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Long-term performance assessment of HPGE detectors used in the neutron activation analysis laboratory of IPEN-CNEN/SP (Brazil)

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A R T I C L E I N F O

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A B S T R A C T

In this work, verification data for 11 HPGe detectors from two different manufacturers and three different intrinsic configurations were analyzed in respect to the stability of both the efficiency and resolution for the 122 keV peak from ^{57}Co and the 1332 keV peak from ^{60}Co. The results allow a discussion about the stability of these parameters over time (in some cases, almost 15 years), their sensitivity to imminent detector failures and their performance after a failure has been corrected; moreover, the results show a clear correlation between the manufacturer or configuration and the long-term performance of the detector.

1. Introduction

In nuclear spectroscopy measurements, the performance of the radiation detectors employed is a key issue for achieving good measurement results. In comparative neutron activation analysis measurements, for instance, much of the methods simplicity depends on the stability of the detector between the measurements of the unknown and the comparator, so that the detectors efficiency can be effectively ruled out of the equation. In the determination of the half-lives of long-lived radioisotopes, instabilities in the detector's efficiency may undermine the resulting values (Siegert et al., 1998).

Factors that might affect the efficiency and/or resolution of HPGe detectors include problems in the detector crystal (as neutron-induced defects or contact migration, for example), in the detector's assembly (vacuum loss is usually the dominant factor) or in the associated electronics (Knoll, 1999; Leo, 1994).

Despite the importance of this matter, few works have been published examining the long-term stability of HPGe detectors. Siegert et al. (1998), have studied a single HPGe detector for efficiency variations over 6 years and determined that its efficiency decrease was less than 0.01% per year (in fact, the variation, if any, was within the limits of the experiment's uncertainty). Sajo-Bohus et al. (2011) reported on the long-term performance of 99 n-type HPGe detectors used mostly in-beam in the INFN-Legnaro, giving details on their performance before and after annealing, showing that in their experience approximately 80% of the detectors recover their nominal performance after annealing, while 20% recover, but not to that same level. In another work, Szymanska et al. (2008) have studied the performance of an HPGe detector under strong magnetic fields for approximately one year, and didn't find any noticeable degradation.

Due to the relevance of the detectors performances, the Neutron Activation Analysis Laboratory of IPEN (LAN-IPEN) has been performing daily verification measurements in its operational detectors since 1999; this verification consists in the measurement of composite $^{(57-60)}$Co sources, in which the position, resolution and cps (counts per second) for the 122 keV peak of ^{57}Co and the 1332 keV peak of ^{60}Co are registered, along with the date and time of the measurement. This verification is performed by whatever user is the first to use the detector on any given day, and the results are manually transcribed into a computer spreadsheet, so mistakes are not unlikely – some are quite evident in a close inspection, some are not, so even after inspection some odd results may appear. Nevertheless, despite these shortcomings, these data may present an excellent opportunity to analyze the long-term stability of these detectors.

2. Experimental procedure

2.1. The LAN-IPEN detector base

The detectors present in the Neutron Activation Analysis Laboratory that have been used in this study are from two different brands (Canberra Industries, identified here as “A”, and Ortec, identified as “B”), have different ages, volumes and configurations. The oldest detectors have verification data available since 1999, and the newest one has these data only since 2011; most of the detectors are connected to regular analogic electronics (amplifier + multichannel analyzer), whereas the three newest ones are directly plugged into Digital Signal Processing combos; moreover, all the detectors except B1 are p-type.

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Table 1
Basic description of the 11 detectors used in this study; the detection efficiency is the nominal relative efficiency; start and end dates refer to the data available, not to the real detector availability; BW stands for Beryllium Window, DSP to Digital Signal Processor, NT to an N-Type Ge crystal and VG to a Variable Geometry assembly ("Pop-Top") – unless otherwise stated, detectors are P-Type, regular coaxials, plugged into regular analogic electronics.

