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Measurement of ^{151}Sm in nuclear decommissioning samples by ICP-MS/MS

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Abstract

^{151}Sm [half-life 94.7 (6) years] is a fission and activation product that requires accurate measurement as part of nuclear decommissioning. A procedure is outlined for the measurement of ^{151}Sm in spiked graphite samples. Digestion is performed by automated lithium borate fusion, followed by extraction chromatography separation of ^{151}Sm from interfering lanthanide isotopes, primarily europium. Measurement is carried out using tandem inductively coupled plasma mass spectrometry (ICP-MS/MS), using the integrated collision/reaction cell as a rapid support to radiochemical separation of ^{151}Sm from isobaric ^{151}Eu using oxygen as a reactive gas, achieving detection limits below the out-of-scope limit for ^{151}Sm.

Keywords Nuclear decommissioning · Radiological characterisation · ^{151}Sm · Lanthanide separation · ICP-MS/MS

Introduction

The first-generation nuclear power plants and reprocessing facilities are coming to the end of their operational lives. In the European Union alone, ninety-one power plants are currently being decommissioned, and the majority of the remaining 129 reactors and fuel cycle facilities are likely to begin decommissioning by 2030. The cost of decommissioning and waste management in the EU is estimated to be in excess of €150 billion [1, 2]. The key to safe and cost-effective disposal of the waste is accurate physical, chemical, and radiological characterisation of nuclear decommissioning waste, including accurate measurement of difficult to measure (DTM) radionuclides in a range of matrices including concrete, graphite, steel, plastics, and resins.

One DTM radionuclide is ^{151}Sm, a fission product radionuclide (0.53% yield) also formed via neutron activation of stable Sm. Samarium-151 decays by low-energy β-particle emission (Eβ endpoint 76.6 keV (99.09%),

Fig. 1) to ^{151}Eu with a half-life of 94.7 (6) years [3], and is measurable by liquid scintillation counting (LSC).

Accurate quantification of ^{151}Sm by LSC can be affected by the presence of other β-emitting lanthanides, which can interfere with the measurement and make the deconvolution of the resulting spectrum rather challenging. Therefore, a chemical separation of ^{151}Sm from interfering radionuclides is normally required prior to the LSC analysis to prevent spectral overlap.

The chemical separation of lanthanide elements represents a significant challenge due to similarities in their physical and chemical properties [4]. A summary of previously published procedures are shown in Table 1. The process of complex formation of lanthanide ions with organic ligands has been used to greatly enhance the selectivity of the separation methods [5]. The most effective complex formatting agent is 2-hydroxyisobutyricacid (α-HIBA), which has been used for the separation of rare earth elements by cation exchange chromatography [6, 7], capillary electrophoresis [5, 8] and high-performance liquid chromatography (HPLC) [9, 10].

Yoshida et al. reported a rapid separation method for the analysis of ^{151}Sm and ^{147}Pm in environmental samples based on HPLC in the presence of 0.2 M α-HIBA at pH 4.6. The two radionuclides were quantified by LSC, while ICP-MS was used for chemical yield determination [9]. HPLC has also been applied to the analysis of

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Fig. 1 ^{151}Sm decay scheme [3]

