

On the applicability of LaBr₃ detectors in the non-destructive characterization of radioactive waste drums

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Abstract The applicability of LaBr₃-detectors for the non destructive characterization of radioactive waste drums is evaluated by comparing their performance with those of HPGe-detectors. It is shown, that relevant easy-to-measure key-nuclides are identified as well as quantified with a LaBr₃-detector even in complex spectra at nearly the same level of precision and reliability than with HPGe-detectors.

Keywords LaBr₃ · HPGe · Radioactive waste characterization · TRISO particle

Introduction

To verify the compliance of radioactive waste packages with national regulations for transportation, intermediate storage or final disposal, assays to characterize their radionuclide inventory are required [1]. The most established non-destructive assay for the radiological characterization of radioactive waste drums is segmented γ -scanning (SGS) [2, 3]. SGS is based on γ -spectroscopic measurements at predefined sectors of a waste drum with a collimated detection system. The identification and quantification of radionuclides is based on the separation of their characteristic γ -peaks in measured spectra. Thus, γ -ray detectors used for SGS must exhibit a sufficient energy resolution to identify key-nuclides expected in radioactive wastes. Currently, SGS systems are equipped with high purity germanium (HPGe) semiconductor detectors

offering the best commercially available energy resolution ranging between 0.19 and 0.30 % at 661.7 keV [4]. Competing thallium doped sodium iodide (NaI(Tl)) scintillation detectors exhibit a significantly lower energy resolution of about 7 % at 661.7 keV [5]. However, they offer considerably higher detection efficiencies and better timing properties than HPGe-detectors. In recent years cerium doped lanthanum bromide (LaBr₃(Ce)) scintillation detectors are provided by the commercial market combining all advantages of scintillation-detectors with a significantly improved energy resolution of about 3 % at 661.7 keV [5]. Hence, a replacement of HPGe-detectors by LaBr₃-detectors yields different potentials for improvement of measurement setups in various fields. First, higher detection efficiencies and a reduction of dead time due to better timing properties offers a potential towards a reduction of measurement time. Second, the avoidance of a cooling system, either electrical or using liquid nitrogen, as it is required for HPGe-detectors economizes maintenance costs as well as reduces the size and weight of the detection system which allows the construction of mobile detection systems. The suitability of LaBr₃-detectors has already been successfully proofed for nuclear physics experiments [6, 7], medical imaging [8, 9] or in industrial applications [10–12]. In order to demonstrate the usability of LaBr₃-detectors for the radiological characterization of radioactive waste drums by SGS, it has to be shown, that γ -emitting easy-to-measure (ETM) key-nuclides can be quantified. The entire radionuclide inventory of a waste drum can be determined by scaling factor methods relying solely on the detection of a few ETM key-nuclides [13]. The most relevant ETM key-nuclides in radioactive wastes from nuclear power plants are the activation product ⁶⁰Co and the fission product ¹³⁷Cs. Thus, the performance of LaBr₃-detectors and HPGe-detectors is compared in terms

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of the detection of these ETM key-nuclides by investigating detection efficiencies, energy resolutions, detection limits, timing properties and intrinsic backgrounds. Finally, the performance of both detectors is investigated with an irradiated UO₂ TRISO (tristructural isotopic) coated particle [14].

