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ELEMENT-LOADED ORGANIC SCINTILLATORS FOR NEUTRON AND NEUTRINO PHYSICS

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New types of element-loaded (B and Gd) organic scintillators for neutron detection and neutrino experiments have been synthesized recently at JINR. Their optical, spectral, scintillation and radiopurity characteristics are presented and discussed. It is shown that the 5 % B-loaded scintillator has a light output as much as 70 % relative to the unloaded one. The same characteristics for the 3 % Gd-loaded sample is equal to 51 %. Transparency and other properties of the produced scintillators did not change at normal conditions for at least one year.

В работе сообщается о новых типах элементосодержащих (B и Gd) органических сцинтилляторов, полученных в ОИЯИ. Обсуждаются их оптические, спектральные и сцинтилляционные свойства. Показано, что световыход пластмассового сцинтиллятора, содержащего 5 % бора, составляет 70 % от световыхода аналогичного по геометрии сцинтиллятора, не загруженного бором. Аналогичная характеристика для пластмассового сцинтиллятора, содержащего 3 % гадолиния, составляет 51 %. Свойства новых сцинтилляторов стабильны в обычных условиях хранения как минимум в течение одного года.

INTRODUCTION

Element-loaded organic scintillators (plastic and liquid) offer some potential advantages as detectors for neutrons and neutrinos. This has led to widespread use of developed materials both in solar and atmospheric neutrino, neutrino oscillations, neutrinoless double beta decay future experiments and for precise neutron spectroscopy with high level of sensitivity as well as for other approaches both in basic and applied physics. Modern neutrino physics became a large-scale operation, requiring large detectors to intercept the dispersed particles. New-look large (kilotons) neutrino detectors could be mounted using state-of-the art detector technologies and must be placed in facilities with very low level of inner neutron and gamma-ray background. Wide application of element-loaded organic scintillators in running and future nonaccelerator experiments is connected with relatively high cross sections of specially loaded isotopes for neutrino and neutron reactions. Such types of scintillators also offer flexible configuration with potential to discriminate between signals from gamma-ray and neutron or neutrino events. Some possible applications of element-loaded organic scintillators are presented in Table 1.

Recently new types of element-loaded (B and Gd) organic scintillators for neutron detection and neutrino oscillation experiments have been synthesized at JINR. Their optical, *Table* 1. **Applications of element-loaded organic scintillators**

Elements or isotopes	Applications
^{6}Li, ^{10}B, ^{113}Cd, ^{155}Gd, ^{157}Gd	Neutron detectors, searching for neutrinos oscillations
^{176}Yb, ^{160}Gd, ^{100}Mo, ^{37}Cl	Detection of solar neutrinos
Pb	Detection of astrophysics neutrinos
^{19}F, ^{73}Ge	Searching for Dark matter
^{150}Nd, ^{160}Gd, ^{100}Mo, ^{130}Te, ^{82}Se	Searching for double β decay
Pb, Sn, W, Hg, Bi	High energy physics

spectral, scintillation and radiopurity characteristics are presented and discussed here. Transparency and other properties of the produced scintillators did not changed at normal conditions for at least one year. Such stability in time is very important for long-term experiments. High efficiency for thermal neutron registration achieved for produced samples makes it possible to use such scintillators as a part of neutron high-sensitive spectrometers. The possibility to built large mass cost-effective detectors on the base of other element-loaded (Li, F, Pb, W, Mo, Nd, Yb) organic scintillators with high radiopurity is discussed as possible application for future nonaccelerator experiments.

Application of such materials can also be useful for neutron flux monitoring in nuclear power stations, nuclear reactors, storage life of long lived radioactive trash, and for measuring of the environment radioactivity inside civilian buildings.

1. B-LOADED PLASTIC SCINTILLATORS

Low energy neutrons ($<$ 100 keV) cannot be detected by proton recoil in usual (unloaded) plastic scintillator since they do not generate enough light. Scintillators are therefore loaded with various additives (for example, B and Gd, see Table 2) so that the neutrons can be detected by exoergic capture reactions.

