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New data concerning the efficiency calibration of a drum waste assay system. Part I: Experimental calibration

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Abstract. The study is focused on the efficiency calibration of the gamma spectroscopy system used in the NIPNE drum waste assay facility. The measurement of a radioactive drum waste is generally difficult because of its high volume, the complex and usually unknown distribution of the waste in the drum and its high self-attenuation. To solve these problems, a complex calibration of the system is required. For this purpose, a calibration drum filled with Portland cement and provided with seven tubes, placed at different distances from its center was used.

For the efficiency calibration appropriate for a uniformly distributed source, the shell method, using a linear source of ^{152}Eu, was applied. The linear calibration source was introduced successively in the seven tubes, the gamma-spectra were collected while the drum was translated and simultaneously rotated. Using the GENIE-PC software, the gamma-spectra were analyzed and the detection efficiencies were obtained.

For the efficiency calibration in the case of a non-homogenous source, supplemental measurements in the following geometries were made. First with a point source of ^{152}Eu placed in front of the detector, measured in all seven tubes, the drum being only rotated. Second with the linear source of ^{152}Eu placed in front of the detector, measured in all seven tubes, the drum being only rotated. For each case the gamma spectra were recorded and the detection efficiency was calculated.

Key words: gamma spectroscopy, waste assay system.

INTRODUCTION

The problem of radioactive waste has become a critical issue in the country and in the world. In order to solve this problem it is necessary to analyze, to classify and to finally dispose the radioactive wastes. The classification and the disposal solution are based on the information concerning the risk degree of these wastes, which depends on the their composition (radionuclides, disintegration type, lifetime, activity) and also on their matrix.

We must underline that in the context of Horia Hulubei National Institute of Physics and Nuclear Engineering Nuclear Reactor's decommissioning the problem of radioactive waste analysis is of utmost importance.

In this context Madera is used for characterization of radionuclide content of containers with radioactive waste, conditioned at the Radioactive Waste Treatment Plant, IFIN-HH, in order to be transported and stored at the National radioactive waste Repository-Baita Bihor.

The gamma spectrometry analysis of the radioactive waste containers is an efficient method to determine the composition of the waste [1]. The qualitative analysis to determine the radionuclides present in the containers can be easily achieved because the energy calibration can be done with common radioactive sources.

The quantitative analysis to determine the activity of each radio nuclide is more difficult due to the efficiency calibration. In principle, it should be carried out with reference sources that have the same geometry and the same matrix with the measured waste container. The non-homogeneous distribution of radioactive waste inside drums poses additional problems [3]. As there are no reference sources with the required properties for direct calibration of the detection system, the effort necessary for the indirect experimental calibration is considerably greater and it cannot accurately describe all real waste configurations inside the drum [4, 5, 6, 7, 8].

General aspects

The radioactive waste is generally packed in type A containers (220l drums). For the intermediate and final disposal of medium and low activity radioactive waste, the Romanian law authorizes this kind of packing.

For the efficiency calibration, a similar drum filled with Portland cement and a linear reference source of ^{152}Eu was used. In order to minimize the effect of non-homogeneous matrix and sources distribution in typical measurements of waste drums, the drum is rotated and translated in front of a HPGe collimated detector, while the measurement is performed. To obtain the best possible accuracy (independent of the distribution of radioactive material in the drum) a double translation is performed.

Calibration drum

A 220 l stainless steel drum is used for efficiency calibration. The drum is provided with 7 hollow parallel tubes of 20 mm diameter, the rest of the drum being filled with Portland cement similar with the one used for the drums conditioned with radioactive waste (ρ=2.00g/cm^3). The tubes are placed in the calibration drum at different radial distances from the drum centerline as follows: 7 (center), 6 (7cm), 5 (12cm), 4 (16cm), 3 (19cm), 2 (21.5cm), 1 (24.5cm) from center, respectively (Fig. 1). The tubes are used to introduce the linear reference source into the drum to perform the efficiency calibration. During the efficiency calibration only one tube at a time was filled with the linear reference source leaving the others empty.