Code	Nominal efficiency	Nominal resolution (keV)	Start month	End	Obs.
A1	20%	2.0	10/2001	06/2016	BW
A2	20%	2.0	07/1999	06/2016	BW
A3	20%	2.0	07/1999	06/2016	BW
A5	20%	1.8	07/2007	06/2016	DSP
A6	20%	1.8	02/2008	06/2016	DSP
A7	30%	2.0	02/2011	06/2016	DSP
B1	20%	1.95	07/1999	12/2010	NT, BW
B2	20%	1.90	09/1999	06/2016	VG
B3	20%	1.90	06/2006	06/2016	VG
B4	20%	1.90	07/1999	06/2016	VG
B5	60%	2.10	10/2001	06/2016	VG

More importantly, although all detectors are used in the vertical position, the detectors from Canberra have a fixed geometry while 4 of the detectors from Ortec (detectors B2-B5) are Pop-Tops, which have the possibility of changing from vertical to horizontal, if required. All detectors are used off-beam, with proper background shielding, in rooms with controlled temperature (20 ± 1 °C) and humidity (50 ± 5%), and are kept permanently refrigerated with liquid nitrogen and under high voltage (except when undergoing a repair procedure). The data for all 11 detectors are presented in Table 1.

All of the detectors from Ortec have had to undergo at least one vacuum recovery/annealing process during this period; as for the detectors from Canberra, none have had vacuum issues so far, but detector A2 had several problems in its associated electronics in the last years.

2.2. Verification measurements

The daily verification measurements are usually performed by the first person to use a certain detector in a given day; the user places a non-calibrated detector-specific piled-up $^{(57+60)}$Co source on the specified position and performs a 600 s livetime acquisition (all detectors work with pile-up reject turned off). The spectra are analyzed using a suitable software (Canberra's Genie-2000 (Canberra, 1999) for $A5-A7$, in-house developed software VISPECT (Zahn et al., 2015) for the others) which calculates peak position, resolution, area (in counts per second of livetime acquisition) and uncertainty. These results for both the 122 keV peak from ^{57}Co and the 1332 keV peak from ^{60}Co are transcribed manually in a paper file, together with the date and time of measurement, then typed into a computer spreadsheet. When the activity of the ^{57}Co source gets too low, this source is changed – but not the ^{60}Co one.

2.3. Data compilation and analysis

The computer spreadsheets, one for each detector, were first checked for obvious typing errors, as problems with dates and/or the decimal separator; in this process some of the data had to be discarded as the results were completely incompatible with the whole, indicating some form of unidentified experimental mistake. The "clean" results were then analyzed in the following way:

- The resolution (in keV) for each peak was plotted against the date, to check for degradation over the years;
- To correct for source changes, sudden pile-up changes (when changing the ^{57}Co for a much stronger one) or other noticeable experimental setup changes, each detector's data was divided in smaller datasets when it seemed necessary – Fig. 1 shows one example;
- To check for overall detection efficiency loss, a simple model assuming linear degradation with time was assumed, so a linear function was added to a modified two-isotope non-paralizable dead time corrected decay function, as seen in Eq. (1), and fitted to the 1332 keV peak's cps data from each subset (where λ_{57} and λ_{60} are the literature decay constants for ^{57}Co (Bhat, 1998) and ^{60}Co (Browne and Tuli, 2013), respectively; $p(1)$-$p(4)$ are the fit parameters, where $p(1)$ is the yearly percent efficiency change, $p(2)$ and $p(3)$ the initial values – somewhat related to the activity, but not explicitly so, as the sources are uncalibrated – for ^{60}Co and ^{57}Co, respectively, and $p(4)$ a dead time correction parameter) – it must be stressed that, as the total count rate isn't registered, an approximation was made in which the background contribution is neglected and the total dead time is assumed to be dependent on the activities of both cobalt isotopes, represented by their peak count rates;
- To check for a possible energy-dependent efficiency loss (for instance, an increase in the dead layer of the detector), a linearly-corrected exponential decay, seen in Eq. (2), was fitted to the ratio between the counts per second for the 122 keV and the 1332 keV peaks, using literature values for both decay constants (Bhat, 1998; Browne and Tuli, 2013) ($q(1)$ and $q(2)$ are the fit parameters, where the first gives the yearly percent change and the second refers to the initial amplitude of the ratio).

$$F(t)=\left[100+\frac{p(1)\cdot t}{365.25}\right]\cdot p(2)\cdot e^{-\lambda_{60}\cdot t}\cdot\frac{e^{-p(4)\cdot[p(2)\cdot e^{-\lambda_{60}\cdot t}+p(3)\cdot e^{-\lambda_{57}\cdot t}]}}{1+p(4)\cdot\ [p(2)\cdot e^{-\lambda_{60}\cdot t}+p(3)\cdot e^{-\lambda_{57}\cdot t}]} \tag{1}$$

$$F(t)=\left[100+\frac{q(1)\cdot t}{365.25}\right]\cdot q(2)\cdot e^{-(\lambda_{57}-\lambda_{60})\cdot t} \tag{2}$$

