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Radiation-resistant plastic scintillator on a polystyrene-polyphenymetylsiloxane mixed base

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In this work, plastic scintillators based on mixed polystyrene-polyphenylmethylsiloxane have been created, and their mechanical and scintillation characteristics have been studied. The created plastic scintillators based on polystyrene containing 30 wt. % polyphenylmethylsiloxane, demonstrated radiation resistance reaching 22 Mrad. The obtained PSs retain their physical and mechanical properties at a level that ensures ease of mechanical processing of the material and makes it possible to obtain samples in a wide range of shapes and sizes.

Keywords: polymer base, polystyrene, polyphenylmethylsiloxane, plastic scintillator, microhardness, light output, radiation resistance.

Радіаційно стійкий пластмасовий сцинтилятор на змішаній основі полістирол-поліфеніметилсилоксан. *Д.А. Єлісєєв, О.В. Єлісєєва, Ю.О. Гуркаленко, П.М. Жмурін, В.Д. Алексєєв, Л.О. Мірошниченко*

У роботі створено ряд пластмасових сцинтиляторів на змішаній основі полістирол-поліфенілметилсилоксан та досліджено їх механічні і сцинтиляційні характеристики. Створено ПС на основі полістиролу, що містить 30 мас. % поліфенілметилсилоксану, радіаційна стійкість якого сягає 22 Мрад. Фізико-механічні властивості отриманих ПС залишаються на рівні, який забезпечує легкість механічної оброки матеріалу та надає можливість отримання зразків у широкому діапазоні форм і розмірів.

1. Introduction

Polystyrene-based plastic scintillators (PS) are widely used for detection of ionizing particles in high-energy physics experiments. However, with the development of accelerator technology, the requirements for radiation resistance of materials used in various detectors have increased, therefore, traditional PS with a radiation resistance threshold of 1-5 Mrad are no longer relevant [1-3]. Modern research in high-energy physics at high-current accelerators requires PS with radiation resistance of more than 10 Mrad and mechanical strength (Vickers microhardness) at a level of at least 100 MPa [4-6]. Standard polystyrene-based plastic scintillators have mechanical properties (Vickers microhardness HV=250 MPa) that provide a number of technological advantages: ease of processing, the ability to obtain scintillators of any shape and size, resistance to external physical impact. However, they have a rather low radiation resistance at the level of 1-2 Mrad [7, 8].

More radiation-resistant polymer bases for creating PS are polysiloxanes (for example, polyphenylmethylsiloxane) [9, 10]; since the Si-O bond is stronger than the C-C bond, the prob-

ability of damage to the polymer chain under the influence of high-energy particles is significantly reduced [11, 12]. In polysiloxane bases of PS (including polyphenylmethylsiloxane), under the action of ionizing radiation, practically no new centers for capturing excitation energy are formed, therefore their radiation resistance can reach tens of Mrad [13, 14]. However, a significant disadvantage of polysiloxanes as a PS base is their mechanical properties. Polysiloxane-based polymers are jelly-like, soft and brittle materials that require specific application conditions, such as, for example, scintillation composition polymerization directly at the site of application [15]. These properties limit the use of polysiloxanes as bases for the creation of radiation-resistant plastic scintillators.

The work attempts to combine the advantages of two polymer bases – the physical and mechanical characteristics of polystyrene and the radiation resistance of polysiloxanes. For this purpose, a series of plastic scintillators on a mixed polystyrene-polyphenylmethylsiloxane base has been created and their mechanical and scintillation characteristics have been investigated.

2. Experimental

PS samples are obtained by the method of thermally initiated free-radical bulk polymerization. In a borosilicate glass ampoule with a diameter of 20 mm, 2 wt. % p-terphenyl (p-Tp), 0.1 wt. % 1,4-bis-2-(5-phenyloxazolyl)-benzene (POPOP), 0.1 wt. % N,N'-azoisobutylonitrile (AIBN) and X wt. % (where X=5, 10, 20, 30 and 35 wt. %, respectively) of polyphenylmethylsiloxane with terminal vinyl groups [viscosity – 300-600 cSt] (hereinafter polyphenylmethylsiloxane) are loaded. Each ampoule is filled with purified styrene to a total mass of 5 g. The prepared reaction mixture is dissolved at a temperature of 55±2 °C. To remove traces of oxygen, the reaction mixture is purged with argon for 10 min, the ampoules are hermetically sealed and placed in a thermostat heated to 60 °C. Polymerization mode: 24 h – 60 °C, 12 h – 75 °C, 12 h – 90 °C, 12 h – 110 °C, 12 h – 125 °C, 2 h – 140 °C; then the ampoule is cooled at a rate of 10 °C/h to room temperature. The cooled ampoule is broken and PS samples are cut out from the resulting blank in the form of polished cylinders with a diameter of 18 mm and a height of 6 mm.

