High Precision Half-Life Measurement of the Extinct Radio-Lanthanide Dysprosium-154

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Abstract

Sixty years after the discovery of ^{154}Dy , the half-life of this pure alpha-emitter was re-measured. ^{154}Dy was radiochemically separated from proton-irradiated tantalum samples. Sector field- and multicollector-inductively coupled plasma mass spectrometry were used to determine the amount of ^{154}Dy retrieved. The disintegration rate of the radio-lanthanide was measured by means of α -spectrometry. The half-life value was determined as $(1.33 \pm 0.07) \cdot 10^6$ y, with an uncertainty reduced by a factor of ~ 10 compared to the currently adopted value of $(3.0 \pm 1.5) \cdot 10^6$ y. This precise half-life value is crucial for the correct estimation of p-process nucleosynthetic reactions in the lanthanide region, as well as for the safe disposal of irradiated target material from spallation facilities. As a first application of the half-life value found in this work, the excitation functions for the production of ^{154}Dy in proton-irradiated Ta, Pb, and W targets were re-evaluated, which found to be in agreement with theoretical calculations.

1. Introduction

Radionuclides with half-lives ($t_{1/2}$) in the order of few millions of years play an essential role in the description and in the dating of recent galactic events [1–3]. Even though such radionuclides became extinct soon after the formation of the Solar System, it is possible from the analysis of the abundances of their stable decay products to reconstruct the nucleosynthetic processes that occurred millions of years ago. Prominent examples can be found in [4–7]. Over the past two decades, there has been a strong effort in the description of the mechanism responsible for the synthesis of p-nuclei. The latter is a group of proton-rich isotopes between ^{74}Se and ^{196}Hg that cannot be produced by either slow neutron-capture (s-process) or rapid neutron-capture (r-process) reactions [8, 9]. Instead, they are produced in a network of photodisintegration and charged particle reactions called p-process [10]. Among the different benchmark radionuclides for p-process studies, ^{154}Dy ($I_{\alpha} = 100\%$ at $E_{\alpha} = 2.7$ MeV) [11] is of particular interest. Through the nuclear reactions $^{154}\text{Dy}(\alpha, \gamma)^{158}\text{Er}$ [12], $^{154}\text{Dy}(\gamma, \alpha)^{150}\text{Gd}$, and $^{154}\text{Dy}(\gamma, 2n)^{152}\text{Dy}$ [13], this exotic radionuclide is crucial in the synthesis of ^{142}Nd , ^{146}Sm , ^{152}Gd , and ^{158}Dy – all nuclides shielded from s- and r-processes by their respective isobars. Furthermore, the pure α -decay series of ^{154}Dy (i.e., $^{154}\text{Dy} \rightarrow ^{150}\text{Gd} \rightarrow ^{146}\text{Sm} \rightarrow ^{142}\text{Nd}$) influences both the abundance of ^{146}Sm [14] and the natural isotopic composition of stable Nd [15, 16], directly contributing thus to the $^{146}\text{Sm}/^{142}\text{Nd}$ chronometer. The latter is one of the most powerful tools to date silicate differentiation and primitive crust formation events occurred during the first hundreds of million years (My) of the Solar System [17–20]. A direct experimental measurement of most of the p-process reaction rates is not yet accessible. Hence, calculations relying on models of stellar nucleosynthesis are applied [21, 22]. These calculations, that correlate the observed isotopic abundances to specific nuclear reaction paths, depend on the reactions cross-sections and on the half-life values of all the nuclides involved. Therefore, for a precise evaluation of the production and destruction processes in the lanthanide region, exact knowledge of the pure α -emitter ^{154}Dy half-life value is essential. Besides important applications in cosmo and geoscience, a precise value for the half-life of ^{154}Dy is also required for safety evaluation of spallation target facilities,

such as Accelerator Driven Systems and/or Spallation Neutron Source Facilities, that utilize targets made of heavy metals – e.g., lead, tantalum, tungsten [23]. During the spallation process, exotic α -emitting radionuclides like ^{154}Dy are produced [24]. Recent theoretical calculations [25] estimated the contribution of ^{154}Dy , ^{146}Sm , and ^{148}Gd to the overall radiotoxicity of spallation targets to be similar to that of Po radionuclides. Precise nuclear data for the production and decay of ^{154}Dy are thus needed in order to estimate operational mode and maintenance of such facilities, as well decommissioning of the irradiated material.

Despite its relevance, since the discovery of ^{154}Dy in 1961 [26] an accurate half-life value for this exotic radionuclide is still not available. Instead, a plethora of half-life values can be found in literature, summarized in Table 1.

Table 1
Half-life values for ^{154}Dy available from literature.

