STUDY OF STRENGTH CHARACTERISTICS AND RADIATION-PROTECTIVE PROPERTIES OF POLYSTYRENE TUNGSTEN COMPOSITE MATERIALS

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Samples of composite materials are made of polystyrene, which is reinforced with dispersed aluminum. The radiation protective additive was powdered tungsten or powdered steel. The tensile strength, for these samples of composites, was investigated. The tensile strength value was obtained at various temperatures. It has been found that the ultimate strength of composites increases with an increase in the metal component. Composites with different component compositions were studied. The relative attenuation of the absorbed dose of gamma radiation by these composite materials has been calculated. Composites were with different mass composition. The code of Geant4 v 4.9.6p03 applied for calculations. It is got, that experimental results coincide with calculation results.

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INTRODUCTION

The problem of radiation protection is one of the important problems that arise in the operation of nuclear power facilities. To create stationary radiationprotective structures, concrete-based materials with various radiation-protective additives are usually used.

To protect against X-rays, gamma radiation, beams of alpha and beta particles, heavy metal additives Pb, W are used. To protect against neutron fluxes, Br, B, Bi are used. These additives make it possible to obtain high radiation-protective characteristics of protective materials.

However, work on the improvement of radiationprotective materials continues. This creates materials that have higher protective characteristics. When creating new protective materials, various technologies and methods are used.

One of methods, which is used at creation of radiation-protective materials, there is combination of different materials technique. Materials which were created on such principle are named composite

Usually, composite materials consist of a base and various additives. A detailed review of publications that present materials on the creation of new radiation-protective materials is given in [1]. At present, a significant number of composite materials have been developed, which are used in various directions.

X-ray units are used in industry and medicine. Therefore, there is a need to develop radiation shielding materials that will provide protection from X-rays. These composite materials consist of rubber with the addition of lead [2].

Composite materials are known in which tungsten, steel, hematite, and lead glass were used as radiationprotective additives [3–7]. Composites can also be used in the construction of stationary protective structures [8, 9]. Ceramic composite materials are also being developed to protect spent nuclear fuel. However, the need for new radiation-protective materials is only increasing. Interest in composite materials is also caused by the fact that their use allows solving many problems.

Previously, work was carried out on the development and creation of polystyrene metal composite materials (PS-W-Al) and (PS-Fe-Al) [10–12]. Some of these materials have already passed laboratory tests and have been used to create radiation shields. It was found that these radiation-protective composites can perform another function.

Polystyrene metal composite materials only absorb electromagnetic radiation, but also scatter it to a large extent. A change in the reflective characteristics of electromagnetic radiation allows not only to mask the real shape of the object, but also to change its signatures on the screens of radar stations (stealth technology). That is, they can protect objects from being detected by radar stations.

PURPOSE OF WORK

1. Improvement in the technology of manufacturing polymer metal composites (PS-W-Al).

2. Determination of the dependence of the strength characteristics of a composite material on the type of components, their composition, structure and operating temperature.

3. Study of the radiation-protective characteristics of composite materials.

CONDUCTING EXPERIMENTS AND DISCUSSION OF RESULTS

The work was carried out in several stages. At the first stage, work was carried out to prepare the technology for the production of composite materials [11]. Further refinement of the technological process was carried out [12]. At the second stage, changes were made to various characteristics of the composite. Composition material which we studied consists of three components: polystyrene, aluminum and tungsten.

Polystyrene is the basis and binding component. materials. When creating composite **PSM-115** polystyrene (GOST 20282-86) was used as the basis. The choice was determined by several factors. Polystyrene is well processed. Registered and approved technical regulations and technical methods for the production of products from polystyrene. Polystyrene has a softening point of 86...92 °C and a melting point of 196...200 °C. Polystyrene has low hygroscopicity and high water resistance. Polystyrene is resistant to acids and alkalis. Therefore, polystyrene is a convenient material for creating composites that are used to protect against ionizing radiation.

Polystyrene has low radiation-protective characteristics. With a layer thickness of 10...20 mm, only gamma quanta with energies up to 40 keV are effectively absorbed. With an increase in the energy of gamma rays (up to 1.5 MeV), the attenuation of the absorbed dose is less than 10%. When the energy of gamma quanta is higher than 1.5 MeV, no more than 2...3% of gamma quanta are absorbed in polystyrene [3, 5, 8, 10–12].