$$^{151}\text{Sm} \ \beta^- \ \text{Decay} \ (94.7 \ \text{y})$$

Decay Scheme

Intensities: I(γ+ce) per
100 parent decays

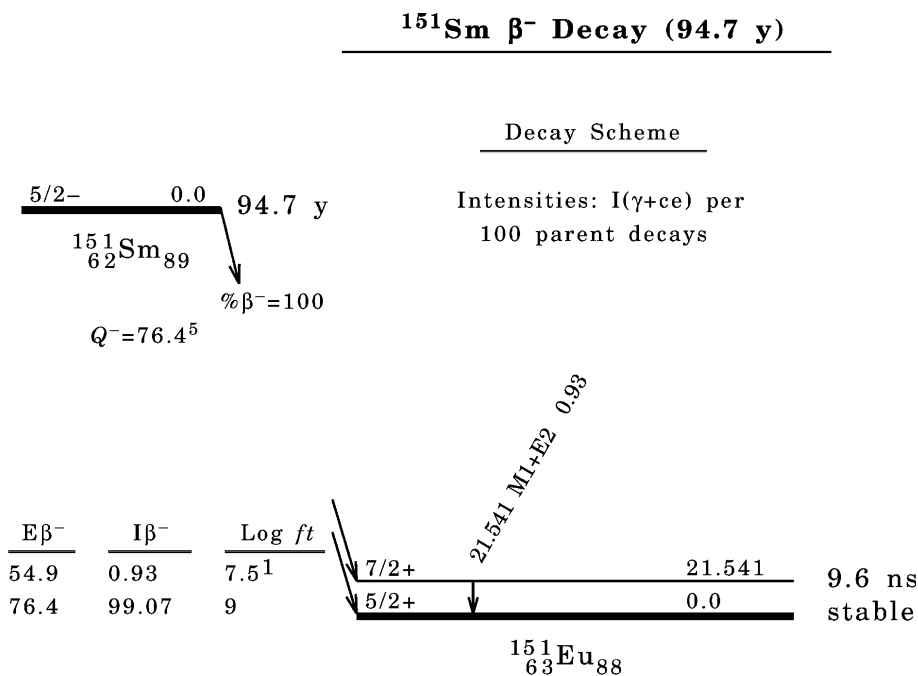| Eβ$^-$	Iβ$^-$	Log ft
54.9	0.93	7.5 1
76.4	99.07	9

Table 1 Recently published studies on radiochemical separations and analysis of lanthanides

References	Separation method	Counting mode	Sm yield	Detection limit
Yoshida, 1995 [9]	HPLC, pH 4.6, 0.2 M, (α-HIBA)	LSC/ICP-MS	99.4%	0.1 Bq kg^{-1}
Martin, 1999 [7]	Extraction chromatography, Ln-resin, 0.15–0.25 M HNO$_3$	LSC/ICP-OES	80%	0.02 Bq g^{-1}
Wolf, 2005 [10]	HPLC, 0.04–0.26 M α-HIBA, flow rate 0.30 mL/min	ICP-MS		
Pitois, 2008 [8]	Capillary electrophoresis, 0.8 mM picolinic acid, 10 mM α-HIBA and 25 mM formic acid at pH 4.7	ICP-SFMS		10 ng mL^{-1} (10 Bq g^{-1})
Vio, 2012 [5]	Isotachophoresis, 14 mM HMBA/10 mM acetic acid, pH 4.5	ICP-MS		

fissiogenic lanthanide elements Nd, Sm, Eu, and Gd in high burn up spent nuclear fuel [10]. The separation utilized a linear gradient elution of 0.04–0.26 M α-HIBA with a 0.30 ml/min flow rate, followed by ICP-MS detection [10].

Capillary electrophoresis (CE) has been successfully applied to the separation of fission products including lanthanide nuclides prior to measurement using quadrupole and sector field ICP-MS [8]. In this study, a CE buffer solution containing 0.8 mM picolinic acid, 10 mM HIBA and 25 mM formic acid at pH 4.7 have been employed for the determination of lanthanides. Separation has also been carried out using isotachophoresis, which, unlike other CE techniques, leads to a self-sharpening effect at the zone boundaries and pre-concentration capability all along the analysis. Lanthanide separation was carried out using 2-hydroxy-2-methylbutyric acid (HMBA) and acetic acid as complexing agents [5].

Extraction chromatography techniques have also been applied to lanthanide element separation. A method was developed for analysis of ^{151}Sm and ^{147}Pm in environmental and nuclear waste samples using LN resin [Lanthanide resin, composed of di(2-ethylhexyl)orthophosphoric acid (HDEHP)], followed by LSC measurement and yield determination by inductively coupled plasma optical emission spectrometry (ICP-OES) [7]. The samples were loaded onto the column in 0.15 M HNO$_3$ before eluting ^{147}Pm with 40 mL of 0.25 M HNO$_3$ while less than 40% of ^{151}Sm was removed from the resin at this stage.

Inductively coupled plasma mass spectrometry (ICP-MS) has been increasingly used over the last few decades for measurement of long-lived radionuclides as a rapid alternative to decay counting techniques [11, 12]. As the technique has advanced, the number of radionuclides measurable and sensitivities have improved, making the technique a valuable tool for nuclear decommissioning [13]. ICP-MS is now in a position where routine measurement of difficult-to-measure radionuclides, including ^{151}Sm, is now a realistic possibility.

The presence of various interferences that overlap with the mass of the radioanalyte remains an issue for accurate ICP-MS measurement. In the case of ^{151}Sm, the major challenge is removal of an isobaric interference from stable ^{151}Eu (47.81% abundance) prior to detection.