Detector setups

The experimental comparison of HPGe- and LaBr₃-detectors is performed using coaxial detector-crystal geometry of same volume (2×2 in.). Although both detector-crystals have same external dimensions, the LaBr₃-crystal has a slightly higher active volume compared to the HPGe-crystal because of the hollow space in the HPGe-crystal, which is necessary for crystal cooling. The detectors are operated in lead chambers as low level counting systems. The wall thickness of the chamber is 10 cm for the HPGe-detector and 5 cm for the LaBr₃-detector. The HPGe-detector (p-type, DSG Detector system GmbH) has a relative efficiency of 20 %. It is cooled by liquid nitrogen and operated with a high voltage of 2 kV. The signals of the detector amplifier (resistive feedback) are processed via a spectroscopy amplifier (C.A.E.N Model N968) and a 1,000 MHz Wilkinson analog-to-digital-converter (FAST ComTec Model 7070) by high performance multi-channel-analyzer (Flir Radiation GmbH webMATE) interfaced to a PC via USB. The shaping time of the spectroscopy amplifier is set to 6 μ s. The LaBr₃-detector consists of a BrillianceCe 380 crystal (Saint-Gobain Crystals) doped with 5 % Cerium activator impurities coupled to a specially selected photomultiplier tube (PMT XP5500). The signals of the detector are processed by a universal multi-channel-analyzer (Flir Radiation GmbH scintiSPEC) containing an integrated 60 MHz Wilkinson analog-to-digital-converter interfaced to a PC via USB. The voltage of the PMT is set to 639 V. For both detectors the γ -ray spectra are recorded with the acquisition software Win TMCA32 (Flir Radiation GmbH). Analysis of the spectra is performed with the spectroscopy software Gamma-W (Westmeier GmbH).

Detection efficiency and energy resolution

The detection efficiency as well as the energy resolution of both detectors is investigated. Reference point sources of ⁶⁰Co (103.0 ± 1.1 kBq), ¹³⁷Cs (56.7 ± 0.6 kBq), ¹⁵²Eu (39.0 ± 0.6 kBq) and ²⁴¹Am (20.1 ± 0.3 kBq) are placed arbitrary 14.3 cm in front of the detector surface and are measured for 2 h each. The results of this study are summarized in Fig. 1. For γ -energies lower than 500 keV the

HPGe-detectors exhibit detection efficiencies lower by a factor of 2.5 compared to the LaBr₃-detector. Up to γ -energies of more than 1 MeV this discrepancy increases to a factor of around 3.0. This effect is mainly due to different effective atomic numbers of Ge ($Z = 32$) and of LaBr₃ ($Z = 47$) resulting into far higher stopping powers of γ -rays and thus far higher detection efficiencies for LaBr₃ than for HPGe. The energy resolution is given in form of FWHM per incident γ -energy. The energy resolution of the HPGe-detector is a factor 5.5 higher compared to the energy resolution of the LaBr₃-detector at 123.1 keV increasing to a factor of about 9.5 at 1,596.5 keV. The energy resolution is mainly driven by the number of electron hole pairs generated by an incident photon. Since the bandwidth of LaBr₃ (5 eV) is far higher than for Ge (0.75 eV), less electron-hole-pairs are generated by an incident photon in LaBr₃ which results into the lower energy resolutions of the LaBr₃-detector.

Background and detection limits

Besides background radiation originating from the surrounding materials, a LaBr₃-detector is affected by intrinsic background radiation arising from the presence of the natural occurring isotopes ¹³⁸La ($T_{1/2} = 1.05 \cdot 10^{11}$ a) and ²²⁷Ac ($T_{1/2} = 21.733$ a) within the crystal itself [11]. These radioactive impurities produce a significant intrinsic background for measurements with LaBr₃-detectors. In order to qualify the suitability of LaBr₃-detectors for the characterization of radioactive waste drums containing low isotope specific activities, the detection limits for relevant ETM key-nuclides have to be investigated.