To fulfill the tasks of protection, it is necessary that the composites have certain characteristics of hardness and strength. The composite must be structured to increase its strength. The frame of the composite material must be created. Powdered aluminum was used to create a frame of a composite material

In our case, in the manufacture of composite materials, several types of aluminum powder were used: ASD-6, 2014, 6111 [13, 14]. These aluminum powders comply with the standards set out in regulatory documents: ISO 209-1, TU 1791-007-49421776-2011. Aluminum particles had the following dimensions: $10...20, 60...90 \mu m$.

Aluminum is a component with the help of which the matrix elements of a composite material are created. Aluminum is wetted with polystyrene. Therefore, when creating a protective composite, aluminum powder was mixed with molten polystyrene. The composite material was obtained when the mixture solidified. The polystyrene was reinforced with aluminum in this composite material. The matrix structure is obtained. The component of tungsten must be added to protect against ionizing radiation.

Tests were carried out with various types of radiation-protective additives. Powder of tungsten was applied as a radiation protective additive (composites of the PS-W-Al type) [15]. Steel powder was applied as a radiation-protective component (PS-Fe-Al composites). Steel powder was obtained from waste in the manufacture of various metal products [16, 17].

Composite materials were studied in which tungsten was used. It was used in powder form. Powdered tungsten (PV-2) was manufactured in accordance with the requirements of TU 14-22-143-2000. Powdered tungsten was used for radiation protection. Tungsten particles had sizes: 50...60, 230...280 µm.

Grains of tungsten are placed in a solution of polystyrene with aluminum. Grains of tungsten are fixed in polystyrene after cooling. We get a new composite material. It has high radiation-protective properties. The composite material consists of polystyrene and metal additives. The composite material is designed to protect against ionizing radiation. A study of the characteristics was carried out [3, 5, 8, 10]. Hardness characteristics are found for composite materials [10].

Improvement of equipment. In the manufacture of composite materials, standard industrial equipment was used. Kuasy 100/25 and Windsor SP 80 automatic machines were used to develop manufacturing technologies and obtain test batches of the composite. The Kuasy 100/25-1 device had the following operating parameters: injection pressure - 180 MPa, injection volume -50 cm^3 . The Windsor SP 80 device had the following operating parameters: injection pressure -300 MPa, injection volume -187 cm^3 . This equipment was chosen due to the fact that it can produce products from reinforced polystyrene. The extrusion method is used to manufacture composite materials. Certified technological processes for the production of polystyrene products are known. The production of composite materials is a challenging task. Therefore, work was carried out to improve and refine the equipment. The results of the improvements are shown in Fig. 1 [12].



Fig. 1. Of principle chart of castable machine for casting under constraint: 1 – material cylinder;
2 – heating elements; 3 – screw (auger); 4 – cooling channels; 5 – material hopper; 6 – hydraulic engine;
7 – gearbox; 8 – hydraulic cylinder injection unit;
9 – manometer; 10, 17 – fixed plates; 11 – guide columns; 12 – injection mold; 13 – movable plate;
14 – wheel-lever mechanism; 15 – hydro-cylinder of the clamping unit; 16 – nuts; 18 – emphasis; 19 – nozzle. Elements that have been added to the equipment:
20 – bunker heaters; 21 – blades for mixing the mixture;

22 – mixer drive; 23 – IR control camera; 24 – drive and rotary device for the entire bunker

Several methods were used to control the uniformity of mixing. IR radiometric methods were applied to determine the degree of mixture heating. For this purpose, thermal imaging devices Lend Ti-814, Fluke-10, Fluke-25 were used [18]. A thermogram was obtained as the temperature on the surface of the mixture changed. The thermogram was obtained using IR radiometric methods.

Based on the thermogram, we can determine the degree of heating of the surface of the mixture, the degree of mixing of the components, the presence of lumps of the components. Thermal inhomogeneities on the surface appear very clearly (Fig. 2).

The thermogram of the surface of the mixture that is being mixed is shown in Fig. 2,a. The mixture is at the initial stage of preparation. Various temperature inhomogeneities are observed on the thermogram.

