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Pawel Sibczynski, Andrzej Dzedzic, Krystian Grodzicki, Joanna Iwanowska-Hanke, Marek Moszyński, et al.. New perspectives for undoped CaF₂ scintillator as a threshold activation neutron detector. EPJ Web of Conferences, 2018, 170, pp.07012. 10.1051/epjconf/201817007012 . cea-01772811

HAL Id: cea-01772811

<https://cea.hal.science/cea-01772811>

Submitted on 4 Apr 2023

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New perspectives for undoped CaF₂ scintillator as a threshold activation neutron detector

Pawel Sibczynski, Andrzej Dziedzic, Krystian Grodzicki, Joanna Iwanowska-Hanke, Marek Moszyński, *Fellow, IEEE*, Lukasz Swiderski, *Member, IEEE*, Agnieszka Syntfeld-Kazuch, *Member, IEEE*, Dariusz Wolski, Frédéric Carrel, Amélie Grabowski, Matthieu Hamel, Frederic Laine, Adrien Sari, Alessandro Iovene, Carlo Tintori, Cristiano Fontana, Felix Pino

Abstract— In this paper we present the prompt photofission neutron detection performance of undoped CaF₂ scintillator using Threshold Activation Detection (TAD). The study is carried out in the frame of C-BORD Horizon 2020 project, during which an efficient toolbox for high volume freight non-intrusive inspection (NII) is under development. Technologies for radiation monitoring are the part of the project. Particularly, detection of various radiological threats on country borders plays an important significant role in Homeland Security applications. Detection of illegal transfer of Special Nuclear Material (SNM) - ²³⁵U, ²³³U and ²³⁹Pu - is particular due to the potential use for production of nuclear weapon as well as radiological dispersal device (RDD) – known also as a “dirty bomb”. This technique relies on activation of ¹⁹F nuclei in the scintillator medium by fast neutrons and registration of high-energy β particles and γ-rays from the decay of reaction products. The radiation from SNM is detected after irradiation in order to avoid detector blinding. Despite the low ¹⁹F(n,α)¹⁶N or ¹⁹F(n,p)¹⁹O reaction cross-section, the method could be a good solution for detection of shielded nuclear material. Results obtained with the CaF₂ detector were compared with the previous study done for BaF₂ and ³He detector. These experimental results were obtained using ²⁵²Cf source and 9 MeV Varian Linatron M9 linear accelerator (LINAC). Finally, performance of the prompt neutron detection system based on CaF₂ will be validated at Rotterdam Seaport during field trials in 2018.

Index Terms — photofission, neutron detection, threshold activation detection, TAD, CaF₂, BaF₂, scintillators, fluorine, border monitoring, SNM, nuclear material, uranium, MCNP

This project has received funding from the European Union’s Horizon 2020 research and innovation programme under grant agreement No 653323. Disclaimer: This text reflects only the author’s views and the Commission is not liable for any use that may be made of the information contained therein.

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I. INTRODUCTION

FOR several years detection of nuclear materials plays an important role in Homeland Security and Border Monitoring. Particularly, detection of Special Nuclear Materials (SNM) is of high importance. Recently, fluorine based scintillation detectors were found to be good candidates for prompt photofission neutrons detection by means of threshold activation (the technique is known as a Threshold Activation Detection – TAD) [1], [2], [3], [4]. Detection of the prompt photofission neutron, emitted several fs after the photofission, is hardly possible directly because of detector blindness during the bremsstrahlung photon pulse from linear accelerator (LINAC). However, prompt photofission neutrons can be registered by the ¹⁹F(n,α)¹⁶N threshold reaction in the scintillator medium using LINAC producing bremsstrahlung photons with endpoint at 9 MeV. By this way, it is possible to detect characteristic β particles (energy endpoint at 4.3 MeV and 10.4 MeV) and γ-rays (6.1 MeV) emitted from the ¹⁹F(n,α)¹⁶N reaction product few seconds after irradiation. Registration of the characteristic radiation can be done after irradiation of nuclear material with high energy bremsstrahlung photons from LINAC due to the fact that the half-life of the ¹⁶N nucleus is 7.1 s. Currently, liquid fluorocarbon [1]–[3], pentafluorostyrene-based plastic [5] and BaF₂ [6] scintillators were proposed for the TAD technique. However, some properties of the fluorocarbon liquid scintillators cause the notably limited potential use for Homeland Security. The main disadvantages of the liquid fluorocarbon scintillators, such as EJ-313 or BC-509, are toxicity of hexafluorobenzene as the main compound, high flammability with the flashpoint at 10°C and risk of leakage. On the other hand, BaF₂ scintillator contains rather small mass fraction of fluorine (about 21%). Thus, we decided to investigate the undoped Ø5” × 3” CaF₂ scintillator for the TAD technique. The half of CaF₂:Eu light yield and light emission peak at 280 nm [7], although diminishes energy resolution when a photomultiplier with borosilicate glass would be used, still allows to be applied for the TAD. The energy resolution plays secondary role in the TAD technique. This technique aims on detection of beta continuum with endpoint at 10.4 MeV and high energy delayed γ-rays emitted for variety of photofission products. Both the mass fraction of

fluorine about 48.6% and the very low light self-absorption, allowing for production in large volumes, are the main advantages of the undoped CaF₂ crystals. Furthermore, we used an MCNP6 code for simulation of (n,α) reaction rate for the CaF₂ and compared with the BaF₂ of volume of Ø5" × 1", studied in [6].

