

# Single-stage technology for granulated foam glass production based on the composition of tripoli and technological microsilica

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**Abstract.** The possibility of foam glass production by means of one-stage technology based on the natural tripoli and technogenic silica is determined. 45 % sodium hydroxide solution is used for the synthesis of foam glass. The addition of microsilica as an extra component in an amount of 10 to 50 wt. % in batch component increases the strength of the obtained material to 4 MPa and reduces foaming temperature from 870 to 830 °C. The increased mechanical strength of the material is due to the residual quartz dissolution and cristobalite crystallization. The samples obtained from batch with 30 wt. % microsilica have maximum strength.

## 1. Introduction

Foam glass is considered to be one of the most environmentally friendly, durable and efficient thermal insulators. Its effectiveness is due to a unique combination of properties, such as low water absorption, frost resistance, resistance to aggressive media, relatively high strength. Currently, the field of foam glass application has been rapidly developing. Granulated foam glass is used for fill insulation, and as an effective filler of various composites, for example, in the production of warm plasters as light concrete and cement slurries.

Glass cullet or finely crashed glass melted in a glass-melting tank is a raw material for foam glass production. In view of the limited resources of waste glass the raw material base development for foam glass production is relevant. Granulate melting is associated with high energy and capital intensity of glassmaking. European researches are mainly focused on the use of various types of glass cullet, for example, glass of electron - ray tubes and computer screens, etc. [1-7]. Some researchers consider not only glass cullet but also various types of natural and technogenic materials as raw material [8-11].

The objective of this study is to obtain foam glass material by means of one-stage technology based on natural silica component in the form of tripoli at a foaming temperature not exceeding 850 °C.

## 2. Materials and Experimental method

Tripoli and microsilica were chosen as objects to study siliceous materials of natural and technogenic origin. Chemical and granulometric composition of silica raw materials are shown in Table 1. The chemical composition of tripoli differed from the standard silica sand used in glass production, as it had a reduced content of glass-forming oxide and an increased content of  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{Fe}_2\text{O}_3$ . Silica fume, which is a byproduct of metallurgical production in melting of ferrosilicon and its alloys, corresponds to the sand grade used for the production of foam glass, cans and semiwhite glass bottles.

Along with chemical composition the high dispersion with average particle size of less than 100 microns was the main requirement to the raw material. Initial tripoli contains coarse particles in the form of clay aggregated in an amount of 3 - 7 wt. %. Therefore, tripoli was previously dried and then milled in a ball mill. The results of the grain-size composition of the prepared tripoli were presented in the following table. Microsilica was mainly presented in a form of particles of less than 0.5 microns without additional preparing. According to the results of X-ray analysis it was determined that microsilica refers to X-ray amorphous material. Crystal silicon ( $d = 0,313$ ;  $0,192$  nm), carborundum ( $d = 0,251$ ;  $0,217$ ;  $0,154$  nm), and graphite ( $d = 0,335$ ) practically without a crystal phase were presented in the results. Small amount of amorphous halo (ring) reflection maxima were shown on the X-ray photograph of tripoli, which



corresponded to different modifications of silica, ie, material was amorphous crystalline. A few of diffraction maxima specific for albite were also presented.

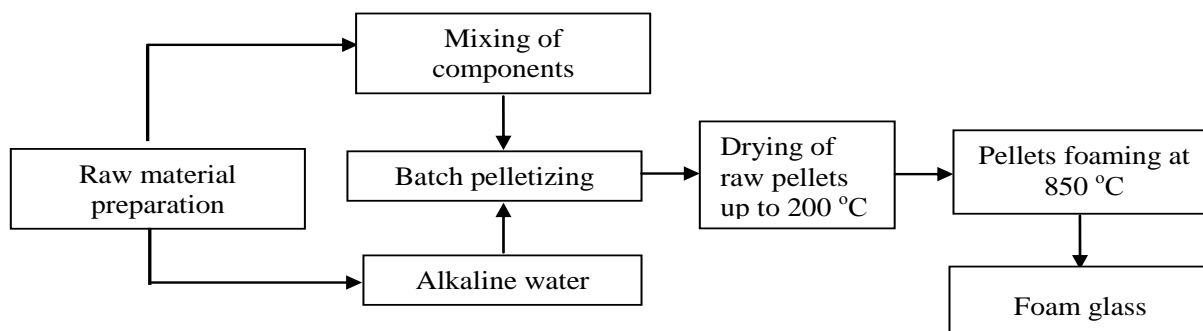
**Table 1.** Chemical and grain-size composition of silica components