Additionally, polyatomic interference from $^{150}Sm^1H$ (7.38% Sm abundance), $^{150}Nd^1H$ (5.6% Nd abundance) must be considered, as well as others including $^{111}Cd^{40}Ar$, $^{135}Ba^{16}O$ and $^{119}Sn^{16}O_2$. Finally, the half-life of ^{151}Sm is relatively short with regards to mass spectrometry measurement, with an activity of 1 Bq/g equivalent to 1.08 pg/g, compared to 8.0×10^7 pg/g for the same activity of long-lived ^{238}U.

There has been a limited number of past measurements of ^{151}Sm by ICP-MS. The majority have focused on coupling high performance liquid chromatography (HPLC) online to ICP-MS [10, 14–16], whilst one study applied CE [8]. An alternative separation approach that has not been investigated is to use ICP-MS equipped with a reaction cell, which can potentially reduce the extent of offline chemical separation required prior to measurement, which is particularly beneficial when wet chemical separation is challenging.

Tandem ICP-MS/MS has been successfully applied to the measurement of multiple radionuclides, including ^{90}Sr, ^{93}Zr, ^{129}I, ^{135}Cs, ^{226}Ra and ^{236}U [17–22]. The instrument layout consists of a collision-reaction cell positioned between two quadrupole mass filters. The inclusion of the first quadrupole means that the ion beam is filtered prior to entering the cell, improving the understanding of the cell chemistry, and preventing the formation of secondary polyatomic interferences that can form in the cell (e.g., formation of oxides when using O_2 reaction gas). Additionally, the first mass filter means the abundance sensitivity has been measured as $< 10^{-10}$ [23], eliminating peak tailing that can otherwise prevent accurate measurement of radionuclides including ^{88}Sr on ^{90}Sr and ^{133}Cs on ^{135}Cs [17, 21].

This study presents the first known procedure for measurement of ^{151}Sm by ICP-MS/MS. The procedure incorporates digestion and chemical separation of ^{151}Sm, focusing on graphite. Graphite is a sample matrix of significant interest for nuclear decommissioning, with approximately 250,000 tons of radioactive graphite worldwide which must be characterized prior to disposal [24]. Although the main nuclides of interest in this matrix are currently volatile radionuclides (i.e., ^3H, ^{14}C and ^{36}Cl), the radioactive inventory will vary depending on the reactor design and the use of graphite as fuel sleeves, making the presence of fission products including ^{151}Sm possible. This study demonstrates that ICP-MS/MS removes the need for complete interference removal prior to measurement, and that high throughput measurement of ^{151}Sm in graphite is achievable.

Experimental

Reagents and materials

Stable single element standards (100–1000 μg g^{-1}) were used for initial testing of digestion and radiochemical separation procedures, and for tuning the ICP-MS/MS. ^{151}Sm was standardised at NPL by the CIEMAT/NIST technique [25], and a range of standards prepared for active runs and as ICP-MS/MS calibration standards. Blank graphite was purchased from Fisher Scientific in pure powder form. Pre-packed 2 mL resins of TRU and Ln- extraction chromatography resins (Triskem International) were tested as part of the radiochemical procedure. A vacuum box with 12 positions was used to increase the separation speed by controlling the flowrate by the use of a vacuum pump (Eichrom Vacuum Box System). Hydrochloric acid, nitric acid and ammonia solution (Fisher Scientific, Analytical Reagent Grade) were used for radiochemical separations, and nitric acid (Fisher Scientific, Trace Analysis Grade) was diluted to 2% (v/v) in deionised water (ELGA, Veolia Water, Marlow, UK, 18 MΩ cm, < 5 ppb Total Organic Carbon) prior to ICP-MS/MS measurement.

Digestion and chemical separation

Samples containing 1–5 g of blank graphite were added to platinum crucibles and left in a furnace (Carbolite RHF 15/8) at 800 °C overnight to incinerate organic material and ash the samples. The resulting ash was then digested by automated lithium borate fusion (SPEX Katanax K2 Prime), and the fused material was then dissolved in 150 mL 5 M HNO_3. The full procedure is described elsewhere [17, 26], and based on a procedure first used in the radioanalytical field by Croudace et al. [27]. Stable elements (including Sm) were measured in aliquots of the digested material using a semi-quantitative ICP-MS/MS screening mode to asses if the elemental concentration increased with the amount of graphite dissolved as an indication that the selected digestion technique was appropriate.

