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PREDICTION OF RADIATION SHIELDING PROPERTIES OF SELF ADHESIVE ELASTIC COATING

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Abstract. Analytical definition of radiation shielding properties of polymeric materials is proposed. This method shows good convergence of theoretical calculations with experiment. Obtained results show, that to receive material with high protective properties it is necessary to add barite or tungsten filler in the amount of 50-70 % of the volume into the composition. Carried out researches show, that at energy of irradiation 59 keV materials with thickness of 2-3 mm are sufficient for radiation protection. At high energy of irradiation 661 keV half radiation minimizing is provided at thickness of 15 mm using oxide of lead and tungsten technical of 50 % in volume as filler and at thickness of 35 mm using barium sulphate of 50 % in volume as filler.

Keywords: gamma shielding, radiation protection, mechanical properties, ethylene propylene diene monomer (EPDM), metal oxide, barite, rubber composites, prediction

1. Introduction

One of the most important problems of ecology is a problem on recycling of radiation-active waste, the final solution of which has not been found yet. Both radioactive materials which have fulfilled the resource, and materials and constructions that have come into contact with radiation must be recycled. It is especially difficult to recycle building constructions from radiation. An optimum variant of surface protection of building constructions from radiation is to use elastic self-adhesive radiation shielding sheetings which will be removed and reclaimed after saturation by radionuclides. The technology of creation of radiation shielding polymeric composites is based on adding particles of the materials possessing high absorption coefficients of energy of ionising radiation into a polymeric matrix. Application of lead or its oxide as a filler is considered to be a traditional way. Production of easy and inexpensive materials with high protective properties in a wide range of energies by adding lead oxide into a polymeric matrix is described in [1,2]. Modern tendencies require to create better and safer lead-free materials [3,4]. Studies [5,6] on the influence of barite content in polymer composite on a linear attenuation coefficient of radiation are carried out. In [7] oxide gadolinium is presented as an effective filler. Works [8] are an example of using bismuth as a filler for protection against gamma-ray. Works [9,10] describe the development and study of properties of a composite material, in which tungsten is a filler. On the basis of polystyrene and titan hydride the composite with neutron-protective properties [11] was developed. Nickel is used as one of layers of composite material [12].

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Not only metals can be fillers. At protecting against an ionising radiation boron is used as a filler. Works [13,14] report about obtaining a composite material on the basis of polyethylene of high density filled with nanoparticles of boron oxide. Application of boron oxide as a filler of a composite material on the basis of EPDM rubber (ethylene propylene diene methylene rubber) is described in [15]. Polymeric composite filled with white graphite, is described in [16]. In [17] polymeric composite was developed on the basis of high-molecular polyethylene, in which antimony oxide is a filler.

The works mentioned above describe the creation of hardened polymeric radiation shielding materials which cannot be quickly mounted and dismantled. One of the important requirements for radiation shielding materials, is the possibility of quick installation and dismantle [18]. From this point of view, the most perspective are elastic self-adhesive radiation shielding In order to obtain such materials, an elastic self-adhesive matrix has been developed [19]. The general disadvantage of creating existing radiation shielding materials is that the protective properties of materials are chosen experimentally.

Problem statement. The aim of the work is to develop a method of designing radiation protection compositions with the necessary protective properties.

There are the following tasks according to the problem:

- theoretical substantiation of definition of radiation shielding properties of a material calculation;
- experimental studies to confirm the theoretical justification.

2. Materials and Methods

Theoretical Part. To obtain composite materials with necessary radiation shielding properties the theoretical substantiation was made. Shielding properties of materials are characterized by a linear attenuation coefficient of a bunch γ-quanta. Intensity of bunch γ-quanta at passage through substance varies under the law

$$J = J_o \mathrm{e}^{-\mu x}, \quad (1)$$

where J_o- intensity of a bunch incidenting on a sample γ-beams, $MeV/cm^2 \cdot c$;J - intensity of a bunch after passage through a sample, $MeV/cm^2 \cdot c$; x - thickness of the sample, *cm*; μ - linear attenuation coefficient, *1/cm*;*e* - basis of the natural logarithm.

The linear attenuation coefficient μ represents the thickness of the material sample, weakening intensity of a stream γ-beams in times and is the sum of three summands: coefficient of photo-electric attenuation μ_{ph}, coefficient Compton attenuation μ_{com} and coefficient of twin attenuation μ_{tw}:

$$\mu = \mu_{\mathrm{ph}} + \mu_{\mathrm{com}} + \mu_{tw}. \quad (2)$$

The linear attenuation coefficient depends on density of substance and order numbers of elements [21] of which the substance consists. In case of a photo effect the linear attenuation coefficient is expressed by the following parity:

$$\mu_{\mathrm{ph}} = 0.089 \cdot \rho \frac{Z^{4.1}}{A} \cdot \lambda^h, \quad (3)$$

where ρ - density of capture substance; Z and A - element order number and atomic weight; λ - wave length γ-quantum; *h* - empirical factor.

Thus, the higher the capture density and the greater order number of elements that the capture consists, the higher its shielding properties will be.

