LiCaAlF₆ scintillators in neutron and gamma radiation fields

L. Viererbl*, V. Klupák, M. Vinš, M. Kolečka and J. Šoltés

*Research Centre Rez Ltd., Hlavní 130,
Husinec-Řež, 25068, Czech Republic
ladislav.viererbl@cvrez.cz

A. Yoshikawa

*Institute for Materials Research (IMR), Tohoku University,
Sendai 980-8577, Japan*

M. Nikl

*Institute of Physics AS CR, Cukrovarnická 10,
16000, Prague, Czech Republic*

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Intentionally doped LiCaAlF₆ (LiCAF) single crystals are prospective scintillators, especially for thermal neutron detection through the ${}^6\text{Li}(n,t){}^4\text{He}$ nuclear reaction. Four different LiCAF scintillator samples were tested in various neutron and gamma fields. Two of the tested samples were LiCAF:Eu and LiCAF:Eu,Na single crystals, and another two samples were made of LiCAF:Eu micro crystals dispersed in transparent rubber, with different rubber dimensions. All LiCAF samples contain lithium enriched to ${}^6\text{Li}$. A plutonium–beryllium source was used as a neutron source. The neutron spectrum was modified by moderator and filter to get different ratios between thermal, epithermal and fast neutron fluence rates. The MCNP code was used for calculations of the fluence rates for different configurations. Radionuclides ${}^{137}\text{Cs}$ and ${}^{60}\text{Co}$ were applied as gamma radiation sources. The light signal from the scintillator was evaluated with a photomultiplier and a multichannel analyzer. The purpose of this work was to study the characteristics of LiCAF scintillators, especially the ability to discriminate signals from neutron and gamma radiation, which is the basic scintillator condition for neutron detection in mixed neutron-gamma radiation fields. Generally, the discrimination can be done by the pulse height and/or the pulse shape of the evaluated signals. Both methods can be used for a LiCAF scintillator. However, only the pulse height discrimination method is discussed in this paper. The possibility of fast neutron detection with LiCAF scintillators was also tested.

Keywords: LiCAF scintillator; thermal neutron detection; fast neutron detection; gamma radiation.

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1. Introduction

Scintillators containing lithium are usually used for thermal neutron detection through the ${}^6\text{Li}(\text{n},\text{t}){}^4\text{He}$ nuclear reaction. The most common scintillator for this purpose is LiI:Eu . Intentionally doped LiCaAlF_6 (LiCAF) single crystals are also prospective scintillators for thermal neutron detection.^{1,2,3} Compared with LiI:Eu , they are not hygroscopic but they have a relative lower content of lithium. Four different LiCAF scintillator samples were tested in various neutron and gamma fields. A plutonium–beryllium source (PuBe) was used as a neutron source. The neutron spectrum was modified by a water moderator and a cadmium filter to get different ratios between thermal, epithermal and fast neutron fluence rates. The radionuclides ${}^{137}\text{Cs}$ and ${}^{60}\text{Co}$ were applied as gamma radiation sources. The light signal from the scintillator was evaluated with a photomultiplier and a multichannel analyzer. The purpose of this work was to study the characteristics of LiCAF scintillators, especially the ability to discriminate the signals from neutron and gamma radiation, which is the basic scintillator condition for neutron detection in mixed neutron-gamma radiation fields. Generally, the gamma-neutron discrimination can be done by the pulse height and/or the pulse shape of the evaluated signals.^{4,5} Both methods can be used for a LiCAF scintillator. In the described work, only pulse height discrimination was studied by detector spectra measurements in different radiation fields. The influence of scintillation quenching and crystal dimensions of the tested scintillators on the pulse height discrimination is discussed. The possibility of fast neutron detection with a LiCAF scintillator was tested. The studied detectors are also to be tested and used in horizontal neutron channel beams⁶ of the LVR-15 research reactor.⁷

2. Materials and Methods

Four LiCAF samples were used for the measurement purchased from Tokuyama Co. Ltd. Japan. The first two samples were LiCAF:Eu and LiCAF:Eu,Na single crystals with dimensions of 10 mm x 10 mm x 1 mm, and another two samples were made of LiCAF:Eu micro crystals dispersed in transparent rubber, with different rubber dimensions of 10 mm x 10 mm x 1 mm and 10 mm x 10 mm x 5 mm. In the following discussion, the four samples will be designated as LiCAF:Eu , LiCAF:Na , LiCAF:R1 , LiCAF:R5 , respectively. All LiCAF samples contain lithium with 95% enrichment of ${}^6\text{Li}$. Also, a LiI:Eu scintillator with dimensions of ϕ 12 mm x 2 mm was used for comparison of some parameters.