The radiochemical separation was developed using aqueous tracer solutions spiked with a known amount of a mix of stable elements tracers to study the behaviour of Sm(III), Eu(III) and other elements on extraction chromatography resins. For this, the uptake of the tracers by the resins was studied in nitric and hydrochloric media as a function of the solution acidity analyzing aliquots of the separated fractions by ICP-MS/MS.

In order to investigate the effect of sample matrix, samples containing 3 g of blank graphite were digested and analyzed using the procedure developed. Samarium recovery was traced by spiking graphite with a stable Sm standard, with aliquots following each digestion and separation stage measured by ICP-MS/MS.

ICP-MS/MS measurement

An Agilent 8800 was used for all measurements in this study. The instrument was fitted with the x-lens setup, with a quartz double-pass spray chamber, MicroMist nebuliser, quartz torch (2.5 mm internal diameter), nickel sample and skimmer cones. The instrument is equipped with four cell gas lines—H_2, He, and corrosive and non-corrosive reaction gas lines. In this study, the corrosive line (10% NH_3 balanced in 90% He) and the non-corrosive line (O_2) were investigated (all gases were grade 6.0 (99.9999% purity), BOC).

Tuning was initially carried out using a mixed 1 ng g^{-1} standard solution in Single Quad mode (i.e., only the second quadrupole mass filter operating). For assessing recovery from chemical separation stages, no further tuning was carried out, with the Sm and Eu signals measured at $m/z = 147$ (14.99% abundance) and 151 (47.81% abundance), respectively. For investigating cell gases, a mixed 10 ng g^{-1} stable Sm and Eu standard was used. A product ion scan was used to assess elemental behaviour in NH_3: the instrument was operated in MS/MS mode, with Q_1 set to $m/z = 147$ and 151 for Sm and Eu, respectively, and then Q_2 scanned the entire all masses up to a maximum of $m/z = 260$, returning the counts per second for each mass. For O_2 reaction gas, Q_1 and Q_2 were set to $m/z = 147$ and 163 for Sm and SmO, respectively, and 151 and 167 for Eu and EuO, respectively. The formation of polyatomic $^{150}Sm^{16}O^1H$ was also monitored in O_2 mode.

Once the optimal instrument conditions were established, active ^{151}Sm calibration standards were run to determine the instrument limit of detection. Active standards were also spiked with increasing concentrations of stable Eu and Sm to assess the cell-based decontamination factors achievable. Lastly, spiked graphite samples were run following digestion and chemical separation to assess the procedures capability for decommissioning samples.

Results and discussion

Digestion and chemical separation

Ashing and lithium borate fusion effectively digested graphite samples, with increasing concentrations measured for multiple elements (including Sm) measured as the graphite mass increased [26]. Volatile radionuclides e.g. ^3H and ^{14}C were lost during the digestion stage, and therefore a suitable extract system was required for the furnace and borate fusion instrument. The trapping of volatile radionuclides could be achieved by using a Pyrolyser prior to digestion,

and the ashed samples prepared for borate fusion as described.

