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RESEARCH OF RADIATION RESISTANCE OF POLYMER COMPOSITE MATERIALS

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Abstract. The results of comprehensive studies of radiation shielding properties and the structure of elastic self-adhesive coverings are presented. It was found that there was no change in the linear absorption coefficient of radiation, structural damage is minor at irradiation doses which are significantly higher than standard (105 Gy).

Keywords: radiation shielding elastic self-adhesive coverings, linear absorption coefficient, radiation resistance, microstructure, EPDM

1. Introduction

Since the testing of the first atomic bomb, the number of industries in which various sources of ionizing radiation are used has steadily increased. In addition to the military-industrial complex, radiation in various forms is currently used in medicine, metrology, energy, food industry, biology and genetics, and agriculture. Ionizing radiation, with all the benefits it brings, has a negative effect on human health, and can lead to the development of serious diseases. In this regard, it is necessary to protect a person from the effects of radiation.

One of the methods of protection against ionizing radiation is distance protection [1,2]. But, if the use of this protection method is impossible, then they resort to creating protective screens. Concrete based on heavy aggregates [3,4] or metal sheets is used as traditional materials to create such screens. This type of protection has a high protective effect, but it has a number of disadvantages: high mass and thickness of screens, sometimes the impossibility of creating complex shapes, difficulties in maintenance, and dismantling.

The desire to overcome the limitations associated with the use of steel and concrete protective barriers forced researchers to pay attention to polymer composites. Composite materials are a combination of a polymer matrix and a filler. The polymer matrix consists of a binder, a plasticizer, and special additives. Due to the large number of substances that can be used as fillers and binders, the creation and use of multicomponent polymer-matrix composites in radiation shielding materials is a promising type of research [5]. The binders in the polymer matrix are high-molecular compounds with radiation resistance: polyethylene [6], synthetic rubbers [7], and resins [8]. Powders of materials with high absorption coefficients of gamma quanta (Pb, W, and their various compounds) are used as fillers [9,10].

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By combining the composition of the polymer matrix, as well as the type and amount of filler, it is possible to create different types of materials with different properties. Such materials can be used to create items for various purposes from elements of X-ray protective clothing to the manufacture of parts and housings for devices and equipment. In [11], a description of the method for determining the linear attenuation coefficient of a protective material depending on its composition is given. Thus, knowing the operating conditions, it is possible to determine in advance the most rational composition of the composite material, as well as the type and amount of filler in it.

Based on the above, it follows that the main requirement for radiation shielding materials is their preservation of the initial parameters during long-term operation under conditions of ionizing radiation [12]. According to the literature [13], in the result of such impact various physical and chemical processes take place in the materials, which can lead to deterioration in their operational parameters. In this regard, much attention is paid to the creation of special polymer composites which are resistant to the effects of radiation of different nature. Besides, one of the important requirements for radiation shielding materials is the ability to be quickly mounted and disassembled.

The materials studied in this work are a composite material with a filler content of 52% by volume. Barite and a mixture of tungsten and its oxide are used as fillers. The polymer matrix of the composite is designed to be elastic and self-adhesive, which allows quick assembly and disassembly of radiation shielding coatings.

The criterion for assessing the radiation shielding properties of the composite material and its components is the linear absorption coefficient. The linear absorption coefficient is a characteristic of the protective properties of a material, which quantitatively characterizes the relative fraction of the energy of radiation quanta lost in a substance due to absorption and scattering per unit of their path.

In this regard, the purpose of this work is to study new elastic self-adhesive radiation shielding coatings, including the dependence of protective properties and structure on the effects of ionizing radiation.

2. Methods and materials

The paper presents the results for two types of tungsten-containing samples, which differed from each other in the type of polymer matrix. Radiation resistance was estimated by the nature of damage of the samples by proton beams of different durations. Also, linear absorption coefficients were determined for the samples before and after irradiation.

A qualitative assessment of the homogeneity (mixing quality) and defectiveness of the samples before and after impact was carried out using a Universal Microscope optical microscope at a magnification of x200.

A comparative analysis of the linear absorption coefficient and defectiveness coefficient of the samples before and after proton irradiation makes it possible to evaluate the resistance of the samples to radiation.

3. The experimental part

Irradiation conditions. Assessment of protective properties. For a preliminary assessment of radiation resistance (RR), the method of accelerated radiation aging was used. The method is based on irradiating samples with an increased absorbed dose of ionizing radiation, for a time significantly less than the control time. In our case, in the sample material, due to irradiation with ionizing radiation, only structural effects associated with ionization of the sample material and uneven heating of the various components of the sample under the action of ionizing radiation will appear. Edge effects that occur at low fluxes of ionizing radiation can be ignored.

The tandem accelerator EGP-10 was chosen as the source of ionizing radiation [14]. The EGP-10 accelerator belongs to direct-acting ion accelerators and is an electrostatic rechargeable generator (tandem). In our case, the accelerator was a source of accelerated protons with an energy of 4 MeV.

