FURTHER DEVELOPMENT OF HEMISPHERICAL CdZnTe DETECTORS FOR SAFEGUARDS APPLICATIONS

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Abstract

CdZnTe (CZT) hemispherical detectors are used for international safeguard verification measurements of different nuclear materials. Small dimensions, good energy resolution and efficiency of these detectors allow to verify fresh and spent fuel assemblies in locations where difficult to access. Possibilities of further increasing of the sensitive volume/area of CZT detectors is also discussed. Spectroscopy characteristics of large and extra small volume hemispherical detectors are resulted.

1. Introduction

Presently CZT detectors of various designs and sizes are available. As it is known this material has strongly different transport characteristic of electrons and holes. The complete charge collection in detectors made of such material is difficult to accomplish. For registration of penetrating gamma radiation the best results are obtained with detectors where conditions of single charge collection are realized. One of the detector geometry where these conditions are realized is a detector with hemispherical geometry of sensitive volume. These detectors are quite simple in manufacturing and do not require special electronic circuits for their application. In practice we use a quasi-hemispherical detector shape. The sensitive volume has a rectangular parallelepiped shape with relation of the sides - AxAxA/2.

One of the commercially available detectors types are hemispherical gamma radiation detectors produced by Ritec Ltd. These are Spectrometric Detection Probes with hemispherical detectors with sensitive volumes of 10 to 20 mm^3 (SDP310/Z/20) and 60 mm^3 (SDP310/Z/60) and Large Volume CZT detectors with sensitive volume 500 mm^3 (CZT/500). These detectors are fabricated of CZT crystals produced by eV PRODUCTS.

Improvement and stabilization of crystals growing technology and technology of hemispherical detectors fabrication allowed to introduce a new commercially available detector grade with improved performance. The typical and the best energy resolutions (FWHM) at 662 keV line obtained on serial Super Grade detectors are presented in the Table1.

But there are problems when detectors with large registration efficiency are required. Those are problems related to registration of weak radiation fields, e.g. of fresh nuclear fuel (uranium and plutonium). Additionally, there are problems where detectors have to work under an extremely high radiation field in difficult to access places where it is not possible to use radiation shielding and collimators, e.g. spent fuel verification under water without its movement. For these applications detectors must have a very less registration efficiency while preservation their high spectroscopy performance. Results of recent development in these directions and results of study of the operational characteristics of hemispherical detectors are presented below.

Table 1

Detector Type	Det. Volume, mm^3	FWHM at 662 keV line and peak-to Compton ratio			
		Serial Super Grade		Best Result	
		Energy resolution, KeV	Peck-to-Compton ratio	Energy resolution, keV	Peck-to-Compton ratio
SDP310/Z/20S	5...20	≤10	≥2.5	5.8	6
SDP310/Z/60S	60	≤15	≥2.5	8	7
CZT/500S	500	≤18	≥4	9.9	6

2. Large volume hemispherical detectors

We have examined a few ways of increasing of the sensitive volume of the detector:

- increasing the single slab hemispherical detector sizes;
- combining of a few single slab detectors to single assembly - segmented detectors.

Utilized CZT crystals have rather high level of electron transport characteristic $(\mu\tau)_e$. The $(\mu\tau)_e$ values of selected CZT crystals lie in a range of $(1\div8)x10^3$ cm^3/V. Such level of electron transport characteristic allows, in principle, to make hemispherical detectors with a sensitive volume up to a few cubic centimeters.

Earlier we reported on manufacturing of detectors CZT/1500 with a sensitive volume of 1500 mm^3 /1/. These detectors have the energy resolution FWHM at a 662 keV line - 22 keV. However the serial production of such detectors appeared to be very difficult, because of considerable difficulties in the selection of CZT crystals suitable for these detectors.

An attempt was nevertheless made to create a detector with larger volume - 4000 mm^3 (size of a crystal 20x20x10 mm). But spectroscopy performance of such a detector appeared to be low, the energy resolution FWHM at a 662 keV line was insufficient and did not exceed 70 keV with peak-to-Compton ratio about 2.

The possible reason for this failure was a too low level of large CZT crystal homogeneity.

