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Efficiency calibration of Ge detectors in wide energy range with natural thorium and ^{137}Cs

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For the efficiency calibration of Ge detectors the ^{232}Th natural radionuclide chain is suggested with a standard source of ^{137}Cs which due to its long half-life requires a change of the calibrated standard only in about 30 to 50 years.

Introduction

Most Ge detector/MCA set-ups are calibrated for energy and efficiency by the use of commercially available radioactive point sources. Several national standard laboratories and commercial firms offer calibrated sources of mixed radionuclides. A favorite mixture[1] consists of ^{109}Cd, ^{57}Co, ^{139}Ce, ^{203}Hg, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{60}Co and ^{88}Y, which covers the range from 88 to 1836 keV. The disadvantage of this mixture lies in the short half-lives of several nuclides, so that the source is only useful over a period of a few months. COURSEY et al.[2] suggested the use of ^{125}Sb +^{154}Eu+^{155}Eu which covers the energy range from 27 to 1596 keV, with half-lives beyond 2.5 years. This mixture suffers from the disadvantage of many cascade transitions which require correction for coincidence summing. In addition, while 2.5 years is longer than the half-life of the previous mixture, it is still too short, requiring replacement of the standard in less than 5 years.

In order to reduce the correction for coincidence summing, it is preferable to use a mixture of radionuclides each emitting preferably only one photon, rather than using one radionuclide (or a few of them), each emitting several photons with various energies. The main problem of preparing a mixture of one-photon emitters for detector calibration is the lack of sufficiently long-lived radionuclides.

The half-life of any mixture like this is given by the half-life of its shortest-lived member. A solution to this situation can be found in the use of a chain of radionuclides decaying one to another, each of them emitting one or several photons, i.e., A→B→C→D→E→... The half-life of an equilibrated chain like this is given by the half-life of A, usually the longest-lived member.

Several papers[3–6] suggested to use ^{226}Ra as the firs member of the chain. However, ^{226}Ra suffers from two disadvantages: (1) it is rather expensive and requires special laboratories to handle it, and (2) if it is not hermetically sealed, its daughter ^{222}Rn ($T_{1/2}$ = 3.825 d) which is a gas, can leak off the sample aborting the equilibrium.

We would like to present the use of another natural radionuclide chain, that of ^{232}Th. It is suggested to be used as a relative energy calibrator, while absolute calibration will be done by a standard source of ^{137}Cs which, due to its long half-life, requires a change of the calibrated standard only in about 30 to 50 years.

The advantages of ^{232}Th

Two natural very long-lived radioelements have a radioactive chain, i.e., a chain of radionuclides decaying one to another. The advantage of using a natural element rather than a separated shorter-lived fraction (like ^{226}Ra which is part of the ^{238}U chain) is that it is considerably cheaper and can be handled in every laboratory.

The disadvantage of the very long-lived elements is that they have lower specific activities, i.e., lower disintegration rate per unit mass, leading to physically larger sources. Such sources are not point sources but as they are used for relative efficiency measurements rather than absolute measurements, the geometry of the source is less important.

Natural Th is preferable to natural U for three reasons: (1) Th has only one natural isotope and hence the ratios of the various γ-lines in an equilibrated sample are constant whereas in a U sample they depend on the isotope ratio of the two natural isotopes ^{235}U and ^{238}U; (2) ^{238}U chain has a relative long-lived isotope of the gaseous Rn(^{222}Rn $T_{1/2}$ = 3.825 d), and due to this long-life it has a higher chance to leak out of the sample and to disturb the equilibrium. The Rn isotope in the ^{232}Th chain (^{220}Rn) has a half-life of only 55.6 seconds and most probably will decay to the non-gaseous Po before it has a chance to escape from the sample, and (3) ^{232}Th has more γ-photons per gram than ^{238}U, hence, smaller samples can be used.

Several compounds of Th can be used. the highest specific gravity has the metal itself, but its use is not recommended as it is pyrophoric.

Thorium oxide has a quite high specific gravity (9.86 g·cm^{-3}) and its use is recommended. ThO_2 powder can be made to pellets by using high pressure dice.

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To reach high specific gravity and reduce the risk of contamination, it is recommended to use ceramic thorium oxide rather than powder or pressurized pellets. The pressurized pellet can be changed to the ceramic form by sintering at high temperatures (>1200 °C) in H_2/Ar atmosphere.

