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Temperature Dependence of NaI(Tl) Radiation Scintillation Detectors' Characteristics

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Abstract:

Scintillators belong to the oldest types of radiation detectors. Nowadays, for spectrometric purposes the use of semiconductive detectors is more common, but scintillation detectors are still in use for various specific measurement purposes. We have investigated the dependence of a gamma spectrum measured by NaI(Tl) scintillation detector on temperature changes. We examined the need of energy or efficiency recalibration and software compensation. Due to temperature dependence, scintillation detectors require energy recalibration before environmental and outdoor measurements or before the use of etalon sources for obtaining spectrum for follow-up calibration. We have elaborated on a simple method for energy recalibration of scintillation detectors at different temperatures. This method was converted into an algorithm and it can be either burned into instrument EPROM or used in software processing.

Keywords:

NaI(Tl) detector, recalibration algorithm, scintillation detector, temperature dependence

1. Introduction

Some of the oldest detection techniques of nuclear radiations are based on the observation of flashes, or so called "scintillations". In 1903, W. Crookes (1832-1919), J. P. L. Elster (1854-1920) and H. F. Geitel (1855-1923) released radiation of radioactive substances onto a thin zinc sulphide (ZnS) layer, and in a dark room they could observe stochastic flashes of light with their bare eyes [1]. Nowadays the flashes are observed not with bare eyes but with the use of electronic data processing devices connected to photoelectron multipliers. These instruments can detect even very weak flashes and generate electric pulses proportional to the strength of light.

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The first really usable photoelectron multiplier was created by a Hungarian physicist, Zoltán Bay (1900-1992) in Budapest, at the Tungsram Company in 1938.

In the last decades, the scintillation technique in nuclear measurements was replaced mainly by the high performance and high purity germanium (HPGe) semiconductor technology, but its importance remained in case of some special areas and measuring tasks, such as:

- special laboratory radiation assignments, where measures of only one or just some isotopes with non-superpositioning, significant peaks are done [2, 3],
- in-situ measures, where easily and rapidly recordable gamma spectra can provide vital information on hotspots, main isotopes determining the field of radiation and specification of suitable places for sampling,

In addition to these, the scintillation technique in military practice is still dominant in the following cases:

- on-board radiation detection modules of mobile CBRN subunits,
- gamma spectrometric measurement devices of SIBCRA (CBRN sampling) groups,
- high-sensitivity gamma detectors of helicopter based or unmanned aerial vehicles' radiation reconnaissance devices.

As it is seen from the given examples, the scintillation technique continues to be the first choice for outdoor gamma spectrometry because of its combination of relatively low cost and high reliability. In case of a short time operation in outdoor environment, this technique is not subjected to a significant temperature variation; therefore, it can be operated in the same way as in laboratory. For their continuous operation in outdoor environment, as in the case of continuous monitoring of environmental radiation, they are likely to be impacted by temperature effect [4-6].

Besides the low resolution, the temperature effect is one of the major drawbacks of NaI(Tl) based systems, or for that matter, of all scintillation spectrometers [7]. This effect is derived from the complex temperature dependence of the system [8, 9] including temperature dependence of the scintillator's light output [7] and decay time constants [10], the photomultiplier tube's temperature drift [11] and the associated electronic components' temperature behaviour [5]. The most significant contribution of the temperature effect coming from the photomultiplier tube is too difficult to handle [12], so the spectrum processing methods would be the most beneficial way to correct the temperature effect.

All of correction methods are related to restoring a shifted spectrum recorded under different temperatures to the position of a reference spectrum which allows the restored spectrum to be processed together with the reference spectrum [13], hence eliminating the effect of the change of temperature between their measurements [5, 6, 9]. The essential condition for spectrum converting methods is the linearity of the overall system that allows to perform linear operations.