The main requirement to crystals, alongside with the high electron transport characteristics and is a high homogeneity of the transport characteristics by the detector volume and the minimal presence of structural heterogeneity. But it is difficult to realize this requirement for rather large crystal sizes. Moreover, to achieve the satisfactory energy resolution, the application of high operation voltage is required, up to 5 keV. Such high voltage in the area of a dot positive electrode can result in effects of a strong field negatively influencing the charge collection and the noise level. E.g., at external and internal radiuses of an ideal hemispherical detectors 10 mm and 0.5 mm respectively and at operation voltage 3000 V, intensity of an electrical field in the area of a small electrode will be about 63 kV/cm.

Therefore we suggest the second way of detector volume increasing – by combining of single detectors in a single assembly - segmented detectors.

There are two possible ways of combining of detectors to an assembly. First is parallel connection of single detectors working under one operation voltage and with one preamplifier. The main requirement to detectors making an assembly is identical or as close as possible values of the detector's charge collection efficiencies. The second way is connection of separate detectors working under one operation voltage, but each detector is connected to its own preamplifier with the subsequent summation of signals in the summarizing unit. In this case the use of detectors with various charge collection efficiencies is possible, as an opportunity of adjusting of a preamplifier's gain is available. However this way requires additional electronic units, is quite labor consuming in adjusting and raises the requirements in stability (temperature and in time) to the preamplifiers.

Therefore a segmented detector was built. For this detector, single detector elements with volumes 500 mm^3, sizes 10x10x5 mm, were used. Ours measurements showed that the scatter of the detector's charge efficiency made from one ingot, by the same technology does not exceed 2%. Single detectors were mounted on a one metal base and joined in parallel. Distance between slabs was 1 mm. The detectors are placed at the case with an external diameter 40 mm. The BNC connector connected the segmented detector to the preamplifier. On a Fig. 1. schematic design of segmented detectors is shown.

The appearance of segmented detector with preamplifier is shown on photograph, Fig. 2.

Spectra of Cs-137 registered by single detectors and spectrum registered by segmented detector are shown on Fig. 3.

The segmented detector has an energy resolution FWHM 23.9 keV at 662 keV line. Scatter of charge collection efficiencies of the used single detector elements did not exceeded 0.3%.

The application of the assembled detector for increase of registration efficiency in a range of rather low energies (100-300 keV) is more preferable than the use the single detector of the larger volume. As in this case the increase of the detector area will result in greater increase of sensitivity than increase of the detector thickness. The assembled detector has a larger

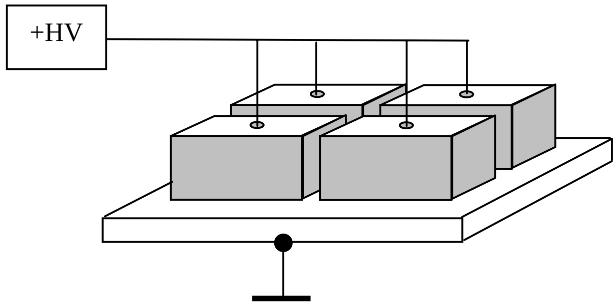

Fig. 1. Schematic design of segmented detector.

Fig. 2. Segmented detector with preamplifier.