Year	Half-life value (My)	Reference
1961	1.5 ± 0.9	[26]
1965	2.9 ± 1.5	[27]
1971	4	[28]
1985	3.0 ± 1.5	[29]
1991	1.2	[30]

The nowadays-accepted half-life value for ^{154}Dy , i.e., $t_{1/2} = 3.0$ My is associated with a 50% uncertainty and derives from the revision made in [29] of the works of MacFarlane [26] and Gono [28]. The half-life value in [30] was calculated by applying a cluster model for the ground state alpha decay of even-even nuclei. In most cases, little is known about the experimental procedure or about the effects taken into account in the calculation of the relative uncertainties, rendering a re-evaluation of the mentioned works difficult. Reasons for such a lack of reliable nuclear data are mainly the limited availability of suitable sample material, together with inherent complications in performing accurate activity measurements with long-lived nuclides. To overcome those difficulties, the initiative “Exotic Radionuclides from Accelerator Waste for Science and Technology – ERAWAST” was launched in 2006 at Paul Scherrer Institute (PSI) [31, 32]. This long-term project aims, among others, to improve the existing nuclear databases, with a special focus on the re-determination of uncertain decay data. For this purpose, the necessary exotic radionuclides are obtained by reprocessing radioactive waste already available at the PSI site. In this work, we report on a high-precision half-life measurement of ^{154}Dy , performed in the framework of ERAWAST. ^{154}Dy material was obtained by reprocessing Ta samples irradiated with protons and spallation neutrons during the SINQ Target Irradiation Program (STIP) at PSI [33]. For the estimation of half-lives in the order of millions of years, the so-called *direct method* is applied. This consists of the

determination of the number of radioactive atoms in a specific sample, combined with the measurement of its radioactivity. Here, the number of atoms of ^{154}Dy was determined using sector field inductively-coupled plasma mass spectrometry (SF-ICP-MS) and multicollector inductively-coupled plasma mass spectrometry (MC-ICP-MS). The specific activity of ^{154}Dy in the sample was measured by means of α -spectroscopy. Thin and homogeneous radioactive sources, necessary to obtain high-resolution α -spectra, were prepared using the molecular plating technique - also referred to as electrodeposition [34, 35]. Following the *Guide to the expression of uncertainty in measurement – GUM* recommendations [36, 37], a realistic and complete uncertainty budget for the measured $t_{1/2}$ is given as well.

2. Experimental Techniques And Methodology

2.1 Separation and purification of the ^{154}Dy sample

The Dy fraction containing ^{154}Dy was obtained from the reprocessing of 4 Ta samples from the STIP-II project. The procedure for the dissolution of the Ta samples is described in detail in [38]. Successively, a series of ion-exchange separation processes allowed us to obtain a purified Dy fraction in 1 M HNO_3 . During the separation process, the γ -emitter ^{159}Dy ($t_{1/2} = 144.4$ d, $I_\gamma = 2.29\%$ at $E_\gamma = 58$ keV [39]) was added as an internal radio-tracer. The separation method for the retrieval of the Dy fraction is reported in detail in [40]. A scheme of the chemical separation steps is reported in Figure 1.

The homogeneous Dy fraction (in 1 M HNO_3) was collected in a scintillation vial (HDPE material, capacity: 20 ml). The total mass of the collected Dy solution – from now onwards referred to as “Dy master solution” – was determined gravimetrically (averaged value of 5 consecutive weightings: 6.79404 g, see Table S1). All the gravimetric steps were performed on a certified Mettler-Toledo XP56 balance (10^{-6} g scale interval), in a room with controlled temperature within 20–23°C.

2.2 Mass spectrometric analysis

The concentration of ^{154}Dy in the Dy master solution was calculated from the amount of ^{161}Dy (deduced by SF-ICP-MS) and from the $^{154}\text{Dy}/^{161}\text{Dy}$ isotope ratio in solution (determined by MC-ICP-MS). ^{161}Dy was chosen as reference nuclide due to the absence of isobaric interferences for mass 161. All gravimetric additions were done on a Mettler-Toledo XP56 balance.

SF-ICP-MS measurements. SF-ICP-MS analyses were conducted using a Thermo Scientific Element 2 spectrometer, applying the medium mass resolution setting in order to minimize potential effects of molecular interferences. The plasma was operated at 1350 W. All analytes were introduced into a cyclonic PFA spray chamber using an ELEMENTAL SCIENTIFIC PFA-ST nebulizer and a peripump set, with a sample consumption of ca. $130 \mu\text{l}\cdot\text{min}^{-1}$. An external linear calibration was used to establish the ^{161}Dy concentration in the Dy master solution. In this procedure, several dilutions of a Dy standard stock solution ($10 \text{ mg}\cdot\text{l}^{-1}$ in 2% HNO_3 , density: $1.00885 \text{ g}\cdot\text{ml}^{-1}$, concentration uncertainty: 2%, Sigma Aldrich) were repeatedly analyzed (before, in-between, and after the replicate analysis of the sample solution). In

all the Dy dilutions used for the external calibration, a Re standard stock solution ($10 \text{ mg} \cdot \text{l}^{-1}$ in 2% HNO_3 , density: $1.00885 \text{ g} \cdot \text{ml}^{-1}$, concentration uncertainty: 2%, ESI, Elemental Scientific, Omaha, NE, USA) was added as an internal reference. This allowed for cancelling potential temporal drift in instrumental signal response or plasma instability. The series of dilutions used for the external calibration scheme is presented in Table S2. An aliquot of the Dy master solution (averaged value of 5 consecutive weightings: 0.03000 g , see Table S3) was used for mass-spectrometry analysis. To this Dy aliquot, the same Re ESI standard stock solution used in the preparation of calibration standards were added as an internal reference. The Dy aliquot was then diluted with a 0.28 M HNO_3 solution, to a total weight of 13.90972 g (averaged value of 5 consecutive weightings – see Table S3). Instrumental background signals (including potential imperfect washout between analytes) were subtracted by repeated analysis of the same acid used to prepare the external standards and the sample analytes. Each of these “blank” measurements preceded the standard and the sample analyses. The ^{161}Dy content in the Dy master solution was obtained by correlating the background-corrected and Re-normalized ^{161}Dy signal to the external calibration line.

