Chapter 12
Neutron Detectors Based on ^{10}B-Containing Nanomaterials

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Abstract Due to of deep penetration ability and strong secondary ionization, neutron-radiation is the mostly dangerous ionizing radiation for humans and environment. Developments in materials science provide neutron-sensor devices with enhanced selectivity and sensitivity. Among them, detectors based on boron-containing thin films are the naturally best because of special neutron-capture properties of ^{10}B isotope. In this work, there are evaluated the key physical-technical characteristics of neutron detectors made from ^{10}B-enriched semiconducting materials: thickness of the effective working layer ~10 μm, releasing rate of the ^{10}B – n interaction products ~10^{15}/cm^3 s, electron–hole pairs generating rate in process of neutron absorption ~10^{22}/cm^3 s, rate of rise in the temperature ~10 K/s, and device mean operating time ~10^{-4} s.

Keywords Neutron detectors • Boron

12.1 Introduction

A number of different types of neutron sources are known: nuclear weapons testing or its use in hostilities; nuclear power plants and research nuclear reactors; spent nuclear fuel; radioisotopes and spread particle accelerators utilized as neutron generators in various areas of human life: radiography and damage-evaluation in materials science, industry, agriculture, environmental protection, medicine; uranium deposits; cosmic radiation.

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Nuclear reactors are most important among sources of peaceful uses. They utilize chain-reaction – neutron-induced decay of a heavy nucleus with additional neutrons emission (e.g., $U + n \rightarrow Rb + Cs + 4n$). In deposits and wastes, it takes place the spontaneous decay reaction also accompanied with neutrons emission (e.g., $U \rightarrow Br + La + 3n$).

Physical damages of living tissue caused by neutron irradiation is more dangerous than that from other types of exposure at the same flux because neutrons tend to provide high values of linear energy transfer. As for the ionization damage effects, due to the lack of electric charge neutrons are unable to cause primary ionization by detaching electrons from the water molecules predominantly constituting a tissue. Instead, they penetrate deeply and detach nuclei of hydrogen atoms – protons, which are charged heavy particles enable to cause the strong secondary ionization damages. As a result, weight factor determining the relative share of living tissue damages in equivalent absorbed dose from the neutron-irradiation is too high: from ~5 up to ~20 depending on the mean energy of neutrons in beam.

Thus, detection of neutron radiation and measuring its fluence are the particularly important tasks caused by two factors: (1) currently, too many neutron-sources are in wide use; and (2) neutron irradiation is mostly dangerous both for human health and life and environment among other types of ionizing radiation. There is an urgent need in means of the monitoring of neutron-active environmental pollutants generated as a result of nuclear power plants operational or waste influences. In addition, with an increase in neutron facilities in various fields, it becomes more important the personal dosimetric control, environmental monitoring near the neutron-active sources, as well as evaluation of leakage neutrons.

Developments in the range of sensor materials have provided devices with enhanced sensitivity for neutron radiation. Current state of the art in technology offers the ability to incorporate micro- and nano-sized neutron-radiation indicators into widely used materials such as paints, coatings, and ceramics to create composites. Among them boron-containing materials are found to be the best mainly because of ^{10}B isotope's special neutron-capture properties.

Since neutrons are uncharged their detection depends on the secondary ionizing processes induced by the products of neutron capture reactions, most important of which is the capture by a ^{10}B nucleus [1]:

$$6\ \%: {}^{10}_{5}\mathrm{B} + {}^{1}_{0}\mathrm{n} \rightarrow {}^{11}_{5}\mathrm{B} \rightarrow {}^{7}_{3}\mathrm{Li}(1.01\ \mathrm{MeV}) + {}^{4}_{2}\mathrm{He}(1.78\ \mathrm{MeV}),$$

$$94\ \%: {}^{10}_{5}\mathrm{B} + {}^{1}_{0}\mathrm{n} \rightarrow {}^{11}_{5}\mathrm{B} \rightarrow {}^{7}_{3}\mathrm{Li}(0.84\ \mathrm{MeV}) + {}^{4}_{2}\mathrm{He}(1.47\ \mathrm{MeV}) + {}^{0}_{0}\gamma(0.48\ \mathrm{MeV}).$$

