Applied Radiation and Isotopes 109 (2016) 114–117

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Metrological tests of a 200 L calibration source for HPGE detector systems for assay of radioactive waste drums

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HIGHLIGHTS

- Large (200 L) volume drum source designed, produced and certified as CRM in 2007.
- Source contains 448 identical sealed radioactive ^{152}Eu sources (modules).
- Two metrological inspections in 2011 and 2014.
- No statistically significant changes of the certified characteristics over time.
- Stable calibration source for HPGe-gamma radioactive waste assay systems.

ARTICLE INFO

Article history:
Received 29 March 2015
Accepted 19 November 2015
Available online 27 November 2015

Keywords:
Certified reference material (CRM)
Large volume source
Test procedures
Radioactive waste

ABSTRACT

In this work we present test procedures, approval criteria and results from two metrological inspections of a certified large volume ^{152}Eu source (drum about 200 L) intended for calibration of HPGe gamma assay systems used for activity measurement of radioactive waste drums. The aim of the inspections was to prove the stability of the calibration source during its working life. The large volume source was designed and produced in 2007. It consists of 448 identical sealed radioactive sources (modules) apportioned in 32 transparent plastic tubes which were placed in a wooden matrix which filled the drum. During the inspections the modules were subjected to tests for verification of their certified characteristics. The results show a perfect compliance with the NIST basic guidelines for the properties of a radioactive certified reference material (CRM) and demonstrate the stability of the large volume CRM-drum after 7 years of operation.

1. Introduction

Proper activity measurements of gamma-ray emitting radionuclides are needed for complete characterization of radioactive waste (ISO, 2007). The accurate activity estimation strongly depends on the efficiency calibration of the assay system. The precision of the efficiency calibration in its turn depends on the precision of the calibration method. The methods usually applied for calibration of gamma-ray assay systems are: experimental, using radioactive certified reference material (CRM) and/or computational (including Monte Carlo simulations, e.g. Sima et al. (2004)). The experimental method requires a CRM of the same form and size as the sample and, generally, with uniform activity distribution. In the case of measurements of radioactive waste where large volume drums (about 200 L) are usually used, the experimental calibration methods often use 'quasi-CRM' whose geometrical form and size are the same as that of the drum, but a uniform activity distribution is simulated by consecutive measurements of a point or linear source placed at several suitable positions in the drum (Bruggeman et al., 1999; Toma et al., 2007). In this case, the only certified source is the linear (or point) source and the so-called 'calibration source-drum' relates to the calibration method rather than to a certified reference material. In this work we comment on a real large volume calibration source-drum which is validated as a radioactive CRM.

The large volume source (drum) was designed and produced in 2007 (Mitev et al., 2010). A total of 470 modules were prepared as sealed radioactive sources from a standard solution of ^{152}Eu with activity certified by the Laboratoire Etalons d'Activité of the Compagnie pour l'Etude et la Réalisation de Combustibles Atomiques (LEA CERCA). The modules were produced from

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http://dx.doi.org/10.1016/j.apradiso.2015.11.039
0969-8043/

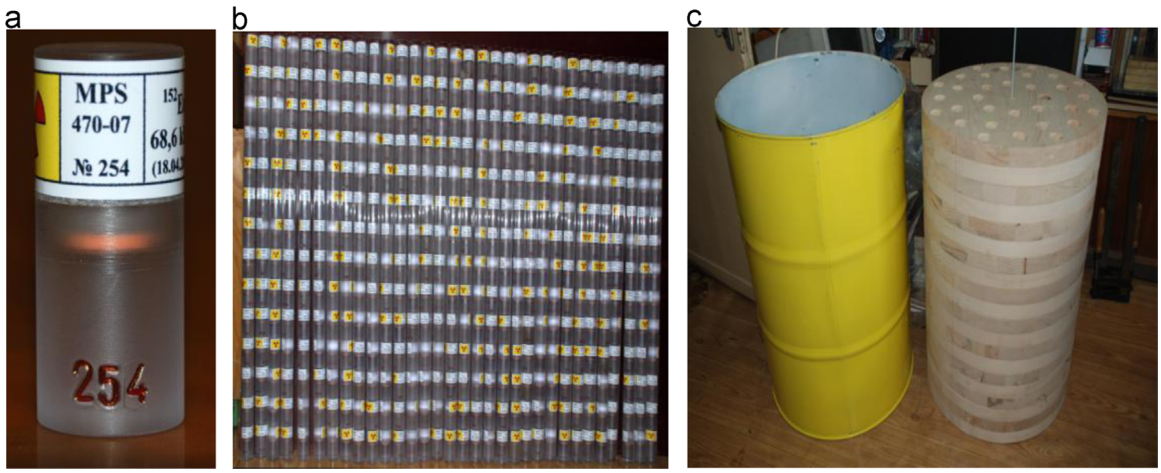

Fig. 1. Photographs of: (a) one module; (b) 32 tubes filled with 448 radioactive modules; (c) the drum and the wooden matrix.

