Non-destructive Method of Characterization of Radioactive Waste Containers Using Gamma Spectroscopy

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Abstract

During the decommissioning of the SATURNE accelerator at CEA Saclay, a number of low or very low activity concrete blocks, containing different radioactive materials, had to be characterized before sending them for storage. A destructive method, being the most precise, is also the most expensive one and not the easiest from the radioprotection point of view. For this reason, in this paper a non-destructive approach, combining gamma spectroscopy and Monte Carlo simulations has been examined in detail. Here we present the case study for a number of typical concrete blocks to be characterized. The limits and uncertainties of the proposed method are quantified for the activity estimates in the case of ^{137}Cs as a tracer element.

1. Introduction

A non-destructive approach, combining gamma ray spectroscopy and Monte Carlo simulations is examined in detail in order to characterize massive concrete blocks containing some radioactive waste, typically of low or very low activity. The major goal of this study is to quantify the limits and uncertainties of the proposed method for the activity estimates in the case of ^{137}Cs as a tracer element.

Fig. 1 presents a cut of a typical concrete block to be examined. Although the thickness of concrete walls can vary, in all cases the waste materials are placed in a metallic cylindrical barrel of ~200 liters. Concrete density is typically of 2.2 g/cm^3, while waste density is not known precisely and can vary in the range of 0.5 - 1.6 g/cm^3.

2. Calibration measurements

2.1. Efficiency of the Ge detector

From the HP Ge detector total efficiency e_{tot}, obtained from the measurements using the ^{152}Eu source, an absolute (intrinsic) detector efficiency e_{Ge} can be calculated taking into account the solid angle correction. If the source is taken as a point like and detector radius *r* is much smaller than the distance between source and detector *l*, then

$$e_{Ge} = e_{tot} / W = e_{tot} / (p\, r^2/l^2 * 1/4p). \qquad (1)$$

The results of our investigations are given in Fig. 2. The efficiency values for *l = 70-106 cm* have been averaged in order to fit the dependence with an empiric function for Ge detectors, namely $e = a\, E_g^{\,b}$ *(a=0.121* and *b= -0.780).* The gamma peaks at *121 keV* have been excluded from the fit due to the different response function of the detector at low energies.

Figure 1. A cross view (cut) of a typical waste container with radioactive materials.

2.2. Measurements with waste barrel

As it was mentioned above, the density of the waste inside the waste barrel can typically vary from 0.5 to 1.6 g/cm^3. For this reason a number of measurements with known source activity of ^{152}Eu and variable material density have been performed. The following densities of the waste have been considered: *0.0, 0.15, 0.70* and *1.20 g/cm^3*. An experimental scheme is presented in detail in Fig. 3. The Ge detector had the following geometrical characteristics: 60.3 mm long and 52.2mm diameter crystal.

As long as simulations are concerned, the geometry of the waste container and detector was modeled exactly as presented above with the MCNP (Monte Carlo N-Particle code) [1]. A number of experiments performed on activity measurements of waste barrels can serve to test our prediction power of the modeling. For comparison, we considered the case when the Ge detector is placed at *106 cm* from the central axis of the cylindrical waste container with variable density of *0.00 (only a gamma source and no container), 0.15, 0.70* and *1.20 g/cm^3* correspondingly. A measured counting rate in each gamma peak in *(#/s)* can be expressed by the following formula:

$$N^{meas}_{\gamma} = A_{source} * \varepsilon_{meas}. \qquad (2)$$

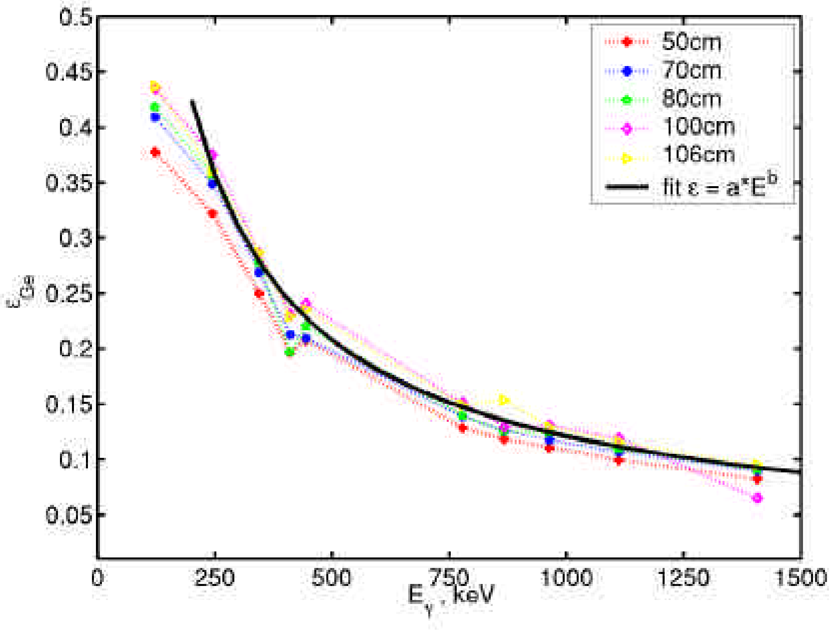

Figure 2. An absolute HP Ge detector efficiency calibrated with the ^{152}Eu source.

