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# Protective properties of a multicomponent material when irradiated by electrons with an energy of 5 MeV and gamma radiation with an energy from 0.570 to 1.252 MeV

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The synthesis of a multicomponent material intended for use as protection for astronauts and spacecraft equipment from cosmic radiation is presented. PTFE-4 was chosen as a binder for the multicomponent material. The following components were introduced into the matrix as fillers: bismuth oxide, tungsten carbide, titanium hydride fraction and boron carbide. The synthesis of the material was carried out through solid-phase compaction, followed by cooling to room temperature, pressing out with further sintering. Its physical and mechanical properties have been studied: the value of erosive wear, Vickers microhardness, elastic modulus. Experimental irradiation of a multicomponent material with gamma radiation was carried out. The values of the linear attenuation coefficient of gamma radiation and the mass attenuation coefficient of gamma radiation are calculated. A multicomponent material was irradiated with fast electrons in a vacuum. It has been established that at an absorbed electron dose of 5 MGy, a twofold decrease in the bending strength of the multicomponent material is observed.

Keywords: multicomponent material, electron impact, y-radiation, linear attenuation coefficient.

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# Introduction

With development of society and technology the space is becoming one of the main areas of interest of humanity, but main reason of containment for its full study is negative factors of outer space. The most dangerous of them is space radiation, because humanity at this moment can not protect completely the cosmonauts from the radiation effects. The radiation effects result in irreversible consequences for the human organism, for example, to the development of cancer, cardiovascular diseases, acute radiation sickness, cognitive and hematopoietic disorders [1-4]. Space radiation can be primary comprising galactic cosmic radiation, solar particle phenomena and particles captured by the Earth's magnetosphere, and secondary which is formed during interaction of the primary radiation with structures of space vehicles due to nuclear fragmentation [5]. The primary radiation comprises mainly protons, electrons, alpha particles, deuterons, <sup>3</sup>He and particles with atomic number greater 2, and secondary radiation comprises protons, neutrons, gamma rays, electrons, mesons, muons and nuclei from He to Ni [6-11].. The main factor affecting the human organism is exactly secondary rations [12], so, currently it is necessary to develop materials for cosmonauts protection against space radiation with focus on protection against protons, neutrons, gamma rays and electrons, since aluminum, which is currently used to protect space vehicles does not ensure sufficient decreasing of the radiation dose [13].

Currently there are many materials, including composite materials, for protection in space against protons and neutrons, rarely against electrons, and their possibility to screen these radiations is widely studied [14-18]. For protection against neutrons special interest is paid to polymer materials filled with neutron absorbers, which is due to high content of hydrogen in polymers, using which the quick neutrons are effectively decelerated under elastic scattering. In addition, polymer composites will be lightweight and have low cost. For protection against definite type of radiation appropriate filler is introduced in polymers [19,20]. As neutron absorbers compounds of boron, cadmium, samarium are widely used due to high transverse cross-section of thermal neutrons absorption [21–25]. We know paper [26] in which neutron protective characteristics of CdO, B<sub>4</sub>C, BN are compared using MCNP simulation code. It is shown that for protection against neutrons BN with density 3.487 g/cm<sup>3</sup> has better parameters relating equivalent dose decreasing. and reaches about  $2\mu$ Sv/h (decrease by 52%).

There are also many studies discussing the possibility of polymer composite materials use to protect against gamma radiation [27–33], but many of them can not be used space due to high density, insufficient degree of protection against neutrons, presence of heavy elements which upon interaction with the primary space radiation form secondary radiation, as well as due to low resistance to negative factors of outer space.