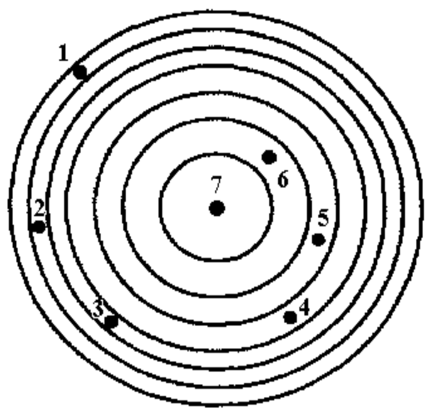

Fig.1 - Section on the calibration drum. 1…7 represent the tubes used to introduce the ^{152}Eu reference source

^{152}Eu-linear reference source

The linear reference source is composed of six ^{152}Eu liquid sources of equal activity, each introduced in a glass ampoule with length l=55mm (Fig 2). The ampoules were placed at equal distances in a plastic tube. The length of the linear source is 620 mm equal with the length of the active volume of the drum. Both ends of the drum are filled with 10 cm of cement.

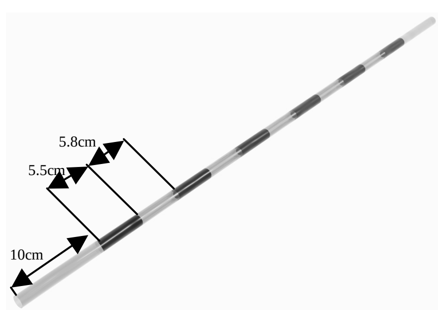

Fig. 2 - ^{152}Eu reference source

Efficiency calibration

The efficiency calibration for a uniformly distributed radioactive waste was performed using the shell-sources method, for two measurement configurations.

In year 2000 an efficiency calibration was performed, using a lead-collimated horizontal HPGe of 20% relative efficiency. The diameter of the collimator window was 78 mm, the wall thickness of the lead shield 50 mm. That measurement geometry, hereafter denominated as initial geometry, is displayed in Fig. 3a, including the detector and the calibration drum.

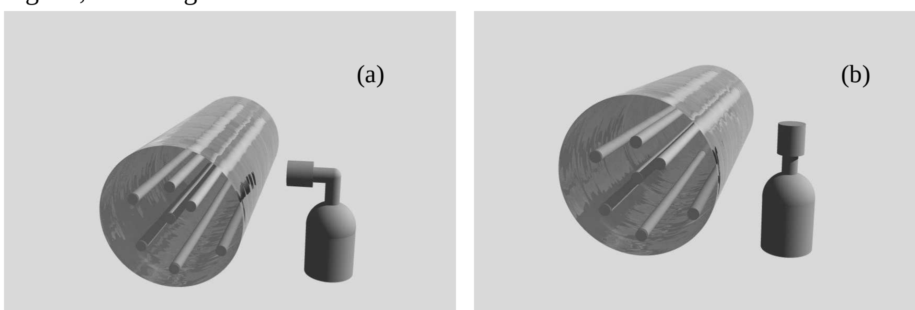

Fig 3 - A schematic representation of the two measurement geometries: (a) initial geometry. (b) present geometry

Due to an accidental damage suffered by the detector, the geometry has been changed, and in 2003 the efficiency calibration was repeated. In the new geometry of the experimental setup we used a lead-collimated vertical HPGe detector (Fig. 3b), of 30% relative efficiency and 1.8 keV energy resolution (at 1332 keV). The rectangular collimator window has the length 10 cm and the width 4 cm. The lead shield has the interior diameter 9 cm and the wall thickness 4.4 cm. The detector was located at 32.5 cm radially from the surface of the calibration drum.

The gamma spectroscopy technique was used for measurements. The gamma ray energies given in Table 1 were used for the efficiency calibration.

