Novel Boron based multilayer thermal neutron detector

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Thin layers of composite material, natural boron and boron carbide (B_4C) with natural abundance of ${}^{10}B$, mixed with binder were used as thermal neutron convertors in a multilayer thermal neutron detector design. Plane metal electrodes of 4 -100 cm² area were covered with thin (20-50 µm thick) layers of converter material and assembled in a 4-12 layers sandwich configuration, also with a pixel structure. High voltage was applied to metal electrodes to create in an interspacing electric field. The spacing volume could be filled with air, nitrogen or argon. Thermal neutrons were captured in converter layers due to the presence of ${}^{10}B$. The resulting nuclear reaction produced α -particles and ${}^{7}Li$ ions which ionized the gas in the spacing volume. Electron–ion pairs were collected by the field to create an electrical signal.

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1. Introduction

Neutron detection and developing neutron imaging systems are an actual problem of the current nuclear physics and neutron detector technology. At present time the most part of neutron detectors are based on gaseous Helium-3 (³He(n, α) H³) which provides outstanding performance as a converter in neutron detectors working in ionisation or proportional mode and excellent neutron/gamma separation. During last years the growth needs of neutron detectors create a problem of additional Helium-3, which receiving is a large technical problem. So, the practical interest now is concentrated on search of possible alternative techniques to detect neutrons. Really, only ¹⁰Boron (${}^{10}B(n,\alpha){}^{7}Li$) and ⁶Lithium (${}^{6}Li(n,\alpha){}^{3}H$) may be considered for use as alternative neutron converters in large area detectors. Resent publication by Bell et al.[1] is presented sufficiently complete review of present situation of neutron detection: different methods of neutron detection, using cryogenics and semiconductor detectors, application of different kinds of neutron-sensitive converter materials, which contain the neutron-capturing nuclides and directly convert neutrons into an energy scale [2,3]. In our previous publication we have submitted for consideration experimental results on thermal neutron composite detectors based on application of pure boron nitride(BN), BN mixed with boron carbide (B₄C) and lithium fluoride (LiF) as a converters[4]. This kind of composite solid-state detectors have demonstrated some advantages such as better relation signal /noise, weak sensitivity to gamma radiation, ability producing detectors of any shape and size.

In current paper we present a further development of boron-based composite thermal neutron detectors in air (here and after) - multilayer thermal neutron detector, which has some advantages over the semiconductor, scintillation or GEM –detector (gas electron multiplier) such as: simple construction, not limited plane area and number of electrodes, possibility to realize pixel geometry of electrodes and low cost. Using ¹⁰B as a converter gives possibility to achieve detection efficiency over 50%. This detector may be successfully operated both in a count regime (in the case of low intensity neutron fields) and current regime (high intensity neutron fields). Multilayer thermal neutron detector may be applied as an image detector to control inside a metallic container combustible, explosive materials and drags (in solid or liquid state) and for the detection neutron radiation on transport control, industry et al.

2. Detection mechanism

Multilayer thermal neutron detector construction consists of parallel metallic plane plates (electrodes), which are placed with spacing of 3-6 mm and fixed in frame from isolated material. This construction looks (Fig.1) like a sandwich. Inside spacing volume is filled with air or any gas. Inner surfaces of metallic plane plates were covered by the thin layers of neutron sensitive materials (converters). Natural Boron or Boron carbide (B₄C) were embedded in organic or inorganic matrices, which act as binders to hold grains together (see details in [4]).

