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Scintillation element for thermal neutron detection based on ZnS:Ag/carborane material

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A scintillation material containing mixtures of zinc sulfide powder activated with silver and a neutron converter dicarba-*closo*-dodecaborane(12) – ZnS:Ag/carborane – was synthesized. Its registration efficiency of thermal neutrons is 49 %. A scintillation element based on the obtained scintillation material was developed. It consists of poly(methyl methacrylate) light guide layers alternating with layers of ZnS:Ag/carborane dispersed in a transparent polymer matrix. The thermal neutron registration efficiency of the created scintillation element is 85 %.

Keywords: carborane, neutron converter, scintillation material, scintillation element, thermal neutron registration efficiency

Сцинтиляційний елемент для реєстрації теплових нейтронів на основі матеріалу ZnS:Ag/карборан. *П.М. Жмурін, В.О. Тарасов, А.Ю. Бояринцев, О.В. Колесніков, М.Л. Сібілєв, Т.Г. Сібілєва, Д.А. Єлісєєв, В.Д. Алексєєв, Ю.О. Гуркаленко*

Синтезовано сцинтиляційний матеріал, що містить суміші порошку сульфіду цинку, активованого сріблом, та конвертора нейтронів дикарба-*клозо*-додекаборану(12) – ZnS:Ag/карборан. Його ефективність реєстрації теплових нейтронів становить 49 %. На основі отриманого сцинтиляційного матеріалу розроблено сцинтиляційний елемент, що складається з шарів поліметилметакрилату, які чергуються з шарами ZnS:Ag/карборан, диспергованого у прозорій полімерній матриці. Ефективність реєстрації теплових нейтронів створеного сцинтиляційного елемента становить 85 %.

1. Introduction

Modern neutron detectors require scintillation elements with high thermal neutron detection efficiency against a gamma-ray background. This will improve the overall efficiency of the detector and optimize its design.

Widespread detectors for the thermal neutrons registration are scintillation elements based on a mixture of zinc sulfide powder activated by silver (ZnS:Ag) and a neutron converter dispersed in an optically transparent polymer matrix such as poly(methyl methacrylate), polystyrene, or epoxy resin-based compositions [1-4]. Neutron converters are isotopes that are capable of absorbing neutrons and emitting alpha particles, to which the ZnS:Ag scintillator is sensitive. The most common neutron converters are ^{6}Li and ^{10}B isotopes. A scintillation material in the form of a polymer film containing a mixture of ZnS:Ag and a thermal neutron converter has been known for more than 65 years [5]. Today, the demand for such material has increased due to the need to replace the expensive and short-lived ^{3}He counters in portal radiation monitors. Scintillation materials based on ZnS:Ag/^{6}Li and ZnS:Ag/^{10}B are at the level of gas-discharge counters in terms of selectivity of thermal neutron detection and can be used quite successfully to detect weak flows of thermal neutrons [6, 7]. In addition, they have a high response speed and lower cost compared to gas-discharge counters [8, 9].

The working principle of the ZnS:Ag converter is based on neutron interaction with atomic nuclei, which have abnormally large thermal neutron capture cross sections. As a result of this interaction, the nucleus decays with the emission of an alpha particle. For example, for ^{10}B:

$$^{10}B + n \rightarrow {}^{7}Li\ (0{,}83\ MeV) + \alpha\ (1{,}47\ MeV) + E_\gamma\ (0{,}48\ MeV)$$

In turn, alpha particles interact with the ZnS:Ag scintillator, which is sensitive to them, and the signal from this interaction is already registered by the photomultiplier.

The most common neutron converters are ^{6}Li and ^{10}B isotopes. Usually, due to the higher total energy of alpha particles emitted as a result of neutron capture, compounds enriched with ^{6}Li isotopes are used as a converter. However, the boron isotope ^{10}B has a number of advantages. The thermal neutron capture cross section of ^{10}B is about 4 times higher compared to ^{6}Li. In addition, the content of the neutron-sensitive isotope ^{10}B in natural boron is almost 3 times higher than the content of ^{6}Li in natural lithium (20 % and 7.5%, respectively). The use of lithium as an effective neutron converter is not possible without a high level of ^{6}Li enrichment, while the use of boron is possible with the natural composition of isotopes. Boron oxide B_2O_3 is traditionally used as a boron-containing converter [1, 2]; the natural content of the ^{10}B isotope in which is 6.3%. This content of the ^{10}B isotope allows the use of boron oxide without additional enrichment, but does not allow achieving the required level of neutron detection efficiency. To increase the efficiency of neutron registration, it is necessary to increase the concentration of the ^{10}B isotope. An alternative is to use a compound with a higher specific boron content as a converter. A promising example of such a compound is dicarba-*closo*-dodecaborane(12) – $C_2B_{10}H_{12}$ (carborane) with a total boron content of 75% and, accordingly, a ^{10}B isotope content of 15%. Therefore, when using carborane, the concentration of the ^{10}B isotope can be increased more than two times, with the same scintillator/converter ratio in the scintillation material. In this case, there is no need to enrich the raw material with the ^{10}B isotope.

