New perspectives for undoped CaF_2 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-Każuch, *Member, IEEE,* Dariusz Wolski, Frédérick 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 CaF_2 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) - ^{235}U, ^{233}U and ^{239}Pu - 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 ^{19}F 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 $^{19}F(n,\alpha)^{16}N$ or $^{19}F(n,p)^{19}O$ reaction cross-section, the method could be a good solution for detection of shielded nuclear material. Results obtained with the CaF_2 detector were compared with the previous study done for BaF_2 and ^{3}He detector. These experimental results were obtained using ^{252}Cf source and 9 MeV Varian Linatron M9 linear accelerator (LINAC). Finally, performance of the prompt neutron detection system based on CaF_2 will be validated at Rotterdam Seaport during field trails in 2018.**

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

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Pawel Sibczynski, Andrzej Dziedzic, Krystian Grodzicki, Joanna Iwanowska-Hanke, Marek Moszyński, Lukasz Swiderski, Agnieszka Syntfeld-Każuch, Dariusz Wolski are with the National Centre for Nuclear Research (NCBJ), Otwock, 05-400 Poland (telephone: +48 22 273 1440, e-mail: pawel.sibczynski@ncbj.gov.pl, pawel.sibczynski@gmail.com)

Frédérick Carrel, Amélie Grabowski, Matthieu Hamel, Frederic Laine, Adrien Sari are with CEA Saclay, LIST, Sensors and Electronic Architectures Laboratory, F-91191 Gif-sur-Yvette, France.

Alessandro Iovene, Carlo Tintori are with the CAEN S.p.A., Via Vetraia 11, 55049 - Viareggio (LU) – Italy.

Cristiano Fontana, Felix Pino are with University of Padua, Department of Physics and Astronomy, Via Marzolo 8, 35131 Padua - Italy

I. Introduction

For several years detection of nuclear materials plays 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 $^{19}F(n,\alpha)^{16}N$ 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 $^{19}F(n,\alpha)^{16}N$ 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 ^{16}N nucleus is 7.1 s. Currently, liquid fluorocarbon [1]–[3], pentafluorostyrene-based plastic [5] and BaF_2 [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, BaF_2 scintillator contains rather small mass fraction of fluorine (about 21%). Thus, we decided to investigate the undoped Ø5" × 3" CaF_2 scintillator for the TAD technique. The half of CaF_2: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 CaF_2 crystals. Furthermore, we used an MCNP6 code for simulation of (n,α) reaction rate for the CaF_2 and compared with the BaF_2 of volume of Ø5” × 1”, studied in [6].

II. MCNP6 SIMULATIONS

The MCNP6 software package was used for investigation of the CaF_2 and BaF_2 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 ^{252}Cf neutron source was situated 15 cm from the front of the detector. The reactions occur mainly in the first layer of the CaF_2 detector, where about 51% of (n,α) reactions take place. However, the BaF_2 has greater density (ρ = 4.88 g/cm^3), resulting in better detection efficiency of 6.1 MeV γ-rays emitted from ^{16}N. Although the BaF_2 would be more efficient considering the same volume of the detectors, the lower cost

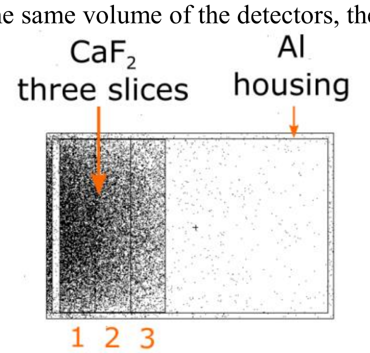

Fig. 1. Visualization of (n,α) reactions in the CaF_2 scintillator medium.

of CaF_2 can play significant role for application in large volume systems. Calculation results were summarized in

TABLE I

THE SIMULATED (N,ALPHA) REACTION RATES, ELECTRON ABSORTPION 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)	Black 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” CaF_2 present much greater (n,α) reaction rate and better γ-ray detection efficiency for 6.1 MeV γ-rays than the previously tested BaF_2.

