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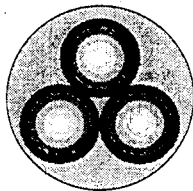
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A Natural Calibration Source for Determining Germanium Detector Efficiencies

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Abstract

In many applications of gamma-ray spectroscopy one is faced with the task of efficiency determinations for large and/or unusually shaped samples. We propose an efficiency calibration technique that takes advantage of the well-understood decays of the long-lived naturally occurring radioisotopes ^{176}Lu , ^{138}La and ^{40}K . Mixtures of known amounts of Lu_2O_3 , La_2O_3 and K_2SO_4 with the sample provide efficiency values for 6 gamma lines ranging from 88 to 1461 KeV. We discuss results and applications of this technique.

1 Introduction

High resolution gamma-ray spectroscopy is widely used as a tool for determining the radioactive contaminants in many different kinds of samples and in detector materials themselves. This technique provides positive contaminant identification, due to the detector's good energy resolution.

In many instances, large and/or odd-sized samples have to be measured, and the correct result depends on the absolute detection efficiency value. Natural uranium and thorium ores have been used for efficiency determination, especially when establishing the activity of naturally occurring radioisotopes. The use of these isotopes as calibration sources is delicate, since uranium and thorium daughters have complex decay schemes and some of them have significant relative uncertainties associated with the absolute gamma-ray intensities. Moreover, when using U and Th as calibration sources, one has to assume the secular equilibrium of the chain, which might have been broken at some point by physical or chemical processes.

Here we describe a method to obtain the absolute detection efficiency by using a mixture of the sample material and known amounts of the naturally occurring long-lived isotopes, ^{176}Lu , ^{138}La and ^{40}K . These isotopes emit gamma-ray lines ranging 88 to 1460 keV and have simple and well understood decay schemes. We present the results of efficiency calibration in two different geometries, using these three isotopes as calibration sources.

2 Methods

The chemical compounds used as sources were Lu_2O_3 , La_2O_3 and K_2SO_4 . The purities were 99.99% for the lutetium and lanthanum oxides and 99.9% for the potassium sulphate (values are from the supplier's chemical analysis). The natural abundances and half lives of ^{176}Lu , ^{138}La and ^{40}K are presented in table 1. Also listed in table 1 are the activities, in Bq, of each one of the radioactive isotopes per gram of chemical compound. Half lives are from references [1], [2], and [3] and the natural abundances of the radioactive isotopes are from references [3], [4], and [5]. The intensities of gamma lines used for the efficiency determination are listed in table 2. The values are from reference [3].

The combination of potassium and lanthanum compounds provides a cross check of the mixing procedure, since they present two gamma-rays of similar energy: 1461 and 1436 keV, respectively.

Table 1: Natural abundances and half lives of ^{176}Lu , ^{138}La , ^{40}K , and the activity in the compounds Lu_2O_3 , La_2O_3 and K_2SO_4 . Values in parentheses are the uncertainties in these quantities

Isotope	$T_{1/2}$ (y)	Abundance (%)	Compound	Activity (Bq/g)
^{176}Lu	$4.08(24) \times 10^{10}$ [1]	2.584 (5) [4]	Lu_2O_3	42.11 (8)
^{138}La	$1.06(3) \times 10^{11}$ [2]	0.089 (2) [5]	La_2O_3	0.68 (2)
^{40}K	$1.277(8) \times 10^9$ [3]	0.0117(1) [3]	K_2SO_4	13.91 (12)

Table 2: Gamma lines used for efficiency determination. Uncertainties quoted in the parentheses include the relative and absolute errors from reference [3].

Isotope	Energy (keV)	Intensity (%)	Uncertainty (%)
^{176}Lu	88.35	13.1	1.3
^{176}Lu	201.82	84	5
^{176}Lu	306.88	93	0.2
^{138}La	788.74	33.3	1.4
^{138}La	1435.79	66.7	1.4
^{40}K	1460.82	10.67	1.0

To illustrate the activity of these chemical compounds, in figure 1 we show a spectrum of a mixture of 0.815 g of Lu_2O_3 , 111.444 g of La_2O_3 and 18.592 g of K_2SO_4 . This sample was placed in a box with approximately the same diameter of the detector's endcap and 2 cm high, and measured for 21.8 h, in a high efficiency geometry. The counting rate in this spectrum was 39.6×10^3 counts per minute, while the background counting rate was 90 counts per minute. Both counting rates were evaluated in the energy range between 30 and 3800 keV.