The final separation procedure for graphite samples is shown in Fig. 2. Extraction chromatography separation was preceded by an iron hydroxide co-precipitation stage to remove the sample matrix. An Fe carrier (5 mg) was added to the digested flux, and adjusted to pH 12–14 by addition of ammonium hydroxide. The precipitate formed was centrifuged at 4000 rpm for 2 min, and then dissolved in 5 mL 1 M HNO_3 to enable direct loading onto TRU resin, pre-conditioned with 20 mL 1 M HNO_3. The eluted load contained Fe and other matrix elements, whilst lanthanides were retained. Following Fe and matrix removal from the TRU resin column, Sm and Eu were then eluted with 5 mL 0.05 M HNO_3. Some tests showed a yellow colouring to the eluted Sm fraction as a result of insufficient Fe and matrix removal, which could potentially affect the performance of Ln resin. To ensure sufficient Fe and matrix removal, a wash volume of 10 mL 1 M HNO_3 was applied to the column prior to Sm and Eu elution. The Sm and Eu fraction was then directly loaded onto an Ln resin column conditioned with 20 mL 0.05 M HNO_3, followed by a 3 mL 0.25 M HCl wash to elute other lanthanides. Samarium was then eluted in 3 mL 0.75 M HCl, whilst the
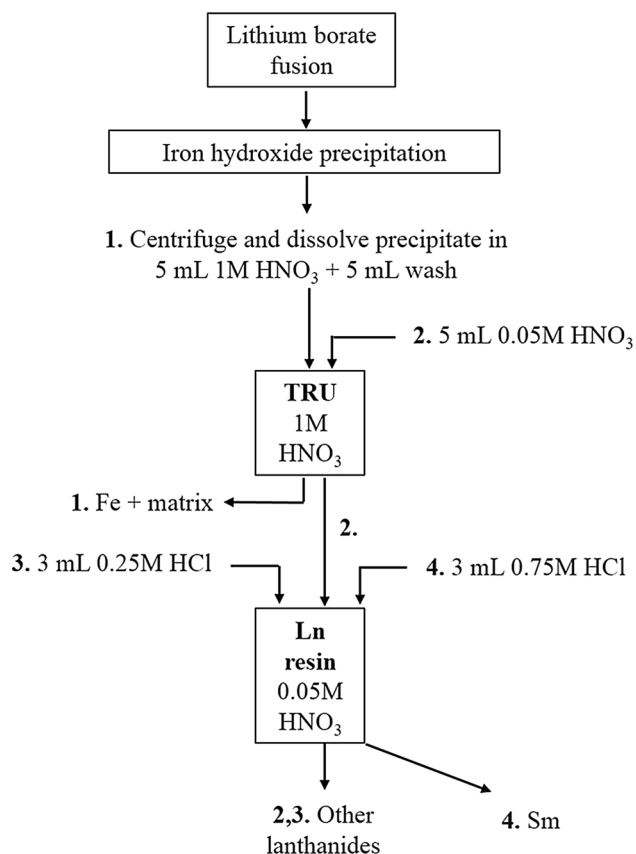
Fig. 2 Separation scheme for ^{151}Sm prior to ICP-MS/MS measurement

majority of Eu was retained on the resin until higher elution volumes were applied.

Direct loading of the eluted Sm fraction from TRU onto Ln resin was critical in achieving the optimal Sm recovery (60–70%). In earlier tests using different elution conditions, if the Sm fraction was evaporated to dryness, the recoveries were as low as 20%, which was improved by evaporating to insipient dryness. The final procedure in Fig. 2 does not require any evaporation stages. Approximately 30% of Eu was recovered in the Sm elution fraction, demonstrating the challenge of Sm/Eu separation by extraction chromatography. The procedure from the resin manufacturer co-elutes Sm and Eu in 0.75 M HCl [28], with the separation in this study achieved by limiting the elution volume to 3 mL. The final sample is therefore contaminated with Eu, and reaction cell-based separation is required.

ICP-MS/MS

Optimisation using stable element standards

By operating in MS/MS mode, no ions other than those at $m/z = 151$ were able to enter the cell, enabling better understanding and control over the cell chemistry. The instrument sensitivity decreased by approximately 45% when operating in MS/MS mode compared to Single Quad mode. When operating with NH_3 reaction gas, multiple cell products were formed for both Sm and Eu, however, the majority of the signal remained on mass for both ions (~ 80 and $\sim 95\%$ for Sm and Eu, respectively), in agreement with results from the instrument manufacturer [29]. The most significant cell product for Sm [$Sm(NH_2)^+$] only represented 5% of the total Sm signal, and therefore NH_3 was not investigated further. By comparison, when operating with O_2 reaction gas, the majority of Sm formed SmO^+, whilst the Eu signal remained on mass. This is because the reaction to form SmO^+ is slightly exothermic (ΔH_r enthalpy of reaction $= -0.70$ eV), whereas the reaction with Eu^+ is not energetically favourable and does not proceed ($\Delta H_r = 1.10$ eV [30]).

Custom tuning of the instrument significantly improved SmO^+ separation from EuO^+, with the optimal operating conditions shown in Table 2. Other than the cell gas flow rate, the parameters that had the most significant impact were the octopole bias, and to a lesser extent the energy discrimination voltage. These parameters affect the ion energy, reactivity and ion rejection in the cell. A negative energy discrimination voltage speeds up the ions and increases their energy in the cell [30]. The reduction of residence time in the cell means that reactions have less time to proceed, suggesting that SmO^+ is more readily formed than EuO^+, which is supported by the thermodynamic data. As the energy discrimination is reduced further, the SmO^+ formation decreases, suggesting that the residence time in the cell is too short. A negative octopole bias accelerates reactant ions so that they gain kinetic energy before reacting with O_2. At more negative octopole bias voltages, the EuO^+ formation rate increased, as the ion energy was high enough for endothermic reactions to proceed. At the optimal value of 2–4 V, Eu^+ did not have sufficient energy to form EuO^+ (Fig. 3).

