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A filter spiked with radioactivity dedicated for use in the EU safeguards laboratory in Luxembourg

A quality assurance tool for the radiation protection and nuclear safety laboratory

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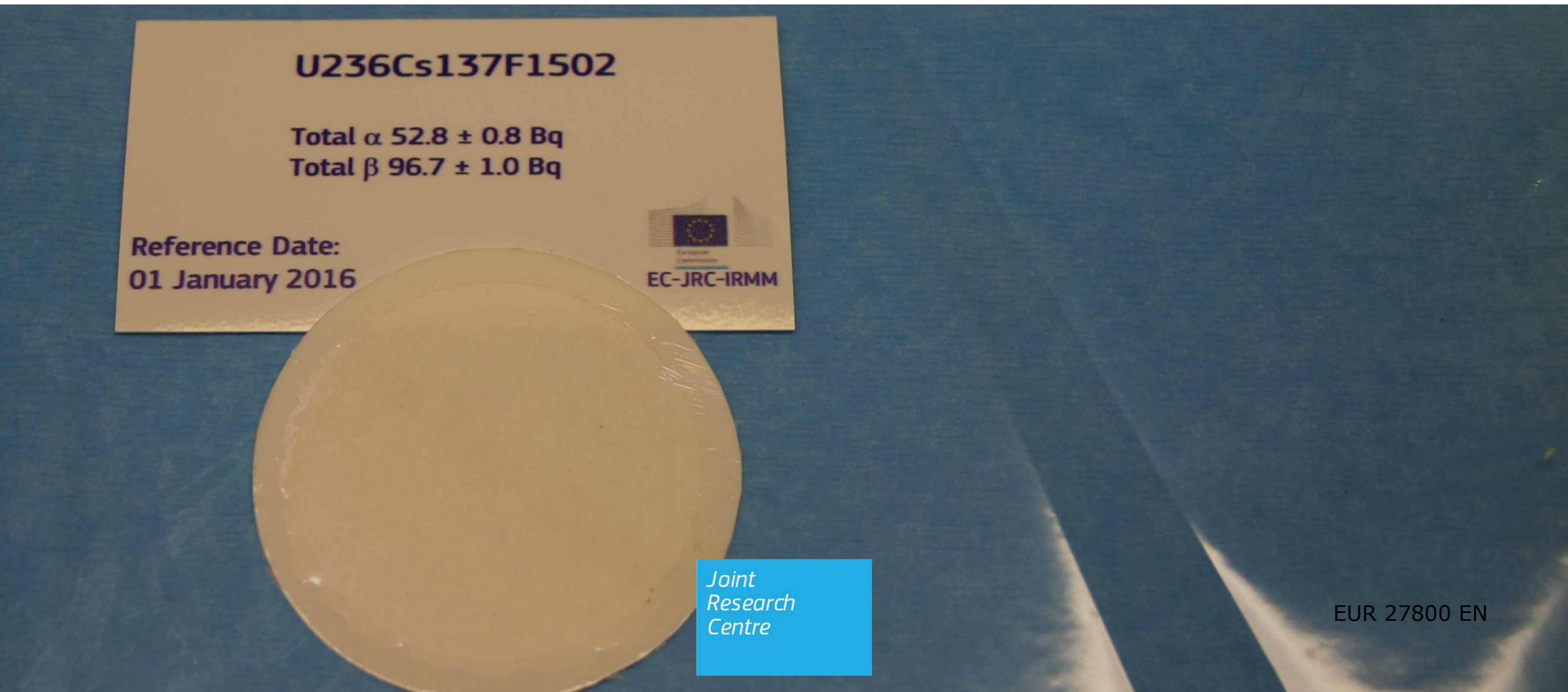

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Abstract

The Radiation Protection and Nuclear Safety Unit DG Energy requested JRC-Geel to make a spiked filter with certified values for the total alpha and beta activity (ARES:(2016)28718). This filter will be used as a quality assurance tool for testing the performance of instruments measuring the radioactive contamination of smear samples taken on equipment returned by nuclear safeguard inspectors.