Numerous researches have shown [20], that material structure defines features of distribution of the captured radiant energy in volume, however radiation shielding properties are defined by average chemical composition and material density. In this connection, average parameters of density and a linear attenuation coefficient of a composite can be defined as the first approximation by a rule of mixtures. So the composite density will be defined by the following expression:

$\rho_{\mathrm{c}} = \rho_{\mathrm{M}} \cdot V_{\mathrm{M}} + \sum_1^n \rho_{\mathrm{f}}^i V_{\mathrm{f}}^i,$ (4)

where ρ_{c}- composite density; ρ_{M} - matrix density; V_{M}- volume content of a matrix; ρ_f^i - density of *i* filler; V_{f}^i - volume content of *i* filler.

The factor of linear attenuation of a composite can be calculated using the following expression:

$\mu_{\mathrm{c}} = \mu_{\mathrm{M}} \cdot V_{\mathrm{M}} + \sum_1^n \mu_{\mathrm{f}}^i V_f^i,$ (5)

where μ_{c} - factor of linear attenuation of a composite; μ_{M} - linear attenuation coefficient of a matrix; V_{M} - volume content of a matrix;μ_{f}^i - linear attenuation coefficient of *i* filler; V_f^i- volume content of *i* filler, %.

Index of shielding efficiency of a material is the lead equivalent. Lead equivalent - an index of shielding efficiency of the material, equal to a thickness of a lead plate in millimeters, in as much time weakening a radiation dose, as well as the yielded material. From definition the conclusion about necessity of comparison of capture coefficients of radiation (relation of an initial stream to the past stream through a sample to a radiation flux) in lead and in a compared material is possible.

From the formula (1):

$J = J_o e^{-\mu x}$

we have:

$J_o / J = e^{\mu x}.$ (6)

By definition of a lead equivalent:

$J_o / J_{(\mathrm{Pb})} = J_o / J_{(\mathrm{sam})}\,,$ (7)

that is

$e_{(\mathrm{Pb})}^{\mu x} = e_{(\mathrm{sam})}^{\mu x},$ (8)

or

$\mu_{\mathrm{Pb}} x_{\mathrm{Pb}} = \mu_{\mathrm{sam}} x_{\mathrm{sam}}.$ (9)

We receive relation for calculation of a lead thickness x_{Pb}, which is equivalent to the sample thickness.

$x_{\mathrm{Pb}} = (\mu_{\mathrm{sam}} x_{\mathrm{sam}}) / \mu_{\mathrm{Pb}},$ (10)

or relation for calculation of sample thickness, which is equivalent to the lead thickness x_{Pb}:

$x_{\mathrm{sam}} = (\mu_{\mathrm{Pb}} x_{\mathrm{Pb}}) / \mu_{\mathrm{sam}}.$ (11)

This expression allows optimizing parameters of radiation shielding covering.

The theoretical studies have shown that to obtain radiation shielding materials with high shielding properties it is necessary to choose a matrix with high linear attenuation coefficient and filler (capture) with high density and high order number. The amount of such filler should be maximum. Considering that capture cost is high, it is necessary to optimize the quantity, taking into account sufficient material protection and its cost.

Experimental Part. Test problems have been developed to confirm the theoretical studies. The first test problem was to define the influence of absorption as a part of a material on radiation shielding properties. The second problem was to state the influence of an order number and atomic weight of absorbing substance on radiation shielding properties of a material.

The important indicator of radiation shielding properties of materials is frequency rate of attenuated radiation. Therefore the next task was to investigate the influence of a material thickness on frequency rate of attenuated radiation.

The next step in the research was to test the analytical expressions. For this purpose calculation of a linear attenuation coefficient from a filler level and its kind was carried out, and then it was compared to the experimental data.

Radiation shielding properties of materials were defined as follows. By means of detector initial intensity of electromagnetic radiation without the sample (I_0) and intensity of incident electromagnetic radiation which has passed through a plate from a radiation shielding material (I) are defined.

Intensity of an incident electromagnetic radiation is defined in the laboratory installation which schematic drawing is specified in Fig. 1.

