

Radiation-attenuating compounds in the system “ $\text{Na}_2\text{O} \cdot \text{Al}_2\text{O}_3 \cdot 8\text{SiO}_2 \cdot 15\text{H}_2\text{O} - \text{WNi} - \text{MoSi}_2 - \text{B}_4\text{C}$ ”

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Abstract. The paper considers the influence of refractory compounds on the gamma attenuation of geocement compounds intended for radioactive waste conditioning. A positive influence of tungsten-nickel alloy and boron carbide on the total gamma attenuation and linear attenuation coefficient is noted, and molybdenum disilicide on thermal scattering, which contributes to the slowdown of radiolysis during the conditioning of radioactive waste and reduces the gamma load on metal drums and the inner surfaces of concrete containers. These requirements are met by a compound containing WNi – 2.85-6.34%, MoSi₂ – 5.7-10.9%, B₄C – 1.82-3.23%.

Keywords: geocement, conditioning, compound, radioactivity, radiolysis, radioactive waste, refractory compounds, absorption coefficient, linear attenuation coefficient, metal and concrete containers.

Currently, a wide range of binding agents is used for conditioning low- and medium-level radioactive waste, selected with a view to ensuring the long-term durability of the compounds. Depending on the type of waste and its impact on the radiation resistance of matrices, the surfaces of metal drums and concrete containers, it is most appropriate to use alkaline binding systems, primarily aluminosilicates [1, 2]. Their selection, in addition to the reliable adsorption-chemical binding of radionuclides in water-insoluble compounds, is also determined by the accelerated processes of polycondensation of zeolite-like new formation under the intense action of gamma radiation [3]. However, as a result of radiolysis [4], a significant increase in temperature in the range of 700-950°C is recorded in the compounds. In addition to radiation, this increase contributes to a decrease in the structural properties of metal container drums. This is a rather pressing issue that needs to be addressed in order to ensure compliance with the conditions for intermediate and long-term storage of radioactive waste [5].

The object of the study is compounds based on mordenite-type geocement [1, 6-9], filled with refractory metallic and non-metallic compounds [10-12] the physical properties of which are given in Table 1.

The oxidation onset temperature of the above compounds is quite high. This is important for the resistance to self-heating of cemented radioactive solutions from radiolysis during long-term storage [12].

Table 1. Physical properties of refractory non-metallic materials

Title	ρ , g/cm ³	λ , W/m·K	C_p , J/g·K	α , 10 ⁻⁶ degrees ⁻¹	T oxidation, °C
WNi	18,75	190	0,14	4,0	1700
MoSi ₂	6,3	0,07	0,45	4,0	1700
B ₄ C	2,52	11,0	-	4,5	1000

Tungsten-nickel alloy, together with boron carbide, scatters gamma radiation and absorbs thermal neutrons. Molybdenum disilicide, in addition to gamma attenuation, acts as a heat insulator. Optimization of the compound compositions was carried out using a three-factor simplex central experimental design in the Statistica 12 mathematical environment with the implementation of a special cubic model that takes into account the nonlinearity of the influence of factors on the properties of the initial parameters.

Variation factors and the matrix for planning the experiment are given in Table. 2 and Table. 3.

Table 2. Variation intervals and values of variable factors

Factors, appearance	Natural	Coded	Levels of variation		Variation interval
			0	1	
W _{Ni}	%	X ₁	2,5	7,5	5
MoSi ₂	%	X ₂	5	15	10
B ₄ C	%	X ₃	1,5	3,5	2

Table 3. Experiment planning matrix

Plan points	Matrix plan in codes			Full-size matrix plan		
	X ₁	X ₂	X ₃	W _{Ni} , %	MoSi ₂ , %	B ₄ C, %
1	0,00	1,00	0,00	2,5	15	1,5
2	0,33	0,33	0,33	4,17	8,33	2,17
3	1,00	0,00	0,00	7,5	5	1,5
4	0,50	0,50	0,00	5	10	1,5
5	0,00	0,00	1,00	2,5	5	3,5
6	0,50	0,00	0,50	5	5	2,5
7	0,00	0,00	0,50	2,5	10	2,5

The following initial parameters were selected as initial parameters: density, porosity, gamma attenuation coefficient (scattering), linear attenuation coefficient, which satisfy the initial criteria: $\rho \rightarrow \max$, $P \rightarrow \min$, $R_{cm} > 4.9$ MPa, $\gamma \rightarrow \max$, $\mu \rightarrow 1$.

The numerical values of the parameters were determined according to the Technical Specifications of Ukraine on cubes measuring 3x3x3 cm after their hardening at a temperature of 80°C for 6 hours. The gamma attenuation and linear attenuation coefficients were determined according to the method and calculated according to the formulas given in [13].

As a result of processing the experimental results, mathematical models were obtained in the form of regression equations (1-5), which describe the influence of varied factors on the properties of the compounds, and their graphical interpretation is shown in the figure:

$$\rho = 2,01x_1 + 1,92x_2 + 1,93x_3 - 0,02x_1x_2 - 0,08x_1x_3 - 0,18x_2x_3 + 2,64x_1x_2x_3 \quad (1)$$

$$P = 7,65x_1 + 10,31x_2 + 7,37x_3 - 3,36x_1x_2 - 0,32x_1x_3 - 2,04x_2x_3 - 4,53x_1x_2x_3 \quad (2)$$

$$R_{cm} = 28,4x_1 + 26,8x_2 + 28,3x_3 - 5,1x_1x_2 + 13,4x_1x_3 + 49,9x_2x_3 - 110,2x_1x_2x_3 \quad (3)$$

$$\gamma = 22,1x_1 + 21,1x_2 + 23,4x_3 - 1,6x_1x_2 + 5x_1x_3 + 0,6x_2x_3 - 6,6x_1x_2x_3 \quad (4)$$

$$\mu = 0,456x_1 + 0,438x_2 + 0,446x_3 + 0,02x_1x_2 + 0,02x_1x_3 + 0,03x_2x_3 + 0,32x_1x_2x_3 \quad (5)$$

From the presented mathematical models (1-5) it is clear that the change in the initial properties of compounds is most significantly influenced by the main variation factors - x_1 , x_2 and x_3 , in some cases, their products.