Background spectra measured for 72 h are shown in Fig. 2 for both detectors operated in the lead chambers. The background spectrum measured with the LaBr₃-detector shows a first peak at 32 keV. This peak is caused by two characteristic X-rays of ¹³⁸La at 31.8 keV ($p_{Ex} = 11$ %) and 32.2 keV ($p_{Ex} = 20.4$ %). This peak is followed by two continua. The first continuum up to 250 keV is generated by the β -decay of ¹³⁸La. The second continuum up to 750 keV is the Compton-continuum of the two characteristic γ -rays from the decay of ¹³⁸La. These characteristic γ -rays are at 788.7 keV ($p_{E\gamma} = 34$ %) and at 1,435.8 keV ($p_{E\gamma} = 66$ %). The characteristic γ -peak at 788.7 keV is measured in random coincidence with the β -decay of the ¹³⁸La. Therefore, the peak at 788.7 keV is smeared to a continuum up to 1,000 keV. At around 1,460 keV a triple-peak arises. The first peak is the characteristic 1,435.8 keV γ -ray of ¹³⁸La. This γ -ray is measured in random coincidence with the X-rays of ¹³⁸La leading to the second peak at around 1,468 keV. The third peak at 1,460.8 keV originates from ⁴⁰K, which is present in the environment. Above 1,500 keV

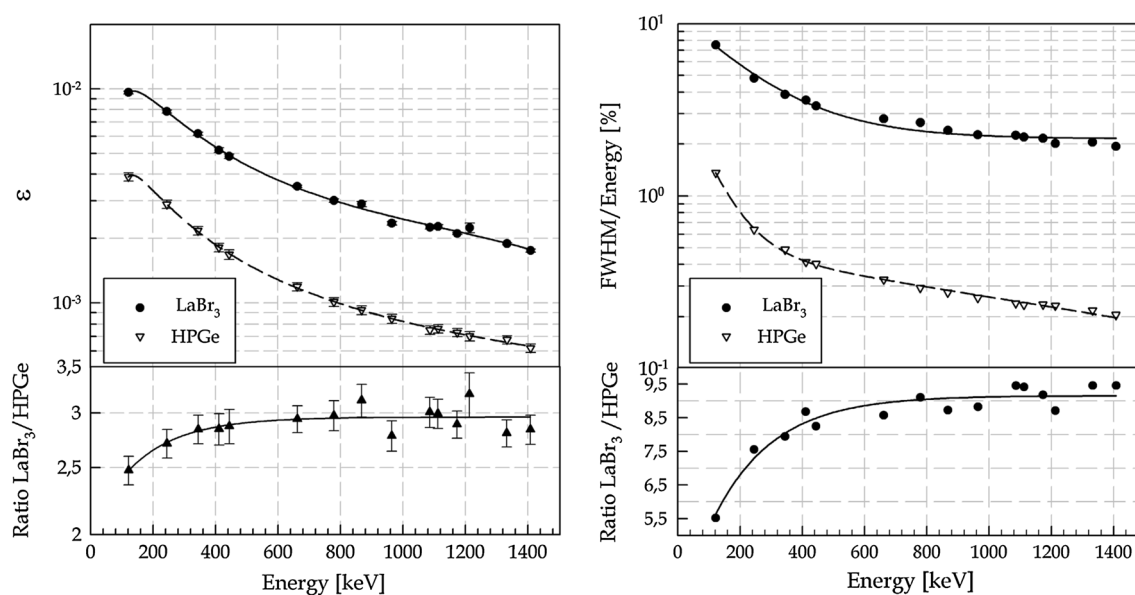


Fig. 1 Detection efficiency (ε) and energy resolution (FWHM/energy) of the HPGe- and LaBr_3 -detector as a function of the γ -energy. The ratios are display in the lower part of each plot

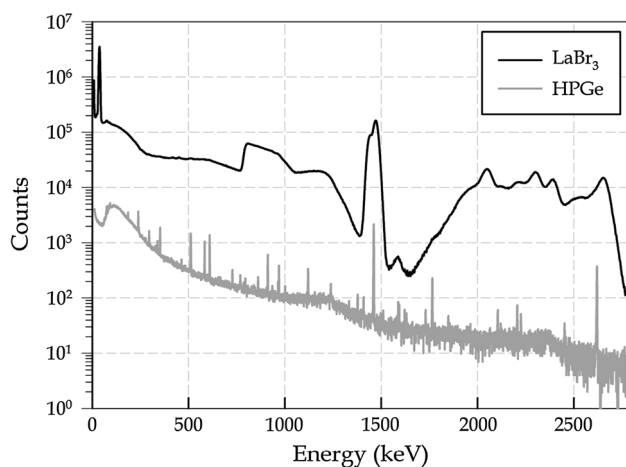


Fig. 2 Background spectra recorded during 72 h with the LaBr_3 - and the HPGe-detector

