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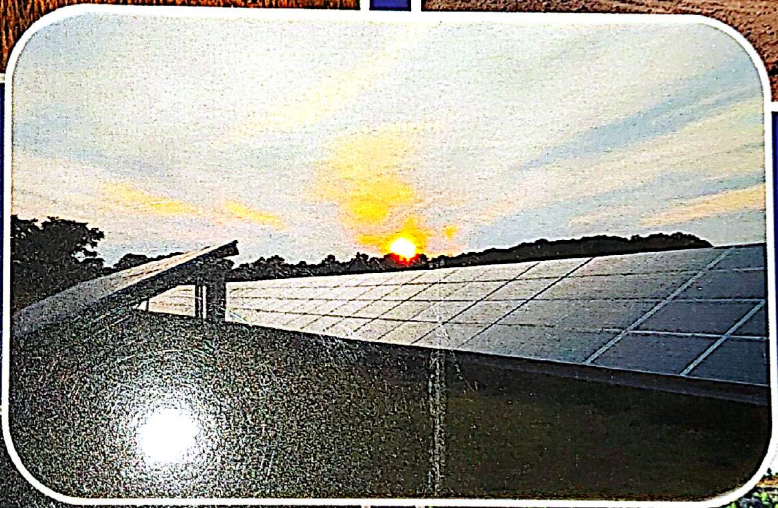

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RECYCLING OF GLASS WASTE INTO FOAM GLASS: A REVIEW

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Abstract Waste glass can be transformed into a foam by addition of suitable substance (foaming agent) which generated gaseous products by decomposition or reaction at temperatures above its softening temperature (corresponding to a viscosity of $10^{6.6}$ Pas). This paper provides a concise overview of the latest research in the field of improving the glass foam production process. Some of the possibilities for reducing the environmental impact of glass foam production are also presented. Some of the new production techniques, new foaming agents as well as the possibility of including other solid waste in the composition of glass foams are presented. At the end, the glass foam produced in the Laboratory for glass and ceramics, Institute for Technology of Nuclear and other Mineral Raw Materials, was shown.

Keywords: foam glass, recycling, waste glass

1. INTRODUCTION

The generation of waste has been growing rapidly with the increase in population. A massive volume of the waste is disposed in landfills. This is creating significant environmental problems such as contamination of soil, water and air and also impacting human health. It is reported that the world's solid waste is responsible for 5% of the total global carbon dioxide emission, also a more rational solid waste management would reduce the total global CO_2 emission by up to 15%. [1]

Approximately, 130 million tons of glass waste is generated annually in the world, about 21% of glass waste is recycled. Glass is a 100% recyclable material. No matter how many times you recycle it, glass never loses its quality. Glass is made from readily available materials, such as sand, soda ash, limestone, and "cullet" recycled glass. Over a ton of natural resources are saved for every ton of glass recycled. One ton of carbon dioxide is reduced for every six tons of recycled container glass used in the manufacturing process [2].

Glass containers for food and beverages are 100% recyclable, but not with other types of glass. Other kinds of glass, like windows, ovenware, Pyrex, crystal, etc. are manufactured through a different process. If these materials are introduced into the glass container manufacturing process, they can cause production problems and defective containers. Whatever, it is not possible to recycle any glass and get the desired product. But, most glass can be converted into foam glass by the foaming process. Any glass can be transformed into a foam by addition of suitable substance (foaming agent) which generated gaseous products by decomposition or reaction at temperatures above its softening temperature (corresponding to a viscosity of $10^{6.6}$ Pas). The foaming agents that are most often used in the process of obtaining glass foam are: carbonates, sulfates, nitrides, carbides and other carbon sources as carbon black, graphite, carbon ash, coal ash [2–4].