A blank filter was laminated with a plastic foil on the backside in order to make it mechanically stronger. Then, the filter was spiked with standardised solutions of mainly two alpha-radiation emitting radionuclides (^{234}U and ^{236}U) and one beta-radiation emitting radionuclide (^{137}Cs). The exact amount of solution dispensed on the filter was determined by weighing the pycnometer containing the standardised solution, before and after dispensing. After spiking and drying, the filter was covered with a thin Mylar-foil to prevent the loss of material. The certified activities were 52.8 ± 0.8 Bq for the total alpha activity and 96.7 ±- 1.0 Bq of total beta activity.

1 Introduction

The Radiation Protection and Nuclear Safety Unit (D.3) of DG Energy is preparing their laboratory for ISO17025 accreditation. Amongst the requirements are regular participation in proficiency testing schemes and documented use of reference materials for internal validation of measurements. To fulfil these requirements for measurements of smear tests collected from equipment returned by nuclear safeguard inspectors, a dedicated reference material with certified values for total activity of alpha- and beta emitting radionuclides is needed. The importance of such a material is highlighted by the fact that there are no proficiency tests available for this type of measurements.

For these reasons a formal request was made to the JRC to make a reference filter suitable as reference materials for internal validation of the smear tests. (ARES: (2016)28718).

After mutual discussions between the people in charge of operating the laboratory in Luxembourg and the ones in charge of source production at JRC-Geel, they agreed on the technical specifications of the source:

Diameter: 55 mm diameter

Material: filter: Whatman® filters, grade 4, Cat No 1004-055

Radionuclides: Certified values for the activity of

alpha-emitting radionuclide: ^{234}U and ^{236}U (about 50 Bq requested)

beta-emitting radionuclide: ^{137}Cs (about 100 Bq requested)

2 Materials and Methods

2.1 Filter preparation

A box of blank paper filters used in the health protection laboratory in Luxembourg was sent to JRC-Geel. The filters are commercially available paper filters having a 55 mm diameter (Whatman® filters, grade 4, Cat No 1004-055). As the reference filter will be subject to frequent manipulations it was first necessary look for a suitable method to strengthen it mechanically. It was also necessary to investigate methods of ascertaining that no loss of radioactive material during the spiking and contamination of the underlying surface would occur.

2.1.1 Filter lamination

In order to both strengthen the filter and prevent leakage of dispensed liquid, the filter was laminated between two 0.125 mm thick plastic foils. The bottom foil was completely covering the backside of the filter while the upper foil had a 45 mm diameter circular opening.

An additional advantage of the lamination is that at the edge of the filter, the operator has a 5 mm border to manipulate the filter with e.g. tweezers, without touching the active area of the filter. In Figure 1 the process of filter lamination is shown.