In this study we have investigated the dependence of the gamma spectrum measured by a NaI(Tl) scintillation detector on temperature changes. For the experiment we used a climate chamber. Here we outline the results of two series of measurements representing the migration of three specific gamma peak centroids of used isotope sources, 662 keV of ^{137}Cs and 1173 and 1333 keV of ^{60}Co.

Based on the results of these experiments, we present an easy-to-implement method for energy recalibration of measured spectra to allow efficiency calibration and for derivation of activity concentrations in unknown samples.

2. Materials and Methods

2.1. Experimental Setup

The detector used in this paper was a 4.9' × 2.5' (125 mm × 63 mm) NaI(Tl) scintillation detector, TARGET nanoSpec model. The detector was coupled to a preamplifier of direct anode type, with 1 μs decay constant and an amplifier with software selected coarse gain, bipolar pulse shape 1 μs peaking time and 0.1 μs fast channel peaking. The Winchester ADC had 2048-channel conversion range and 512-2048 software selected range. The detector was connected to a PC through a serial port. Spectra were analysed in EXCEL environment with codes written in Visual BASIC. The experiment was carried out in a well-shielded climate chamber, which provided ideal conditions and measuring environment for the tests. The chamber temperature varied from 243 K to 313 K (–30 °C to 40 °C) in increments of 10 K. Temperature changing rate was set to 20 K/h and each set of measurements at a given temperature was taken after 2 h of holding time to allow excellent thermal equilibration. The experimental data were obtained using two radioactive sources: ^{137}Cs (21.77 kBq) and ^{60}Co (1.86 kBq). These sources emit gamma-rays at 662 keV (^{137}Cs), 1173 and 1333 keV (^{60}Co).

2.2. Data Collection

To investigate the spectrum changing phenomenon, we collected two sets (series) of 8 spectra (altogether 16 spectra) in the temperature range of –30 °C to 40 °C, which is the manufacturer's recommended operating range. The first set of spectra (see Fig. 1) was collected using the ^{137}Cs source and the second set using a combined source containing ^{137}Cs, and ^{60}Co. Each spectrum was collected after thermal stability was achieved (at least 2 hs of constant temperature). Detailed analysis of the first set served for elaborating on a simple method for energy recalibration and finally for the compensation of the temperature effect on the detector. The second series was recorded two months later in the same conditions as the first one and it served for validating the developed recalibration method.

2.3. Data Processing

The energy calibration coefficients for a second-order polynomial of the first series were calculated (Tab. 1).

Thus, the energy calibration has the form:

$$E = ax_i^2 + bx_i + c \tag{1}$$

where

x_i – the index of corresponding energy channel,

a, b, c – the coefficients of the energy calibration function.

As the coefficients of the second-order part (curvature) have relatively low values with exponent values of –5 the spectra have good linearity and we can assume that there is the linear relation between channels and energy. We used this fact in the further analysis.