The process of obtaining foam glass begins with the mixing of glass, foaming agent and other additives. After that, the mixture is placed in molds. The production of foams finishes in the sintering and cutting process. The starting glass must be ground and sieved to grain size of less than 0.4 mm. If the grains are larger, the foaming process will be disabled. Also, very important that the foaming agent be finely ground. The mixture is heated above the glass softening temperature, foaming agent particles are wrapped by the softened glass until the reaction or decomposition temperature of the foaming agent is reached. After the foaming agent reacts or decomposes, it releases gases that form gas bubbles in the softened glass. As temperature increases, glass viscosity decreases continuously and as a result, the surface tension of glass decreases. The reduction in surface tension of glass decreases the pressure over the gas bubbles, which favors their growth, and increases expansion and pore coalescence. At the end of the process, after cooling, a material that looks like a sponge is formed, that is foam glass [5–19].

Foam glasses are unique materials, lightweight, non-flammable, chemically inert, non-toxic, etc. They are easy to transport and cut, and are resistant to the action of insects, rats and bacteria. Due to their specific properties, foam glass has found application as thermal and sound insulation materials in construction industry.

This paper presents a brief overview of the latest knowledge about foam glasses as well as various improvements in the production process. Also, foam glass obtained at the Institute for Technology of Nuclear and Other Mineral Raw Materials, Belgrade, is shown. To obtain foam glass, beer bottles were used as raw material, and waste calcium carbonate from the sugar factory was used as a foaming agent.

2. TECHNICAL REQUIREMENTS

The latest research on foam glasses obtained from glass waste, pays the most attention to foaming agents. Research is focused on finding an eco-frendly foaming agent. The most common foaming agents currently used in industries are the emission of SO_3, CO and CO_2 into the environment during sintering. These gases are responsible for greenhouse effect and acidification, in contrast to the benefits generated by the production of foam glass from glass residues. Therefore, NaOH was introduced as an alternative to traditional foaming agents [5] to produce low density environmentally-friendly foams. As a result, NaOH began to be used as a foaming agent in other studies [6]. The authors of paper [7] propose a new chemical route to obtain foam glasses from soda-lime waste glass (float glass) using sodium hydroxide and borax as foaming and flow agents, respectively. This research aims to investigate the chemical reactions of soda-lime waste glass with NaOH and borax during the sintering process. The research results showed that during the initial-heating stage, the mixture was dehydrated. Between 318 °C to 587 °C, the sample expanded until its glass transition temperature. The sample shrank due a crystallization process generating $Na_2CaSiO_6 \cdot 2H_2O$ and $Na_2CaSi_3O_8$. Increasing the temperature (at 686 °C), the sample expanded more than 75%, being the water (from dehydration) the unique expansion agent. Finally, a new crystalline phase was formed. This foaming process provided a foam glass with 86% of porosity, low permeability and bulk density of 0.3 g/cm^3.

A group of authors from Brazil used waste glass bottles as a raw material for obtaining foam glasses and as an foaming agent, eggshells [8]. They made several mixtures with different mass contents of eggshells (1, 3, 6, 9, 15 and 30 wt%), which they heated at 900 °C for 30 min. By comparing the physical characteristics of the newly obtained glass foams with the characteristics of glass foams in which pure calcium carbonate was used as a foaming agent, the authors concluded that eggshells can be successfully used as foaming agents. Thermal conductivity of the glass foams is in the range from 0.055 to 0.177 W/mK and compressive strength is 0.15 to 1.5 MPa. The produced foams have glass structures with partially open porosity (interconnected pores) in the compositions up to 15 wt% egg shell.

Wang et al. (2016) [9] examined the effect of the addition of KNO_3 on the bulk density, crystalline phase, and microstructure of glass foams. They prepared glass foams by using waste glass and SiC powder as a foaming agent. The foaming ability of SiC was directly dependent on the content of the oxygen around, so the addition of KNO_3 could notably promote the foaming process of glass foam. The decomposition products of KNO_3 are oxygen and K_2O, both improved the performance of glass foam. Glass foams were obtained by sintering process at different temperatures, 900, 925 and 950 °C. KNO_3 improve the foaming process which has a great influence on the microstructure and physical properties of glass foams. Good correlation between physical properties and microstructure (size, distribution and interconnectivity of pores) was observed with 1 wt% KNO_3 addition. The emission of gaseous pollutants (e.g. CO_2) from the most used foaming agents can be harmful to the environment.