<i>Chemical composition</i>						
Material name	Content of oxides, wt. %					
	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	CaO	MgO	Δm
tripoli	76,16	7,52	4,10	1,05	0,75	8,73
microsilica	91,70	0,50	0,40	1,20	-	2,00
<i>Grain-size composition</i>						
Size of particles, μm	Percentage of fractions, %					
	> 0,5	0,5 – 1	1 – 5	5 – 10	10 – 50	50 – 100
tripoli	17,0	30,8	26,4	6,6	18,4	0,8
microsilica	73,0	8,0	1,0	1,0	1,0	16,0

In the study, granular sodium hydroxide, a white crystalline powder with the composition of NaOH - 99.5 wt. %; Na<sub>2</sub>CO<sub>3</sub> - 0.5 wt. %; NaCl, Na<sub>2</sub>SO<sub>4</sub>, Fe<sub>2</sub>O<sub>3</sub> – traces, was used as an alkali component. The study of phase composition of the raw and obtained materials was carried out using a X-ray diffractometer DRON-3M in copper radiation. Quantitative X-ray analysis was performed using the Match program. Macro- and microstructure analysis of porous samples was performed on a scanning electron microscope (JCM – 6000).

### 3. Experimental results

Preparation of foam glass was carried out according to the scheme shown in Figure 1. First, the source raw material was dried, then sieved, and, if necessary, milled. The prepared materials were mixed in a bowl of the mixer-granulator, then granulated with binding substance in an amount of 30 wt. 2 %. The alkaline component adding in a form of solution did not affect the batch homogeneity obtained in the stage of component mixing. At the same time it resulted in intimate contact of the mixture components through the alkaline solution, which led to more complete silication reactions in each pellet. The resulting raw pellets were dried at a temperature not exceeding 200 °C to final moisture content of less than 1.5 %. Foaming was carried out in an electric-tube furnace at different temperatures.



**Figure 1.** One-stage production of foam glass

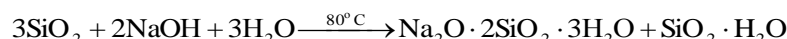
Use of only microsilica as a silica component showed that microsilica was completely dissolved in alkali due to structural features and high dispersion. In addition, the pellets foaming occurred at a temperature of 200 °C. However, the foam density exceeded 800 kg/m<sup>3</sup>, which didn't comply with the requirements of thermal insulation materials. Therefore, microsilica can be used only as an additional component. Its amount in the batch was varied from 10 to 50 % by weight in 10% increments. The batch compositions were presented in Table 2.

**Table 2.** Chemical and compositional analysis of batch

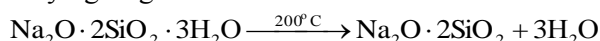
№ compound	Compositional analysis of batch, wt. %			Oxide composition of glass phase, wt. %					
	tripoli	microsilica	45 % NaOH	SiO <sub>2</sub>	Na <sub>2</sub> O	CaO	MgO	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>

1.1	100	0	23	73,67	13,36	1,02	0,73	7,26	3,96
1.2	90	10	23	74,86	13,31	1,03	0,65	6,56	3,59
1.3	80	20	23	76,05	13,25	1,03	0,57	5,88	3,22
1.4	70	30	23	77,24	13,19	1,04	0,5	5,17	2,86
1.5	60	40	23	78,41	13,14	1,05	0,43	4,48	2,49
1.6	50	50	23	79,57	13,09	1,06	0,36	3,79	2,13

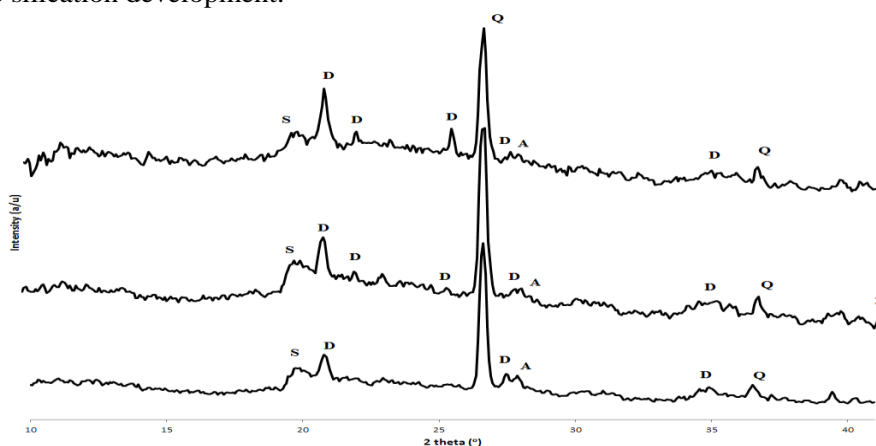
Self-heating of the moisturized batch up to 80 °C was observed in the stage of preparation and granulation of the batch with formation of sodium hydrosilicate and silica gel in a colloidal state. Interaction of the alkaline solution with the amorphous silica component of the batch occurred due to the following reaction:



Silication reactions occurred due to dehydration of crystalline sodium silicate upon heating the granulated material up to 200 °C at the drying stage:



According to X-ray analysis it was determined that reflection peaks corresponding to different sodium silicate were presented on the X-ray photographs of the dried granules (up to 200 °C) of all compositions (Fig. 2). Diffraction peak intensity increased with an increase of microsilica in the batch, which indicated more intensive silication development.

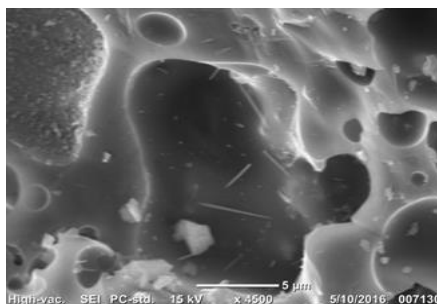


**Figure 1.** The XRD patterns of the dried pellets due to the content of microsilica in the batch of wt. %: 1 - 10; 2 - 20; 3 - 50 (Q - quartz, D - sodium disilicate; A - albite; S -  $\text{Na}_2\text{Si}_3\text{O}_7$ )

When the granulated material was heated up to 750 °C and above, a glass foaming process was observed at the foaming stage, and the obtained porous material had a vitrified surface. The process proceeded in stages. The first phase was obtaining of eutectic melt by ternary eutectic melting formed among  $\text{Na}_2\text{O} \cdot 2\text{SiO}_2$  and  $\text{SiO}_2$  and albite, at a temperature of 740 °C and the binary eutectic among  $\text{Na}_2\text{O} \cdot 2\text{SiO}_2$  and  $\text{SiO}_2$  at a temperature of 793°C. In the second stage, the residual silica was dissolved in the primary melt, then, the granules were cooled to form sodium- aluminosilicate glass at foaming temperatures (800-900 °C).

Quantitative X-ray analysis of the synthesized foam glass showed that the amount of the glass phase in the obtained product depended on the content in the starting batch of microsilica. The glass phase content increased from 74 % to 78 % with microsilica increasing from 10 to 20 %, respectively. The amount of microsilica in the starting material of 30 % resulted in a decrease up to 64 % of glass phase, and when the content of microsilica was 40% and 50 %, the amount of the glass phase increased up to 69 %. Different content of the glass phase in the final product was due to the glass crystallization processes occurring at the stage of pellets foaming. The X-ray photographs of all samples presented reflection peaks corresponding to quartz, and new reflection peaks corresponding to cristobalite appeared (Fig. 3). With an increase of silica in the batch the intensity peaks of cristobalite increased.

On the electronic micrographs of the samples obtained with the addition of microsilica, it can be seen a new crystal phase in foam glass. The picture showed needle-like crystals, which were absent in the samples without microsilica (Fig. 3). According to the X-ray diffraction analysis and the needle shaped crystals one can determine the cristobalite crystallization.



**Figure 3.** SEM micrograph of the foam glass obtained with addition of 50% microsilica

The granular material strength values specified by the standard techniques showed the following: with an increase of the content of microsilica in the batch up to 30 % the mechanical strength of the granules increased to 4 MPa, the strength reduced to 3.3 MPa with more content of microsilica, but the strength of the granulated foam glass without microsilica addition of 1.5 MPa became higher. The increased strength of the foam glass was due to crystallization processes that occurred at the foaming stage. At a density of 500 kg/m<sup>3</sup> the strength of foam glass was two times larger than the strength of the foam glass obtained by one-stage technology based on tripoli, and was 1.3 times larger than the strength of the foam glass obtained from the glass cullet.

#### 4. Conclusions

The use of highly fine natural amorphocrystalline tripoli and amorphous technogenic microsilica provides obtaining of foam glass material by one-stage technology with 45 % sodium hydroxide solution as an alkaline component at foaming temperatures of 830-850 °C. Addition of microsilica as a minor constituent in the batch in the amount of 10 to 50 wt. % increases the strength of the obtained material to 4 MPa and reduces the foaming temperature from 870 to 830 °C. The increased mechanical strength of the foam glass is due to dissolving of residual quartz and cristobalite crystallization. The samples obtained from the batch containing 30 wt. % of microsilica have the maximum strength of 2 MPa.

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