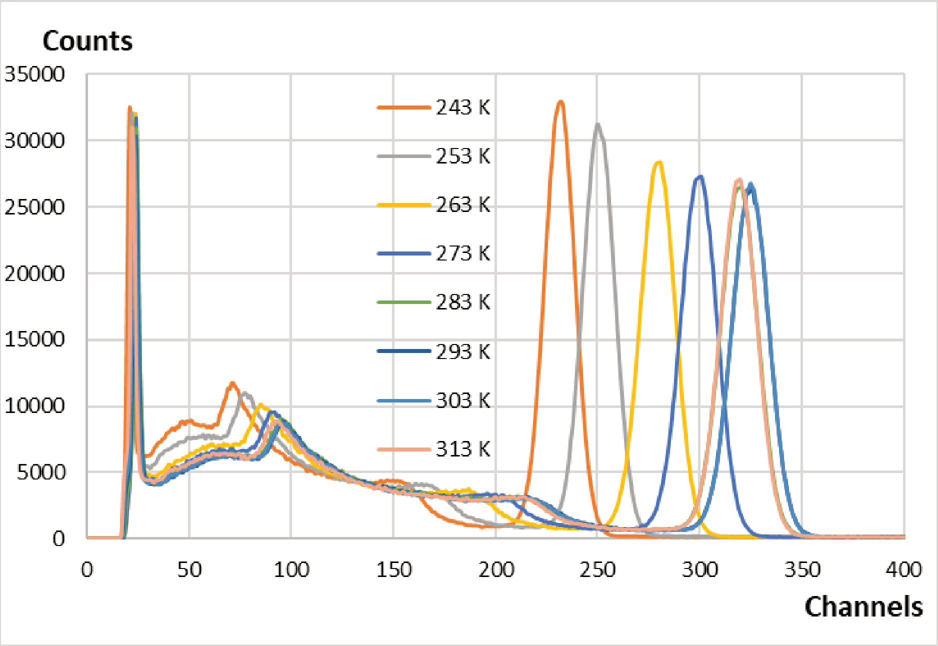

Fig. 1 Temperature dependence of the scintillation detector. Spectra at 293 K - 303 K *overlap each other*

Tab. 1 Energy calibration coefficients for the first series

Temperature [K]	**a**	**b**	**c**
243	3.695×10^{-5}	2.982	–25.20
253	5.865×10^{-5}	2.726	–17.58
263	-1.732×10^{-5}	2.501	–31.32
273	-5.503×10^{-5}	2.372	–39.90
283	-6.990×10^{-5}	2.242	–43.54
293	3.690×10^{-5}	2.111	–23.68
303	6.326×10^{-5}	2.085	–18.04
313	1.885×10^{-5}	2.160	–26.67

Efficiency calibration was performed using simulation software “Geometry Composer” with an empiric correction factor obtained from test measurements (see Fig. 2).

The efficiency coefficient for the 661.7 keV energy of the test source is: $\eta = 1.03 \times 10^{-2}$ (1.03%). The measured spectra during the experiment were evaluated with the formerly detailed method. Tab. 2 shows the results:

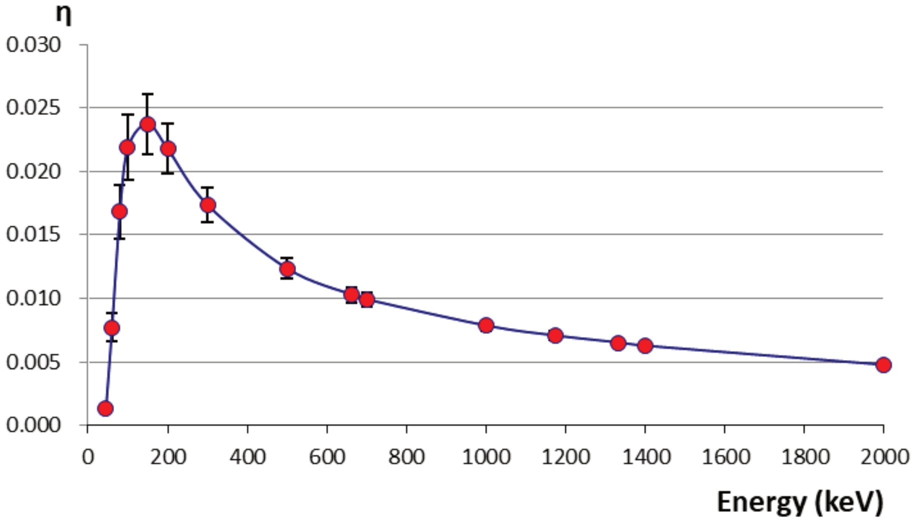

Fig. 2 Efficiency calibration of the measurement system

Tab. 2 Evaluated data of the first measurement series

Temperature [K]	**243**	**253**	**263**	**273**	**283**	**293**	**303**	**313**
Gross Peak Intensity I_G [cps]	700.7	700.0	689.6	687.7	690.3	685.7	686.4	686.3
Background I_B [cps]	39.5	39.3	33.8	34.3	35.2	33.6	33.6	34.4
Net Peak Intensity I_N [cps]	661.3	660.7	655.8	653.4	655.2	652.1	652.8	651.8
Uncertainty of the Peak Intensity ΔI_N [cps]	0.91	0.91	0.90	0.90	0.90	0.89	0.89	0.89
Relative uncertainty Δ [%]	0.137	0.137	0.137	0.137	0.137	0.137	0.137	0.137
Total spectrum intensity [cps]	1945	1947	1952	1959	1963	1958	1959	1965

It can be seen in Fig. 3 that temperature dependence of the energy calibration on the temperature is very strong below 0 °C. The shifting, migration of the spectra's gamma peak centroids in the function of the temperature can be seen on the following diagram Fig. 3.

The centroid migration can be well approximated with a fourth-order-polynomial; therefore the regression equation may be used for energy recalibration.

After the measurements the evaluation of the spectra indicated, that in spite of the strong temperature dependence of the detector's energy calibration, the detector's efficiency has practically no temperature dependence at all. The net intensity of the 662 keV ^{137}Cs peak had only negligible changes (Fig. 4).

