Single crystal HPGe (80%) versus BGO shielded CLOVER detector for high precision decay rate measurements: a comparative study

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Abstract

In this study, various detector configurations have been investigated in order to explore the optimal condition for decay rate measurements of radioactive samples using gamma spectroscopy technique. A limitation of detecting low energy gamma rays from decaying radioactive nuclei, is the Compton background which can be significantly reduced by rejecting Compton scattered events through active Bismuth germanate (BGO) shielding. On the other hand, for a CLOVER detector without BGO shielding, one can place the radioactive samples very close to the detector for enhancing geometrical efficiency. A single crystal High Purity Germanium (HPGe) detector can also be used for decay rate measurements. In order to measure the decay rate of nuclei decaying via gamma emission with reasonable intensity, optimal close geometry options have been investigated for various HPGe detector configurations.

Introduction

Among the parameters characterizing a radioactive decay, half-life or decay constant can be considered as the most fundamental property and continual efforts have been carried out for obtaining more and more precise measurement of this quantity. The exponential-decay law had to withstand numerous tests as researchers investigated the possibility of dependence of the radioactive decay constant on static factors like temperature, pressure, magnetic field or temporal factors like solar influence. Experimentalists advocating the constancy of nuclear decay constant [1–3] claimed that the decay of radionuclide can be characterized solely by a single decay constant which is independent of its physical and chemical conditions. However, it was pointed out quite early that the nuclei which undergo decay via internal conversion or electron capture might be influenced by external environment as they involve direct contributions from atomic electrons. There is a long history of ^{7}Be decay measurements where small but detectable effects on the radionuclide's decay constant caused by the surrounding environment have been reported [4–9].

Recent measurements claimed to observe relatively large changes in half-lives of α, β⁻, β⁺ and electron capture decays. These claims have attracted considerable interest as they could possibly lead to the improved disposal of radioactive wastes and it provides inspiration for new physical theories and applications [10]. From a metrological point of view, factors to be considered are instruments, electronics, geometry and background which may vary due to external influences such as temperature, pressure, humidity and natural or man-made sources of radioactivity. However, the claim of change in half-lives and any deviation of decay curve from an exponential decay law can only be considered when the instrumental effects have been fully compensated and/or accounted for in the uncertainty budget [11, 12].

The study of the change in nuclear decay rates under compression is a topic of fundamental significance and it has application in several fields like astrophysics, geophysics, condensed matter etc. There are only a few measurements on the decay rate change of electron capturing nuclei under compression and change in decay rate ~ 1% have been observed [4, 6, 8]. The radioactive samples

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could be compressed by two methods: (a) applying external pressure on the radioactive sample using a Diamond Anvil Cell (DAC) and (b) implanting the radioactive atoms in the octahedral and tetrahedral spaces of a small lattice (Pd) and large lattice (Pb) and subsequent change in decay rate under compression could be studied. As gamma-spectroscopic technique is one of the commonly used techniques for such decay rate change measurements, to perform high precision decay rate measurement optimal configuration for higher statistics and reduction of pile up events is an important consideration.

In a gamma spectroscopy measurement, the source should be placed at a position considering efficiency, dead time, summing effects and detector resolution. In a nuclear decay rate measurement, the intensity variation of a particular gamma-ray is generally followed with time, so the sample is placed at a closed geometry with respect to the detector for enhancement of geometrical efficiency. A BGO shielded CLOVER detector reduces Compton background and enhances photo-peak efficiency using addback option. Whereas without a BGO shielding, one can place the radioactive samples very close to a CLOVER detector or a single crystal HPGe detector for enhancing the geometrical efficiency. We present here a comparative study of single crystal HPGe (80%) versus CLOVER detector with and without active BGO shielding to find the optimal configuration for nuclear decay rate measurements.