Table 2. **Properties of B and Gd as targets for thermal neutrons detection and searching for neutrinos oscillations**

Isotope	Natural abundance, %	Reaction	Cross section,b	Signatures
^{10}B	19.6	(n,α)	$3.8\cdot10^{3}$	$\alpha(1.74\ \mathrm{MeV}) + {}^{7}\mathrm{Li}(0.84\ \mathrm{MeV})$
^{155}Gd	14.7	(n,γ)	$6.1\cdot10^{4}$	γ burst: up to 8 MeV
^{157}Gd	15.7	(n,γ)	$2.6\cdot10^{5}$	γ burst: up to 8 MeV

Up to now widely used organic scintillators for thermal neutron detection were boron-loaded materials. It is connected with high (n,α) cross section of ^{10}B as well as relatively high natural abundance of it (19.6 %). Thermal neutrons may subsequently be captured in boron-loaded scintillator by ^{10}B, which has a thermal cross section of 3838 b. The Q-value of $^{10}\mathrm{B}(n,\alpha)^{7}\mathrm{Li}$ reaction is 2.79 MeV with 2.31 MeV going to the charged particles (1.48 MeV for α) along with a 480 keV γ ray (94 % of the time):

$$n + {}^{10}\mathrm{B} \longrightarrow {}^{7}\mathrm{Li}^{*} + \alpha + 2.31\ \mathrm{MeV}$$
$$\longrightarrow{}^{7}\ \mathrm{Li}^{*} + \gamma\ (480\ \mathrm{keV}).$$

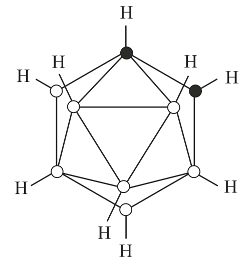


Fig. 1. Chemical structure of o-carborane: • — carbon atoms; ∘ — boron atoms

Several boron compounds have been considered for use in plastic scintillators [1–8]. As a result of our investigation o-carborane (Fig. 1) has been selected as one of the most perspective chemical compounds with mass fraction of boron as much as 75 %.

First samples of polystyrene-based plastic scintillators containing 0.38 %, 0.75 %, 2 % and 5 % mass fraction of boron have been produced by using a method of thermal polymerisation. p-Terphenyl and POPOP have been used as activator and waveshifter.

The results of investigations of the optical, spectral, scintillation, and neutron capture characteristics showed good perspectives of such scintillator. It was shown that 5 % B-loaded scintillator has a light output as much as 70 % relative to the unloaded one and is very effective for thermal neutron detection. Some characteristics of the obtained B-loaded scintillator samples (30 mm in diameter, 9 mm thickness) are presented in Table 3 and in Fig. 2.

Table 3. **Properties of B-loaded plastic scintillators**

Characteristics	Mass fraction of boron, %				
	0	0.38	0.75	2.00	5.00
Density, g/cm^3	1.05	1.05	1.05	1.05	1.05
Index of refraction	1.575	1.575	1.575	1.575	1.540
Number of B atoms per cm^3, 10^{22}	0	0.02	0.04	0.12	0.28
Number of H atoms per cm^3, 10^{22}	4.85	4.85	4.85	4.86	4.86
Number of C atoms per cm^3, 10^{22}	4.85	4.83	4.82	4.75	4.58
$\lambda_{\max}$ of luminescence, nm	421	421	421	422	423
Transmission ($\lambda_{\max}^{\mathrm{lum}}$), %	86.8	86.3	86.9	85.7	84.8
Light output, %	100	97	88	78	70
Thermal neutrons ($E \leq 0.5$ eV) detection efficiency, %	0	3	5	13	22

Influence of the boron concentration in scintillators on their light output has been carefully investigated with using a set of α, β, γ sources (see Table 4).

Using of different types of radiation makes it possible to study surface and inner uniformities of the scintillation medium as well as energy resolution and α/β quenching factor (relative energy conversion coefficient). It was obtained that α/β value is equal to (0.082 ± 0.008) for the energy $E_\alpha = 5$ MeV. Light output of the boron-loaded scintillator samples as a function of B mass fraction is presented in Fig. 3.