MC-ICP-MS measurements. Dy isotopic ratio analysis was conducted on the Nu Instruments Plasma 3 MultiCollector Inductively Coupled Plasma Mass Spectrometer (MC-ICP-MS) equipped with an inductively coupled Ar-plasma ion source, 16 Faraday cups, 3 Daly detectors, and 3 secondary electron multipliers. These instrumental characteristics allow for the simultaneous measurement of up to 22 ion beams. Analytes were introduced into the system using an Elemental Scientific Apex HF desolvating nebulizer and a self-aspiring Elemental Scientific PFA-ST Microflow at a consumption rate of ca. $50 \mu\text{l} \cdot \text{min}^{-1}$. The plasma was operated at 1350 W forward power. Ion beams of masses 149 (Sm), 152 (Sm, Gd), 154-164 (Sm, Gd, Tb, Dy), 166-167 (Er), and 170 (Er, Yb) were collected simultaneously in Faraday cups connected to amplifier systems with a $10^{11} \Omega$ resistors in their feedback loop. To assess potential isobaric interferences of Yb, mass 172 was monitored using a Daly ion counting detector. An aliquot of the Dy master solution was diluted by a factor of ca. 500 by addition of a pure 0.28 M HNO_3 solution. To the diluted Dy aliquot, $^{\text{nat}}\text{Er}$ was added, allowing for an empirical semi-external mass discrimination correction. Successively, six analyses of the so-prepared Dy sample were bracketed by 10 analyses of mixed Er-Dy solution standards. Each analysis consisted of 60 ten-second-long integrations of the ion beam intensities. Instrumental background signals were removed using interspersed analysis of the Dy sample and of the 0.28 M HNO_3 solution used in the preparation of the analytes. Online recorded $^{170}\text{Er}/^{166}\text{Er}$ values of the admixed Er were used to determine the magnitude of instrumental mass discrimination during the analysis of the Dy sample.

2.3 Preparation of the ^{154}Dy α -source for activity measurements

For the preparation of a thin radioactive source with the molecular plating technique, an aliquot of the Dy master solution (averaged value of 5 consecutive weightings: 2.77410 g - see Table S4) was used. The estimation of the deposition efficiency was necessary to determine the effective number of ^{154}Dy atoms

plated. The deposition yield was determined by monitoring the activity of the γ -tracer ^{159}Dy added during the separation process (see Section 2.1). Specifically, the activity of ^{159}Dy in the Dy aliquote before molecular plating was measured, and compared to the activity of the ^{159}Dy plated on the deposition foil. Since isotopes of the same element behave chemically identically, the yield of deposited ^{159}Dy is thus equal to the yield of deposited ^{154}Dy . For a reliable deduction of the deposition yield, both ^{159}Dy γ -activity measurements had to be performed in equal geometries. This was achieved by using a custom-made holder made of two interchangeable parts (see Figure 2), that allowed for performing γ -spectrometry measurements in two geometrically equivalent positions, namely Position A (used to quantify the activity of ^{159}Dy before electrodeposition), and Position B (used to quantify the activity of ^{159}Dy after electrodeposition). Technical drawings in scale of the holder are given in the Supplementary Information, Figure S1. In both positions, the distance between the radioactive source and the detector endcap was 5 cm. This distance was large enough to consider true coincidence summing effects negligible.

γ -spectroscopy measurements were performed with a BEGeTM (Broad Energy Germanium γ -detector, Mirion Technologies (Canberra), Inc.). Data acquisition and analysis were done using the GenieTM 2000 Gamma Acquisition & Analysis Software. Energy calibration was done using a ^{152}Eu ($t_{1/2} = 13.53$ y, $I_\gamma = 28.41\%$ at $E_\gamma = 121.78$ keV) point-source (Physikalisch-Technische Bundesanstalt – PTB). The energy resolution FWHM (Full Width at Half Maximum) was 0.540 keV at 58.1 keV. Efficiency calibration was performed with a ^{133}Ba ($t_{1/2} = 10.54$ y, $I_\gamma = 32.9\%$ at $E_\gamma = 80.99$ keV) calibrated source in HNO_3 1 M (see Section 5 of the Supplementary Information for details). For Position A, a known amount of the ^{133}Ba calibrated liquid source was put into the PEEK vial, evaporated to dryness under a N_2 flow at $T = 70^\circ\text{C}$, and dissolved in 400 μl of 1 M HNO_3 . Efficiency calibration in Position A gave a FEPE (full energy peak efficiency, ϕ) of $(15.75 \pm 0.25)\%$ at 58 keV. For Position B, a known amount of the ^{133}Ba calibrated liquid source was drop-deposited onto a graphite foil (thickness: 15 μm , purity: 99.8%, Flexible Graphite, GoodFellow). The liquid was evaporated by heating the graphite foil at $T = 70^\circ\text{C}$. Efficiency calibration in Position B gave a $\phi = (15.36 \pm 0.24)\%$ at 58 keV. Details on the efficiency calibrations in Position A and B are given in Section 5 of the Supplementary Information.