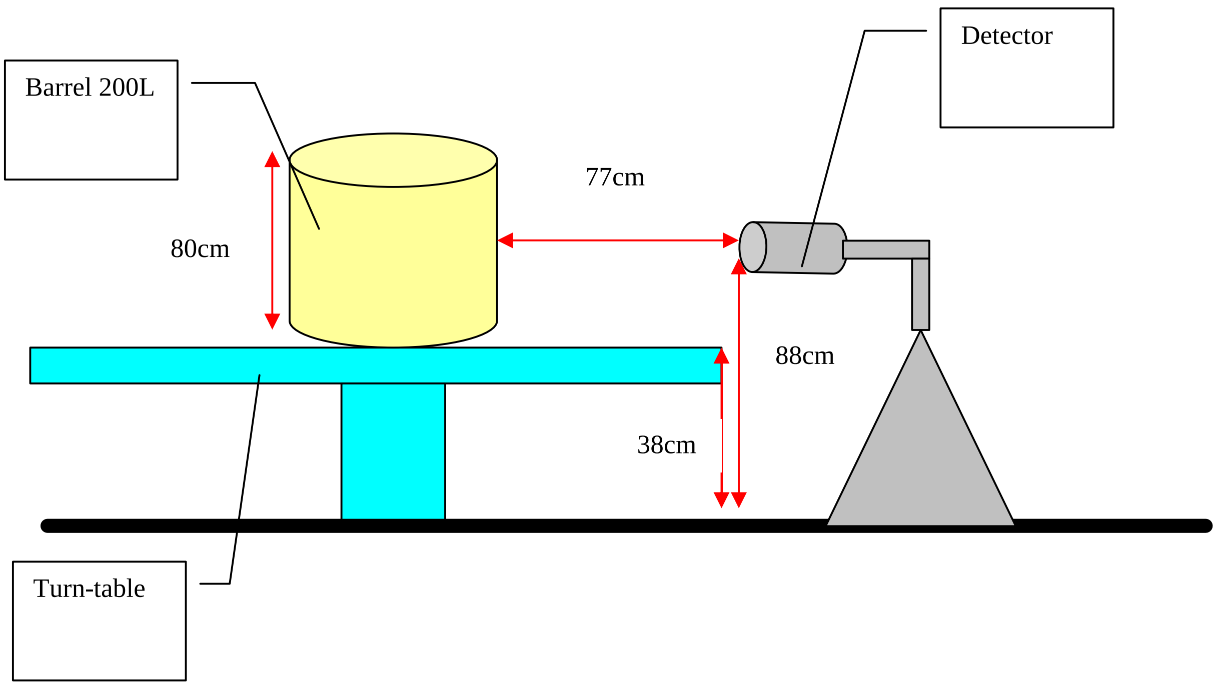

Figure 3. An experimental scheme of test measurements with a known source activity and variable waste density.

Here A_{source} is a source activity in *(Bq)*, and e_{meas} is the total detection efficiency including a solid angle, attenuation, Ge intrinsic efficiency and gamma peak intensity.

Similarly, a calculated counting rate in each gamma peak in *(#/s)* can be expressed by the following formula:

$$N^{calc}_{\gamma} = A_{source} * \varepsilon_{calc} * \varepsilon_{Ge} * S_{Ge}. \qquad (3)$$

Here e_{calc} = $M_\gamma * \phi_\gamma$ with ϕ_γ as a calculated detection efficiency in *(#/(cm^2 * s))*, including a solid angle, attenuation, and gamma peak intensity. ϕ_γ is proportional to the gamma flux (per one source gamma and per *cm^2*) crossing an active surface S_{Ge} of the Ge detector. M_γ is gamma-ray multiplicity per one Bq, e_{Ge} an intrinsic Ge detector efficiency.