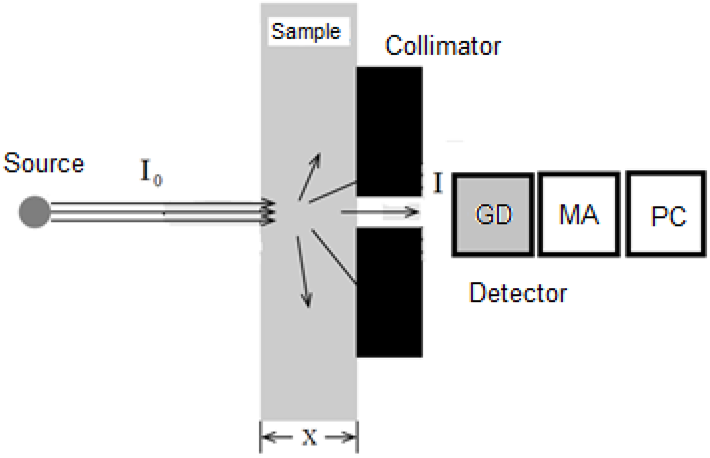


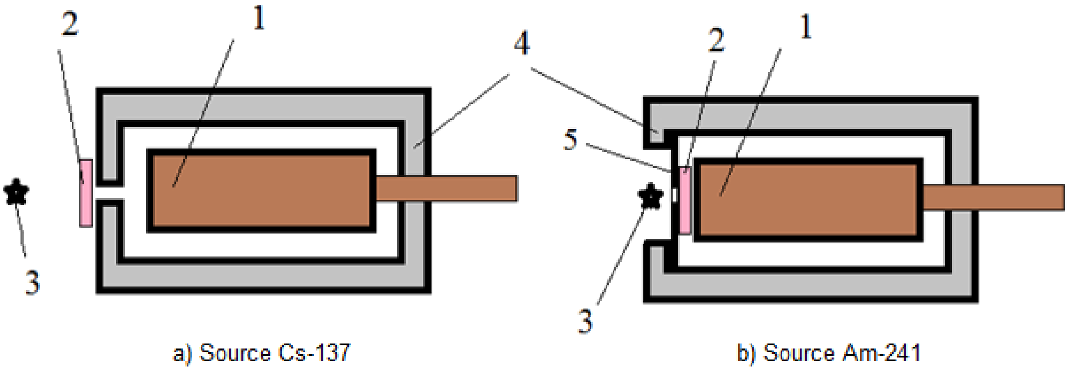
Fig. 1. Scheme of laboratory equipment. GD – a germanium detector; MA – multichannel analyze; PC–personal computer

For test from a material strip, with in advance measured thickness, samples of the required dimensions [(100x100) ±2] mm are cut down.

Isotope sources of gamma quanta Am-241 and Cs-137 (energy of gamma quanta 59 and 661 kV accordingly) are applied as radiation sources. (Isotope Am-241 corresponds to radiant energy of an x-ray tube with pressure on the anode 100 kV.)

At carrying out of tests with the sample at its irradiation by radiation source Am-241 the distance from a source to detector butt should make 8 mm (Fig. 2).

At use Cs-137 the yielded distance should be equal to 80 mm (Fig. 2).



a) Source Cs-137 b) Source Am-241

Fig. 2. Scheme of sample arrangement at testing: 1 – detector of gamma quanta; 2 - sample; 3 – source of gamma quanta; 4 – lead protection; 5 – collimator

For each source the initial flow of gamma quanta and optimum distance from detector butt to a source were defined. Then the sample and a source of gamma quanta are placed in front of detector butt. Thus the detector should be surrounded by lead protection serving by a primary collimator in diameter of 5 mm and protecting from background sources of gamma quanta and X-rays. At measurements using Am-241 as a collimator the lead plate with thickness of 0.5 mm with a collimator in diameter of 5 mm is applied. After that a gamma quanta flow which has passed through the sample is measured.

The linear attenuation coefficient of a material (3) in cm^{-1} is defined under the formula:

$$\mu = \frac{1}{x}\ln\frac{I_0}{I}, \tag{12}$$

where x - the sample thickness, I_0- initial intensity of an electromagnetic radiation; I- intensity of the incidenting electromagnetic radiation which has passed through a plate from a radiation shielding material.

The lead equivalent (δ_{pb}) in mm is defined by the formula:

$$\delta_{\mathrm{pb}} = \frac{\mu_{sam}\cdot \mathrm{x}}{\mu_{pb}}, \tag{13}$$

where μ_{sam} - a linear attenuation coefficient of tested material, cm^{-1}; μ_{Pb}- linear attenuation coefficient of lead at set radiation energies, cm^{-1}; h - sample thickness, mm.

3. Results and Discussion

As it follows from a theoretical substantiation, protective properties of a material depend on quantity of absorbant, its order number and atomic weight. In this connection, the influence of the amount of the filler on a linear attenuation coefficient of radiation is studied. Barite was taken as a filler. Its amount in the matrix has changed from 30 % to 70 %.

The results of the research are shown in Figs. 3,4 and in Table 1.