Several groups of authors investigated the possibilities of using animal remains in the process of obtaining foam glass. A group of American and Thai authors [10] used waste amber glass as raw material and dried porcine bones as a foaming agent to obtain foam glass. Amber glass has been used as containers or bottles for protection from the deteriorating effect of light, such as beverages, pharmaceutical products and chemicals. Most of the amber glass is disposed of in landfills and only a small part is recycled. Fast sintering at a temperature of 850 °C for 600 s, resulted in foam glass with a high strength of 16.71 to 29.69 MPa. The porosity increases with increasing the amount of foam agent added and flexural strength shows the opposite trend. In paper [11] the structural, physical, and mechanical characteristics of glass-ceramic foams, obtained from waste soda-lime-silica glass and ark clam shell were used as a foaming agent, have been observed and characterized. Ark clam shell (ACS) consists mainly of $CaCO_3$ at 95–99 wt% with other minor oxides such as K, Si, Fe, Sr. Foam glass-ceramics with varied ark clam shell compositions (1–9 wt%) were utilized as a foaming agent and sintered at 800 °C for 60 min using soda-lime-silica cullet glass waste. Cristobalite and wollastonite crystals have been discovered in XRD analysis, with cristobalite crystals decreasing and wollastonite crystals increasing as the ACS composition increases from 1 to 9 wt%. As the ACS composition grew from 3 to 9 wt%, the linear expansion reduced because the gas partial pressure that had accumulated in the glass matrix had ruptured the pore walls, allowing pores to be incorporated. At 3 wt% ACS content, maximum total porosity (59.05%) and minimum apparent density of 0.699 g/cm^3 were achieved successfully. Furthermore, the lowest compressive strength value (0.33 MPa) was reached at 3 wt% ACS, which also had the highest porosity value (59.05%).

Waste glass is very often a mixture of different types of glass. The different chemical composition of these mixtures significantly affects the process of obtaining glass foam. The high production costs of foamed glass are, among other factors, related to the need for a precise control of the glass´s composition, as compositional fluctuations are directly reflected in product quality. During the heating of the mixture of waste glass and calcium carbonate (the most widely used foaming agent), the decomposition of calcium carbonate and the formation of CO occur. Also, calcium oxide is formed, which can lead to glass crystallization in some mixtures (depending on the composition of glass). Furthermore, the foaming with carbonates is difficult to control, as a rapid release of the gases results in rupturing of the pore walls and thus, in an open structure. Köning et al. [12] showed that it is possible to fabricate foams glass from CRT panel glass and C as a foaming agent and Fe_2O_3 as a oxidating agent, additionally, calcium phosphate was added to the powder mixtures to suppress the crystallization tendency of the soda lime silicate compositions. Also, they changed the composition of the mixture, i.e. they added container glass and flat glass to the mixture and reduced the proportion of CRT glass. This glass foams have the same density or pore structure as glass foams with only CRT panel glass, by adding up to 50 wt% container cullet or 70 wt% flat glass to the mixture. The foam samples prepared from the CRT panel glass are amorphous. In the foamed samples with a low content of panel glass, crystals form, resulting in an increased open porosity, density and inhomogeneous pore structure. The sample prepared from flat glass has a relatively homogeneous porosity while the sample prepared from container glass does not. In this samples the pore size is smaller than in the sample prepared from CRT panel glass. The thermal conductivity decreases with a decrease in the density of the samples as well as with an increasing content of CRT panel glass. Same authors showed that the use of foaming agent/oxidizing agent couples and crystallization inhibitors represents a great potential for decreasing the effect of the glass composition on the formation and properties of the foam glass, thus stabilizing the production of foamed glass. Also, König [13] used the TG-MS analysis, to show the impact of different oxidation states of manganese on the foaming process. For experiments he used, glass powder from obsolete CRT panels mixed with different foaming additives: activated charcoal, carbon black, MnO_2, Mn_2O_3 and Mn_3O_4. MnO_2 only partially contributes to the foaming reaction and has a mainly negative impact on the foaming process as it shifts the foaming to a higher temperature, increases the mass loss rate leading to open pores, and burns out the carbonaceous foaming agent before the sintering of glass particles. Mn_2O_3 should be used as the foaming agent in an oxidizing atmosphere, and Mn_3O_4 as the oxidizing agent in an oxygen-free atmosphere. Based on Koning's research, Qu et al [14] prepared a series of novel glass foam with microwave absorption properties from soda lime silica waste glass, C and Fe_2O_3 by a sintering method. The glass foams were prepared with the sintering temperatures of 740, 750, 760, 770, 780, 790 and 800 °C. The glass foams with magnetic crystals were found to have excellent efficiency of microwave absorption. They showed that the sintering temperature significantly affects the physical and absorbing properties of glass foam.