Under the optimal instrument conditions, 89–91% of the total Sm signal was measured as an oxide, compared to 3–4% for Eu. At O_2 flow rates > 0.25 mL min^{-1}, the EuO^+ signal increased, whilst also reducing the SmO^+ sensitivity (Fig. 4). Q_1 was set to $m/z = 150$ and Q_2 set to $m/z = 150$, 151, 166 and 167 to determine the Sm^+:SmH^+:SmO^+:$SmOH^+$ ratio. The majority of Sm formed SmO^+, with negligible formation of $SmOH^+$. The measured Sm^+:SmH^+:SmO^+:$SmOH^+$ ratio was 0.11:5.14×10^{-6}:0.89:5.36×10^{-4}. For graphite samples, there was an increased background at $m/z = 151$ when operating in no gas mode, which was most likely the result of $^{135}Ba^{16}O^+$ formed during sample introduction as a result of Ba present in the sample. The low (pg g^{-1}) levels of Sn, Nd and Cd meant that the increased background was unlikely to be due to the presence of $^{119}Sn^{16}O_2^+$, $^{150}Nd^1H^+$, or $^{111}Cd^{40}Ar^+$, respectively. However, when operating in O_2 mode, there was no increase in background at $m/z = 167$ for unspiked graphite samples, suggesting no formation of $^{135}Ba^{16}O_2^+$, or indeed other polyatomics. Of the polyatomics described, Cd should be closely monitored for graphite samples, given its use as a neutron absorber.

Active ^{151}Sm and spiked graphite measurement

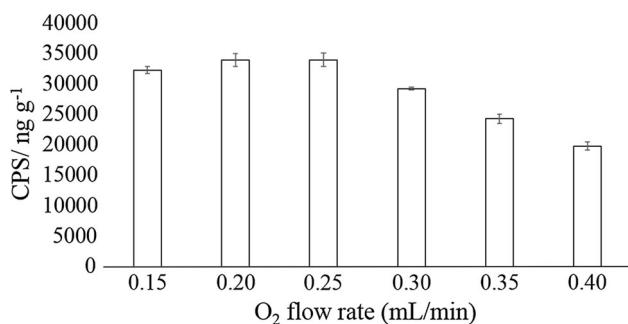Calibration standards for ^{151}Sm were run in the optimal instrument setup at activity concentrations from 0.25 to 250 Bq g^{-1}, equivalent to 0.27–270 pg g^{-1} (Fig. 5). The

Table 2 Instrument setup for ^{151}Sm measurement

Instrument parameter	Operating window
Scan mode	MS/MS ($Q_1 = 151$, $Q_2 = 167$)
Forward power (W)	1550
Carrier gas flow rate (L min^{-1})	1.10–1.15
Extraction lens 1 (V)	0
Extraction lens 2 (V)	− 175
Omega bias (V)	− 90
Omega lens (V)	10
Octopole bias (V)	2–4
Cell line 4 flow rate (mL min^{-1})	0.2–0.25
Octopole RF (V)	200
Energy discrimination (V)	− 4 to − 7
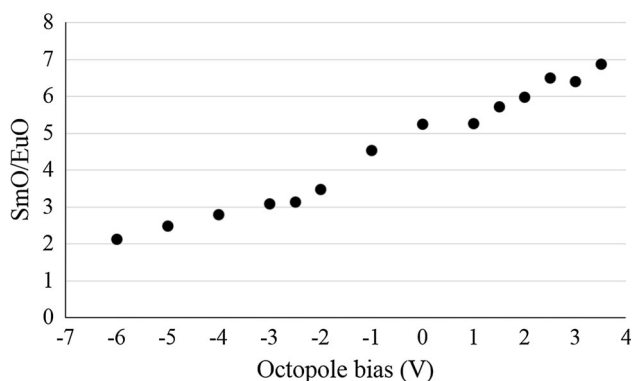
Fig. 3 Impact of octopole bias voltage on SmO:EuO ratio for a mixed 10 ng g^{-1} Sm + Eu standard at a O$_2$ flow rate of 0.25 mL/min