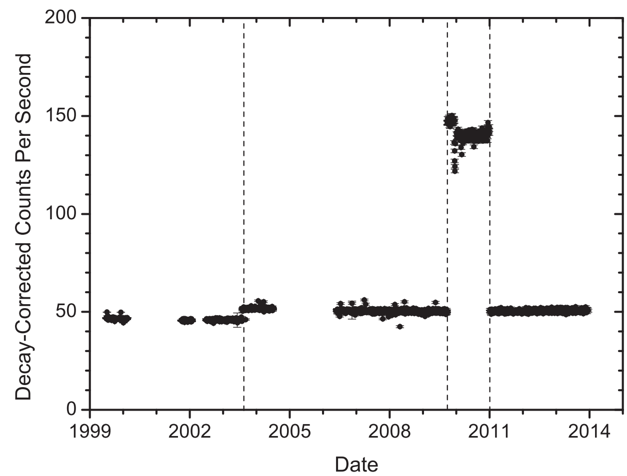

Fig. 1. Complete decay-corrected 1332 keV efficiency dataset from detector A3 showing the division in smaller datasets.

3. Results and discussion

In the analysis of the resolution dependence with time, the results from both the 122 keV and the 1332 keV peaks showed the same overall features, but as the data from ^{57}Co had larger dispersions due to the reduction in counting statistics after some time of source decay, only the results for ^{60}Co are displayed here.

The analysis of the resolution plots for the 6 Canberra detectors, seen in Fig. 2, reveals that detectors A1 and A3 have experienced an essentially smooth 10–20% increase in their resolutions over a period of approximately 15 years, while detectors A5 and A6, much more recent,