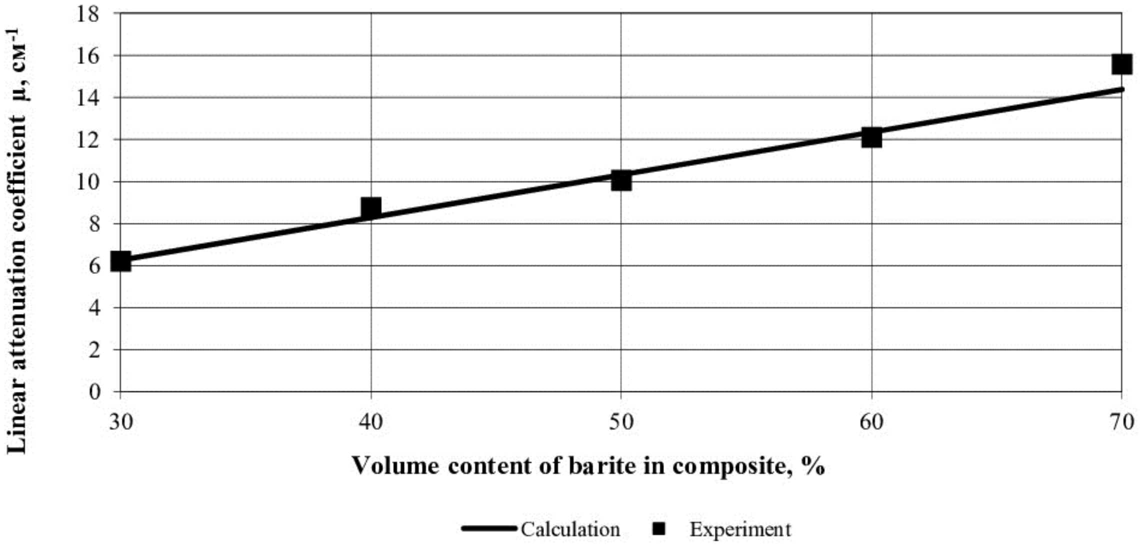


Fig. 3. Change of a linear attenuation coefficient At E=0.059 MeV depending on volume content of barite

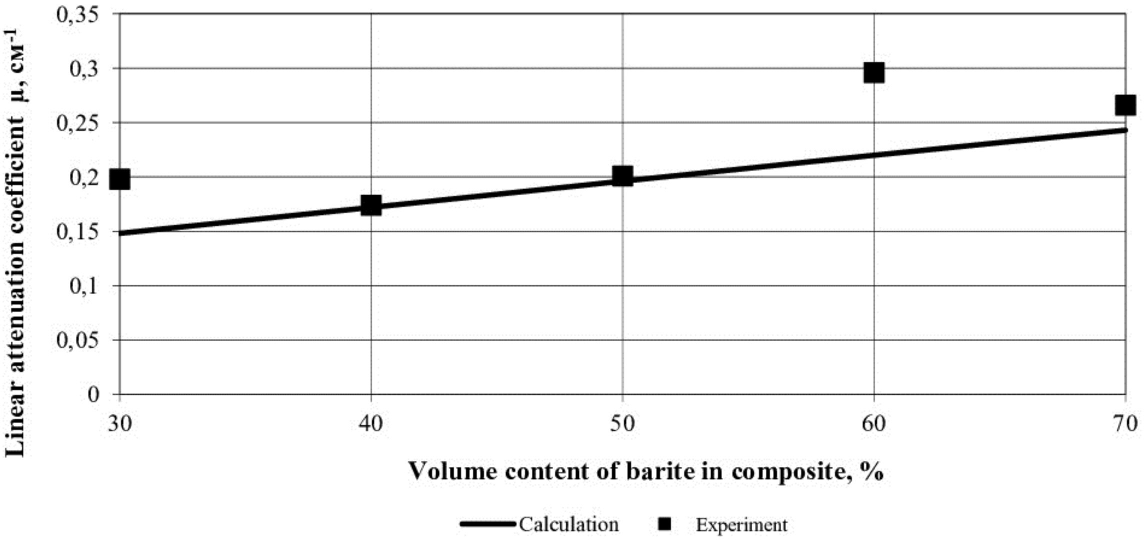


Fig. 4. Change of a linear attenuation coefficient at E=0.661 MeV depending on volume content of barite

Table 1.Sample test results on radiation shielding properties

Barite content, %	Density, g/sm^3	Sample thickness, mm	Linear attenuation coefficient, SM-1. at energy		Material thickness equivalent to1 mm Pb, at energy		Lead thickness equivalent to sample thickness, mm, at energy		Lead thickness equivalent to 1 mm of material, mm, at energy	
			59 keV	661 keV	59 keV	661 keV	59 keV	661 keV	59 keV	661 keV
30	1.9	5.2	6.22	0.198	8.16	5.96	0.64	0.87	0.12	0.17
40	2.23	5.1	8.765	0.174	5.79	6.78	0.88	0.75	0.17	0.15
50	2.56	5.0	10.06	0.201	5.05	5.87	1.02	0.87	0.2	0.17
60	2.88	4.7	12.1	0.295	4.19	4.0	1.12	1.18	0.25	0.25
70	3.21	4.8	15.58	0.266	3.26	4.43	1.47	1.08	0.31	0.23

The obtained results show (Table 1), that with filler content increasing the density of material and a linear attenuation coefficient increase, as well as the material thickness, equivalent to 1 mm of lead thus decreases. From this follows, that in order to obtain a material with high protective properties it is necessary to add into composition filler 50-70 % of the volume.

To state influence of order number and atomic weight of absorbant substance samples with fillers were made, and it is presented in Table 2. Volume content of filler was 50 %.

Table 2. Order numbers and atomic weight of filler substance

№	Substance	Order number in Periodic table	Atomic weight
1	Iron	26	55.847
2	Barite	56	137.33
3	Tungsten	74	183.85
4	Lead	82	207.19

Results of samples test with various types of fillers are presented in Table 3.