Experiment

In order to compare the efficiency between the HPGe (80%) and BGO shielded CLOVER detector assembly, the radioactive sample was kept between the two detectors. The single crystal HPGe detector from M/S Baltic Scientific Instruments is a p-type liquid nitrogen cooled coaxial detector used in this experiment and shown in Fig. 1a. The sensitive area of the HPGe crystal (80% efficiency compared to (3″×3″) NaI(Tl) detector) is 78 mm (dia)×72.8 mm (depth). The CLOVER detector used in this experiment is a n-type coaxial HPGe detector (Model No. EGNC-25-200-R of M/s. Canberra, France) with Aluminium window having BGO Anti-Compton shield (ACS) in the horizontal plane. In this detector, 4 HPGe crystals, each of size 50 mm (dia)×70 mm (length), constitute the CLOVER which is placed inside an Aluminium housing [13] shown in Fig. 1b. In the experimental set up, the detectors are placed facing each other so that the front face of Aluminium housing the crystals of CLOVER detector is at a distance of 7.65 cm inside the BGO shield collimator as shown in Fig. 1c. The marked portion of the detectors in Fig. 1a, b are shown in Fig. 1c with a
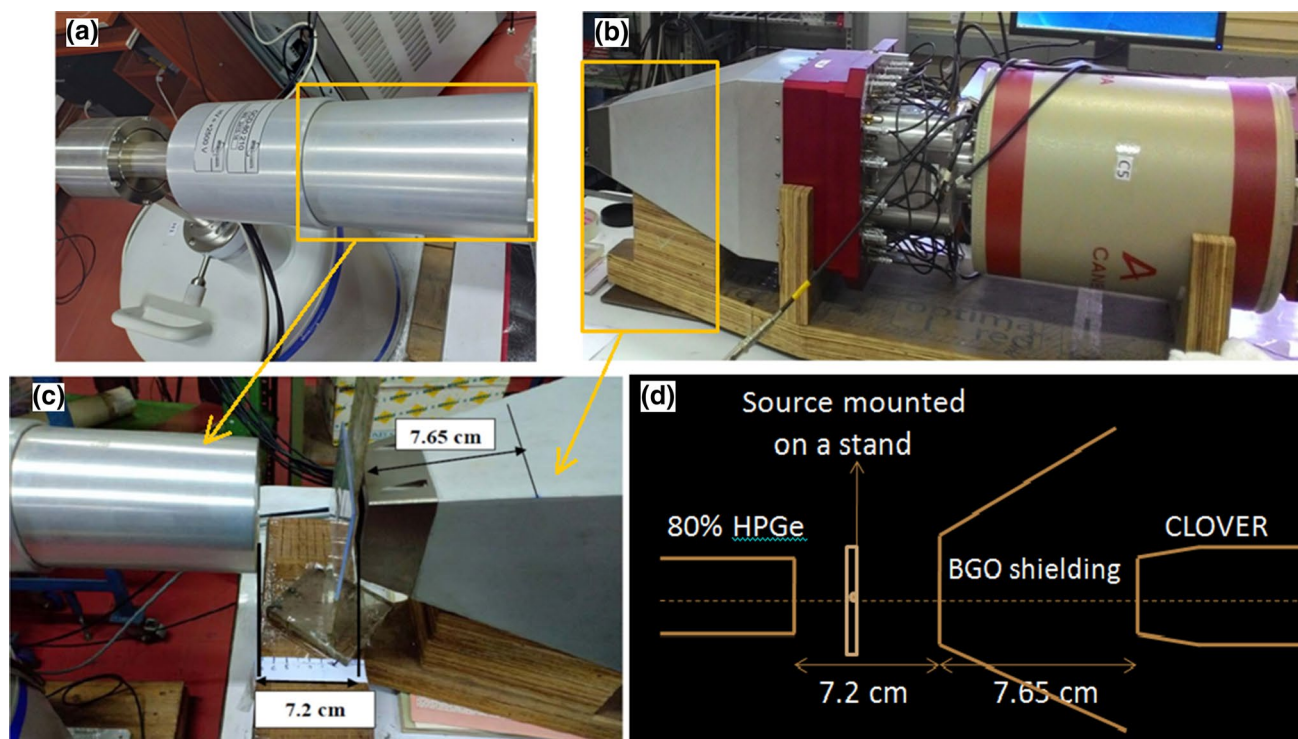

Fig. 1 **a** HPGe detector, **b** BGO shielded CLOVER detector, **c** experimental setup: HPGe detector and BGO shielded CLOVER detector facing each other with a source mounted on a stand in between, **d** schematic layout of the experimental setup

sample holder placed in between. Schematic diagram of the experimental setup is shown in Fig. 1d.