γ -activity measurements before molecular plating (Position A). For the γ -spectrometry measurement in Position A, the Dy aliquot was transferred from the HDPE vial to a custom-made polyether ether ketone (PEEK) vial (internal diameter: 20 mm, thickness at the bottom: 1 mm), and evaporated to dryness at 70°C under a N_2 gas flow. To ensure a complete transfer of the Dy aliquot, the HDPE vial was rinsed with 10 ml 1 M HNO_3 , transferring the washing solution to the PEEK vial, and evaporating the liquid to dryness. This process was repeated 5 times. Then, 400 μl of 1 M HNO_3 were added in order to dissolve the dried Dy solid. The added volume corresponded to the minimum volume that would entirely cover the bottom of the PEEK vial. This step was necessary to avoid attenuation of the γ -rays of ^{159}Dy at 58 keV due to the presence of $\text{Dy}(\text{NO}_3)_3$ crystals, as well as to ensure a specific geometry equivalent to the one of the electrodeposited radioactive source. The PEEK vial containing the Dy dissolved in 1 M HNO_3 was placed in the custom-made holder. A graphite foil (thickness: 15 μm , purity: 99.8%, Flexible Graphite,

GoodFellow) was inserted between the bottom of the PEEK vial and the detector endcap, as shown in Figure 2a. The γ -measurement of the ^{159}Dy contained in the PEEK vial was performed for 5400 s.

Molecular plating. After the γ -spectroscopy measurements, the Dy solution was transferred from the PEEK vial to a HDPE vial and evaporated to dryness at 70°C under a N_2 gas flow. To ensure a complete transfer of the Dy, the PEEK vial was rinsed with 5 ml 1 M HNO_3 , the washing solution was transferred to the HDPE vial, and the liquid was evaporated to dryness at 70°C under a N_2 flow. This process was repeated 5 times. The dried Dy was then dissolved in 6 M HNO_3 to promote the formation of nitrate species and again evaporated to dryness at 70°C under a N_2 flow. Any organic species that might derive from the separation process described in Section 2.1 was digested by the addition of modified aqua regia, i.e., 1.5 ml 30% (w/w) H_2O_2 + 4.5 ml conc. HCl + 1.5 ml conc. HNO_3 . The solution was evaporated to dryness at 80°C under a N_2 flow, and the residual solid was re-dissolved in a mixture of 2 ml conc. HNO_3 , 6 ml conc. HCl , and 2 ml conc. HF for the destruction and removal of any silica compound that might derive from the ion exchange resins used in the separation of the Dy fraction from the Ta matrix. The solution was then evaporated to dryness (80°C under a N_2 flow), dissolved in 1 M HNO_3 , and re-evaporated to dryness (70°C in N_2 flow). Finally, the electroplating solution was obtained by adding a 50:50 methanol (MeOH) / isopropanol (iPrOH) mixture to the dried solid residue, for a total volume of 10 ml. The liquid was then transferred to the electrodeposition cell made of polytetrafluoroethylene (PTFE). A description of the molecular plating setup can be found in [41]. Before electrodeposition, a cleaning procedure (stepwise rinsing in 1 M HNO_3 , MilliQ water, and iPrOH) was applied to the PTFE cell and to the spiral Pt wire (anode). The cathode, made of a copper block, was cleaned with 0.1 M citric acid, washed with MilliQ water, and rinsed with iPrOH . The graphite deposition foil (thickness: 15 μm , diameter of deposition area: 20 mm, GoodFellow Cambridge Ltd.) was cleaned with iPrOH before molecular plating. For a constant deposition temperature, the setup was implemented with a Peltier cooler at the cathode, maintaining the graphite foil at 15°C during the entire plating procedure. The distance between the two electrodes was approximately 10 mm. The electrodeposition of Dy on the graphite foil was achieved in 8 hours by applying a constant voltage of 550 V.

γ -activity measurements after molecular plating (Position B). The activity of the ^{159}Dy contained in the Dy deposited on the graphite foil was measured by placing the foil in Position B (see Figure 2b). In between the graphite foil and the BEGeTM detector endcap, a PEEK disk (thickness = 1 mm, identical to the bottom of the PEEK vial) was inserted, as shown in Figure 2b. The γ -spectrometry measurement of the ^{159}Dy deposited on the graphite foil was conducted for 2'160'000 seconds.

2.4 ^{154}Dy α -activity measurement

The graphite foil with the electrodeposited Dy was then transferred to an α -chamber for the measurement of the α -activity of ^{154}Dy . α -spectrometry was performed using the Alpha Analyst Integrated Alpha Spectrometer (model A-450-21AM, Canberra) equipped with a silicon semiconductor detector (Passivated Implanted Planar Silicon – PIPS. Detector area: 450 mm^2). Data acquisition and analysis were done

using the Genie™ 2000 Alpha Analysis Software. Energy calibration of the detector was performed with an α -source of ^{148}Gd ($t_{1/2} = 74.6$ y, $I_{\alpha} = 100\%$ at $E_{\alpha} = 3.182$ MeV), and a mixed ^{239}Pu ($t_{1/2} = 2.41$ y, $I_{\alpha} = 70.77\%$ at $E_{\alpha} = 5.157$ MeV), ^{241}Am ($t_{1/2} = 432.8$ y, $I_{\alpha} = 84.8\%$ at $E_{\alpha} = 5.486$ MeV), and ^{244}Cm ($t_{1/2} = 18.1$ y, $I_{\alpha} = 76.90\%$ at $E_{\alpha} = 5.805$ MeV) α -source. A FWHM of 24.73 keV was reached, with a sample detector distance (SDD) of 10.4 mm. The efficiency calibration of the detector was performed with a certified ^{241}Am standard source (PTB, calibration reference n° PTB-6.11-2016-1769, $A = (539 \pm 11)$ Bq @ 01.11.2016), having the same diameter (20 mm) as the Dy deposition area on the graphite foil. Geometrical differences between the ^{241}Am standard source and the Dy electrodeposited sample were further minimized by using holders with the same SDD for both samples. The activity of the ^{154}Dy electrodeposited sample was measured at a defined solid angle. The α -spectrometry measurement of the electrodeposited Dy layer was conducted for 500'000 seconds.