the background spectrum is dominated by α -radiation produced by daughter nuclides in the decay chain of ^{227}Ac . The background measured with the HPGe-detector is rather low in comparison to the LaBr_3 -background. The peaks in the background spectrum arise from radioactive isotopes like ^{40}K , ^{208}Tl , ^{212}Pb , ^{212}Bi and ^{228}Ac which naturally occur in the environment. Apart from ^{40}K none of these radioactive isotopes can be identified in the background spectrum of the LaBr_3 -detector.

For the calculation of detection limits, it is assumed, that a peak can be identified if the number of registered counts associated to the γ -ray exceeds the background $B_{E\gamma}$ at the investigated region around the γ -energy by three standard-

deviations. Thus, the minimum detection activity LD for the given counting geometry can be calculated by.

$$LD = \frac{3 \cdot \sqrt{B_{E\gamma}}}{\varepsilon_{E\gamma} \cdot p_{E\gamma} \cdot t_m} \quad (1)$$

where $\varepsilon_{E\gamma}$ and $p_{E\gamma}$ are the detection efficiency and the intensity of the γ -ray respectively and t_m the counting time. The detection limits for the relevant ETM key-nuclides ^{60}Co , ^{137}Cs and for ^{152}Eu are determined for the measured background spectra of both detectors. In a second measurement set, spectra of ^{60}Co , ^{137}Cs and ^{152}Eu point sources are recorded for 4 h each with a detector to source distance of 14.3 cm for both detectors. For each of these spectra the detection limits of the other two nuclides are determined. The achieved detection limits are listed in Table 1. The LD values based on background measurements are below 10 Bq for both detectors. Nevertheless, the detection limits for the HPGe-detector are lower by factor 4–5 compared to those for the LaBr_3 -detector. For the detection limits evaluated from the spectra of the point sources this factor is around 2. In all cases the detection limits for the given counting geometry are less than 200 Bq. It has to be noted, that the absolute value of detection limits depends on sample properties as well as on parameters of the measurement. Hence, it is not possible to draw any conclusion on the absolute detection limits of both detectors for isotope specific activities in waste drums based on this data. But it is shown that the detection limits in presence of other sources calculated for both detectors are of the same order of magnitude. Therefore we can assume that the quantification of ETM key-nuclides may be

Table 1 Detection limits LD estimated for ETM key-nuclides in different spectra recorded with the LaBr₃- and the HPGe-detector in the lead chambers

Spectrum	Counting time (h)	LD ⁶⁰ Co (Bq)		LD ¹³⁷ Cs (Bq)		LD ¹⁵² Eu (Bq)	
		LaBr ₃	HPGe	LaBr ₃	HPGe	LaBr ₃	HPGe
Background	72	4	1	9	2	5	1
⁶⁰ Co	4	–	–	172	109	83	60
¹³⁷ Cs	4	28	12	–	–	43	29
¹⁵² Eu	4	15	3	119	77	–	–

The detection limit for ⁶⁰Co is based on the 1,333.5 keV γ -ray, for ¹³⁷Cs on the 661.7 keV γ -ray and for ¹⁵²Eu on the 344.3 keV γ -ray. The source to detector distance is 14.3 cm

achieved with LaBr₃-detectors at the same level of reliability offered by HPGe-detectors.