ThO_2 can be purchased or freshly prepared from $Th(NO_3)_4$ solution by precipitation of thorium oxalate and calcination of the precipitate at about 450 °C.[7]

Looking at the thorium chain (Fig. 1) it is clear that a full equilibrium requires about 23 years (four half-lives of the longest-lived radionuclide in the chain after ^{232}Th). Many of the chemicals commercially supplied are probably not in equilibrium due to separation in the purification processes and shelf time even considerably shorter than the 23 years. This is the reason why thorium and its daughters cannot be used for absolute calibration. However, after ^{224}Ra ($T_{1/2}$ = 3.66 d) all the radionuclides have half-lives shorter than 12 hours and hence the ratios of these nuclides are constant. Due to the short half-life of ^{220}Rn there is very little danger of its leaking out as a gas. Consequently, the ratios of the rates of emission of photons originating from these radionuclides are constant and they can be used to measure the relative dependence of the efficiency of detection on the energy of photons in the range of 39.9 keV to 2614 keV. The γ-ray spectrum of a ThO_2 disc are shown in Fig. 2. From the point of view of equilibrium of the nuclides in the chain, most samples can also use the lines of ^{228}Ac, since it requires only the equilibrium of ^{228}Th, which takes less than 8 years and many samples of thorium nitrate or thorium chloride of this age can be found. However ^{228}Ac is a bad radionuclide for efficiency calibration due to its many γ-lines which will lead to a large correcting factor for cascade summing.[8–12] Corrections for cascade summing are needed also for the lines of ^{212}Pb and ^{208}Tl, but they are smaller.

SCHIMA and HOPPES[8] gave an equation for calculating the correction for the various γ-lines of ^{208}Tl. Examination of the correction factors found by GEHRKE et al.[11] for ^{208}Tl shows that the ^{208}Tl correction factors are about the same as for ^{152}Eu or ^{60}Co and are smaller than that for the 1275 keV line of ^{22}Na used in many commercial calibration mixtures.

Methodology

The chain starting from ^{224}Ra has γ-lines of 39.9, 74.8, 77.1, 87.2, 238.6, 277.4, 300.1, 583.1, 727.2, 860.4, 1620.6 and 2614.6 keV. There is also a line of 510.8 keV but it is mixed with 511 keV annihilation from pair production by the 2614.6 keV photon in the sample and thus cannot be used for efficiency calibration.

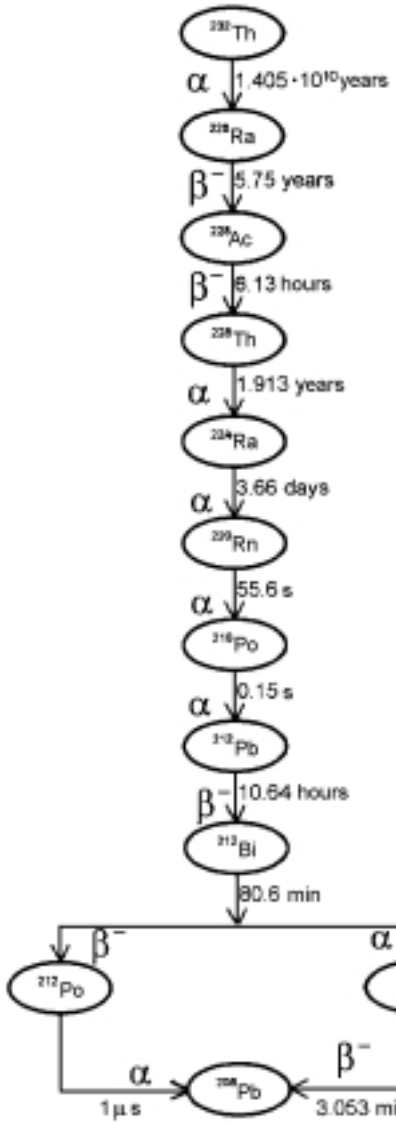

Fig. 1. The ^{232}Th chain

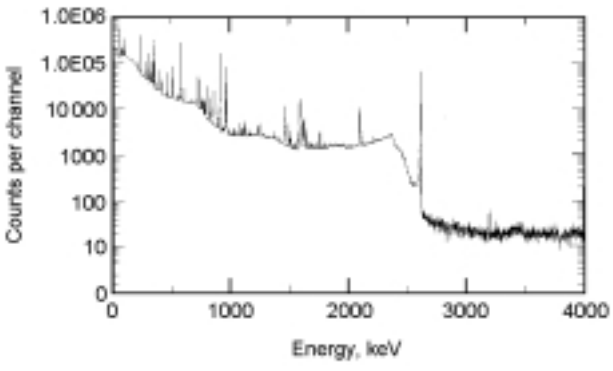

Fig. 2. The γ-ray spectra from ThO_2 pellet, R = 0 cm, H = 12.8 cm

The count rate C_i in the i-th photopeak is given by

$$C_i = \varepsilon_i \cdot \eta_i \cdot N \quad (1)$$

where N is the rate of disintegration of ^{228}Th atoms, η_i is the intensity of the i-th γ-line (in equilibrium with ^{228}Th) and ε_i is the geometric efficiency of the detection of a photon of energy E_i, which is the energy of the i-th γ-line. The geometric efficiency as a function of energy for photons with energy above 150 keV is given usually by