Table 3. Results of samples test with various kinds of filler

Sample marking	Density, g/sm^3	Sample thickness, mm	Linear attenuation coefficient, cm^{-1}. at energy		Lead thickness equivalent to sample thickness, mm, at energy		Sample thickness equivalent to 1 mm of material, mm, at energy		Material sample thickness equivalent to1 mm Pb, at energy	
			59 keV	661 keV	59 keV	661 keV	59 keV	661 keV	59 keV	661 keV
Iron	3.08	5.1	2.243	0.263	0.23	1.14	0.044	0.22	22.6	4.48
Barite	2.56	5.1	10.06	0.201	1.01	0.87	0.2	0.17	5.05	5.87
Tungsten	4.04	5	12.774	0.486	1.26	2.1	0.25	0.41	3.98	2.43
Lead	5.15	4.7	14.7	0.51	1.36	1.99	0.29	0.42	3.45	2.35

Studies have shown, that the higher order number and atomic weight of substance, the higher radiation shielding properties of a material. Using a capture with a high order number and atomic weight, it is possible to obtain a material with high radiation shielding properties. Thus, to obtain a material with necessary protective properties it is necessary to choose a certain type of filler and its quantity.

From table 3 it is seen, that applying corresponding filler and adding it in composition in certain quantity it is possible to obtain materials with high radiation shielding properties.

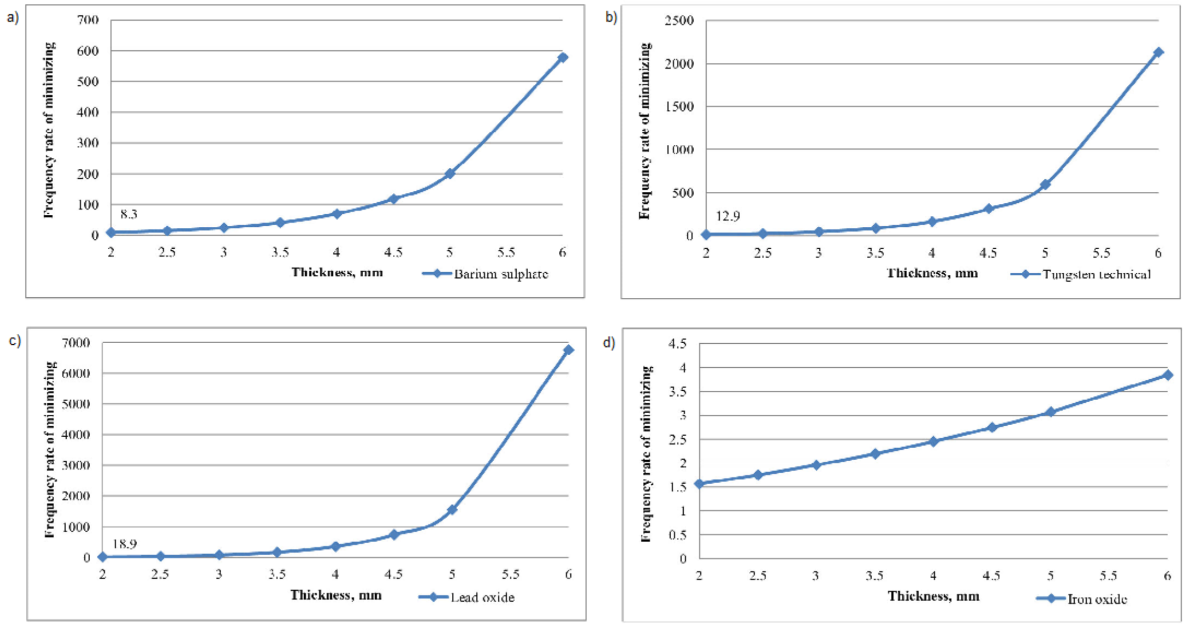


Fig. 5. Dependence of frequency rate of minimizing of dosage rate of gamma radiation from a material thickness with filler: a) barium sulphate; b) tungsten technical; c) lead oxide; d) iron oxide. Irradiation energy 59 keV

The important indicator of radiation shielding properties of materials is the frequency rate of radiation minimization. Carried out researches (Figs. 5, 6) have shown, that at energy of an irradiation 59 keV materials with thickness of 2-3 mm (Fig. 5) are sufficient for radiation protection. Thus quantity of gamma quanta, which have passed through a material, makes 0-0.5 %. At high energy of irradiation 661 keV frequency rate of minimizing increases with increase of material thickness (Fig. 6). Apparently from the presented graphs, half radiation minimizing is provided at thickness of 15 mm using oxide of lead and tungsten technical of 50 % of the volume (Fig. 6) as filler and at thickness of 35 mm using barium sulphate of 50 % of the volume as filler (Fig. 6).