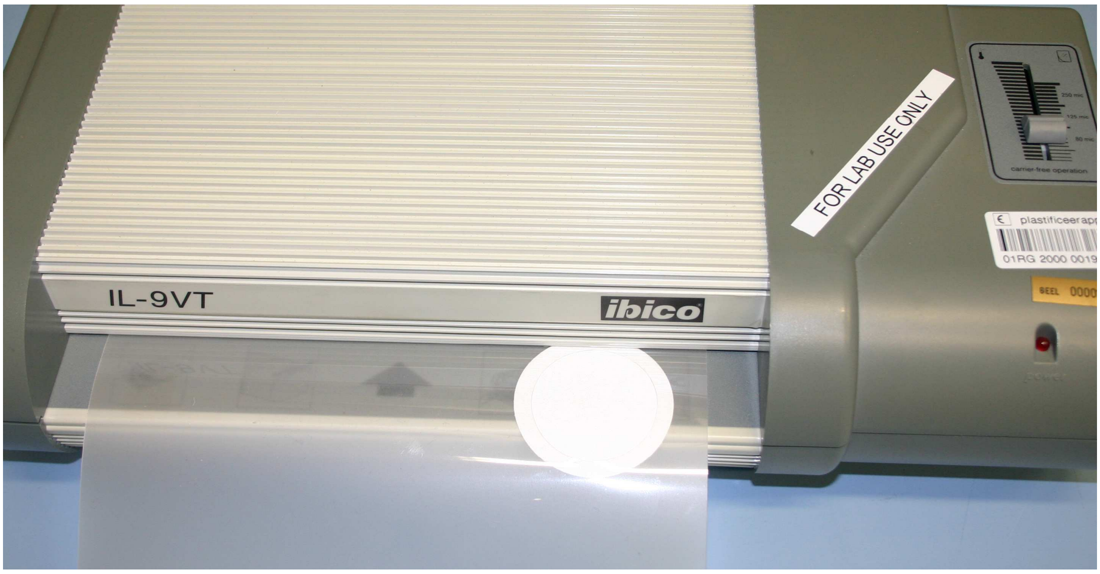

Figure 1: Lamination of the paper filter

2.1.2 Adsorption capacity test

In order to distribute the activity over the area of the filter several drops of the alpha- and beta-particles emitting solutions had to be dispensed on the filter. The liquids had to be dispensed in such a way that the spread of activity remains within the 45 mm circular area at the upper side of the laminated foil and that it is rather homogenous. Tests were carried out to determine how much liquid could be dispensed without spreading outside the 45 mm area. They indicated that no more than 100 μL could be dispensed at once. If more liquid needs to be dispensed on one filter, the filter has to be dried in between consecutive dispenses.

2.2 Standard solutions

For the spiking of the filter two standard solutions were used: one containing alpha-particle emitting radionuclides and one containing a beta-particle emitting radionuclide.

2.2.1 Alpha-particle emitting solution

The alpha-particle emitting solution that was used to spike the filter contained a mixture of different alpha-particle emitting uranium isotopes. The activity fractions of the U-isotopes as determined using mass spectrometry in 2007 are listed in Table 1 (Certificate IM/MeaC/07/117). The dominating U-isotopes are ^{236}U (88% by activity) and ^{234}U (11% by activity).

The solution was standardised using defined solid angle α-particle counting according to WI-D-00016 "Alpha particle counting in defined low solid angle". The total alpha activity was 1030 ± 10 Bq/g on the reference date 1 January 2016.

Table 1 : Activity fraction of the different U-isotopes contributing to the total α–particle emission in the solution used for the spiking.

α-radiation	**Activity fraction 1 January 2016 at 00:00:00 UTC**
^{233}U	0.00164 (0.00003)
^{234}U	0.11385 (0.00009)
^{235}U	0.003074 (0.000003)
^{236}U	0.88137 (0.00009)
^{238}U	0.0000690 (0.0000001)

The purity of the solution was checked using gamma-ray spectrometry. The spectrum did not reveal any gamma-ray not originating from the Uranium isotopes present in the solution or from the natural background.

2.2.2 Beta-particle emitting solution

The beta-particle emitting solution used was a pure ^{137}Cs solution. Its radiopurity was checked by measuring the solution by gamma-ray spectrometry on a Hyper Pure Germanium (HPGe) detector. No other gamma-ray emitting radionuclides could be detected. The solution was standardised by liquid scintillation counting. The total beta activity of the solution is 270 ± 3 Bq/g on 1 January 2016.

2.3 Spiking of the filter

A blank laminated filter was prepared for spiking. The activity requested on the filter was 50 Bq total α–particle emitting solution and 100 Bq total β–particle emitting solution. Taking in consideration the activity concentrations of both solutions we had to dispense about 3 drops of the alpha-solution and about 20 drops of the beta-solution. The drops were dispensed in a pattern as shown in Figure 2. The spiking of the filter was done according to WI-D00471 "Preparation of radioactive sources".