The calculated detection efficiency ϕ_γ is plotted in Fig. 4 as a function of the source gamma energies and different densities of the waste container. Gamma source was taken as a volumetric source uniformly distributed over entire volume of the container. It is clearly seen that energetic photons may be attenuated by a factor of 5 (0 density versus 1.2 g/cm^3), while for low energy photons this attenuation may reach nearly one order of magnitude. See ~1408 and ~122keV rays correspondingly in the same Fig. 4.

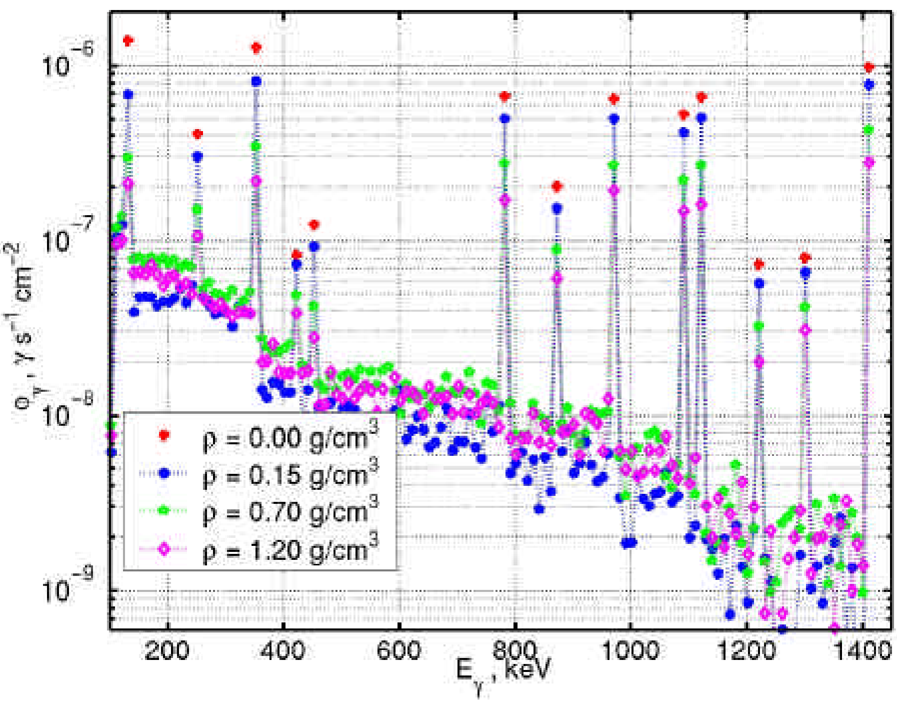

Figure 4. Photon flux at the detector position as a function of different densities of the waste container with the ^{152}Eu source.

Below we intend to compare N^{calc}_g and N^{meas}_g (see formulas 2 and 3 above) for a number of gamma rays. Note that both N^{calc}_g and N^{meas}_g depend on the same A_{source}. Therefore, arbitrarily we took it equal to *1.79e+7 Bq* for *^{152}Eu* in both cases. Tables 1 to 4 present our direct comparison of the results.

E_γ (keV)	N_{meas} (#/s)	N_{calc} (#/s)	N_{meas} / N_{calc}
121.6	1186.8	1036.7	1.14
344.1	780.4	688.7	1.13
778.0	404.5	376.0	1.08
1112.0	313.3	286.7	1.09
1408.0	256.0	236.7	1.08

Table 1. r=0.00 g/cm^3, i.e. no waste container present in this configuration.

E_γ (keV)	N_{meas} (#/s)	N_{calc} (#/s)	N_{meas} / N_{calc}
121.6	737.5	489.0	1.51
344.1	619.3	463.7	1.34
778.0	343.7	317.7	1.08
1112.0	279.2	239.6	1.17
1408.0	247.0	195.9	1.26

Table 2. The waste container with r=0.15 g/cm^3 ("coton/plastique").

E_γ (keV)	N_{meas} (#/s)	N_{calc} (#/s)	N_{meas} / N_{calc}
121.6	100.1	140.8	0.71
344.1	138.7	176.3	0.78
778.0	113.7	171.8	0.66
1112.0	106.3	124.9	0.85
1408.0	99.0	107.6	0.92

Table 3. The waste container with r=0.70 g/cm^3 ("cecacite").

E_γ (keV)	N_{meas} (#/s)	N_{calc} (#/s)	N_{meas} / N_{calc}
121.6	64.3	93.3	0.69
344.1	78.0	96.2	0.81
778.0	61.4	110.2	0.56
1112.0	58.4	68.5	0.85
1408.0	55.7	69.6	0.80

Table 4. The waste container with r=1.2 g/cm^3 ("sable").

