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Thermal neutron detection using thin PEN film doped with high cross section materials

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Poly(ethylene 2,6-naphthalate) (PEN) is promising as a new plastic scintillator, which emits deep-blue photons. Its photoluminescence emission peak (434–436 nm) and the decay time is of the order of 3 ns, as well as it is resistant to harsh environment. In this study, thin PEN film with high neutron cross section dopants was used for the thermal neutron detection. Iron boride, lithium tetraborate and lithium metaborate layers were coated on a 125 μm PEN film. Reaction particles from the boron neutron and lithium neutron reaction were detected by scintillation in PEN, and photomultiplier (PMT) pulses were registered and analysed. Energy deposition in the film samples was calculated using MCNP6 code taking into account losses in the source and air gap for the alpha particle source and incomplete energy deposition for the electrons. It was found that a small quantity of particles from the thermal neutron reaction could be detected in the strong neutron, gamma ray and recoil proton background.

Keywords: scintillation, irradiation detection, neutrons

INTRODUCTION

Sensitive, inexpensive and simple detectors with fast response and photoluminescence short decay times are needed for identification of different ionizing radiation particles, e.g. neutron/gamma in situ measurements for neutron activation, neutron dose evaluation and neutron sources detection. The development of organic scintillators started in the last century and it continuously has improved till nowadays [1–6]. Organic scintillator operation is based on high scintillation performance (of active material) due to the incident radiation event and the photon emission registration using photoelectrical multipliers. The organic scintillator detectors with modifications could be used to detect all kinds of ionizing radiation [7, 8]. For neutron detection, materials with high neutron capture cross section could be used. It is important from economical point of view because currently ^{3}He gas proportional detectors are mostly used. Due to production limitations ^{3}He price is increasing very fast.

Poly(ethylene 2,6-naphthalate) as a possible material for ionizing radiation detection has been recently taken into consideration. This material is well resistant to the environment degradation and emits enough photons per radiation incident to detect scintillation using a photomultiplier. Moreover, it has a higher oxygen barrier than polyethylene terephthalate (PET). This polymer emits up to 10500 photons per MeV and has the energy resolution of approximately

10% in the 1 MeV region [9]. In this study, we used thin PEN film with high neutron cross section materials (based boron and lithium) coated on it.

METHODOLOGY

The main problem in neutron detection is the neutron reaction separation from the gamma ray reactions in detector material because the neutron flow is always accompanied by gamma flow. Moreover, schemes of interaction with material are similar. To detect neutron interaction we used the spectrum analysis method, which is based on specific energy area analysis, which depends on the converter material [9].

We obtained three modifications (Q51, Q83, and Q65HA) of polyethylene naphthalate (PEN) samples from Teijin DuPont Films and GoodFellow of different thickness. In this paper, data from experiments with Q83 modification and a 125 μm thickness film were used because Q83 film showed better results in the previous radiation registration and neutron radiation identification experiments, as well as better neutron and gamma separation results were obtained.

Natural, not enriched iron boride, lithium tetraborate and lithium metaborate layers were coated on a 125 μm PEN film. ^{10}B isotope concentration in natural boron is 19.9%, and that of ^{6}Li isotope is 7.5%. 100 μm thickness FeB was coated on film using the prepared paste. A 500 nm thickness $LiBO_2$ film was coated using the physical vapour deposition (PVD) technology, and electron beam evaporation in a vacuum chamber was used. Together with PEN film, Makrofol® film was coated with the same $LiBO_2$ layer.

Makrofol® is a film which is used for ionizing tracking of radiation particles. It was placed in the neutron flow for 48 hours. After exposition in the neutron flow, Makrofol® film was etched for 2 hours in 70°C PEW solution. Ionizing radiation created tracks were scanned with the digital microscope.

Thermal neutron and converter material reactions used in this experiment:

$$^{10}_{5}B + n \rightarrow {}^{7}_{3}Li(1.47\ \text{MeV}) + {}^{4}_{2}He(0.84\ \text{Mev}) \quad (1)$$

$$^{6}_{3}Li + n \rightarrow {}^{3}_{1}H(2.73\ \text{Mev}) + {}^{4}_{2}He(2.05\ \text{Mev}) \quad (2)$$

The experimental set-up is described in Fig. 1. Measurement was performed with the irradiating scintillator and a Pu-Be source (~4.7 × 10^7 n/s). The detector was placed at 0.15 m from the source. The lead layer was used to eliminate the low energy gamma background. The cadmium layer was used to absorb thermal neutrons.