It can be the sticking together pieces of polystyrene, lumps of tungsten, and lumps of aluminum. They have different heat capacity and thermal conductivity and therefore heat up at different rates. Their temperature is different, and they reflect differently on thermograms. Graphs of temperature changes along the selected lines are also shown. On Fig. 2,b shows the change in temperature along section 1. The temperature peak (41.6 °C) corresponds to the lump of aluminum, which warmed up the fastest. On the thermogram, it corresponds to the image of a white dot. We observe similar temperature peaks on the graph of temperature changes along line 2 (see Fig. 2,c). In this case, the heating of the entire material has not yet occurred.



Fig. 2. The thermo gram surface temperature of the mixture (a). The graphs of temperature change along the sections 1 (b) and 2 (c)

The uniform distribution of the tungsten and aluminum components over the volume of the composite material depends on the quality of mixing of these components. Also, the uniformity of distribution also depends on the size of the grains of the composites. This is especially noticeable for tungsten. A slight increase in the size of tungsten particles (from 30 to $280 \mu m$) greatly changes the mass of these particles.

To perform research, several modifications of composite materials were made. The samples were made in the form of plates $(70 \times 40 \times 15 \text{ mm})$. The appearance of samples C120201 and C100401 is shown in Fig. 3.

The plates were used to measure the hardness of composite materials. Also, plates of composite materials were used when measuring the absorbed dose of ionizing radiation. Sources of ionizing study were used in experiments. The following sources were used: ²⁴¹Am source (gamma rays with energy of 60 keV) and ⁹⁰Sr

[12]. In all composite materials, aluminum particles had a size of $60...90 \ \mu\text{m}$, and the particles of tungsten had a size of $50...60 \ \mu\text{m}$.



Fig. 3. Original appearance of samples with different component composition (a - C120201, b - C100401)

Strength is an important characteristic of composites. A tensile testing machine was used to determine the tensile strength. The tests were carried out in accordance with GOST 11262-80 "Plastics. Tensile test method". The composite material was made in the form of rods by the diameter 10 mm and length of 120 mm. The stretch was uniaxial. Stretch curves obtained.

When testing pure polystyrene, tension was found with the formation of a neck. The relative elongation was 95...96%. The limit of fluidity is equal 24...25 MPa. The index of limit of fluidity turned out to be high. The tensile strength of pure polystyrene was 22...23 MPa. The elastic modulus of pure polystyrene was 2.1 GPa. These data were obtained at temperatures of 290 K. We have other results at other temperatures.

Composite materials of the C12YYZZ series were manufactured. The amount of components is given in units of volume. This is due to the peculiarities of the manufacture of composite materials. The composites contained 12 parts by volume of polystyrene (80% by volume) and three parts by volume of metal powder (20% by volume).

Composite materials can be used both indoors and on open space. Therefore, the tests were carried out at different temperatures.

At first, the work was performed at temperatures of 250 K (-23 °C). The fracture was fragile for all types of composite materials. All composites fail without necking. For composites of this type, the relative elongation has decreased. It was 0.5...1%. Tensile strength is given in Table 1 (at a temperature 250 K, first column). The fragility of polystyrene has increased.

A fragile break appears at points of inhomogeneity in the thickness of the composite. The tensile strength of composites is higher than that of pure polystyrene. The break of long polystyrene molecules and the formation of a reinforced structure occur when grains of metal are added to the polystyrene.

Also, studies were carried out at a temperature of 290 K. The rupture occurs with the formation of a neck. The relative elongation was 50...56%. Limit of fluidity 20 MPa. The modulus of elasticity was 2 GPa. The tensile strength for these composites is given in Table 1 (second column). The study of surface of break was performed. It has been found that a significant number of microscopic necks are formed on the surface of break. The sample is elongated, and the polystyrene is elongated at separate points. That is, in addition to

stretching the sample as a whole, there is also the effect of stretching the composite material at individual points. This effect is consistent with the theory of gaps that occur in graft copolymers. The tensile strength of composites of this type is higher than tensile strength that of pure polystyrene. This is due to the reinforcement of polystyrene with aluminum and tungsten powder.