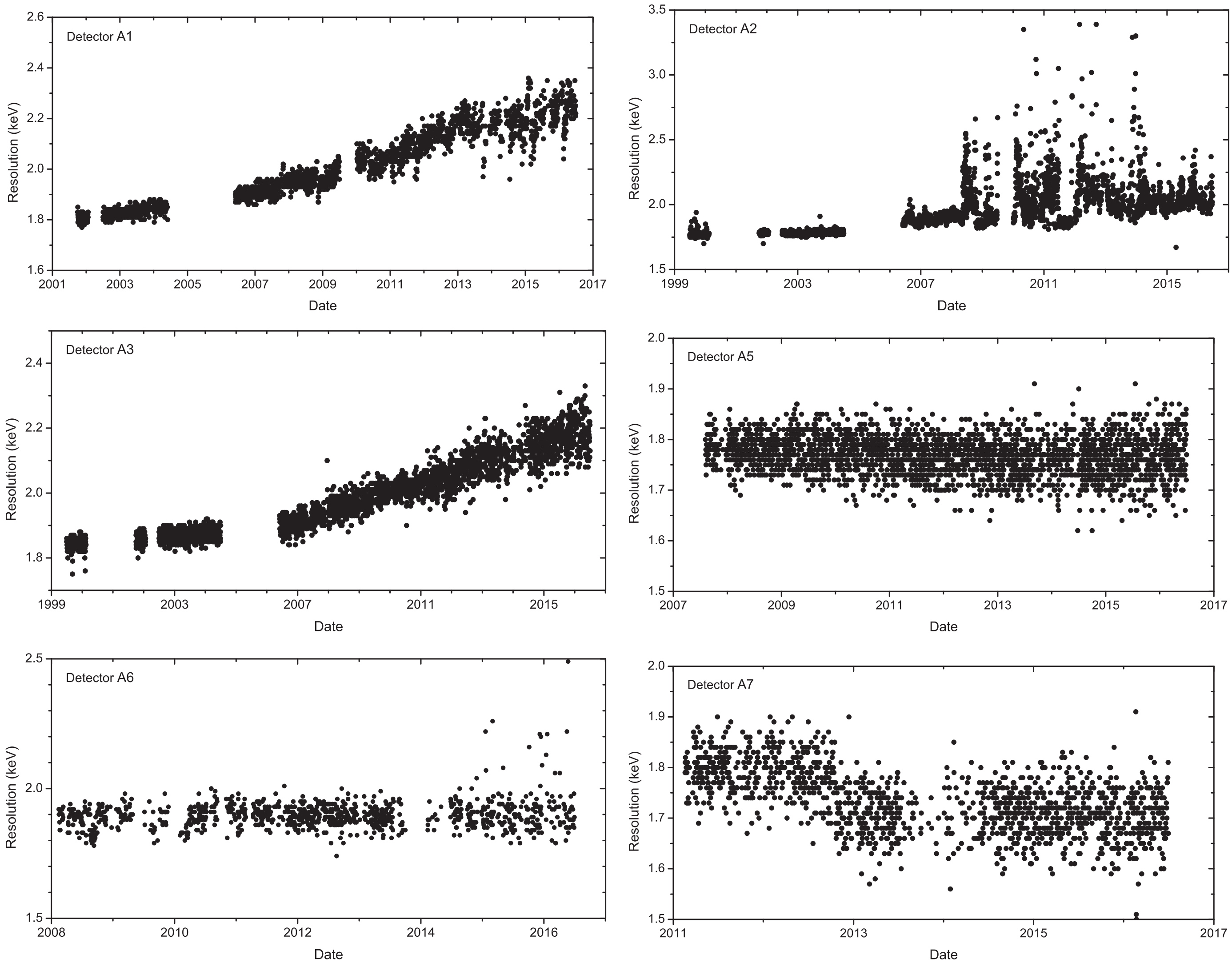

Fig. 2. Evolution of the resolution of Canberra Industries detectors (A1-A7) for the 1332 keV transition from the decay of ^{60}Co.

showed no sign of resolution worsening; detector A2 showed several "spikes" in the resolution from 2008 on, and these are most likely due to a series of bad contact problems that arose in the external electronics; as for detector A7, the sudden improvement in the resolution is due to changes in some of the acquisition parameters of the Digital Signal Processor – it should be noticed that none of these detectors underwent any vacuum-recovery procedure.

The resolution plots for the detectors from Ortec (Fig. 3), however, tell a completely different story: all these detectors show a much faster worsening of their resolutions, and all of them required several vacuum-recovery procedures during this period – in fact the sudden increase in the resolution is usually considered a sign of vacuum loss, and this has been consistently the case with these detectors. It should be noted, too, that detector B1 is no longer in use and that detector B3 is now hardly used after a 2-year maintenance stop in 2012–2013, in which its operating voltage was decreased in order to prevent further damage. These vacuum issues could be related to the more fragile vacuum assembly of these geometry-variable detectors when compared to the more robust single-geometry vertical detectors A1-A7. One important trend is that in all cases the resolution was clearly improved with the vacuum recovery process, returning to its initial value.

In the test of the overall detector stability for the 1332 keV transition, the function in Eq. (1) could not be properly fitted for all subsets from each detector, so only the subsets where a proper fit was obtained were used, and a weighted average was taken from the results for all subsets for each detector. The resulting values for the parameter $p(1)$ are shown in Table 2 and indicate that, while for detectors A1, A7 and B5 no detection efficiency loss could be identified with 95% confidence (2σ interval), for the other detectors it was mostly below 1% per year. One fact that should be stressed, though, is that there is no noticeable change in the efficiency even when a detector was in a pre-failure state - Fig. 4 shows an example of such a case in detector B4, where it is clear that the worsening of the detector's resolution is not reflected in the efficiency results.

In the analysis of the ratio between the efficiencies at 122 keV and 1332 keV, the results were mostly inconclusive, due to the necessary segmentation of the datasets and, mostly, due to the low activity of the ^{57}Co sources in a great part of the data, which resulted in high uncertainty in many of the 122 keV results. Nevertheless, there was no evidence of degradation in the efficiency ratio in any of the detectors which seems to be an indication that the dead layer of these detectors remained quite stable with time.