The neutron detection is realized through the conversion of neutrons into a detectable radiation due to the presence of 10 B atoms in natural boron and boron carbide. Reactions which were used in present devices are:

$${}^{10}\mathbf{B} + n \rightarrow {}^{7}\mathbf{Li} + \alpha$$
$${}^{10}\mathbf{B} + n \rightarrow {}^{7}\mathbf{Li}^* + \alpha + \gamma$$

As a result of these nuclear reactions [5] α -particles and ⁷Li ions flow into the spacing volume and ionized air. Electron–ion pairs are collected by applied electric field

creating an electrical signal. ¹⁰B has a cross-section of 3840 b for thermal neutrons (0.025eV) and it reacts with thermal neutrons producing alpha particles and ⁷Li. Only 6% of the ¹⁰B (n, α) reaction induced by thermal neutrons produce the ⁷Li isotope in the ground state directly with a kinetic energy of 1.01 MeV, while the energy of an alpha particle is 1.78 MeV. The major part of the reaction (94%) produces lithium nuclei in an excited state, ⁷Li*, with a kinetic energy of 0.84 MeV. The alpha particle has an energy of 1.47 MeV in this case. The excited ⁷Li* nucleus has a very short half lifetime of about 10^{-13} s, and it returns to the ground state by emitting a gamma photon of 0.48 MeV energy. In course of the nuclear reaction induced by thermal neutrons, heavy charged particles (alpha particle and ⁷Li) are emitted in opposite directions from the interaction point [6]. These particles ionize air in spacing volume and generate positive air ions and electrons which are collected at the electrode by the applied electric field creating current electrical signal (current impulse) [5] as shown on Fig.1. The electrical signal (number of electron-ion pairs) is proportional to the energy of the initial heavy charged particles.



Fig.1. Principal schema of creating signal in 2-layers boron-based composite thermal neutron detector.

It is necessary to note that gamma photons of 0.48MeV practically do not ionize gas in spacing volume and do not affect on ratio signal/noise. Low sensitivity to gamma radiation was tested in [4] and shows an additional advantage of presented detector.

3. Experimental arrangement.

There were produced and tested multilayer thermal neutron detectors with neutron converters of thin layers from composite boron or boron carbide.

4-layers construction was taken as a basic element of thermal neutron detector (Fig.2). It consists of 3 metal plane plates electrodes, assembled as sandwich with spacing 3-6 mm, and are hold in the frame from isolated material. This basic element has 2 volumes filled with air or gas (nitrogen, argon, butane). The inner surfaces of metal plates forming these volumes were covered by thin (20-50 μ m thick) composite layer of neutron converter. Bias voltage was applied to upper and lower metal plates (electrodes) and created an electric field in spacing with central electrode. The central electrode is connected to preamplifier. The electrical signal created by absorbed thermal neutrons after preamplifier next to the standard measuring schema: spectroscopy amplifier (with shaping time of 10 μ s), multichannel analyzer (MCA) with threshold discriminator. 4-layer thermal neutron detector was connected with preamplifier and formed assembly for the following testing.



Fig. 2. Basic element - (4-layers) thermal neutron detector and connection schema.

Let us suppose that every converter layer is based on natural boron enriched of ${}^{10}B$. According to the results in ref. [7-9] further usage of thin layers (1-5 μ m) of converter materials enriched with ${}^{10}B$ up to 95% permits to increase thermal neutron detection efficiency about 5-6 times.

In the case of normal flow of neutron flux on detector surface, the thickness of every layer must be optimize to reach maximum of detection efficiency. The best results of detection efficiency may be achieved in the uniform neutron field.

The thermal neutrons were received as a result of moderating in paraffin the fast neutrons (2.2 x 105 n/4 π s) emitted by radioactive AmBe alloy (²⁴¹Am + Be, activity of 100 mCi). AmBe neutron source known to emit also gamma rays (60 keV and lower energy ²⁴¹Am isotope gamma radiation, as well as 4.4 and 9.6 MeV gamma photons from ¹²C* isotope produced during α (⁹Be, ¹²C)n reaction [10]. Therefore gamma shielding was used. The AmBe source was ring-shaped and 4cm in diameter. The source was placed in the centre of the cylindrical container with 2-mm-thick Pb walls to reduce the gamma background. Accurate testing gamma-sensitivity of

4-layer detector with electrode area 100 cm^2 and $15 \mu \text{m}$ thick convertor along with the our previous results [4] have demonstrated efficiency of this Pb -shielding (60

keV and lower energy radiation was blocked near completely).