polycarbonate material in cylindrical shape (ϕ 25 × 56 mm) with an activity spot (ϕ 14 × 2 mm) centred at the geometrical centre of the module (Mitev et al., 2010). The activity per module was 68.6 kBq at the reference date of 18.04.2007 with a 2.1% relative expanded uncertainty (k=2). The activity was estimated through the solution mass activity and the mean mass of the solution dropped in each module. The uncertainty includes the uncertainty of the certified solution (1.5%, k=2) and the uncertainty of the mass of the solution dropped in a module (1.4%, k=2). After a number of tests the modules were certified as Certified Reference Materials (CRM-modules) by the Bulgarian National Metrological Institute in 2007. Of these 470 modules, 448 were chosen to be built into the drum. Thus, the large volume source consists of 448 identical small radioactive ^{152}Eu sources (CRM-modules) with 30.7 MBq total activity and a 2.1% relative expanded uncertainty (k=2). The modules were apportioned in 32 transparent plastic tubes which were placed in holes drilled in a homogeneous wooden matrix which filled the 200 L drum (Fig. 1). A specially designed arrangement of the CRM-modules in the drum ensures that the emission of the drum is practically the same as the emission of a drum with homogenously distributed activity as fully described in Mitev et al. (2010).

After a number of tests the drum was also certified as Certified Reference Material (CRM-drum) by the Bulgarian National Metrological Institute in 2007. Since then the CRM-drum is in operation in the Bulgarian state enterprise “Radioactive waste”. The entire construction of the CRM-drum ensures easy and stable usage over time while allowing the tubes with the radioactive modules to be easily removed from the drum and tested.

The objective of this work is to assess the long term stability of the certified characteristics of the CRM-modules and consequently of the CRM-drum. The results of the metrological tests are presented and discussed.

2. Method and material

A full description of the CRM-drum, including the characteristics of the modules, container and of the wood matrix, have been described previously (Mitev et al., 2010). The proper usage of the CRM-drum requires periodic tests of its certified characteristics. Essentially the verification of the CRM-drum requires the verification of the CRM-modules themselves. If the certified activity of the modules is demonstrated, then, by design, the verification of the drum requires only a simple check of the total number and the correct positioning of the modules in the matrix (Mitev et al., 2010). Two inspections of the CRM-drum were performed since its production: 150 modules were tested in 2011 and 150 – in 2014. During the inspections, the modules were subjected to the following tests:

2.1. Leak test

A leak test (wipe test) was performed in order to prove that the sealed sources (modules) are uncompromised as per ISO (1992). The surface of each module was wiped and the swab was measured by a low-level gross beta counting system with a typical background count rate 5.5(5) min^{-1} and MDA (*a priori* estimate) 0.6 Bq for 20 min counting time. The approval criterion requires the swab activity to be $<$0.2 kBq (ISO, 1992). In our case, this value is less than 20% of the stated uncertainty of the activity of the module.

2.2. Activity test

The main test was directed to a verification of the certified activity of the modules. All measurements were made with a gamma-ray spectrometry system, consisting of an HPGe detector (relative efficiency 24.9% and resolution 1.9 keV for the 1332 keV line of ^{60}Co). The system was calibrated using a certified by the Czech National Metrological Institute point source (^{152}Eu, uncertainty 2.2%, k=2.58) placed in a blank module so as to ensure the right measurement geometry.

It should be noted that all modules were measured right after their production in 2007 and the mean (with respect to all modules) activity was estimated. This measurement was an independent test of the certified activity at the production stage and can be regarded as a confirmatory measurement in the terms of NIST (2014). The relative difference between measured (69.092 kBq) and certified (68.6 kBq) values of the activity was 0.7% which is entirely within the range of the stated uncertainty.

Presuming that the same detector is used during recurrent inspections and the detector efficiency is stable, one could measure only the full-energy peak counting rate in order to check whether the activity of the modules change over time (apart from the radioactive decay). Unfortunately the detector efficiency is not stable – in most cases it decreases over time or a detector repair may be needed. For example, the change of the relative efficiency of the detector used in this work is shown in Fig. 2. It can be seen that the detector efficiency is not the same in 2007, 2011 and 2014. Because of that, a new efficiency calibration was performed whenever the activity of the modules had to be verified.

