

Composite polycrystalline boron nitride for alpha and neutron detectors

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Hexagonal Boron Nitride polycrystalline material can act as semiconductor neutron detector which converts neutrons to alpha particles as a result of the neutron reaction with ^{10}B . Therefore by testing for the response to alpha radiation BN or any other similar compounds, which have a relative large cross section to absorb neutrons; one can obtain a preliminary indication on its ability to detect neutrons. Among the successful compounds we studied and reported here is composite BN. The polycrystalline studied BN were bound as planar detectors with different binders, such as Polystyrene or Nylon-6, and coated with different metal electrodes such as ITO, Aquadag, Al and Cu. The planar BN detector plates which contained initially only the natural amount of the active ^{10}B of 20% were further diluted by the binder to only 10% of ^{10}B . Despite the low content of neutron active conversion isotope and very low signal to noise (S/N) ratio of about 2, the neutron caused events could be counted by subtracting the noise from the signal. In addition, the BN was also irradiated on an intensive neutron beam and the results will be shown.

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1. Introduction

^{10}B is an isotope of Boron which has a very large cross section for neutrons, of 3840 barns. In natural Boron it has a rather high abundance of 19.8%. ^{10}B reacts with thermal (slow) neutrons producing alpha particles and ^7Li . 94% of Lithium nuclei is produced in an excited state $^7\text{Li}^*$ with a kinetic energy of 0.84 MeV. Alpha particles bring then an energy of 1.47MeV. The $^7\text{Li}^*$ nucleus returns after a very short half life of about 1×10^{-13} sec to its ground state emitting a gamma radiation of 0.48MeV. The remaining 6% of the $^{10}\text{B}(n,\alpha)$ reactions induced by thermal neutrons, produce ^7Li directly in the ground state with kinetic energy of 1.02MeV and 1.78 MeV in case of alpha particle. Nuclear reaction products induced by thermal neutrons are emitted in opposite directions [1] forming a trail of electron-hole pairs with a cumulative charge proportional to the total energy of reaction products. By applying an electric field to the semiconductor, the charge carriers can be collected and create electric signals. Thus, in principle, any compound or metallic boron containing ^{10}B material can be used for preparation of a thermal (slow) neutron detector with a direct conversion of energy of neutron interaction into an electric charge within the detector body itself. The present paper will show results obtained with natural occurring boron compounds such as a (wide band gap) semiconductor, BN, which is efficient enough to detect thermal neutrons without any enrichment of the natural Boron.

An important novelty of the approach shown here is the use of polycrystalline semiconductor layers rather than single crystals. Nevertheless, the semiconductor

polycrystalline materials used in this paper are not pure, but mixed with inorganic or organic polymer binders. The use of the organic or inorganic binder reduces an effective thickness of the ^{10}B neutron conversion layer from 20%, which corresponds to the natural Boron, to 10%. The use of polycrystalline semiconductors mixed with binders used as X-ray imaging detectors has already been shown earlier with HgI₂ films [2-5]. Summarizing the advantages of Composite Semiconductor Neutron Detectors is their low cost of production at lower temperatures and use of natural Boron (not enriched in ^{10}B) and the ability to produce the detectors in any shape and size. First results presented by the present Authors [6-8] showed the response obtained with composite polycrystalline semiconductor hexagonal BN to low fluxes of slow neutrons. The general state of the art of neutron detection and the different methods used so far are described in a review paper [9] which contains many pertinent references on this subject. Since in ^{10}B based neutron detectors the (n, alpha) reactions are responsible for a possible signal generation, it is natural to study a response of neutron detectors such as BN to alpha particles. Some of these alpha detection studies will be published elsewhere [8]. The present paper will summarize responses of the detector based on hexagonal polycrystalline Boron Nitride powders bound with Polystyrene tested to 5.5MeV and 4.8MeV alpha particles from ^{241}Am source and from ^{226}Ra source, respectively. The main novelty of the present paper, are the results obtained here for the first time with the composite semiconductor BN detector in a pure thermal neutron

beam. These results are compared with some neutron detection data obtained with weak ^{241}Am - ^9Be source.