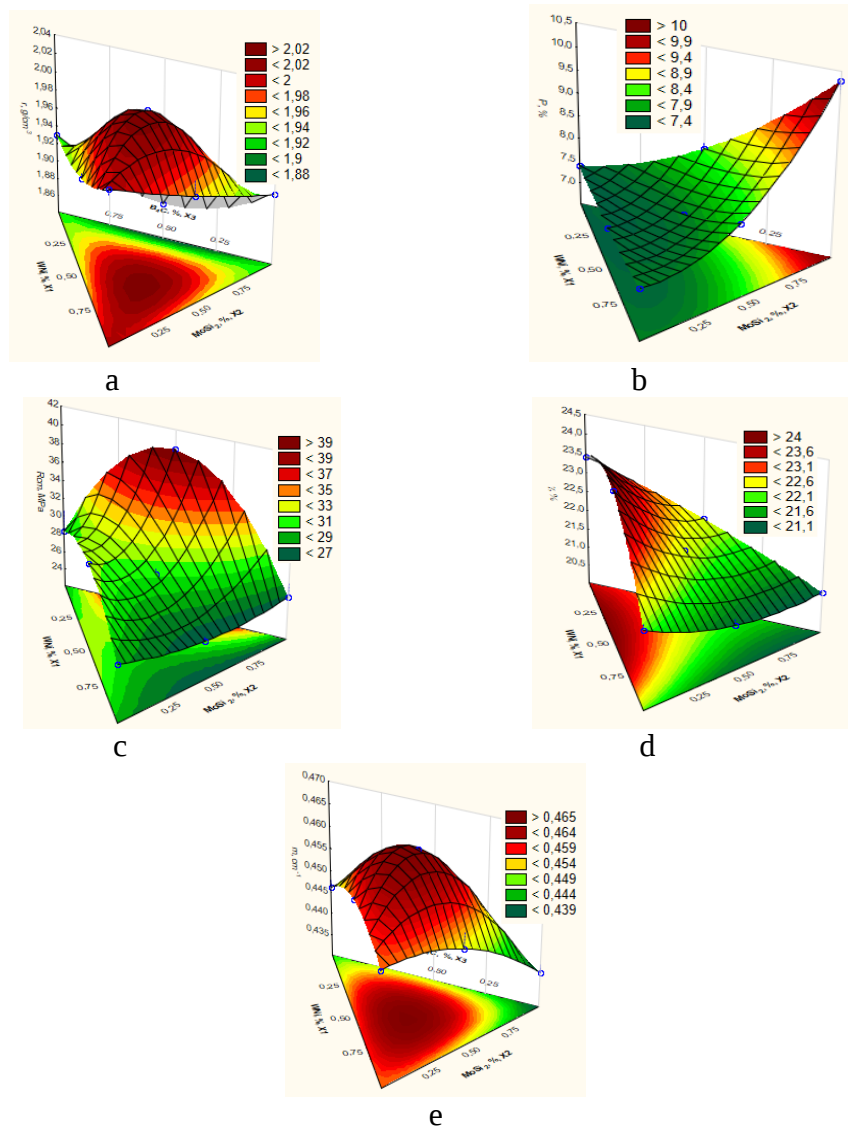


Fig 1. Ternary response surfaces of the influence of variation factors on the change in the indicators of structural materials: a – density; b – porosity; c – compressive strength; d – attenuation gamma; e – linear attenuation coefficient

As can be seen from the figure, the maximum density values (up to 2.2 g/cm³) are achieved when the compound contains WNi in an amount from 3.75 to 7.5% (factor x1), MoSi₂ in an amount from 5 to 12.5% (factor x2) and B₄C in an amount from 1.5 to 3.25% (factor x3). The minimum porosity values (up to 7%) are achieved when the compound contains WNi in an amount from 2.5 to 7.5% (factor x1), MoSi₂ in an amount from 5 to 10% (factor x2) and B₄C in an amount from 1.5 to 3.55% (factor x3). The maximum values of compressive strength (up to 40 MPa) are achieved when the compound contains WNi in an amount of 2.5 to 3.25% (factor x1), MoSi₂ in an amount of 7.5 to 12.5% (factor x2) and B₄C in an amount of 2 to 3% (factor x3). The maximum values of the gamma attenuation coefficient (up to 25%) are achieved when the compound contains WNi in an amount of 2 to 7% (factor x1), MoSi₂ in an amount of 5 to 8% (factor x2) and B₄C in an amount of 2 to 3.5% (factor x3). The maximum values of the linear attenuation coefficient (up to 0.468 cm⁻¹) are achieved when the compound contains WNi in an amount from 3 to 6.5% (factor x1), MoSi₂ in an amount from 6 to 11.5% (factor x2) and B₄C in an amount from 2.1 to 2.7% (factor x3).

By superimposing the ternary response surfaces on each other, the optimal region is determined that provides the criterion requirements for compounds based on mordenite-type geocement, namely: WNi – 2.85-6.34%, MoSi_2 – 5.7-10.9%, B_4C – 1.82-3.23%.

Mordenite-type geocement modified with an optimal amount of refractory compounds has been successfully tested in the conditioning of low-level radioactive waste.

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