Dead time

In routine characterization of radioactive waste drums with a priori unknown source activities and different count rates versatile detectors are required. Especially, for measurements of waste drums with radionuclide inventories of high activities, the detector dead time may significantly influence the measurement. Thus, the timing properties of both detectors are investigated by measurements of a ¹³⁷Cs point source with an activity of 306 kBq. The detector to source distance is varied in 0.5 cm steps from 5 to 40.5 cm and a spectrum is measured at each position for 300 s (real time). The detector dead time is calculated as the difference between the real time and the live time divided by the real time. Low detector to source distances corresponds to a

very high level of count rates. As shown in Fig. 3 the HPGe-detector dead time reaches up to nearly 80 % for low detector to source distances whereas the LaBr₃-detector dead time is around 25 %. As expected the dead time decreases for increasing detector to source distances and reaches almost 0 % for both detectors at a detector to source distance of 40 cm.

Precision and reliability at different live times

In previous parts of this work all spectra are recorded during comparably long measurement times. However, the routine characterization of radioactive waste drums by SGS requires short measuring times. To study the performance of both detectors at different measurement times, spectra of a ¹⁵²Eu point-source (39.0 ± 0.6 kBq) are recorded with both detectors for measurement times between 15 s and 2 h. The distance between the source and the detector is 14.3 cm for all measurements. Figure 4a shows the deviations of the measured to the reference activity for different live times based on the analysis of the second most intense γ -ray of ¹⁵²Eu at 344 keV. It can be seen, that all determined activities with both detectors are within one standard-deviation in good agreement with the reference activity. The deviations are between -3 and 2 %. With increasing live time, uncertainties become smaller due to the increased counting statistics. It can be noted, that the uncertainties estimated from the measurement with the LaBr₃-detector are slightly smaller than that estimated from the measurements with the HPGe-detector. This effect is expected, since the LaBr₃-detector has a higher efficiency than the HPGe-detector, as shown previously. The higher efficiency leads to a higher level of counting statistics and thus to a more reliable activity evaluation. However, the poor energy resolution and the complexity of the LaBr₃-spectrum leads to worse uncertainties in the peak fitting compared to the HPGe-spectrum. In a second approach, the activity of the ¹⁵²Eu point-source is estimated using all identified γ -rays with intensities $p(E_\gamma)$

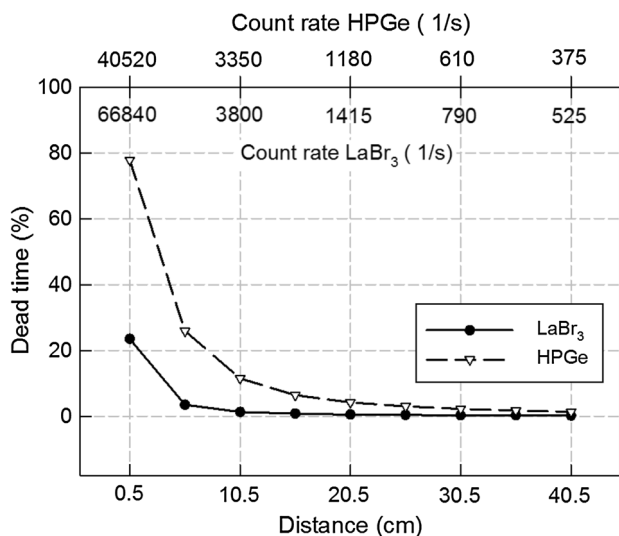


Fig. 3 Dead time of the LaBr₃- and the HPGe-detector as a function of the detector to source (306 kBq ¹³⁷Cs) distance. The counting (real) time is 300 s