To investigate thermal neutron detection efficiencies of the B-loaded samples both Pu-Be neutron source with moderators and time-off-flight technique with using the Pulse Fast Neutron Reactor (IBR-30, JINR) have been applied. Scheme of measurements with the Pu-Be source set-up is given in Fig. 4. The spectra accumulated with Pu-Be source for all B-loaded samples are presented in Fig. 5.

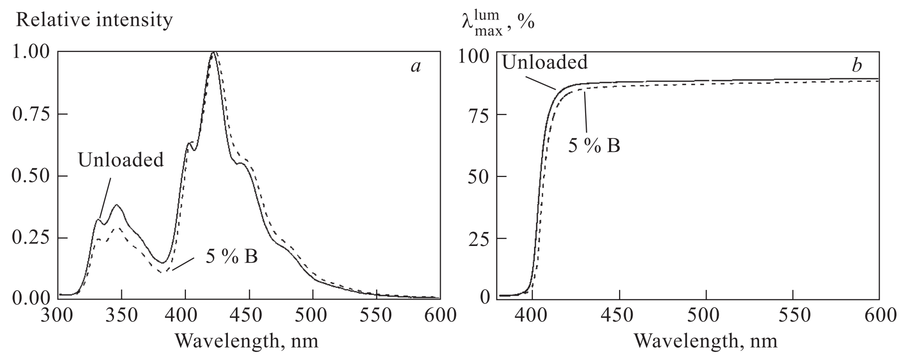
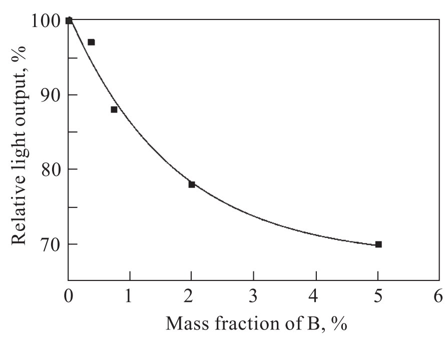


Fig. 2. Photoluminescence (*a*) and transmission (*b*) (relative to air) spectra of the B-loaded plastic scintillators samples

Table 4. **Calibration sources used for light output measurements**

Type of source	Isotope	Energy, MeV
α	^{148}Gd + ^{241}Am	$3.18 + 5.49$
α	^{244}Cm	5.80
β	^{207}Bi	$e_k = 0.976$
β	^{137}Cs	$e_k = 0.625$
γ	^{137}Cs	0.662
γ	^{60}Co	$1.17 + 1.33$



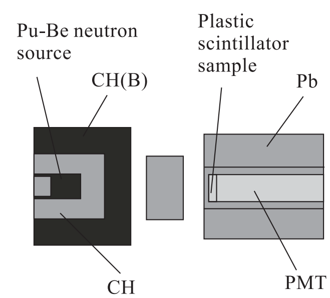
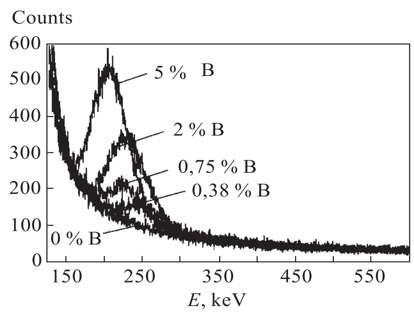


Fig. 3. Light output of the boron-loaded scintillator samples vs B mass fraction

Fig. 4. Scheme of measurements with the Pu-Be source set-up

One can see that the net area under the neutron induced α peak depends on boron's contamination. To estimate thermal neutron detection efficiencies (Table 3 and Fig. 6) the net α peak areas obtained after subtraction of the baseline (unloaded scintillator) were used. As it followed from data presented in Table 3, the efficiency for thermal neutron detection is achieved as high as 22 % just for the small size control sample with 5 % of B.