All our experiments have been hold in the measuring camera filled with the paraffin as shown on Fig.3. Detector assembly (thermal neutron detector with preamplifier) was mounted in the centre of the camera and was surrounded about 12 cm of paraffin layer. The neutron source was placed above upper surface of the detector as shown on Fig.3. It was possible to create a vacuum or air pressure in the camera as well as change air atmosphere to argon or nitrogen. The fast neutrons from AmBe source after moderating in the paraffin layer creating in the centre of camera thermal neutron cloud.



Fig. 3. Geometry of measurements in vacuum camera: 1- detector assembly (detector +amplifier), 2-Pb-cilidric container with neutron source, 3 - ring - shape AmBe neutron source, 4 - paraffin layer.



Fig. 4. Thermal neutrons spectrum detected by the tube detector BF_3 (2 cm diameter, 10 cm long) in the centre of measuring camera, emitted by AmBe source + 12 cm paraffin layer.

We have used the standard thermal neutrons tube gas detector BF₃ (2 cm diameter , 10 cm long) to test thermal neutrons. Fig.4 presents measured thermal neutron spectrum. Two peaks corresponds to different types on nuclear reaction neutrons with ¹⁰B atoms. First occurs in

94% of events – when the energy of flying alpha particles and ⁷Li ion is equal to their sum (1.47+0.84 = 2.31 MeV). Second occurs in 6% of events – when the energy of flying alpha particles and ⁷Li ion is equal to their sum (1.78+1.01 = 2.79 MeV). This reaction took place only in the case of neutron trapping in the thin gas layer near the tube wall : or alpha particle or ⁷Li ion sticks in the tube's wall and gas is ionized by only alpha particle or only by ⁷Li ion. Therefore on spectra are presented two peaks of alpha particles with energy 1.47 MeV (94%) and of ⁷Li ions with energy 0.84 MeV (94%) in the case of neutron trapping near tube's wall. As a result of our measurement there was received that AmBe source + 12 cm paraffin layer create in the testing configuration low thermal neutron flux ~200 cm⁻²s⁻¹ in the centre of the camera.

4. Results and discussion

There were manufactured multilayer thermal neutron detectors based on composite natural boron or boron carbide (B₄C). Thin layers of converter (20-50 μ m thick) on the surface of metal plane plates were fabricated by mixing neutron sensitive materials (particles dimensions 1-2 μ m) with a polymeric binder as reported elsewhere [6]. Prepared metal plates covered with converter were used to fabricate 2, 4, 8 and 12 layers detectors. Thin layers of converter (2-10 μ m thick) may be also received by vacuum deposition or spattering.

Spectrum testing and detection efficiency measurements of multilayer detectors were realized in the vacuum camera. All measurements were executed in 2 steps: counts measure with and without presence of AmBe neutron source for the same period of time. Information about summarize level of electronics noise and radiation background was tested without presence of AmBe neutron source. The net measured neutron flux was obtained by subtracting the noise counts from the total neutron and noise counts.

Table 1 presents technical parameters of fabricated multilayer thermal neutron detectors.

Number of layers (converter) in detector	Converter material	Single layer area (cm ²)	Thickness of a ingle layer (µm)	Detection efficiency measured in air (%)
2	Boron natural	24	50	2.6
4	Boron natural	16	20	6
4	Boron carbide	16	20	5
8	Boron carbide	4.5	50	7.5
12	Boron carbide	6	20	10

Table 1. Technical parameters of fabricated multilayer thermal neutron detectors.



Fig. 5. Pulse height response of thermal neutrons obtained with multilayer (4-layers) detector.

 Table 2. Specifications of the multilayer (4-layers)

 detector and obtained thermal neutron detection results (measured in air).

