Nuclear Instruments and Methods in Physics Research A 652 (2011) 319–322

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research A

journal homepage: www.elsevier.com/locate/nima

Thermal neutron detection with Ce^{3+} doped $LiCaAlF_6$ single crystals

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ARTICLE INFO

Available online 5 March 2011

Keywords:
Neutron detection
Neutron/gamma discrimination
Intrinsic gamma-neutron detection efficiency

ABSTRACT

Cerium-doped $LiCaAlF_6$ (Ce:LiCAF) crystals have been studied as scintillators in application to thermal neutron detection. Three crystals: high-doping Ce:LiCAF, low-doping Ce:LiCAF with 50% enrichment of ^6Li (both 10 mm × 10 mm × 2 mm, rectangular) and high-doping Ce:LiCAF with 95% enrichment of ^6Li (Ø50.8 mm × 2 mm, discus) coupled to Photonis XP5300B PMT, were tested. The response of these crystals to neutrons emitted from a paraffin moderated $^{238}PuBe$ source has been investigated. Thermal neutron peaks have been found at a Gamma Equivalent Energy (GEE) of ~2.5 MeV for high-doping Ce:LiCAF (50% ^6Li), ~2 MeV for low-doping Ce:LiCAF (50% ^6Li) and ~1.9 MeV for high-doping Ce:LiCAF (95% ^6Li). The light output of Ce:LiCAF was also measured (175–250 phe/MeV from sample to sample). Lithium-6 glass GS20 from Saint Gobain was used as a reference scintillator (Ø50 mm × 2 mm, circle). Relative neutron efficiency, normalized to that of GS20 lithium glass, as well as gamma-neutron intrinsic efficiency for all tested samples was calculated. Intrinsic efficiency on thermal neutron detection for small Ce:LiCAF samples was estimated at about 32–35% of that of GS20 and for large Ce:LiCAF sample as about 82% of that of GS20.

1. Introduction

Neutron detection techniques are essential in the case of border monitoring against illegal smuggling of nuclear materials. Commonly used helium detectors are likely to be replaced in the near future due to the lack of ^3He in world market. Therefore, researcher's community is looking for new thermal neutron detectors that will be based on scintillation crystals. Cerium-doped $LiCaAlF_6$ (Ce:LiCAF) single crystals are sensitive to thermal neutron detection because of the presence of ^6Li, which has a high thermal neutron capture cross-section of 940 b. This fact makes Ce:LiCAF an attractive proposal for border monitoring. Generally, a neutron scintillator is characterized by a low stopping power for gamma-rays in order to discriminate thermal neutrons from gamma-ray background. For this reason, the host lattice of neutron scintillator should constitute of light elements. LiCAF meets these requirements as it has density below 3 g/cm^3.

The main aim of the present work is to study gamma-ray discrimination and to estimate the efficiency of thermal neutron detection for Ce:LiCAF samples:

- high-doping Ce:LiCAF 10 mm × 10 mm × 2 mm, with 50% enrichment of ^6Li,
- low-doping Ce:LiCAF 10 mm × 10 mm × 2 mm, with 50% enrichment of ^6Li,
- high-doping Ce:LiCAF ∅50.8 mm × 2 mm, with 95% enrichment of ^6Li.

Results obtained with Ce:LiCAF samples are compared to lithium-6 glass GS20 (∅50 mm × 2 mm) from Saint Gobain with 95% content of ^6Li.

Table 1 presents a comparison of basic properties of Ce:LiCAF and GS20 lithium glass.

Lithium-6 based scintillators are sensitive to thermal neutrons, which utilize the nuclear reaction of ^6Li. Thermal capture reaction with ^6Li nucleus is as follows:

$$^6_3Li + ^1_0n \rightarrow ^4_2He + ^3_1H + Q\,(4.78\text{ MeV}) \quad (1)$$

and the resulting charged particle spectrum corresponds to neutrons registered in the detector. The energy Q released in reaction (1) is determined and divided between the alpha and the triton particles. The method of growing of Ce:LiCAF crystals and their optical and scintillation properties have been reported in the paper from the group at Tohoku University and Tokuyama Corporation [1].