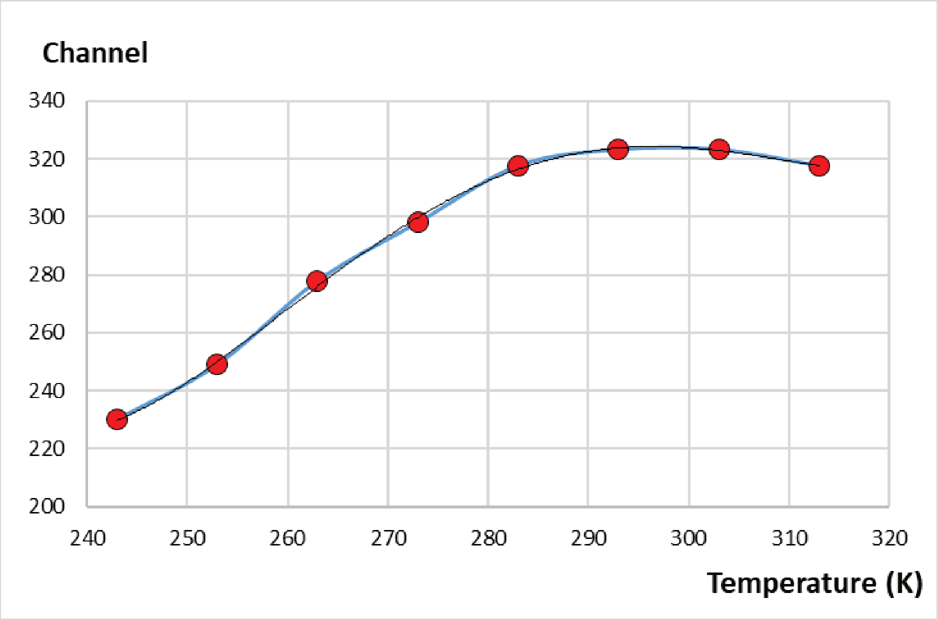

Fig. 3 Temperature effect on the migration of the peak centroids

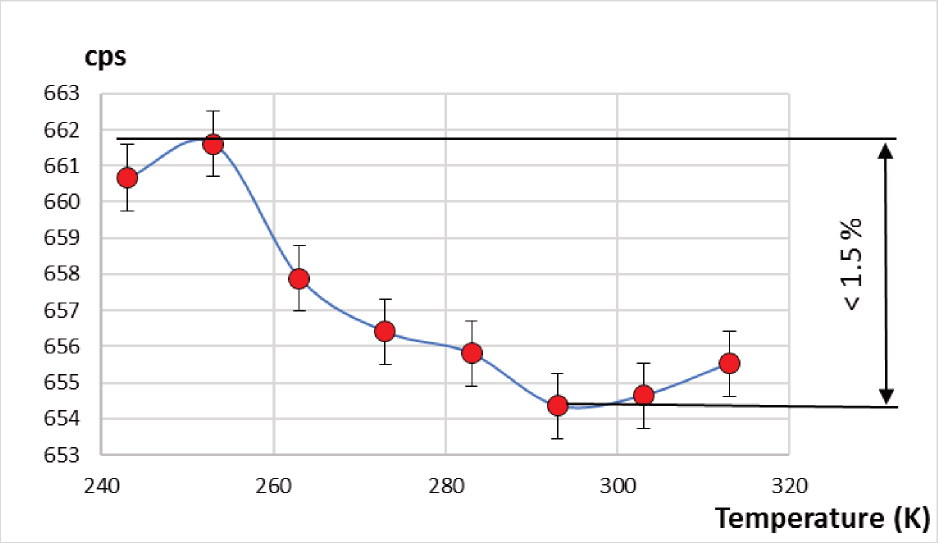

Fig. 4 Temperature dependence of the ^{137}Cs 662 keV *peak intensity*

Similarly, the same tendency can be observed for the intensity change of the total spectrum. However, a small increase can be seen, it remained below 1% and was still negligible (Fig. 5).

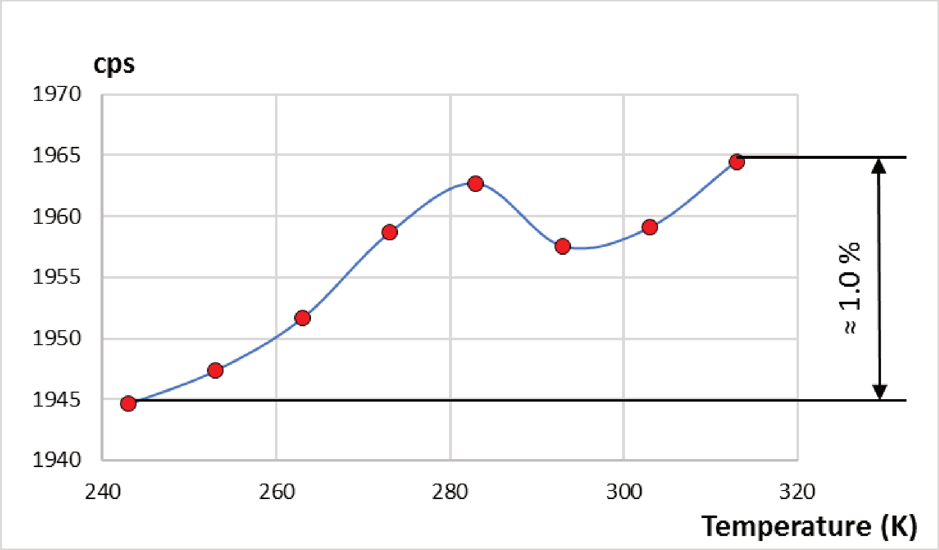

Fig. 5 Temperature dependence of the total spectrum intensity

The relative uncertainty of the peak areas during the whole experiment remained on the same level for 3 decimal values on Δ = 0.137%.

2.4. Energy Recalibration Method

At a constant reference temperature T_0 (e.g. T_0 = 243 K) of the detector, the position x_{i0} of the i^{th}-channel of the MCA remains constant. Thus, the energy calibration is constant. However, temperature changes lead to a channel shift that may invalidate the energy calibration and lead to the misidentification of radionuclides. Thus, the i^{th}-channel position x_{ik} in a gamma-ray spectrum measured at the temperature T_k is displaced with respect to the reference position at T_0, x_{i0}.