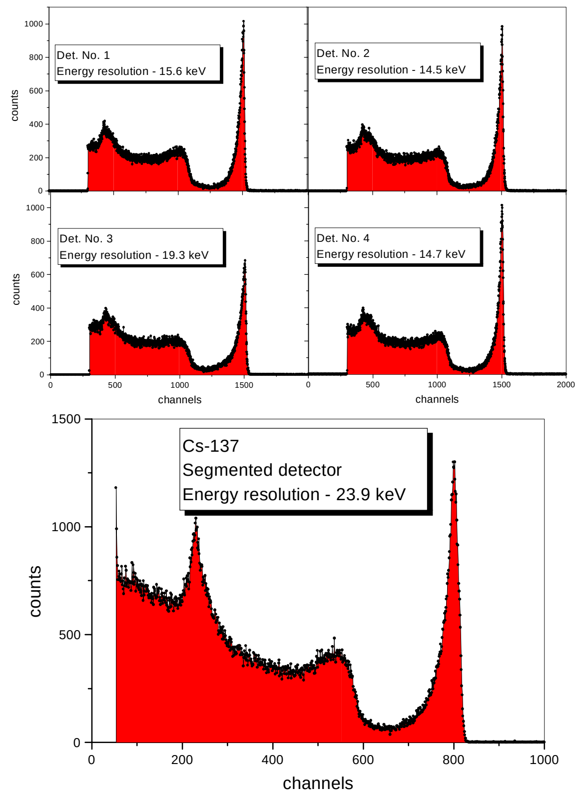

Fig. 3. Spectra of Cs-137 registered by four single detectors, volume 500 mm^3 each and spectra of segmented detector. Time of measurements is the same for all cases.

sensitive area than a large single detector with volume equal to a total volume of assembled detector. The total area of the assembled four element detector is 400 mm^2, thickness 5 mm and volume 2000 mm^3, area of the single hemispherical detector of same volume would be about 2600 mm^2 and thickness 16 mm. Joining of a few single detectors allows significantly increase registration efficiency.

Spectra of U-235 registered by four element segmented detector and by NaJ detector, ∅1"x1" (sensitive area - about 500 mm^2) at the same conditions are shown on Fig. 4.

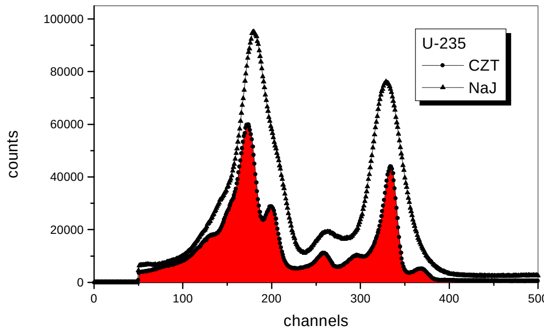

Fig. 4. Spectra of U-235
registered by segmented and NaJ detectors.

The energy resolution of segmented detector at 186 keV line is 7.8 keV.

Spectrum of Eu-152 registered by segmented detector is shown on Fig. 5.

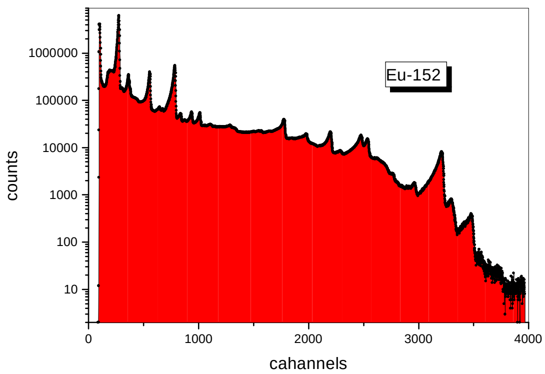

Fig. 5. Spectra of Eu-152
registered by segmented detector.

Spectrum of Ra-226 is shown on Fig. 6. As a Ra-226 source was used old wristwatch.

Measured segmented detector registration efficiency at different energy lines is shown on Fig. 7. Results of efficiencies calculation are shown on the same figure.

An opportunity of combining several standard detectors CZT/500(S) in an assembly by using a special adapter was considered too. The adapter allows parallel installation of several detectors and connection to input of the preamplifier.

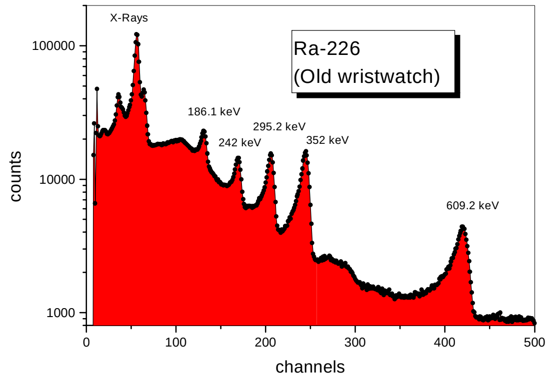

Fig. 6. Spectra of Ra-226 (old wristwatch)
registered by segmented detector.