The pulse processing from the HPGe detector was carried out with conventional NIM electronics consisting of high resolution Spectroscopy Amplifier (ORTEC 672), Timing Filter amplifier (ORTEC 474) and Constant fraction discriminator (ORTEC 484) modules. For Clover detector, IUAC make Clover module was used which is a compact version of all necessary modules required for signal processing [14]. For both the detectors, the gain was adjusted to the energy range ~ 2 MeV. The MASTER trigger for data acquisition was generated by using the NIM standard logic modules (Philips Scientific 756 and ORTEC CO4020). The data collected with a 13 bit VME ADC (Mesytec MADC32) and a VME based data acquisition system. The list mode data was collected and analyzed with LAMPS software package [15].

A standard ^{60}Co (activity $= 2.5 \times 10^4$ Bq on 07/08/2018) was moved towards the CLOVER detector with BGO shielding and the data were collected at different distance from the clover detector for an equal interval of time. A CLOVER detector has four crystals and it comes with addback option where temporally coincident events could be added leading to detection of those photons which deposit its total energy in two or more crystals through compton scattering. Due to this addback option, the photo peak efficiency of CLOVER detector gets enhanced and Compton background gets reduced. Thus, addback option has been used for CLOVER detector throughout this work. The variation of intensity of 1173.2 keV line of ^{60}Co source has been plotted with distance from the front face of CLOVER detector as shown in Fig. 2.
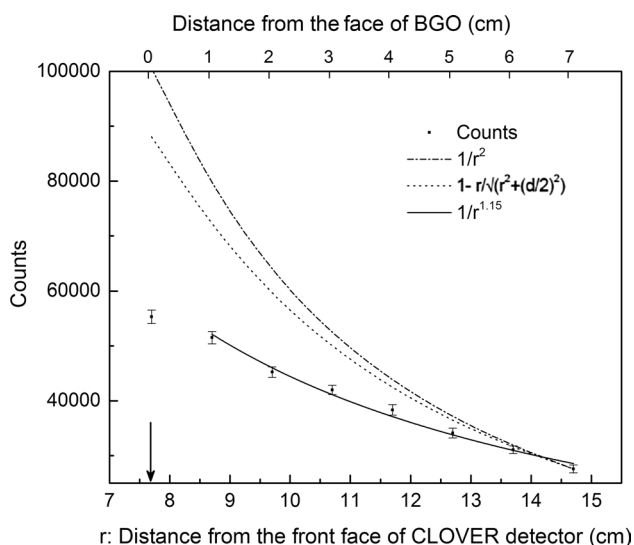
It is observed in Fig. 2 that the counts under 1173.2 keV peak inceases with decreasing distance between source and detector but the trend could not be explained either by $1/r^2$ or by introducing the geometrical efficiency factor (Ω) given in Eq. (1) as follows

$$\Omega = 2\pi \left(1 - \frac{r}{\sqrt{r^2 + (d/2)^2}} \right) \tag{1}$$

where r is the distance between the detector and the source and d is the diameter of the face of detector which is 10 cm for this CLOVER detector. Since the distance between the front face of CLOVER detector and the front face of BGO shield was 7.65 cm, shown in Fig. 1, the source could not be placed nearer to the detector. Moreover, it can be clearly observed from Fig. 2, that the photopeak counts tend to saturate when the source is placed very close to BGO shield. The possible reasons can be due to the close proximity of the radioactive source, (1) there is an increase in the number of γ-rays falling directly on the BGO, (2) there are more Compton events being registered by BGO shield, leading to higher rejection rate of actual events recorded by the crystals of CLOVER detector.

The variation of peak area under 1173.2 keV line with the distance of source from the front face of CLOVER detector without BGO shielding has been shown in Fig. 3. In this configuration, the source could be placed very close to the detector and one could get relatively much higher photopeak count compared to BGO shielded configuration when the data was taken using same source for equal duration
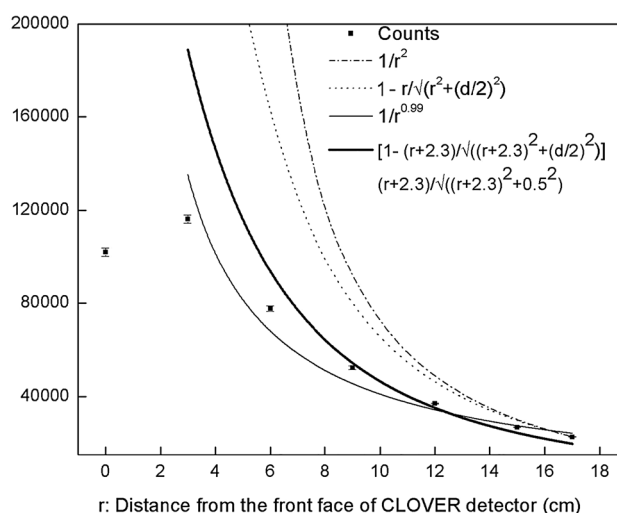