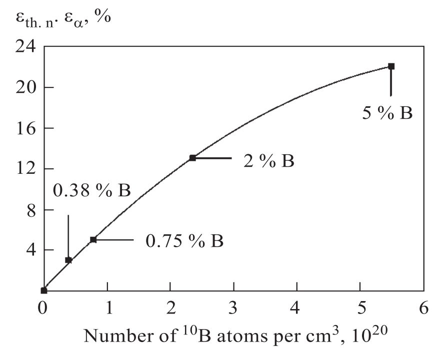


Fig. 5. Spectra of α particles induced by thermal neutron capture in ^{10}B

Fig. 6. Dependence of thermal neutron detection efficiency upon the ^{10}B concentration (for the samples 30 mm in diameter, 9 mm thickness). Detection efficiency of the unloaded sample is assumed to be equal to zero

Owing to this fact, a prototype of thermal neutron detector (70 mm in diameter, 70 mm thickness) on a base of the 5 % B-loaded plastic scintillator has been produced recently. This pilot detector has very suitable scintillation characteristics. Namely, it has 70 % light output relative to a control unloaded scintillator, good uniformity of light collection, and 12 % of energy resolution (at 1 MeV) in comparison with 11 % for the unloaded one. Investigation of the thermal neutron capture efficiency is in progress now by using both neutron sources and neutron beam. Our special interest was to measure radioactive contaminations in the produced scintillators aimed to test their ability to use in low background nonaccelerator experiments. For such purposes a radiopurity of the pilot B-loaded detector has been investigated in the underground low background laboratory of the Baskan Neutrino Observatory (BNO INR RAS). As a result only limits on the ^{232}Th, ^{238}U, and ^{40}K contamination at the level of $< 10^{-9}$ g/g were obtained that gives a good chance for using such scintillators in very low background set-ups.

2. Gd-LOADED PLASTIC SCINTILLATORS

Synthesis of gadolinium-loaded plastic scintillators is considered as one of other perspective directions of such investigations. Scintillators contained gadolinium are very interested for thermal neutron detection too, because this element has two isotopes with high enough natural isotope abundance: ^{155}Gd (14.7 %) and ^{157}Gd (15.7 %) as well as very high thermal neutron capture cross-sections ^{155}Gd ($6.1 \cdot 10^4$ b) and ^{157}Gd ($2.6 \cdot 10^5$ b).

Scintillators with more than 0.5 % Gd have not yet been produced [9–11]. It has been shown that the solubility of the complex of gadolinium nitrate with hexamethylphosphotriamide in methylmetacrylate can be achieved as much as 30 %. This compound has been used in present work as Gd-contained additive. Total chemical composition used for Gd-scintillator synthesis on the base of polymethylmethacrylate is the following: naphthalene, 2.5-diphenyloxazole (PPO) and POPOP. The obtained results showed us a high probability of successful synthesis of gadolinium-loaded plastic scintillators.

Table 5. **Properties of Gd-loaded plastic scintillators**

Characteristics	Mass fraction of Gd, %			
	0	1	2	3
Density, g/cm^3	1.172	1.182	1.195	1.204
Index of refraction	1.480	1.475	1.466	1.457
Number of Gd atoms per cm^3, 10^{22}	0	0.0047	0.0095	0.0143
Number of H atoms per cm^3, 10^{22}	5.41	5.39	5.38	5.35
Number of C atoms per cm^3, 10^{22}	3.82	3.73	3.64	3.53
$\lambda_{\max}$ of luminescence, nm	424	424	425	425
Transmission ($\lambda_{\max}^{\text{lum}}$), %	82.8	80.7	78.2	75.2
Light output, %	100	79	62	51
Thermal neutrons ($E \leq 0.5$ eV) detection efficiency, %	0	9	10.5	12.5