Note, above reactions usually are used to detect slow, so-called thermal, neutrons. As for the general method of measuring intermediate and fast neutrons, it is to first moderate their energies to thermal values so they can be detected with a thermal neutron detector. The one of principal measurement systems is a long counter, containing a boron fluoride BF_3 tube, surrounded by an inner paraffin

moderator. Incident neutrons cause a direct response after being thermalized in the paraffin layer. Those from other directions are either reflected or thermalized by the outer paraffin jacket and then absorbed in a layer of boron oxide B_2O_3. The disadvantage of this arrangement is that the probability that a moderated neutron will be counted is not dependent on the initial energy and, therefore, no information on the spectral distribution of neutron energies is obtained.

The description on applications of boron isotopes nuclear properties had been done as early as in the brief overview [2]. Those included counters of neutrons and energy sensors for neutron-detectors. Some neutron-capture applications of boron cluster compounds were mentioned in [3, 4] as well. Neutron sensor device-structures based on boron-containing materials also were described in the newer Review [5] on the isotopic effects of boron in solid materials. Recently, in special issue [6] of the SPIE Proceedings, devoted to the achievements in radiation detectors physics, it has been described the boron-containing epitaxial layers proposed for neutron-sensor applications.

Crystalline elemental boron, which is semiconducting, was used in neutron-thermometers [7]. Neutron-sensors constructed from boron and boron-rich semiconductors offered advantages over the conventional (gas-filled) ones being more effective due to easily detectable products of the nuclear reaction ^{10}B – n.

At the first time, a boron-based neutron-detector was described in [8]. Two members of a pair of thermoresistors were matched with respect to semiconductor properties, but differed in nuclear properties: one thermistor was made of ^{10}B, the other of ^{11}B. When exposed to neutrons with thermal energies, only ^{10}B nuclei undergo transformation and the significant energy released (~2.79 MeV per a single act) causes the ^{10}B-thermistor to become warmer than the ^{11}B-thermistor. This temperature difference can be converted into an electrical signal and measured.

Formation of the rectifying contact on ^{10}B sample allowed its application in a different-type solid-state neutron detector [9]. Its principle of operation is based on the generation charge carriers due to absorbing He-nuclei, i.e. α-particles, ejected in process of neutrons interaction with ^{10}B nuclei. In the neutron detector [10] based on semi-insulating boron-containing crystal biased by an external voltage, both produced nuclei He and Li are rapidly stopped (within ~10 μm) exciting thousands of electron–hole pairs. These free charges then drift in the electric field generating a measurable current-pulse.

Boron carbide-based thermoelectric device for the detection of a thermal-neutron flux proposed in [11] exploited, on the one hand, very high melting temperature of boron carbides in combination with their radiation tolerance and, on the other hand, anomalously large Seebeck coefficient of boron carbides in proposing a relatively sensitive detector of the local heating that follows the absorption of a neutron by a ^{10}B nucleus. The possibility of elaboration of the boron carbide-based neutron detectors also was considered in [12]. Thermal neutrons cannot penetrate for more than ~1 mm into ^{10}B-enriched material before being absorbed and upon absorbing a neutron, ^{10}B decays heating the material in the immediate vicinity. A temperature difference is thereby established between the faces of a boron carbide sample. It can be monitored using large Seebeck effect. All told, boron carbides may serve as simple, small, and robust neutron detectors.

Real-time solid-state neutron detectors were fabricated [13] from different semiconducting boron–carbon alloy layers (with the natural isotopic abundance or enriched in ^{10}B) prepared by plasma-enhanced CVD method from the ortho-carborane source gas. Single neutrons were detected, while the signals induced by concomitant γ-rays were determined to be insignificant. The last is important because at many working places thermal neutrons coexist with γ-rays. Such kind of active diodes could use the inherent micro-scale spatial resolution, increasing the range of applications.