Fig. 2 Variation of intensity of 1173.2 keV γ-peak of ^{60}Co source with the distance from the front face of CLOVER detector with BGO shielding with arrow indicating the nearest position at which source could be placed

Fig. 3 Variation of intensity of 1173.2 keV line of ^{60}Co source with the distance from the front face of CLOVER detector without BGO shielding

of time. It is evident from Fig. 3 that the counts tend to decrease when the source was placed closer than 3 cm from the detector and the trend could not be explained either by $1/r^2$ or by correcting the geometrical efficiency factor (Ω) given by Eq. (1). From MCNP simulation studies [16], the distance of the detector crystals from Al end cap is found to be 2.3 cm. Considering this detector crystals to Al end cap distance and source off-set from central line, modified geometrical efficiency factor (Ω_{MG}) with distance was calculated using Eq. (2) as follows:

$$\Omega_{MG} = 2\pi \left(1 - \frac{(r+2.3)}{\sqrt{(r+2.3)^2 + (d/2)^2}} \right) \left(\frac{(r+2.3)}{\sqrt{(r+2.3)^2 + 0.5^2}} \right) \tag{2}$$

where 0.5 cm has been taken as off-axis position of the source from central line. It is observed that the photo peak counts are in reasonable agreement when modified geometrical efficiency factor is taken into account as shown in Fig. 3.

For comparing the detection efficiency in close proximity with single crystal HPGe (80%) detector, the intensity variation of 1173.2 keV γ-peak with the distance of source from the front face of HPGe detector was measured as shown in
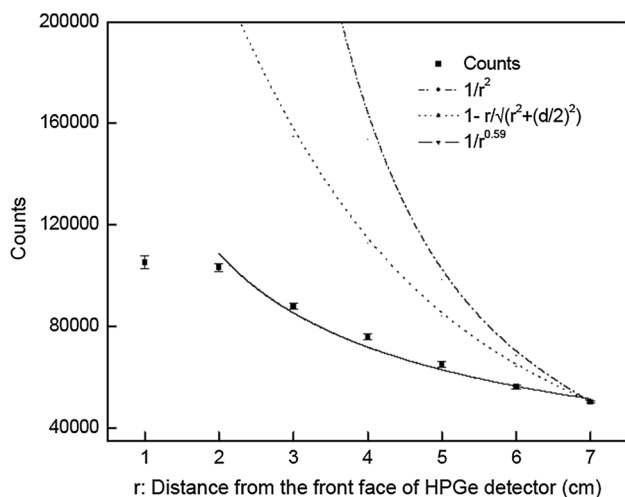

Fig. 4 Variation of intensity of 1173.2 keV line of ^{60}Co source with distance from the front face of 80% single crystal HPGe detector

Fig. 4. The source could be placed very close to the detector similar to the configuration where BGO shielding was not used with CLOVER detector. It is seen from Fig. 4 that the counts tend to saturate when the source was placed closer than 2 cm from the detector. It is expected that the photo peak count will be in agreement if modified geometrical efficiency factor is considered.

The trend followed by each detector configuration before a saturation is reached is given in Table 1. For all the three detector configurations, it has been observed that the counts tend to saturate or decrease when the source was placed closer to a certain distance and this distance is defined here as optimal position to place the source with respect to (w.r.t) front face of the detector. The optimal distance for CLOVER with BGO shielding, without BGO shielding and 80% single crystal HPGe detectors are listed in Table 1. Since the source used for each detector characterization was same and the data was taken for equal duration of time for all the three configurations, it can be seen from Table 1 that the highest photo-peak count was obtained for 1173.2 keV γ-ray peak when the ^{60}Co-source was placed at the optimal position from bare CLOVER detector (detector without BGO shielding). Since HPGe detector has 80% efficiency compared to ($3'' \times 3''$) NaI(Tl) detector and CLOVER detector has four crystals each having efficiency greater than 25%, thus higher statistics obtained for source placed at optimal distance from CLOVER detector seems justified.

