NEUTRON DETECTION USING CONVERSION LAYERS AND YAP:Ce AND YAG:Ce CRYSTALS

by

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The work deals with the detection of thermal neutrons using a conversion layer of LiF on YAP:Ce and YAG:Ce scintillation crystals of various sizes. Enriched LiF (95 at. % ^{6}Li) in the form of a mixture with 5 % polyvinyl alcohol was deposited on the crystal face by spraying it in a thin layer. The ^{252}Cf isotope with paraffin moderation and lead shielding was used as a neutron source. The detection is based on the nuclear reaction of ^{6}Li with the neutron. The conversion leads to emitting an alpha particle and triton according to the scheme ^{6}Li (n,)^{3}H + + 4.5 MeV which are detected by the scintillation crystal. Due to gamma ray interaction, the optimum thickness of the crystal was estimated to 70 μm.

Key word: YAP:Ce, YAG:Ce, conversion layer, indirect thermal neutron detection

INTRODUCTION

The detection of neutrons is based on the so-called conversion which is most often done via the nuclear reaction of a neutron with a corresponding nucleus being followed by the emitting of the desired particle. It can, for instance, refer to a proton, deuteron, tritium, alpha particle, fission products and/or the quanta of gamma radiation. Concerning the contribution of gamma radiation to the spectra while measuring neutrons, it is desirable to use a suitable discriminator.

The solid-state scintillation detectors of neutrons are scintillation crystals with additives such as ^{10}B, ^{6}Li or ^{157}Gd [1, 2] which have the highly efficient cross-sections for the conversion of thermal neutrons into charged particles or gamma rays. These are subsequently detected in the volume of the crystal. The next possible method of detection is to use the so-called conversion layers [3-6] which are applied in a suitable way onto a scintillation crystal. The subsequent reaction of a neutron with the given atom emits a particle which is afterwards detected in the crystal. One of the possible detection mechanisms is based on the reaction

$$^{6}\mathrm{Li}(\mathrm{n},\alpha)^{3}\mathrm{H} \quad 4.6\,\mathrm{MeV} \qquad (1)$$

The aim of the work is to verify the hypothesis of simple detection of thermal neutrons using the conversion layer formed with lithium fluoride enriched with ^{6}Li isotope applied on the head of selected inorganic crystals in the form of the mixture of a 5 % water solution of polyvinyl alcohol.

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EXPERIMENT

Devices and equipment

The neutron detection was performed by inorganic scintillation crystals produced and supplied by Crytur Lmtd, Czech Republic; specifically, cerium doped yttrium aluminum perovskite (YAP:Ce) [7] and cerium doped yttrium aluminum garnet (YAG:Ce) [8] of a cylindrical shape with the sizes shown in tab. 1. The conversion layer consisted of enriched LiF (95 at. % ^{6}Li) and 5 % water polyvinyl alcohol solution (trademark Sloviol R).

All measurements were conducted in a light-tight chamber with an inbuilt 5.08 mm (2") photomultiplier (PMT) type 9266KB, (ET Enterprises, GB), which was connected to the base digiBase (Ortec, USA), and the subsequent evaluation of acquired counts was conducted with GammaVision v. 6.07 (Ortec, USA) software.

Table 1. Size and active volume of the used crystals

Crystal size	Type		
	YAP:Ce	YAP:Ce	YAG:Ce
Diameter [mm]	25.4	10	10.9
Thickness [mm]	34	0.1	0.2
Volume [mm^3]	17300	7.9	18.7

As a neutron source, ^{252}Cf with the activity of $A = 37$ MBq (1 mCi) on January 1st, 2015, and a calculated neutron flow of 3.7 10^6 1/s with 10 cm paraffin moderation and 1 cm lead shielding, was used. Point sources type EG3 ^{241}Am, ^{137}Cs, and ^{60}Co (all manufactured by CMI-IIZ, Prague) were chosen for gamma calibration. Alpha calibration was performed by an ^{241}Am source produced by electro-deposition techniques [9] on a thin stainless steel disc. For higher energy calibration than the energy lines of the used sources, linear extrapolation was assumed.