Paper [34] is of particular interest. In it the polymer composite polyester/baryte/tungsten boride for protection

against gamma radiation is studied with gamma spectrometer and sources <sup>22</sup>Na, <sup>133</sup>Ba, <sup>137</sup>Cs and <sup>60</sup>Co, as well as for protection against neutrons and charged particles using simulation by Monte Carlo method. Practically in all studies the best composition is BaWB-50 with 50% For example, results of full macroscopic crossfilling. section, the mean free path and the number of transmitted neutrons are 2.0515 cm<sup>-1</sup>, 4.8745 mm and 8601998 [L1] respectively. half-value layer of gamma radiation for this composition is 2.543 cm, which by 2.74 time is lower than in pure polymer. So, as per all protection parameters the addition of filler BaWB to polyester material has positive effect on the protection characteristics. Such study was performed by Oğul [35] who studied the effect of additions of nanopowders B and Sn on protection against gamma and neutron radiation. To test shielding ability of the prepared samples energy of gamma radiation (neutrons) in range of 59.5 to 1332.5 keV (0.1-10 MeV) was used. Besides the experimental measurements the theoretical (simulation) calculations were performed using programs WinXCOM (GEANT4 and FLUKA). The measurements showed that the shielding effect of the polymer increases with addition of B and Sn, at that maximum values are in composition with maximum content of Sn.

So, the composition materials intended for use in space industry shall be studied comprehensively. Characteristics of their protection against not only space radiation, but from secondary radiation shall be studied. Actual possibility of these materials use on space vehicles shall be evaluated considering volume occupied by them and weight.

Previously team of authors developed a multicomponent material which has high neutron protective characteristics and can be used for protection against neutron radiation in space [36,37]. This paper presents the results of the impact of electron and gamma radiation on this multicomponent material. Protection properties of multicomponent material were studied using gamma sources with energies  $E(^{207}\text{Bi}) = 0.570 \text{ MeV}$ ,  $E(^{137}\text{Cs}) = 0.662 \text{ MeV}$ ,  $E(^{60}\text{Co}) = 1.252 \text{ MeV}$ , and radiation resistance under electron radiation was studied.

# 1. Materials and methods

### 1.1. Synthesis of multicomponent material

Fluoroplast-4 press-powder was used as a binder for the multicomponent material. Fluoroplast use as binder is determined by exclusive resistance of fluoroplast materials against chemical effect, excellent mechanical, dielectric and antifriction properties, as well as low gas emission during vacuuming [38].

As fillers the following components were used: bismuth oxide  $(\alpha-Bi_2O_3)$ , tungsten carbide (WC), titanium hydride shot TiH<sub>1.7</sub> with diameter up to 2.5 mm, boron carbide F2500 (B<sub>4</sub>C). Bismuth oxide is a p-type semiconductor with high density 8900 kg/m<sup>3</sup>. Unlike the lead the bismuth oxide is not toxic and has high radiation protection characteristics

in relation to  $\gamma$ -radiation, practically not inferior to lead. The boron carbide is intended to absorb thermal neutrons, and titanium hydride — to decelerate quick neutrons. tungsten carbide is necessary to increase wear resistance of composite, which is especially actual under action of the oncoming flow of atomic oxygen in space and micrometeorite particles [39].

For manufacturing sample of multicomponent material we used substance at following mass ratios (mass.%): fluoroplastic — 38.5; bismuth oxide  $Bi_2O_3 - 42.2$ ; tungsten carbide WC — 2.9, titanium carbide shots  $TiH_{1.7} - 15.3$ ; boron carbide  $B_4C - 1.1$ . It was previously established that this particular composition has high neutron protective properties [36,37].

The components (except titanium hydride shots) were mixed using cryogenic milling for at least 15 min. Use of cryogenic milling ensures mechanical activation of surface of all components, which significantly improves physical and mechanical properties of final material [40]. After cryogenic mixing the titanium hydride shot was added to the mixture and milled manually to exclude shot deformation. Mixture was loaded into the mould and heated to 280°C with this temperature keeping for at least 60 min. To form the finished product a solid phase compaction under pressure at least 195 MPa was performed. After pressing the mold was gradually cooled under pressure to 100°C and pressure was removed with further cooling to room temperature. Following pressing, the sample of the multicomponent material was sintered at temperature 360-370°C for at least 3 h, with further slow cooling to room temperature directly in the heating equipment.

### 1.2. Objects and methods of study

Micrographs of the materials were made using TES-CAN MIRA 3 LMU high resolution scanning election microscope. Micrographs were made in SE mode— Everhart–Thornley secondary electron detector.

Grain size of powder materials was studied by the grain size analysis method using Analysette 22 NanoTec plus laser light scattering particle size analyzer.