2.1 The river sediment measurement

In order to test our technique, we used a standard reference material which contains known radioactive contamination, SRM 4350B which is available from the National Institute of Standards and Technology, NIST. Our goal was to show that the known activities in SRM 4350B could be reproduced using ^{176}Lu and ^{138}La for the efficiency calibration. SRM 4350B consists of

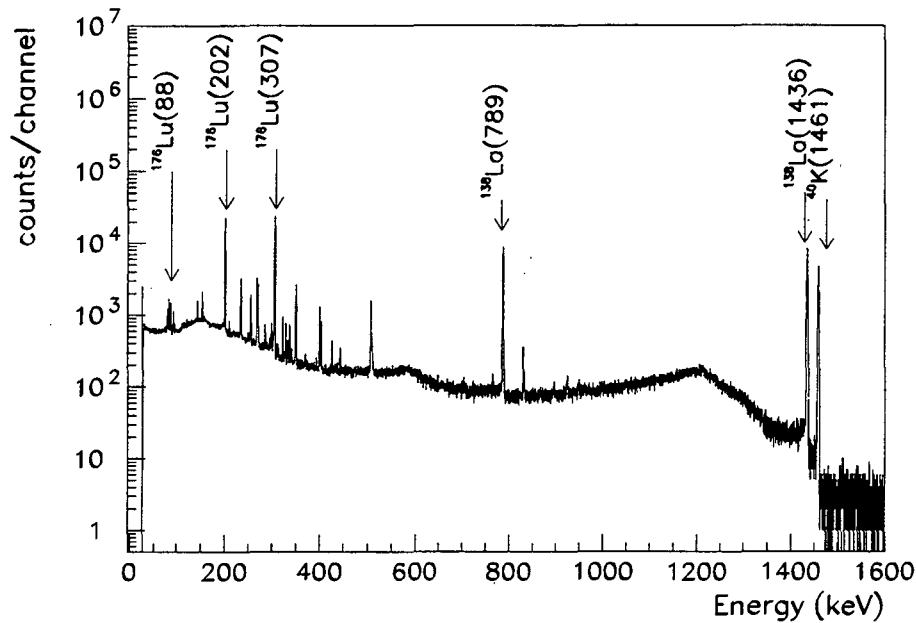


Figure 1: Observed spectrum from a mixture of 0.815 g of Lu_2O_3 , 111.444 g of La_2O_3 and 18.592 g of K_2SO_4 . Data were accumulated for 22.8 h and the counting rate was 39.6×10^3 counts per minute.

freeze-dried, pulverized river sediment from the Columbia river in the state of Washington, with certified activities of ^{60}Co , ^{137}Cs , ^{152}Eu , ^{154}Eu , ^{226}Ra , ^{238}Pu , ^{239}Pu , ^{240}Pu and ^{241}Am . The reference time for these activities is Sep/9/1981, 14.166 y before our measurement was made. Other radioactive isotopes are present but their activities were not certified, because there were no corroborative measurements for comparison. They are: ^{40}K , ^{55}Fe , ^{90}Sr , ^{228}Th , ^{230}Th , ^{232}Th , ^{234}U , ^{238}U , and ^{235}U . Details about the river sediment are available in ref. [6].

The sample was prepared by thoroughly mixing 0.257 g of Lu_2O_3 , 22.449 g of La_2O_3 (both in powder form) and 78.990 g of the river sediment (SRM 4350B). These three components were dried at 60°C for 24 hours prior to weighing. For this measurement K_2SO_4 was not added to the mixture because we also wanted to extract the activity of ^{40}K in the river sediment.