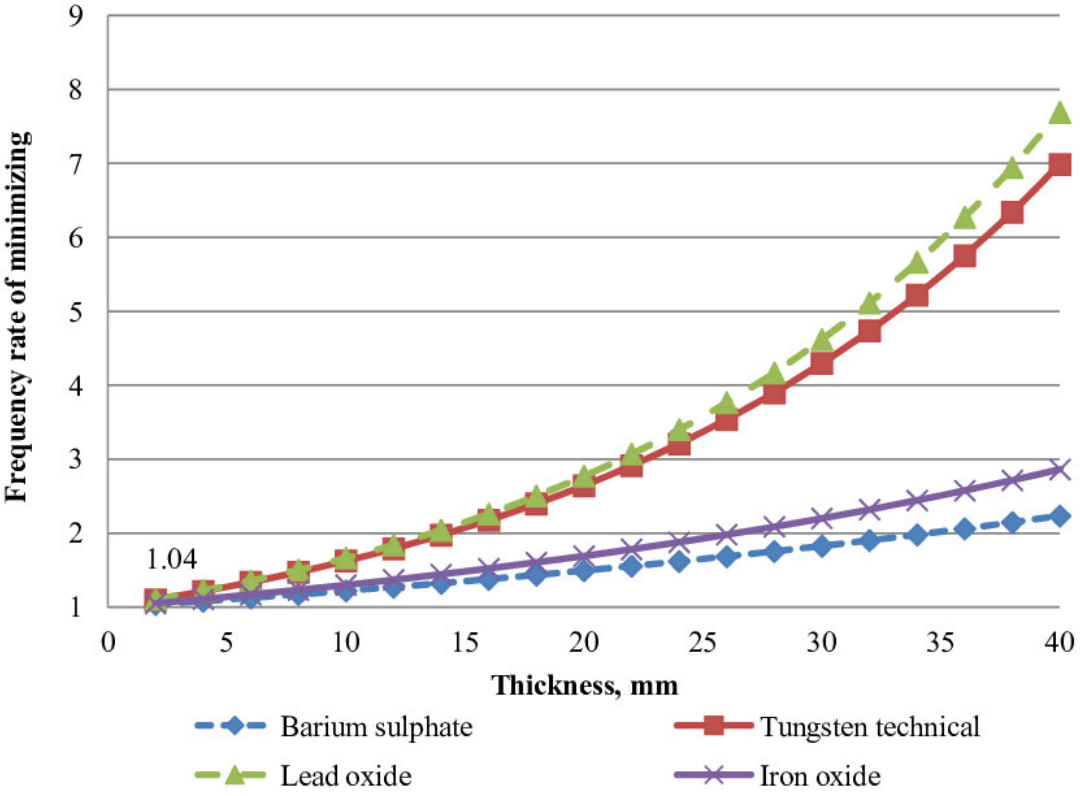


Fig. 6. Dependence of frequency rate of minimizing of dosage rate of gamma radiation from a material thickness with filler: a) barium sulphate; b) tungsten technical; c) lead oxide; d) iron oxide. Irradiation nary661 keV

The next stage of research was testing of analytical expressions obtained in the previous part. For this purpose, calculation of a linear attenuation coefficient from a filler level and its type was carried out, and then it was compared to experimental data. Results of calculations and experiments are presented in Table 4 and in Figs. 7-10.

Table 4 – Results of numerical calculations and experimental researches

Volume content of filler (Barite)	Linear attenuation coefficient μ, cm $^{-1}$			
	0.059 MeV		0.661 MeV	
	Calculation	Experiment	Calculation	Experiment
30	6.261	6.220	0.148	0.198
40	8.290	8.765	0.172	0.174
50	10.320	10.06	0.196	0.201
60	12.345	12.1	0.220	0.296
70	14.379	15.58	0.243	0.266