In brief, an agreement between calculations and measurements is acceptable. The biggest uncertainty in the simulations is that one does not know an exact distribution/position of a gamma source in a non-negligible volume of the waste container. Our discussion below gives a more quantitative estimate on the dependence of the results on this uncertainty.

3. Prediction uncertainties

As it was mentioned above, one of the prediction uncertainties, what can change the final results by a factor of 2 or bigger, is that one will not know precisely the average density inside the waste barrel (see Fig. 4).

Another uncertainty is related to the assumptions on modeling of a radioactive source within ~200 liters of a waste barrel. Below we will try to quantify this problem. The ^{152}Eu gamma source was modeled, and 13 gamma rays, i.e. all with intensities >1%, were considered. Gamma source was distributed homogeneously all over an active volume of the container. Photons were emitted isotropically from the entire active volume. A number of different (from the geometrical point of view) source specifications were tried (see Fig. 5).

The waste container density of *1.2 g/cm^3* was chosen to maximize possible variations due to different source definitions as above. This dependence on the geometrical source specification is presented in Fig. 6. It is seen that the calculations vary strongly with this particular parameter. For the highest energy photons this difference can change by a factor of 2, while for lower energies it can increase to a factor of 3 or higher. If higher material densities were used, this difference could be even higher.

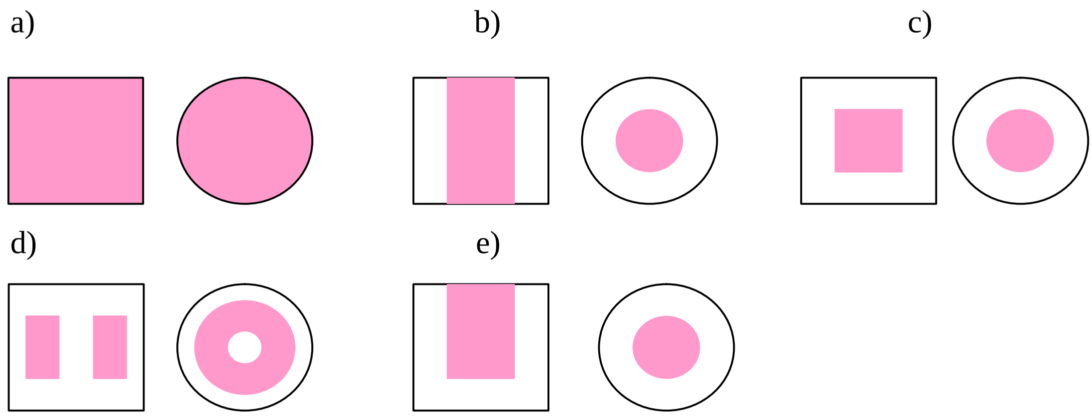

Figure 5. Different gamma source specifications-distributions within the 200 l cylindrical waste barrel.

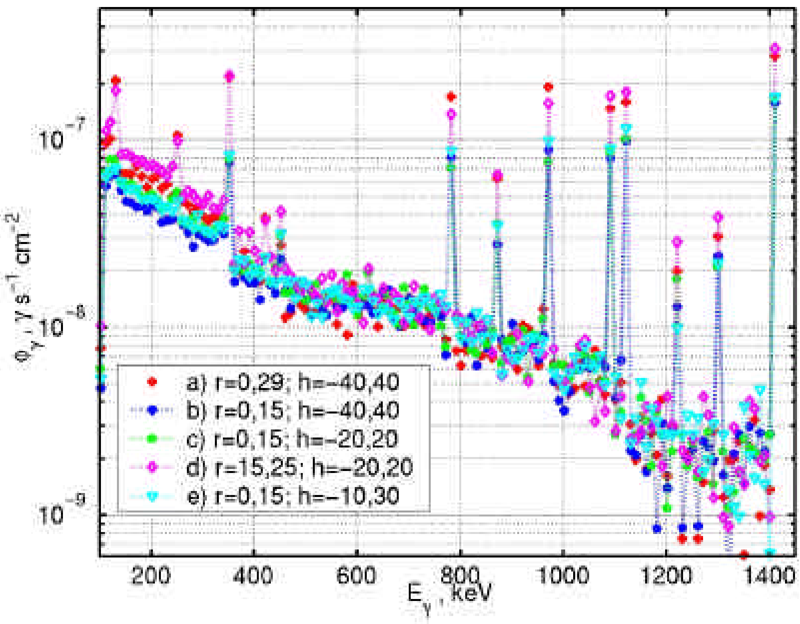

Figure 6. Photon flux at the detector position as a function of different source specifications (different geometrical distributions of the ^{152}Eu source); r and h stands for the radial and height intervals (cm) correspondingly.