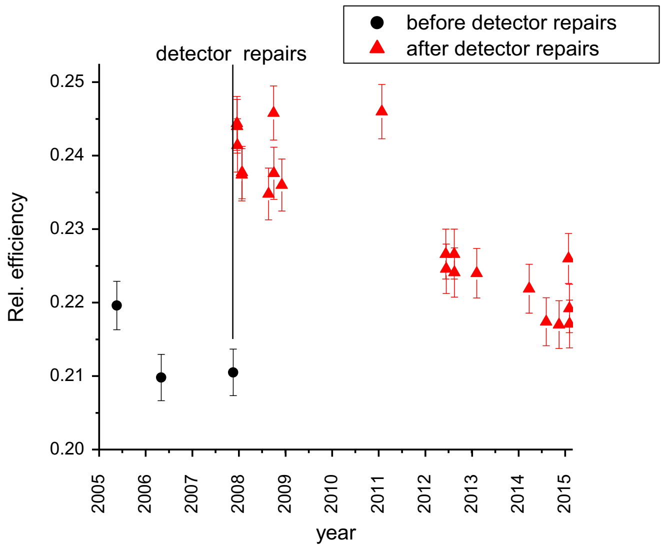

Fig. 2. Changes of the relative efficiency of the detector used in this work. The relative efficiency was estimated according to IEC (1995).

The mean activity of the modules initially measured in 2007 (69.092 kBq) was accepted as a control value ($A_{control}$) which allowed checking whether the properties of the modules alter over time (apart from radioactive decay). During the inspections in 2011 and 2014, the modules were measured again and the new estimated mean activities (test values A_{test}) were compared to the control value. The approval criterion suggests that the relative difference between the test value and the control value should be less than 25% of the stated certified uncertainty (NIST, 2014). In our case this means that the relative difference between the values measured during the inspections (A_{test}) and the control value ($A_{control}$) of the activity is to be less than 0.5%.

3. Results from the inspections

3.1. Results from the leak test

None of the swabs measured during the inspections in 2011 and 2014 gave a statistically significant net count rate (Fig. 3). The critical level (1.25 min^{-1}, α=0.05) was estimated according to Currie (1968) and the estimated MDAs (*a posteriori* estimates) were each less than 0.5 Bq. The results show that the modules are

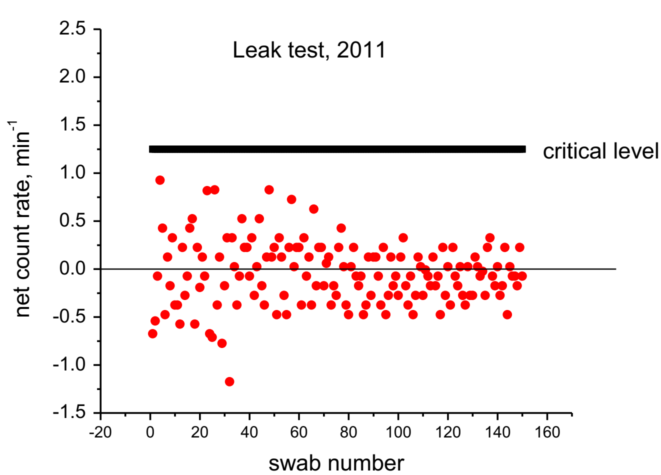

Fig. 3. Net counting rate of the swabs compared to the critical level. The results from the leak test of 2014 are similar and are not shown.

leaktight in the sense of ISO (1992).

3.2. Results from the activity test

The activity of each of the measured modules was evaluated as a mean value $\bar{A}$of the activity estimates A_i by ^{152}Eu gamma lines at energies E_i: 121.78, 344.28 and 1408.01 keV. The uncertainty of $\bar{A}$ was estimated as the higher value between the standard deviation $s(\bar{A})$ and the propagated uncertainty $\sigma(\bar{A})$, where $s(\bar{A}) = \sqrt{\frac{\sum_i (A_i - \bar{A})^2}{m(m-1)}}$, $\sigma(\bar{A}) = \frac{\sqrt{\sum_i \sigma^2(A_i)}}{m}$ and $\sigma(A_i)$ are the individual uncertainties based on the counting statistics of the net area of the full energy peak at energy E_i. Finally, the control value of the module activity (2007), as well as the activities measured during the inspections in 2011 and 2014 were estimated as a mean (with respect to all measured modules) value. The respective uncertainty of the mean value was estimated as the higher value between the standard deviation and the propagated uncertainty, using the same equations as above. But, in this case A_i and $\sigma(A_i)$ are the activity of each module and its uncertainty, which were estimated in the previous step. The expanded (k=2) uncertainties U' include only the uncertainty due to counting statistics of the modules measurement. It does not include the uncertainty due to the calibration by the point ^{152}Eu source, since one and the same point source, the stability of which was verified over the same time period, was used for HPGe-detector calibration in 2007, 2011 and 2014 and the uncertainty of the efficiency due to the counting statistics was negligible. All activities are recalculated to the reference date (the uncertainty of the decay correction is negligible). The results of activity test are presented in Table 1. It can be seen from Table 1 that the relative differences (Δ, %) between the measured test values A_{test} and the control value $A_{control}$ are less than 0.5% and are entirely within the range of the statistical uncertainty.