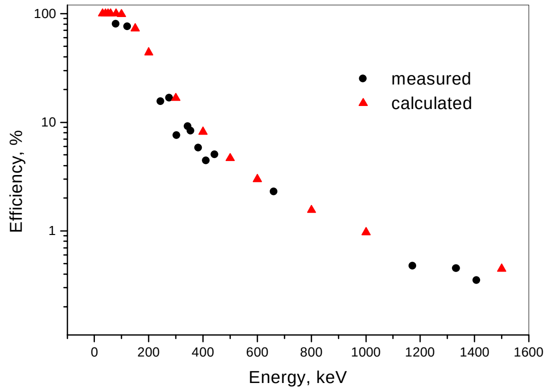

Fig. 7. Measured and calculated segmented detector
efficiency versus energy.

The design of four single CZT/500 detectors, connection adapter with the preamplifier is shown on photograph, Fig. 8.

Fig. 8. Four element of assembled detector with
connecting adapter and preamplifier.

The energy resolution of four element detectors assembly at 662 keV line is 20.3 keV. The scatter of charge collection efficiency of used detectors does not exceed 1 %.

The given way of detectors association may be useful to the users which have a lot of CZT/500(S) detector making it possible to select detectors with about equal charge collection efficiencies.

Some energy resolution degradation of the assembled detectors is related to increasing of contribution of current and capacitor noise. Capacity of the single CZT/500S detector is about 6pF, capacity of four detectors with the adapter is about 28 pF.

A detectors assembly with 6 detectors CZT/500, total volume 3000 mm^3 was also tested. But energy resolution FWHM at 662 keV line was not good, about 50 keV with peak-to-Compton ratio of 1.7. The degradation of the spectrometric characteristics is mainly related to a too big difference in charge collection efficiencies of assembly elements. Scatter of charge collection efficiency of 6 used detectors was 2.9%.

3. Small sizes hemispherical detectors

As it was mentioned already, detectors with the smallest sensitive volume are required for verification of objects having very intensive gamma radiation field. Thus the spectrometer characteristics should not considerably be worsened.

The technological process of extra small hemispherical detectors fabrication requires the large accuracy in performance of an optimum relation between the outer and central electrode sizes.

Moreover, for small detectors the maximum reachable peak-to-Compton ratio is smaller in comparison with bigger detectors. It is caused by reduction of a number of pulses in a total absorption peak connected with Compton absorption. The Compton absorption process of gamma-quanta is a multi-event process. With reduction of the detector volume probability of the repeated interaction acts of secondary gamma-quantum is decreased.

Hemispherical detectors with volume less then 5 mm^3 were investigated. Fig. 9. shows the spectrum of Cs-137 recorded by detector with volume about 4 mm^3.

Energy resolution at 662 keV line is 5.5 keV with rather good peak-to-Compton ratio of 4.4. The hemispherical detector was placed inside the standard 8 mm diameter case of SDP310 probe.

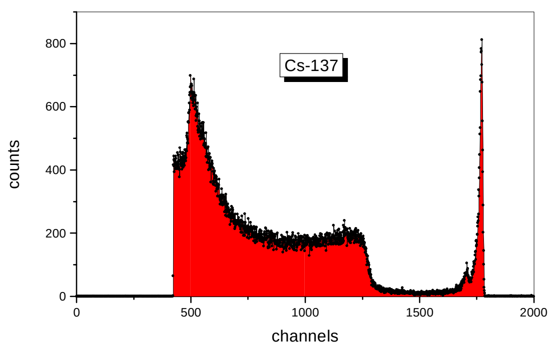

Fig. 9. Spectrum of Cs-137
registered by hemispherical detector, volume 4 mm^3.

The minimum reachable detector size was 1.2x1.2x0.6 mm (volume 0.86 mm^3). Energy resolution FWHM of this detector remained rather good 8 keV at 662 keV line, but peak-to-Compton ratio was reduced up to 3.6.

Fig. 10 shows change of ratio of total absorption peak (662 keV) area A_p to total spectrum area A_p+A_c. versus hemispherical detector volume. A_c is Compton distribution area.