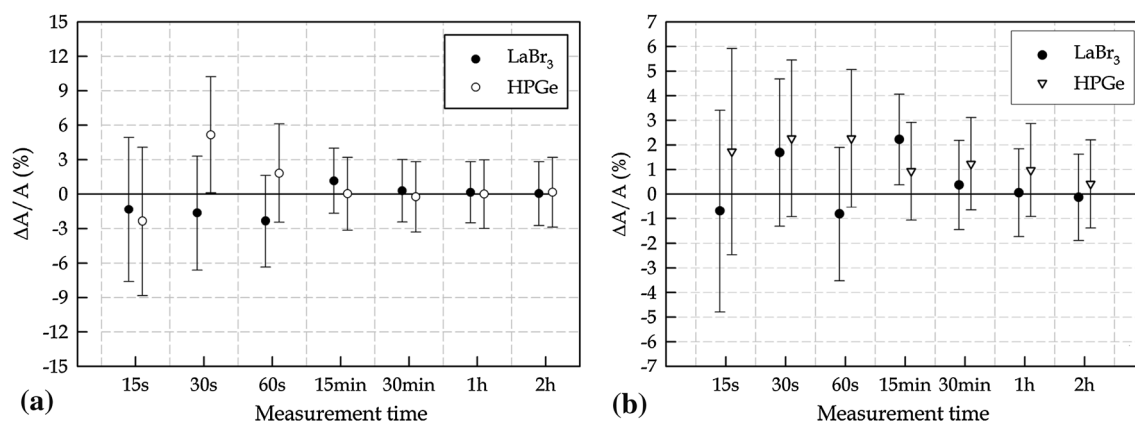


Fig. 4 Deviations of the calculated activity from the reference activity for the measured ^{152}Eu -point source. **a** Based on the analysis of the 344.3 keV γ -ray; **b** based on the analysis of all identified γ -rays having intensities higher than 1 %

greater than 1 %. The activity is determined by weighted mean of the activities estimated for each characteristic γ -ray. For the 15 s measurement 11 γ -rays are considered with the HPGe-detector whereas only 6 with the LaBr₃-detector. For the 2 h measurement the number of identified γ -rays increases to 14 and 10, respectively. The results are shown in Fig. 4b. At a first glance the determined activities are slightly more reliable in a range between -1 and 2.5 % in comparison to the results obtained from the analysis of the γ -ray at 344 keV. On average, uncertainties are reduced by a factor of 1.5–2. Thus, LaBr₃-detectors can be used in SGS to determine the activity of ETM key-nuclides with similar precision and reliability than provided by HPGe-detectors.

TRISO coated particle spectra

In contrast to the measurements performed in the previous sections, the radionuclide inventory of a waste drum contains usually more than one radioactive isotope. The applicability of LaBr₃-detectors is strongly relying on the reliable identification of relevant ETM key-nuclides. To investigate the performance of both detectors in terms of complex spectra, the spectrum generated by an irradiated UO₂ TRISO coated particle containing fission products and minor actinides is investigated as an example for real radioactive waste. The UO₂ TRISO coated particle contains originally 0.60476 mg uranium with an enrichment of 16.74 % ^{235}U and has a diameter of 919 μm . It consists of a spherical UO₂ kernel ($\varnothing = 502.2$ μm , $\rho = 10.86$ g/cm³) and coating layers of porous pyrolytic carbon ($d = 92.3$ μm , $\rho = 1.012$ g/cm³), inner dense pyrolytic carbon ($d = 40.6$ μm , $\rho = 1.87$ g/cm³), silicon carbide ($d = 35.9$ μm , $\rho = 3.20$ g/cm³) and outer dense pyrolytic carbon ($d = 35.9$ μm , $\rho = 1.87$ g/cm³) [14].

The particle is measured during 24 h at a detector to source distance of 60 cm. The dead time of the HPGe-detector is 13.7 %, whereas the dead time of the LaBr₃-detector is only 1.7 %. The spectra recorded with the two detectors are shown in Fig. 5. Both spectra give a good overview on the previously discussed properties of the detectors. Due to the higher efficiency, the count rate of the LaBr₃-detector exceeds that of the HPGe-detector. However, the energy resolution of the LaBr₃-detector is not sufficient to resolve all peaks. For example in the zoom of Fig. 5 the left peak in the LaBr₃-spectrum is the sum peak of the γ -rays of ^{154}Eu at 996.3 keV and 1,004.7 keV. Hence the intensities of the two γ -rays are here simply summed for evaluation of the ^{154}Eu activity. The right peak is a sum peak of the γ -rays of ^{134}Cs at 1,038.6 keV and ^{106}Rh at 1,050.4 keV. In this case the activity of ^{106}Rh may be only evaluated after knowing the ^{134}Cs activity. Using the HPGe-detector 9 radionuclides (^{106}Rh , ^{122}Sb , ^{134}Cs , ^{137}Cs , ^{144}Ce , ^{144}Pr , ^{154}Eu ,