Samples microhardness was determined by Vickers method  $\left(HV\right)$  using Nexus 4504-IMP hardness tester.

The density was studied by hydrostatic weighing.

Erosion wear of samples was tested in Air Jet Erosion Testing Machine model TR-471-400 (2015, Ducom Instruments, India). Test time — 1 h, abrasive material consumption 2.2 g/min, as abrasive material the corundum power (Al<sub>2</sub>O<sub>3</sub>) was used with average fraction  $50 \,\mu$ m. Rate of abrasive powder was 65 m/s, incident angle of abrasive particles is 90°.

Bend strength was measured in universal test machine REM-100-A-1-1 (manufacturer LLC "Metrotest", Republic of Bashkortostan, Nefteyugansk, with high measurement range of load 100 kN) by method of three-point bending. The sample was placed on two supports and loaded in the center with a bending punch.

Filler	Size range of particles, $\mu m$	Modal diameter of particles, $\mu m$	Specific surface area, cm <sup>2</sup> /cm <sup>3</sup>
Bismuth oxide $Bi_2O_3$	0.08-27.9	0.09	106489
Tungsten carbide (WC)	0.09-24.7	10.31	108268
Boron carbide B <sub>4</sub> C	0.09-120.7	57.64	2957

Table 1. Data on granulometry of fillers

The modulus of elasticity was calculated using equation (1) and PULSAR-1.2 ultrasound propagation time meter:

$$E = \frac{\gamma V^2}{0.95} \cdot 10^3,$$
 (1)

where E — modulus of elasticity, [Pa], V — ultrasound speed value, [m/s],  $\gamma$  — bulk mass, [g/cm<sup>3</sup>].

Contact angle of wetting was measured in Krüss DSA 30 device (KrüssGmbH, Germany).

Radiation protective characteristics in relation to  $\gamma$ radiation were determined using DKS-96 dosimeterradiometer with BDKS-96b detection unit. Initially background was measured, it was stored in device memory, then in lead container sources of  $\gamma$ -radiation were loaded, the exposure dose was measured without protective shield, after which the container was hermetically sealed with sample of the multicomponent material being studied 1 cm thick using special gas, creating the protective shield, and then the exposure dose was measured. As sources we used:  $E(^{207}\text{Bi}) = 0.570 \text{ MeV}, \ E(^{137}\text{Cs}) = 0.662 \text{ MeV},$  $E(^{60}\text{Co}) = 1.252 \text{ MeV}$ . Each source was made in geometry OSGI 4 and is in form of a disk with diameter of 25 mm, its center comprises active spot with diameter of 2.5 mm of radionuclide sealed with a polymer material with a total thickness of 3 mm. Sources were manufactured by JSC "Khlopin Radium Institute", Saint-Petersburg.

Irradiation of multicomponent material by quick electrons in vacuum ( $P = 1.4 \cdot 10^{-4}$  Pa) with energy 5 MeV (beam power 1.5 kGy/s) was performed at microwave electron accelerator "Raduga". Time of samples holding in chamber was 1.5-2 s. The electron fluence was  $3.5 \cdot 10^{15}$  electrons/(cm<sup>2</sup> · s). Single absorbed dose varied from 10 to 20 kGy depending on time of staying in chamber. So, maximum absorbed dose 6 MGy was accumulated for 2 months during about 350 cycles (irradiation cycles).

Radiation resistance of the multicomponent material was evaluated by decrease by two times of bend strength of samples subjected to radiation exposure of quick electrons flow.

# 2. Results and discussions

# 2.1. Studies of physical and mechanical properties of multicomponent material

To synthesize the multicomponent material, fluoroplastic grade F-4 in the form of a finely dispersed powder was

used as the binder. Size of particles of fluoroplastic powder is in range of 0.1 to  $666.7 \,\mu\text{m}$ , at that major portion of particles with size of 20 to  $150 \,\mu\text{m}$ . Modal diameter of particles is  $211.35 \,\mu\text{m}$ , and specific surface area of particles is  $1614 \,\text{cm}^2/\text{cm}^3$ .

See data on grain size of fillers, except titanium hydride shot, in Table 1.

