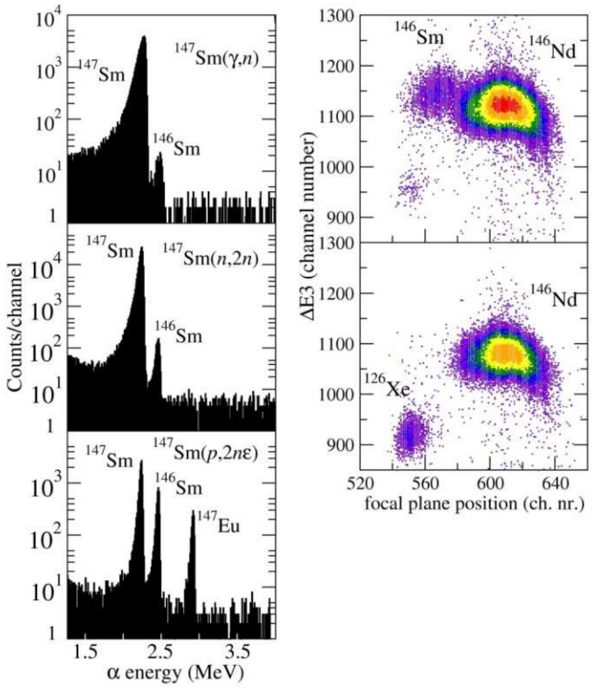

Figure 1: (left panel) Alpha energy spectra measured for (top to bottom) the gamma, neutron and proton activated samples, determining their (A_{147}/A_{146}) activity ratio; (right panel) Ion identification spectra of differential energy loss versus position along the focal plane of the gas-filled magnet for the *n*-activated sample (top). The lower spectrum corresponds to an unactivated (natural) Sm sample. The groups corresponding to ^{146}Sm and stable isobaric ^{146}Nd (from chemical impurities) are indicated. The ^{126}Xe group shown results from residual gas in the ion source.

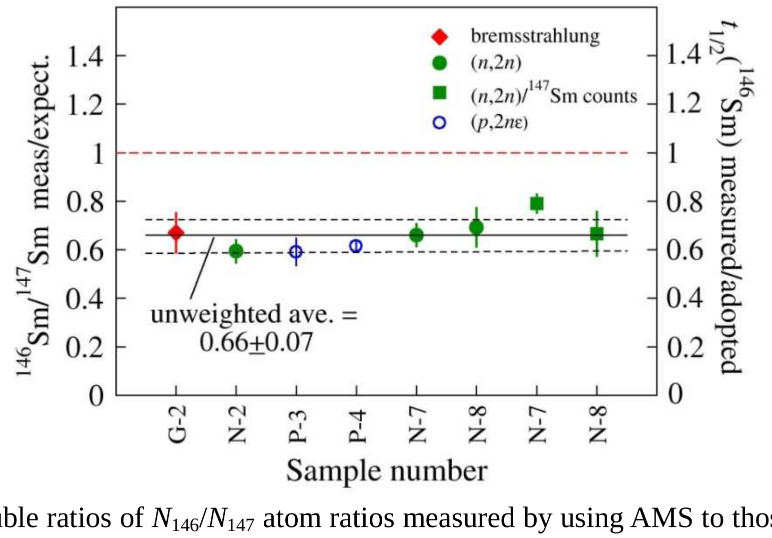

Figure 2: Double ratios of N_{146}/N_{147} atom ratios measured by using AMS to those expected from the α activity ratio of the different samples. G-x, N-x and P-x represent the gamma, neutron and proton activated samples. The double ratios are equal to the ratios of the determined half-life for each sample to that adopted in the literature. Data points represented by a diamond and circles were measured as the ratio of ^{146}Sm counts versus $^{147}Sm^{22+}$ ion beam current; square symbols represent ratios of ^{146}Sm counts versus quantitatively attenuated ^{147}Sm counts in the same detector.

3. Implications to Solar System chronology

The initial abundance (r_0) of ^{146}Sm at the time of Solar System formation, usually expressed relative to the other Sm *p*-process nuclide ^{144}Sm (stable), can be determined by extrapolation of ^{146}Sm/^{144}Sm values extracted from individual meteorites of known ages to the age of the Solar System, 4,568 My. In these studies, the r_0= (^{146}Sm/^{144}Sm)$_0$ value is determined from the correlation between the ^{142}Nd/^{144}Nd isotopic anomaly of the ^{146}Sm alpha daughter ^{142}Nd and the chemical Sm content (expressed as ^{144}Sm/^{144}Nd) in different mineral phases of the meteorite. Boyet et al. [14] selected a set of suitable meteorites (i.e., that remained a closed system with respect to Sm-Nd) and derived a value r_0= 0.0085 ± 0.0007 (2σ) for the initial ^{146}Sm/^{144}Sm ratio in the Solar System. Figure 3 presents a re-interpretation of the available data using our measured value of the ^{146}Sm half-life, leading to a higher initial ratio of 0.0094 ± 0.0005 (2σ).