The ferrimagnetic glass foam has the dual functions of thermal insulation and electromagnetic wave absorption, and realizes the high value-added recycling of waste glasses.

A large amount of research is also focused on examining the possibility of including other types of solid waste in mixtures for foam glass. Bai et al. [15] were studied the utilization of waste glass and fly ash as starting materials to prepare foam glass. Commercially available SiC powders with grain size 3.5 μm, was used as high-temperature foaming agents. The macro-structure of the newly obtained foam glass was typically closed foam structure. The density of foam glass is 267.2 kg/m^3, compressive strength is 0.9829 MPa and porosity is 81.55%.

During the processing of fluorite ore, large amounts of fluorite tailings are produced. It mainly consists of CaO, SiO_2, Al_2O_3 and CaF_2. The inappropriate disposal of fluorite tailings in landfills has already caused major problems in Mexico, South America and China because in the infiltration of residual fluorine in fluorite tailings into land and groundwater resources causes their acute contamination. Li et al. [16] produced lightweight glass-ceramic foams from fluorite tailings, waste glass, calcined kaolin, calcium phosphate and SiC as a foaming agent. The obtained glass-ceramic foams sintered at 1110 °C exhibited relatively homogeneous pore sizes and distributions, low densities and thermal conductivities, and high compressive strengths. The thermal conductivity, compressive strength, and bulk density of the prepared glass-ceramic foams were 0.06 W/mK, 1.2 MPa and 0.3 g/cm^3. The fluoride leaching concentration of the glass-ceramic foams reached 1.12 mg/L, which satisfied the requirement of fluorine emission concentration as per the ISO standard. In this way, fluorite tailings can be successfully recycled into an inert material that can be used in the construction industry. Another group of authors examined the possibility of using titanium-bearing blast furnace slag to obtain glass foam. Titanium-bearing blast furnace slag is a major waste produced by the iron and steel industry. In this study, foam glass using waste glass and titanium-bearing blast furnace slag with 8 wt%, 10 wt%, 12 wt% $Na_2B_4O_7{\cdot}5H_2O$ as flux agent and $Na_3PO_4{\cdot}12H_2O$ as foaming stabilizer are successfully prepared at 900 °C [17]. The experimental results showed that $Na_2B_4O_7{\cdot}5H_2O$ addition has a significant influence on the properties of the foam glass. The addition of 10 wt% $Na_2B_4O_7{\cdot}5H_2O$ is suitable for the preparing foam glass at temperature of 900 °C. The specimens with 10 wt% $Na_2B_4O_7{\cdot}5H_2O$ addition exhibits the excellent comprehensive properties such as low bulk density (0.52g/cm^3), high porosity (79.13%), low water absorption (11.14%) and super compressive strength (8.26 MPa).