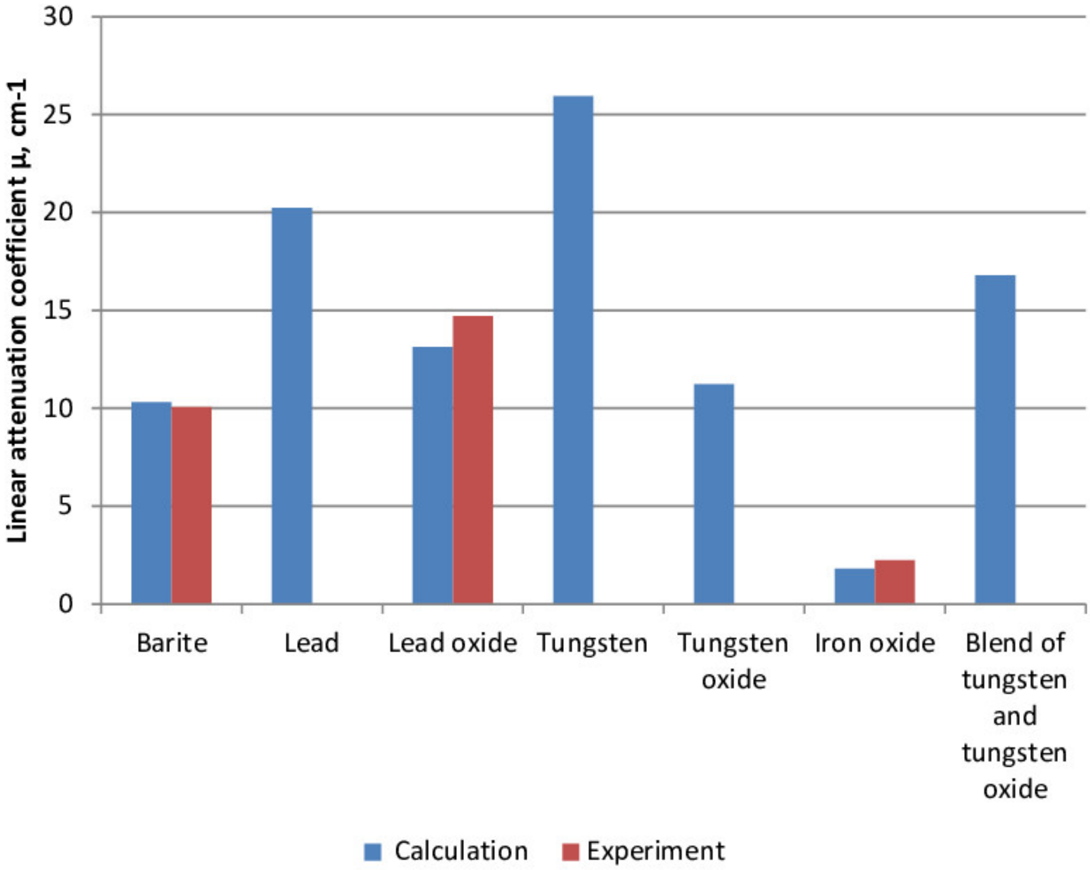


Fig. 7. Efficiency of fillers at E=0.059 MeV at their volume content in a composite 0.5 in volume

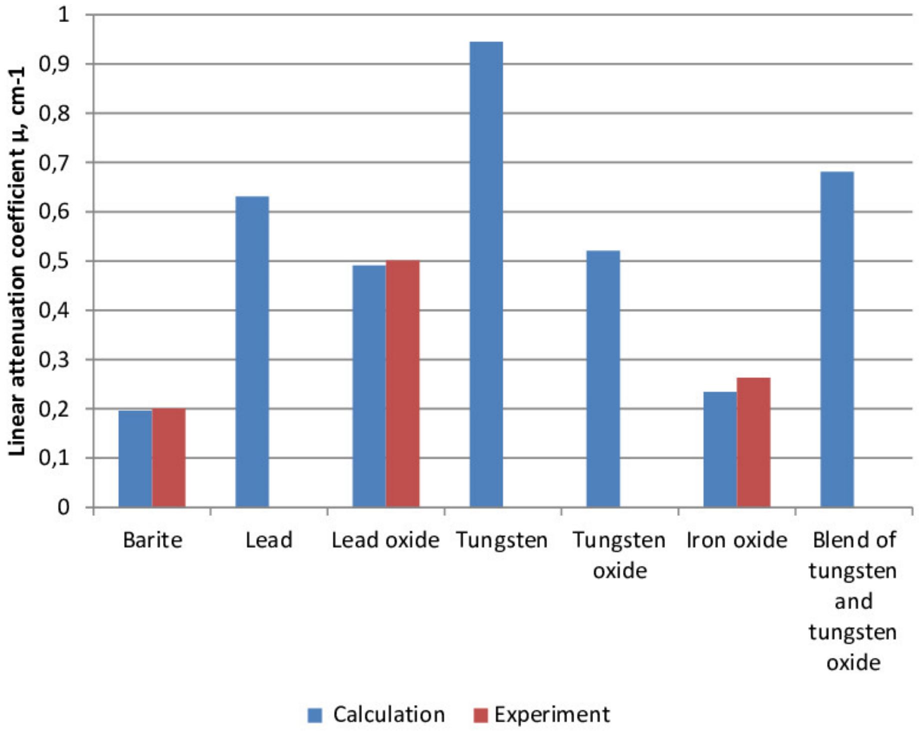


Fig. 8. Efficiency of fillers at E=0.661 MeV at their volume content in a composite 0.5 in volume

