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Inaccuracy Determination in Mathematical Model of Labsocs Efficiency Calibration Program

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Abstract. The study of radioactive materials quantitative inaccuracy determination caused by semiconductor detector aging is presented in the article. The study was conducted using a p-type coaxial GC 1518 detector made of a high-purity germanium produced by Canberra Company and LabSOCS mathematical efficiency calibration program. It was discovered that during 8 years of operation the efficiency of the detector had decreased due to increase of the dead layer of the germanium crystal. Increasing the thickness of the dead layer leads to 2 effects, which influence on the efficiency decrease: the shielding effect and the effect of reducing the active volume of the germanium crystal. It is found that the shielding effect contributes at energies below 88 keV. At energies above 88 keV the inaccuracy is connected with the decrease of the germanium crystal active volume, caused by lithium thermal diffusion.

1. Introduction

Gamma spectrometry is one of the most common methods of non-destructive analysis of nuclear materials and radioactive substances in the field of nuclear non-proliferation, as well as in the field of accounting and control of nuclear materials. Germanium detectors came into widespread acceptance in the gamma spectrometry because of their high resolution and efficiency. Eventually, the outer layer of the germanium crystal undergoes some changes because of the influence of various external factors.

These changes may be mechanical, chemical and radioactive. Mechanical changes mainly refer to micro-scratches and abrasions that occur during the operation process. Chemical changes are usually meant the detector outer surface oxidation, which leads to increase of the germanium crystal dead layer.

Also, there is an accumulation of irreversible radiation damage under the influence of ionizing radiation. Various forms of ionizing radiation interaction with the detector material are the reason of the defects in its crystal lattice structure, and, in most cases, these injuries are increasing proportionally to the radiation dose. The larger the detector, the earlier the degradation of its energy resolution begins when it is exposed by ionizing radiation. As a result of these changes, the detector changes its characteristics. In particular, its own detection efficiency changes, as well as the effectiveness of gamma-ray absorption by the detector. [1,5,6]

Efficiency calibration and measurement technique significantly affect the accuracy of measurements. There are 2 methods of efficiency calibration:

- Mathematical – using mathematical models and calculations.
- Experimental – using special calibration sources.

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2. Mathematical method of calibration

Mathematical methods make unnecessary the calibration sources. However, these methods require certain mathematical models to be created. There are many methods that offer the simulation algorithms for the profile function of the detection system energy and spatial response. Simulating such functions is a challenge, so the mathematical packages are developed to make the user free from self-calculation.

In most cases, the manufacturer creates its own mathematical model of the profile function of the detection system energy and spatial response and the user, with help of the software package, creates a geometric model of the experiment. Then, the software package calculates the detection efficiency of detectors radiation using a mathematical model developed by the manufacturer and the geometric one created by the user.

3. Experiment

The aim of the work is to determine the inaccuracy of mathematical model of LabSOCS (Laboratory Sourceless Object Calibration Software) calibration program, developed in June, 17, 2007 for a coaxial high-purity GC1518 germanium detector produced by Canberra for 2015.

All the measurements and analysis presented in this paper were performed using the LabSOCS program of mathematical efficiency calibration in Genie-2000 software environment. The use of the program provides the most accurate method to conduct qualitative and quantitative gamma-analysis.

The software package is based on a mathematical model of the detector, which describes the profile function of the detector system spatial and energy response to ionizing radiation. Such model is created when the detector is in a particular state, so when we change the detector efficiency it is necessary to correct a mathematical model, because the profile function of the detector system spatial and energy response to ionizing radiation varies over time due to the detector parameters changes.

GC1518 detector is made of high purity germanium. While the detector is produced, the cylindrical sample external surface is sputtered with lithium that diffuses into the sample. The properties of lithium embedded in germanium allow creating sufficiently large areas (with the thickness of more than 1 cm) of almost full compensation and, consequently, the areas with the conductivity close to its own. It is connected both with extremely high mobility of lithium ions into tetravalent crystals and a low energy of its ionization [2].

When the detector is regularly kept at a room temperature, deep lithium thermal diffusion into the germanium crystal actively occurs. As a result, the detector dead layer has increased during 8 years and the active volume of the germanium crystal has decreased. The experimental part presents 2 measurements series of the gamma radiation standard samples in the source-detector geometry using high-purity germanium coaxial detector. 3D source-detector geometric model was created (Figure 1) for each series of the experiments in LabSOCS software package.