4. Conclusions

The analysis of the long-term performance of 11 HPGe detectors from two different major brands over periods up to 15 years showed that the detectors from one of the manufacturers (Canberra Industries) suffered much less degradation in the resolution than those from the other (Ortec), but this could be due to the fact that detectors from this

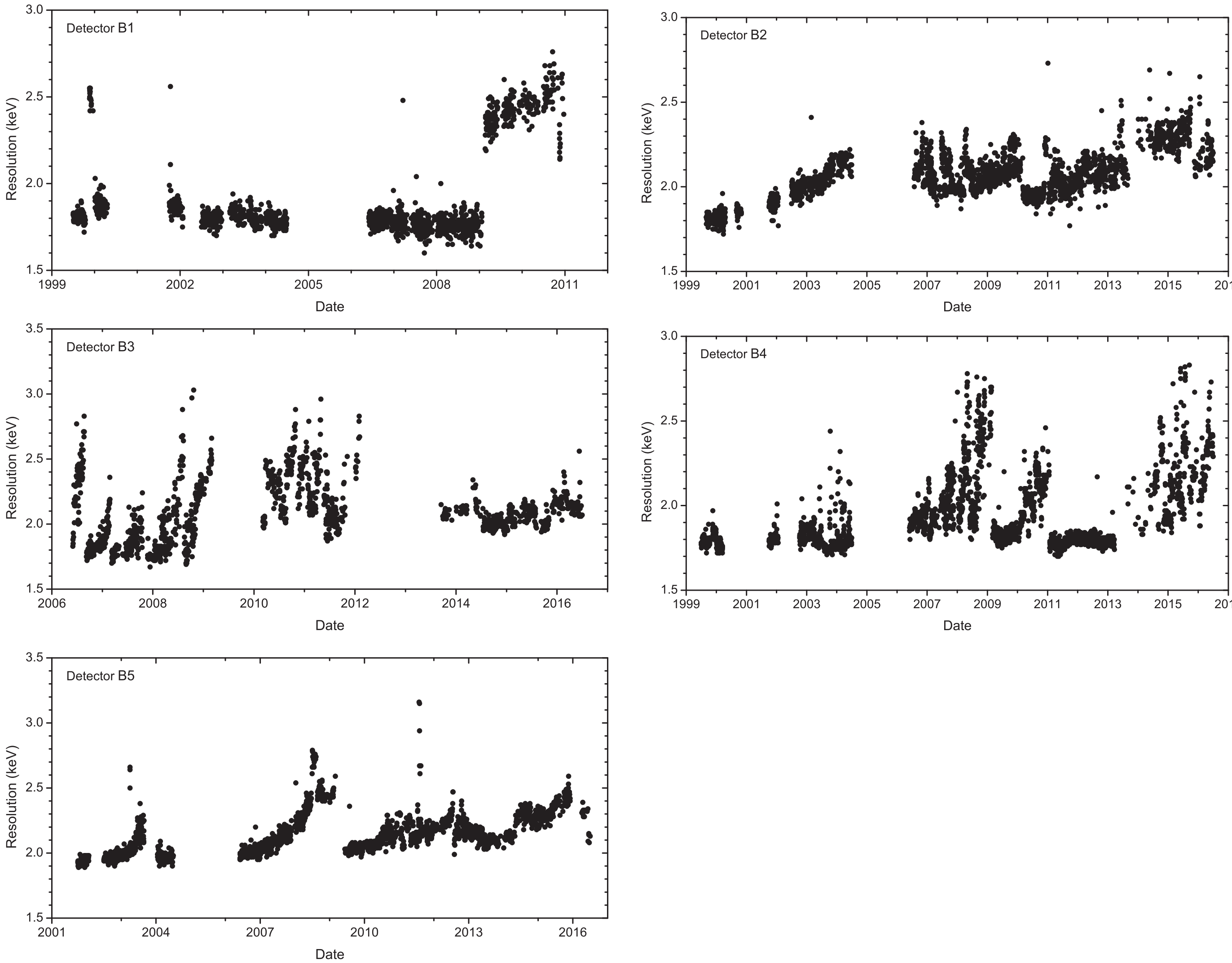

Fig. 3. Evolution of the resolution of Ortec detectors (B1-B5) for the 1332 keV transition from the decay of ^{60}Co.

Table 2
Results obtained in the ^{60}Co efficiency test for the 11 detectors; the total period shown includes only the periods that were effectively used in this evaluation and $p(1)$ is the result from the fit of Eq. (1), with its 1σ uncertainty.