If we assume for a fixed voltage and gain that the channel positions only depend on the temperature, we can establish a simple relationship between x_{ik} and x_{i0}:

$$x_{ik} = x_{i0} f_i\left(T_k\right) \tag{2}$$

where $f_i(T_k)$ is the function that depends only on the temperature. Based on the experimental evidence (see Section 3.2), we assume that the relative channel displacement due to the temperature changes is approximately the same for all of the n channels conforming the spectrum. Thus, the $f_i(T_k)$ functions become:

$$f_1\left(T_k\right) \approx f_2\left(T_k\right) \approx f_n\left(T_k\right) = f\left(T_k\right) \tag{3}$$

In addition, we assume that we have the spectrum recorded for example at 243 K as the reference one and we will investigate the migration of full energy peak centroids of nuclides appeared in the spectrum.

To stabilise the spectrum, we move the $x_{ik}^{\text{reference}}$, which is the reference position at T_0, i.e., $x_{ik}^{\text{reference}} = x_{i0}$ to its new position x_{ik}.

Using the assumption of Eq. (2) in Eq. (1), the corrected channel position is given by:

$$x_{ik} = x_{ik}^{\text{reference}} f\left(T_k\right) \tag{4}$$

With the corrected spectrum, the energy calibration obtained at the reference temperature T_0 is still valid. Thus, the objective of the proposed method is to find an approximation to $f(T_k)$ that corrects the energy calibration for the measured spectrum.

This method calculates energy calibration coefficients for the measured spectrum using an algorithm based on previous measurements in the laboratory. Thus, no radioactive or pulse reference is needed during the measurements. However, data must be collected in the laboratory under controlled temperature conditions, which is not always possible. This method assumes a normalized by the reference position fourth-order polynomial approximation (see Fig. 6) to the functions $f_i(T_k)$. Thus, Eq. (1) becomes:

$$f\left(T_k\right) = \frac{x_{ik}}{x_{i0}} = \sum_{j=0}^{4} a_{ij} T_k^j = a_{i4} T_k^4 + a_{i3} T_k^3 + a_{i2} T_k^2 + a_{i1} T_k + a_{i0} \tag{5}$$

where corresponding coefficients are:
$a_4 = 5.111 \times 10^{-8}$
$a_3 = -5.812 \times 10^{-5}$
$a_2 = 2.459 \times 10^{-2}$
$a_1 = -4.583$
$a_0 = 3.182 \times 10^{2}$

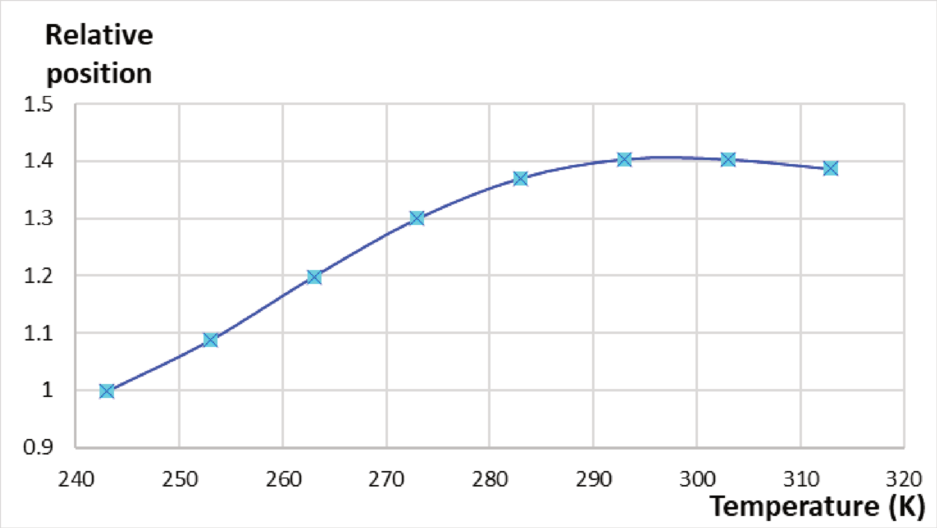

Fig. 6 Fourth-order reference polynomial for calculating relative centroid positions

3. Results and Discussion

3.1. Correction of Temperature Effect on Energy Calibration

For the energy calibration of scintillation spectra, the most common method is to use second-order polynomial approximation. It supposes to use at least three reference full-energy peak covering the energy range of the spectrum.