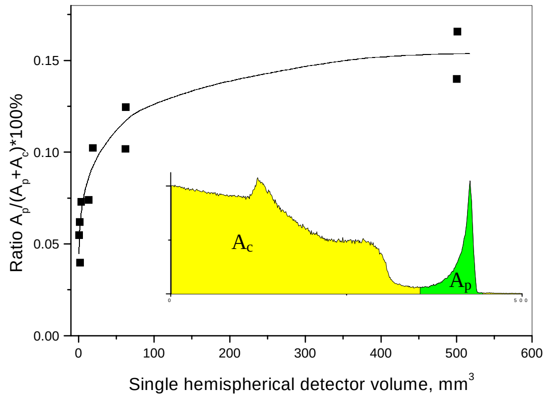

Fig. 10. Ratio of total absorption peak area to total spectrum area versus detector volume.

A strong decrease of this ratio with the detector volume reduction is observed.

The application of small detectors can be useful for registration of low-energy gamma-radiation at the presence of intensive high energy gamma radiation, on an example at registration of uranium characteristic radiation (UK_α and UK_β lines) in spent fuel. In this case reduction of detector thickness together with reduction of detector volume leads to a more essential reduction of registration efficiency in high-energy region in comparison with low-energy region. The intensity of the background Compton distribution in the low-energy region connected with registration of high-energy radiation is decreased. This leads to peak-to-background ratio increasing for low-energies lines.

Spectra of lead characteristic radiation PbK_α and PbK_β lines excited by Cs-137 for two detectors with different sizes are shown on Fig. 11.

Spectra were normalized by the 662 keV peak area. For measurement the radiation of Cs-137 was passed through a 3 mm thick lead plate. A significant relative raise of low energies lines was observed.

For these measurements detectors with a similar energy resolutions (9-13 keV) at a 662 keV line were selected.

On Fig. 12. dependence of the Pb Kα peaks (about 80 keV) area to background ratio versus detector volume is shown. The maximal value of this ratio was observed for detector size about 1.8x1.8x0.9 mm.

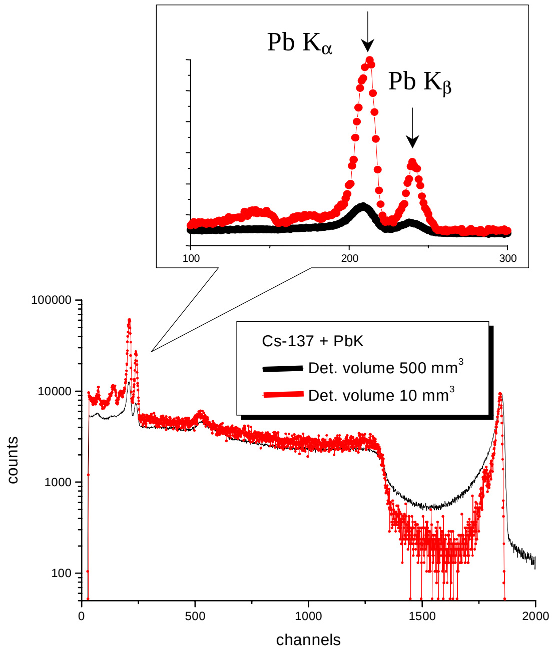

Fig. 11. Normalizes spectra of Cs-137 and Pb K lines registered by two detectors with different sizes.

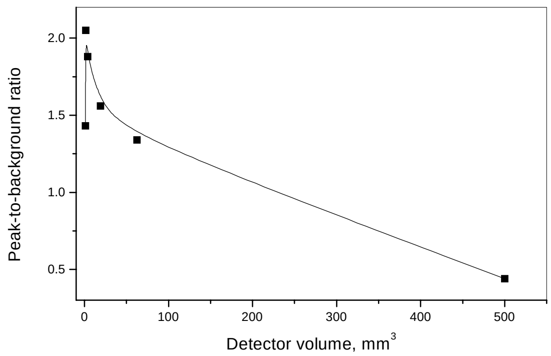

Fig. 12. Pb Kα peaks-to-background ratio versus detector volume.