The basic setting of electronic instrumentation was as follows: the number of channels of the multichannel analyzer – MCA unit was given by unchangeable factory settings with the value of 1024, the gross gain had the value of 1, the shaping time was 0.75 μs and the LLD was set on the value of the 5th channel. The optimal setting of the parameters was set for each crystal separately, as shown in tab. 2. The basic criterion for choosing the said setting was the formation of an energetic spectrum of the detected particles with a suitable range.

Table 2. Setting parameters of the base digiBase

Type	Voltage [V]	Gain	Measurement time [s]
YAP:Ce[(a)]	600	1.0	1000
YAP:Ce[(b)]	800	1.0	10000
YAG:Ce	800	1.2	9000

Note: [(a)]crystal of volume of 17300 mm^3, [(b)] crystal of volume of 7.9 mm^3

MEASUREMENT METHODOLOGY

The selected crystals were applied with a conversion layer. The coating was conducted by Envinet PLC, via the method of repeated spraying with a nozzle, 0.3 mm in diameter. Proper homogenization was achieved by modifying the ZnS:Ag coating process, the method verified by Envinet for the preparation of particle sensitive scintillators. The thickness of the layer was determined by a high-performance ultrasonic thickness gauge.

The prepared crystal was placed on the window of the photomultiplier (PMT). To ensure a better optical contact between crystal and window, glycerin was used. The neutron source was placed upon a 6 mm thick, low-density polyethylene pad placed on a 15 cm thick hollow polystyrene base. The measurement was conducted at preset voltage and time. The counts spectrum measured this way was marked as non-moderated and, subsequently, the measurement repeated with 10 cm paraffin moderation, a 5 cm hollow polystyrene base and a 6 mm thick lead beaker which provided a distance of approximately 15 cm, as in the previous case (fig. 1).

Alpha and gamma radiation calibration was performed using above-mentioned sources in the same geometry as the one used for energy calibration, *i*. *e*., at a distance of the pad of 1.5 mm. The tritons/protons calibration has not been done, due to the absence of a suitable source. Therefore, their position in the spectrum has been estimated according to the values of the function dE/dx. To confirm the measured results, a set of measurements with a 2 mm thick hollow cadmium cylinder containing the neutron source and, then, measurements with Bonner spheres of various radii containing the same neutron source [10], were conducted. The measurement of the background counts of both the crystal and separate PMT without a crystal was conducted on the day of measurement to eliminate possible further interferences in the counts spectra. To improve the rate of photons striking the photocathode PMT, as well so as to increase the distinguishing properties, a bigger crystal, YAP:Ce, was coated with a reflective layer fashioned out of a teflon tape. After finishing the measurements stated above, the layer of lithium fluoride was washed off with methanol and the same measurement performed again, but without the lithium layer at the face of the crystals. The conditions and setting were identical to (tab. 2) those of the lithium layer.

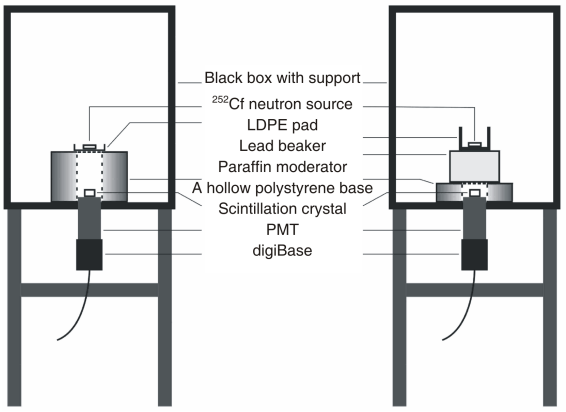

Figure 1. Schematic drawing of the geometrical layout of the measurement

RESULTS AND DISCUSSION

To verify the detection of neutrons, the spectra with and without a conversion layer were compared. A significant obstacle to the analysis can be attributed to the fact that it was impossible to eliminate the interfering gamma radiation from a neutron source and the induced radiation from the surroundings. This resulted in the loss of all particles with lower energies resulting from the attenuation in the conversion layer and the dead layer of the crystal. Due to this, very thin planar detectors with a minimal cross-section for gamma rays are relevant in this type of detection.