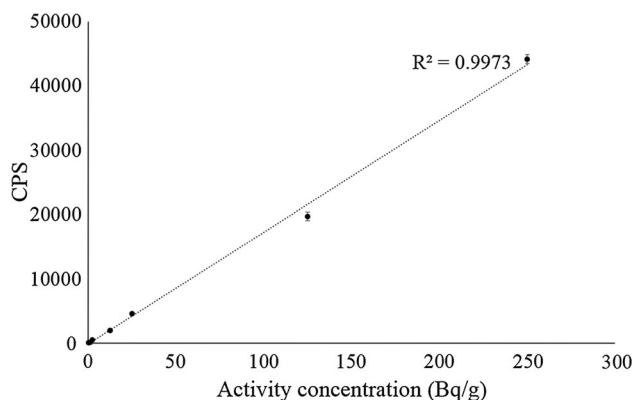Fig. 5 Instrument sensitivity for ^{151}Sm under optimal instrument conditions

Fig. 4 SmO$^+$ sensitivity with increasing O$_2$ gas flow rate (0.15 ng g^{-1} Sm standard measured as ^{147}Sm^{16}O$^+$)

standards were free from Eu contamination, which was monitored at $m/z = 153$ (52.19% abundance). The instrument detection limit was calculated as 1.0 Bq g^{-1} (1.08 pg g^{-1}).

Multiple 100 Bq g^{-1} (108.09 pg g^{-1}) ^{151}Sm standards were prepared and then spiked with stable Sm at concentrations from 1 to 1000 ng g^{-1}, and the signal monitored at $m/z = 167$. Even at a concentration of 1000 ng g^{-1}, there was no increase in signal at $m/z = 167$ as a result of ^{150}Sm^{16}O^{1}H$^+$ formation (Fig. 6). This is in agreement with stable element results, with the majority of Sm forming SmO$^+$, and the remainder staying on mass, with negligible hydride and hydroxide formation. Given that the stable Sm concentration in graphite following chemical separation ranged from the levels of 13.4–22.6 ng g^{-1}, polyatomic ^{150}Sm^{16}O^{1}H$^+$ is not a significant concern for the samples studied. Additionally, given that the abundance sensitivity of ICP-MS/MS in MS/MS mode has been measured as $< 10^{-10}$ [23], there is no tailing from ^{150}Sm$^+$ present as ^{150}Sm^{16}O$^+$.

Repeat standards of 100 Bq g^{-1} (108.09 pg g^{-1}) ^{151}Sm were also spiked with 0.1–100 ng g^{-1} Eu and the signal monitored at $m/z = 167$ (Fig. 7). The majority of the Eu$^+$ signal remained on mass, with only 3.4–3.7% of the total signal measured as EuO$^+$, in good agreement with the

results achieved during tuning. However, given the low activities of Sm being investigated, even a small concentration of Eu contamination can significantly contribute to the final Sm signal.

An Eu decontamination factor of one order of magnitude was achieved using ICP-MS/MS in O$_2$ mode. The lowest Eu concentrations measured in final samples was around 1 ng g^{-1}, enabling ^{151}Sm quantification at the 100 Bq g^{-1} (108.09 pg g^{-1}) concentration level. This is one order of magnitude lower than the ^{151}Sm exemption limit of 1000 Bq g^{-1} [31], demonstrating that ICP-MS/MS is applicable to quantification of ^{151}Sm in decommissioning matrices. Complete separation of Sm from Eu prior to measurement is not required, which is significant given the difficulty in separating lanthanide elements. Additionally, the procedural time is reduced, as reaction-cell separation is instant, and eliminates the need for additional and relatively time-consuming offline separation techniques, as well as reducing the number and volume of resins and reagents used. However, the relatively short half-life of ^{151}Sm with regards to ICP-MS measurement means that even low-level Eu contamination has a significant impact on the detection limits achievable.

Alternative approaches to Eu interference removal include mathematical correction by monitoring contamination at $m/z = 153$ (52.19% abundance). In order to overcome instrumental mass bias, a reference solution with known ^{151}Eu/^{153}Eu ratio would need to be run prior to the samples in order to calculate the mass bias correction factor. This approach will increase the measurement uncertainty compared to the approach presented in this study. Alternative reaction gases may offer improved Sm/Eu separation compared to O$_2$. For example, gases such as CH$_3$F and N$_2$O have been successfully applied to ICP-MS/MS separations of multiple elements [20, 32], but were not investigated as part of this study. An improved Eu separation may also be achieved by combining reaction cell