To estimate the reached bonding between the fluoroplastic and fillers, as well as to determine uniform distribution of fillers in multicomponent material we studied microstructure of surface of the obtained sample (Fig. 1).

Analysis of the surface microstructure of the obtained multicomponent material showed that the fillers are distributed uniformly over the entire surface of the composite. Large amount of titanium hydride shot is well visible, is covers about 30% of surface, which ensures the assumption on high probability of interaction of neutrons passing through the polymer composite with hydrogen atoms contained in shot. This says about correctness of selection of percentage content of fillers. But on titanium hydride shot cracks are visible, they are filled by other fillers, which can say about low exit of hydrogen from the shot volume. No other defects were found on the surface of the polymer composite. The material surface is smooth, which indicates that the bond between the fillers and the binder was achieved. Also, for a more qualitative determination of the fillers distribution the energy-dispersive analysis (EDS) map was made (Fig. 2).

On material surface two regions with increased concentration of tungsten particles were detected (Fig. 2). Note that tungsten particles present only together with bismuth particles. Several agglomerates of bismuth and boron particles were also identified. Presence on surface of multicomponent material of titanium particles is explained by its integrity damage occurred during samples pressing. For detailed evaluation of structural integrity of irradiated sample and to detect internal defects the rate of distribution of ultrasound oscillations was studied, based on it the modulus of elasticity by formula (1) was calculated. Besides for comparison of the obtained values the similar studies were performed with samples made of pure fluoroplastic, the results are given in Table 2. Density of the multicomponent material for calculations was  $3.56 \text{ g/cm}^3$ .

From Table 2 we can see that introduction of fillers by 1.67 times increases the modulus of elasticity, i.e. the ability of obtained multicomponent material to restore the initial view after force application increased by 1.67 times. Besides the stable values of wave distribution rates in three



Figure 1. SEM images of surface of multicomponent material.



**Figure 2.** Multilayer map of energy dispersive analysis of multicomponent material surface.

**Table 2.** Data on modulus of elasticity of fluoroplastic and multicomponent material

Sample of multicomponent material			
$V_{med.}, m/s$	$5108\pm1$		
R, MPa	$54.4\pm0.5$		
E, GPa	$97.7\pm0.5$		
Density, g/cm <sup>3</sup>	$3.56\pm0.03$		
Sample of pure fluoroplastic			
$V_{med.}, m/s$	$5083\pm1$		
R, MPa	$54\pm0.5$		
E, GPa	$58.4\pm0.5$		
Density, g/cm <sup>3</sup>	$2.15\pm0.03$		

different points of the obtained sample confirm the absence of internal defects, and homogeneity of structure.

To study the deformation and physical-mechanical properties of the developed multicomponent material the bending strength of samples in the form of rectangles of size  $50 \times 10 \times 7 \text{ mm}$  was tested by method of three-point bending (Fig. 3).

5 samples of multicomponent material were tested, and average values for maximum load 347.2 N, maximum deformation 1.16 mm were determined; ultimate strength is 14.6 MPa.

Then properties of multicomponent material surface were studied using contact angle of wetting (Fig. 4).

Fluoroplastic is known by its low wettability, but if filler is introduced in about over 60 mass.% the wettability parameters can significantly increase. We can conclude that surface of the multicomponent material for water is unwetted (Fig. 4).

Values of basic physical and mechanical characteristics of multicomponent material are given in Table 3.

Thus it is shown that the polymer composite has good physical and mechanical properties. Then protective properties of this composite against gamma and electron irradiation were tested.



**Figure 3.** Applied load vs. deformation for sample of multicomponent material.



**Figure 4.** Images of laying water droplet on surface of multicomponent material.

**Table 3.** Values of physical and mechanical characteristics ofmulticomponent material

Parameter	Value		
Density, g/cm <sup>3</sup>	$3.56\pm0.03$		
Vickers microhardness HV/1 (Load 200g), HV	$13.08\pm0.5$		
Erosion wear, $10^{-3} \text{ mm}^3/\text{g}$	$0.052\pm0.002$		
Ultimate bending strength, MPa	$14.6\pm0.89$		
Maximum deformation, mm	$1.16\pm0.05$		
Modulus of elasticity, GPa	$97.7\pm0.5$		
contact angle of wetting with water, $^{\circ}$	$105.40 \pm 2.42$		

# 2.2. Study of protective properties of multicomponent material in relation to gamma radiation

Based on the experimental data the linear gamma radiation attenuation coefficient, the half-value layer of gamma radiation, the mass attenuation coefficient for gamma radiation, and the percentage of gamma radiation attenuation behind the sample were calculated. Also for comparison of the obtained characteristics the same experiment and calculations were made for the sample made of pure fluoroplastic and presented in Table 4.