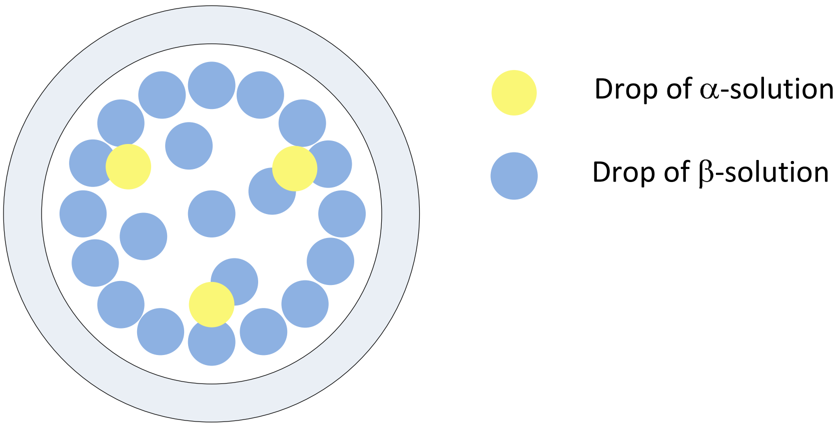

Figure 2: Spiking pattern of the solutions on the laminated filter

2.3.1 Quantitative dispensing of the solutions

The exact amount of solution dispensed on the filter was determined gravimetrically. Two pycnometers, each filled with one of the two different solutions, were used.

The three drops of α-solution were dispensed first. The exact mass of the dispensed solution, 51.282(20) mg, was determined by weighing the pycnometer on a calibrated microbalance before and after dispensing the solution on the filter. The difference in mass is the amount of dispensed solution.

From the β-solution 358.225(20) mg was dispensed. As mentioned in paragraph 2.2, this amount could not be dispensed in one step. Therefore, we dispensed the amount in 5 steps. Between each step the filter was dried under an infrared lamp to accelerate the drying process. The difference in mass of the pycnometer, before the first dispense and after the fifth dispense, corresponds to the exact amount of beta-solution dispensed. Table 2 gives the exact amount of solutions dispensed including the uncertainties (k=1) on the dispensed masses. The weighings were carried out according to WI-D-00155 "Drop deposition" and WI-D-00010 "Use of balances".

Table 2: Dispensed masses of the solutions including uncertainties (k=1)

Solution	Dispensed mass (g)	Absolute uncertainty (g)	Relative uncertainty (%)
α-solution	0.051282	0.000020	0.040%
β-solution	0.358225	0.000020	0.006%

2.4 Filter covering

To prevent loss of material from the filter, resulting in loss of activity, it had to be covered. The cover had to be mechanical strong enough to keep all the material on the filter. On the other hand it should not stop (or reduce its energy significantly) the alpha radiation coming out of the source.

A Mylar® foil with a thickness of 0.5 μm was used to cover the spiked filter. This foil is thin enough to not stop the alpha-particles coming out of the source. As the foil is too thin to be directly glued on the source, the foil was first glued on a support ring using 2-component epoxy glue and left to cure for 24 hours. To glue the foil on the filter the 2-component epoxy glue was applied in a very thin line on the edge of the filter using a turntable and a pneumatic glue dispenser. The ring with the foil was placed gently on top of the filter. The filter was put on a small support to allow the ring with the Mylar® foil to stretch nicely over the whole filter area as shown in Figure 3. After 24 hours of curing of the glue, the Mylar® foil was cut at the edge of the filter. The filter was labelled with identification number: *U236Cs137F1502*.

