Low-Temperature Synthesis of Porous Materials from Mortar Sands

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It is established that the eliminations of construction sand with the content of SiO2 about 70 wt.% and particle size less than 60 μm are suitable for the production of a foam-glass–crystal material on the basis of the low-temperature frit, which was synthesized at the temperature of 900°C. The obtained foam-glass–crystal material exceeds foam-glass (by 3.0 times) and clayite (by 1.5 times) by strength and is characterized by the low value of water absorption (0.1%).

INTRODUCTION

The production of lightweight and efficient building materials, in particular of lightweight granulated materials, is a constant and modern subject of scientific investigations. One of the high-efficiency heat-insulating materials that meet the requirements of environmental safety is foam-glass. Raw material for production of foamed glass can serve various types of waste. Processing of industrial waste is an important topic not only from the point of view of reducing dangerous environmental pollution but also from the point of view of their potential beneficial use as an alternative source of raw materials. Therefore, as initial raw materials for producing heat-insulating materials, the preference is given to technogenic and substandard raw materials. The most frequently used lightweight aggregates are made from expanded clay, shale, perlite, vermiculite and different kinds of sintered waste. The publications Refs. 1 and 2 suggest the use of building wastes in the production of aggregates for concrete. The results of studies in which concrete aggregates were obtained from ash waste are presented in Refs. 3–9. The authors of Ref. 10 suggest the possibility of using sludge from a decorative quartz industry hot bituminous mixtures.

When the temperatures necessary to reach the pyroplastic state of the melt and active formation of a gas phase coincide, the mix foams and stabilizes with the subsequent decrease of temperature. The high dispersity of sand sludge allows the expectation of the possibility of a low-temperature production of the frit. The results of preliminary studies are encouraging, since they confirm the possibility of using this type of raw material for the production of lightweight granulated material, that can be used as an aggregate for concrete and as a heat insulation filler.

The aim of this study is to investigate the possibility of the preparation of foam material similar to foam-glass on the basis of frit synthesized from quartz sands screenings. Industrial technology of foam-glass is based on the use of glass cullet or frit with the composition of sheet and container glasses. The high dispersiveness of quartz sand wastes allows one to suppose the possibility of low-temperature synthesis of frit. Therefore, one of the assigned tasks of this research was the development of blend compositions suitable to obtain frits at temperatures not exceeding 900°C, excluding the traditional glass-melting process.

RAW MATERIALS

When one selects the chemical composition of low-temperature frit, it is necessary to meet the following requirements. The first condition which defines the component composition of a blend is provision sufficient quantity of glass former (60–75 mass%) and the alkali metal oxides (13–22%). It is also necessary to take into account that foaming occurs at a viscosity of 105–107 Pa s, and therefore it is necessary to use the glasses which reach such a viscosity at temperatures of 750–900°C. It is possible to estimate the preliminary viscosity properties of the glass by its composition using a viscosity modulus, the value of which can be in the range 1.6–1.8.11 The second selection condition is the formation of not less than 70% of
melting at a temperature not exceeding 900°C, which has been shown by the results of previous experiments. The third condition is the hydrolytic stability of glass (not below the 3rd class) and the presence of an active oxidizing component in sufficient quantity to carry out foaming reactions, for example, the presence of SO₃ in quantities not less than 0.2%. One of the main requirements for all blend components are their dispersiveness; particle size must not exceed 100 μm.

Glass composition corresponding to the abovementioned requirements, selected by the state diagram of Na₂O-CaO-SiO₂, has the following content of components: Na₂O, 14 mass%; CaO, 13 mass%; and SiO₂, 73 mass%. The composition of sand under investigation according to the results of chemical analysis is presented by a relatively low quantity of the main glass-forming oxide (SiO₂) and a sufficiently high quantity of the alkali metal oxides, aluminas, and iron oxides (Table I). According to the data of x-ray phase analysis, sand contains, together with quartz and illite, tiff, plagioclase, and feldspar in about equal proportions. The high dispersiveness of sand is confirmed by the results of screen analysis according which 50% of sand is presented by a fraction of particles less than 60 μm which meet the necessary requirements.

As a foaming agent in the production of lightweight aggregate, we used carbon black type 220 (ASTM D1765). This carbon black represents technically highly active carbon with a high dispersity and good structural properties, obtained from thermal-oxidative decomposition of liquid hydrocarbon raw materials. The specific surface of carbon black is of the order of 1.14 × 10⁵ m²/kg.