This mixture was placed in a plastic box of 8.5 cm inner diameter and 2.2 cm high and positioned on the detector's endcap. Data were acquired for 126.111 h at the low activity cave at the LBNL Low Background Facility. The detector used was a p-type ORTEC-HPGe detector of 7.46 cm diameter and 9.36 cm length with a resolution of 1.85 keV at 1.33 MeV. The whole set-up was shielded by 10 cm of lead with an inner copper shielding 2 cm thick. The

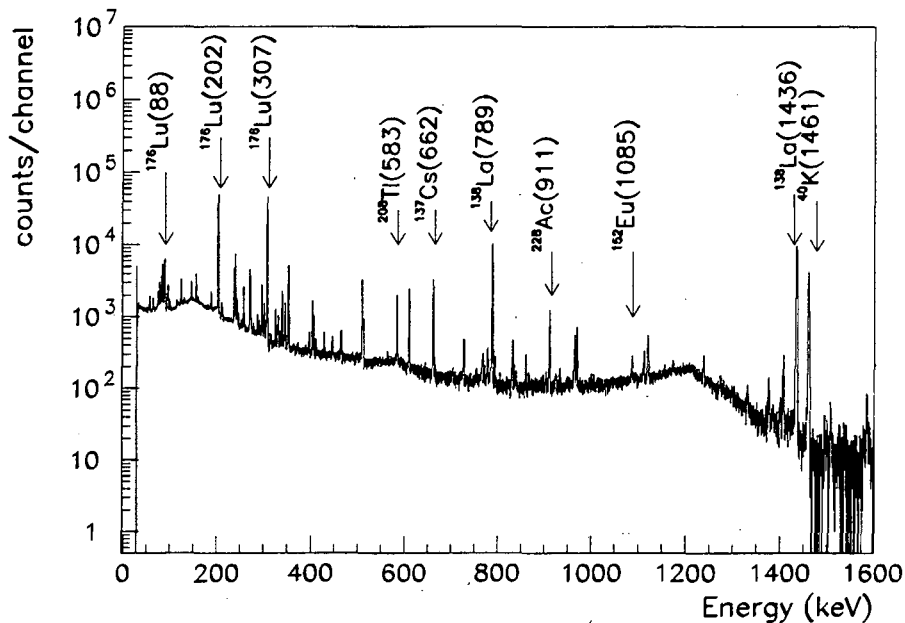


Figure 2: Spectrum from the Columbia River sediment sample, SRM 4350B, 0.257 g of Lu_2O_3 and 22.449 g of La_2O_3 . Room background was subtracted. Data were accumulated for 126.111h.

background was measured without the sample for 317 h and corresponds to 90 counts/minute in the energy range from 30 to 3800 keV. The spectrum obtained, after background subtraction is shown in figure 2.

2.1.1 Analysis and Results

We obtained the efficiency curve for the SRM 4350B using the gamma lines from ^{176}Lu and ^{138}La . Because our sample consisted of an extended source, the mixing procedure can influence the efficiency calibration at low energies. We verified the homogeneity of our mixture experimentally, by re-orienting the sample and re-mixing it and re-counting it. The variation of the measured efficiency in both cases was less than 1% for the three low energy ^{176}Lu gamma-ray lines.

Due to the geometry of the sample and its position close to the detector, the efficiency points from the ^{176}Lu decay, 88, 202 and 307 keV, had to be corrected for summing effects. We performed the summing corrections using KORSUM, a program for calculating corrections for summing coincidences [7].

The lanthanum oxide showed a high level of ^{227}Ac contamination. ^{227}Ac has a