So, we see the dependence of decrease in linear attenuation coefficient on energy increasing, which correspondingly increases the half-value layer. We also see that all calculated characteristics for the multicomponent material, except for the mass attenuation coefficient, exceed the same of the pure fluoroplastic practically by two times. If for gamma radiation with energy 0.570 MeV decreasing by 50% a screen made of pure fluoroplastic 3.65 cm thick is required, then such parameter will be achieved by screen made of the multicomponent material only 2.17 cm thick, which is lower by 1.48 cm, and at energy 1.252 MeV - lower by 2.03 cm. Besides, percentage of gamma radiation attenuation by sample made of the multicomponent material also exceeds the same for the pure fluoroplastic in the full range of studied energies by 1.5 and more times. This result was achieved by the selection of bismuth oxide Bi<sub>2</sub>O<sub>3</sub> as the basic filler with percentage content in the material 42.2 mass.%.

We know paper where nanofilled polymer composite materials based on thermoplastics are studied for protection against gamma radiation [41]. Linear attenuation coefficient of given material at energy 0.661 MeV and density  $4.5 \text{ g/cm}^3$  is  $0.25 \text{ cm}^{-1}$ , which by  $0.05 \text{ cm}^{-1}$  is lower than in developed by us multicomponent material at density  $3.56 \text{ g/cm}^3$ . Also paper [41] presented linear attenuation coefficients of gamma radiation at energy 0.661 MeV of polymer materials: composite material "Nejtronstop SO–Pb" ("Kovo", Czech

	Multicomponent material		Pure fluoroplastic			
Parameter	$E(^{207}{ m Bi}){=}0.570$ MeV	$E(^{137}Cs)=0.662$ MeV	<i>E</i> ( <sup>60</sup> Co)=1.252 MeV	<i>E</i> ( <sup>207</sup> Bi)=0.570 MeV	$E(^{137}Cs)=0.662$ MeV	<i>E</i> ( <sup>60</sup> Co)=1.252 MeV
Linear attenuation coefficient, $cm^{-1}$	$0.32\pm0.01$	$0.30\pm0.01$	$0.21\pm0.01$	$0.18\pm0.01$	$0.17\pm0.01$	$0.13\pm0.01$
Half-value layer, cm	$2.17\pm0.07$	$2.31\pm0.09$	$3.30\pm0.17$	$3.85\pm0.2$	$4.08\pm0.23$	$5.33\pm0.45$
Mass attenuation coefficient, $cm^2/g$	$0.089\pm0.002$	$0.084\pm0.003$	$0.058\pm0.002$	$0.083\pm0.004$	$0.079\pm0.005$	$0.060\pm0.005$
Percentage of gamma radiation attenuation by sample, %	$27.38\pm0.73$	$25.91\pm0.74$	$18.94\pm0.80$	$16.47\pm0.83$	$15.63\pm0.84$	$12.19\pm0.89$

Table 4. Comparative characteristic of protective properties of fluoroplastic and multicomponent material against gamma radiation

Table 5. Comparison of mass attenuation coefficient of gamma radiation of known materials

Material	Mass attenuation coefficient of gamma radiation, cm <sup>2</sup> /g at energy 0.662 MeV		
Obtained multicomponent material	0.084		
High-density polyethylene [42]	0.070		
40% High-density polyethylene/bulk zinc oxide [42]	0.065		
40% high-density polyethylene/bulk nanoparticles of zinc oxide [42]	0.077		
Composite polyacrylamide/zinc oxide 20% [43]	0.080		
Polyester concretes filled with bismuth oxychloride (10%) [44]	0.079		
Polymethyl methacrylate with filler content $Bi_2O$ , equal to 10% [45]	0.081		
30 mass.% nanoGd <sub>2</sub> O <sub>3</sub> /10 mass.% B <sub>4</sub> C/60 mass.% HDPE [46]	0.062		