Table 1. The selected gamma rays emitted by ^{152}Eu and their yields, used for efficiency calibration

Energy (keV)	121.78	344.2	778.9	964	1112	1408
Y(E) (%)	28.4	26.5	12.9	14.6	13.5	20.8

The linear calibration source was introduced successively in the seven tubes. For the source placed in a given tube, the spectrum acquisition was started when the detector was placed at one end of the drum and was continued while the drum was rotated and translated in front of the collimated detector until a complete cycle (forward and backward) was finished. Using the GENIE-PC software, the gamma-spectra were analyzed and the detection efficiencies for shell-sources were obtained [4,6]. Using these efficiencies, the total response of the detector and the detection efficiency appropriate to a uniform volume source were calculated for each of the six gamma ray energies [2].

Besides the efficiency values for a drum completely filled with radioactive waste, the shell-sources method provides the efficiency values for the case when only a part of the drum, namely a cylinder of radius *x*, coaxial with the drum, contains radioactive sources. In Tables 2 and 3 the corresponding efficiencies are presented for several values of *x*, for both geometries. The data are also displayed in Fig. 4.

Table 2. The detection efficiency for several values of *x* in the case of the initial geometry

E (keV)	**Efficiency**					
	28.5(cm)	**24.5(cm)**	**21.5(cm)**	**19.5(cm)**	**16.5(cm)**	**12.5(cm)**
122	$1.265\cdot10^{-6}$	$5.080\cdot010^{-7}$	$2.000\ 10^{-7}$	$9.724\cdot10^{-8}$	$3.180\cdot10^{-8}$	$7.199\cdot10^{-9}$
344	$1.269\cdot10^{-6}$	$6.866\cdot10^{-7}$	$3.834\cdot10^{-7}$	$2.465\cdot10^{-7}$	$1.224\cdot10^{-7}$	$4.404\cdot10^{-8}$
779	$1.106\cdot10^{-6}$	$7.038\cdot10^{-7}$	$4.587\cdot10^{-7}$	$3.364\cdot10^{-7}$	$1.953\cdot10^{-7}$	$8.558\cdot10^{-8}$
964	$1.105\cdot10^{-6}$	$7.360\cdot10^{-7}$	$5.056\cdot10^{-7}$	$3.800\cdot10^{-7}$	$2.353\cdot10^{-7}$	$1.110\cdot10^{-7}$
1112	$1.136\cdot10^{-6}$	$7.712\cdot10^{-7}$	$5.419\cdot10^{-7}$	$4.134\cdot10^{-7}$	$2.612\cdot10^{-7}$	$1.260\cdot10^{-7}$
1408	$1.131\cdot10^{-6}$	$7.969\cdot10^{-7}$	$5.759\cdot10^{-7}$	$4.451\cdot10^{-7}$	$2.929\cdot10^{-7}$	$1.469\cdot10^{-7}$

Table 3. Same as Table 2 for the present geometry

E (keV)	**Efficiency**					
	28.5	**24.5**	**21.5**	**$19.5\cdot10^{-8}$**	**16.5**	**12.5**
122	$1.250\cdot10^{-6}$	$5.540\cdot10^{-7}$	$2.190\cdot10^{-7}$	$9.930\cdot10^{-7}$	$2.530\cdot10^{-8}$	$3.570\cdot10^{-9}$
344	$1.470\cdot10^{-6}$	$7.760\cdot10^{-7}$	$4.020\cdot10^{-7}$	$2.370\cdot10^{-7}$	$9.920\cdot10^{-8}$	$3.230\cdot10^{-9}$
779	$1.330\cdot10^{-6}$	$8.010\cdot10^{-7}$	$4.960\cdot10^{-7}$	$3.430\cdot10^{-7}$	$1.860\cdot10^{-7}$	$7.730\cdot10^{-8}$
964	$1.350\cdot10^{-6}$	$8.270\cdot10^{-7}$	$5.250\cdot10^{-7}$	$3.710\cdot10^{-7}$	$2.090\cdot10^{-7}$	$9.070\cdot10^{-8}$
1112	$1.350\cdot10^{-6}$	$8.500\cdot10^{-7}$	$5.580\cdot10^{-7}$	$4.060\cdot10^{-7}$	$2.380\cdot10^{-7}$	$1.080\cdot10^{-7}$