Detector	Total period (days)	p (1) (see Eq. (1)) (% per year)
A1	5077	0.009 ± 0.016
A2	5268	−0.230 ± 0.017
A3	4720	−0.45 ± 0.04
A5	2168	−0.77 ± 0.11
A6	2028	−1.28 ± 0.17
A7	1777	−1.02 ± 0.65
B1	4111	−0.21 ± 0.03
B2	4911	−0.64 ± 0.09
B3	2895	−0.93 ± 0.12
B4	6704	−0.062 ± 0.018
B5	5354	0 ± 0.005

manufacturer were from a variable geometry line ("Pop-Top"), and that characteristic could affect the longevity of the vacuum insulation in these detectors. As for the detection efficiency, there was no noticeable difference between the detectors of both manufacturers, and the overall efficiency loss ranged from zero to 1% per year. Qualitatively, also, there was no evidence of change in the ratio between the efficiencies at 122 keV and 1332 keV, indicating that the dead layer of the detectors appeared to remain stable over the years.

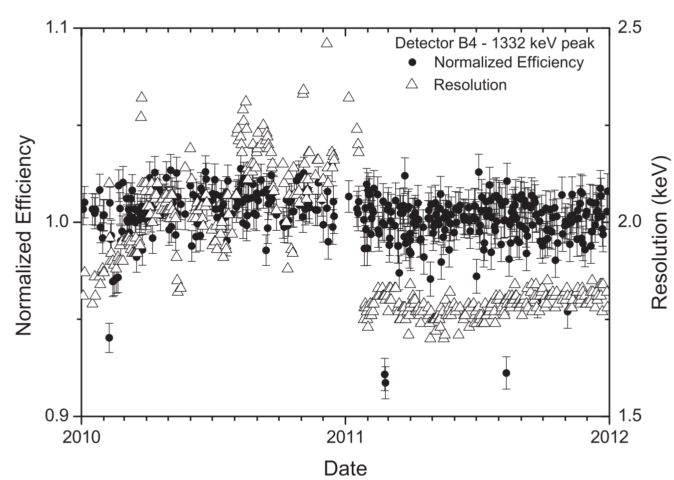

Fig. 4. Normalized efficiency (black dots) and resolution (gray triangles) results for the 1332 keV transition in detector B4 in a two-year period, where the vacuum loss can be clearly noticed in the worsening of the resolution, with no noticeable effect in the efficiency; the detector underwent an annealing process in the period of 21–25/january/2011 and its resolution went back to the values obtained prior to the problem.

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References

Bhat, M.R., 1998. Nuclear data sheets for A = 57. Nucl. Data Sheets 85, 415–536.
Browne, E., Tuli, J.K., 2013. Nuclear data sheets for A = 57. Nucl. Data Sheets 114, 1849–2022.
Canberra, 1999. Genie-2000 Spectroscopy System – operations manual. Meriden, USA.
Knoll, G.F., 1999. Radiation Detection and Measurement, 3rd edition. Wiley, New York.
Leo, W.R., 1994. Techniques for Nuclear and Particle Physics Experiments. Springer, New York.
Sajo-Bohus, L., Rosso, D., Castelli, A.S., Napoli, D., Fioretto, E., Menegazzo, R., Barros, H., Ur, C., Palacios, D., Liendo, J., 2011. HPGe detectors long time behaviour in high-resolution γ spectrometry. Nucl. Instrum. Methods A 648, 132–138.
Siegert, H., Schrader, H., Schotzig, U., 1998. Half-life measurements of europium radionuclides and the long-term stability of detectors. Appl. Radiat. Isot. 49, 1397–1401.
Szymanska, K., Achenbach, P., Agnello, M., Botta, E., Bracco, A., Bressani, T., Camera, F., Cederwall, B., Feliciello, A., Ferro, F., Gerl, J., Iazzi, F., Kavatsyuk, M., Kojouharov, I., Pochodzalla, J., Raciti, G., Saito, T., Lorente, A.S., Tegner, P.-E., Wieland, O., 2008. Resolution, efficiency and stability of HPGe detector operating in a magnetic field at various gamma-ray energies. Nucl. Instrum. Methods A 592, 486–492.
Zahn, G.S., Genezini, F.A., Moralles, M., 2015. Evaluation of Peak-fitting Software for Gamma Spectrum Analysis. URL 〈https://arxiv.org/abs/1511.04362v1〉.