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Standardisation of the ^{129}I, ^{151}Sm and ^{166m}Ho activity concentration using the CIEMAT/NIST efficiency tracing method

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HIGHLIGHTS

- Standardisation of ^{129}I, ^{151}Sm and ^{166m}Ho by Liquid Scintillation Counting.
- Use of the CIEMAT/NIST efficiency tracing method and the CN2005 and MICELLE2 codes.
- Complete uncertainty budgets for each radionuclide standardisation.
- Work performed for the EMRP project "Metrology for Radioactive Waste Management".

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ABSTRACT

The ^{129}I, ^{151}Sm and ^{166m}Ho standardisations using the CIEMAT/NIST efficiency tracing method, that have been carried out in the frame of the European Metrology Research Program project "Metrology for Radioactive Waste Management" are described. The radionuclide beta counting efficiencies were calculated using two computer codes CN2005 and MICELLE2. The sensitivity analysis of the code input parameters (ionization quenching factor, beta shape factor) on the calculated efficiencies was performed, and the results are discussed. The combined relative standard uncertainty of the standardisations of the ^{129}I, ^{151}Sm and ^{166m}Ho solutions were 0.4%, 0.5% and 0.4%, respectively. The stated precision obtained using the CIEMAT/NIST method is better than that previously reported in the literature obtained by the TDCR (^{129}I), the 4πγ-NaI (^{166m}Ho) counting or the CIEMAT/NIST method (^{151}Sm).

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1. Introduction

Accurate nuclear decay data are important input parameters in the development of technical solutions for a safe long-term disposal of nuclear waste. One of the aims of the European Metrology Research Program (EMRP) project "Metrology for Radioactive Waste Management (MetroRWM)", executed in 2011–2014, was to determine the half-lives of three long-lived radionuclides, ^{129}I, ^{151}Sm and ^{166m}Ho, present in nuclear waste.

^{129}I is one of the ^{235}U fission products with high fission production yield of 0.7%. Due to its long half-life ($T_{1/2}$=16.1 (7)·10^6 a, DDEP), chemical reactivity and mobility, it is considered as one of the most important dose contributors in the nuclear waste disposal storages in the future. The ^{129}I half-life has been determined four times between 1951 and 1973, with values ranging from 15.6 (6)·10^6 a (Katcoff et al., 1951) to 19.7 (14)·10^6 a (Kuhry and Bontems, 1973) that were not consistent within their uncertainty limits.

^{151}Sm is a relatively long-lived nuclide ($T_{1/2}$=90 (6) a, DDEP) produced during the irradiation of uranium fuel rods in nuclear reactors. Its half-life was determined several times between 1950 and 1968. The last time it was measured in 2009 and the value of the half-life was determined to be 96.6 (2.4) a (He et al., 2009).

^{166m}Ho ($T_{1/2}$=1200 (180) a, DDEP) is produced in nuclear fuel as the result of the neutron activation of erbium admixtures added to the fuel to improve the burn out of the uranium. Its half-life was determined once in 1965 (Faler, 1965). Recently, the ^{166m}Ho half-life has been measured again, resulting in a value with lower uncertainty ($T_{1/2}$=1132.6 (3.9) a, Nedjadi, Bailat, Caffari et al., 2012).

The currently available data in the literature on the ^{129}I, ^{151}Sm and ^{166m}Ho half-lives are either scarce or inconsistent and new measurements are needed to obtain accurate values with reliable uncertainties. For such long-lived nuclides the half-life value is best obtained by independently measuring the activity concentration and the mass concentration of the given radionuclide in the same solution. In the course of the MetroRWM project the radionuclide activity concentrations were determined using a number of standardisation techniques at the highest level of

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accuracy and precision in collaboration with national metrology institutes. The ^{129}I and ^{151}Sm standardisations were registered as the Bureau International des Poids et Mesures (BIPM) supplementary comparisons EURAMET.RI(II)-S6.I-129 and EURAMET.RI (II)-S7.Sm-151, respectively, and the ^{166m}Ho standardisation was registered as the BIPM Key Comparison EURAMET.RI(II)-K2.Ho-166m (Ratel et al., 2015). At the Joint Research Centre Institute for Reference Materials and Measurements (JRC-IRMM), the activity concentrations were measured both by the Triple-to-Double Coincidence Ratio (TDCR) Liquid Scintillation Counting (LSC) method and the CIEMAT/NIST efficiency tracing LSC method (Grau Malonda, Garcia-Toraño, 1982), the latter being the subject of this article.

There are a few publications on the standardisation of the above mentioned radionuclides using the CIEMAT/NIST method. For example, there exists only one publication on the standardisation of the ^{129}I solution, and this describes the determination of the radionuclide activity concentrations using the TDCR method and 4π(LS)-γ coincidence counting (Cassette et al., 1994). The relative combined standard uncertainties of the standardisation of the 50 kBq g^{-1} solution using the TDCR method and the 4π(LS)-γ counting were 0.5% and 0.3%, respectively.

There has been published so far one paper on the standardisation of the ^{151}Sm solution and this concerned the application of the CIEMAT/NIST efficiency tracing method, using ^{14}C as tracer (He et al., 2009). The reported relative combined standard uncertainty of the standardisation solution was 2.5%.