Apart from the usual way of obtaining foam glass, it was shown that it is also possible to obtain foam glass using other methods. Using the replica method, it is also possible to obtain foam glass. This was shown by Pereira da Costa et al. [18] Using waste glass bottles and bentonite, they created an aqueous suspension in which they immersed polyurethane foams. After multi-stage heating, during which the polyurethane matrix is removed, glass-ceramic foams are obtained. The bentonite was used as a plasticizer. The glass-ceramic foams with high contents of a waste glass bottle had a lower porosity. The X-ray diffraction showed that sodium-lime-silicate was the main crystalline phase. Gel-casting technique is known as an eco-friendly method for making glass foams by vigorous mechanical foaming. The waste glass is alkaline activated (most often with NaOH). After intensive mixing, a foaming agent is added to the mixture. The mixture (activated media) is then dried at lower temperatures, at which the mixture stabilizes (stabilized media). Finally, the mixture is sintered at higher temperatures, foam glass is obtained [19] . Glass contains amorphous silica, with a suitable additive, hydration or geopolymerization can be accelerated during the alkali activation. Calcium silicate hydrates are formed by alkaline activation of mixtures rich in calcium carbonate. Mixtures with a large proportion of Al_2O_3 form aluminosilicate products during alkaline activation. Therefore, the foam stability is achieved without any other stabilizing agents [20].

2.1. Foam glass ITNMS

Discarded green beer bottle was used as glass raw material. Beer bottle was first crushed in agate mortar into cullet and then pulverized into fine particles. Obtained glass powder was sieved in order to obtain glass powder particles size under 48 μm. Sugar beet factory lime (SBFL) was obtained from sugar factory in Crvenka, Serbia and it was used as foaming agent. SBFL was dried in oven at 110 °C, then crushed in agate mortar and sieved to particles size under 48 μm. Glass powder was mixed SBFL in agate mortar and it was uniaxially pressed in laboratory hydraulic press at 20 MPa with addition of 5% moisture. The obtained pelets were placed in an electric furnace. Samples were heated at T=800 °C for t=30 min using heating rate of 10 °C/min. After the heat treatment the samples were removed from the furnace, and cooled in air. In Fig 1 is shown foam glass.

Our goal was to obtain glass foam with good physical and chemical characteristics while using only waste materials.

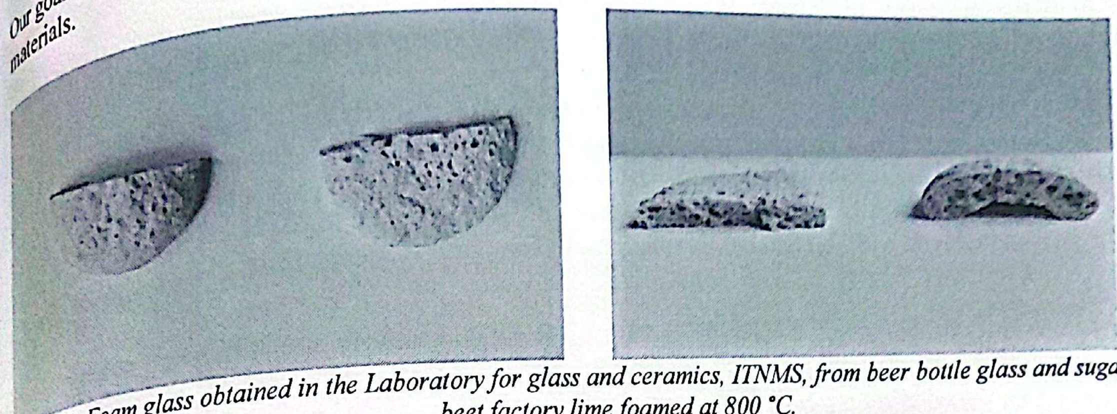

Figure 1. Foam glass obtained in the Laboratory for glass and ceramics, ITNMS, from beer bottle glass and sugar beet factory lime foamed at 800 °C.

3. CONCLUSION

Incorporation of glass wastes provides a systematic solution for glass-disposal waste management. Glass waste might be wielded for the fabrication of foam glass. By using waste glass instead of natural raw materials in the production of glass foam, it reduces the cost of production, the use of natural raw materials, and therefore protects the environment. It can be concluded that glass foam obtained from waste glass can be used as sound and heat insulation materials in construction devoid of any supplementary modification beyond cutting. This paper represents an overview of the latest literature related to the production of glass foams, new foaming agents and additives, as well as new procedures for the production of glass foams from waste glass.

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