Coated films were connected to the PMT tube (ФЕУ-101) photocathode using optical, liquid contact. The photomultiplier cathode was connected to Ortec high voltage supply, and the electrical chain was configured to work in a hot cathode

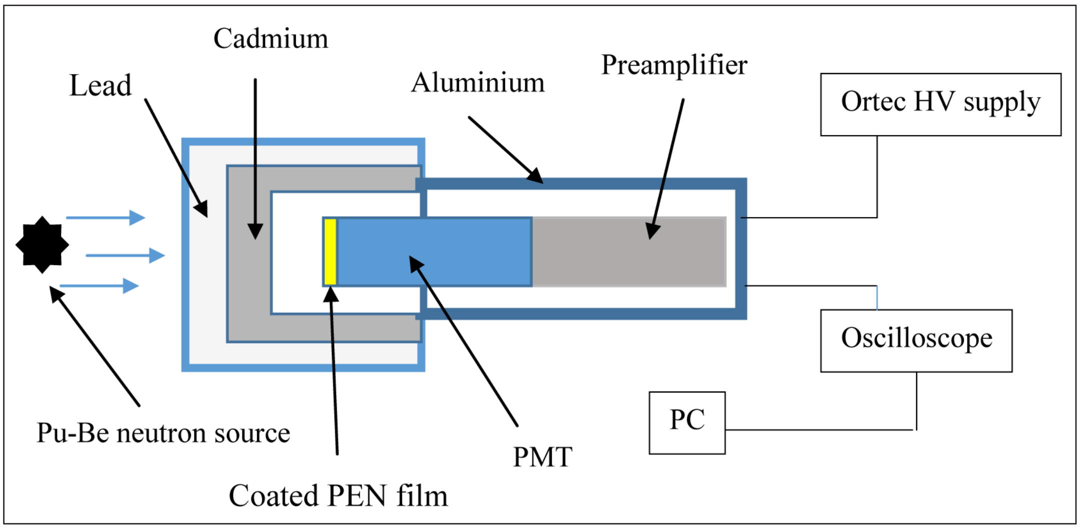

Fig. 1. Experimental set-up of thermal neutron measurement

mode. Such configuration was used because of organic scintillators, which usually have a short photoluminescence decay time. PEN photoluminescence decay time is 3 ns [9].

The pulse from PMT was registered in the oscilloscope. The oscilloscope ADC output was analysed using a special PC software. The total pulse was integrated and saved as energy (channel) counts. Same pulse counts were used for the spectrum measurement to have a possibility of comparing spectra with and without coatings. Measured spectra from the neutron – boron reaction were compared to the spectra from a 1.5 MeV α particles Pu source.

MCNP6 code was used to simulate the ionizing radiation particles energy deposition in the detector material. MCNP is a general-purpose Monte Carlo N-Particle code that can be used for neutron, photon, electron, or coupled neutron/photon/electron transport. Specific areas of application include radiation protection, dosimetry and radiation shielding [10].

RESULTS AND DISCUSSION

The ionizing radiation particles energy deposition was simulated using MCNP6. Simulation results are presented in Fig. 2. It shows that using the FeB layer the PEN absorbs more energy from the same neutron flux. The graph shows that (starting) from about 1.5 MeV more energy is absorbed because α particles created in the thermal neutron reaction could reach the detector surface.

Experimental results with the FeB layer on PEN film are presented in Fig. 3. It shows that more pulses were registered in the 1.5 MeV area, the same area where pulses from the alpha source are situated, when PEN with FeB was used. Moreover, in the graph it could be seen that the Cd absorber has influence on the measured spectrum. Fewer pulses were registered in the 1.5 MeV area with the Cd absorber than without it.

Experimental results are correlating with simulation and show that particles created in the converter layer could reach the PEN detector surface and the detector can detect these particles.