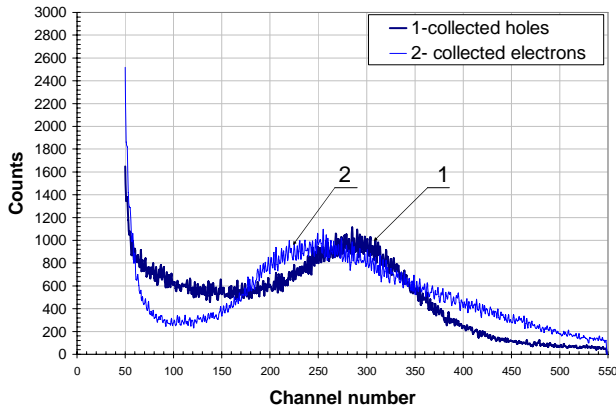


Fig. 1. Alpha spectra (hole collection) of 5.5 MeV alpha particles emitted by ^{241}Am detected by a 0.8 mm composite BN in a polystyrene binder. Both contacts are Aquadag, top and bottom contacts are 5 mm², applied bias 1100 V. Negative polarity on top contact is for electrons and positive for holes. Total amplification is 500,000; Shaping time = 10 μs . The scale is 10 mV/channel. 1- collected electrons 2- collected holes.

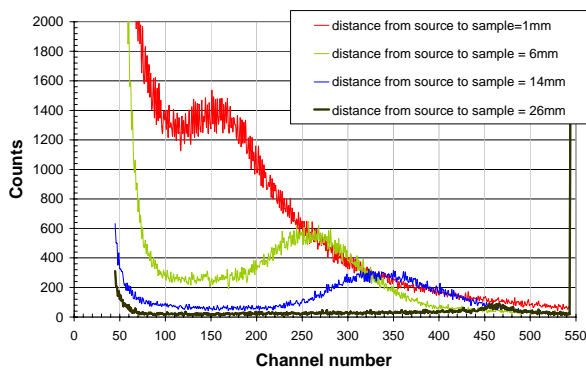


Fig. 2. Pulse height spectra of alpha particles from the ^{241}Am source measured with BN detector at atmospheric pressure in different distances between the source and the detector top contact. A 0.29 mm thick composite BN in a polystyrene binder was illuminated by alpha particles through a thin collimator of 5 mm in diameter. The distances between alpha source and the detector varied between 6-24 mm. Top contact is Aquadag, bottom contact is ITO. Effective area is 3 mm², detector biased to 1200 V. Negative polarity on top contact is for electrons and positive for holes. Total amplification is 400,000. Shaping time is 10 μs .

2. Experimental procedure and results

The composite hexagonal BN bound in Polystyrene or in Nylon-6 were produced as shown in [4] and tested in a nuclear spectroscopic setup shown in [6-8] for both, alpha particles and neutrons. Fig. 1 shows a composite BN detector (0.8 mm in a polystyrene binder) response to 5.5 MeV from ^{241}Am . Both contacts are Aquadag, top and bottom contacts of about 5 mm² areas are biased to 1100 V.

Negative polarities on top contact are favorable for collection of electrons, the positive ones for holes. Total amplification was set to 500,000, shaping time to 10 μs . One can see that the spectra at both polarities look quite similar. Nevertheless, most of our measurements for BN were performed with positively biased device (hole collection). Another measurement was done to determine pulse height spectra (hole collection) of alpha particles from the ^{241}Am source positioned at atmospheric pressure in different distances between the source and the detector top contact, see Fig. 2. A 0.29 mm thick composite BN in a polystyrene binder was illuminated by alpha particles through a thin collimator of 5 mm in diameter. The distances between alpha source and the detector varied between 6-24 mm. One can see that the shape of the pulse height spectrum depends on the source-detector distance. At smaller distances, the number of counts increases and the alpha peak becomes sharper and positioned at lower channels. For example, at the distance of 24 mm compared to the response at 6 mm, the number of counts is smaller and the alpha peak is broadened and placed at higher energy channels. Table 1 shows the actual number of counts as a function of distance for a larger number of energy channels (45-550) and a smaller number of channels (100-530). The smaller number of counts is a more conservative estimate, trying to avoid the counting of possible noise signals.

Table 1. The total number of counts from Channels 45-550 and 100-530 as a function of the distance between the Alfa source of 5.5 MeV from ^{241}Am and the top contact of the BN detector shown in Fig. 2.

Distance from source to the sample [mm]	Total number of counts from	
	Channels 45 - 550	Channels 100-530
1	386014	248193
6	209829	100924
14	67040	54524
26	18290	11454

Generally speaking, since the experiments were performed in air at atmospheric pressure, three effects have to be taken into account for their results description: 1. Energy losses of alpha particles in air (before entering into the detector body); 2. Deviations from the true values of the $1/(R^2)$ law, if the collimation was not very precisely adjusted and 3. A charge generation of electron-hole pairs which obeys the Bragg's law, and where the Charge Collection Efficiency (CCE) depends strongly on the depth from which the charge (holes or electrons) are collected.