The microhardness of PS samples is determined by the Vickers method on a PMT-3 microhardness tester (measurement error ±2.5 %). Measurements are carried out at room temperature.

The light yield of the PS was measured on a scintillation spectrometer made in the CAMAC standard. The signal from the anode of the Hamamatsu R1307 photomultiplier (PMT) is fed directly to the input of the QDC LeCroy 2249A digital charge converter. The PS samples are placed directly on the surface of the PMT photocathode, optical contact is provided using an immersion liquid. Irradiation is carried out with conversion electrons with energy of 975 keV from a monoenergetic source of the radionuclide Bi-207. The light yield is determined by the maximum peak position in the amplitude scintillation spectrum. The relative light yield of the obtained samples is determined relative to the light yield of the standard plastic scintillator UPS-923A [8]. To establish the level of radiation resistance, PS samples were irradiated on a LU-40 linear accelerator with electron energy of 40 MeV (National Science Center «Kharkiv Institute of Physics and Technology» of the National Academy of Sciences of Ukraine). The integral radiation dose for all samples was 15 Mrad. The light yield of irradiated PS samples was measured the next day after irradiation.

3. Results and discussion

A series of PS was obtained on a mixed basis containing simultaneously two polymer matrices – polystyrene and polyphenylmethylsiloxane (Fig. 1). p-Tp and POPOP were used as primary and secondary luminescent additives, respectively. N,N'-azoisobutylonitrile (AIBN) was chosen as the polymerization initiator, which provides the technological possibility of obtaining a homogeneous copolymers suitable for creating PSs (the concentration of AIBN is determined by the generally accepted conditions of its use and does not significantly affect the characteristics of PSs). The formation of the polystyrene part of the PS polymer base occurs directly during the preparation of samples due to radical polymerization. The chain size of the polysiloxane part is determined by the structure of the polyphenylmethylsiloxane introduced into the composition. In this work, polyphenylmethylsiloxane with an average molecular weight of 2000-3000 g/mol was used. The combination of polystyrene and polyphenylmethylsiloxane into a single polymer base occurs due to the terminal vinyl groups of

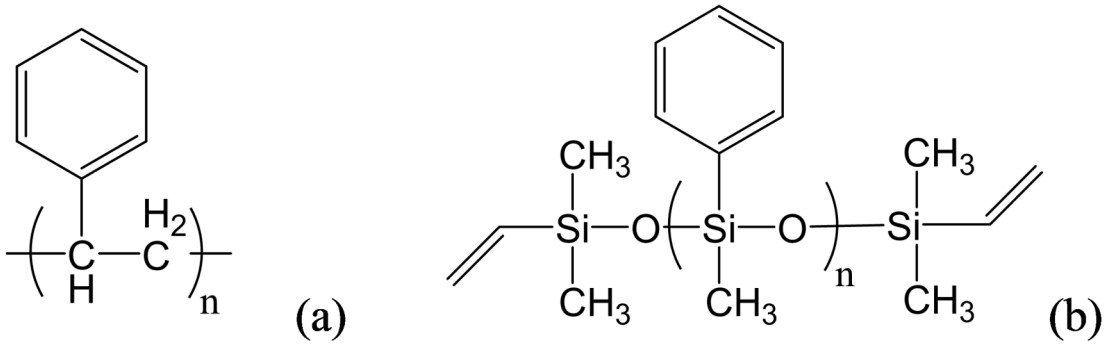

Figure 1 – Schematic representation of the structure of polystyrene (a) and polyphenylmethylsiloxane with terminal vinyl groups (b)

polyphenylmethylsiloxane, which can participate in radical copolymerization with styrene.