Having n calibration centroid positions in the reference spectrum, we can convert them for the measured at T_k obtaining a new set of channel positions for the second-order regression. This provides the new energy calibration equation.

3.2. Validation of the Method

To test the validity of our method, we have recorded a new set of spectra (see Fig. 7) under the same conditions as the first one. The sources used for the second series were ^{137}Cs and ^{60}Co.

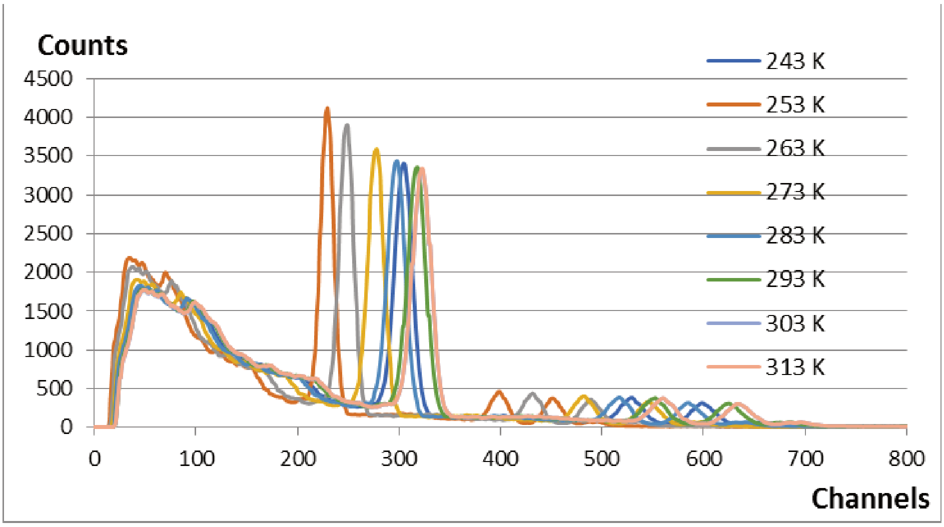

Fig. 7 Spectra of the second series

We determined the displacement of the characteristic energy peaks, then normalised them to the unity at T_0 = 243 K (662 keV, 1 173 keV and 1 333 keV, see Fig. 8).

After normalising we compared the relative centroid positions with calculated ones using the fourth-order reference polynomial $f(T)$ described in Section 2.4 and obtained the relative deviation of the peak centroid positions as it depicted on the diagram (Fig. 9).

Comparison shows that the calculated and measured peak positions have relatively small deviations. This may result in maximum 3-5 channel uncertainty in the higher energy interval, which does not significantly affect the energy calibration.

4. Conclusions

In this paper, we presented a method to handle the peak migration that does not require adjustment of the gain. Thus, the method is useful to manage changes of gamma-ray spectra obtained under unstable temperature conditions.

The method requires previous measurements under controlled temperature conditions, which cannot always be performed and may be specific to each detector.

