# Metodă eficientă de încălzire cu microunde pentru fabricarea spumei de sticlă din deșeu de sticlă

## Effective microwave heating method for manufacturing glass foam from glass waste

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**Rezumat.** *In lucrare este prezentată o tehnică originală de fabricare a spumei de sticlă prin sinterizarea la temperatură înaltă a deșeului de sticlă datorită încălzirii cu microunde. Încălzirea materiei prime este realizată predominant direct și parțial indirect utilizând un tub ceramic din SiC cu o grosime optimă a peretelui de 2,5 mm. Această metodă contribuie la reducerea semnificativă a consumului specific de energie, a vitezei de incălzire și a duratei procesului comparativ cu tehnicile convenționale de încălzire. Caracteristicile spumei de sticlă sunt similare cu acelea ale spumelor fabricate conventional și pot fi utilizate ca înlocuitori ai materialelor de construcție existente.*

**Cuvinte cheie:** spumă de sticlă, microundă, încălzire predominant directă, agent de spumare, eficiență energetică

**Abstract.** An original technique for manufacturing glass foam by high temperature sintering of glass waste due to the microwave heating is presented in the paper. The heating of the raw material is performed predominantly direct and partially indirect by using a SiC ceramic tube with an optimal wall thickness of 2.5 mm. This method

contributes to the significant reduction of the specific energy consumption, the heating rate and the process duration compared to the conventional heating techniques. The characteristics of glass foam are similar to those of conventionally manufactured foams and can be used as replacements for existing building materials.

**Key words:** glass foam, microwave, predominantly direct heating, foaming agent, energy efficiency

## 1. Introducere

The manufacture of glass foam using recycled glass waste is a process that emerged and imposed in the last decades of the 20th century as an objective consequance of the need to reduce the amont of a waste with a very high annual generation rate, capable of causing major environmental damage. According to [1], in 2018 the container glass generation was 12.3 million tons in the United States representing 4.2 percent of all municipal solid waste generation. Also, in the same period, the global oil crisis [2] has produced a reorientation of the energy policy of the world's states by replacing traditional energy-intensive industrial products obtained by recycling waste incorporating much lower amounts of primary energy. This is also the case for building materials. It was found that the glass foam manufactured by sintering at 750-1150 ºC the recycled powder glass waste has remarkable physical, thermal, mechanical and morphological characteristics better compared to those of thermal insulation building materials and the energy consumption is significantly lower. The glass foams are lightweight, with very good thermal insulation properties, resistance to fire and moisture, non-toxic, physical and chemical stability, resistance to attack of rodents, insects, bacteria, acids, etc. [3, 4]. Several companies in the world (Misapor, Pittsburgh Corning, Geocell, Glapor, etc.) began to industrially produce different varieties of glass foam. The manufacturing method is based exclusively on conventional heating techniques [4].

Unlike the method of manufacturing glass foam used in the industrial production, the team of researchers from the Romanian company Daily Sourcing & Research has performed in recent years several experimental tests applying the unconventional method of microwave heating. Although known since the 1930s and recognized in the literature [5] as a fast, economical and „clean" heating technique, it has been industrially used only in drying or low temperature heating processes. It has been experimentally found at the beginning of the new millennium that several types of materials are susceptible to efficient microwave heating (organics, ceramics, metals, glass, polymers, etc.) [5]. Despite this, the industrial application of microwaves is still in different experimental stages.

An explanation of the low interest shown by industrial manufacturers for the use of microwaves as an energy source in the process of making glass foam could be the theory presented in 1997 in the paper [6], which considers that the commercial glass (soda-lime glass), the main raw material, is poorly microwave susceptible at room temperature due to its high content of transparent microwave materiala ($SiO_2$,

$Al_2O_3$). The electrical conductivity of the glass and implicitly, the dielectric properties of this material, increase rapidly with increasing the temperature, so that at about 500 ºC the microwave susceptibility reaches a high energy efficiency level [7-9]. The theory developed in the paper [6] was taken over in a market study [4], which concluded that the microwave application to the industrial glass foam furnaces would be interesting only for areas with temperatures above 500 ºC, which would mean equipping the furnace with a conventional heating system in the temperature areas below 500 ºC. Experimental results obtained by Daily Sourcing & Research [10] challenged the theory of the poor microwave susceptibility of soda-lime glass at low temperatures. Due to the presence in the composition of this glass of some inherent contaminants ($Fe_2O_3$, $Cr_2O_3$, etc.) even in low weight proportions that absorb more efficiently the electromagnetic radiation at room temperature, the microwave heating process can occur with normal efficiency starting from ambient temperature [9-11].