Polystyrene is a primary polymer base, the task of which is to provide such a level of mechanical properties of PS that will make their relatively simple mechanical processing possible. The parameter for assessing the mechanical properties of PS was the microhardness of the sample on the Vickers HV scale (MPa). As a rule, plastic scintillators with microhardness at the HV level of more than 100 MPa, can be mechanically processed without significant problems [8].

The purpose of polyphenylmethylsiloxane is to increase the radiation resistance of PS by introducing a more stable component into the polymer composition. As a parameter characterizing the radiation resistance of PS, the light yield half attenuation dose ($D_{1/2}$, Mrad) is used; this parameter does not depend on the irradiation dose and can be calculated if the light yield before and after irradiation, as well as the irradiation dose, are known [1]:

$$L = L_0 exp(\alpha D) \quad (1)$$

where L_0 and L are the light yields of the PS sample before and after irradiation; α is the degradation constant ($Mrad^{-1}$); D is the irradiation dose (Mrad).

$$D_{1/2} = (\ln 2)/\alpha \quad (2)$$

The degradation constant α is calculated from experimental data using formula (1).

A series of PSs with different contents of polystyrene and polyphenylmethylsiloxane was obtained to determine the composition with the optimal ratio of mechanical strength and radiation resistance characteristics. Table 1 shows the scintillation and mechanical characteristics of the obtained PS samples.

A series of PS samples with a polyphenylmethylsiloxane concentration of up to 30 wt. % was obtained. An increase in the polyphenylmethylsiloxane content in the scintillation composition more than 30 wt. % resulted in the production of inhomogeneous PS due to the phase separation and sample turbidity during the polymerization process (Table 1, sample 5).

The microhardness and relative light yield of the obtained PS samples were measured. After irradiation with a dose of 15 Mrad, the half attenuation dose of the light yield $D_{1/2}$ was calculated (Table 1, samples 1-4).

Table 1 – Composition of samples; relative light yield of PS before (L_0) and after irradiation (L) with a dose of 15 Mrad; ratio of light yields after and before irradiation (L/L_0); doses of half attenuation of light yield ($D_{1/2}$); microhardness according to Vickers (HV)

Sample	[Polyphenylmethylsiloxane] wt. %	L_0	L	L/L_0	$D_{1/2}$, Mrad	HV, MPa
1	5	80.1	15	0.19	4.5	230
2	10	69.2	25	0.36	10.5	215
3	20	52.5	30.6	0.58	20.0	192
4	30	49	30.5	0.62	21.9	174
5	35	–				Heterogeneous sample
UPS-923A	–	100	-	-	1-2 [8]	250
*the rest is polystyrene						

In all samples, the microhardness (by Vickers) decreases with increasing polyphenylmethylsiloxane concentration, but remains at a level not lower than 170 MPa (Table 1). This parameter value will ensure ease of material processing and the possibility of obtaining scintillators of any shape and size.

As expected, the radiation resistance of PS increases with increasing concentration of polyphenylmethylsiloxane in the scintillation composition. For a sample with 30 wt. % polyphenylmethylsiloxane, the half attenuation dose of the light yield was 21.9 Mrad, which is almost an order of magnitude higher than the radiation resistance of the standard polystyrene-based scintillator UPS-923A [8].

Thus, the obtained results indicate that the use of two polymer bases – polystyrene and polyphenylmethylsiloxane – in one scintillation composition made it possible to combine the advantages of each of the polymer bases. The use of up to 30 wt. % polyphenylmethylsiloxane in the polystyrene scintillation composition made it possible to create PS, whose light yield half attenuation dose was almost 22 Mrad. At the same time, the physical and mechanical properties of the obtained PS remain at a level that ensures ease of mechanical processing of the material and makes it possible to obtain samples in a wide range of shapes and sizes.

4. Conclusions

A series of plastic scintillators have been created on mixed polystyrene-polyphenylmethylsiloxane bases with a polyphenylmethylsiloxane content of up to 30 wt.%.

In the range of polyphenylmethylsiloxane concentration of 0-30 wt. % in polystyrene, the Vickers microhardness of the obtained PS remains at a level of not less than 170 MPa. Such parameter values ensure ease of mechanical processing of the material and provide the possibility of obtaining samples in a wide range of shapes and sizes.

PSs created on the base of polystyrene with 30 wt. % polyphenylmethylsiloxane demonstrate the light yield half attenuation dose of 22 Mrad.

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