The second limiting case is the case of elevated temperatures. The experiment was carried out at a temperature of 320 K. At these temperatures, polystyrene is still in a solid state. The structure of the fracture and the appearance of the fracture surface have changed. The neck was formed at break, for all types of composite materials.

The relative elongation value has increased. The relative elongation was 65...70%. The fluidity of the sample appeared. The value of the limit of fluidity was 23 MPa. The number of microscopic necks increased on the fracture surface. The number of microscopic necks is greater than at a temperature of 290 K. Microscopic necks have a high degree of elongation

The tensile strength of composites has decreased (at a temperature of 320 K). The tensile strength is given in Table 1 (at a temperature of 290 K, third column). The tensile strength, at a temperature of 320 K, is much less than at temperatures of 250 K. The tensile strength at a temperature of 320 K differs slightly from the tensile strength at a temperature of 290 K.

Table 1

Tensile strengths composition materials C12YYZZ, MPa

ivii u						
PS	38.0	23.0	22.0			
C120300	39.0	27.5	26.5			
C120201	39.2	30.0	29.5			
C120102	40.0	31.5	31.2			
C120003	40.5	33.5	33.5			

Graphs of changes tensile strength are presented on Fig. 4.



Fig. 4. Graphs of changes in the tensile strength of composite materials C12YYZZ

From Fig. 4 evidently, that the tensile strength is different for composite materials with different component composition. The volume content of the metal component in all types of composites identically.

The ratio changes between the volumes of aluminum and tungsten. Composition material of C120201

included two volume parts of tungsten powder and one volume part of aluminum powder.

Composition material C120102 contained one volume part of tungsten powder and two volume parts of aluminum powder volume. From the graphs, we see that the tensile strength of composite materials increased with an increase in the aluminum component.

The tensile strength changes insignificantly with increasing temperature from 290 to 320 K. The tensile strength did not change for composites with the maximum content of the aluminum component.

Composition materials were investigated, in which the amount of polystyrene was 10 parts by volume (65% by volume). The metal component was 5 parts by volume (35% by volume). These composite materials were designated C10YYZZ. This included composite materials C100104, C100203, C100302, C100401.

The researches were carried out at three fixed temperatures (250, 290, 320 K).

The samples were tested at a temperature of 250 K. Destruction occurred without the formation of a neck for all samples. The fracture is brittle for all composites. No elongation of the polystyrene neck was found. No rupture necks were formed either. The relative elongation was no more than 2%.

The tensile strength increased due to polystyrene glazing. The sensitiveness of samples was also increased to the incisions. That is, a brittle fracture appeared in areas where there were delaminations of the material or a decrease in the thickness of the samples. Tensile strength values for C10YYZZ composites at a temperature of 250 K are given in Table 2 (first column).

Tensile strength measurements were also carried out at a temperature of 290 K. The destruction of composite materials occurred with the formation of a neck. Analysis of fracture photographs was performed. Microscopic stretches of polystyrene appear on the fracture surface.

The relative elongation has a value of not more than 20%. The limit of fluidity has a value of not more than 21 MPa. The tensile strength increased to values of 29...33.5 MPa. For composite materials of this class, we observe a significant spread in the magnitude of the tensile strength.

The tensile strength strongly depends on the amount of aluminum powder. Aluminum powder sticks together to polystyrene better than tungsten powder sticks together to polystyrene.

Tensile strength values for these composites, at a temperature of 290 K are given in Table 2 (second column).

Table 2 Tensile strengths composition materials C10YYZZ, MPa

ivii u							
Tape∖T, K	250	290	320				
PS	37.0	23.0	22.0				
C100401	40.8	28.5	27.5				
C100302	41.0	31.0	30.5				
C100203	41.5	33.5	33.2				
C100104	42.5	34.5	34.4				

For a more complete understanding of the structure of composite materials, studies were performed at elevated temperatures. The experiment was carried out at a temperature of 320 K. The structure of the surface of break of the samples at a temperature of 320 K differs from the structure surface of break at a temperature of 290 K.