Republic) — mixture of polyethylene with lead powder, density  $3.5 \text{ g/cm}^3$ ; "Light-Lead" (USA) — mixture of lead in inert polymer, density  $3.03 \text{ g/cm}^3$ ; "Lead Blanket" (USA) —  $\gamma$ -protective elastomer with high content of lead, density  $3.45 \text{ g/cm}^3$ . Attenuation coefficients were 0.22, 0.19, 0.20 cm<sup>-1</sup> respectively, which practically by one third is lower than in obtained multicomponent material at practically same density [41].

Mainly, the comparison of the efficiency of gamma protective properties is used through the mass attenuation coefficient, which is presented in Table 5.

So, based on data in Table 5 we can conclude that the obtained multicomponent material surpasses modern analogues in terms of the mass attenuation coefficient of gamma radiation. It is evident that composites based on the polyethylene are worser in terms of this characteristic.

### 2.3. Study of effect of electron radiation on multicomponent material

Results of strength characteristics decreasing of multilayer composite depending on the absorbed dose are given in Fig. 5.



**Figure 5.** Bend strength of multicomponent material vs. absorbed dose of electrons.

Radiation resistance is the material ability do not destruct under exposure to ionizing radiation, i.e. up to such absorbed dose the material integrity will not be critically damaged. So, the expected change at the absorbed dose 5 MGy will be bend strength decreasing by two times of the multicomponent material. Accordingly, the radiation resistance of the multicomponent material is 5 MGy.

For example, it is known that polyethylene terephthalate loses up to 30% of ultimate strength at dose of irradiation by electrons only 1 MGy [47], at same exposure the obtained multicomponent material losses 18.5% of strength. At the same time in polyethylene even at dose of irradiation by electrons 0.3 MGy the fracture elongation decreases by 50%, an in fluoroplastic the fracture elongation decreases by 40% at dose of irradiation by electrons 0.005 MGy [48].

# Conclusion

It is confirmed that mixing of components of the multicomponent material using cryogenic milling, its synthesis by the method of hot solid-phase compaction under pressure of 195 MPa followed by sintering at temperature  $360-370^{\circ}$ C provide the necessary physical and mechanical, and operational characteristics: density —  $3.56 \text{ g/cm}^3$ , erosion wear —  $0.052 \cdot 10^{-3} \text{ mm}^3/\text{g}$ , Vickers hardness under load 200 g = 13.08 HV, bend strength — 14.6 MPa, modulus of elasticity — 97.7 GPa, maximum deformation — 1.16 mm. By obtained values we can say that components for the synthesis are selected correctly; and optimal parameters are selected for solid-phase compaction.

According to experimental data obtained under  $E(^{207}\text{Bi}) = 0.570 \,\text{MeV},$ action of gamma sources  $E(^{137}Cs) = 0.662 \text{ MeV},$  $E(^{60}Co) = 1.252 \text{ MeV}$ on multicomponent material the linear  $(0.32, 0.30, 0.21 \text{ cm}^{-1})$ respectively) and mass  $(0.089, 0.084, 0.058 \text{ cm}^2/\text{g})$ respectively) attenuation coefficients of gamma radiation are calculated, as well as half-value layer (2.17, 2.31, 3.30 cm respectively). Comparison of values of mass attenuation coefficient of gamma radiation of multicomponent material with the modern analogues demonstrates its superiority.

results of radiation resistance under action of quick electrons in vacuum with energy 5 MeV showed that at absorbed dose 5 MGy the bend strength of the obtained material decreases by 50%.

The developed material can be used to provide protection for cosmonauts and electronic equipment from the effects of negative factors in outer space, primarily from space radiation, including secondary radiation arising from the interaction of high-energy space ray particles (primarily protons and heavy charged particles) with the materials of the structural elements of space vehicles. Also, this material can be used to protect space bases built on Moon or Mars surface; besides it can be used to protect the crew of vessels with mobile reactors, for example, on nuclear submarines and nuclear icebreakers.

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