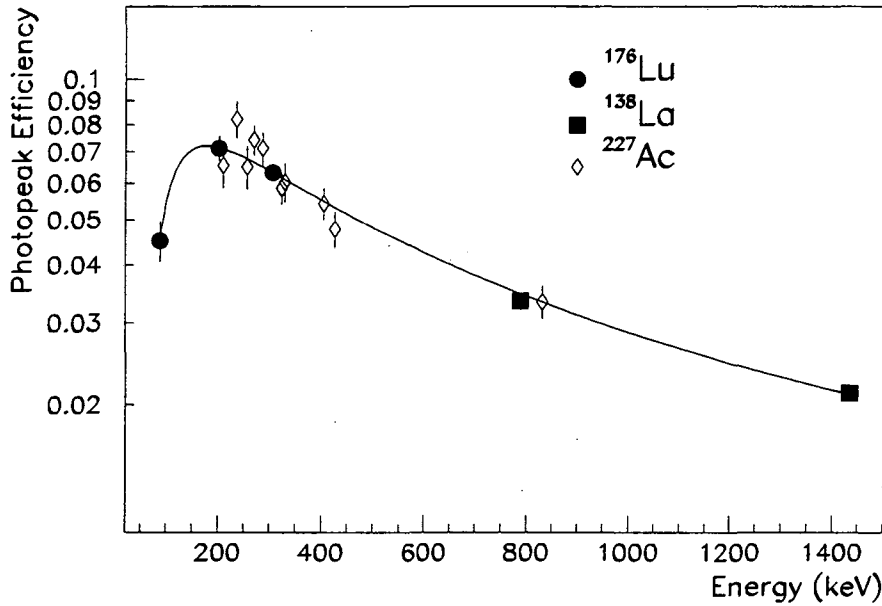


Figure 3: Efficiency curve obtained with ^{176}Lu and ^{138}La points for the SRM 4350B sample. Points from ^{227}Ac contamination are also plotted but were not considered in the fit and subsequent calculations.

half live of 22 y and the equilibrium with its daughters is reached fairly rapidly, in around 200 days. This contamination is common in lanthanum compounds since actinium and lanthanum are located in the same column of the periodic table and have similar chemistry. ^{227}Ac in lanthanum compounds was also observed by Norman and Nelson [2] and Kukoč [8].

Figure 3 shows a photopeak efficiency curve fit using ^{176}Lu and ^{138}La points and an empirical function:

$$\epsilon = \frac{A}{(E + C)} + \frac{B}{(E + C)^2}, \quad (1)$$

where E is the gamma-ray energy and A, B and C are free parameters. This analytical form allows for a simple and practical way to interpolate between the experimental efficiency points.

Using the fitted efficiency curve we obtained the ^{227}Ac contamination by using only the line at 832 keV from ^{211}Pb . The value for the activity of ^{227}Ac and daughters is 0.21 ± 0.02 Bq/g. The ^{227}Ac chain has many gamma-ray lines in the low energy range of the spectrum, below 832 keV, that can also be used as efficiency calibration points as suggested by Kukoč and collaborators [8]. However in this work we only used these points as a cross-check of our

Table 3: Activities obtained from our measurements and the certified reference activities from sample SRM 4350B. Numbers in parentheses are the 1 σ uncertainties for the activities measured in 1995 by us.

Radionuclide	Measured activity (Bq/g) (1995)	Reference Activity (Bq/g) (1995)
^{60}Co	$7.78(29) \times 10^{-4}$	$7.22(36) \times 10^{-4}$
^{137}Cs	$1.88(8) \times 10^{-2}$	$2.10(13) \times 10^{-2}$
^{152}Eu	$1.32(7) \times 10^{-2}$	$1.48(6) \times 10^{-2}$
^{154}Eu	$1.78(49) \times 10^{-3}$	$1.19(18) \times 10^{-3}$
^{40}K	0.536(18)	0.56

procedure at low energies. Efficiency values from low energy gamma lines from the ^{227}Ac contamination are also plotted, although they were not used for the fit. Within the errors, data points from ^{227}Ac and daughters are consistent with the efficiency determined from ^{176}Lu .

Using the fitted efficiency curve and gamma ray lines from the contaminants in the SRM 4350B, we are able to obtain the measured activities of the contaminants in the sample today. The reference activities in 1995 were obtained from the reference activity in 1981, given in the sample certificate. The uncertainties in the NIST reference are quoted as percent errors, and were converted to the errors listed in table 3.

Our results are in good agreement with the certified activities in the sample. It is worth noting that since the reference activity was established by NIST, large fractions of some isotopes have decayed. Nevertheless, the accuracy of our measurement is comparable or better than that of the NIST reference, although we worked with less activity. An exception is ^{154}Eu which has only very little activity left.

Other uncertified activities in the sample are heavy long-lived alpha emitters and do not produce significant gamma-ray emission. These values were uncertified by NIST because only one alpha particle spectrometry measurement was made. However we were able to identify the daughters of these isotopes by their gamma-ray spectrum. In table 4 we list the identified gamma emitters and their activity in the sample as well as the parent isotope and the quoted activity in the SRM 4350B reference.

