Thermophysical Properties of Radiation-Protective Composite Materials

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Abstract. The paper discusses the development of compositions and studies of the properties of heavy composite materials with atypically high heat insulating performance. When choosing the suitable matrix material, epoxy resin, sodium liquid glass and Portland cement were tested. Filler in all compositions was a lot of tonnage waste of the glass industry - heavy flint. The degree of filling of the matrix material in the preparation of press composites reached 95...97% (by volume). This allowed to increase the average density of the material to 3800 ... 3850 kg/m³ and provided the required level of protection against penetrating radiation.

1. Introduction

Because of man’s anthropogenic activity, the natural background of ionizing radiation has increased substantially in vast territories with a large population density. Its negative impact on human health requires the development of new building materials for buildings, located in potentially hazardous areas.

Wall structures from conventional materials do not have the function of protecting internal environment from external ionizing radiation. The solution of the problem consists in using for this purpose specially designed materials of increased density for effective attenuation of gamma radiation. Currently used technological methods for obtaining superheavy concrete, consisting in the use of metal containing aggregates, lead to a sharp decrease in their thermal protection qualities, which are the most important indicators of the enclosing structures of modern energy-efficient houses.

Making the composite difficult to combine indicators of high density and low thermal conductivity was achieved by selection of the structure, composition and volume content of the filler in areas, exceeding the percolation threshold.

The theory of the thermal conductivity of dielectric materials and the empirical data, used in the design of glass, allowed us to formulate the principles for optimizing the chemical composition of the filler. It was found, that to improve the thermophysical parameters of heavy composites as fillers, it is expedient to use [1, 2]:

- vitreous substances, in which there are oxides of alkali metals, reducing the length of free movement of phonons due to an increase in the number of isolated silicon-oxygen tetrahedra and fragmentation of the microstructure;
substances with a high atomic mass (PbO, Pb₂O₃), which due to considerable thermal inertia effectively dissipate the energy of the heat flux [3].

As a filler, satisfying the requirements, a finely ground waste of substandard optical glass (heavy flint) of the following oxide composition was selected, %: (SiO₂=25...30; K₂O=3...4; PbO=68...70; ZnO=0,3...0,4) with a density ρ=3850...3900 kg/m³ and the coefficient of thermal conductivity λ=0,35...0,38 W/(m·°C).

2. Evaluation of the thermal conductivity of a heavy composite

2.1. Thermal conductivity of dispersed phase

The calculated dependences, based on the additive approach to the formation of thermophysical and strength parameters of glass, have the form:

- for strength evaluation:

\[ R_{compr(p)} = k_1 \cdot C_1 + k_2 \cdot C_2 + \ldots + k_i \cdot C_i \]  

(1)

- for evaluation of thermal conductivity:

\[ \lambda = \sum C_i \cdot \lambda_i \, , \]  

(2)

where \( \lambda_i \) and \( k_i \) – the coefficients; \( \lambda_i \) – the content of the i-th oxide, % by weight.

The values of the coefficients of equations (1) and (2) are given in table 1.

| Oxide   | \( \lambda_i \) | \( k_i \) | Oxide   | \( \lambda_i \) | \( k_i \) |
|---------|-----------------|---------|---------|-----------------|---------|
| Na₂O    | 0,6             | 0,02    | 0,0065  | Al₂O₃           | 1       | 0,05   |
| K₂O     | 0,05            | 0,01    | 0,0024  | B₂O₃           | 0,9     | 0,065  | 0,0066 |
| MgO     | 0,1             | 0,01    | 0,0134  | P₂O₅           | 0,76    | 0,075  | 0,0056 |
| CaO     | 0,2             | 0,2     | 0,0116  | SiO₂          | 1,23    | 0,09   | 0,0087 |
| PbO     | 0,48            | 0,025   | 0,0020  | BaO            | 0,62    | 0,05   | ---    |

2.2. Estimation of the total thermal conductivity of the composite material

Vitreous filler is an absolutely dense material (\( \Pi_{\text{total}} = 0 \)), i.e. the entire volume of air pores is concentrated in the matrix. The coefficient of thermal conductivity of such a matrix with air inclusion (\( \lambda_{\text{matr+air}} \)) is calculated by the formula [4]:

\[ \lambda_{\text{matr+air}} = \lambda_{\text{matr}} \cdot \frac{\lambda_{\text{air}}}{\left(1 - \sqrt[3]{\Pi_{\text{total}}} \right) + \lambda_{\text{matr}} \cdot \sqrt[3]{\Pi_{\text{total}}}} + \lambda_{\text{matr}} \cdot \left(1 - \sqrt[3]{\Pi_{\text{total}}} \right) , \]  

(3)

where \( \Pi_{\text{total}} \) – the total porosity of the heavy composite, relative units; \( \lambda_{\text{matr}} \) and \( \lambda_{\text{air}} \) – the coefficients of thermal conductivity of matrix matter and air, W/(m·°C).