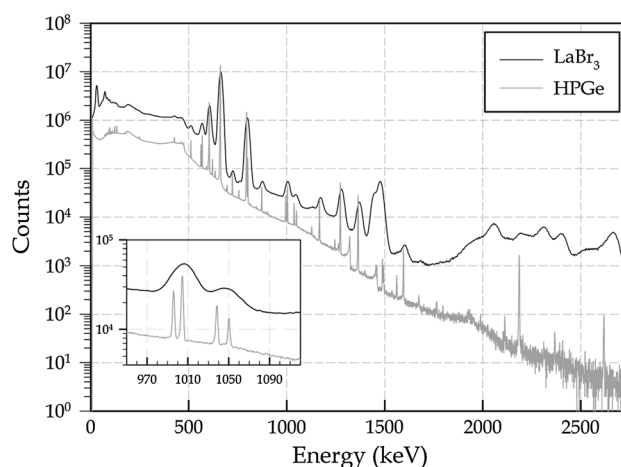


Fig. 5 Spectra of the irradiated TRISO particle measured for 24 h with the LaBr₃- and with the HPGe-detector

^{155}Eu , ^{241}Am) are identified whereas only 4 (^{122}Sb , ^{134}Cs , ^{137}Cs , ^{154}Eu) with the LaBr_3 -detector. The corresponding detected gamma-rays are listed in Table 2. For activity determination the reference point sources ^{60}Co , ^{137}Cs , ^{152}Eu and ^{241}Am mentioned in the first part of this work are used

to determine the detection efficiency of both detectors. The correction factors for gamma-ray self-absorption are given in Table 2. They are calculated for spherical geometry by means of the Eq. (17) given in [15] using an apparent density of 3.36 g/cm^3 for the particle. The mass attenuation

Table 2 Radionuclides and corresponding gamma-rays identified in the spectra of the TRISO coated particle measured with the HPGe- and LaBr_3 -detectors, measured activities and calculated activities with the OCTOPUS code [17]

