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Distribution of radionuclides in an iron calibration standard for a free release measurement facility

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HIGHLIGHTS

- A 246-kg reference standard for a free release measurement facility is described.
- The distribution of ^{110m}Ag and ^{60}Co inside each of 12 iron tubes was measured.
- Silver-110m was homogeneously distributed along the main axis of the tubes.
- There was 3% higher concentration of ^{110m}Ag on the inside of each tube.
- Cobalt-60 was homogeneously distributed both in radial and axial directions.

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ABSTRACT

A Europallet-sized calibration standard composed of 12 grey cast iron tubes contaminated with ^{60}Co and ^{110m}Ag with a mass of 246 kg was developed. As the tubes were produced through centrifugal casting it was of particular concern to study the distribution of radionuclides in the radial direction of the tubes. This was done by removing 72 small samples (swarf) of ~0.3 g each on both the inside and outside of the tubes. All of the samples were measured in the underground laboratory HADES.

1. Introduction

The decommissioning of nuclear installations is a very complex and costly undertaking. However, most materials from old nuclear power plants (NPP) have radioactivity content below exemption/clearance levels. With the global trend towards sustainable use of resources, recycling is of utmost importance but must be carried out using robust techniques and applying a high safety margin. To be able to clear all recyclable material from a NPP, Free Release Measurement Facilities (FRMF) based on spectrometric techniques are necessary. Within the EMRP (European Metrology Research Programme) project named MetroRWM (Metrology for Radioactive Waste Management) such a facility was developed and tested (MetroRWM, 2015). In the new EMRP project named Metrology for Decommissioning Nuclear Facilities (MetroDecom, 2015), a FRMF will be further developed and tested at the site of a decommissioning object at the JRC (Joint Research Centre) in Ispra, Italy. In order to perform robust quantitative characterisation of waste drums or Europallet-sized containers with waste, it is necessary to use suitable calibration standards. One standard that has been developed is composed of 12 grey cast iron tubes (Fig. 1). It has a total mass of 246 kg and is contaminated with ^{60}Co (71 kBq on 30 Sept, 2013) and ^{110m}Ag (749 kBq on 30 Sept, 2013). The flexibility of the calibration standard is shown in Fig. 1b, where eight of the tubes have been placed inside a waste barrel. Its production and certification was described recently by Tzika et al. (2015). For the calibration standard to perform well it is essential that the radionuclides are homogeneously distributed in the tubes. In this grey cast iron, one would expect ^{60}Co to distribute more evenly than ^{110m}Ag for solubility reasons. As the tubes were produced through centrifugal casting, it was of particular concern to study the distribution in the radial direction of the tubes, which

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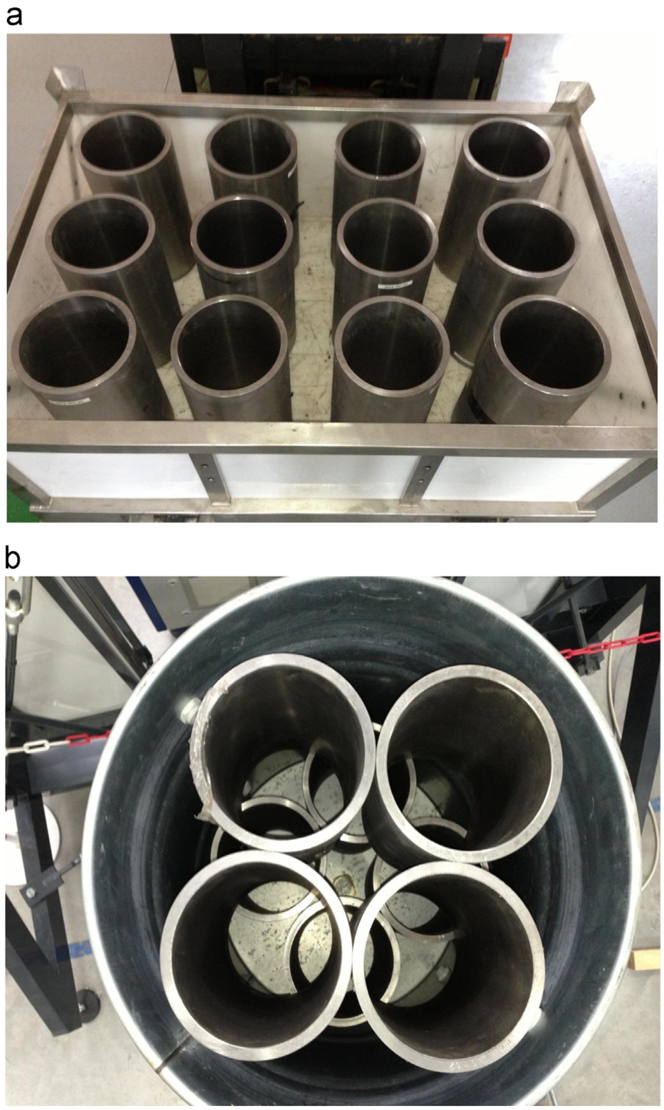