2. Experimental details

The XP5300B Photonis photomultiplier (PMT) was used due to its high blue sensitivity (up to 14.8 μA/lmF) and high quantum efficiency (about 40%). This was necessary because Ce:LiCAF samples have a very low light output (~175–250 phe/MeV depending on the sample). Experimental details are shown in Fig. 1. This simple setup

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doi:10.1016/j.nima.2010.09.182

consists of a preamplifier (Canberra 2005E) and an amplifier (Canberra 2022). The signals from scintillators were recorded by the Tukan8k Multi-Channel Analyzer [2]. Each sample was coupled with a photomultiplier by silicon grease and coated around and on the top by several layers of Teflon® tape to enhance light collection.

The estimation of the number of photoelectrons (phe) per energy unit (phe/MeV) in the case of Ce:LiCAF crystals was done by using the Compton edge of gamma-rays from ^{137}Cs source and in the case of lithium glass GS20—by using the full energy peak from ^{137}Cs (Bertolaccini method [3]). The response to thermal neutrons was measured with the ^{238}PuBe source ($\sim 5\times 10^5$ n/s) put into a paraffin ball and shielded by 5 cm of lead (against high energy gamma-rays mainly from the ^{238}PuBe source).

3. Results

3.1. Photoelectron yield

The first step of Ce:LiCAF study was to estimate the photoelectron yield as a function of shaping time constant set in an amplifier (Fig. 2). It is seen that the photoelectron yield is independent of the shaping time and we can conclude that Ce:LiCAF crystals have a short pulse decay time for gamma-rays. On the other hand, the reference scintillator, GS20 lithium glass reveals slight decrease in the photoelectron yield for short shaping time constants (see Fig. 3). However we have to remember that measurements made at very high gamma-ray rates require short shaping time constants to minimize the build-up of pile-up events, especially in a neutron detection window. This is an important issue in the case of border monitoring, where we expect masking of neutron source high-activity gamma-rays.

Table 1
Basic properties of Ce:LiCAF and GS20 lithium glass.

Scintillator	Wavelength of emission maximum (nm)	Decay time (ns)	Density (g/cm^3)
Low-doping Ce:LiCaF 10 mm × 10 mm × 2 mm	300	~40	2.94
High-doping Ce:LiCaF 10 mm × 10 mm × 2 mm	300	~40	2.94
High-doping Ce:LiCaF ∅50.8 mm × 2 mm	300	~40	2.94
GS20 lithium glass ∅50 × 2	395	~75	2.5

The number of photoelectrons per energy unit (phe/MeV) was obtained for all of the scintillators. The average values are shown in Table 2. Ce:LiCAF crystals are characterized by a very low light yield from gamma-rays, but for neutron scintillators it may not be a disadvantage, provided that they have considerably higher light yield for thermal neutron capture (which is a condition for good neutron/gamma discrimination).

3.2. Neutron response

The tested Ce:LiCAF scintillators are designed for thermal neutron detection; therefore for neutron measurements we used a paraffin moderated ^{238}PuBe source, shielded by 5 cm of lead bricks to reduce the flux of 4.4 MeV gamma-rays following the reaction in the ^{238}PuBe source. The source emitted neutrons up to about 11 MeV with an intensity maximum in the 3–5 MeV range [4]. A further test was performed to demonstrate the tested crystals in gamma-rays from ^{60}Co source. The results are presented in Fig. 4 for Ce:LiCAF crystals and Fig. 5 for the reference lithium glass GS20. The continuous spectrum on the left of the neutron peak corresponds to the gamma-ray background. For Ce:LiCAF samples neutron peaks appear at a Gamma Equivalent Energy (GEE) of 2 MeV for low-doping Ce:LiCAF

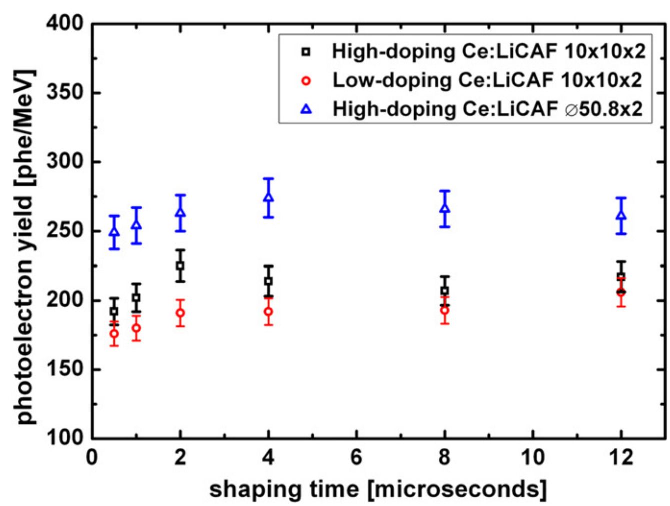

Fig. 2. Number of photoelectron per energy unit as a function of shaping time constant for tested Ce:LiCAF crystals. In principle, for Ce:LiCAF crystals photoelectron yield is independent of the shaping time.