3. Results And Discussion

3.1 Mass spectrometry analysis

The ^{161}Dy content in the Dy master solution, resulting from the average of six analyses (see Table S6), was deduced to be (0.01164 ± 0.00036) nmol $\cdot\text{g}^{-1}$. Final concentration uncertainties were derived by Monte-Carlo propagation, and include: 1) the gravimetrically defined Dy contents of the external standards, 2) the assumption of the accepted natural isotope composition of Dy [42] for the external standards, and 3) the background-corrected and Re-normalized ^{161}Dy signals obtained for the standards. Possible isobaric interferences from Sm and Gd on the ^{154}Dy ion-beam were considered negligible, since the ratios of the signals $^{149}\text{Sm}/^{154}\text{total}$ (where $^{154}\text{total}$ stands for the bulk signal on mass 154 without element distinction), $^{152}(\text{Sm,Gd})/^{154}\text{total}$, $^{155}\text{Gd}/^{154}\text{total}$, and $^{157}\text{Gd}/^{154}\text{total}$ are all below 10^{-3} . Therefore, a correction of the $^{154}\text{total}/^{161}\text{Dy}$ value for isobaric interferences from Sm and Gd – assuming natural isotope compositions of the interfering species – would result to a 0.02% lower value. Therefore, no isobaric correction was undertaken. Likewise, potential interferences from Yb on mass 170 were insignificant since the signal ratios $^{172}\text{Yb}/^{170}\text{total}$ (where $^{170}\text{total}$ stands for the bulk signal on mass 170 without element distinction) were below 10^{-5} . The signal ratio $^{170}\text{Yb}/^{170}\text{total}$ was in the order of 10^{-6} . The relation between the exponential mass discrimination factors for Er and Dy [43] was established from the analyses of the natural Er-Dy solution standards, considering the natural isotopic abundances reported in [42, 44] and the nuclide masses listed in [45]. This (linear) relation, together with the exponential mass discrimination factors for Er, allowed for accurate mass discrimination corrections of the $^{154}\text{Dy}/^{161}\text{Dy}$ ratio in the Dy master solution. As a final result from the average of six analyses (see Table S7), the $^{154}\text{Dy}/^{161}\text{Dy}$ ratio was determined as (0.27732 ± 0.00010) . The combined results from SF-ICP-MS and MC-ICP-MS analyses are shown in Table 2. The concentration of ^{154}Dy in the Dy master solution was assessed as 1.497 nmol $\cdot\text{g}^{-1}$, with an uncertainty of 3.7% (1 SD %). The latter includes the 2% concentration uncertainty of the Dy standard used for SF-ICP-MS.

Table 2
Combined SF- and MC-ICP-MS results for the Dy fraction.

^{154}Dy concentration in (SF+MC)-ICP-MS Dy sample	0.00323 nmol•g⁻¹
1 SD (including 2% uncertainty on Dy standard)	3.7%
Dilution factor	463.60182
1 SD	0.01%
^{154}Dy concentration in the Dy master solution	1.497 nmol•g⁻¹
1 SD	3.7%

3.2 Estimation of the molecular plating efficiency

From the knowledge of the ^{154}Dy concentration in the master solution, it follows that the amount of ^{154}Dy contained in the aliquot (mass: 2.77410 g, see Table S4) used for the preparation of the thin radioactive α -source corresponds to (4.153 ± 0.154) nmols. The activity of the ^{159}Dy tracer before electrodeposition contained in the PEEK vial was quantified to be (779.89 ± 6.77) Bq @13.08.2020, whereas the activity of the ^{159}Dy tracer electrodeposited on the graphite foil amounted to (1.30 ± 0.04) Bq @30.03.2021. Details on the procedure adopted for the estimation of the γ -activities can be found in Section 7 of the Supplementary Information. Considering the decay of ^{159}Dy in the time elapsed between the measurement in Position A and the measurement in Position B (i.e., 229 days), a deposition yield of $(0.50 \pm 0.01)\%$ was calculated (see Table 3). Taking into account the uncertainties deriving from (SF-MC)-ICP-MS and from the ^{159}Dy activity measurements, (0.021 ± 0.001) nmols of ^{154}Dy were electrodeposited on the graphite foil, equivalent to $(1.265 \pm 0.06) \cdot 10^{13}$ atoms of ^{154}Dy .

Table 3
Initial activity of ^{159}Dy measured in Position A and the activity of the electrodeposited ^{159}Dy , measured in Position B.