1408	$1.420\cdot10^{-6}$	$9.180\cdot10^{-7}$	$6.190\cdot10^{-7}$	$4.590\cdot10^{-7}$	$2.780\cdot10^{-7}$	$1.310\cdot10^{-7}$

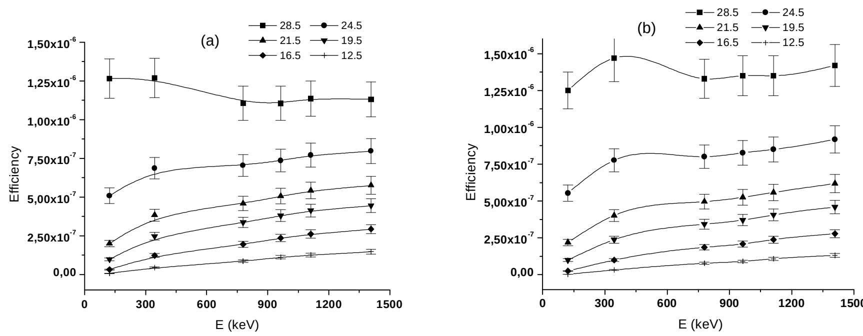

Fig. 4 - The dependence *ε=f(E)* for six waste cylinders having different radii, embedded into the drum; (a) initial geometry. (b) present geometry

From Fig. 4 it can be observed that for a source distributed in the entire volume of the drum, the calibration curve has a pattern approximately similar to a normal one. For a source distributed in smaller volumes (cylinders of the same length as the drum, but with the radius *x* smaller than the radius *R* of the drum), the attenuation of gamma rays of low energy in cement layer is much stronger than that of the high-energy gammas. As a result, the efficiency curve presents a slowly increasing pattern with energy, though the relative efficiency of the detector decreases with gamma ray energy.

In order to underline the gamma ray attenuation contribution to the efficiency value, the detection efficiency for a cylindrical source of radius *x*, embedded in the drum, is displayed in Fig 5 in function of *R-x*, which represents the thickness of the inactive cement layer surrounding the cylindrical radioactive source. It can be seen that, in the given geometry - a high density of sample matrix and a collimated detector - the efficiency curve presents a sharply decreasing pattern for low energies, compared to a slowly decreasing pattern in the high-energy case.

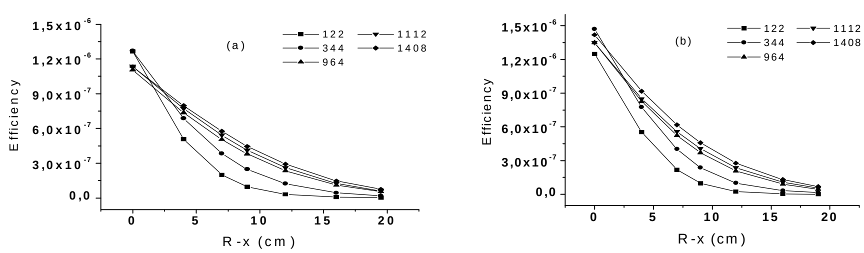

Fig. 5 - The variation of the detection efficiency for a cylindrical source of radius *x* in function of the thickness *R-x* of the inactive cement layer; (a) initial geometry, (b) present geometry

For the present measurement geometry (vertical HPGe detector) supplemental measurements were carried out in order to obtain informations about the detection efficiency for a non-homogenous source. The following cases were studied:

a) with the detector placed at half-distance between drum ends, using a quasi-point source, the drum being only rotated. (As the point source, one of the six liquid ^{152}Eu sources, from the linear reference source, was used).

b) with the detector placed in the same position as in the case a), but using the ^{152}Eu-linear reference source, the drum being only rotated.