From the results in table 4, it should be pointed out that ^{208}Tl has a branching ratio of 36% with respect to ^{228}Ac and ^{212}Bi . The activity of the ^{208}Tl parent

Table 4: Measured activities of gamma emitting daughters of heavy contaminants in SRM 4350B. Numbers in parentheses are the 1σ uncertainties in the activities measured in 1995 by us.

Gamma emitter	Measured Activity (Bq/g)	Parent Isotope	Reference Activity(Bq/g)
^{208}Tl	$1.08(8) \times 10^{-2}$	$^{232}\text{Th}, ^{228}\text{Th}$	$3.32 \times 10^{-2}, 3.35 \times 10^{-2}$
^{228}Ac	$2.59(20) \times 10^{-2}$	$^{232}\text{Th}, ^{228}\text{Th}$	$3.32 \times 10^{-2}, 3.35 \times 10^{-2}$
$^{212}\text{Bi}/^{212}\text{Pb}$	$3.09(7) \times 10^{-2}$	$^{232}\text{Th}, ^{228}\text{Th}$	$3.32 \times 10^{-2}, 3.35 \times 10^{-2}$
$^{214}\text{Bi}/^{214}\text{Pb}$	$3.25(22) \times 10^{-2}$	$^{238}\text{U}, ^{226}\text{Ra}$	$3.08 \times 10^{-2}, 3.32(36) \times 10^{-2}$

is then $3.00(22) \times 10^{-2}$ Bq/g, in agreement with the activity measured for ^{228}Ac and ^{212}Bi . Also the measured activities of ^{214}Bi and ^{214}Pb agree with the values quoted for the parent nuclei ^{238}U and ^{226}Ra .

2.2 Large sample measurement

In our first application of the mixed source method for efficiency determination, we measured the unknown radioactive contamination of a sample consisting of 5006 g of 99.999 % pure Nd_2O_3 powder. The sample was measured for 1268.67 h in a Marinelli beaker geometry, using an n-type HPGe detector of 8.03 cm of diameter and 7.8 cm long, located at the LBNL Low Background Facility under the Oroville dam (700 meters-water-equivalent). The detector's shielding consists of 20 cm of lead and an inner copper shielding 1.3 cm thick. This measurement was part of a study of the double beta decay of ^{150}Nd to the excited states of ^{150}Sm and is described in detail elsewhere [9].

In order to obtain accurate values for the efficiency, we assumed that the characteristics of the 5 kg of pure Nd_2O_3 were similar to those of mixture of 14.85 g of Lu_2O_3 , 2014 g of La_2O_3 , 330 g of K_2SO_4 , and 2706 g of Nd_2O_3 of lower grade. This mixture was placed in the same container used for the pure sample and measured for 64 h. Because of the size of the sample, the efficiency points from the ^{176}Lu decay do not need to be corrected for summing effects since the total efficiency is low. The measured radioactive contamination levels of the pure Nd_2O_3 are listed in table 5. Note that the pure Nd_2O_3 contained some contamination of ^{176}Lu . We stress that in this application the absolute photopeak efficiency was determined with a different measurement which reproduces the conditions of the final experiment.

Our resulting efficiency curve for this Marinelli beaker geometry is shown in figure 4. The solid line represents a fit using equation 1. The diamonds rep-