Figure 1. 3D geometric model of HpGe detector with the source

In the first series of measurements, standard gamma-radiation samples were in geometry of point source at a distance of 0 mm from the center of the detection unit housing normally. In the second series of measurements, the same standard gamma radiation samples were at a distance of 100 mm from the center of the detection unit housing normally. In particular, the following point sources of gamma radiation were measured: Mn-54, Co-57, Co-60, Zn-65, Cd-109, Sn-113 and Cs-137, Am-241. The measuring time for each source was 300 seconds. This spectrum range time is sufficient, because during this time the number of the counts in the peak is more than 10 thousand. Further, the measured samples spectra were processed using existing mathematical model of the detector, developed in June, 17, 2007, in the energy range from 50 keV to 2000 keV in LabSOCS software package. Obtained activity of measured sources was recalculated at the date of certification to compare it with certified values.

Table 1. Experimental results

Source	E (w), keV (%)	Certified activity at the date of certification, kBq	Measured activity at the date of certification (distance 0 mm), kBq	Inaccuracy (distance 0 mm), %	Measured activity at the date of certification (distance 100 mm), kBq	Inaccuracy (distance 100 mm), %
^{241}Am	59.0 (36.3)	104.0	89.7	13.7	74.1	28.7
^{109}Cd	88.0 (3.7)	118.0	114.9	2.6	98.7	16.4
^{57}Co	122.0 (85.5) 136.0 (10.6)	131.0	124.7	4.8	117.2	10.5
^{113}Sn	391.0 (64.9)	155.0	141.8	8.5	143.9	7.2
^{137}Cs	661.6 (85.1)	101.9	95.1	6.7	91.7	10.0
^{54}Mn	834.0 (99.9)	134.0	125.9	6.1	119.5	10.8
^{65}Zn	1115.0 (50.7)	135.0	116.9	13.4	111.9	17.1
^{60}Co	1173.2 (99.8) 1332.5 (99.9)	90.9	68.2	24.9	81.7	10.1

The table shows that obtained active source values are different from their true certified values with an inaccuracy of more than 5%, indicating that mathematical model is not in accordance with actual profile function of the detector system spatial and energy response to ionizing radiation.

Am-241 high measurement inaccuracy at source-detector distance of 100 mm could be explained by the shielding effect arising from the detector dead layer increase. The shielding effect is usually observed at low energies. Therefore, for gamma rays energies of above 88 keV, this effect makes a small contribution, as it is shown in the Table.

Co-60 high measurement accuracy at the source-detector distance is due to the cascade summation effect. To avoid it, it is necessary to carry out measurements at the distance of, at least, 10 cm from the source.

The Table shows that at energies above 88 keV the measurement inaccuracy is no more than 15%. This inaccuracy is connected with the decrease of the germanium crystal sensitive volume, caused by lithium thermal diffusion.

Thus, the detector has changed its properties since a mathematical model of the detection system was developed. As a result, the effectiveness of gamma-ray detection by the detector has changed.

4. The shift of the calibration curve on semiconductor detector efficiency

Figure 2 and Figure 3 show the efficiency curve of the detector detection in the point geometry for 2007 and 2015 at the source-detector distance of 0 mm and 100 mm normally, respectively.