III. MEASUREMENTS WITH A ^{252}CF SOURCE AT NCBJ

Spectra from the ^{252}Cf for the Ø5” × 3” CaF_2 and previously tested BaF_2 [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 CaF_2 and BaF_2 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.

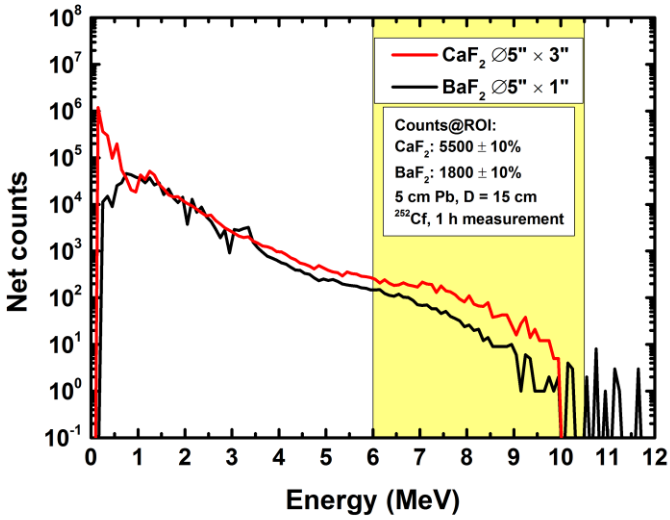

Fig. 2. The energy spectra of BaF_2 and CaF_2 exposed to the ^{252}Cf source. The β continuum up to 10.4 MeV is clearly seen.

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, CaF_2 detector was placed 1 meter from the DU. Data from the tested CaF_2 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 CaF_2 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 CaF_2, BaF_2 and ^3He detectors are shown in Table 2. The number of counts between 6 and 10.5 MeV for the CaF_2 and BaF_2 showed in the table were summed from the 3 consecutive measurements.

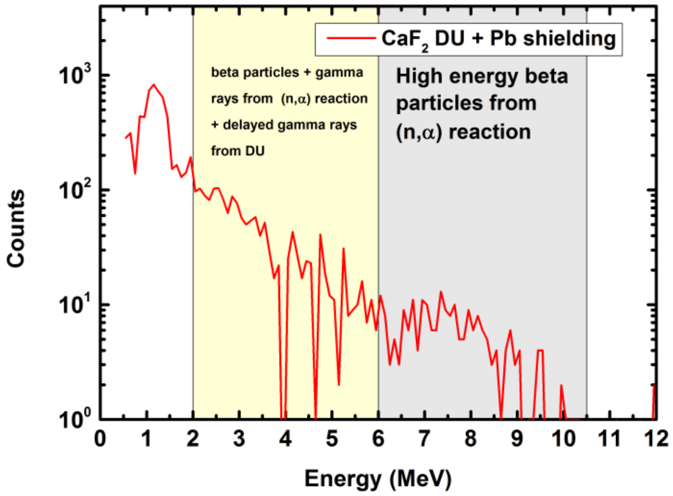

Fig. 4. Net spectrum of depleted uranium after photofission registered with the CaF_2 detector.

VI. SUMMARY

TABLE II
NUMBER OF NET COUNTS FROM DU REGISTERED WITH THE CaF_2, BaF_2 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]
^3He tube + 5 cm Pb	Ø2.5 cm × 1m	3600 ± 60	[6]
^3He tube without Pb	Ø2.5 cm × 1m	4450 ± 67	[6]

Better performances were achieved with the Ø5” × 3” CaF_2 in comparison with the previous solution based on Ø5” × 1” BaF_2. Much greater (n,α) reaction rate, resulting in greater high energy β particles emission, was observed for CaF_2 in comparison with the previously tested BaF_2. The Ø5” × 3” CaF_2 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 ^{252}Cf source are in reasonable agreement. Although measurements at SAPHIR facility confirmed better performance of the CaF_2 over the BaF_2 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 CaF_2 detectors will be integrated and tested in the field trial at Rotterdam Maasvlakte Seaport in 2018.

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