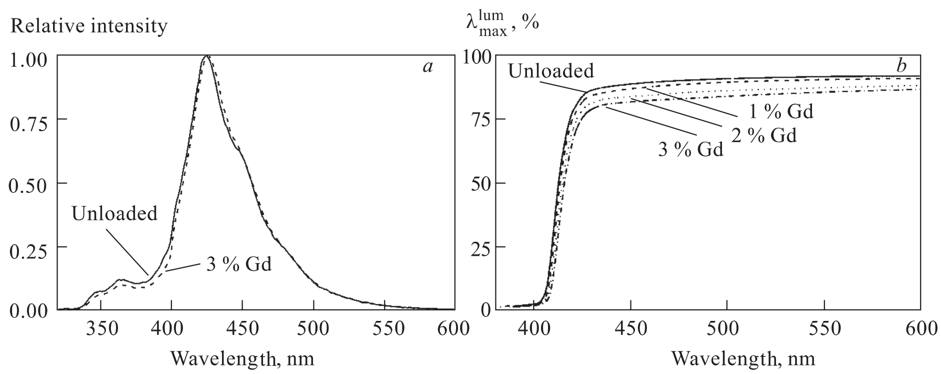
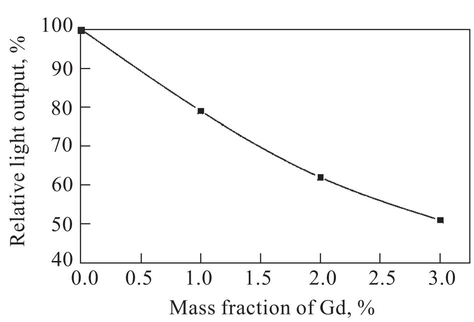


Fig. 7. Photoluminescence (*a*) and transmission (*b*) (relative to air) spectra of the Gd-loaded plastic scintillators samples



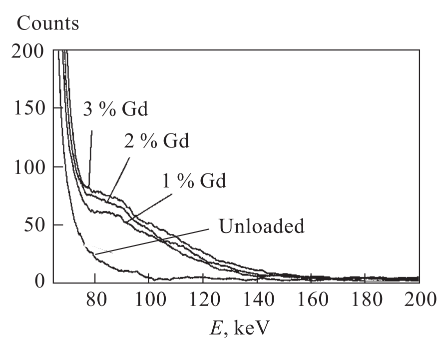


Fig. 8. Light output of the Gd-loaded plastic scintillator samples vs Gd mass fraction

Fig. 9. Spectra of gammas induced by thermal neutron capture ^{155}Gd and ^{157}Gd (for the samples 30 mm in diameter, 10 mm thickness)

Samples of plastic scintillators containing 1 %, 2 % and 3 % of gadolinium have been produced recently. It was shown that 3 % Gd-loaded scintillator has a light output as much as 51 % relative to the unloaded one. The characteristics of the produced Gd-loaded scintillator samples (30 mm in diameter, 10 mm thickness) are presented in Table 5 and in Figs. 7, 8, 9, and 10. To measure light output and thermal neutron detection efficiency the same methods as for the B-loaded samples were used. The difference is that for the case of the Gd-loaded scintillators a total efficiency depends in higher degree on a sample size due to smaller gamma detection efficiency in comparison with an alpha one.

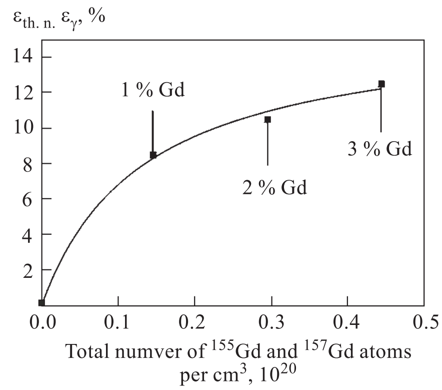


Fig. 10. Dependence of total (thermal neutron and gamma) detection efficiency on the ^{155}Gd and ^{157}Gd concentration for the Gd-loaded samples (for the samples 30 mm in diameter, 10 mm thickness). Detection efficiency of the unloaded sample is assumed to be equal to zero

3. Gd-LOADED LIQUID SCINTILLATORS WITH HIGH FLASH POINT

Liquid scintillators containing Gd are also needed for several planned solar neutrino and neutrino oscillation experiments. One of the main requirements for such scintillators is their high value of a flash point. So, our next attempt was to find a set of multicomponent organic solvents, which can satisfy above-mentioned requirement. As a result the composition of gadolinium nitrate with α methylnaphthalene (flash point 82 °C), tributylphosphate (flash point 193 °C), and 2-(-biphenylyl)-5-phenyloxazole (BPO) has been chosen for such a purpose.