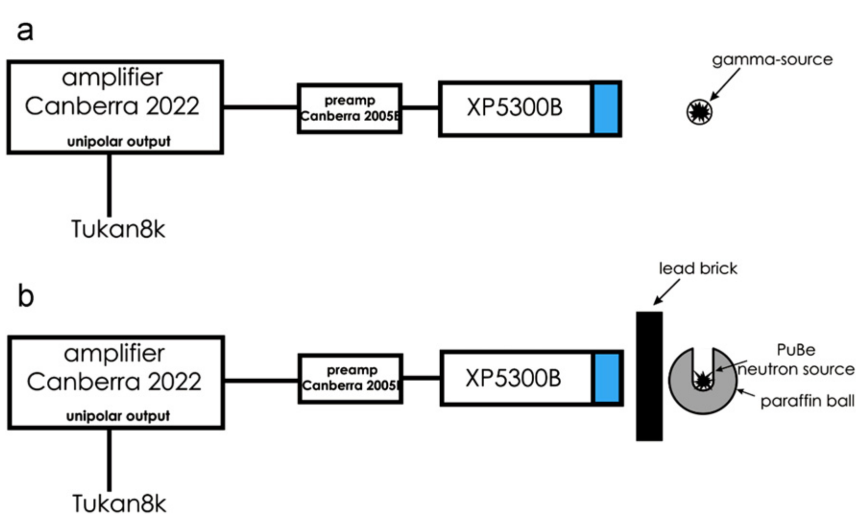

Fig. 1. Diagram of the experimental setup: (a) for gamma measurements, and (b) for neutron measurements.

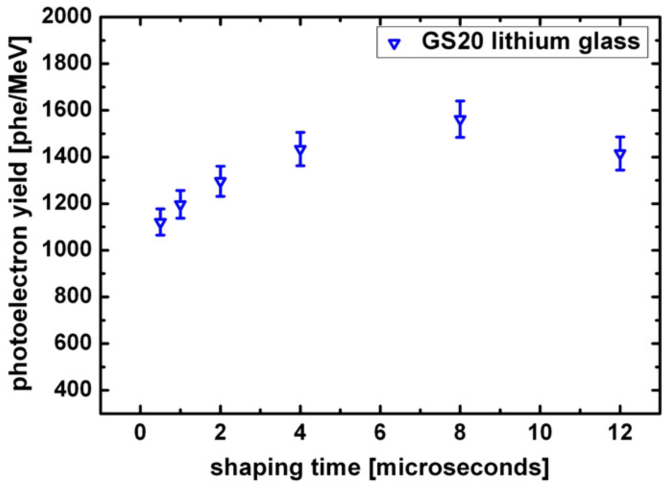

Fig. 3. Photoelectron yield as a function of shaping time constant set in amplifier for a reference GS20 lithium glass.

Table 2
Number of phe/MeV for all tested scintillators.

Scintillator	Number of phe/MeV
Low-doping Ce:LiCaF 10 mm × 10 mm × 2 mm	191 ± 10*
High-doping Ce:LiCaF 10 mm × 10 mm × 2 mm	225 ± 11*
High-doping Ce:LiCaF ∅50.8 mm × 2 mm	254 ± 13*
GS20 lithium glass	1296 ± 65*

* Shaping time $\tau = 2$ μs.

10 mm × 10 mm × 2 mm, 2.5 MeV for high-doping Ce:LiCAF 10 mm × 10 mm × 2 mm and 1.9 MeV high-doping Ce:LiCAF ∅50.8 mm × 2 mm.

The distribution of thermal neutron events in Fig. 4 has the FWHM of about 16% for high-doping Ce:LiCAF 10 mm × 10 mm × 2 mm, 18% for low-doping Ce:LiCAF 10 mm × 10 mm × 2 mm and 28% for high-doping Ce:LiCAF ∅50.8 mm × 2 mm. Despite the fact that neutron peaks are very broad, in the case of both 10 mm × 10 mm × 2 mm samples, neutron/gamma separation is good. Only in the case of large sample of Ce:LiCAF the neutron peak is partly covered by the Compton continuum from ^{60}Co. But, as it will be mentioned later, this sample has better efficiency for thermal neutron detection than small samples, because of a higher content of ^{6}Li. For the reference lithium glass GS20 resolution of neutron peak is significantly better than for Ce:LiCAF samples ($\Delta E/E = 7.35\%$, see Fig. 5), mainly due to a larger photoelectron yield, but the neutron/gamma separation is worse (GEE for GS20 is about 1.5 MeV) and neutron peak is slightly covered by the Compton continuum from ^{60}Co source.