The thermal conductivity of the next scale level of the structure of a heavy composite includes a porous matrix and filler particles (heavy flint). By analogy with equation (3) we have [5, 6]:

\[ \lambda_{hc} = \lambda_{\text{matr+air}} \cdot \left(1 - \sqrt[3]{\Pi_{\text{matr}}} \right) + \lambda_{\text{matr+air}} \cdot \left(1 - \sqrt[3]{\Pi_{\text{matr}}} \right) \]  

(4)

where \( \lambda_{hc} \) и \( \lambda_{\text{fil}} \) – coefficients of thermal conductivity of a heavy composite and filler, W/(m·°C); \( \Pi_{\text{fil}} \) – the relative degree of filling of the matrix:

\[ V_{\text{fil}} = \frac{V_{\text{fil}}}{V_{\text{matr}} + \Pi_{\text{total}}} , \]  

(5)

where \( V_{\text{fil}} \) and \( V_{\text{matr}} \) – content of filler and matrix in the composite, % by volume.
Investigation of the effect of the granulometric composition of the filler on the thermal conductivity of the composite was carried out using the method of mathematical design of the experiment. As the variable parameter the particle size of the filler was taken, mm: 2,5 \( (Y_1) \); 0,63 \( (Y_2) \); 0,14 \( (Y_3) \).

As a result of the experiments, the dependencies of the coefficient of thermal conductivity of the composites on the granulometric composition, the amount and type of binder are obtained. For a heavy composite on an epoxy matrix with a resin content of 6% of the mass of the filler the thermal conductivity has the form:

\[
\lambda_{\text{eff}} = \exp(-1,207Y_1 - 1,378Y_2 - 1,276Y_1 + 0,262Y_1Y_2 + 0,236Y_2Y_3)
\]

Figure 1 shows the concentration diagrams of the value of \( \lambda_{\text{eff}} \) for an epoxy composite

![Concentration Diagrams](image_url)

**Figure 1.** Influence of the granulometry of the filler on the thermal conductivity of epoxy composite with different degree of filling, %: a – 97; b – 94

The results of the studies showed, that the thermal conductivity of composites increases with an increase in the binder content and an increase in the density of the material. Thus, by changing the amount of binder and granulometric composition of the filler, it is possible to control the process of forming the thermal conductive properties of extremely filled composites to protect against radiation [7, 8].

In the structure of extremely filled composites there is often a deficit of matrix material, which significantly reduces the strength properties of the material. The experiment showed, that the minimum content of epoxy binder, providing sufficient for the wall material strength (5...6 MPa), is 3%.

Because of studies of the sorption ability of composites, it was found, that the nature of the absorption of moisture from the ambient air is determined by the type of matrix and the degree of its filling. Increased sorption capacity is characterized by composites on a liquid-glass matrix, and minimal – on an epoxy matrix.

The thermal conductivity of the epoxy composite varies insignificantly with the change in the moisture content of the ambient air: its increase to 98...99% is accompanied by an increase in thermal conductivity by 2...3%. For compositions with liquid-glass and cement binders the analogous dependence is significantly higher and amounts to 4...5 and 10...11%, respectively.

At a density of 3500...3600 kg/m\(^3\) the developed compositions show thermal conductivity \( \lambda = 0,21...0,27 \text{ W/(m\(^\circ\)C)} \) comparable to the thermal conductivity of widely used wall materials – cellular concrete with a density of 800 kg/m\(^3\).

Given the characteristics of the properties of heavy composites, it is advisable to use them as a heat-accumulating layer of the external enclosure structures of the building in combination with a low-density insulation. In such constructions, an effective thermal insulation material (for example, mineral wool or foam-glass) prevents the rapid loss of heat by the composite material, which in this case serves as a heat accumulator [9].
The use of heavy composites of reduced thermal conductivity contributes to the effective damping of the temperature flow in the outer enclosures. Being as a part of the enclosing structures, such composites with an extremely low coefficient of thermal diffusivity \( \alpha = \frac{\lambda}{c \cdot \rho} \) ensure the formation of more comfortable thermal microclimate of an internal air environment. Calculations show, that the value of \( \alpha \) for heavy composites is 1.5...3.5 times smaller in comparison with cellular concrete.

With cyclic temperature changes, the reliability of the external enclosing structure is ensured by the joint operation of its structural layers [10]. This is possible with the equality of temperature stresses, arising in them and caused by the thermal expansion coefficients of materials \( \alpha_t \). The values, obtained as a result of laboratory tests, make it possible to conclude, that the thermal expansion of composites depends on the degree of their filling: for all the compositions studied, the value of \( \alpha_t \) increased with the content of the binder.

The developed composites with a matrix on epoxy resin (3%), liquid glass (11%) or cement (20%) are well combined in temperature deformation with traditional materials (concrete, reinforced concrete, ceramics, etc.).

**Summary** The conducted studies have revealed the practical expediency of using heavy composites, with composition and structure optimized for thermophysical properties, in external enclosure structures not only to protect the internal environment from radiation but also as thermal protection of buildings. The regulation of thermal conductivity in the selection of composites allows them to be used both in enclosing structure to reduce the heat loss of buildings and in the construction of the protection of special installations for faster heat dissipation. In conventional buildings, designed for use in areas with an elevated background of radioactive radiation, the most effective properties of heavy composites are shown as heat accumulating layers in combination with low-density insulation.

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