A standard source of ^{152}Eu (activity $= 3.8 \times 10^3$ Bq on 09/08/2018) was placed along with ^{60}Co source (activity $= 2.5 \times 10^4$ Bq on 07/08/2018) and variation in the intensity of several γ-ray peak with distance was studied. In Fig. 5, variation in photo-peak counts of two typical γ-rays, 1332.5 keV and 121.78 keV with distance for bare CLOVER detector (detector without BGO shielding) have been shown and it is observed the the efficiency of detection is maximum at a distance of ~3 cm from the front face of the detector.

The intensity of 121.78 keV and 1173.2 keV line recorded by the individual crystal of bare CLOVER detector were added and compared with the addback value. This ratio of added to addback value is defined as addback factor and it is plotted with distance as shown in Fig. 6. A comparative study of added to addback spectra at 25 cm [17, 18] indicates a decrease in addback efficiency in a CLOVER detector.

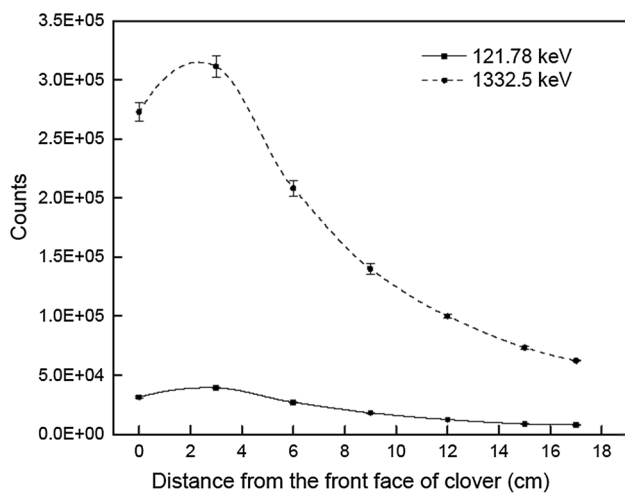| Table 1 Variation of Intensity of 1173.2 keV γ-peak with distance in close geometry configuration and optimal position for placement of source for maximum efficiency	Sl no.	Detector configuration	Optimal source position w.r.t. detector face	Intensity comparison	Intensity variation up to optimal position
	1	CLOVER with BGO shielding	~8.7 cm (1 cm from face of BGO)	0.4433	$1/r^{1.15}$
	2	CLOVER without BGO shielding	~3 cm	1	$1/r^{0.99}$
	3	HPGe single crystal (80%)	~2 cm	0.8880	$1/r^{0.59}$

Fig. 5 Variation of intensity of 121.78 keV line of ^{152}Eu source and 1332.5 keV line of ^{60}Co source with the distance from the front face of CLOVER detector without BGO shielding
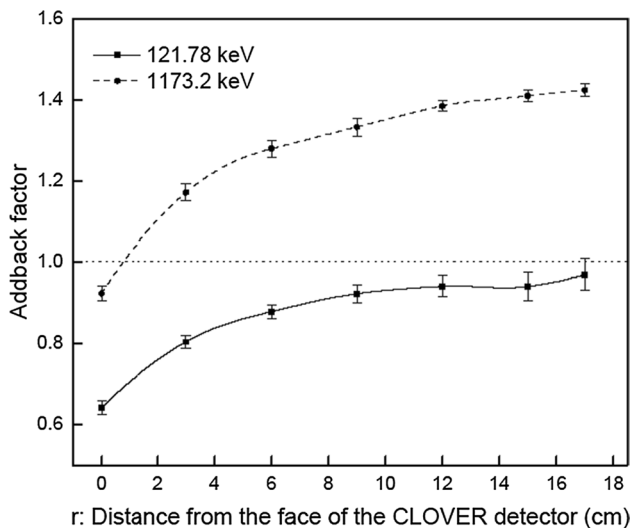

Fig. 7 Spectrum with ^{152}Eu and ^{60}Co source placed at optimal distance (~ 3 cm) from bare CLOVER detector. Sum peak of 121.78 keV and 1173.2 keV is shown in the inset

γ-ray peak of natural background ^{40}K decreases as the source is brought closer to the bare CLOVER detector. This confirms the increasing effect of dead-time when the source is placed at close proximity. Thus, the saturation or decrease in the intensity of the γ-ray peak observed in all detector configurations for bringing source closer to the detector could be qualitatively understood as summing and dead-time effect.