In [14], a plastic track detector was applied to thermal neutron dosimetry by combining with boron nitride BN converter using (n, α) reactions. BN seems to be a promising converter because of high boron-concentration and smooth surface. It is important to detect separately thermal neutron flux and γ-dose, in particular, to evaluate prescript dose for so-called boron-neutron-capture-therapy, which uses a high intensity neutron radiation. Recently, it has been proposed [15] the method exploiting a glass-rod-dosimeter with thermal neutron shielding: in order to evaluate thermal neutron fluence, the combination with BN as a thermal neutron converter has been developed. Epitaxial layers of h-BN synthesized by metal-organic CVD have been used in [16] for the neutron-sensor application. To address the key requirement of long carrier lifetime and diffusion length for a solid-state neutron detector, on this basis the micro-strip metal–semiconductor–metal detectors have been fabricated and tested. A good current-response was generated using continuous irradiation with a thermal neutron beam corresponding to an effective conversion efficiency of ~80 % for absorbed neutrons. Utilizing the graphene-oxide property to provide a sufficient change in luminescence when exposed to neutron radiation, it was investigated [17] the integration of h-BN with some graphene-based structures to evaluate neutron-radiation induced conductivity in nanoscale devices and discussed the successful integration of h-BN with large area graphene electrodes as a means to provide the foundation for large-area sensors.

Boron phosphide BP single crystals were studied [18] to develop refractory electric devices, such as solid-state neutron detectors utilizing a large cross section of ^{10}B isotope. Isotopic composition of the wafer used was $(^{10}BP)_{0.95}(^{11}BP)_{0.05}$.

Thermo-luminescent thermal-neutron dosimeters, which are one of known few types of personnel dosimeters commonly used for protection purposes, also utilize (n, α) reaction of ^{10}B nucleus [19]. Such kind of neutron dosimetry can be performed, e.g., by the spark counting of tracks in boron-doped film [20]. For this purpose, thin cellulose nitrate films doped with a boron compound. After thermal neutron irradiation, the films etched and then the etch-pits caused by ^{10}B (n, α) ^{7}Li reactions punched and counted at high voltages. As a highly sensitive, simple and non-radioactive neutron dosimeter, the plastic plates doped with other boron compound – ortho-carborane – were prepared in [21]. The etch-pits generated then counted using an optical microscope or an automatic track counting system. It is important to note that, these plates are insensitive to other types of rays (visible, UV, X-, β- and γ-) and are easy to handle because detector and converter are incorporated together.

Alternative boron compound, lithium tetraborate $Li_2B_4O_7$, was also considered [22] as promising converter for neutron dosimeters. The poor reliability of sintered crystals as dosimeter was overcome by making it forms of glass ceramics $Li_2B_4O_7 + 5$ % $SiO_2 + 0.01$ % Pt or thin evaporated layers on a LiF single crystal.

From the information on the thermally-stimulated-electron-emission glow pattern of the duplicated structure sample it may be possible to measure the dose of each kind of radiation separately in a mixed radiation field.

Nanostructured films of germanium and silicon doped with isotope ^{10}B are known as naturally the best materials for novel high effective and sustainable neutron-radiation sensors preparation [23]. Boron atoms, originally shallow acceptors for Ge and Si, change their charge because of nuclear reaction on ^{10}B stimulated by neutron-irradiation: it forms Li, the shallow donor for these semiconductors. Charge carriers concentration changes are easy to fix by an electrical measuring.

The detailed Review on above mentioned neutron-detectors has been given elsewhere [24].