Isotope	E_γ (keV)	Shielding factor	HPGe-detector A (MBq)	LaBr_3 -detector A (MBq)	OCTOPUS code A (MBq)
^{106}Rh	621.9	0.9876	0.318 ± 0.014		
(^{106}Ru)	1,050.4	0.9922	0.299 ± 0.017	0.319 ± 0.024^f	0.603
			0.308 ± 0.013^e		
^{125}Sb	176.3	0.9046	0.079 ± 0.004		
	380.5	0.9770	0.099 ± 0.018		
	427.9	0.9806	0.081 ± 0.003	0.081 ± 0.004	
	463.4	0.9826	0.082 ± 0.004		
	636.6	0.9878	0.082 ± 0.003		
			0.085 ± 0.008^e		0.239
^{134}Cs	475.7	0.9831	0.845 ± 0.072		
	563.2 ^a	0.9862	0.827 ± 0.037	0.652 ± 0.030	
	569.3 ^a	0.9863	0.845 ± 0.037		
	604.7	0.9872	0.835 ± 0.033	0.777 ± 0.033	
	795.9 ^b	0.9902	0.855 ± 0.034	0.884 ± 0.033	
	801.9 ^b	0.9902	0.839 ± 0.035		
	1,038.6	0.9921	0.828 ± 0.042	0.851 ± 0.054	
	1,167.9	0.9928	0.813 ± 0.033	0.850 ± 0.034	
	1,365.2	0.9935	0.850 ± 0.031	0.742 ± 0.025	
			0.837 ± 0.013^e	0.793 ± 0.087^e	0.772
^{137}Cs	661.7	0.9883	6.569 ± 0.261	6.478 ± 0.253	6.178
^{144}Ce	80.1	0.8308	0.399 ± 0.048		
	133.5	0.8300	0.353 ± 0.021		
			0.376 ± 0.032^e		0.346
^{144}Pr	696.5	0.9889	0.371 ± 0.017		
	1,489.16	0.9938	0.447 ± 0.030		
			0.409 ± 0.054^e		0.346
^{154}Eu	123.1	0.7994	0.094 ± 0.006		
	247.9	0.9521	0.116 ± 0.006		
	723.3	0.9893	0.122 ± 0.005	0.084 ± 0.004	
	756.8	0.9897	0.114 ± 0.005		
	873.2 ^c	0.9909	0.131 ± 0.006	0.130 ± 0.005	
	892.8 ^c	0.9911	0.136 ± 0.017		
	904.1	0.9912	0.107 ± 0.008		
	996.3 ^d	0.9918	0.114 ± 0.005	0.133 ± 0.005	
	1,004.7 ^d	0.9919	0.118 ± 0.005		
	1,128.6	0.9926	0.118 ± 0.009		
	1,246.2	0.9931	0.109 ± 0.006		
	1,274.4	0.9932	0.118 ± 0.004	0.123 ± 0.004	
	1,596.5	0.9940	0.131 ± 0.006	0.091 ± 0.004	
			0.117 ± 0.011^e	0.112 ± 0.023^e	0.189
^{155}Eu	105.3	0.9045	0.054 ± 0.004		0.091
^{241}Am	59.5	0.6928	0.033 ± 0.004		0.039

^{a,b,c,d} Identified as a single peak in the LaBr_3 -spectrum, the intensities of the gamma-rays are summed for activity determination

^e Mean values

^f Corrected for the contribution of the 1,038 keV gamma-ray of ^{134}Cs

coefficients are calculated from the XCOM: Photons cross section database [16] with the following composition: 52.78 wt% UO_2 , 30.11 wt% C and 17.11 wt% SiC. The calculated radionuclide activities are presented together with the activities calculated with the OCTOPUS code [17, 18]. The activities of the radionuclides identified in both measurements agree well. In particular the activity ETM key-nuclides ^{134}Cs , ^{137}Cs and ^{154}Eu used to determine the activities of difficult-to-measure (DTM) isotopes [19] are determined consistently within one standard-deviation with both detection systems. Furthermore there is a good agreement between the measured activities and the activities calculated with the OCTOPUS code for ^{134}Cs , ^{137}Cs , ^{144}Ce , ^{144}Pr and ^{241}Am .

Conclusion

Within this work it has been shown, that LaBr_3 -detectors are generally applicable in the non-destructive characterization of radioactive waste drums. The activity determination of ETM key-nuclides based on spectra recorded for same measurement times, exhibit similar results for LaBr_3 - as well as for HPGe-detectors. Despite higher counting statistics due to a 2–3 time higher efficiency, it is not possible with LaBr_3 -detectors to achieve a higher level of reliability in activity determination compared to HPGe-detectors at short measurement times. Nevertheless, it has been shown, that relevant ETM key-nuclides are reliably identified and quantified even in complex spectra taken by the LaBr_3 -detector. The response of the LaBr_3 -detector to lower counting statistics is worse than the response of the HPGe-detector, but sufficient for the expected counting statistics in the assay of radioactive waste drums. Furthermore, far better timing properties of the LaBr_3 -detector and thus less dead time offers an improvement over HPGe-detectors especially for the measurement of waste drums exhibiting very high count rates. Considering the additional savings of maintenance costs and higher flexibility LaBr_3 -detectors are a compelling alternative to standard HPGe-detectors for the routine characterization of radioactive waste drums.

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