For both measurement geometries the sources were measured in each of the seven positions, the gamma spectra were collected and the efficiency was calculated.

In Fig. 6 the efficiency curves in the two supplemental measurement geometries are represented for the cases when the ^{152}Eu source is placed in the tubes 1 (r=24.5 cm) to 6 (r=7 cm). This figure shows that the calibration curves have the same pattern with the calibration curves obtained in the case of uniformly distributed sources.

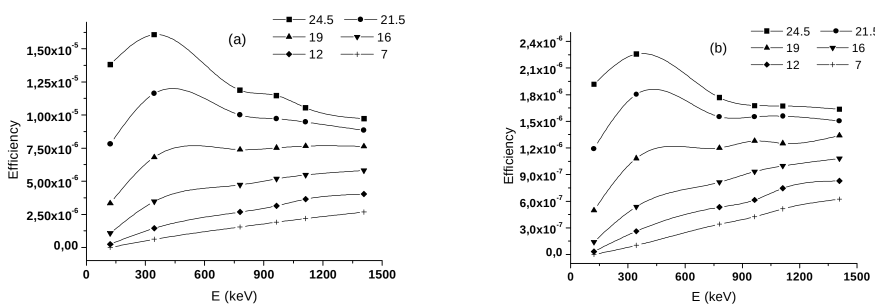

Fig 6 - The efficiency curves obtained in the supplemental measurements, (a) point source geometry, (b) linear source geometry

The efficiency values obtained in the two supplemental measurement geometries are presented in Tables 4 and 5, respectively.

Table 4. The efficiency values for the point source placed in several tubes inside the drum

E (kev)	Efficiency						
	24.5	21.5	19	16	12	7	0

122	$1.38\cdot10^{-5}$	$7.81\cdot10^{-6}$	$3.33\cdot10^{-6}$	$1.07\cdot10^{-6}$	$2.40\cdot10^{-7}$	0.00	0.000
344	$1.61\cdot10^{-5}$	$1.16\cdot10^{-5}$	$6.81\cdot10^{-6}$	$3.47\cdot10^{-6}$	$1.45\cdot10^{-6}$	$6.20\cdot10^{-7}$	$4.09\cdot10^{-7}$
779	$1.19\cdot10^{-5}$	$1.00\cdot10^{-5}$	$7.39\cdot10^{-6}$	$4.72\cdot10^{-6}$	$2.68\cdot10^{-6}$	$1.55\cdot10^{-6}$	$1.19\cdot10^{-6}$
964	$1.15\cdot10^{-5}$	$9.72\cdot10^{-6}$	$7.52\cdot10^{-6}$	$5.18\cdot10^{-6}$	$3.15\cdot10^{-6}$	$1.91\cdot10^{-6}$	$1.58\cdot10^{-6}$
1112	$1.06\cdot10^{-5}$	$9.47\cdot10^{-6}$	$7.69\cdot10^{-6}$	$5.48\cdot10^{-6}$	$3.65\cdot10^{-6}$	$2.18\cdot10^{-6}$	$1.83\cdot10^{-6}$
1408	$9.72\cdot10^{-6}$	$8.83\cdot10^{-6}$	$7.63\cdot10^{-6}$	$5.81\cdot10^{-6}$	$4.03\cdot10^{-6}$	$2.67\cdot10^{-6}$	$2.31\cdot10^{-6}$