The relative elongation has increased. Its value is 55... 60%. An increase in the limit of fluidity was found. The limit of fluidity is equals 22 MPa. When examining the fracture surface, we observe an increase in the number of microscopic necks. However, their amount is less than, at the composites of C12YYZZ. This is due to the fact that the C10YYZZ composite contains a larger amount of the metal component and a smaller amount of polystyrene. That is, most of the polystyrene component is bound in clusters with aluminum and tungsten. And necks are formed only where there is free polystyrene. Values of tensile strength for these composites, at a temperature of 320 K, are given in Table 2 (third column).

The graphs of changes of tensile strength are presented on Fig. 5.



Fig. 5. Graphs of changes in the tensile strength of composite materials C10YYZZ

The results were obtained when the aluminum and tungsten particles had a fixed grain size. The particles of aluminum had a size of $60...90 \mu m$. The particles of tungsten had a size of $50...60 \mu m$. Also, work was carried out to study the tensile strength at other particle sizes of the components.

The powder of aluminum had a grain size of 10...20 or $60...90 \mu m$. The powder of tungsten had a grain size of 50...60 or $230...280 \mu m$.

Composite materials of class of C120102 and C100104 were produced. Composite material C120102 contained 12 volumes of polystyrene, 1 volume of tungsten, and 2 volumes of aluminum. Composite material C100104 contained 10 volumes of polystyrene, 1 volume of tungsten, and 4 volumes of aluminum. Composites with different sizes of grains were made.

The measurements were executed out at a temperature of 290 K. Tensile strengths are given in Table 3.

Table 3

Tensile strengths composition materials C120102 and C100104, MPa

Mat/Tap	1	2	3	4
C120102	32.0	31.5	29.5	28.5
C100104	36.0	34.5	33.0	32.5

The results of measurings are presented on graphs (Fig. 6).

The types of composition materials are marked along the horizontal axis. These materials differ in the size of tungsten grains and aluminum grains. Composition material 1 had a size of grain of W (50...60 μ m), Al (10...20 μ m); 2 – W (50...60 μ m), Al (60...90 μ m); 3–W (230...280 μ m), Al (10...20 μ m); 4–W (230...280 μ m), Al (60...90 μ m).

It can be seen from the graphs that the size of the grains has a significant effect on the strength of the composite material. Composite C100102 contains two volumes of aluminum powder more than composite material C100104. Connection of aluminium with polystyrene is significantly higher than that of tungsten.



Fig. 6. Graphs of changes the tensile strength of composition materials. Dependence on the size of grains of metal

Therefore, all tensile strength for composite material C100104 are greater than for composite material C120102. At point 1, the values of the tensile strength of composites are given, in which the grains of tungsten and aluminum have the smallest dimensions (Al (10...20 μ m), W (50...60 μ m)). The tensile strength had maximum values at point 1.

For both types of composites, the minimum tensile strength values were at point 4. Grains of aluminum and tungsten (Al ($60...90 \mu m$), W ($230...280 \mu m$)) had maximum dimensions at point 4.

From the graphs Fig. 6 confirmed the following assumptions. Basic influence on the increase in the strength of composite materials is provided by such characteristics.

a) Strength depends on the amount of metal component. How more metal components, the higher the strength of the composition material. This is due to an increase in the degree of reinforcement of the composition material.

b) Strength depends on the type of metal component. Polystyrene moistens aluminum powder well and the strength of the composite material is higher. Polystyrene poorly moistens tungsten and the strength of the composite material is lower.

c) The strength depends on the grain size of the metal component. Strength increases when the size of the metal grains decreases.

d) The strength depends on the homogeneity of the distribution of the metal component in the volume of the composite material. Strength increases with increasing homogeneity of the composite material. What higher homogeneity of distributing of компонент on volume composition material, the higher tensile strength

Calculation of radiation-protective characteristics. The most important description of compos are their radiation-protective properties. At the practical use of composition materials it is necessary to know the exact meaning of protective properties. Because measuring of protective properties is a labour intensive task, then it is necessary to apply theoretical calculations.

Software packages are used to calculate the absorption of ionizing radiation. The Geant4 v 4.9.6p03 package was used for our calculations [19, 20]. Geant4 v 4.9.6p03 software package has been improved. [21]. In the study, the relative attenuation of the dose of ionizing radiation (η) was found. The relative attenuation of the dose of ionizing radiation the expression:

$$\eta = 1 - \frac{D}{D_{air}}.$$
 (1)

In expression (1) D_{air} – is the calculated dose, in the absence of protection. D – is the calculated dose when protected. η – is the degree of relative attenuation of the absorbed dose of gamma radiation by a layer of protection made of a composite material.