We also tested an influence of the polymeric binder used to produce the composite detector on its detecting performance. Fig. 3 shows that the amplitude and shape of the alpha spectrum (hole collection, of 5.5 MeV alpha particles from ^{241}Am) obtained with composite BN in a

Nylon-6 [4] binder, is quite similar to that of the detector bound in polystyrene.

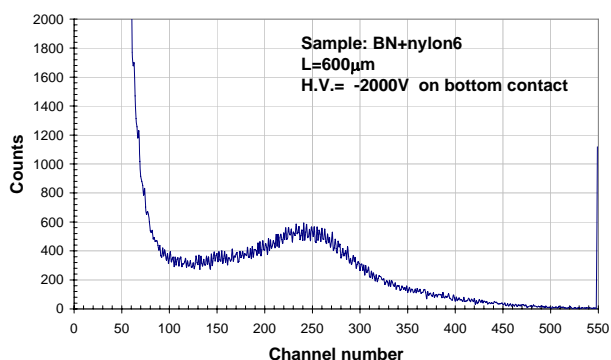


Fig. 3. Alpha spectrum (hole collection) of 5.5 MeV of ^{241}Am measured by a 0.6mm thick composite BN in a nylon-6 binder. Both contacts are Aquadag, top contact 4 mm^2 bottom contact 20 mm^2 bias 2000 V. Negative polarity on bottom contact is for holes.

The much reduced polarization of the alpha response of the composite BN detectors was already reported [8]. Some of the results will be summarized here. After an initial drop of the alpha signal in the first 0-2minutes of the measurement from 100% to 95%, there was very little additional polarization and the amplitude of the alpha peak is reduced further from 0~10 minutes, from 100% to 85% and it stayed stable at this level for another 10 minutes, at a saturation level of ~85%. Fig. 4 shows another measurement of the polarization expressed as the amplitude peak in Volts as a function of time. One can see that the amplitude is reduced from 2.98 V to its final value of ~2.75 V in about 2 minutes, and it stayed constant for another 8 minutes.

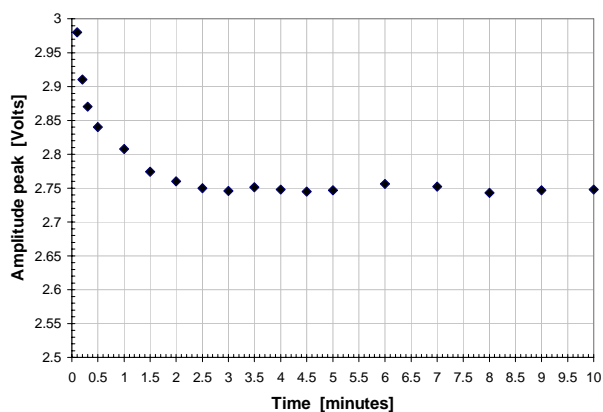


Fig. 4. The amplitude peak in Volts of the Alpha spectrum (hole collection) of 5.5 MeV of ^{241}Am measured by a 0.45mm thick composite BN in a polystyrene binder, SampleN47, area 4 mm^2 , bottom contact ITO and top contact Aquadag. H.V bias = +1600V on bottom contact, total amplification = 400,000, shaping time = 10 μsec

Details about the alpha irradiation results of different detectors measured were also reported [8]. In addition to the 5.5 MeV alpha from ^{241}Am reported in this paper, also other alpha sources such as ^{210}Po (alpha particles of 5.3MeV) and ^{226}Ra (4.8MeV) were tested and the pulse heights were linear within the multichannel energy scale.

Some of these BN detectors were tested with thermal neutrons produced by rather weak ^{241}Am - ^9Be source. The neutrons of these radionuclide sources were always thermalized using a block of paraffin [6-7]. The pulse height responses obtained with BN composite detector (0.8mm thick, effective area of 5 mm^2 , biased at 1000V for hole collection) in measurement with ^{241}Am - ^9Be source are shown in Fig. 4. The results are similar as those already reported with weak sources of neutrons [6-8].