Table 5. Same as Table 4 in the case of the linear reference source

E (kev)	**Efficiency**						
	24.5	**21.5**	**19**	**16**	**12**	**7**	**0**
122	$1.92\cdot10^{-6}$	$1.19\cdot10^{-6}$	$4.96\cdot10^{-7}$	$1.41\cdot10^{-7}$	$3.29\cdot10^{-8}$	0.00	0.00
344	$2.26\cdot10^{-6}$	$1.80\cdot10^{-6}$	$1.08\cdot10^{-6}$	$5.38\cdot10^{-7}$	$2.62\cdot10^{-7}$	$1.03\cdot10^{-7}$	$8.07\cdot10^{-8}$
779	$1.77\cdot10^{-6}$	$1.55\cdot10^{-6}$	$1.20\cdot10^{-6}$	$8.14\cdot10^{-7}$	$5.32\cdot10^{-7}$	$3.41\cdot10^{-7}$	$2.67\cdot10^{-7}$
964	$1.68\cdot10^{-6}$	$1.55\cdot10^{-6}$	$1.28\cdot10^{-6}$	$9.34\cdot10^{-7}$	$6.14\cdot10^{-7}$	$4.25\cdot10^{-7}$	$3.47\cdot10^{-7}$
1112	$1.67\cdot10^{-6}$	$1.56\cdot10^{-6}$	$1.25\cdot10^{-6}$	$9.98\cdot10^{-7}$	$7.46\cdot10^{-7}$	$5.14\cdot10^{-7}$	$4.24\cdot10^{-7}$
1408	$1.64\cdot10^{-6}$	$1.50\cdot10^{-6}$	$1.34\cdot10^{-6}$	$1.08\cdot10^{-6}$	$8.29\cdot10^{-7}$	$6.23\cdot10^{-7}$	$5.28\cdot10^{-7}$

From the Tables 4 and 5 it can be observed that the efficiency values obtained using the point-source are approximately six times greater than the ones obtained with the ^{152}Eu-linear reference source.

The ratios between the efficiency values obtained in the point source geometry and the efficiency values obtained in the linear reference source geometry are presented in Table 6.

It can be observed that the ratios are decreasing with the increase of the distance between the detector and the position of the source inside the drum (0=center of the drum, 24.5=the position nearest to the surface of the drum). This behavior is explained by the fact that the collimated detector “sees” the sources under different solid angles, smaller for the source position closer to the detector, bigger for the source placed at grater distances. The variation of the ratio with energy is due to gamma ray attenuation: the attenuation of low energy photons in the cement layer is much stronger than that of the high-energy photons.

Table 6. The ratios between the efficiency values obtained in the point source geometry **(**ε_{PS})
and the efficiency values obtained in the linear source geometry (ε_{LS})

E (keV)	**ε_{PS}/ ε_{LS}**						
	24.5	**21.5**	**19**	**16**	**12**	**7**	**0**
122	7.21	6.55	6.72	7.60	7.29	0.00	0.00
344	7.12	6.44	6.29	6.45	5.53	6.04	5.06
779	6.71	6.45	6.15	5.80	5.03	4.54	4.48
964	6.84	6.26	4.82	5.55	5.13	4.50	4.56
1112	6.31	6.08	6.12	5.49	4.89	4.25	4.33
1408	5.95	5.87	5.69	5.38	4.86	4.29	4.37

Conclusions

The efficiency calibration was performed for a uniformly distributed source for two measurement geometries. The efficiency calibration curves have a similar pattern.

For the efficiency calibration in the case of a non-homogenous source, supplemental measurements using a point source of ^{152}Eu and the linear reference source were performed. The calibration curves have a similar behavior with the calibration curves obtained in the case of uniformly distributed sources.

The ratios between the efficiency values obtained in the point source geometry and the efficiency values obtained in the linear reference source geometry were computed and analyzed.

These ratios are decreasing with the increase of the distance between the detector and the position of the source inside the drum due to the different solid angles in which the detector "sees" the sources, smaller for the source position closer to the detector, bigger for the source placed at grater distances.

The variation of the ratios with energy is due to the different gamma ray attenuation in the cement layer.

Monte Carlo calculations carried out to complement experimental calibration presented in this work are in progress.

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