We can summarize that, ^{10}B isotope-enriched elemental boron crystalline modifications, semiconducting boron compounds – boron carbides, nitrides, phosphides, etc., and boron-doped common semiconductor or insulating materials can serve as working bodies for effective neutron-fluence detectors of various types. They exploit effects related to the nuclear reaction ^{10}B (n, α) ^{7}Li such as transmutation, heating, and generation of charge carriers. Present work aims to estimate corresponding physical-technical characteristics of these devices.

12.2 Neutron Capture Properties of Materials

There are several advantages of ^{10}B-containing materials making them useful in designing the neutron detector devices.

Boron is a chemical element with two stable isotopes ^{10}B and ^{11}B. The natural abundance of boron stable isotopes ^{nat}B is: ~19.6 % ^{10}B and ~80.4 % ^{11}B. Both interact with neutron through elastic and inelastic scattering processes. But, ^{10}B also interacts with neutron non-elastically – through capturing. This difference makes the total cross section of a ^{10}B – n interaction strongly exceeding that for ^{11}B, as well as most of isotopes of other chemical elements. For example, the total neutronic cross-section of ^{10}B, ^{11}B, and ^{nat}B for the "room-temperature" neutron beam, i.e., with mean kinetic energy of $E \sim 0.025$ eV which is translates into mean velocity of $v \sim 2200$ m/s, equals to 3835, 0.0055, and 767 barns, respectively [25].

Thus, ^{10}B and ^{11}B being chemically identical are quite different by neutron-absorbing characteristics. The effectiveness of boron as a neutrons absorber is due to the ^{10}B isotope, while ^{11}B is essentially non-reactive with neutrons. This provides a flexibility in the use of boron-containing materials in nuclear systems, since the mixture of boron isotopes can be adjusted.

Neutronic cross section of ^{10}B exhibits the monotonous energy-dependence, $\sim 1/\sqrt{E}$, over the almost entire examined incident neutrons energy range 10^{-5}–10^{7} eV, what is not the case for isotopes of other elements: most nuclides show abrupt increases in cross section at certain narrow ranges due to resonances.

Boron belongs to the least abundant chemical elements: its content in the Earth crust makes up no more than 0.005 wt. %. However, the role of boron in forming of various solid structures is incommensurably great. Understanding of the structural diversity of boron-containing phases reduces to the B-atom's distinct electron-acceptor property. It is a reason, why all-boron materials are electron-deficient and exhibit very complex atomic structures, in which icosahedron B_{12} serves as a main building block. Acceptor behavior of B-atoms and their clusters favors formation of the huge number of borides, i.e. compounds of boron with metals, which usually are characterized by the electron-donor behaviour. Only binary B/Me crystalline compounds with chemical formulas from Me_5B to MeB_{66} can be counted approximately 250. Boron compounds with non-metals characterized by the higher electron affinity than boron, i.e., boron carbides, nitrides, oxides, etc. show lesser complicated structures. Strong B – B bonds makes all the boron-rich solids refractory and resistive against aggressive-environment, while diversity of geometric and electronic structures and, consequently, the diversity of sets of their physical properties yields extremely large sphere of their technical and technological applications.

Boron is a light element and, consequently, boron-rich compounds are low-density materials.

One more advantage of the low atomic weight is that products of the nuclear reaction ^{10}B – n are stable – non-radioactive particles.

Neutrons detection ability by any boron-containing material is determined by the higher-limit of concentration n of ^{10}B atoms. Its values can be calculated on the basis of appropriate geometric models for atomic structures of important boron-rich materials, which were described previously in [26]. As for the obtained results of estimations, they are presented in Table 12.1.