II. MCNP6 SIMULATIONS

The MCNP6 software package was used for investigation of the CaF₂ and BaF₂ detector sensitivity. The calculations were divided into following processes:

- F4 tally - (n,α) reaction rate for prompt neutrons
- F1 tally - β particle detection efficiency
- F8 tally - 6.1 MeV γ-ray detection efficiency

Visualization of (n,α) interactions is presented in Fig. 1. A ²⁵²Cf neutron source was situated 15 cm from the front of the detector. The reactions occur mainly in the first layer of the CaF₂ detector, where about 51% of (n,α) reactions take place. However, the BaF₂ has greater density (ρ = 4.88 g/cm³), resulting in better detection efficiency of 6.1 MeV γ-rays emitted from ¹⁶N. Although the BaF₂ would be more efficient considering the same volume of the detectors, the lower cost

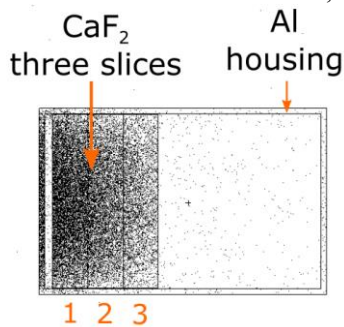


Fig. 1. Visualization of (n,α) reactions in the CaF₂ scintillator medium.

of CaF₂ can play significant role for application in large volume systems. Calculation results were summarized in

TABLE I

THE SIMULATED (N,ALPHA) REACTION RATES, ELECTRON ABSORPTION AND 6.1 MEV GAMMA-RAY DETECTION EFFICIENCY IN DETECTOR SLICES, Ø5" × 1" EACH. VALUES ARE NORMALIZED TO ONE-RADIATION SOURCE.

Detector	Front slice (1)	Central slice(2)	Back slice(3)
CaF ₂ (n, α) rate	2.20E-4 (50.9%)	1.34E-4 (30.9%)	7.87E-5 (18.2%)
CaF ₂ e ⁻ absorption	0.994%	100%	0.994%
CaF ₂ 6.1 MeV gamma absorption	1.78E-2	2.30E-2	1.78E-2
CaF ₂ efficiency – front slice	2.57E-6	-	-
CaF ₂ efficiency – whole detector	-	5.44E-6	-
BaF ₂ (n, α) rate	5.88E-5	-	-
BaF ₂ e ⁻ absorption	0.997%	-	-
BaF ₂ 6.1 MeV gamma absorption	3.87E-2	-	-
BaF ₂ efficiency – whole detector	-	1.49E-6	-

Table 1. Finally, Ø5" × 3" CaF₂ present much greater (n,α) reaction rate and better γ-ray detection efficiency for 6.1 MeV γ-rays than the previously tested BaF₂.

III. MEASUREMENTS WITH A ²⁵²Cf SOURCE AT NCBJ

Spectra from the ²⁵²Cf for the Ø5" × 3" CaF₂ and previously tested BaF₂ [6] are presented in Fig. 2. Comparing the relative number of counts, the experimental results and the one obtained with MCNP6 are in reasonable agreement. The number of counts ratio for CaF₂ and BaF₂ in the region of interest (ROI, 6 – 10.5 MeV) from the experiment is 3.1. The total value ratio result from the MCNP simulation for whole detectors, summarized in Table I, is 3.7.

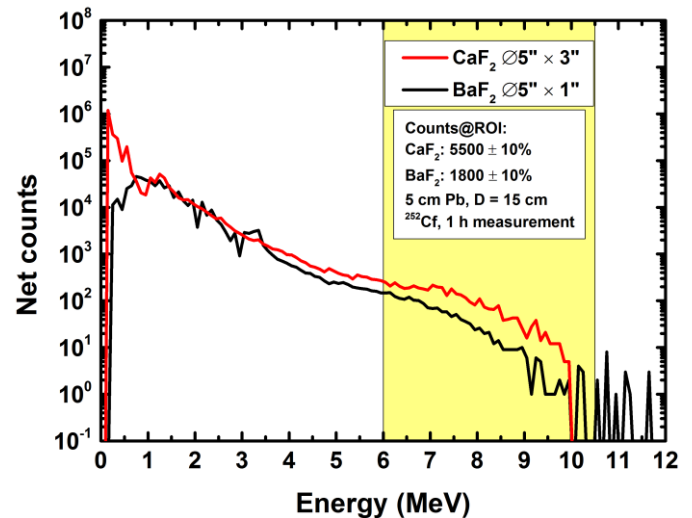


Fig. 2. The energy spectra of BaF₂ and CaF₂ exposed to the ²⁵²Cf source. The β continuum up to 10.4 MeV is clearly seen.