Resolution of the 1173.2 keV γ-ray peak was also measured and variation in resolution as the source moves closer to the CLOVER detector without BGO shielding is shown in Fig. 8. At optimal position resolution for 1173.2 keV γ-ray peak was found to be 2.6 keV. Detoriation of resolution as the source is moved closer to the detector is due to increase in count rate [19].

Fig. 6 Variation of addback factor with distance for bare CLOVER detector

Conclusions

Three different configurations, (a) single crystal HPGe (80%), (b) CLOVER HPGe with BGO shielding, (c) without BGO shielding have been studied to find an optimal configuration required for high precision decay rate measurements. The optimal position to obtain maximum intensity has been determined for each detector configuration using standard sources. Through systematic study, it was found that the source placed at optimal position from the bare CLOVER detector provides higher statistics compared to other configurations and this position remains invariant with energies. For a source placed in a close proximity to a detector, geometrical factor cannot explain the variation in efficiency when distance is calculated from front face of the detector. However considering modified geometrical reasonable

The true coincidence summing effect in a CLOVER HPGe detector has been determined as a function of sample to detector distance [16]. For both the energies, the addback factor reduces with decrease in distance indicating summing effect. Effect of summing on the photo peaks has been considered by taking the summing correction factor at different distances from Chhavi et al. [16]. The spectrum recorded at optimal distance from bare CLOVER detector shows the presence of summed peak of 121.78 keV and 1173.2 keV lines, shown in the inset of Fig. 7. The summing effect of γ-ray peaks with X rays from the source observed at 3.8 cm in an earlier work [17] has not been detected due to threshold voltage settings. It is observed that the intensity of 1460 keV
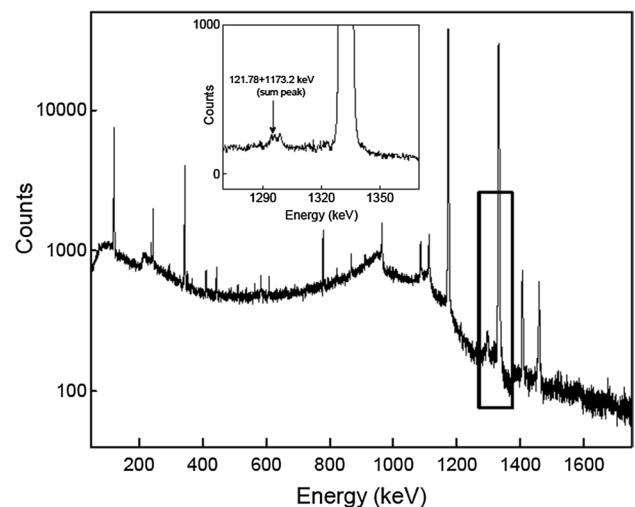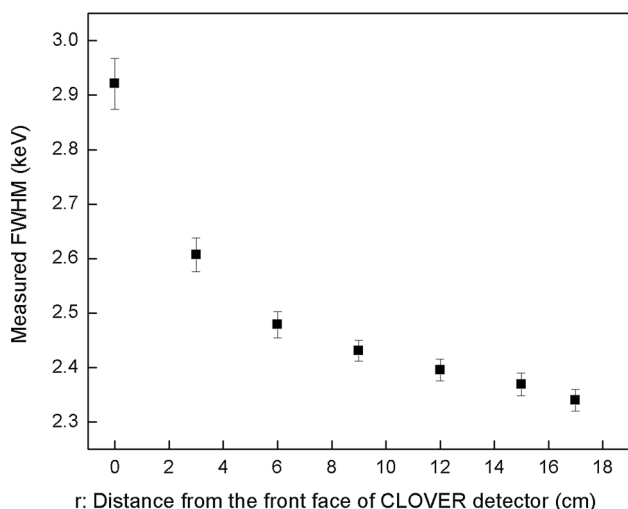
Fig. 8 Variation in energy resolution of 1173.2 keV peak with distance for bare CLOVER detector

agreement has been obtained for bare CLOVER detector. The saturation or decrease in intensity observed at closer proximity is possibly due to summing, dead-time effects and off-centering of the source. For each detector configuration, simulations studies are required for understanding the variation in detection efficiency quantitatively.

Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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