4. Operation temperature range

Variations of the various hemispherical detectors spectroscopy characteristic in a range of an operation temperature -20 °C to + 40 °C were investigated. There are some possibilities of spectroscopy characteristic improvement at lowered temperatures. It may be connected with optimization of relation between electrons and holes transport properties and with reduction of current noise. For hemispherical detectors transport properties for holes must be as less as possible, transport properties as high as possible /2/.

The measurements have shown essential distinction of temperature dependencies of detectors made of different ingots. On Fig.13. dependencies of the energy

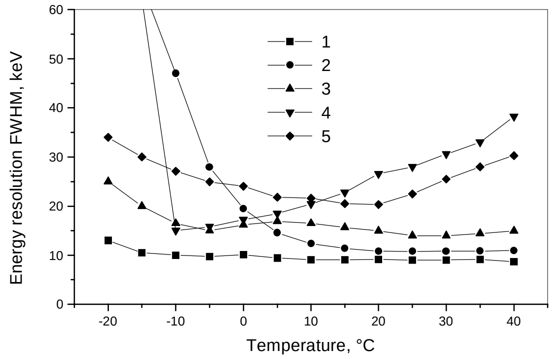

Fig. 13. Energy resolution FWHM at 662 keV line versus operation temperature. 1,2 - detector volume 10 mm^3, 3,4 - detector volume 500 mm^3, 5 - segmented detector volume 2000 mm^3.

resolution FWHM versus operation temperature of several detectors are shown.

The spectrometer characteristics of some detectors were somewhat improved at cooling down to 0 C. The best energy resolution received on the detector with the size 3x3x1.5 mm at this temperature was 5.5 keV. At further cooling the detector spectroscopy characteristic are significantly decreased. The optimum operation temperature for the majority of detectors lies in a range + 15 °C to + 30°C.

At higher temperatures deterioration of detectors spectroscopy characteristic was observed. This deterioration is connected with increase of current noise.

The deterioration of the detector spectroscopy characteristics at lowered temperatures is connected to capture of charge carriers on deep trap levels. At lowered temperature, instability in time of the spectroscopy characteristics of detectors is observed too. The essential difference in temperature dependencies of the spectroscopy characteristics of detectors made of different ingots can be connected to change of quantity and structure of deep levels of CdZnTe crystals from an ingot to an ingot.

The standard temperature operation range of serial hemispherical detectors is 0 °C ...+30 °C. For some tasks is necessary to enable operations at higher temperatures. For these purposes a special detection probes with miniature one or two-stage Peltier element was designed. This probe can operate at higher temperatures up to + 70 °C. The outside diameter of the probe case is 10 mm. Inside of probe case there are a charge sensitive preamplifier and a Peltier element with small size hemispherical detector placed on a cold side. The design of this probe is shown on photograph, Fig. 14.

The Peltier element consumption power is low about 0.3 W at maximum cooling regime.

For operation at highest (+60 °C…+70 °C) temperatures is necessary to use small size (outside diameter 14 mm) removable radiator. It is possible to use metal elements of environmental designs (e.g. of metal

Fig14. Detection probes with Peltier element.

wall of protected case used for under water measurements) as a radiator.

For power supplying and thermal stabilization a small-sized electronic unit is used. As a power 6 V battery can be used.

The given thermostabilized probe can be useful at measurements at higher temperatures, e.g. in nuclear reactor storage pool where temperatures can reached up to + 60 °C.

6. Summary and outlook

For 1999, the following major goals with respect to CZT hemispherical detectors development have been formulated:

i) Improvement spectrometric characteristic of serial CZT detectors: large volume detectors, type CZT/500s; sensitive volume 500 mm^3 and spectrometric detection probes, types SDP310/Z/20S and SDP310/Z/60S, sensitive volumes 20 mm^3 and 60 mm^3;

ii) Large volume segmented detectors, sensitive volume 2000 mm^3, sensitive area 2000 mm^2 testing and evaluation;

iii) Miniature or small size detectors, sensitive volume <5 mm^3 for operation under extra high radiation flux, testing and evaluation;

iv) Investigation of CZT detectors range of an operation temperatures;

v) Specification and design of small size detection probe with a miniature two-stage Peltier element for application under increased operation temperatures up to +70 °C.

7. Acknowledgements

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8. References

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