The coated Makrofol® film photo is presented in Fig. 4. A picture from the digital microscope is presented in Fig. 5. In this picture, enlarged borderline between coated and non-coated Makrofol® sides is shown. The picture demonstrates that the concentration of particles tracks is higher on the coated side. It

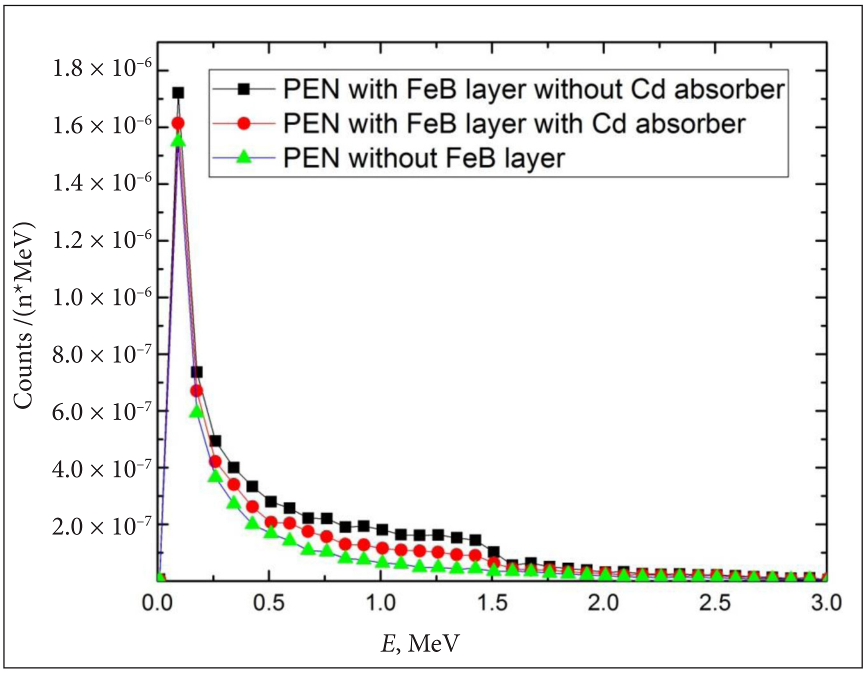

Fig. 2. Simulated energy deposition in the PEN film

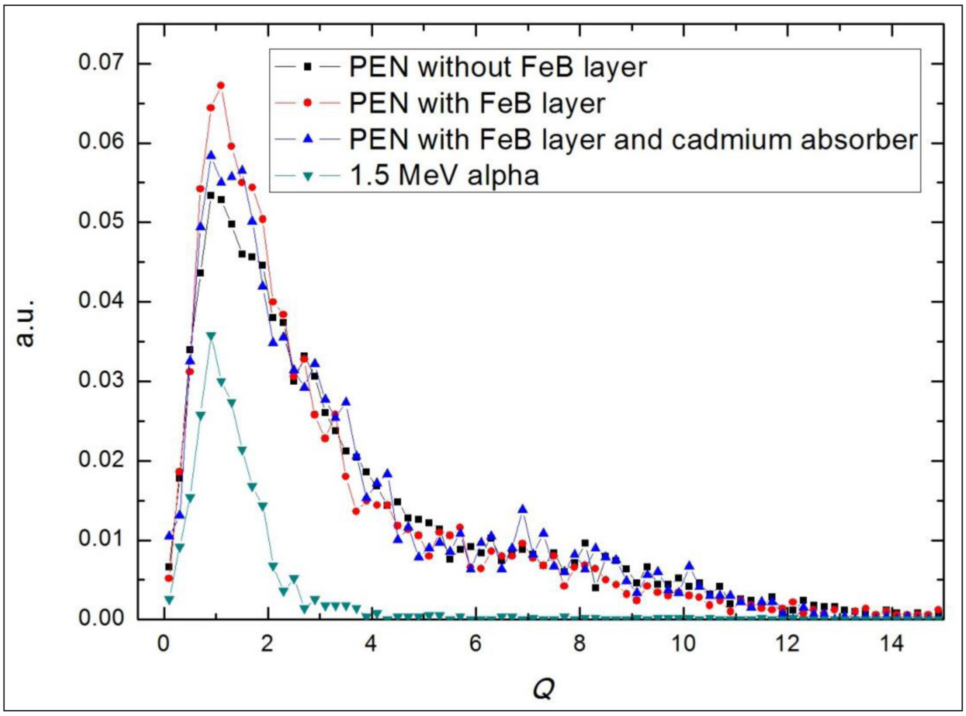

Fig. 3. Ionizing radiation from neutron source spectra measured by using thin PEN film coated by a 100 μm FeB layer. Green line demonstrates 1.5 MeV α particles spectra from sealed source measured by using 125 μm PEN without FeB layer. *Q* – total signal integral