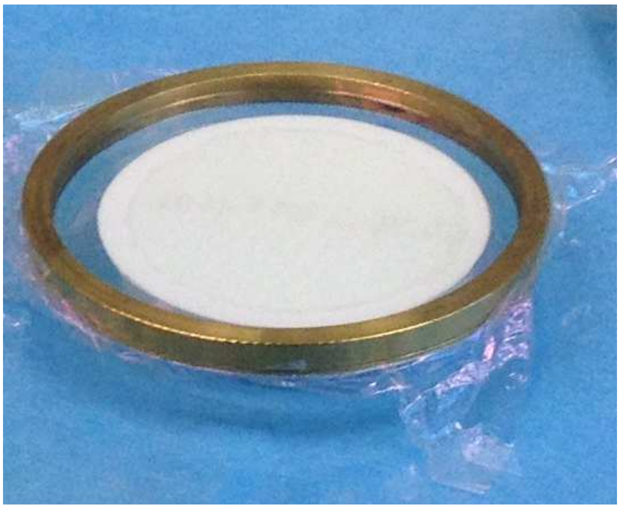

Figure 3: Covering the filter with a Mylar® foil

2.5 Activity dispensed on the filter

To determine the activity on the filter the exact mass of the solutions as mentioned in Table 2 was multiplied with the activity concentration of the solutions. The total uncertainties on the activities are calculated in Table 3. It shows that the dominating uncertainty contribution is coming from the uncertainty on the activity concentration of the used solutions as e.g. the uncertainty on the dispensed mass is very small.

Table 3: Calculation of the activities dispensed on the filter

Solution	*Activity conc (Bq/g)*	*Abs. uncert. (Bq/g)*	*Rel uncert.*	Dispensed mass (g)	Rel. uncert	Total Activity (Bq)	Rel uncert.	Abs. uncert.
α–solution	*1030*	*10*	*0.97 %*	0.0512816	0.040%	52.8	0.97 %	0.51
β–solution	*270*	*3*	*1.11 %*	0.3461114	0.006%	96.7	1.11 %	1.00

2.6 Quality Control

The activity of the dispensed alpha-emitters could not be verified using gamma-ray spectrometry as ^{236}U emits only a few very weak gamma-ray that could not be detected by gamma-ray spectrometry. We performed a check of the composition of the solution using alpha-particle spectrometry. The collected spectrum is shown in Figure 4 (WI-D-00017 "Alpha-particle spectrometry with αSPEC1 and αSPEC2").

The spectrum shows the different alpha peaks coming from the five different uranium isotopes and the fitted alpha curves. The spectra showed no presence of alpha emitters not mentioned on mass spectrometry certificate. The alpha activity on the filter could not be checked by measuring it in one of our alpha defined solid angle measurement set-ups as they are not designed to measure filters.

For this reason and to include unknown sources of errors, we decided to increase the uncertainty on the alpha activity to 0.8 Bq.