The light signal from the scintillators was evaluated with a photomultiplier (photocathode diameter of 40 mm) and the multichannel analyzer InSpector 2000. All analyzer setup parameters were fixed during the measurements (the amplifier gain was 3, high voltage of 700 V, a range 2048 channels, and rise time and flat top of trapezoidal pulse both 0.6 μs). The acquisition time was from 6 h to 24 h. The Genie 2000 V3.1 software was used for the spectra acquisition and evaluation.

A plutonium–beryllium neutron source with a neutron emission of $5 \times 10^6 \text{ s}^{-1}$ was used for the neutron field generation. The neutron spectrum was modified by a water

moderator and a cadmium filter to get different ratios of thermal, epithermal and fast neutron parts of fields. The schematic experimental arrangement for measurement with the PuBe neutron source is shown in Fig. 1. Four different thicknesses (including 0 cm) of water moderator were used, and the neutron field was either filtered by a Cd filter (thickness of 1 mm) or remained unfiltered; thus we had eight different neutron fields.

Neutron spectra in the scintillator position for these eight neutron fields were calculated by the MCNP code. The calculated fluence rates are in Tables 1 and 2, where the energy groups are: Thermal neutrons (0 eV, 0.5 eV); Epithermal neutrons (0.5 eV, 10 keV); Intermediate neutrons (10 keV, 1 MeV); and Fast neutrons (1 MeV, 20 MeV).

Fig. 1. The schematic experimental arrangement for measurement with the PuBe neutron source.

Table 1. Calculated fluence rates for different neutron fields **without** the Cd shielding.

		Fluence rate (cm ² .s ⁻¹)			
Water thickness	0 cm	3 cm	6 cm	9 cm	
Thermal neutrons	2206.8	2131.4	2043.5	1941.3	
Epithermal neutrons	953.0	832.7	735.3	655.6	
Intermediate neutrons	1460.2	971.0	757.3	645.0	
Fast neutrons	1094.3	783.8	581.7	453.1	

Table 2. Calculated fluence rates for different neutron fields **with** the Cd shielding.

	Fluence Rate (cm ⁻² .s ⁻¹)			
	0 cm	3 cm	6 cm	9 cm
Water thickness	0 cm	3 cm	6 cm	9 cm
Thermal neutrons	0.6	0.6	0.6	0.5
Epithermal neutrons	892.8	779.3	687.1	607.4
Intermediate neutrons	1587.8	1054.9	814.7	688.7
Fast neutrons	1086.3	778.5	577.7	449.7

¹³⁷Cs and ⁶⁰Co radionuclides were applied as gamma radiation point sources. Their activities were 200 kBq and 1100 kBq, the distances from scintillator center 50 mm and 100 mm, and the corresponding gamma photon fluence rates 550 cm⁻¹ s⁻¹ and 1750 cm⁻¹ s⁻¹, respectively.

3. Measurement in Neutron Field

3.1. Pulse height spectra

Pulse height spectra were measured for the four LiCAF samples in eight different neutron fields. An example of spectra for the LiCAF samples in the PuBe neutron field with a 6 cm thick water moderator and without the Cd shielding is shown in Fig. 2. Peaks correspond to the reaction of thermal neutrons and ⁶Li with a released energy of 4.78 MeV. The peak shape parameters are shown in Table 3 where the “Min/Max ratio” means the ratio of count rates in the local minimum below the peak and in the peak maximum. The LiCAF:Eu sample shows the best values in all the parameters from the table (for gamma radiation influence, see Sec. 4).

An example of the LiCAF:Eu spectra for neutron fields with a 6 cm thick water moderator, one spectrum with the Cd shielding and second without the Cd shielding, is shown in Fig. 3. In this case the count rate is in logarithmic scale and the x-axis is in energy units calibrated according to peak position. The part of the spectra below the peak (below 3800 keV) corresponds to gamma radiation, to cases when only a part of the ⁶Li(n,α)⁴He nuclear reaction energy is absorbed in the scintillator, and to other nuclear reactions occurring in the scintillator. The peak in the spectrum with the Cd (sometimes called an epithermal peak) is slightly shifted relative to the spectrum without the Cd (thermal peak) due to the addition of the energy of epithermal and intermediate neutrons. The part of the spectra above the peak (6000 keV to 13000 keV, denoted as “tail” in the next text) should mainly correspond to fast neutrons (see Sec. 3.5) and partly to gamma radiation.