$$\log \varepsilon_i = \sum_{j=0}^{n} a_j (\log E_i)^j \quad (2)$$

with $n = 1$, 2 or 3 depending on the accuracy required and the range of the energy.

Coupling Eqs (1) and (2) gives:

$$\log(C_i / \eta_i) = \log N + \sum_{j=0}^{n} a_j (\log E_i)^j \quad (3)$$

The seven peaks beyond 240 keV can be used to obtain an efficency – energy equation by least squares of the polynominal

$$\ln(C_i / \eta_i) = \sum_{j=0}^{n} k_j (\ln E_i)^j \quad (4)$$

where the coefficients η_i are taken from one of the compilations, such as those of REUS and WESTMEIER[15] or DEBERTIN and HELMER's[14] book or the IAEA compilation[13] since it gives all the γ-rays with the right intensities including those of the daughters in equilibrium.

The factor k_j obtained from the regression analysis is equal to a_j of Eq. (2). However, k_0 from the regression is equal to $\log N + a_0$, rather than to a_0 itself. a_0 cannot be calculated from the ^{232}Th pellet measurement since N is not known. For this purpose additional measurement of a sample containing a known amount of radionuclides is required. The best radionuclide for absolute calibration is ^{137}Cs both from the point of view of its long half-life (30 y) and due to its unique photon, thus not requiring any summing-coincidence correction:

$$a_0 = \log(C_{667} / 0.851) - \log N - \sum_{j=1}^{n} a_j (\log 667)^j \quad (5)$$

This a_0 together with a_1, a_2 and a_3 obtained from the regression analysis, yields a complete efficiency function of the detector-vessel set-up.

Experiments

Several cold-pressed pellets of ThO_2 were prepared about 10 years ago by sintering at high temperature (1600 °C) in the atmosphere of hydrogen/argon. The pellets are cylinders of 6.4 mm diameter and 0.5 to 1.0 mm length. These cylinders contains about 0.12 to 0.24 g ThO_2 (average density 7.3 g·cm^{-3}). For measurements, a ThO_2 cylinder of 6.4 mm diameter was sealed inside an epoxy cylinder. This source gave a dead-time correction of about 9.5% with our detector when measured on the cap.

Results

Since the ThO_2 sample is not a real point source, we have to prove first that it can be used for calibration. Since it is not used for an absolute calibration but only for relative energy calibrations it has to be shown that removing the source either off-center on the cap or separating it from the cap does not change considerably the relative energy counts. The experimental data of the ratio of the counts (after background substraction) of the source located at either $r \neq 0$ (off - center) or $d \neq 0$ (far from the cap) to that on the center of the cap ($r = 0$, $d = 0$), normalized to 1 for 238.6 keV, are given in Table 1.

The measured data were corrected for the effect of self-attenuation of gamma-rays in the source. This was done by dividing of the number of counts in the photopeaks in foreground by the source attenuation factor. The precise calculation of the self-attenuation is difficult and rather uncertain due to the non-point geometry of the ThO_2 source. Assuming negligible radius the correction factor was $[1–\exp(–\mu x)]/\mu$, where μ is the energy dependent linear attenuation coefficient and x is the thickness (height) of the ThO_2 source. Except for the 238 keV peak this correction was negligible. For a 1 mm thick ThO_2 source at 238 keV the correction factor was 0.77.

As can be seen, the changes with the energy up to 1 cm are quite small. As our sample is much smaller ($d \leq 0.32$ cm and $h \leq 0.2$ cm), it can be concluded that even for a calibration on the detector cap we can use our ThO_2 pellet for relative energy calibrations.

Table 2 gives the emission probabilities of ^{228}Th and its daughter, as compiled by IAEA[13] and given also in DEBERTIN and HELMER.[14]

Regression of our data with a polynomial of log E up to $(\log E)^3$ shows for all measurements a deviation for the 727 keV data point. Inspection of the data of ^{232}Th in REUS and WESTMEIER's compilation[15] shows that besides the γ-line of 727.2 keV with an emission probability of 0.0665 there is a line of 727.0 keV with emission probability of 0.008, due to ^{228}Ac. As our pellets are more than 15 years old and hence ^{228}Ac is almost equilibrated, we take for this line an emission probability of 0.0745.