IV. TEST SITE AT CEA SACLAY

Fig. 3 presents the test site at CEA Saclay SAPHIR Facility. A Varian LINATRON M9 LINAC working in 9 MeV mode was used to induce photofission in depleted uranium (DU). The DU was placed 1 meter from the conversion target, then, CaF₂ detector was placed 1 meter from the DU. Data from the tested CaF₂ detector were registered with CAEN DT5730 8 channel 500 MS/s desktop digitizer with dedicated FPGA firmware allowing for onboard VETO signal gating using the LINAC trigger.



Fig. 3. Test site at SAPHIR LINAC facility at CEA Saclay, France.

V. ACTIVE MEASUREMENTS RESULTS AT SAPHIR

The single energy spectrum after background subtraction for the CaF₂ for 5 cm Pb shielded DU is showed in Fig. 4. Single measurement was based on 10 s of irradiation, 1 s of cooling time and 20 s of data recording after the global irradiation. Number of net counts for the CaF₂, BaF₂ and ³He detectors are shown in Table 2. The number of counts between 6 and 10.5 MeV for the CaF₂ and BaF₂ showed in the table were summed from the 3 consecutive measurements.

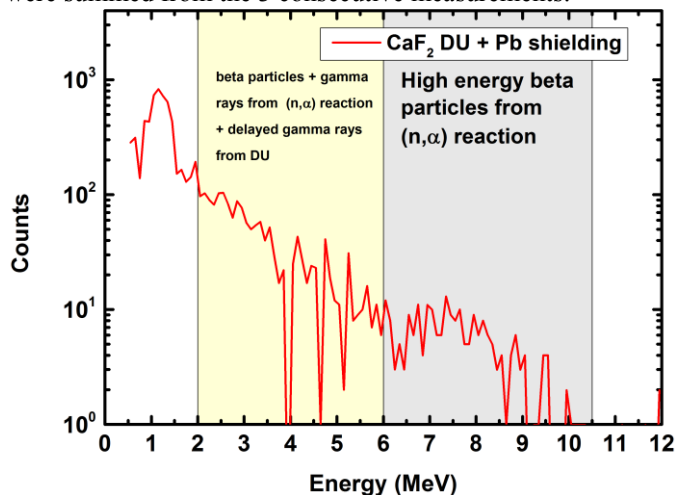


Fig. 4. Net spectrum of depleted uranium after photofission registered with the CaF₂ detector.

VI. SUMMARY

TABLE II
NUMBER OF NET COUNTS FROM DU REGISTERED WITH THE CaF₂, BaF₂ AND SINGLE ³He 150NH100 TYPE DETECTOR

Detector	Size	Net counts	Reference
CaF ₂ + 5 cm Pb	Ø 5" × 3"	205 ± 14	
CaF ₂ without Pb	Ø 5" × 3"	420 ± 20	
BaF ₂ + 5 cm Pb	Ø 5" × 1"	189 ± 14	[6]
BaF ₂ without Pb	Ø 5" × 1"	275 ± 17	[6]
³ He tube + 5 cm Pb	Ø2.5 cm × 1m	3600 ± 60	[6]
³ He tube without Pb	Ø2.5 cm × 1m	4450 ± 67	[6]

Better performances were achieved with the Ø5" × 3" CaF₂ in comparison with the previous solution based on Ø5" × 1" BaF₂. Much greater (n,α) reaction rate, resulting in greater high energy β particles emission, was observed for CaF₂ in comparison with the previously tested BaF₂. The Ø5" × 3" CaF₂ also has better γ-rays detection efficiency, which is crucial in detection of delayed γ-rays as well as 6.1 MeV γ-rays from (n,α) reaction. The MCNP6 simulations and measurements of relative net counts with ²⁵²Cf source are in reasonable agreement. Although measurements at SAPHIR facility confirmed better performance of the CaF₂ over the BaF₂ detector, the dominance is evidently smaller. Despite the fact that the same amount of DU was used in both experiments, the geometry of the DU samples used was

different, what could be the reason of the discrepancy. Finally, in the frame of C-BORD project, the prompt neutron detection system based on CaF₂ detectors will be integrated and tested in the field trial at Rotterdam Maasvlakte Seaport in 2018.

ACKNOWLEDGMENT

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 653323. Disclaimer: This text reflects only the author's views and the Commission is not liable for any use that may be made of the information contained therein.

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