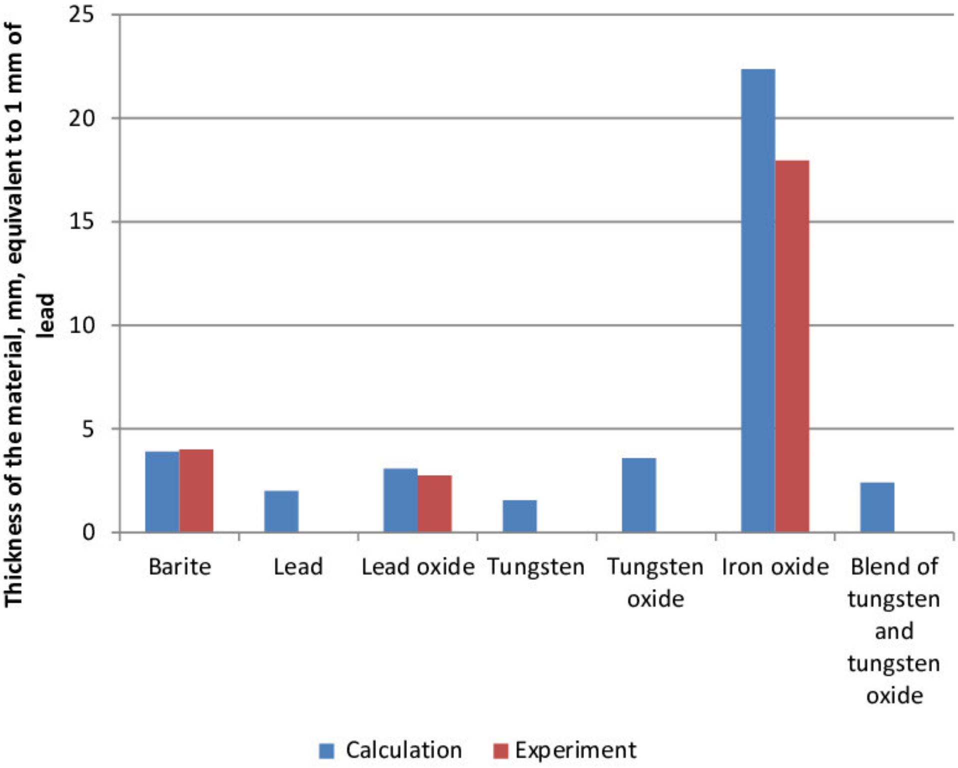


Fig. 9. Efficiency of fillers at E=0.059 MeV at their volume content in a composite 0.5 in volume

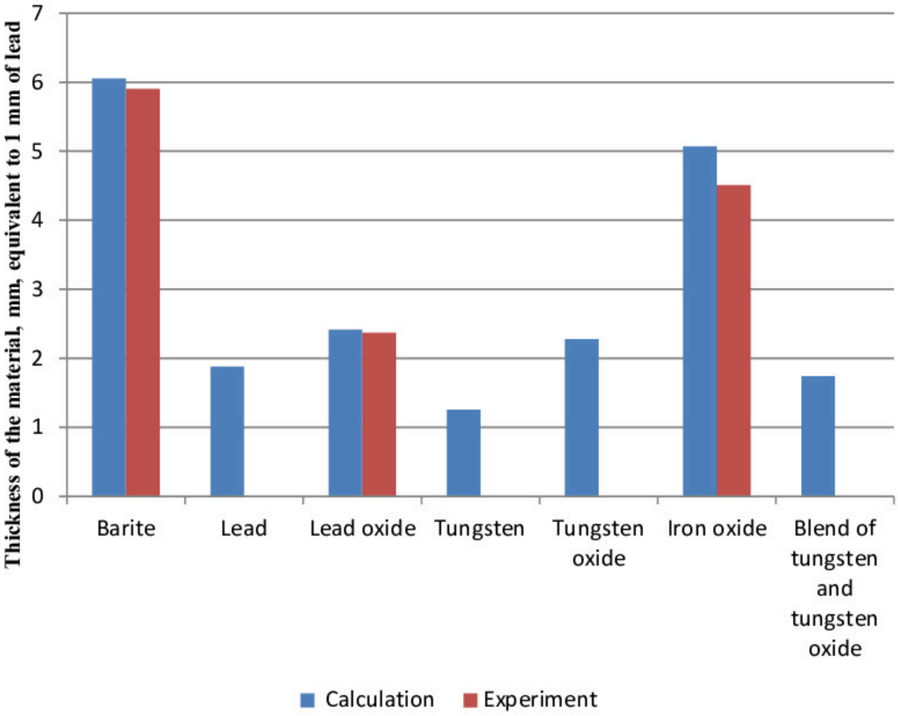


Fig. 10. Efficiency of fillers at E=0.661 MeV at their volume content in a composite 0.5 in volume

The analysis of graphs shows good convergence of theoretical calculations with experiment. Values of linear coefficients of filler substance were taken from [21]. Values of linear coefficients of substance are resulted for various energies. For calculations two radiant energies 60 keV and 661 keV were taken.

All these data testify to correctness of a theoretical substantiation.

On the basis of theoretical substantiation the computer program «Designing of compositions of radiation shielding» was developed (certificate of state Registration №2019614058), allowing to design material compositions with necessary radiation shielding properties.

4. Conclusions.

1. With filler content increasing, the density of material and a linear attenuation coefficient increase, and material thickness, equivalent to 1 mm of lead thus decreases. From this follows, that in order to obtain material with high protective properties it is necessary to add filler of 50-70 % volume into the composition.
2. Applying a capture with a high order number and atomic weight it is possible to obtain material with high radiation shielding properties. Applying barite or tungsten filler and adding it in composition in certain amount allows to obtain materials with high radiation shielding properties. Carried out researches have shown, that at energy of an irradiation 59 keV materials with thickness of 2-3 mm are sufficient for protection. At high energy of irradiation 661 keV half radiation minimizing is provided at thickness of15 mm using oxide of lead and tungsten technical 50 % volume as filler and at thickness of 35 mm using barium sulphate of 50 % volume as filler.
3. The analysis of graphs shows good convergence of theoretical calculations with experiment.

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