Fig. 2. An example of spectra for the LiCAF samples in moderated PuBe neutron field without Cd shielding.

Table 3. The peak shape parameters.

	LiCAF:Eu	LiCAF:Na	LiCAF:R1	LiCAF:R5	LiI:Eu
Peak position (channel)	519	508	360	344	
Count rate in peak (s ⁻¹ /channel)	0.503	0.394	0.079	0.353	
Min/Max ratio (%)	4.3	5.3	18.7	20.5	7.0
Energy resolution (%)	8.7	10.6	19.7	17.6	13.7

Fig. 3. An example of the spectra for the LiCAF:Eu sample with and without Cd in neutron field.

3.2. Count rates for peaks and tails

Peak areas for neutron reaction were evaluated using the Genie 2000 software. “Tail” count rates were taken in the energy interval of 6 MeV to 18 MeV (more precisely, the lower limit was set to $4.78 \text{ MeV} + 2 \times \text{FWHM}$). The count rates are listed in Tables 4 to 6 where counting 1σ uncertainties were from 0.1 % to 1 %. Count rates for tail without the Cd (not presented in the paper) did not differ significantly from values with the Cd shielding.

Table 4. Count rates for the peak area, **without** Cd shielding.

Moderator	Count rates (s^{-1})			
	LiCAF:Eu	LiCAF:Na	LiCAF:R1	LiCAF:R5
0 cm	28.09	27.10	5.42	21.02
3 cm	25.44	24.95	5.44	20.42
6 cm	25.07	23.68	4.92	19.62
9 cm	23.72	22.28	4.86	18.37

Table 5. Count rates for the peak area, **with** Cd shielding.

Moderator	Count rates (s^{-1})			
	LiCAF:Eu	LiCAF:Na	LiCAF:R1	LiCAF:R5
0 cm	9.06	8.60	1.45	6.07
3 cm	6.98	6.81	1.21	4.75
6 cm	5.99	5.83	1.09	4.04
9 cm	6.36	5.21	0.99	3.74

Table 6. Count rates for the tail, **with** Cd shielding.

Moderator	Count rates (s^{-1})			
	LiCAF:Eu	LiCAF:Na	LiCAF:R1	LiCAF:R5
0 cm	2.562	1.900	0.164	1.591
3 cm	2.210	1.637	0.135	1.225
6 cm	1.945	1.361	0.111	0.813
9 cm	2.018	1.248	0.086	0.679

3.3. Thermal Neutron Fluence Rate

Theoretically, the thermal neutron fluence rate should be proportional to subtraction: “count rate without Cd” minus “count rate with Cd”. In spite of this theoretical assumption, comparison of calculated and measured count rates gives slightly better result without subtraction of count rates with Cd. In addition, this is also the more simple

method since for thermal neutron fluence measurement, only measurement of count rates without Cd is sufficient. Because the detectors were not absolutely calibrated, only the ratios of the measured and the calculated values of thermal neutron fluence rate could be compared. Values of the MCNP calculated fluence rates (Table 1, Thermal neutrons) and the peak count rates without Cd shielding (Table 4) were normalized to values for 0 cm thick water moderator and given scintillator sample. Relative differences of the measured from the calculated values are in Table 7 (i.e. a positive difference means that the measured value is greater than the calculated one). The calculated values were confirmed with standard experimental method of activation detectors with agreement up to 5 % for thermal neutrons therefore we can take them as “true values”. Then the relative differences in the table are relative errors for measurement with the specific scintillator and neutron field. Very good agreement is for LiCAF:R1 and LiCAF:R5 scintillators. The differences under 7 % (in absolute values) for LiCAF:Eu and LiCAF:Na are also acceptable, the slightly greater values are probably due to influence of gamma radiation signal to peak count rate evaluation.

Table 7. Relative difference of the measured and the calculated values of thermal neutron fluence rate.

Moderator	Relative Difference (%)			
	LiCAF:Eu	LiCAF:Na	LiCAF:R1	LiCAF:R5
0 cm	0.0	0.0	0.0	0.0
3 cm	-6.3	-4.7	3.9	0.6
6 cm	-3.6	-5.6	-2.0	0.8
9 cm	-4.0	-6.6	2.0	-0.7

3.4. Epithermal Neutron Fluence Rate

The epithermal (and partly intermediate) neutron fluence rate should be connected with the peak count rate with Cd shielding. Similarly as for thermal neutrons in Sec. 3.3, calculated values (Table 2, Epithermal neutrons) are compared with the measured peak count rates with Cd (Table 5), and the relative differences are given in Table 8. Here the differences are higher, compare to Table 7, for thermal neutrons. The reason can be mainly in greater influence of gamma radiation signal.