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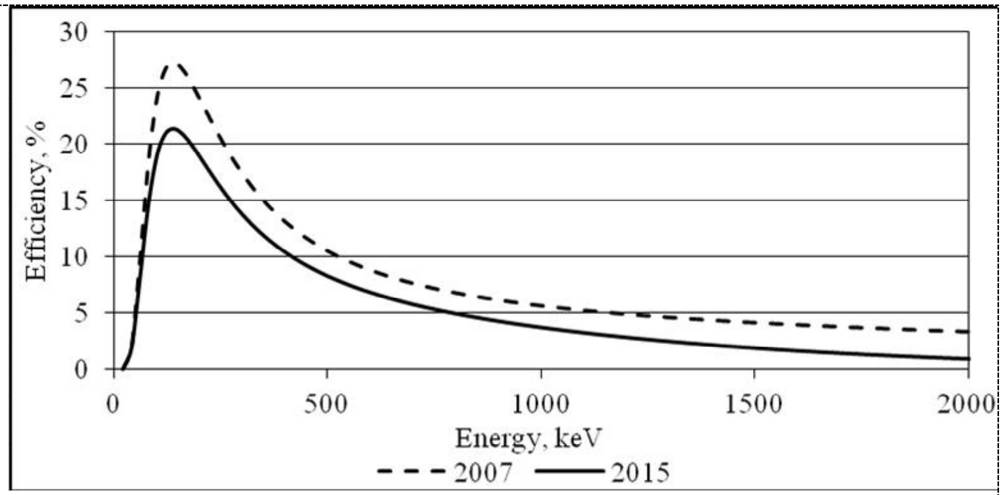

Figure 2. Detector efficiency at the source-detector distance of 0 mm

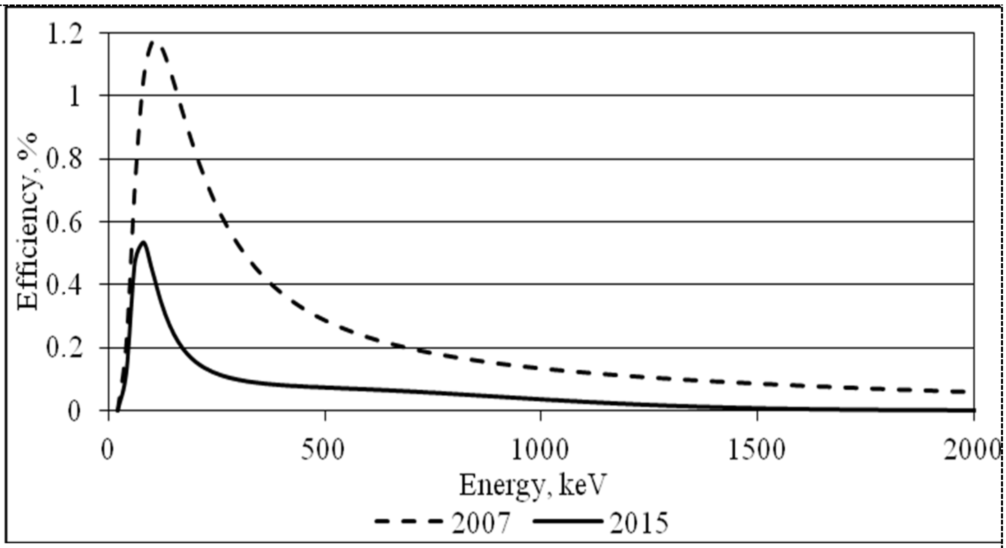

Figure 3. The detector's efficiency at the source-detector distance of 100 mm

The figures show that the efficiency of the detector has changed. There is a general decrease of the absolute detection efficiency of the detector in the whole energy range, which is connected with reducing the sensitive area of the detector. In addition, there is a greater reduction in the detection efficiency of the detector in the low energy region, where the shielding effect occurs due to the increase of germanium crystal dead layer.

Thus, the necessity to create an actual mathematical model of the detector that meets the real function of the detector system spatial and energy response is obvious. Also, it should be noted that it is necessary to increase samples measuring time, since the absolute detection efficiency has decreased, which will affect the rapidity of analysis.

5. Conclusion

As a result of the study, the following conclusions are made. The effectiveness of gamma-ray detection by the detector has changed during 8 years. There is a significant reduction in the detection efficiency of the detector at absolute value over the entire range of energies, but the decrease is

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stronger in low-energy region than in the high-energy one. This is due to the dead layer thickness increase, and, consequently, to the decrease in the sensitive volume of the detector [3].

Measurements inaccuracy of more than 5% occurs in almost the entire area. With such a high inaccuracy it is not recommended to conduct quantitative analysis of nuclear materials and radioactive substances, especially in the field of accounting and control of nuclear materials, as in this field the mass of nuclear materials must be determined up to the gram. [4] Consequently, for further detector use to obtain the most accurate measurements, it is necessary to develop its actual mathematical model corresponding to the actual profile function of the detector system spatial and energy response to ionizing radiation.

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