	Activity ^{159}Dy @13.08.2020 (Bq)	Activity ^{159}Dy @30.03.2021 (Bq)
Position A	778.89 ± 6.77	259.80 ± 2.26
Position B	-	1.30 ± 0.04
Deposition yield	$(0.50 \pm 0.01) \%$	

3.3 ^{154}Dy α -activity measurement

As shown in Figure 3, a slight contamination of ^{148}Gd was visible in the α -spectrum. Thus, a deconvolution method based on a combination of [46] and [47] for a precise peak-shape fitting (see Section 8 of the Supplementary Information for further details), was applied. The contribution of the ^{148}Gd peak tailing to the ^{154}Dy α -peak was estimated as 2.02%. The activities of ^{154}Dy and ^{148}Gd , together with the one of the ^{241}Am standard source used for efficiency calibration, are given in Table 4.

Table 4
Activity (in Bq) for ^{154}Dy electrodeposited on the graphite foil, together with the activity of the ^{148}Gd impurity. The measured cps (in counts•s⁻¹) and the corresponding activity of the ^{241}Am standard source used for efficiency calibration are indicated as well.

	cps (counts•s ⁻¹)	Activity (Bq)
^{241}Am	79.000 ± 0.395	535 ± 11 ^a
^{154}Dy	0.0307 ± 0.0002	0.208 ± 0.005
^{148}Gd	0.0201 ± 0.0001	0.136 ± 0.003
^a Activity of the source at the date of the efficiency calibration (20.07.2021)		

3.4 ^{154}Dy half-life

A reliable half-life value is obtained by applying the following equation:

$$t_{1/2} = \ln 2 \cdot \frac{N}{A} \tag{1}$$

where N is the number of atoms of ^{154}Dy - i.e., $(1.265 \pm 0.06) \cdot 10^{13}$, and A is their activity - i.e., (0.208 ± 0.005) Bq. Here, the half-life value for ^{154}Dy was determined as (1.33 ± 0.07) My, with an estimated total uncertainty of 5.19%. The uncertainty budget is given in Table 5. For the half-life calculation, 1 year was considered equal to 365.242198 days.

Table 5
Relative uncertainties for the ^{154}Dy half-life measurement.

Source of uncertainty	Partial uncertainty	Combined uncertainty
Numbers of atoms		3.7%
ICP-MS Standard	2%	
ICP-MS analysis	3.11%	
Weightings	0.01%	
Molecular plating		2.88%
^{159}Dy activity Position A	0.87%	
^{159}Dy activity Position B	2.75%	
α-activity		2.23%
α -ref. source	2.06%	
Counting statistics ^{241}Am	0.50%	
Counting statistics ^{154}Dy	0.68%	
Total		5.19%

3.5 Re-evaluation of the production cross-sections of ^{154}Dy from proton irradiated Pb, Ta, and W targets

The production cross-sections of ^{154}Dy in proton-irradiated Ta, Pb, and W targets previously reported in [38, 48, 49] were re-evaluated applying Equation 2:

$$\sigma^* = \sigma \cdot \frac{t_{1/2}^*}{t_{1/2}} \quad (2)$$

where σ^* is the re-calculated cross section, σ is the experimental cross section from literature, $t_{1/2}^*$ is the half-life value of ^{154}Dy determined in this work, and $t_{1/2}$ is the currently adopted half-life value for ^{154}Dy . As shown in Figure 4, a significant decrease in the uncertainty of the experimental excitation functions for ^{154}Dy was achieved, with the new results being in agreement with theoretical calculations obtained using INCL++ and ABLA 07 codes [50, 51]. For the sake of completeness, the comparison with the cross-section results derived using $t_{1/2} = (3.0 \pm 1.5) \text{ My}$ is depicted as well.

5. Conclusions

In the framework of the ERAWAST project, the half-life of ^{154}Dy was re-evaluated. By applying state-of-the-art radiochemical separation techniques, in conjunction with high precision activity measurements, the half-life of the pure α -emitting radio-lanthanide ^{154}Dy was determined as (1.33 ± 0.07) My. Compared to the currently adopted value of (3.0 ± 1.5) My, the new measurement drastically reduced the uncertainty by a factor of almost 10. As a first application, the experimental cross sections for the $\text{Ta}(p,x)^{154}\text{Dy}$, $\text{Pb}(p,x)^{154}\text{Dy}$, and $\text{W}(p,x)^{154}\text{Dy}$ nuclear reactions were re-evaluated. The resulting cross-sections are now in good agreement with the theoretical values obtained with INCL++ and ABLA 07 codes. Hence, for future calculations or derivations using the half-life of ^{154}Dy , it is recommended to adopt the value deduced in this work.

Declarations

Data availability

All relevant data are within the paper and its Supporting Information file. The datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.