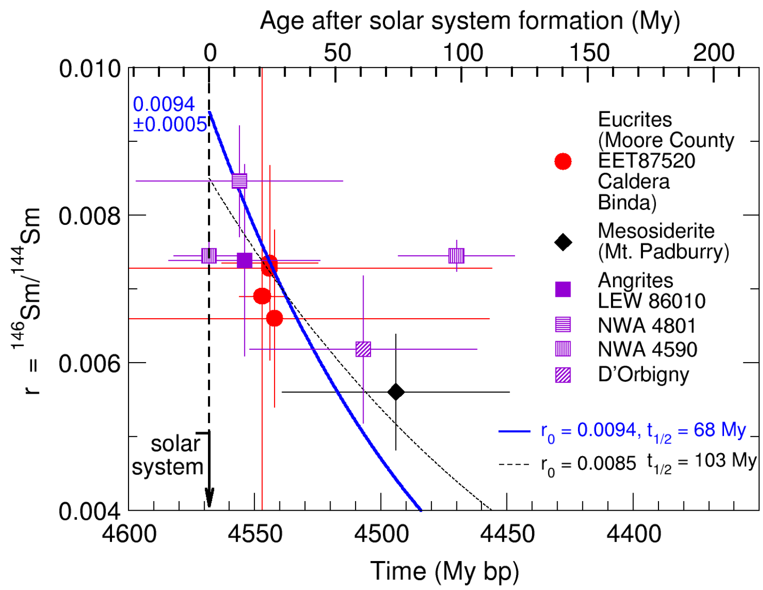

Figure 3: Reinterpretation of the (^{146}Sm/^{144}Sm)$_0$ initial ratio measured in selected meteorites ([14] and references therein) plotted against time in My before the present (bp). The time of solar system formation is taken as 4,568 My bp. The set of meteorites selected by Boyet *et al.* [14] as closed Sm-Nd systems is used here: 4 eucrites (Moore County, EET87520, Caldera, Binda), one mesosiderite (Mt. Padburry) and one angrite (LEW 86010). The data are fitted here by a new decay curve (solid blue line) using the ^{146}Sm half-life value measured in this work ($t_{1/2}^{146}$= 68 My) and yield an initial solar system ratio r_0= (^{146}Sm/^{144}Sm)$_0$ = 0.0094 ± 0.0005 (2σ), compared to the curve from Boyet *et al.* [14] (dashed), who used $t_{1/2}^{146}$= 103 My and yielded r_0= 0.0085 ± 0.0007 (2σ). Recent data [15,16] on angrite meteorites NWA 4801, 4590 and D'Orbigny are included in the figure. NWA 4590, whose ages determined by ^{147}Sm (4568 ± 27 My [15]) and by Lu-Hf (4470 ± 23 My [16]) are inconsistent and D'Orbigny (reported to be disturbed [16]) were not included in the present fit.

The initial abundances of short-lived nuclides in the Solar System, combined with estimated production yields obtained from nucleosynthesis calculations, provide constraints on the time evolution of Galactic nucleosynthesis. Wasserburg et al. [5] used the Uniform Production (UP)

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closed-box model (without Galactic-disk enrichment in low-metallicity gas from the halo) which assumes a quasi-steady state of short-lived nuclides abundance in the Interstellar Medium (ISM). The steady-state nuclide abundances are achieved by a balance between stellar production and free decay. With the previous ^{146}Sm values ($t_{1/2}^{146}$ = 103 My and (^{146}Sm/^{144}Sm)$_0$ = 0.01), Wasserburg et al. [5] estimated an isolation time interval Δ ~ 70 My between formation of the Solar nebula and formation of the first solids. With the ^{146}Sm values derived here, $t_{1/2}{}^{146}$ = 68 My and r_0 = 0.0094, Δ decreases to ~5 My. This value is closer to the values calculated from some other nuclides, e.g., ^{182}Hf/^{180}Hf. Huss et al. [6] have used a more comprehensive 3-phase open-box mixing model, proposed by Clayton [17]. Figure 4 (adapted from [6]) shows initial Solar System abundances normalized to production rates for a number of short-lived