Each sample was irradiated in three zones. The area of each irradiation zone was 0.0016 cm^2. The exposure time of each zone is 30, 300 and 3000 seconds, respectively. The beam current of accelerated protons was 1 nA and was recorded with a picoammeter. The calculation of the absorbed dose of proton radiation was carried out according to the formula:

$$D = E/m, \tag{1}$$

where E is the energy of absorbed radiation (J), m is the mass of the substance in which this radiation was absorbed (kg).

Mass m is calculated by the formula:

$$m = \rho \times S \times h, \tag{2}$$

where: ρ is the density of the sample (g/cm^3); S is the area of the sample in the irradiation zone (0.0016 cm^2); h is the thickness of the sample in the irradiation zone.

The energy absorbed by the sample during irradiation is calculated by the formula:

$$E = E_l \times t \times I/q_l, \tag{3}$$

where E_l is the proton energy (4 MeV) expressed in joules and is equal to 6.4×10^{-13} J; t is the irradiation time of the sample (seconds); I is the proton current (A); q_l is the charge of one proton 1.6×10^{-19} (cal).

The absorbed doses calculated by the formula (1) for the samples B52T and SM4B5 are presented in Table 1. The total error in determining the dose load during proton irradiation can be from 20 to 25%.

Table 1. Doses absorbed by samples

Sample	Absorbed dose, Gray		
	30 seconds	300 seconds	3000 seconds
B52T	42×10^3	42×10^4	262×10^5
SM4B5	40×10^3	43×10^4	493×10^5

The excess of the maximum absorbed dose, compared with the preset (105 Gy), amounted to two orders of magnitude, while the exposure time was only 3000 seconds. Such test conditions, accelerated radiation aging, are more stringent for the sample, because in this case the quantity, speed of formation of irreversible radiation defects increase in proportion to the absorbed dose.

4. Results and discussion

To estimate the radiation resistance of the samples, quantitative and qualitative measurement methods were used.

The quantitative method means in measuring linear absorption coefficients after irradiation at the EGP-10 accelerator and comparing them with the data before irradiation by protons. To study the linear absorption of gamma radiation samples as sources of gamma rays, Am-241 and Cs-137 isotope sources were used (gamma-ray energies of 59 and 661 keV, respectively).

Tables 2 and 3 present linear absorption coefficients and lead equivalents for samples B52T and SM4B5 before and after proton irradiation. The thickness of the B52T sample is 4.3 mm, and that of the SM4B5 sample is 5.3 mm.

The coefficients of linear absorption of gamma rays practically did not change (the values are within the statistical error). Based on the preliminary data obtained, it can be stated that, in the result of irradiation, the samples B52T and SM4B5 did not lose their protective properties.

Table 2. Characteristics of samples before irradiation

№	Sample	Linear coefficient μ, sm^{-1}	Lead sample coefficient, mm
For gamma rays with energy of 59 keV			
1	SM4B5	23.338	1.99
2	B52T	22.203	2.32
For gamma rays with energy of 661 keV			
3	SM4B5	0.763	2.78
4	B52T	0.773	3.47

Table 3. Characteristics of samples after irradiation

№	Sample	Linear coefficient μ, sm^{-1}	Lead sample coefficient, mm
	For gamma rays with energy of 59 keV		
1	SM4B5	23.197	1.98
2	B52T	22.283	2.32
For gamma rays with energy of 661 keV			
3	SM4B5	0.724	2.78
4	B52T	0.743	3.47

Microstructural studies. The issue of radiation shielding properties is directly related to the quality and internal structure of the material. It is known that the composite structure determines material properties, and, as a consequence, its behavior during operation.

Traditionally, the quality assessment of the structure of the sample before and after irradiation is carried out by using optical microscopy. Figure 1 shows the microstructure of the sample's surface before irradiation. As can be seen from the figure, the surfaces are morphologically identical. In the samples, the filler is distributed evenly; there are single clusters of finely dispersed filler.

After irradiation on the EGP-10 accelerator, the shape and the size of the samples did not change. Analysis of the surface structure after irradiation showed that the same changes occur in the samples. After irradiation, the interface between the structural formations of the filler in the polymer bond and the elastic matrix is more clearly revealed. The first disk-shaped cracks appear along the boundaries in the interstructural zones. Figure 2 shows the main defects that appear on the sample's surface after irradiation.

At an absorbed dose of up to 42×10^3 gray, partially opened blisters (swellings) appear, which may indicate the beginning of the destruction of the polymer matrix, which is accompanied by increased gas evolution (Fig. 2a).

At a dose of ... ~ 42×10^4 gray, cracks are observed, the pore size increases, and large filler aggregates appear (Fig. 2b).