The samples of liquid scintillators with concentration of gadolinium 17.3, 51.3, 61.8, and 85.9 mg · ml^{-1} have been produced and tested. The characteristics of the produced Gd-loaded liquid scintillator samples are presented in Table 6 and in Figs. 11, 12.

Table 6. **Properties of Gd-loaded liquid scintillators**

Characteristics	Concentration of Gd, mg · ml^{-1}				
	0	17.3	51.3	61.8	85.9
Boiling point, °C	245	245	245	245	245
Flash point, °C	> 82	> 82	> 82	> 82	> 82
Density, g/cm^3	0.99	1.02	1.07	1.09	1.12
Index of refraction	1.500	1.503	1.508	1.515	1.520
Number of Gd atoms per cm^3, 10^{22}	0	0.0066	0.0196	0.0263	0.0328
$\lambda_{\max}$ of luminescence, nm	395	397	398	398	400
Transmission* ($\lambda_{\max}^{\text{lum}}$), %	47.5	54	54.5	53.6	55.8
Light output, %	100	67	41	37	26

*Quartz cell, length — 5 cm, relative to air.

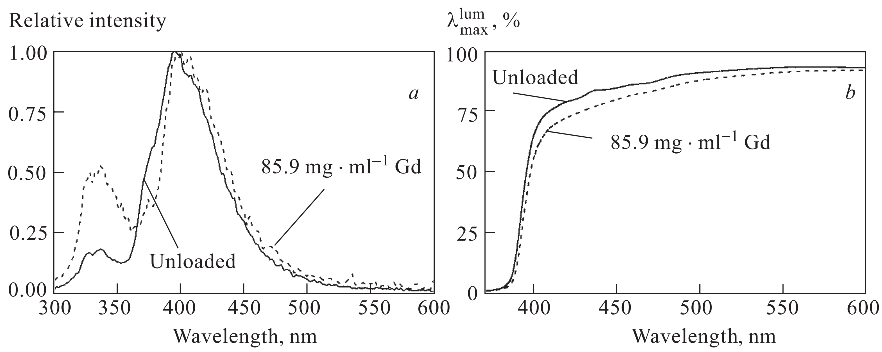


Fig. 11. Photoluminescence (*a*) and visible range transmission (*b*) spectra (quartz cell, length — 5 cm, relative to air) of the liquid Gd-loaded scintillators

CONCLUSION

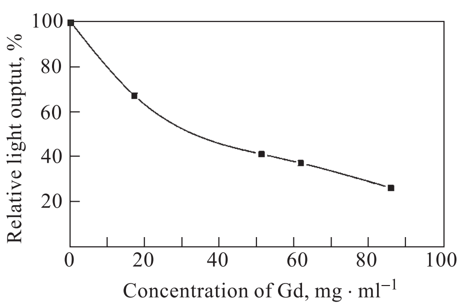


Fig. 12. Light output of the Gd-loaded liquid scintillators vs concentration of Gd

Recent achievements of chemistry and technology of organoelement and polymeric compounds and complexes have been involved to produce new types of boron- and gadolinium-loaded organic scintillators. The developed methods allowed us to load in plastic scintillators as much as 5 % B and 3 % Gd practically without losses of their quality. Transparency and other properties of the produced scintillators did not change at normal conditions for at least one year.

The Gd-loaded liquid scintillator with relatively high (> 82 °C) flash point and contamination of Gd up to 85 mg · ml^{-1} has been produced, too. High efficiency for thermal neutron registration achieved for produced samples makes it possible to use such scintillators as a part of high sensitive neutron spectrometers. It is shown that the level of measured radiopurity of the produced scintillators is low enough for using in large mass cost-effective detectors for future nonaccelerator experiments.

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