RESULTS AND DISCUSSION

To obtain frit of the selected composition we accordingly prepared a blend containing the sand under investigation and afluxing addition in the form of calcined soda in quantities of 80 and 20 mass%. The calculated composition of the glass had following oxide content (in mass%): SiO₂, 66.3; Na₂O, 13.4; CaO, 6.5; Al₂O₃, 6.8; Fe₂O₃, 3.7; K₂O, 1.8; MgO, 0.8; MnO, 0.1; and TiO₂, 0.5. The value of the viscosity module, calculated according to the formula, was 1.8, which falls within the recommended interval.

\[ M_B = \frac{(M_{SiO_2} + 2M_{Al_2O_3})}{(2M_{Fe_2O_3} + M_{CaO} + M_{MgO}) + 2M_{K_2O} + 2M_{Na_2O})} \]  

where \( M_B \) is the viscosity module, and \( M_Rbo \) is the quantity of the corresponding oxides in mass%.

Preparation of foam crystalline material has been carried out by the two-stage technology proposed in Ref. 11. The first stage includes synthesis of frit consisting of glass (not less than 70%) and a residual crystalline phase (not more than 30%), which is provided by the chemical composition of a blend. The second stage includes preparation of a foam-forming blend from frit powder to obtain the finished foam material.

The results of the differential-thermal analysis (DTA) show that, at the heating of the blend of the selected composition up to 1000°C, endo-effects were observed corresponding to the removal of hygroscopic and crystallization water of the blend (8°C and 245°C), polymorphic transitions of quartz (571°C), and the melting of forming double salts and eutectics (733°C and 797°C). The main mass losses occur in the temperature range of 500–800°C, corresponding to silication reactions. At the temperature of 800°C, the thermogravimetric curve is horizontal, which shows the total binding of sodium carbonate and the completion of the silication reaction according to Eq. 2.

\[ nSiO_2 + Na_2CO_3 \rightarrow Na_2O \cdot nSiO_2 + CO_2 \]  

It is stated by the data of thermal analysis that thermal treatment of the blend under investigation at temperatures of 800–900°C secures total

<table>
<thead>
<tr>
<th>Table I. Characteristics of the surveyed sands</th>
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<tbody>
<tr>
<td>Chemical composition, content of oxides, mass%</td>
</tr>
<tr>
<td>SiO₂</td>
</tr>
<tr>
<td>69.67</td>
</tr>
<tr>
<td>Mineralogical structure, mass%</td>
</tr>
<tr>
<td>Quartz</td>
</tr>
<tr>
<td>50</td>
</tr>
<tr>
<td>Granulometric structure, mass%</td>
</tr>
<tr>
<td>&lt;2 μm</td>
</tr>
<tr>
<td>6.8</td>
</tr>
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</table>
completion of the silication processes. To confirm this supposition, we carried out x-ray phase analysis of the frit prepared at temperatures of 800–900°C. It is seen from the obtained results (Fig. 1), that an amorphous halo indicating the presence of a glass phase and reflection maxima responsible for the crystalline phase were observed on all x-ray images. The results of quantitative x-ray phase analysis show that the frit consists of a glass phase in the range from 66% to 72%, while the remaining crystalline phase presented the residual quartz (d = 3.342 nm; 2θ = 26.7°), feldspar, and silicate, Na$_2$OÆCaOÆ3SiO$_2$.

Differential-thermal analysis of the frit showed no exothermic effects either at heating up to 1000°C, or at cooling to 200°C, which confirms the absence of crystallization processes. DTA of foam-forming mixtures prepared on the basis of frits prepared at various temperatures (from 800°C to 900°C) with the addition of soot (0.5%) showed the presence of exo- and endo-effects corresponding to the processes of oxidation of gas-forming agents and the melting of the glassy phase. An increase of frit preparation temperature biases the exo-effect temperature in the higher field, so for an admixture from the frit (800°C), the effect occurs at 463.7°C, while for the frit (900°C), the value is 498.6°C, which is conditioned by differences of the phase composition of the frits (Fig. 2). According to quantitative x-ray analysis, when the sintering temperature of the frit increased through 800°C, 825°C, 850°C to 900°C, the content of the crystalline phase decreased through 34%, 32%, 29% to 27%.

Bias of oxidation temperature in the higher field is favorable for the foaming process since the probability of early burning out of the gas-forming agent decreases. Total mass losses for frit (800°C) is 5 times more in comparison with the foam-forming admixture prepared from frit (900°C). In addition, thermograms of admixtures of low-temperature frits (800°C and 825°C) show some endo-effects, while thermograms of frits with temperatures of 850°C and 900°C show one endo-effect which points to a large inhomogeneity of low-temperature frits and the presence of some phases differing with melting temperature.