As part of this work, the scintillation material ZnS:Ag/carborane was obtained and investigated. A scintillation element on the basis of the obtained material was manufactured and its registration efficiency of thermal neutrons was established.

2. Experimental

To obtain the scintillation material, o-carborane was used. But it should be noted that there is no fundamental difference which of the carborane isomers or their mixture are used to obtain the scintillation material. Additionally, the following materials were used: ZnS:Ag, synthesized on the basis of ISMA NAS of Ukraine, and Toluene (analytically pure) according to TUU 20.1-05761264-320:2019.

Preparation of scintillation material ZnS:Ag/carborane. 2 ml of toluene is poured into a 50 ml beaker, the calculated amount of carborane is added and stirred until complete dissolution. Then 2 g of ZnS:Ag is added and heated to a temperature of 80 ºC with constant stirring, and maintained at this temperature with stirring until a dry powder is obtained. The resulting powder is placed in a drying oven heated to 120 ºC and kept for 30 min for the final removal of toluene.

Obtaining a scintillation element based on ZnS:Ag/carborane. To manufacture a scintillation element, three light guide plates measuring 44 mm × 29 mm × 3 mm are cut from a poly(methyl methacrylate) (PMMA) sheet and the surfaces that will be in contact with the layers of the ZnS:Ag/carborane scintillation material are polished.

To create one scintillation layer, 0.42 g (70 wt. %) of the scintillation material ZnS:Ag/carborane is dispersed in 0.18 g (30 wt. %) of the optically transparent two-component epoxy glue Magic Crystal 3D Clear. The resulting mixture is vacuumed for 5 min to remove air bubbles and applied evenly to the light guide using a doctor blade device; then another light guide is placed on top, forming a scintillation layer with a thickness of 200 µm, which is left to harden under normal conditions for 12 hours. In the same way, the next scintillation layer alternating with the light guide is formed. Then the scintillation layers are applied to the two outer surfaces of the light guides and left to cure.

Table 1 – Registration efficiency of thermal neutrons for different compositions of ZnS:Ag/carborane scintillation material in a layer with a thickness of 200 μm

№	Composition of scintillation material, wt. %		Thermal neutron registration efficiency η_n, %
	ZnS:Ag	Carborane	
1	97	3	22
2	93	7	43
3	86	14	49
4	80	20	32
5	75	25	26

The manufactured multilayer structure is ground and polished on all end faces, after which its total size is about 40 mm × 25 mm × 9.8 mm. The resulting blank of the scintillation element is covered with two layers of a 200-μm-thick reflective film of polytetrafluoroethylene (PTFE) on five sides; the sixth side (the smaller end side) is left free (Fig. 1).

Measurement of thermal neutron registration efficiency of scintillation material ZnS:Ag/carborane and scintillation element based on it. To measure the efficiency of neutron registration η_n, a layer of scintillation material is placed between a glass window and an aluminum substrate measuring 40 mm × 40 mm. Measurements of the efficiency of neutron registration η_n are carried out on a setup with a Lecroy 2249a analog-to-digital converter, a Hamamatsu R1307 photomultiplier tube (PMT) and a certified ^{239}Pu-Be source of fast neutrons with a flux of $1\times10^5\times$n/s ±10% according to the passport. To obtain thermal neutrons, the ^{239}Pu-Be source is placed in the center of a spherical neutron moderator made of polyethylene with a diameter of 150 mm with a certified transformation coefficient of fast neutrons into thermal ones (8 % with an error of ±5%). A sample of scintillation material is placed directly on the entrance window of the PMT. Thermal neutrons are absorbed using a screen (1 mm thick) made of metallic cadmium with a natural isotopic composition. The neutron count rate is measured with and without the cadmium screen. The difference in the resulting count rates («cadmium difference») is the thermal neutron count rate. The ratio of this value to the calculated value of the flow of thermal neutrons through the scintillation material layer is the thermal neutron registration efficiency η_n.

The efficiency of neutron registration of the scintillation element is measured similarly to the efficiency of neutron registration of scintillation material. However, in the case of a scintillation element, it is mounted directly on the entrance window of the PMT with the side free from the reflective coating.

3. Results and discussion

The mean free path of alpha particles in a solid is from one to several tens of microns. This length determines the thickness of the layer of neutron-sensitive substance covering the particles of the ZnS:Ag scintillator, at which the maximum efficiency of neutron registration is achieved. If the thickness of the layer is greater than the optimum, then not all alpha particles will be able to reach the scintillator, and if it is less, then the amount of ^{10}B atoms may not be enough to capture all neutrons. In both cases, the efficiency of neutron registration will decrease. That is, this thickness of the layer determines the optimal ratio of ZnS:Ag and boron-containing compounds.