Table 1. Ratio of counts for source located off center ($r \neq 0$) and far from the cap ($d \neq 0$) to that on the center of the cap, normalized to 1 for 283 keV

Energy, keV	238.6	300.1	583.2	727.3	860.6	1620.7	2614.5
h = 1.2 cm	1	1.01	1.11	1.04	1.06	1.02	1.10
h = 2.3 cm	1	1.03	1.18	1.06	1.11	1.04	1.19
d = 1 cm	1	1.01	1.02	1.03	1.04	0.94	1.01
d = 2 cm	1	1.02	1.15	1.12	1.10	1.06	1.12

Table 2. Emission probabilities (η) of photons from ^{228}Th and its daughter

Energy, keV	238.6	300.1	583.2	727.3	860.6	1620.7	2614.5
η	0.435	0.0325	0.3058	0.0664*	0.045	0.0149	0.3588

*The value used was 0.0745 (see text).

For a small ThO_2 pellet placed at a distance of 12.8 cm from the cap, regression at the seven data points (3 degrees of freedom) gives a correlation coefficient $R^2 = 0.990$ and the regression formula is log(counts) emission probability = –10.562+14.966·logE–5.028 (log E)^2+0.528(log E)^3, where E is in keV. Absolute calibration was done with a ^{137}Cs source. The measured efficiency is $7.574 \cdot 10^{-4}$, i.e., log ε = –3.1207. Since the regression formula yields at 661.66 keV log (counts/emission probability) = 3.5029, the regression formula should be corrected by substracting 6.6232 (3.5025–(–3.1207)). Hence:

$$\log \varepsilon = -17.1852 + 14.966 \log E - 5.028 (\log E)^2 + 0.528 (\log E)^3$$

Comparison of this equation with the efficiencies measured for ^{22}Na and ^{60}Co standard sources yields the following values:

Energy, keV	511.0	1173.2	1274.5	1332.5
Efficiency (calculated)	$9.125 \cdot 10^{-4}$	$4.532 \cdot 10^{-4}$	$4.181 \cdot 10^{-4}$	$4.000 \cdot 10^{-4}$
Efficiency (measured)	$9.123 \cdot 10^{-4}$	$4.563 \cdot 10^{-4}$	$4.222 \cdot 10^{-4}$	$4.048 \cdot 10^{-4}$

References

1. B. M. COURSEY, NBS Special Publication 456, 1976, p. 173.
2. B. M. COURSEY, D. D. HOPPES, F. J. SCHIMA, Nucl. Instr. Meth., 193, (1982) 1.
3. G. T. EWAN, A. J. TAVENDALE, Can. J. Phys., 42 (1964) 2286.
4. G. WALFORD, C. E. DOUST, Nucl. Instr. Meth., 62 (1968) 353.
5. G. WALLACE, G. E. COOTE, Nucl. Instr. Meth., 74 (1969).
6. V. ZOBEL, J. EBERTH, U. EBERTH, E. EUBE, Nucl. Instr. Meth. 141 (1977) 329.
7. J. BELLE, R. M. BERMAN (Eds), Thorium Dioxide: Properties and Nuclear Applications, Government Printing Office, Washington, DC, 1984.
8. F. J. SCHIMA, D. D. HOPPES, Intern. J. Appl. Radiation Isotopes, 34 (1983) 1109.
9. J. MOREL, B. CHAUVENET, A. KADACHI, Intern. J. Appl. Radiation Isotopes, 34 (1983) 1115.
10. K. DEBERTIN, U. SCHÖTZIG, Nucl. Instr. Meth., 158 (1979) 471.
11. R. J. GEHRKE, R. G. HELMER, R. C. GREENWOOD, Nucl. Instr. Meth., 147 (1977) 405.
12. D. S. ANDREEV, K. I. EROKHINA, V. S. ZVONOV, I. K. LEMBERG, Izv. Akad. Nauk SSR Ser. Fiz, 37 (1973) 1609. English Translation in the Bulletin of the Academy of the USSR, Physics Series.
13. X-ray and Gamma-ray Standards for Detector Calibration, IAEA-TECDOC-619, IAEA, Vienna, September 1991.
14. K. DEBERTIN, R. G. HELMER, Spectrometry with Semiconductor Detectors, North-Holland, Amsterdam, 1988, p. 364.
15. U. REUS, W. WESTMEIER, Catalog of Gamma Rays from Radioactive Decay, Part II, Atomic Data and Nuclear Data Tables, 29 (1983) 193.