Fig. 1. Photo of the cast iron tube reference material for a free release measurement facility when (a) placed inside a Europallet-sized container and (b) when placed inside a waste drum.

was not done in the preceding study (Tzika et al., 2015). It was also considered important to study the longitudinal distribution using more sampling points compared to the previous study. This new in-depth study of the distribution is important for, and triggered by, future work in the MetroDecom project. It is essential to decide on optimal shape and size of the next generation of this type of calibration standard including the possibility of using e.g. ^{108m}Ag, which is more long-lived (438a) than ^{110m}Ag (0.68a) but also suitable when it comes to the γ-rays that are emitted[1].

2. Materials and methods

2.1. Production of iron tubes and subsamples

It was not possible to produce this material using conventional steel- (or cast iron-) making technologies like using an electric arc furnace casting as ingots. Conventional melting furnaces require several tens of tonnes of material. The subsequent formation of the material includes many steps like casting and rolling which would cause contamination in several places in the plant. Only foundry technologies are suitable for producing these tubes as they involve a short route from raw material to final product. Consequently, centrifugal casting is most suitable for producing these materials as it generates very little waste. The basic principle is that the molten metal is poured into a rotating metallic mould (Fig. 2). For these tubes the mould had a diameter of 200 mm and length of 1131 mm and was made from a creep-resistant low-alloyed steel.

The raw material was grey cast iron with lamellar graphite grade DIN 1691 GG25. A piece of iron containing ^{60}Co coming from a nuclear installation was melted gradually into a non-active iron melt. The ^{110m}Ag was added to the melt in the form of activated silver wires. Cobalt-60 is highly suitable for inclusion in this material as cobalt forms a solid solution in iron. Silver, however, is relatively insoluble both in the liquid and solid form of iron. In order to reach a homogeneous distribution of silver in the iron tubes, the melt was carefully homogenised. Ten small discs produced directly from the melt showed no sign of inhomogeneous activity distribution (Tzika et al., 2015).

For this study, 72 subsamples (so-called swarf) were produced by drilling small indents at specific locations in the tubes. These locations were chosen such that on each of the twelve tubes one sample came from the inside of the top of the tube and one from the outside at the top of the tube (Fig. 3). In a similar way, 2 samples were produced from the bottom of each tube, which in total gave 48 samples of about 0.3 g each.

For two of the tubes (#2 and #5), six additional locations equally distributed along the length of each tube was sampled both on the inside and outside. The intention with the latter samples was to study in more detail the distribution of radionuclides along the main axis of the tubes.

2.2. Gamma-ray spectrometry

As the activities of the samples were low (~80 mBq and ~250 mBq for ^{60}Co and ^{110m}Ag, respectively, at the time of these measurements at the end of 2014), the activity measurements were performed in the 225 m deep underground laboratory HADES (Andreotti et al., 2011). Compared to ground level, the muon flux in HADES is reduced by a factor of 5000 and the neutron flux reduction is of the same order of magnitude. Three detectors were employed in this study, Ge-3 (Manufacturer: Eurisys, relative efficiency: 60%; crystal configuration: coaxial), Ge-4 (Canberra, 106%; coaxial so-called XtRa) and Ge-8 (Canberra, 19%; planar, so-called BEGe). They all have an endcap of high purity aluminium. In addition to the 225 m overburden, the detectors are shielded by about 15 cm of old lead and 10 cm to 15 cm of freshly produced electrolytic copper.

The small samples were placed in specially made Plexiglass containers with an inner diameter of 15 mm. The sample height was around 2 mm so the sample geometry was not much different compared to point sources. Therefore, point sources from PTB were employed for the efficiency calibration. The effect from coincidence summing was handled by performing Monte Carlo simulations using the EGSnrc code.

3. Results

All activities are given with a reference date of September 30, 2013 to facilitate comparison with the previous study (Tzika et al., 2015). Fig. 4a shows the massic activity for each swarf for ^{60}Co. Every odd number is from the inside and the subsequent even

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[1] Major ($P\gamma > 10$ %) γ-rays of ^{110m}Ag: 658, 678, 707, 885, 937, 1383, 1505 keV. Minor ($P\gamma$: 1 % - 10 %) γ-rays of ^{110m}Ag: 447, 620, 687, 744, 1476, 1562 keV. Major ($P\gamma > 10$ %) γ-rays of ^{108m}Ag: 434, 615, 723 keV (all with Pg of > 90 %). Minor ($P\gamma$ 1 % - 10 %) γ-rays of ^{108m}Ag: 79 keV (6.9 %).