^{166m}Ho has been standardized using a variety of standardisation techniques. In the BIPM publication (Michotte et al., 2009) the results of the international comparison of the ^{166m}Ho activity determination using four primary standardization techniques 4πγ (NaI), 4πβ(proportional counter)-γ(NaI) coincidence, 4πβ(pressurized proportional counter)-γ(NaI) coincidence and 4πβ(LS)-γ (NaI) coincidence counting were presented. The relative combined standard uncertainty of the determination of the ^{166m}Ho activity concentration using the most precise 4πβ(LS)-γ(NaI) coincidence counting technique was 0.24% (Silva et al., 2012). In the papers (Nedjadi, Bailat, Caffari et al., 2012; Nedjadi, Bailat, Bochud, 2012; Nedjadi, Spring et al., 2008) the uncertainty of the standardisation using the 4πγ(NaI) counting technique was 0.55% and 0.2–0.3% in case of other used techniques such as the 4πβ(plastic scintillator)-γ(NaI) coincidence, 4πβ(proportional counter)-γ(NaI) coincidence counting. The most recent publication is that of (Kossert, Cassette et al., 2014); the standardization of a ^{166m}Ho solutions was performed using liquid scintillation counting by both the TDCR and the the CIEMAT/NIST efficiency tracing methods to achieve an overall relative standard uncertainty of about 0.3%.

The aim of the work presented here was to standardize the ^{129}I, ^{151}Sm and ^{166m}Ho solutions using the CIEMAT/NIST efficiency tracing method and with improved uncertainties of the radionuclide activity concentration determination compared with the uncertainties reported in the literature.

2. Method

2.1. Source preparation

The radioactive counting sources were prepared from the radionuclide solutions provided by the pilot laboratories according to the EMRP Joint Research Protocol. The ^{129}I solution of an approximate activity concentration of 32 kBq · g^{-1} was provided by CIEMAT, the ^{151}Sm solution of about 80 kBq · g^{-1} was provided by the Laboratoire National de Métrologie et d'Essais – Laboratoire National Henri Becquerel (LNE-LNHB) and the ^{166m}Ho solution of an activity concentration of about 119 kBq · g^{-1} by the Physikalisch-Technische Bundesanstalt (PTB).

After receiving the ampoules with the radioactive solutions at JRC-IRMM, the activities of the gamma-ray emitting impurities in the solutions were measured using high-resolution gamma-ray spectrometry employing HPGe detectors. There were no impurities detected in the ^{129}I and ^{166m}Ho solutions. In the ^{151}Sm solution the ^{154}Eu and ^{155}Eu admixtures were measured and their relative activities were 0.03% and 0.01%, respectively, relative to the activity of ^{151}Sm. The amount of ^{151}Sm impurities was considered negligible and only a component was included in the uncertainty budget of the radionuclide standardisation to take it into account.

The radioactive sources were prepared using the pycnometer method (Sibbens and Altzitzoglou, 2007; Campion, 1975) by gravimetrically dispensing the radionuclide solution to the 20-mL low-potassium glass LSC vials containing the scintillation cocktail. One part of the sources was prepared in 15 mL of the Ultima Gold® (PerkinElmer, Boston, MA, USA) liquid scintillation (LS) cocktail, mixed with 1 mL of deionized water (hereafter Ultima Gold, for simplicity). The rest of the sources were prepared in 15 mL of InstaGel™ Plus (PerkinElmer) LS cocktail (hereafter InstaGel Plus). The aliquots dispensed into each LS source ranged from 10 to 50 mg. The traceability of the mass measurements was achieved via the calibration of the balance with certified mass standards.

The so prepared sources were shaken and left to dark adapt in the LSC instruments at a temperature between 12 and 14 °C. The sources were visually homogeneous and colourless. In total, the following radioactive sources were prepared:

- for ^{129}I, 8 sources in Ultima Gold and 8 in InstaGel Plus with activities ranging from 0.3 to 1.7 kBq per sample,
- for ^{151}Sm, 8 sources in Ultima Gold and 8 in InstaGel Plus with activities ranging from 0.8 to 3.2 kBq per sample, and
- for ^{166m}Ho 6 sources in Ultima Gold and 3 in InstaGel Plus with activities ranging from 1.3 to 3.8 kBq per sample.

2.2. Source measurement

The sources were counted using a Wallac 1220 Quantulus (PerkinElmer) and a Packard TriCarb 3100 TR/AB (PerkinElmer) LS counter. The quench parameter of each source was determined with the external standard method, using the external ^{152}Eu and ^{137}Cs radiation sources of the two LSC, respectively. Each of the ^{129}I and ^{166m}Ho sources was measured for 30 min and each ^{151}Sm source for 20 min. All sources were measured repeatedly during a period of 4–6 weeks. Typical beta-particle energy spectra of the three radionuclides as measured with the Quantulus LSC are shown in Fig. 1. Blank sources were measured for the same time as the radioactive sources. Each source count rate was corrected for the background by subtracting the count rate of the blank, for dead time and for decay.

2.3. Radionuclide adsorption

The adsorption of the given radionuclide on the original glass ampoule walls was determined by rinsing the original emptied ampoule twice with 1 mL of 2 M HCl and once with 1 mL of deionized water. Then the ampoule was placed in an LSC vial, filled with 15 mL of Ultima Gold LS cocktail and counted. The remaining activity measured was less than 0.04% of the activity of the solution contained in the ampoule in the case of ^{129}I and less than 0.01% for ^{151}Sm and ^{166m}Ho. In all cases, the amount of adsorption was considered negligible and it was not taken into account in the activity calculations, but only included in the uncertainty budget.

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