4. A new application of the SRM modules

A total of 470 modules were prepared and certified as radioactive CRM and 448 of them were loaded in the CRM-drum. Each of the remaining modules could be used for efficiency calibration of HPGe detectors. In a recent study, we found that the geometry of the modules is very convenient for HPGe measurements of radon absorbed in small-size thin foils. The HPGe measurement of ^{222}Rn activity absorbed in thin polymer foils is a useful technique in the studies of radon absorption in plastics (Pressyanov et al., 2011). We found that the CRM-modules are suitable for calibration of such measurements because blank modules with the geometry of the CRM-modules are easy to produce and because the polymer foils can easily be cut to fit the right measurement geometry. Thus, the CRM-modules have shown to be long term stable efficiency calibration sources for HPGe studies of radon absorption in thin polymer foils.

Table 1
Comparison between the test values A_{test} measured during the later inspections and the control value of the module's activity $A_{control}$ measured right after production.

	A, kBq ± U′,% (k=2)	$\Delta, \% = \frac{A_{test} - A_{control}}{A_{control}} \cdot 100$
Control value (2007)	69.092 ± 0.056%	–
Test value (2011)	69.058 ± 0.11%	−0.049%
Test value (2014)	69.111 ± 0.095%	0.027%

5. Conclusions

This work demonstrates that the design of the large-volume CRM-drum with many easy-to-remove sealed sources (CRM-modules) makes easier the metrological tests of its certified characteristics and offers years of stable operation. The results of the two inspections show a perfect compliance with the NIST basic guidelines for the properties of a radioactive CRM: the CRM-modules are stable – the test results show no statistically significant changes of the certified characteristics; the entire CRM-drum is similar to the waste-drums and allows quick and reliable calibration of the assay system; the CRM-modules can also be used for other applications.

Acknowledgments

This work is supported in part by the Bulgarian National Science Fund under contract DFNI T02/13”POLYRAD”. The tests in 2011 and 2014 are performed in the framework of contracts between “Three sigma” LTD and SE “RAW”. We acknowledge the participation of Mr. L. Minev in the gross beta measurements of the swabs.

References

Bruggeman, M., Gerits, J., Carchon, R., 1999. A minimum biased shell-source method for the calibration of rad waste assay systems. Appl. Radiat. Isot. 51 (3), 255–259.

Currie, L.A., 1968. Anal. Chem. 40 (3), 586.

IEC 6 1452 1995-08, Nuclear instrumentation – Measurement of gamma-ray emission rates of radionuclides – Calibration and use of germanium spectrometers.

ISO 21238:2007 (E). Nuclear energy- Nuclear fuel technology- Scaling factor method to determine the radioactivity of low- and intermediate-level radioactive waste packages generated at nuclear power plants.

ISO9978:1992 (E). Radiation protection – Sealed radioactive sources – Leakage test methods.

Mitev, K., Boshkova, T., Minev, L., 2010. Design, Production, Metrological Tests and Certification of a Large-volume (200 L) Calibration Source for Gamma-spectrometry Systems for Assay of Radioactive Waste Drums, N18-227. IEEE Nucl. Sci. Symp. Conf. Rec., 464–471.

NIST, 2014. Radiation Physics Division, Radioactivity Standard Reference Materials, SRM Series 4xxx, IRD-P-15, Procedure 15v305, Version 3.05/2014, 〈http://www.nist.gov/pml/div682/qsprocedures.cfm〉.

Pressyanov, D., Georgiev, S., Dimitrova, I., Mitev, K., Boshkova, T., 2011. Determination of the diffusion coefficient and solubility of radon in plastics. Radiat. Prot. Dosim. 145 (2–3), 123–126.

Sima, O., Cazan, I.L., Dinescu, L., Arnold, D., 2004. Efficiency calibration of high volume samples using the GESPECOR software. Appl. Radiat. Isot. 61, 123–127.

Toma, M., Sima, O., Olteanu, C., 2007. Experimental and simulated studies for the calibration of a radioactive waste assay system. Nucl. Instr. Methods A 580, 391–395.