Our measurements have proved that an applied conversion layer absorbs the majority of formed alpha

particles and that they cannot be reliably identified in recorded counts. On the other hand, it is possible to calculate that the range of an alpha particle with the energy of 2 MeV lithium fluoride in a mixture with polyvinyl alcohol amounts to approximately 25 μm. The method of repeated coating has proved difficult if the desired uniform thickness of the conversion layer is to be achieved, making it therefore unsuitable for subsequent alpha particle detection. For instance, the application of a layer using a deposition from the gaseous state or the use of physical vapor deposition (PVD) [11, 12] seems to be a more viable option. In these cases, thicknesses from 2 μm to 10 μm can be achieved. The disadvantage of this method of coating of a conversion layer lies in the high temperature during its application (150 °C-1000 °C) which can lead to the damage of the crystal lattice and possible ignition and explosive flare of the formed dispersions. A triton interacts with the material in a way similar to alpha radiation; however, after a given period, triton disintegrates into a proton and two neutrons [13, 14], with each of these particles carrying a part of its original energy. It can thus be calculated that a separate triton (proton) interacting with the conversion layer achieves a wider range than an alpha particle and, due to this, unlike most alpha particles, manages to pass through the conversion layer into the detection volume of the crystal.

As observed in fig. 2, the region of higher counts with the YAP:Ce crystal of an active volume of 17300 mm^3 extends to the energy of up to approximately 9 MeV. This is caused by the summation of gamma radiation and the contribution of formed alpha particles and tritons in lower energies. Therefore, they are not positively identified. The cause of the summation is the high activity of a neutron source and, possibly, the partial unsuitability of the signal processing via digiBase, due to electronics not designed for such extreme high-count rates as the ones they were exposed to. The change in the exponential curve of the

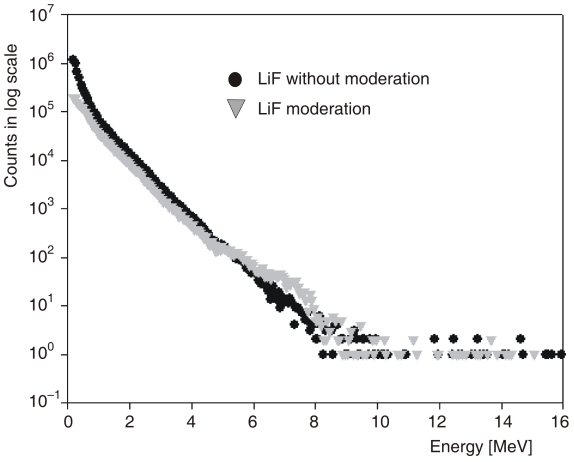

Figure 2. Cf counts spectrum with moderation and without moderation; 17300 mm^3 (1") crystal YAP:Ce

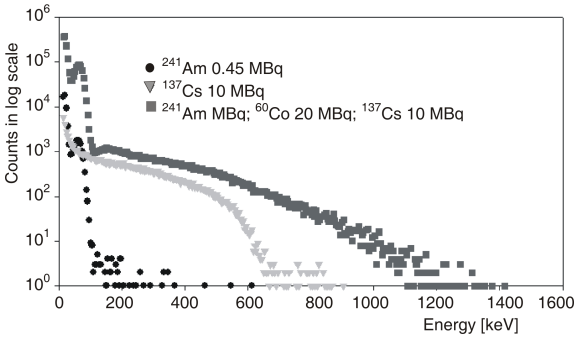

Figure 3. The influence of high photon flux on the summation effect in the counts spectrum of YAG:Ce 200 μm

counts spectrum of LiF moderation can be attributed to the gamma radiation interference originating from the shielding.