In order to obtain a maximum energy efficiency of the glass powder heating process, the direct microwave heating is necessary. In the case of glass (generally, silicate materials) this desideratum could not be experimentally achieved, because the direct contact of the microwave field with the material subjected to irradiation caused the severe destruction of its internal structure at the foaming temperature [12]. The adopted solution was the placement between the material and the microwave emission source of a high microwave susceptible ceramic screen in the form of a crucible or cylindrical tube. The distribution of the waves that penetrated the screen coming in direct contact with the material and those that were absorbed in the wall of the screen was adjusted by the thickness of its wall. Experimentally, it was found a thickness of 2.5 mm ensures a predominantly direct heating and a partially indirect heating suitable for a very high energy efficiency, without affecting the structure of the glass foam [13].

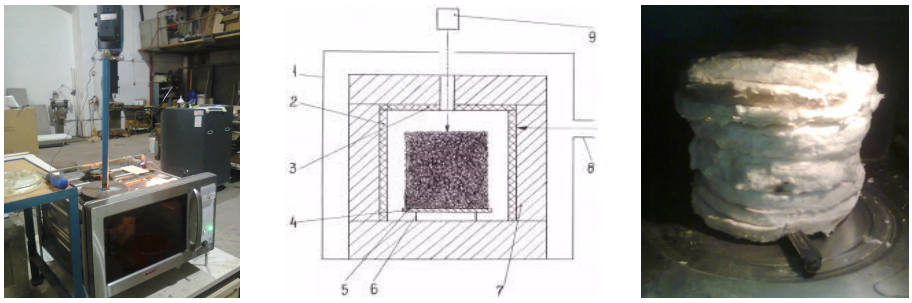
Further, experimental results obtained in the manufacturing process of glass foam under the condition of using the predominantly direct and partially indirect microwave heating method are presented.

## 2. Methods and materials

Commonly, the manufacture of glass foam from recycled glass waste consists of the sintering at high temperature (750-1150 ºC) of a pressed powder mixture containing glass waste, a foaming agent and, where appropriate, mineral additives to facilitate the foaming process. The foaming agent (solid or liquid) releases a gas or a gaseous compound at a temperature approximately similar to that of the softening point of the mixture containing the raw material. The gas bubbles penetrate the viscous mixture, but they cannot leave it being blocked in this space. Continuing he heating process, the material expands increasing its ititial volume. At the end of heating, by the slow cooling that follows, a porous structure specific to glass foam is formed [3, 4].

The experimental equipment on which the tests were performed at Daily Sourcing & Research was a 0.8 kW-microwave oven of the type used in the household for food preparation, adapted for operation at high temperature (up to 1200 ºC). The

powder mixture previously pressed into a metal mold and then released was freely deposited on a metal plate on a thermal insulating bed composed of several ceramic fiber matresses. A cylindrical ceramic tube made of a high microwave susceptible material (SiC and $Si_3N_4$ in the 80/20 weight ratio) with an outer diameter of 1200 mm, a height of 100 mm and a wall thickness of 2.5 mm was placed between the pressed material and the microwave emission source. The upper opening of the tube was covered with a ceramic lid of the same material provided with a 30 m hole for visualizing with a radiation pyrometer the surface temperature of the heated material. To avoid the heat loss of the heated material as well as of the ceramic tube and lid outside the system, the tube and lid were covered with ceramic fiber mattresses. It should be mentioned that the method of direct microwave heating is completely different compared to the conventional methods. The energy of the microwave field that comes in direct contact with the material is converted into heat, the heating being initiated in its core. The heat propagation takes place volumetrically throughout the material mass from the inside to the peripheral areas [14]. The heating process is fast. Also, the direct microwave heating is selective. Only the microwave susceptible material absorbs the electromagnetic waves. The other components of the oven (walls, vault, etc.) are not heated. This heating mode ensures a very good energy efficiency of the process [11]. The process temperature control was performed with a radiation pyrometer mounted above the oven at about 400 mm. To visualizing the material, the upper metal wall of the oven was provided with a 30 mm hole on the same vertical axis as the ceramic lid hole of the tube that includes the heated material. The experimental microwave equipment described above is shown in Figure 1.




a b c

Fig. 1. Experimental microwave equipment
a – overall image of the experimental equipment; b – constructive scheme of the equipment: 1 – microwave oven; 2 – ceramic tube; 3 – ceramic lid; 4 – metal plate; 5 – pressed powder material; 6 – metal support; 7 – ceramic fiber mattress; 8 – waveguide; 9 – radiation pyrometer; c – thermal insulation protection.

The foaming agent adopted for manufacturing glass foam in this experiment was calcium carbonate ($CaCO_3$). The decomposition reaction of $CaCO_3$ occurs at temperatures above 750 ºC [15, 16] with the formation of calcium oxide (CaO) and carbon dioxide ($CO_2$) according to reaction (1).

$$CaCO_3 = CaO + CO_2 \qquad (1)$$

CaO is treated in the molten glass, while $CO_2$ is released into the viscous mass of the glass.

In principle, the foaming process of a common commercial glass using $CaCO_3$ as a foaming agent takes place between 800-900 ºC being mainly influenced by the weight ratio, quality and fineness of the glass granulation and the weight ratio of the foaming agent [3].