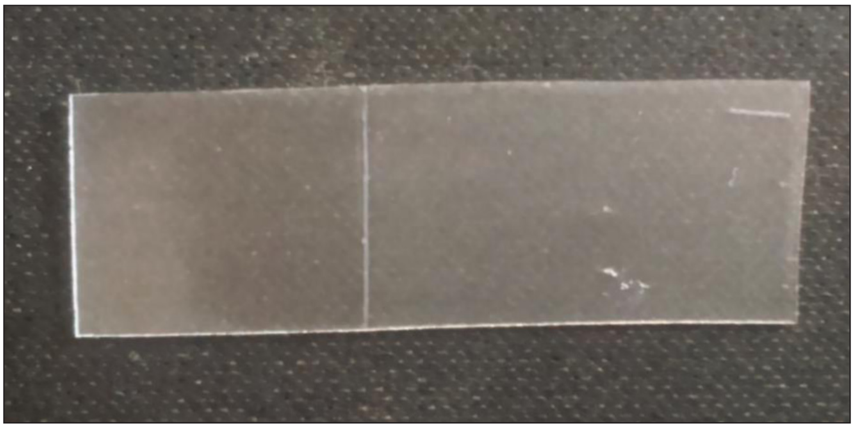

Fig. 4. Makrofol® film. The coated part is shown on the left and the non-coated part on the right

shows that the $LiBO_2$ layer creates particles due to thermal neutron reactions with boron and lithium. The particles could reach the detector surface.

The spectrum measurement using the PEN detector with $LiBO_2$ coating is presented in Fig. 6. It shows that there is no difference between the spectra measured with the coated detector and non-coated detector. Using this detector we could not detect a small quantity of particles from the thermal neutron reaction with the converter layer. Probably, the problem is in the detector thickness, because not enough particles are created to separate them from the background. It is planned to perform the same experiment using a thicker converter layer. For that purpose a different coating procedure must be used because it is complicated to coat thick layers (more than 10 μm) using the electron beam evaporation in a big vacuum chamber.

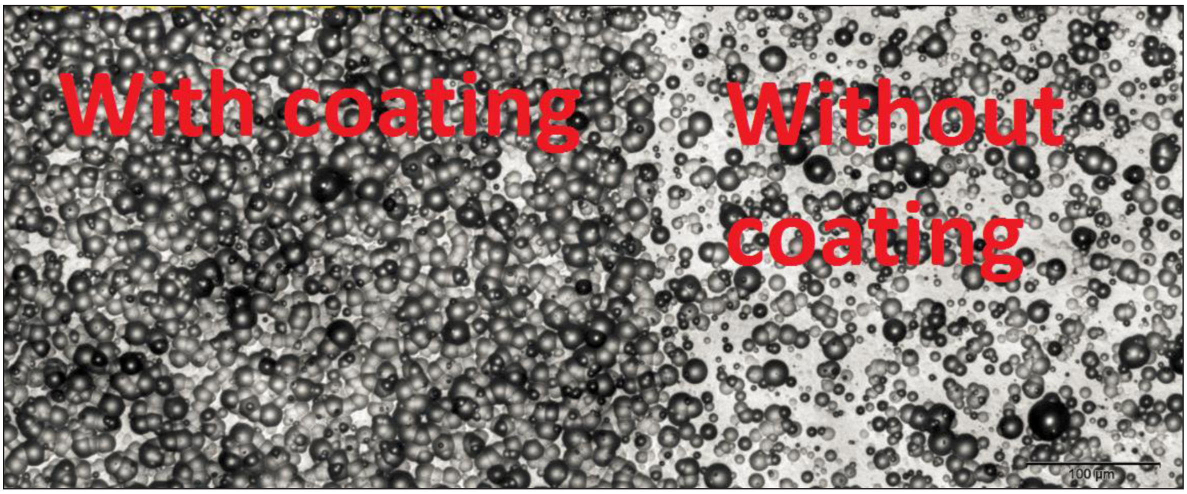

Fig. 5. Microscope picture of Makrofol®. The coated part is shown on the left and the non-coated part on the right