The influence of the count rate and the summation on the shape of the counts spectrum is partially represented in fig. 3 where the gamma sources of various energies and activities were measured. In this case, the measurement was conducted with the crystal YAG:Ce with the thickness of 200 μm and it can be observed that the increase of activity of separate sources is related to the increase in the number of summation pulses.

Considerably different results were measured with crystals of the same composition, but of smaller sizes (tab. 1). According to fig. 4, it can be observed that the conversion layer does not significantly affect the shape of the counts spectrum of a non-moderated source. However, a significant difference is observed in fig. 5 which represents the comparison of the moderated source spectra.

The increase of the count rate in the entire counts spectrum with a conversion layer is observed. Concerning the fact that the measurements were conducted under the same conditions, the only difference being the presence or absence of a conversion layer

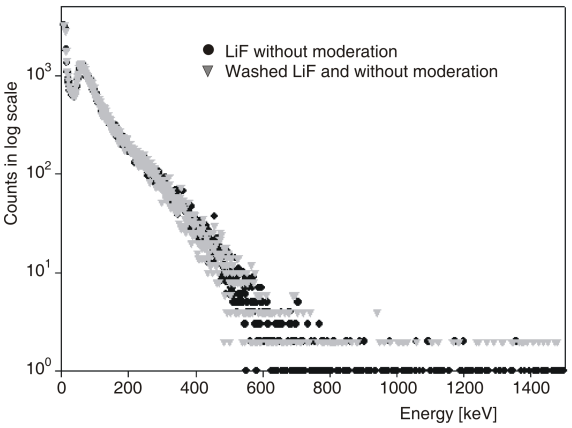

Figure 4. ^{252}Cf counts spectrum without moderation; crystal YAP:Ce 100 μm

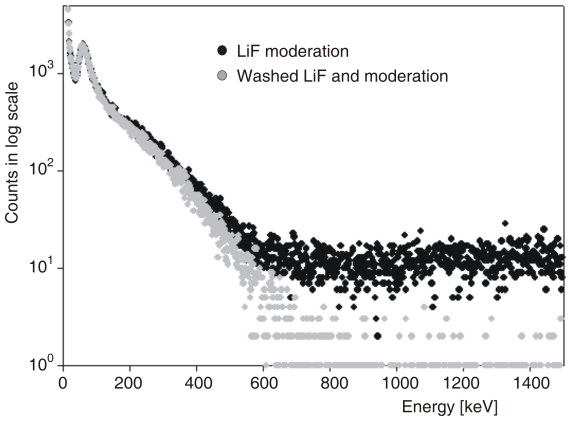

Figure 5. ^{252}Cf counts spectrum moderated with paraffin and lead; crystal YAP:Ce 100 μm

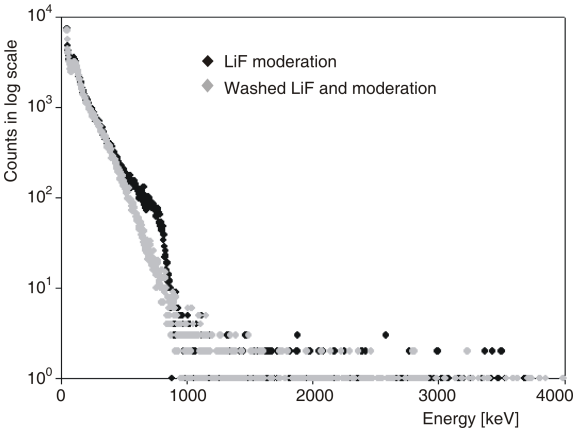

Figure 6. ^{252}Cf counts spectrum moderated with paraffin and lead; crystal YAG:Ce

and, due to the fact that the contribution of the formed alpha particles to the counts spectrum is, due to the influence of self-absorption very small, the ingrowths can be attributed to the tritons formed in said reactions (1).