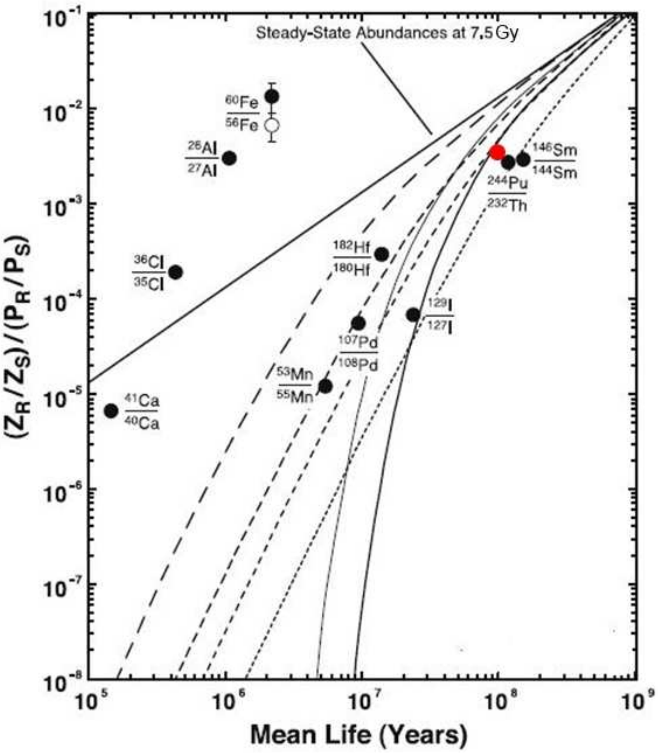

Figure 4: (Adapted from [6]) Initial abundance (solid dots) of short-lived radioactive nuclides (Z_R) relative to a stable species (Z_S) normalized by the respective nucleosynthetic production ratio (P_R/P_S) plotted versus the radioactive nuclide mean life. The solid lines (left to right) correspond to the quasi-steady state model (equivalent to the UP model, see text) with an isolation time interval Δ of 0, 50 and 100 My). The dashed lines correspond to a 3-phase model of the ISM with two mixing times between these phases, (left to right) $T_1 = T_2$ = 10, 50, 100 and 300 My (see [6,17] for details).The solid red dot represent the ^{146}Sm/^{144}Sm ratio using our present values $t_{1/2}^{146}$ = 68 My and r_0 = 0.0094. Within this model, the point lies close to the Δ= 100 My curve.

nuclides vs. their mean lives. With the previous ^{146}Sm values, the ^{146}Sm/^{144}Sm point (upper right part of plot) is positioned close to 3-phase model of the ISM [6,17] with $T_1 = T_2$ = 300 My curve (rightmost dashed curve), as are the ^{129}I/^{127}I and ^{244}Pu/^{232}Th points. T_1 and T_2 represent respectively mixing time between a (star-forming) molecular cloud phase and large neutralhydrogen (H I) clouds not affected by supernovae shocks, and between the latter clouds with a phase made of smaller H I clouds heated by supernovae shocks. The revised ^{146}Sm/^{144}Sm point (solid red circle) is positioned now close both to the free-decay curve with Δ = 100 My and to the 3-phase model with $T_1 = T_2$ = 100 My. Figure 4 clearly shows the complexity of the issue of explaining simultaneously the abundances of short-lived nuclides in the Early Solar System. The values derived here reduce significantly the ^{146}Sm residence times, independent of model.

The ^{146}Sm/^{142}Nd system has also become an important tool for chronology of early silicate mantle differentiation processes in planetary bodies [7,18]. Terrestrial, Lunar, and Martian planetary silicate mantle differentiation events dated with ^{146}Sm-^{142}Nd converge to a shorter time span and in general to earlier times, due to the combined effect of the new ^{146}Sm half-life and (^{146}Sm/^{144}Sm)$_0$ values [10]. The revised ^{146}Sm-^{142}Nd age of lunar ferroan anorthosite 60025, recently dated with precision [19], becomes consistent with ages based on the ^{147}Sm-^{143}Nd and Pb-Pb chronometers (see fig. 5).

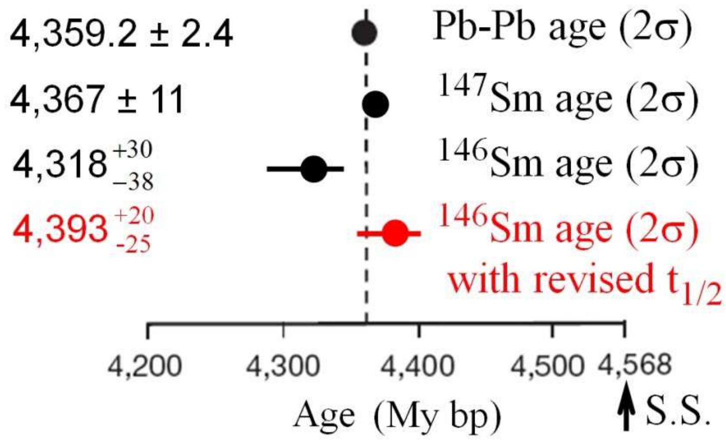

Figure 5: Dating of Lunar ferroan anorthosite 60025 by Pb-Pb, ^{147}Sm and ^{146}Sm chronometers [19]. The ^{146}Sm age (175 $^{+25}_{-20}$ My), derived using the revised values $t^{146}_{1/2}$= 68±7 My and r_0= 0.0094±0.0005 is in better agreement with the precise Pb-Pb and ^{147}Sm ages (208.8±2.4 and 201±11 My, respectively [19]). The horizontal axis represents the sample age in millions of years before the present (My bp).

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