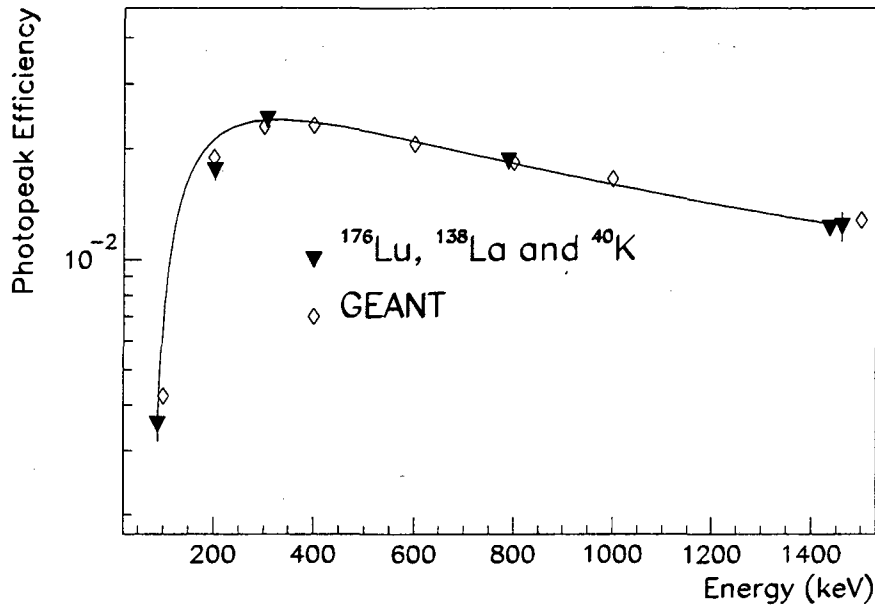


Figure 4: Efficiency curve obtained from ^{176}Lu , ^{138}La and ^{40}K gamma lines in a mixture of Lu_2O_3 , La_2O_3 , K_2SO_4 and Nd_2O_3 . The total weight of the mixture was 5064.85 g. The sample was measured in a Marinelli beaker geometry with a n-type HPGe detector. Monte Carlo points are also shown.

resent Monte Carlo calculation points obtained using GEANT 3.21 [10]. The agreement between simulated and experimental points is very good, showing that our technique can also be employed for testing Monte Carlo calculations of odd geometries.

For completeness, we present in table 6 the experimental values for the absolute photopeak efficiencies obtained in this work for the gamma-rays from ^{176}Lu , ^{138}La and ^{40}K for the two geometries we studied.

3 Conclusions - Comments

Kukoč [8] and collaborators have suggested the use of ^{138}La and its ^{227}Ac contamination for efficiency calibration. The ^{227}Ac and daughters provide low energy calibration points, but this contaminant (^{227}Ac) is not present in a measurable concentration in all lanthanum compounds, lanthanum chloride for example. The use of lutetium compounds allows for calibration points in the low energy range and due to the relatively large natural abundance of

Table 5: Radioactive contaminations observed in the high purity Nd_2O_3 sample. Contaminations are in Bq per gram of Nd_2O_3 . Numbers in parentheses are the 1σ uncertainties in these quantities.

Isotope	Activity (Bq/g)	Isotope	Activity (Bq/g)
^{214}Pb	$8.0(8) \times 10^{-6}$	^{208}Tl	$2.0(5) \times 10^{-6}$
^{212}Pb	$5.8(8) \times 10^{-6}$	^{176}Lu	$2.0(5) \times 10^{-6}$
^{214}Bi	$9.3(13) \times 10^{-6}$	^{228}Ac	$8(5) \times 10^{-6}$
^{40}K	$2.6(3) \times 10^{-5}$	^{212}Bi	$6.3(15) \times 10^{-6}$

Table 6: Experimental values for the absolute photopeak efficiencies for both geometries studied in this work, the geometry of SRM 4350B and the Marinelli beaker geometry for the pure Nd_2O_3 measurement. Numbers in parentheses are the 1σ uncertainties in these quantities.

E_γ (keV)	Efficiency SRM 4350B	Efficiency Marinelli
88.35	0.045 (4)	0.0035 (3)
201.82	0.071 (4)	0.017 (1)
306.88	0.0631 (2)	0.0239 (1)
788.74	0.033 (1)	0.0185 (8)
1435.79	0.0212 (4)	0.0121 (4)
1460.82		0.012 (1)

^{176}Lu , can be used in very small quantities, thus not contributing to the self absorption in the sample under study.

The use of Lu_2O_3 , La_2O_3 and K_2SO_4 as calibration sources provides a reliable efficiency calibration in the energy range between 88 and 1461 keV for powder-like samples. It also provides a ready to use natural calibration source which can be adapted to many different geometries and sample volumes. One of the major advantages of the use of these isotopes as sources is the simplicity of the decay schemes. These chemical compounds are readily available, are not hazardous, and are not considered to be radioactive. Thus, although the source activities will not decrease over the timescale of any research program, disposal of the source is simplified. We would also like to point out that when dealing with liquid samples, it is possible to obtain a similar calibration source by using lutetium, lanthanum and potassium chloride, which have high solubilities in water.

Aknoledgements

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