Fig. 6 Signal measured for a 100 Bq g^{-1} ^{151}Sm in the presence of stable Sm under optimal instrument conditions (Table 2)
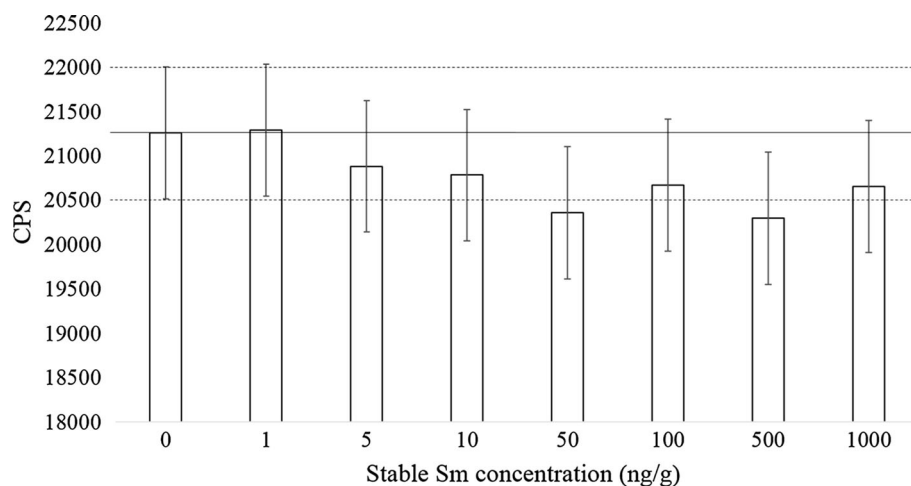
Fig. 7 Signal measured at $m/z = 167$ for a 100 Bq g^{-1} ^{151}Sm in the presence of stable Eu
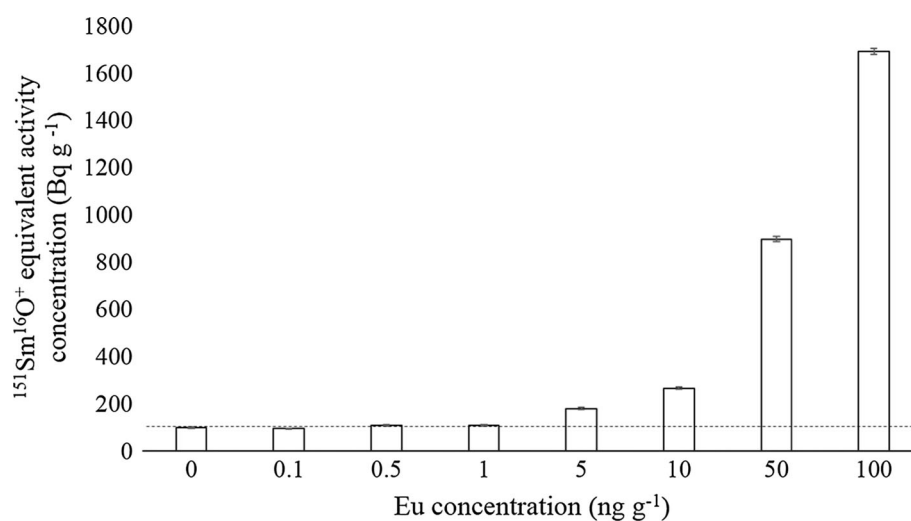
separation with online separation, such as HPLC or CE, which have been proven to be efficient techniques for lanthanide separation.

Conclusions

The decommissioning of nuclear power plants represents a significant challenge with regards to the range of matrices and difficult-to-measure radionuclides that must be accurately measured. One such radionuclide is ^{151}Sm, with interferences from other lanthanide elements (primarily Eu) significantly impacting accurate measurement. This study demonstrates the effectiveness of lithium borate digestion of graphite samples, and the difficulties related to the separation of Sm and Eu using offline extraction chromatography techniques. For the first time, ICP-MS/MS has been used for final quantification of ^{151}Sm. By using O$_2$ in the reaction cell, ^{151}Sm$^+$ is shifted to ^{151}Sm^{16}O$^+$, whilst interfering ^{151}Eu remains on mass. This approach offers a

one order of magnitude cleanup, meaning that complete separation is not required prior to sample introduction. This improves the sample throughput through a reduction in the total procedural time. A detection limit of 1 Bq g^{-1} was achieved for interference-free ^{151}Sm samples, increasing to 100 Bq g^{-1} in samples contaminated with 1 ng g^{-1} Eu. Further improvements in Eu decontamination may be achieved by combining ICP-MS/MS with online separation via HPLC or CE.

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