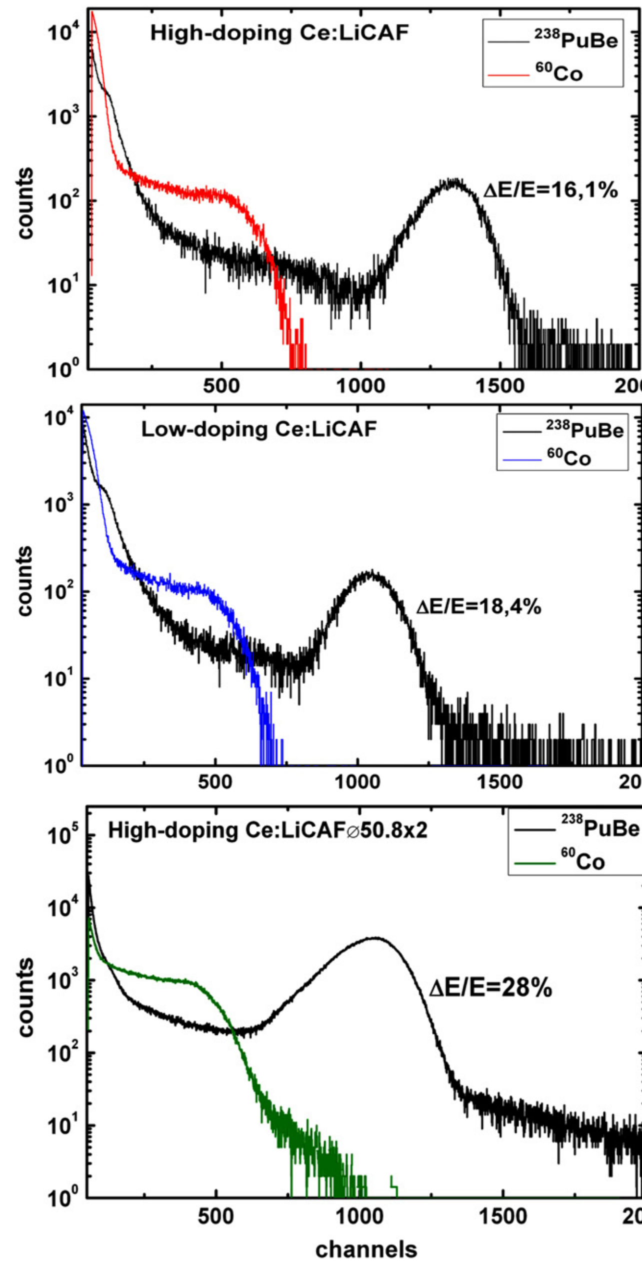

Fig. 4. Neutron response of Ce:LiCAF samples: high-doping Ce:LiCAF 10 mm × 10 mm × 2 mm (upper spectrum), low-doping Ce:LiCAF 10 mm × 10 mm × 2 mm (middle spectrum) and high-doping Ce:LiCAF ∅50.8 mm × 2 mm (lower spectrum).

3.3. *Intrinsic neutron detection efficiency and gamma-neutron intrinsic efficiency*

Intrinsic neutron efficiency for Ce:LiCAF samples was estimated. Moderated ^{238}PuBe source was placed at a distance of 66 cm from the face of the center of detector. The results for tested Ce:LiCAF crystals are normalized to those of the reference GS20 lithium glass (see Table 3).

Intrinsic efficiency for thermal neutron detection by the scintillator $\varepsilon_{thermal}$ is given by the formula

$$\varepsilon_{thermal} = 1 - e^{-n\sigma x} \quad (2)$$

where n is the number of ^{6}Li atoms per cm^3, σ is the cross-section for ^{6}Li on thermal neutron capture and x is the thickness of the scintillator.