The absorbed dose distribution was got on the depth of sample. The biological phantom approximation was used in the calculations. The calculation results are in high agreement with the experimental results [12].

Calculations were carried out for separate types of composite materials. The research results are presented in the works [3, 5, 10–12]. This paper presents numerical results for composite materials of the C10YYZZ type. The values of attenuation of the absorbed dose of ionizing radiation were obtained. Graphs of the absorbed dose change are presented in Fig. 7.



Fig. 7. Graphs of changes in the attenuation of the absorbed dose of gamma radiation

Continuous samples were considered in the calculations. The thickness of the samples was 10 mm (see Fig. 7). The choice of this thickness is due to several factors. Protective barriers are produced faster from 10 mm thick plates. The rate of calculation of the absorbed dose in a composite material increases for such thicknesses. Samples of this thickness are used in experimental setups. On Fig. 7, pure polystyrene (continuous curve of black) and composite C051000 (continuous curve of black with markers) are given as

limit cases. The choice of limit values is determined by the following factors. There are no radiation-protective additives in pure polystyrene. Therefore, polystyrene has minimal attenuation characteristics of ionizing radiation. Composite C051000 contains 10 volumes of tungsten. Therefore, its radiation protection characteristics have maximum values.

All composite materials absorb gamma radiation up to energies of 100 keV. Consequently, all composite materials of this type (thickness – 10 mm) are an effective radiation shielding material for a large number of instruments and equipment.

This equipment includes medical x-ray machines that are used in dentistry (68 keV). It is also possible to use for protection against radiation that comes from X-ray machines that are used for diagnostics in metallurgy. As the energy of gamma radiation increases, the effectiveness of protection deteriorates.

Thus, for the composite material C100104, a half attenuation of the absorbed dose occurs for gamma rays with an energy of 200 keV. At energies of gamma quanta equal to 500 keV. The attenuation of the absorbed dose has a value of no more than 20%, at energies of gamma quanta equal to 500 keV. At an energy of gamma rays equal to 1 MeV, the attenuation of the absorbed dose is no more than 10%. The maximum characteristics of radiation protection were material C100402.

Composite material with the required characteristics can be determined. For this, mathematical modeling methods, methods of measurement of the absorbed dose were used.

CONCLUSIONS

1. The experimental samples of composition materials (C12YYZZ and C10YYZZ) which are intended for protecting from an ionizing radiation are made.

2. Mechanical tests of samples of composite materials were carried out. The values of the ultimate strength of specimens at temperatures of 250, 29, and 320 K.

3. It was found that the aluminum component has the main effect on the strength of the composite

4. Tests of the strength of composites, which included powders with grains of various sizes, were carried out. An increase in the tensile strength was found with a decrease in the size of the grains of the metal component.

5. Numerical calculations were made of how the absorbed dose of ionizing radiation changed for composite materials of the C12YYZZ and C10YYZZ types.

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ВИВЧЕННЯ ХАРАКТЕРИСТИК МІЦНОСТІ І РАДІАЦІЙНО-ЗАХИСНИХ ВЛАСТИВОСТЕЙ ПОЛІСТИРОЛ-ВОЛЬФРАМОВИХ КОМПОЗИЦІЙНИХ МАТЕРІАЛІВ

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Зразки композиційних матеріалів виготовлені з полістиролу, армованого дисперсним алюмінієм. Радіаційно-захисною добавкою був порошковий вольфрам або порошкова сталь. Межу міцності для цих зразків композитів досліджували. Значення межі міцності отримано за різних температур. Знайдено, що межа міцності композитів збільшується із збільшенням металевої компоненти. Вивчалися композити з різним компонентним складом. Проведено розрахунки щодо ослаблення поглиненої дози гаммавипромінювання цими композиційними матеріалами. Композити були із різним масовим складом. Код Geant4 v 4.9.6p03 застосовували для розрахунків. Отримано, що експериментальні результати збігаються із розрахунковими.