Matrices of radiation-protective composites using bismuth oxide

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Abstract. The article presents the results of investigations of radiation-protective composites with two types of matrices based on chamotte and aluminous binders. The synthesis of binders was carried out according to the principles of the production of ceramic concrete based on the artificial ceramic binders (ACB). Bismuth oxide was selected as filler. Basic physical and mechanical, as well as radiation-protective characteristics, of composites with different ratios of ACB and Bi₂O₃ were shown. It was found out that binder of high-alumina chamotte can be used as an optimal matrix base. Composites on its basis have higher structural and radiation-protective properties.

1. Introduction
Nuclear power has long proved its economic efficiency in virtually all areas of the globe. Together with thermonuclear power, it is currently the most promising method of generating electricity. However, their development must be ensured by a high level of safety, including the development of new materials with high radiation-protective and structural properties [1-4].

The materials for protection against gamma radiation are of particular interest. As is known, gamma radiation has relatively weak ionizing ability and a very large penetrating power (for example, it may pass through a layer of lead with a thickness of 5 cm). As the energy of the γ-quanta (Eγ exceeds 0.5 MeV) increases, the main process of interaction with the medium would be the photon scattering by electrons (the Compton effect). In this case, the decisive role in characterizing the protective properties of material is due to its density. For such “heavy” radiation-protective materials, it is important to maintain sufficient structural properties. One of the promising materials are the ceramic composites with a given spectrum of initial properties, which can be further modified, depending on the purpose and conditions of the application field [5-6].

Taking into account everything mentioned above, the aim of this work is to study radiation-protective ceramic composites in the Bi₂O₃ - Al₂O₃ - SiO₂ system with further choice of the optimal matrix composition [7-9].

2. Materials and methods
In the course of the study, it was proposed to use the ceramic binders of two types: 1) on the basis of chamotte mullite; 2) on the basis of technical alumina. Synthesis of artificial ceramic binder (ACB) was carried out by wet grinding in a batch ball mill with the material loaded in a step by step way. This principle of obtaining the ceramic concrete makes it possible to obtain a matrix with the required phase
composition, high physical and mechanical characteristics; besides, it provides wide possibilities for the application of various fillers [10, 12]. Rheological properties of the suspension were studied using the Reotest-2 flow meter. A suspension based on alumina has a thixotropic nature of the flow (the viscosity of the liquid decreases with time); a suspension based on chamotte has a thixotropic-dilatant nature of the flow (destruction of the initial thixotropic structure and the subsequent dilatant structuring). The main properties of ACB are presented in Table 1.

| ACB type | Density, g/cm³ | Relative humidity, % | Flow time, s | Content of Al₂O₃ | Content of SiO₂ |
|----------|----------------|---------------------|-------------|-----------------|---------------|
| Aluminous| 1.99           | 18                  | 65          | 99.6            | 0.3           |
| Chamotte | 2.45           | 13.8                | 98          | 75.6            | 23.8          |

The bismuth oxide powder was used as filler with spherical particles and a size of less than 35 µm. Electron microscope images of both ACB and filler are presented in Fig.1. The shooting was carried out in the Center for High Technologies of the BSTU named after V.G. Shukhov with the help of scanning electron microscope TESCAN MIRA 3 LMU.

![Figure 1. Electron microscope images of Bi₂O₃ filler (a), particles of ACB chamotte (b) and aluminous ACB (c)](image)

The evaluation of radiation-protective characteristics of composites was carried out according to the following procedure. A plate of material (composite) with thickness was placed between the collimated radiation source and the collimated detector. Such experiment (and its model) is called an “experiment in the geometry of a narrow beam” in accordance with GOST 25146-82.

3. Study of ceramic and radiation-protective composites

The results of the study of ceramic composites with a matrix of fireclay composition were presented in this article [12]. Besides, this article lists the main physical and mechanical, as well as radiation-protective, characteristics. Moreover, the optimal conditions for obtaining and correlaton of components were revealed. However, along with the bismuth oxide, alumina was additionally used as a filler in the prototypes. Therefore, to compare the results, the compositions shown in Table 2 were used to mould the test beam applying the method of static pressing with a specific pressure of 100 MPa.
Table 2. Compositions of radiation-protective materials

| No. of composition | Aluminous ACB | Chamotte ACB | Bi$_2$O$_3$ |
|--------------------|---------------|--------------|-------------|
| 1                  | 40            | --           | 60          |
| 2                  | 30            | --           | 70          |
| 3                  | 20            | --           | 80          |
| 4                  | --            | 40           | 60          |
| 5                  | --            | 30           | 70          |
| 6                  | --            | 20           | 80          |

The molded samples were first dried at a temperature of 100-110 °C and fired afterward. Basing on some other research works [12, 13], the optimal firing temperature for both matrices was determined to be 800 °C. The samples fired at this temperature were characterized by the highest density, strength and minimum water absorption. The general view of samples after firing is shown in Fig. 2.

![Figure 2. General view of samples after firing: a – on the basis of alumina suspension, b – on the basis of chamotte suspension](image)

After the heat treatment, using X-ray phase analysis, the mineralogical composition of the samples was determined. Corundum (α-Al$_2$O$_3$) and bismuth aluminates (Al$_2$O$_3$ · 12 Bi$_2$O$_3$ anf 2Al$_2$O$_3$ · Bi$_2$O$_3$) are observed in the samples based on alumina ACB. In the samples based on chamotte, one may observe silicates of bismuth (12Bi$_2$O3·2SiO$_2$ and Bi$_2$O$_3$·SiO$_2$), the formation of which occurs due to decomposition of the mullite component of the chamotte part. In both cases, the reflections typical for bismuth oxide are completely absent.

After this, the basic physical and mechanical, as well as radiation-protective properties presented in Table 3 were determined, basing on the samples with different content of bismuth oxide after heat treatment. The radiation-protective properties were estimated from the value of the linear attenuation coefficient of a point gamma source (a narrow beam with a power of 1120 keV).

Table 3. The main characteristics of ceramic radiation-protective materials

| No. of composition | Water absorption, % | Density, g/cm | Bending strength, MPa | Linear attenuation coefficient, cm$^{-1}$ |
|--------------------|---------------------|---------------|-----------------------|------------------------------------------|
| 1                  | 23                  | 4.1           | 1.8                   | 0.58                                     |
| 2                  | 25.8                | 4.4           | 1.9                   | 0.67                                     |
From the data provided, it can be seen that the samples with a matrix based on alumina ACB with Bi$_2$O$_3$ content of 60% possess the lowest density, strength and linear attenuation coefficient. As the content of heavy aggregate increases, the indicators improve, however, insignificantly.

In the case of the chamotte suspension, a reverse trend is observed. The highest physical and mechanical parameters are those with a minimum Bi$_2$O$_3$ content of 40%. This composition can be considered as optimal out of those that were proposed as it shows the maximum strength having sufficiently high linear coefficient of attenuation.

### 4. Summary

After carrying out the research, it can be concluded that in the process of synthesis of ceramic radiation-protective composites with heavy filler (Bi2O3), a chamotte binder should be used. In general, samples on its basis, in comparison with the samples based on alumina binder, have the best physical and mechanical, as well as radiation-protective parameters. It should be noted that the most effective composite is the composition of 60% bismuth oxide and 40% chamotte ACB. This composition is characterized by density of about 4600 kg/m$^3$ and bending strength up to 15 MPa. It satisfies the conditions for the development of new radiation-protective materials with high structural properties.

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