4. General discussion and preliminary conclusions

From equations (2) and (3) follows that, if $N^{calc}_{g} \gg N^{meas}_{g}$, then

$$A_{source} = N^{meas}_{\gamma}/(\varepsilon_{calc} * \varepsilon_{Ge} * S_{Ge}). \qquad (4)$$

This formula, in principle, can be used to define the radioactivity of a source placed in any geometrical configuration for the measured gamma peak intensity N^{meas}_{g} . It is important to note that e_{calc} is strongly dependent on the modeling of the source distribution and density of the waste barrel (see discussion in the previous section). To minimize this effect, in any measurement of N^{meas}_{g} the experiment should be performed by turning the waste container around its symmetry axis and averaging the detector counting rate over all geometrical configurations. In this way, we believe, that the final A_{source} value could be determined with a precision not worse than a factor of 2.

5. Test cases

In the following section we will test our method for predicting the ^{137}Cs activity in realistic geometry configurations. As it was explained above, in order to determine the activity one needs N^{meas}_{γ} in (#/s) in the peak of ^{137}Cs.

A series of measurements with a few massive blocks (type T.E.N.) were performed before and after destruction. Before the destruction all measurements were done over 4 sides of the block at 50 cm. It is important to note that an average counting rate in the peak for different blocks (or for different sides of the same block) could vary by 1-2 orders of magnitude for comparable activity. This suggests that neither the distribution of radioactive material is homogeneous nor the density of it is unique.

In the case of simulations, the parameters used are as follows: detector efficiency ε_{Ge} (661keV) = 0.167, detector active surface S_{Ge} = 21.4 cm^2, $\varepsilon_{calc} = M_{\gamma} * \phi_{\gamma} = 0.851 * \phi_{\gamma}$. We modeled the experimental conditions as precise as possible resulting in ϕ_{γ} predictions both before and after the destruction of waste containers. These results together with corresponding measurements let us predict the ^{137}C source activities for different blocks (see formula 4).

Block ID	$A^{Cs}_{meas,}$ Bq (destructive method)	$A^{Cs}_{calc} / A^{Cs}_{meas}$ (destructive method)	$A^{Cs}_{calc} / A^{Cs}_{meas}$ (non-destructive method)
TEN 10	6.54e+7	0.66	0.57
TEN 230	1.21e+7	1.38	0.49
TEN 430	1.56e+8	0.76	1.01
TEN 444	1.29e+8	1.05	2.02

Table 5. Comparison of the measured A^{Cs}_{meas} *and calculated* A^{Cs}_{calc} *activities of waste containers in the case of destructive and non-destructive approaches.*

Table 5 presents our major results. In brief, the predicted values, based on both destructive and non-destructive measurements, are consistent with the one, extracted from the experiment, within the uncertainties discussed in the previous section. In other words, it seems that by performing full-scale Monte Carlo calculations one can estimate the source activity based on the measured counting rate in particular gamma peaks. The uncertainty of this estimation should not be bigger than a factor of 2. If this level of uncertainty is satisfying, one can characterize the waste containers in a non-destructive way as it was shown in this work.

6. Conclusions

A non-destructive approach, combining gamma ray spectroscopy and Monte Carlo simulations have been examined in detail in order to characterize massive concrete blocks containing some radioactive waste, typically of low or very low activity. Our method was applied to estimate the waste activity due to ^{137}Cs (being a tracer element) and to compare it with existing experimental data. We conclude that, by performing full-scale Monte Carlo calculations and by measuring the outside counting rate of particular gamma peaks, one can estimate the source activity inside the concrete container without destroying its shielding structure. The uncertainty of this estimation should not be bigger than a factor of 2. If this level of error is satisfying, one can easily characterize the waste containers in a non-destructive way for some tracer elements as ^{134}Cs, ^{137}Cs, ^{60}Co, etc. In addition, the method uncertainties could be decreased if one knew better the distribution of radioactive material inside the waste barrel and also if the waste density were known precisely.

Finally we note that the above study, in the case of waste containers present at INB 48 of CEA Saclay, let one decrease the characterization and corresponding storage costs from ~6.0M Euros to ~1.6 M Euros (when compared to a destructive method), i.e. for about 500 waste containers in total [2].

References

[1] J.F. Briesmeister, “MCNP - A General Monte Carlo N-Particle Code”, Technical Report LA-12625-M, LANL (1997).

[2] F. Damoy, S. Feray, “INB 48: Bilan des Expertises Complementaires des Blocs T.E. Contenant des Dechets F.A./T.F.A.”, report DAPNIA/SDA/D/03.124/NT, CEA Saclay, France (January 2003) (in French) ; F. Damoy, private communication.