Foaming of pellets (d = 10 mm), prepared from foam-forming admixtures (specific surface 6000 cm$^2$/g), has been performed in the temperature range of 900–975°C (with steps of 25°C) with exposure at maximal temperature in 20 min. It has been stated that foam material with a relatively uniform fine porous structure is obtained at a foaming temperature of 950°C. The macrostructure of the samples obtained at 900–925°C has a dense crust with a thickness more than 3 mm, and the samples obtained at 975°C are deformed in consequence of vitrification and show a random structure with the presence of large pores with sizes more than 5 mm. It has been shown by x-ray analysis data that if the foaming temperature increases from 900°C to 975°C, the quantity of the crystalline phase in the finished foam material decreases from 23% to 8%.

There is a connection between the foam material macrostructure and the temperature of the preparation of the frit, from which the foam-forming
admixture is obtained. Photographs of the macrostructures of samples prepared when foaming at 950°C from frit synthesized at 800°C, 825°C, 850°C, and 900°C are presented on Fig. 3. The foam material obtained from the frit at 900°C is the most optimal from the point of view of pore size and the uniformity of their distribution.

Values of the main physical–mechanical properties of samples prepared from frit synthesized at various temperatures (Fig. 4) allow one to note the following laws in ranges of properties. If the temperature of frit synthesis increases from 800°C to 900°C, the density of pellets decreased on an average by two times, the strength by four times, and water absorption by 13 times.

Comparing the characteristics of the samples prepared on the basis of mortar sand screenings with other heat insulating materials shows that the material occupies an intermediate position between foam-glass and expanded clay (Table II). The value of the strength coefficient of the foam crystalline material, which is the ratio of strength to density, exceeds that of foam-glass and expanded clay. The material differs with its high strength, low water absorption and relatively low heat conduction coefficient that allows the recommending of it as a heat-insulating and construction material.

**CONCLUSION**

The main objective of the investigations described in this report was to analyze the feasibility of using waste from the sand industry in lightweight aggregate production. After analyzing the properties of the lightweight aggregates produced in the course of the present studies, the following conclusions can be drawn:

1. Screenings of mortar sands with a content of SiO$_2$ of about 70 mass% and the size of particles less than 60 μm are suitable for the preparation of a foam crystalline material from low-temperature frit synthesized at a temperature of 900°C. The glass phase of the frit is not crystallized in the range of foaming temperature, and has sufficient viscosity and temperature of its pyroplastic state coinciding with the temperature of oxidation of the gas-forming agent.

2. If the synthesis temperature increases from 800°C to 900°C, the quantity of the glass phase in the frit increases from 66% to 73%. The crystalline phase of the frit is represented by residual quartz and feldspar as well as by the appearance of a new phase in the form of silicate Na$_2$O-CaO-3SiO$_2$. The content of the crystalline
phase in the finished foam material decreases from 23% to 8% in dependence on the increase of the foaming temperature from 900°C to 975°C.

3. Frit synthesized at 900°C and a foaming temperature 950°C is optimal for the preparation of foam material with improved properties. The

![Fig. 3. Macrostructure of the foam material received from frit, synthesized at: (a) 800°C, (b) 825°C, (c) 850°C, and (d) 900°C.](image)

![Fig. 4. Properties of the foam material received from frit, synthesized at: 800°C, 825°C, 850°C, and 900°C.](image)

<table>
<thead>
<tr>
<th>The granulated material</th>
<th>Bulk density (kg/m³)</th>
<th>Strength at compression in the cylinder (MPa)</th>
<th>Heat conduction coefficient (W/m K)</th>
<th>Water absorption volume (mass%)</th>
<th>Strength coefficient</th>
</tr>
</thead>
<tbody>
<tr>
<td>Foam-glass–crystal</td>
<td>300–450</td>
<td>4.5–5.5</td>
<td>0.09–0.10</td>
<td>0.5–0.7</td>
<td>1.3</td>
</tr>
<tr>
<td>Foam-glass from a cullet</td>
<td>100–250</td>
<td>1–1.5</td>
<td>0.06–0.08</td>
<td>5</td>
<td>0.7</td>
</tr>
<tr>
<td>Clayite</td>
<td>300–800</td>
<td>0.6–4.5</td>
<td>0.10–0.16</td>
<td>8–20</td>
<td>0.5</td>
</tr>
</tbody>
</table>
prepared foam crystalline material has a higher strength up to 5.5 MPa at a bulk density of 450 kg/m$^3$ and low water absorption not exceeding 1 mass%.

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**REFERENCES**