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Additional Information

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References

1. Banner, J. L. Radiogenic isotopes: systematics and applications to earth surface processes and chemical stratigraphy. *Earth-Sci. Rev*, **65**, 141–94 (2004).
2. Patchett, P. J. Radiogenic isotope geochemistry of rare earth elements in *Geochemistry and mineralogy of rare earth elements* (ed. Lipin, B. R. and McKay, G.A.) 169-200 (De Gruyter, 1989)
3. McLennan, S. M. Rare earth elements in sedimentary rocks: influence of provenance and sedimentary processes in *Geochemistry and mineralogy of rare earth elements* (ed. Lipin, B. R. and McKay, G.A.) 169-200 (De Gruyter, 1989)
4. Wallner, A. *et al.* Fe and ^{244}Pu deposited on Earth constrain the r-process yields of recent nearby supernovae. *Science*. **372**, **60**, 742–745 (2021).
5. Côté, B. *et al.* I and ^{247}Cm in meteorites constrain the last astrophysical source of solar r-process elements. *Science*. **371**, **129**, 945–948 (2021).
6. Wallner, A. *et al.* Recent near-Earth supernovae probed by global deposition of interstellar radioactive ^{60}Fe . *Nature*, **532**, 69–72 (2016).
7. Korschinek, G. *et al.* Supernova-Produced Mn-53 on Earth. *Phys. Rev. Lett*, **125**, 031101 (2020).
8. Burbidge, E. M., Burbidge, G. R., Fowler, W. A. & Hoyle, F. Synthesis of the elements in stars. *Rev. Mod. Phys*, **29**, 547 (1957).
9. Lambert, D. L. The p-nuclei: abundances and origins. *Astron. Astrophys. Rev*, **3**, 201–256 (1992).
10. Arnould, M. & Goriely, S. The p-process of stellar nucleosynthesis: astrophysics and nuclear physics status. *Phys. Rep*, **384**, 1–84 (2003).
11. Reich, C. W. Nuclear data sheets for A= 154. *Nucl. Data Sheets*, **110**, 2257–2532 (2009).
12. Rauscher, T. Branchings in the γ process path revisited. *Phys. Rev. C*, **73**, 015804 (2006).
13. Woosley, S. E. & Howard, W. M. Sm-146 production by the gamma-process. *Astrophys. J*, **354**, L21–4 (1990).
14. Travaglio, C. *et al.* Radiogenic p-isotopes from Type Ia supernova, nuclear physics uncertainties, and galactic chemical evolution compared with values in primitive meteorites. *Astrophys. J*, **795**, 141 (2014).
15. Nicodemi, M. & Coniglio, A. Macroscopic glassy relaxations and microscopic motions in a frustrated lattice gas. *Phys. Rev. E*, **57**, R39 (1998).
16. Arlandini, C. *et al.* Neutron capture in low-mass asymptotic giant branch stars: cross sections and abundance signatures. *Astrophys. J*, **525**, 886 (1999).
17. Bouvier, A. & Boyet, M. Primitive Solar System materials and Earth share a common initial ^{142}Nd abundance. *Nature*, **537**, 399–402 (2016).
18. Harper, C. L. & Jacobsen, S. B. Evidence from coupled ^{147}Sm – ^{143}Nd and ^{146}Sm – ^{142}Nd systematics for very early (4.5-Gyr) differentiation of the Earth's mantle. *Nature*, **360**, 728–32 (1992).

19. Sprung, P., Kleine, T. & Scherer, E. E. Isotopic evidence for chondritic Lu/Hf and Sm/Nd of the Moon. *Earth Planet. Sci. Lett*, **380**, 77–87 (2013).
20. Brandon, A. D. *et al.* Re-evaluating $^{142}\text{Nd}/^{144}\text{Nd}$ in lunar mare basalts with implications for the early evolution and bulk Sm/Nd of the Moon. *Geochim. Cosmochim. Acta*, **73**, 6421–6445 (2009).
21. Côté, B. *et al.* The Impact of Nuclear Physics Uncertainties on Galactic Chemical Evolution Predictions. *J. Phys.: Conf. Ser*, **1668**, 012008 (2020).
22. Matteucci, F. Modelling the chemical evolution of the Milky Way. *Astron. Astrophys. Rev*, **29**, 1–90 (2021).
23. Filges, D. & Goldenbaum, F. Handbook of spallation research. Theory, experiments and applications. Wiley-VCH, Weinheim (Germany).
24. Stankovsky, A., Saito, M., Artisyuk, V., Shmelev, A. & Korovin, Y. Accumulation and transmutation of spallation products in the target of accelerator-driven system. *J. Nucl. Sci. Technol*, **38**, 503–10 (2001).
25. Artisyuk, V., Saito, M., Stankovskii, A., Korovin, Y. & Shmelev, A. Radiological hazard of long-lived spallation products in accelerator-driven system. *Prog. Nucl. Energy*, **40**, 637–45 (2002).
26. Macfarlane, R. D. Dysprosium-154, a long-lived α -emitter. *J. Inorg. Nucl. Chem*, **19**, 9–12 (1961).
27. Mahunka, I. & Fenyes, T. Investigation of the Alpha Spectrum of Dy Isotopes. *Bull. Acad. Sci. USSR, Phys. Ser*, **29**, 1126 (1966).
28. Gono, J. & Hiruta, K. Non-existence of Isomeric Alpha Decay in ^{154}Dy . *Phys. Soc. Japan*, **30**, 1241–1242 (1971).
29. Holden, N. E. *Long-lived heavy mass elements half-lives ($A > 125$)* (Brookhaven National Lab., Upton, NY (USA), 1985 Jan).
30. Buck, B. & Merchant, A. C. Ground state to ground state alpha decays of heavy even-even nuclei. *J. Phys. G: Nucl. Phys*, **17**, 1223 (1991).
31. Schumann, D. & Neuhausen, J. Accelerator waste as a source for exotic radionuclides. *J. Phys. G: Nucl. Phys*, **35**, 014046 (2007).
32. Schumann, D., Stowasser, T., Dressler, R. & Ayrarov, M. Possibilities of preparation of exotic radionuclide samples at PSI for scientific investigations. *Radiochim. Acta*, **101**, 501–8 (2013).
33. Dai, Y. *et al.* The second SINQ target irradiation program, STIP-II. *J. Nucl. Mater*, **343**, 33–44 (2005).
34. Parker, W. & Falk, R. Molecular plating: a method for the electrolytic formation of thin inorganic films. *Nucl. Instr. Methods*, **16**, 355–357 (1962).
35. Ko, Y. G. Preparation and characterization of electrodeposited layers as alpha sources for alpha-particle spectrometry. *J. Radioanal. Nucl. Chem*, **10**, 1–7 (2020).
36. ISO/IEC Guide 98-3: 2008. Uncertainty of measurement - Part 3: Guide to the expression of uncertainty in measurement (GUM:1995). <https://www.iso.org/standard/50461.html> (2008)
37. Bich, W. *et al.* Revision of the 'Guide to the Expression of Uncertainty in Measurement'. *Metrologia*, **49**, 702 (2012).