Table 8. Relative differences of the measured and the calculated values of the epithermal neutron fluence rate.

Moderator	Relative Difference (%)			
	LiCAF:Eu	LiCAF:Na	LiCAF:R1	LiCAF:R5
0 cm	0.0	0.0	0.0	0.0
3 cm	-11.8	-9.4	-4.7	-10.5
6 cm	-14.3	-12.1	-2.2	-13.8
9 cm	2.0	-12.0	-0.6	-10.3

3.5. Fast Neutron Fluence Rate

The fast neutron fluence rate should be roughly proportional to the count rate of spectrum tails.⁸ These count rates did not differ significantly when the Cd shielding was used or not, which corresponds with the calculated fluence rates for fast neutrons. For comparison in Table 9, the calculation and the measurement with Cd was used (Table 2, Fast neutrons and Table 6). Larger differences for LiCAF:Eu and LiCAF:Na are probably caused by gamma radiation interference. Acceptable agreement between calculated and measured values is only for LiCAF:R5 and partly for LiCAF:R1.

Table 9. Relative differences of the measured and the calculated values of fast neutron fluence rate.

Moderator	Relative Difference (%)			
	LiCAF:Eu	LiCAF:Na	LiCAF:R1	LiCAF:R5
0 cm	0.0	0.0	0.0	0.0
3 cm	20.4	20.2	14.4	7.5
6 cm	42.7	34.7	27.2	-3.9
9 cm	90.3	58.6	26.7	3.1

4. Measurement in Gamma Radiation Field

Precise relations between gamma and neutron spectra depend on many factors, such as the detector dimensions, the dopant type and concentration, and analyzer settings. In Fig. 4, pulse height spectra for the LiCAF:Eu and the LiCAF:R5 scintillators are shown in neutron and gamma fields. For better comparison, the count rates were normalized to the same fluence rate ($10^4 \text{ cm}^{-2}\text{s}^{-1}$) for neutrons and photons. The gamma energy of ^{137}Cs is 662 keV, and for ^{60}Co energies are 1173 keV and 1332 keV. The graphs show that gamma radiation could influence the neutron measurement results for gamma energies roughly from 1000 keV for LiCAF:Eu (comparable results e.g. in Iwanowska *et al.*⁹) and 1400 keV for LiCAF:R5. Similar results were gained for LiCAF:Na and LiCAF:R1, respectively.

Fig. 4. Comparison of the gamma and neutron pulse height spectra for the LiCAF:Eu (left) and the LiCAF:R5 (right) scintillator.

5. Discussion and Conclusion

The measurements confirmed the possibility to use all the tested samples of LiCAF scintillators for thermal neutron detection with a pulse height discrimination of neutron and gamma radiation. Also, epithermal neutron detection appears promising when a Cd shielding is used. The attempt to evaluate the fast neutron fluence rate from the pulse height spectrum tail seems hopeful for the LiCAF:Eu micro crystals dispersed in transparent rubber, while for the LiCAF single crystals, measurement and calculation comparison shows high differences. The reason is probably in higher influence of prompt gamma radiation from the water moderator to large single crystals, which are more sensitive to gamma radiation with prompt gamma energy of hydrogen of 2200 keV.

From the point of view of neutron and photon discrimination by pulse height, the results for the studied samples show not very satisfying characteristics. Gamma radiation with energies above 1000 keV interfere with the neutron reaction peak for LiCAF single crystals, which is roughly two times worse than for LiI:Eu (2000 keV). The samples with the LiCAF:Eu micro crystals dispersed in rubber provide a better option than the LiCAF single crystals in this parameter (1400 keV). The difference between the ratio of the absorbed energy and the light yield for gamma radiation and heavy charged particles from the neutron reaction is caused by the quenching dependence on linear energy transfer (LET). On the other hand, this difference could be connected to the greater variance in shapes for the gamma and neutron light pulses, and, therefore, good conditions for the pulse shape discrimination method are provided.

The used relative comparison of measured integral count rates and the fluence rates in four energy intervals helps to evaluate the applicability of the studied scintillators, but it is a simplification, which could contribute to the differences between calculated and measured values. A more precise and complex approach is to compare the theoretically calculated pulse height spectrum with the measured spectrum in absolute units or to calculate the neutron fluence spectrum from the pulse height spectrum by deconvolution. This is a task for future work.

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