Converter compo- sition	Nat. B + binder	Amplification coefficient	5·10 ⁵	Count rate with source (sec. ⁻¹)	326
Upper and bottom contacts	Fe	Pulse shaping time (µs)	10	Count rate without source (sec. ⁻¹)	75
Central contact	Fe	Voltage scale (mV/channel)	10	Calculated neutrons rate (s ⁻¹)	318
One layer converter thickness (µm)	20	Bias voltage (V)	420	Threshold channel	35
Area (cm ²)	16	Measurement time (s)	3·10 ³	Signal to noise ratio	42

Fig.5 shows the thermal neutron response of the multilayer (4-layer) composite detector (see Fig. 2) measured in air. Additional characteristics of the measured sample including the obtained results can be found in Table 2.

There was fabricated and tested 12-layers thermal neutron detector. Its construction and electrical connection schema are demonstrated on Fig. 6. Fig. 7 shows the thermal neutron response of this composite detector measured in air. Additional characteristics of the measured sample including the obtained results can be found in Table 3.

Table 3.	Specifications of	f the	multilayer	(12-layers)			
detector and obtained thermal neutron detection results							
(measured in air).							

Converter	B_4C	Amplification		Count rate	
Compo-	+	coefficient	$5 \cdot 10^{5}$	with source	
sition	binder			(sec.^{-1})	124
Material		Pulse shaping		Count rate	
of electric		time (µs)	10	without	
contact	Copper			source	
				(sec. ⁻¹)	7.9
Material		Voltage scale		Calculated	
of	Delrin	(mV/channel)	10	neutron	
plastic				rate (s^{-1})	116
One layer		Bias voltage		Threshold	
converter		(V)	800	channel	40
thickness					
(µm)	20				
Area		Measurement		Signal to	
(cm^2)	60	time (s)	10^{4}	noise ratio	16



Fig. 6. 12-layer thermal neutron detector and electrical connection schema.

3- pixel detection structure was created based on idea of multilayer thermal neutron detector. The construction of central electrode (see Fig. 2) was changed: it was used plane plastic plate with metalized electrodes from both sides. These metal electrodes by the etching were divided to three (symmetrically on both sides) isolated areas – pixels, which are covered with composite converter. Pixel's area may be varied in wide range. The form of upper and lower electrodes stay without changes, only its area must be equal to summarized area of pixels. Fig. 8 presents schema and electrical connection of 3-pixel thermal neutron detector and Fig. 9 shows the thermal neutron response of this composite detector measured in air. Additional characteristics of the measured sample including the obtained results can be found in Table 4.



Fig. 7. Pulse height response of thermal neutrons obtained with multilayer (12-layers) detector.



Fig. 8. 3-pixels thermal neutron multilayer detector and electrical connection schema.

All three spectra on Fig. 5, Fig.7 and Fig.9 are demonstrated peaks corresponding to α -particles with energy 1.47 MeV, which are flied from the surface of converter layers (as a result of thermal neutrons absorption), ionized air in contacts spacing and formed an electrical signal. When α -particles and ⁷Li ions flied from the volume of converter layer, they have loosed part of their energy in converter and made a contribution into the continuous spectrum below α -peak 1.47 MeV.



Fig. 9. Pulse height response of thermal neutrons obtained with 3-pixels multilayer detector.

Table 4. Specifications of the 3-pixels multilayer detector and obtained thermal neutron detection results (measured in air).