In the case of the YAG:Ce crystal, no significant difference in non-moderated counts spectra is observed. The different shapes of counts spectra with and without a conversion layer (fig. 6) while measured with paraffin moderation can be attributed to the contribution of tritons. Concerning the double thickness of the YAG:Ce (200 μm) crystal in comparison with the YAP:Ce (100 μm) crystal, the thicker one is in its detection efficiency capable of capturing gamma radiation at higher energies. This was observed in the counts spectrum in the superposition of counts originating from gamma radiation and tritons.

CONCLUSIONS

The use of conversion is generally one of the most commonly used ways of neutron detection. In solid-state scintillation spectrometry, mainly crystals containing a certain percentage of lithium, boron, and gadolinium isotopes are used [15, 16]. The main problem of inorganic scintillation crystals is their relatively high sensitivity to gamma radiation, which increases the demands on the instrumentation set-up and the evaluation of measured results.

While using neutron conversion layers, it is inevitable to consider two significant factors: the thickness of a crystal and the nature of the conversion layer. It can be said that the thinner crystal is more sensitive to the lower energy gamma radiation and charged particles absorption and vice versa. Due to this, the crystals with lower thicknesses are used to detect X-ray radiation, low-energy gamma radiation and charged particles.

The conversion layer should be applied in a thickness of about 30 μm, in such a manner that its surface ensures maximum homogeneity, without bubbles or other defects. Proper homogenization was achieved by modifying the ZnS:Ag coating process, the verified method used being in Envinet, so as to prepare particle sensitive scintillators. The thickness of the layer was determined by a high performance ultrasonic thickness gauge.

Based on the measured results, the significance of crystal thickness is critical when the increase of approximately100 μm leads to the increase in the absorption of gamma radiation to such an extent that it causes a significant interference of gamma radiation counts to triton counts in the spectrum, otherwise possible to be determined with a thinner crystal. With crystals of bigger thickness, the positive detection of neutrons is impossible due to their ability to absorb gamma radiation at higher energies. Furthermore, the significance of energy summation dramatically grows as the summation hides the contribution in the counts spectrum formed by charged particles.

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AUTHOR CONTRIBUTIONS

The idea for the study was put forward by L. Fiserova. The measurements and theoretical calculations were carried out by L. Fiserova and J. Janda. The data were evaluated and interpreted by D. Sas.

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Луција ФИШЕРОВА, Јиржи ЈАНДА, Данијел САС

ДЕТЕКЦИЈА НЕУТРОНА ПРИМЕНОМ КОНВЕРЗИОНИХ СЛОЈЕВА И YAP:Ce И YAG:Ce КРИСТАЛА

У овом раду бавимо се детекцијом термичких неутрона употребом конверзионог LiF слоја на YAP:Ce и YAG:Ce сцинтилационим кристалима различитих величина. Обогаћени LiF (95% атома ^{6}Li) у смеши са 5 % поливинил алкохолом нанет је спрејом на кристал у танком слоју. Као неутронски извор коришћен је радионуклид ^{252}Cf са парафинским модератором и оловном заштитом. Детекција се заснива на нуклеарној реакцији ^{6}Li са неутронима. Конверзија води ка емитовању алфа честице и трицијума према релацији $^6\mathrm{Li}(n,\alpha)^3\mathrm{H} + 4.5$ MeV, који се детектују сцинтилационим кристалом. Имајући у виду интеракцију гама зрачења, процењено је да оптимална дебљина кристала износи 70 μm.

Кључне речи: YAP:Ce, YAG:Ce, конверзиони слој, индиректна детекција термичких неутрона