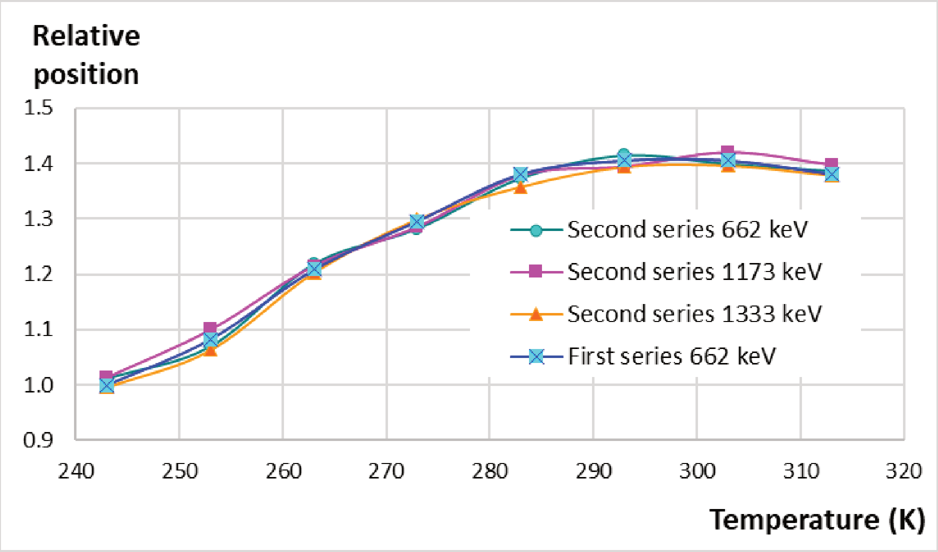

Fig. 8 Relative peak-displacement as a function of temperature for the tested detector. The peak positions are normalised to unity at $T_0 = 243$ K *and are named according to their gamma-ray energy in* [keV]

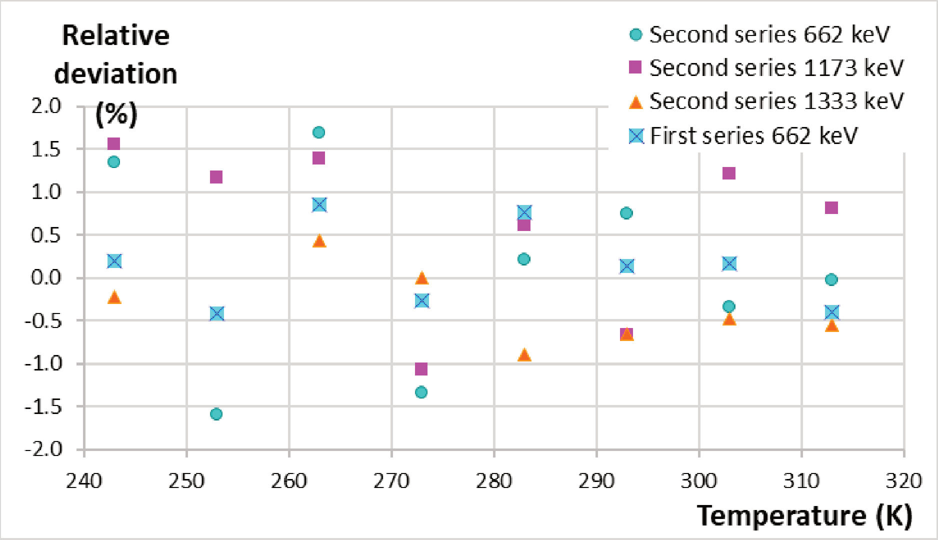

Fig. 9 Relative deviation $(\mathrm{RD}(\%) = (x_{i0}-x_{ik})/x_{i0} \times 100\%)$ *of the peak centroid positions as compared to their calculated positions. The calculation uses regression equation*

Besides, to handle thermal impact on the spectra, continuous temperature monitoring of the detector is required; furthermore, measuring the temperature is effective if the thermal equilibrium is established during the recording spectrum.

The thermal equilibrium can be achieved and may be more stable, if the detector and the thermal sensor are shielded in a container of relatively high thermal inertia.

Finally, this method cannot replace the conventional methods of energy calibration, because they are related to detectors' actual state. In fact, it is useful for automatic monitoring systems when performing calibration procedures is difficult or impossible.

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