Converter	B ₄ C	Amplification		Count rate	
compo-	+	coefficient	5·10 ⁵	with	
sition	binder	coefficient	5 10	source	
Shion	omuur			(sec.^{-1})	66.5
Upper		Pulse shaping		Count rate	
and		time (µs)	10	without	
bottom				source	
contacts	Cu			(sec.^{-1})	4.1
Central		Voltage scale		Calculated	
contact	Fe	(mV/channel)	10	neutrons	
				rate (s^{-1})	62.4
One layer		Bias voltage		Threshold	
converter		(V)	530	channel	35
thickness					
(µm)	50				
Area		Measurement		Signal to	
(cm^2)	13.5	time (s)	6000	noise	
				ratio	16.2

Additional testing was fulfilled to check multilayer detector gamma-sensitivity. For this purpose there was fabricated 4-layer detector (see Fig.2) with geometry: electrode area 100 cm², natural boron composite layer thickness 15 µm and electrodes spacing 6 mm. ²⁴¹Am (activity 10 µCi) gamma source was mounted at the height of 2 cm above upper electrode. This gamma source created 59.6 keV photons flux ~1.53x10⁶ s⁻¹cm⁻² through the lead aperture 5 mm diameter. The measurements were hold under the next conditions: amplification coefficient-500000, pulse shaping time -10 µs, voltage scale -10 mV/channel, bias voltage -700V, measurement time -3000sec, and threshold channel -30. The received detection efficiency less than 5x10⁻⁶ confirm our previous conclusion [4] of low gamma-sensitivity for composite detectors.

With the help of basic element (4-layers) thermal neutron detector (see Fig. 2) we have tested influence of gas atmosphere (gas kind and its pressure) on thermal neutron spectrum forming. The volume of vacuum camera with detector was successively filled under pressure 1 atm. by air, nitrogen or argon and thermal neutron spectra were measured.



Fig.10. Pulse height response of thermal neutrons obtained with multilayer (4-layers) detector, with filled spacing volume by air, nitrogen or argon. Converter – natural boron ~ 20 μ m thick, electrode area ~ 16 cm², bias voltage 600 V.



Fig.11. Dependence of signal amplitude on gas (argon, nitrogen, air) pressure for the spacing 5 mm and bias voltage 600 V.

Spectra on Fig.10 shows that the signal amplitude grows 3 times in argon and about 2.5 times in nitrogen in comparison with signal amplitude in air. This effect may been explained by presence of oxygen in air, which considerably worse electrons collection. The signal amplitude in nitrogen less as the signal amplitude in argon due to difference of energy E_p , which is necessary to create electron-ion pair: for argon $E_p= 26.3$ eV, and for nitrogen $E_p= 36.4$ eV. It was received result for the current pulse duration about 1 µs in the nitrogen or argon atmosphere and 50 µs for air atmosphere. So the count rate of multilayer detector may be considerably increased by the filling of spacing volume with nitrogen or argon. There was also detected that the signal amplitude nearly liner growth with gas (nitrogen or argon) in the range of pressure 0.6-1.2 atm. (Fig.11).

5. Summary and conclusion

Boron based composite materials were used to fabricate multilayer thermal neutron detector, operated in air. Suggested basic 4-layers element of detector made a possible to fabricate detectors of different area as also of different volume by the increasing number of layers (4 and more). Fabricated pixel detection structure based on idea of multilayer detector has demonstrated possibility to create thermal neutron imaging systems to control inside metallic container combustible, explosive materials and drags (in solid or liquid state) and for detection neutron radiation on transport control, industry et al. Earlier measurements [11] have demonstrated a good detection in thin boron based efficiency of thermal neutrons composite layers in comparison with widely used ³He based detectors which have much larger dimensions to reach similar detection efficiency. In addition application of thin layers of boron-based composite materials leads to reducing sensitivity to background radiation, commonly composed of gamma rays and energetic electrons.

The main advantages of the multilayer thermal neutron detector are simple construction, compact size, low cost fabrication, reproducibility and high detection efficiency.

In present paper also was shown that changing of air atmosphere in spacing volume to another gas allow to increase signal amplitude and signal to noise ratio. Using composite converter materials enriched with ¹⁰B permits sufficiently improve detection parameters.

Current work was focused on the multilayer detector and its modification fabrication technology. Comparison investigation of its parameters with widely used thermal neutron detectors is a task of future work.

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