Therefore, the efficiency of neutron registration is determined by the ratio of scintillation and neutron-sensitive components in the mixture and requires determination of the optimum.

In the work, neutron-sensitive scintillation materials ZnS:Ag/carborane with different ratio of scintillator and neutron converter were obtained and their efficiency of neutron registration was determined. The converter content in the scintillation material varied within the range of 3 - 25 wt. %. For each composition, the efficiency of neutron registration was determined (Table 1).

Analysis of the obtained results showed that the maximum neutron registration efficiency of 49 % (Table 1) is achieved when the carborane

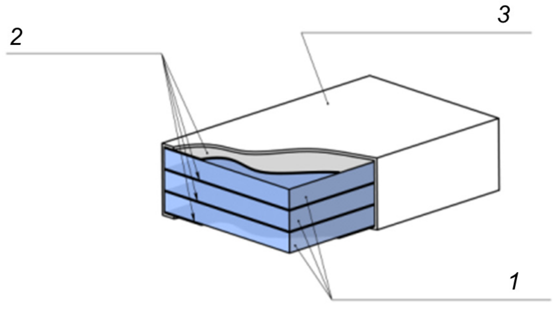

Fig.1. Scheme (a) and photo (b) of a scintillation element for thermal neutrons registration: 1 – PMMA light guide; 2 – layer of ZnS:Ag/carborane scintillation material dispersed in a transparent polymer matrix; 3 – light-reflecting PTFE film.

content in the scintillation material is at the level of 14 wt. %.

The obtained level of neutron registration efficiency for the scintillation material ZnS:Ag/carborane exceeds the characteristics of similar materials in which neutron converters were boron oxide $^{10}B_2O_3$ (37-39 %) [1-2] and ^{6}LiF (29 %) [3-4]. In addition, as noted earlier, the use of carborane does not require additional enrichment with a neutron-sensitive isotope to achieve high neutron detection efficiency.

However, the neutron detection efficiency of a single layer of scintillation material is still insufficient to create an efficient scintillation element. This problem is solved by increasing the overall efficiency of the detector by using several layers of scintillation material in one scintillation element. A multilayer thermal neutron detector was manufactured using the ZnS:Ag/carborane scintillation material composition, which has the highest neutron detection efficiency (Fig. 1). The detector consists of three layers of PMMA alternating with four layers of ZnS:Ag/carborane scintillation material dispersed in a transparent polymer matrix; it is coated on five sides with a reflective material made of PTFE. The use of several layers of scintillation material in the detector allows for an increase in the overall efficiency of neutron detection. Polished plates of polymethyl methacrylate act as a retarder and an efficient light guide for outputting the scintillation signal to the PMT. The created design of the detector with four layers of ZnS:Ag/carborane scintillation material has an efficiency of thermal neutrons registration at the level of 85 %, which is quite a high result.

The ZnS:Ag/carborane scintillation material and the neutron-sensitive scintillation element based on it can be used to develop high-precision neutron detectors for various applications. At the same time, the use of the ZnS:Ag/carborane scintillator makes it possible to further increase the efficiency of thermal neutron registration of the scintillation element due to the optimization of its structure.

Further development of the manufacturing technology of the considered neutron-sensitive scintillation element based on the ZnS:Ag/carborane material has a promising continuation in the field of using additive manufacturing methods (3D printing). Thus, the use of ZnS:Ag/carborane scintillation material dispersed in a thermoplastic transparent polymer matrix creates the possibility of using FDM (Fused Deposition Modeling) printing technology. Further research in the field of using 3D printing to create scintillation elements will significantly reduce the labor intensity of the manufacturing process of detectors of any geometric complexity, which is aimed at increasing the economic efficiency of production.

The ZnS:Ag/carborane scintillation material and a neutron-sensitive scintillation element based on it can be effectively used to develop high-precision neutron detectors for various applications.

4. Conclusions

A ZnS:Ag/carborane scintillation material has been developed, with a thermal neutron detection efficiency of 49% in a single 200 μm thick layer.

A scintillation element was manufactured, consisting of three layers of light guide made of poly(methyl methacrylate) alternating with four layers of the obtained ZnS:Ag/carborane scintillation material. The efficiency of thermal neutrons registration of the developed scintillation element is at the level of 85 %.

The obtained ZnS:Ag/carborane scintillation material and a neutron-sensitive scintillation element based on it can be used to create highly efficient detectors for registering thermal neutrons against a gamma-ray background for high-energy physics, nuclear power, environmental monitoring, and other fields of science and technology.

The possibility of dispersing the scintillation material ZnS:Ag/carborane in a thermoplastic transparent polymer matrix opens up prospects for its application in the field of additive manufacturing methods (3D printing), namely in the FDM (Fused Deposition Modeling) technology.

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