The responses of the composite semiconductor BN detector were also studied in a pure thermal neutron beam of a flux of about 10^7 neutrons per sec per cm^2 . The measurements were carried out using neutron beam at the Nuclear Physics Institute of the Czech Academy of Sciences at Rez near Prague (NPI). Results of these first tests are shown in Figs. 5 and 6. In Fig. 5 the thermal neutron induced pulse height spectrum is compared to background one, both obtained at the bias of 800V. Exposure time was 100s. Similar spectra obtained at significantly lower biased BN detector (450V) shown in Fig. 6 demonstrate a significant decrease of detector background signal. It supports an idea, that this background signal is generated due to some kind of electric discharges in the dielectric binder. The shape of the neutron spectra in intensive thermal neutron reactor beam of about $10^7\text{ sec}^{-1}\text{ cm}^{-2}$ (Figs 5 and 6) is very similar to the shape of the spectrum shown in Fig. 4 for a very weak neutron source. We plan to study the possible electrical discharges at high bias voltage in the future to understand this phenomenon and finally, to eliminate them. However, the result shown in Fig. 6 clearly demonstrates the use of the BN device for thermal neutron detection already in the present level of technological progress.

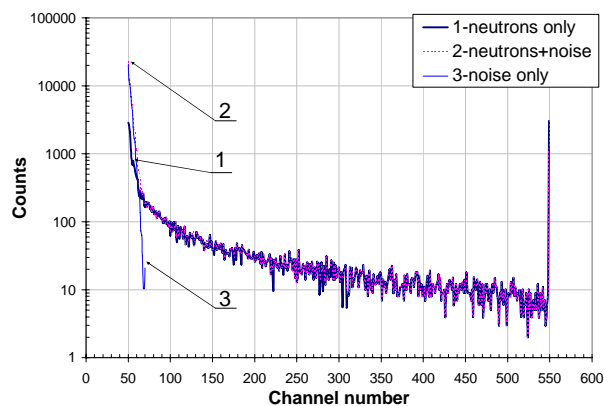


Fig. 5. The pulse height responses obtained with BN composite detector (0.8mm thick, effective area of 5 mm^2 , biased at 1000V for hole collection) in measurement with ^{241}Am - ^9Be source (activity of 100 mCi) with paraffin. The time of measurement is 34000 sec.

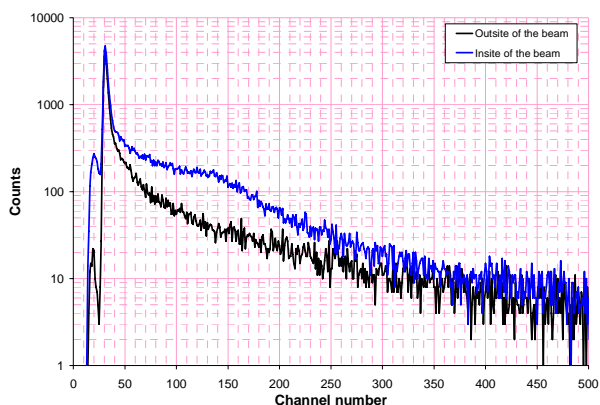


Fig. 6. Pulse height spectrum (the blue line) obtained with positively biased (+800V) BN detector when illuminated by thermal neutrons at neutron flux of about $10^7 \text{ sec}^{-1} \text{ cm}^{-2}$. The black graph shows the response of the device when placed out of the neutron beam. Exposure times were 100s.

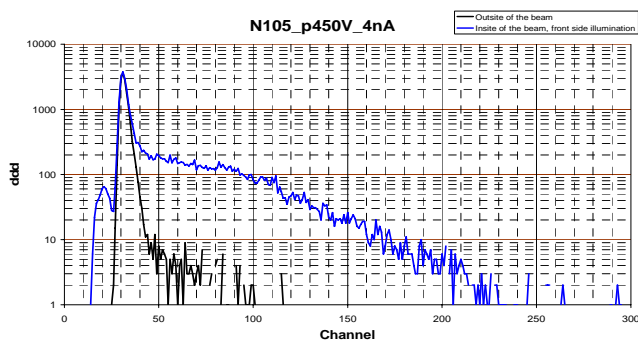


Fig. 7. Similar pulse height spectra as in Fig.5 obtained at significantly lower biased BN detector (450V). One can clearly see a significant decrease of detector background signal (the black curve, detector is placed outside of neutron beam). This result demonstrates usability of the BN device for thermal neutron detection already on the present level of technological progress.

3. Conclusions

The composite hexagonal BN device is an excellent Alpha particle detector with very little polarization showing also spectral responses. The main novel results reported here in this paper, prove that it is possible to detect both low and very high fluencies of neutrons using a planar composite natural occurring semiconducting BN imbedded in a polymeric matrix, although its content of ^{10}B is about or even less than 10%. The low temperature required to produce the device and the fact that one can use it without the expensive enriched ^{10}B can make it as a suitable candidate for large area neutron detectors.

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