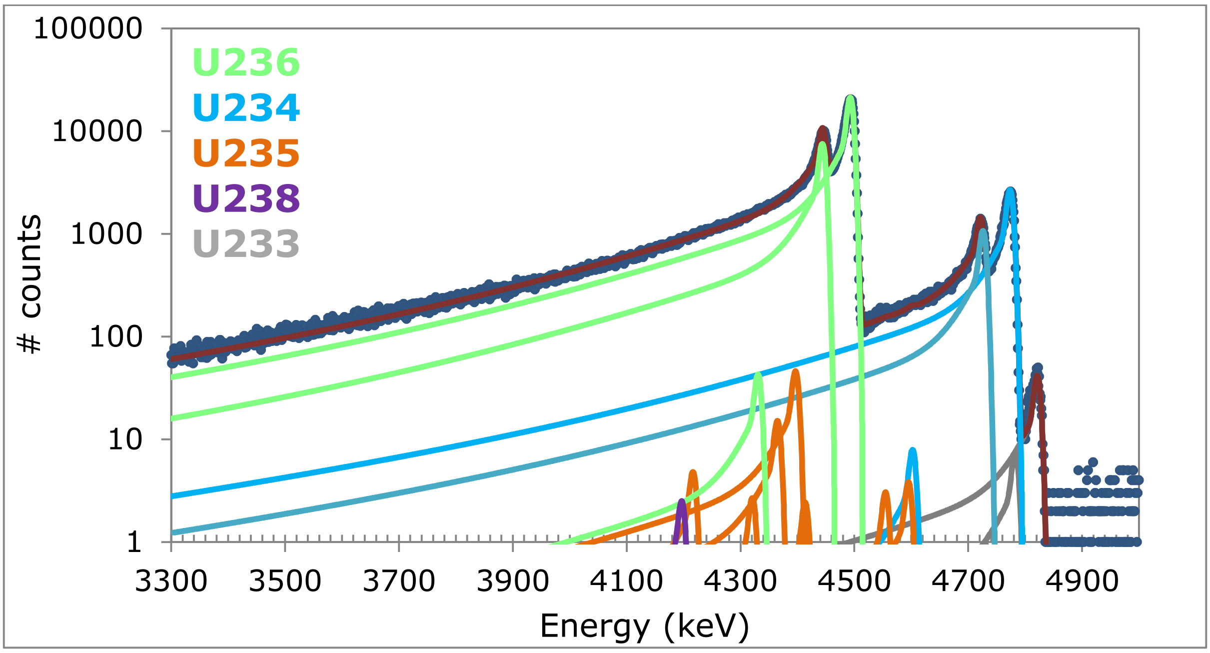

Figure 4: Alpha particle spectrum including fitting of the different alpha peaks.

The beta-activity dispensed on the filter could be checked by measuring it on a HPGe-detector. The 661.7 keV gamma-ray of ^{137}Cs could easily be detected.

The filter was measured on a low-background coaxial HPGe detector (46% relative efficiency) at a distance of 45.2 mm from the endcap of the detector. The full energy peak efficiencies of the detector were determined by measuring a liquid solution from the NPL 2012 environmental radioactivity proficiency testing exercise. The geometry, density and true coincidence summing corrections were calculated using the Monte Carlo (MC) simulation. The MC calculations were conducted using EGSnrc code based on a detector model validated through several proficiency tests.

The results obtained for the beta-particle emitter (96.7 Bq ± 2.9 Bq) confirmed the value obtained from the weighing and standardisation. As the value could be confirmed by an independent measurement we decided to keep the uncertainty unchanged.

The final results are listed in Table 4 containing the certified values for the total alpha– and beta-activity including the uncertainties (k=1).

Table 4: Total alpha and total beta activities of the reference filter.

Parameter	Activity on 1 January 2016 at 00:00:00 UTC
Total α-emitters	(52.8 ± 0.8) Bq
Total β-emitters	(96.7 ± 1.0) Bq

Conclusion

The Radiation Protection and Nuclear Safety Unit of DG Energy requested to produce a reference source to be used as quality control tool to validate their measurements. This is necessary to comply with the requirements of the ISO 17025 laboratory accreditation. For the type of measurements they perform no proficiency tests could be found. In this case a customised solution needed to be provided.

A spiked filter with certified values for the total alpha and beta activities is a suitable alternative to validate internally the analytical method performed in the laboratory. A specially designed source was produced at JRC-Geel. A filter provided by DG Energy was made mechanically stronger by laminating it between two plastic foils leaving an opening on the upper side. A known amount of standardised solution was gravimetrically dispensed over the uncovered filter area. After drying, the filter was covered to prevent possible loss of material. The activities were calculated from the dispensed mass and the activity concentration of the solutions used. The activity of the beta-particle emitting radionuclide was checked by gamma-ray spectrometry confirming the value obtained from the weighings and the standardisation.

The filter will serve as a quality assurance tool to assess the performance of the analysis made by Radiation Protection and Nuclear Safety Unit of DG Energy. The filter will be a key tool in their measurement validation. It will enhance the trust in the analytical results of the laboratory.

References

ARES: (2016)28718 : registered e-mail

Certificate IM/MeaC/07/117, NPL-Uranium-236 Ampoule E-5557

WI-D-00010 Use of balances

WI-D-00016 Alpha particle counting in defined low solid angle

WI-D-00017 Alpha-particle spectrometry with αSPEC1 and αSPEC2

WI-D-00155 Drop deposition

WI-D-00471 Preparation of radioactive sources

List of abbreviations and definitions

Abs. uncert. = absolute uncertainty

Rel. uncert. = relative uncertainty

Bq = Becquerel

k = coverage factor

g = gram

DG-Energy = Directorate-General for Energy

SN3S unit = Standards for nuclear safety, security and safeguards unit

HPGe = Hyper Pure Germanium detector

MC = Monte Carlo

ISO = International Organization for Standardisation

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