The polymer matrix is partially loosened; small areas appear where large pores merge with cracks; filler segregation in the free volume between the particles of the polymer matrix is observed at a dose of ~250×10^5 gray (Fig. 2c).

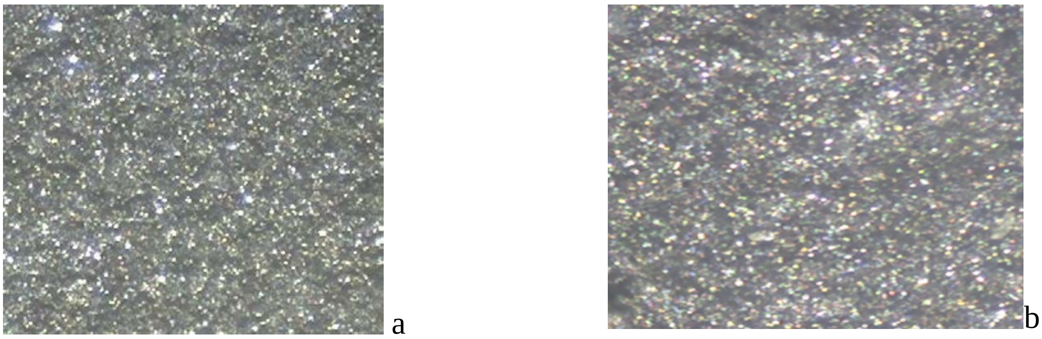

Fig. 1. Sample microstructure: a – B52T; b – SM4B5

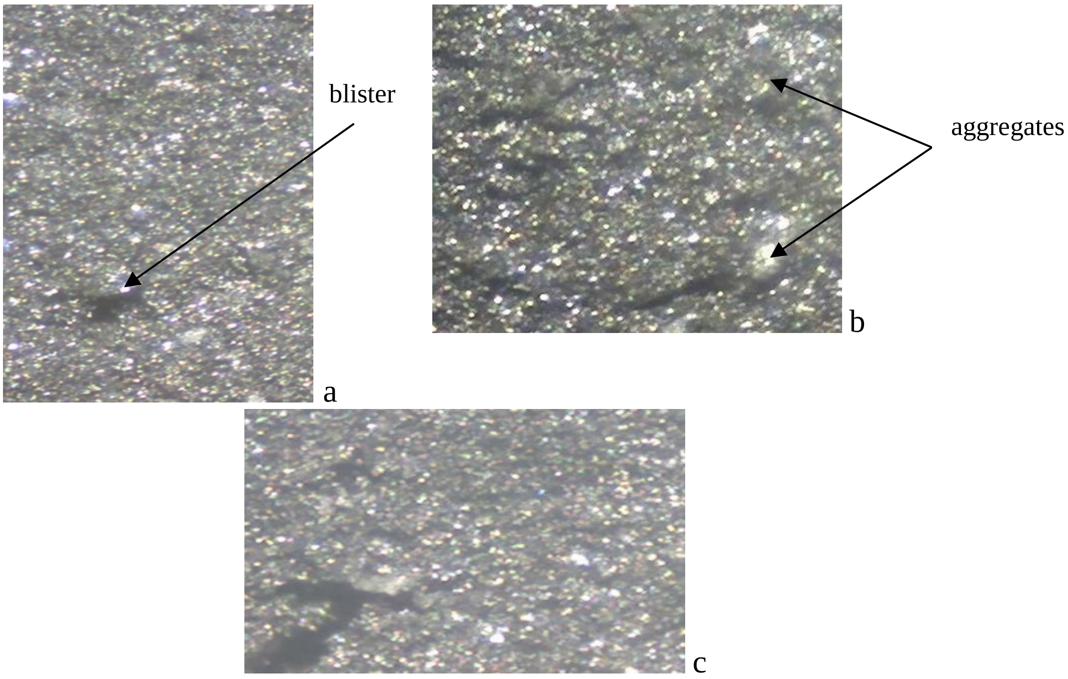

Fig. 2. Defects in the microstructure after irradiation, x200:
a – zone with blister; b – the beginning of crack formation, large aggregates;
c – fusion of pores with a crack

As the dose increases, surface imperfection generally increases. However, the changes are local and minor. The presented radiation shielding materials meet the requirements for radiation resistance. According to GOST 9.711-85 about stratification resistance, the material of the samples can be classified into resistance groups I or II.

5. Conclusion

The studies showed that at high doses of radiation (100 times higher than the standard 10^5 Gy), the linear radiation absorption coefficients in elastic self-adhesive radiation shielding coverings do not change. An analysis of the material structure using optical microscopy after irradiation with these doses has established that minor changes in the material structure occur at this scale level. According to the resistance to delamination after irradiation, the material of the samples can be classified into resistance groups I or II (GOST 9.711-85). Elastic self-adhesive radiation shielding coverings meet the requirements for radiation resistance.

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