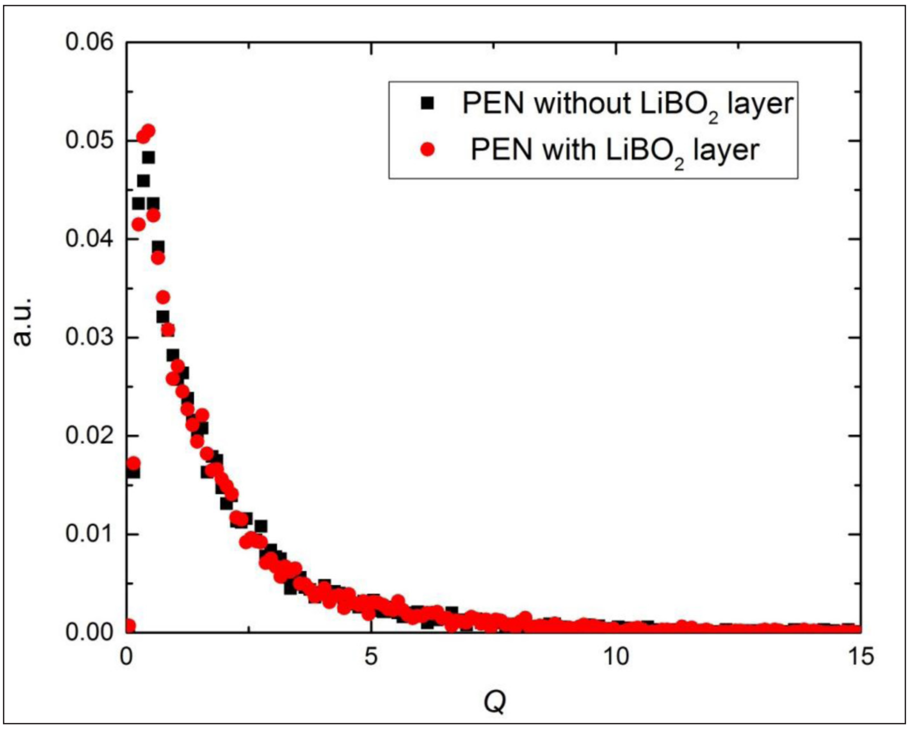

Fig. 6. Ionizing radiation from neutron source spectra measured by using thin PEN film coated with a 500 nm $LiBO_2$ layer

CONCLUSIONS

It has been revealed that thin, coated PEN film could be used as a thermal neutron detector. MCNP6 simulation has shown that the FeB layer due to the neutron interaction creates reaction particles that could reach the detector surface. Experiments showed that a small quantity of α particles from the thermal neutron reaction could be detected in the strong background of neutrons, gamma rays and recoil protons. A thick FeB layer could be used to create charged ionizing particles, which could be detected with the PEN scintillation detector.

The Makrofol® detector microscope pictures have shown that the $LiBO_2$ layer creates thermal neutron reaction products. Created particles could not be detected with the PEN detector because of strong background and a small quantity of particles. The 500 nm thickness of the $LiBO_2$ layer is too thin to detect thermal neutron reaction products during a short exposition time. A thicker converter layer is needed to detect the particles.

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ŠILUMINIŲ NEUTRONŲ REGISTRAVIMAS NAUDOJANT PLONĄ PEN PLĖVELĘ, PRATURTINTĄ DIDELIO PAGAVOS SKERSPJŪVIO MEDŽIAGOMIS

Santrauka

Polietileno naftalatas (PEN) yra medžiaga, kuri gali būti naudojama kaip scintiliatorius, emituojantis mėlynos šviesos spektro fotonus. Fotoliuminescencijos maksimumas yra ties 434–436 nm, išspinduliavimo laikas – 3 ns, medžiaga atspari agresyvios aplinkos poveikiui. Šiame darbe neutronams registruoti buvo naudojama plona PEN plėvelė, praturtinta didelio neutronų pagavos skerspjūvio medžiagomis. 125 μm PEN plėvelė buvo padengta geležies borido, ličio tetraborato ir ličio metaborato sluoksniais. Dalelės, atsirandančios po šiluminių neutronų reakcijos su boru ir ličiu, buvo registruojamos pagal PEN plėvelėje susidariusias scintiliacijas. Šviesos impulsai iš plėvelės buvo registruojami naudojant fotodaugintuvą, elektriniai impulsai iš fotodaugintuvo išanalizuoti kompiuterinių programų pagalba. Dalelių energijos pasiskirstymas PEN plėvelėje buvo modeliuotas naudojant MCNP6 programinę įrangą. Siekta, kad modeliavimo sąlygos kuo labiau atitiktų eksperimento sąlygas, taip pat įvertintas dalelių energijos praradimas konvertuojančioje medžiagoje. Tyrimai parodė, kad naudojant aprašytą technologiją, net ir intensyviame gama spinduliuotės ir atatrankos protonų sraute, mažas šiluminių neutronų kiekis gali būti užfiksuotas.

Raktažodžiai: scintiliatorius, jonizuojančiosios spinduliuotės registravimas, neutronai