38. Talip, Z. *et al.* Radiochemical Determination of Long-Lived Radionuclides in Proton-Irradiated Heavy-Metal Targets: Part I-Tantalum. *Anal. Chem*, **89**, 13541–9 (2017).
39. Reich, C. W. Nuclear data sheets for A= 159. *Nucl. Data Sheets*, **113** (1), 157–363 (2012).
40. Chiera, N. M., Talip, Z., Fankhauser, A. & Schumann, D. Separation and recovery of exotic radiolanthanides from irradiated tantalum targets for half-life measurements. *Plos one*, **15**, e0235711 (2020).
41. Maugeri, E. A. *et al.* Preparation of ^7Be targets for nuclear astrophysics research. *J. Instrum*, **12**, 02016 (2017).
42. Chang, T. L., Li, W. J., Zhao, M. T., Wang, J. & Qian, Q. Y. Absolute isotopic composition and atomic weight of dysprosium. *Int. J. Mass Spectrom*, **207**, 13–17 (2001).
43. Russell, W. A., Papanastassiou, D. A. & Tombrello, T. A. *Geochim. Cosmochim. Acta.*, **42**, 1075 (1978).
44. Chang, T. L., Zhao, M. T., Li, W. J., Wang, J. & Qian, Q. Y. Absolute isotopic composition and atomic weight of erbium. *Int. J. Mass Spectrom*, **177**, 131–136 (1998).
45. Audi, G. *et al.* The Ame2012 atomic mass evaluation. *Chin. Phys. C*, **36**, 1287 (2012).
46. Pomme, S., Fitzgerald, R. & Keightley, J. Uncertainty of nuclear counting. *Metrologia*, **52**, S3 (2015).
47. Pöllänen, R., Karhunen, T., Siiskonen, T., Toivonen, H. & Pelikan, A. Deconvolution of alpha spectra from hot particles in *Radioactive Particles in the Environment* (eds. Oughton, D.H., Kashparov, V.) 209-220 (Springer, 2009).
48. Talip, Z. *et al.* Analysis of the ^{148}Gd and ^{154}Dy content in Proton-Irradiated Lead Targets. *Anal. Chem*, **89**, 6861–6869 (2017).
49. Talip, Z. *et al.* Radiochemical Determination of Long-Lived Radionuclides in Proton-Irradiated Heavy Metal Targets: Part II Tungsten. *Anal. Chem*, **93**, 10798–10806 (2021).
50. Unruh, D. M. *et al.* (eds) (a), Lu-Hf and Sm-Nd evolution in lunar mare basalts *J. Geophys. Res.: Solid Earth*, **89**, B459-B477 (1984); (b) Fujimaki, H., Tatsumoto, M., Aoki, K.I. Partition coefficients of Hf, Zr, and REE between phenocrysts and groundmasses. *J. Geophys. Res.: Solid Earth*, **89**, B662-B672 (1984).
51. Salters, V. J. $^{176}\text{Hf}/^{177}\text{Hf}$ determination in small samples by a high-temperature SIMS technique. *Anal. Chem*, **66**, 4186–9 (1994).

Figures

Figure 1

Separation scheme adopted for the retrieval of a pure Dy fraction (in 1 M HNO₃) containing the exotic radio-lanthanide ¹⁵⁴Dy. The so-obtained Dy fraction is suitable for mass spectrometric analysis without further processing.

a) POSITION A

2D section, front view

3D view

b) POSITION B

2D section, front view

3D view

Figure 2

Schematic drawing of the custom-made holder, made of two interchangeable parts (namely, "Holder part 1" and "Holder part 2"), used to quantify the activity of ^{159}Dy via γ -spectrometry. a) 2D front section and 3D view of Position A, used to measure the γ -activity of ^{159}Dy contained in the vial before molecular plating. b) 2D front section and 3D view of Position B, used to quantify the γ -activity of ^{159}Dy deposited on a graphite support after molecular plating. Both positions are geometrically equivalent. The acronym PEEK stands for polyether ether ketone.

Figure 3

α -spectra (SDD = 10.4 mm, counting time = 500'000 seconds) of the electrodeposited ^{154}Dy on a graphite foil. The slight contamination of ^{148}Gd , contained in traces in the Dy separated fraction, is indicated.

Figure 4

Excitation functions for the production of ^{154}Dy in proton irradiated Ta (a), Pb (b), and W (c) targets. Solid lines: theoretical values obtained with INCL++ and ABLA 07 codes; filled dots: experimental values using $t_{1/2} = (1.33 \pm 0.07) \text{ My}$; open squares: experimental values using $t_{1/2} = (3.0 \pm 1.5) \text{ My}$.

Supplementary Files

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