One can see that, higher-limits of the ^{10}B isotope content in materials, from which should be made neutron detectors, are too high: (0.1–2.5) $10^{23}/cm^3$. It means that, the

Table 12.1 Higher-limit of concentration of ^{10}B atoms in boron-rich materials

Material	$n/cm^3 \times 10^{23}$
Boric acid H_3BO_3	0.14
Bundle of B (2,1) nanotubes	0.27
Zirconium diboride ZrB_2	0.65
Amorphous B	1.2
α-tetragonal B	1.3
β-tetragonal B	1.3
β-rhombohedral B, solid solutions of metals in B	1.3
Boron oxide B_2O_3	1.3
α-rhombohedral B, carbides and related compounds of B	1.4
Bundle of B (2,0) nanotubes	1.6
Boron nitrides h-BN and r-BN	1.7
Bundle of B (2,2) nanotubes	2.1
Bundle of B (1,0) nanotubes	2.2
Bundle of B (1,1) nanotubes	2.4
"Close-packed" B	2.5

neutrons should be rapidly stopped in a ^{10}B-enriched material [27]. Consequently, in all the solid-state neutron-sensors based on ^{10}B-rich materials, working body should be a micro- or nano-layer.

12.3 Physical-Technical Characteristics

Let evaluate the key physical-technical characteristics of the neutron detectors made from ^{10}B-containing materials.

The depth of penetration l of neutrons into the material, in which concentration of ^{10}B nuclei is n, equals to

$$l = \frac{1}{n\sigma}$$

where σ is the cross section of ^{10}B – n interaction at the given neutron velocity.

Rate W of releasing of the nuclear reaction products is $Jn\sigma$ with J for incident intensity of neutrons. Taking into account the previous formula we get

$$W = \frac{J}{l}$$

If E, the energy released during a single act of ^{10}B – n interaction, is spent only on the thermal generation of the electron–hole pairs, their number N is equal to the ratio E/E_g, where E_g is the band gap width of the irradiated material. It is evident that the product NW is the rate R of generation of electron–hole pairs:

$$R = \frac{E}{E_g}W$$

Let C is the heat capacity per unit volume of the irradiated material. If the energy released in result of ^{10}B – n interactions is spent only on the heating, then the heating rate can be expressed as CdT/dt, where T is the temperature and t is the time. But, the same equals to EW as well. Consequently, the rate of rise in the temperature in process of neutron absorption in material should be

$$\frac{dT}{dt} = \frac{EW}{C}$$

As for the neutron detectors' typical operation time, apparently it can be estimated from the following relation:

$$\tau = \frac{l}{v}$$

where v is the neutrons mean velocity.

Calculations were performed for following input data: total neutronic cross-section of a ^{10}B nucleus for thermal neutrons $\sigma = 3835$ b; incident neutron flux intensity $J = 10^{13}/\text{cm}^2$ s, which is the value characteristic for radiation of a small nuclear reactor; $E = 2.79$ eV; band gap width of β-rhombohedral boron, the boron ground-state modification, $E_g = 1.55\,\text{eV}$ [28]; specific heat of β-rhombohedral boron at room temperature $C = 1.83 \cdot 10^{19}$ eV/K cm^3 [29]; and the velocity of thermal neutrons $v = 2200$ m/s. Values of the concentration n were taken from Table 12.1.

Estimated values of above introduced parameters are:

- thickness of the effective working layer in a semiconductor device ~10 μm;
- releasing rate of the ^{10}B decay products, lithium and helium, induced by neutrons absorption ~$10^{15}/\text{cm}^3$ s;
- generating rate of electron–hole pairs in process of neutron absorption ~$10^{22}/\text{cm}^3$ s;
- rate of rise in the temperature in process of neutron absorption ~10 K/s;
- device mean operating time ~10^{-4} s.

Using obtained results and relevant multipliers, engineers and designers of solid-state detectors of neutron radiation can easily recalculate parameters for devices of different designs and different values of the incident neutron flux.

12.4 Conclusion

On the basis of computations performed, certain recommendations can be made about improvements in physical-technical characteristics of neutron-radiation flux detecting and measuring devices made from ^{10}B-containing materials.

The general conclusion is that ^{10}B-isotopically enriched semiconducting modifications of elemental boron, semiconducting boron compounds, and boron-doped common semiconductor materials can serve as working bodies for thin-film, high-reliable, high-sensitive, and fast-acting robust solid-state electronic neutron-detectors of various types.

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