Considering 2 mm thick samples, quotient of thermal neutron efficiency for Ce:LiCAF to GS20 will be 60%:95% according to the formula (2), whereas we observed a ratio of 35%:100%, which is close to the ratio of the ^{6}Li content in Ce:LiCAF 10 mm × 10 mm × 2 mm to GS20. This means that $n\sigma x \ll 1$, so we can assume that $\sigma \ll 940$ b. It results from the fact that the neutron flux from the ^{238}PuBe source after passing the paraffin is not fully thermalized, which agrees with our expectations.

Measurements with an intense ^{137}Cs radionuclide source were also performed. Using the gamma source we increased the ambient background to 10 mR/h (according to ANSI standards [5]) and we checked how many events from gamma-rays occur in the neutron window (*gamma-neutron intrinsic efficiency* $\varepsilon_{\gamma n}$, according to Ref. [6]). The gamma-neutron intrinsic efficiency measures the response of a neutron detector to gamma-ray field when no neutron source is present. This is a very important requirement

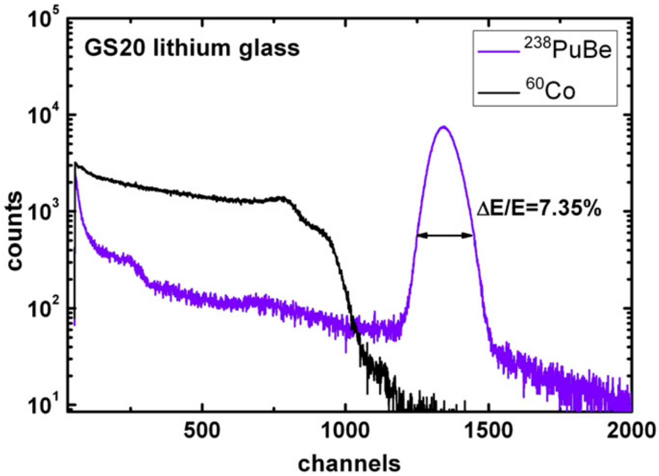

Fig. 5. Neutron response of GS20 lithium glass. Neutron peak is partly overlapped by ^{60}Co Compton continuum.

Table 3
Intrinsic neutron detection efficiency for tested scintillators.

Scintillator	Number of ^{6}Li atoms per cm^{3}	Intrinsic thermal neutron efficiency (%)
GS20 ∅50 mm × 2 mm	$\sim 1.58\times10^{22}$	100
High-doping Ce:LiCAF 10 mm × 10 mm × 2 mm	$\sim 0.5\times10^{22}$	35 ± 5
Low-doping Ce:LiCAF 10 mm × 10 mm × 2 mm	$\sim 0.5\times10^{22}$	32 ± 5
High-doping Ce:LiCAF ∅50.8 mm × 2 mm	$\sim 1\times10^{22}$	82 ± 5

for neutron detectors, specifying their sensitivity to gamma-rays. The value of $\varepsilon_{\gamma n}$ should be very small, typically $\varepsilon_{\gamma n}\leq 10^{-6}$. Laboratory background was subtracted from all the spectra. In these conditions, gamma-neutron intrinsic efficiency for all tested scintillators is kept below 10^{-7}.

4. Conclusions

Samples of Ce:LiCAF were studied as scintillators for thermal neutron counting. Crystals detected the neutrons emitted from the ^{238}PuBe source, which were subsequently thermalized by paraffin. Ce:LiCAF crystals are characterized by low photoelectron yield for gamma-rays, which is not a disadvantage until neutron/gamma discrimination is good. Consequently, high neutron/gamma discrimination was observed, as well as very small gamma-neutron intrinsic efficiency, which is one of the parameters defining neutron detectors used in Radiation Portal Monitor in Homeland Security. Intrinsic neutron detection efficiency for Ce:LiCAF crystals was evaluated, where we used Saint Gobain GS20 lithium glass as a reference. Results were on the level of 32–35% of those of GS20 for small samples, because they have less content of ^{6}Li in lithium ions than GS20. In the case of a large sample of Ce:LiCAF, with 95% content of ^{6}Li, the intrinsic efficiency is better than for small samples (82% of GS20). Ce:LiCAF scintillators have properties, that make them an attractive proposal for high efficiency thermal neutron detectors.

Acknowledgements

This study was supported in part by the International Atomic Energy Agency, Research, Contract no. 14360 and by EU Structural Funds Project no POIG.01.01.02-14-012/08-00 and by Polish Ministry of Science and Higher Education, Grant no. 657/W-IAEA/2010/0.

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