As a fluxing agent, borax (sodium borate) was used in the starting mixture. Due to the boron content of over 13 wt.% [17], borax contributes to the increase of mechanical strength of glass foam.

The post-consumer container glass waste is the most widespread waste type in the world. In the current experiments, this type was used, being selected only the colorless glass waste. The chemical composition of colorless commercial glass according to the previous determinations [13] is shown in Table 1.

*Table 1*

**Chemical composition (wt.%) of colorless commercial glass**

| $SiO_2$ | $Al_2O_3$ | CaO | $Fe_2O_3$ | MgO | $Na_2O$ | $Cr_2O_3$ |
|---|---|---|---|---|---|---|
| 71.7 | 1.9 | 12.0 | 0.05 | 1.0 | 13.3 | 0.05 |

The glass waste was processed before using in experiments in Bilmetal Industries Company in Popesti Leordeni-Ilfov (Romania) including its selection by color, breaking, grinding in a ball mill and sieving at a granulation of less than 100 μm.

$CaCO_3$ purchased from the market at a fine grain size (below 40 μm) was used in experiments without other mechanical processing.

Borax was purchased from the market at a grain size below 400 μm, being ground in a laboratory electric device and sieved for a granulation of less than 130 μm.

The experimental variants composition was adopted taking into account the usual range of $CaCO_3$ proportions as a foaming agent in the glass foam manufacture (0.7-2.5 wt.%). The weight ratio of borax was adopted at 1.5 wt.% being kept constant for all variants. A water addition as a binder (9.0 wt.%) to facilitate the cold pressing of the powder mixture was also adopted. The composition of experimental variants is presented in Table 2.

*Table 2*

**Composition of experimental variants**

| Variant | Colorless glass waste (wt.%) | Calcium carbonate (wt.%) | Borax (wt.%) | Water addition (wt.%) |
|---|---|---|---|---|
| 1 | 96.0 | 2.5 | 1.5 | 9.0 |
| 2 | 96.6 | 1.9 | 1.5 | 9.0 |
| 3 | 97.4 | 1.1 | 1.5 | 9.0 |
| 4 | 97.8 | 0.7 | 1.5 | 9.0 |

Usual techniques have been applied in the process of characterizing experimentally manufactured glass foam samples. The gravimetri method [18] was used to measure the apparent density and the comparison method between the apparent density and the density in compact state (true density) of the same material [19] was used to determine the porosity. The use of a TA.XTplus Texture Analyzer allowed to measure the compressive strength and for determining the thermal conductivity the guerded-comparative-longitudinal heat flow (ASTM E1225-04) was applied. The water immersion method (ASTM D570) was used to determine the water absorption of the samples. Using an ASONA 100X Zoom Smartphone Digital Microscope the glass foam microstructures were examined.

## 3. Results and discussion

The functional parameters of the experimental process of manufacturing glass foam by predominantly direct microwave heating are shown in Table 3.

*Table 3*

**Main functional parameters of the experimental process**

| Variant | Dry raw material amount (g) | Process temperature (ºC) | Process time (min) | Average rate (ºC/min) | | Glass foam amount (g) | Specific energy consumption (kWh/kg) |
|---|---|---|---|---|---|---|---|
| | | | | Heating | Cooling | | |
| 1 | 500 | 826 | 29 | 27.8 | 5.0 | 486 | 0.62 |
| 2 | 500 | 824 | 29 | 27.7 | 5.3 | 485 | 0.62 |
| 3 | 500 | 825 | 31 | 26.0 | 5.1 | 487 | 0.66 |
| 4 | 500 | 827 | 33 | 24.5 | 5.3 | 487 | 0.71 |

According to the data in Table 3, the amount of dry raw material was kept constant at 500 g. The energy efficiency of the microwave heating process was remarkable, the heating rate having values between 24.5-27.8 ºC/min. Given that the temperature of the sintering/foaming process was between 824-827 ºC, the duration of the process was very low (29-33 min). The specific energy consumption decreased to very low values (up to 0.62 kWh/kg) corresponding to variants 1 and 2 with $CaCO_3$ weight ratio of 2.5 and 1.9 wt.%, respectively. The literature provides insufficient data on the specific consumption of industrial processes for the manufacture of glass foam by conventional heating techniques. The market study [4] mentions an average energy consumption in Misapor Company of 100 kWh/m$^3$, i.e. about 0.66-0.75 kWh/kg and the bibliographic source [20] indicates an average consumption of Energocell Company of 140 kWh/m$^3$, i.e. about 0.8 kWh/kg. Thus, the energy efficiency of the application of predominantly direct microwave heating technique was superior to conventional techniques, especially since it is known that the working conditions of an experimental process are disadvantageous in energy terms compared to an industrial scale process [5].

Pictures of the cross section of the four glass foam samples are shown in Figure 2.

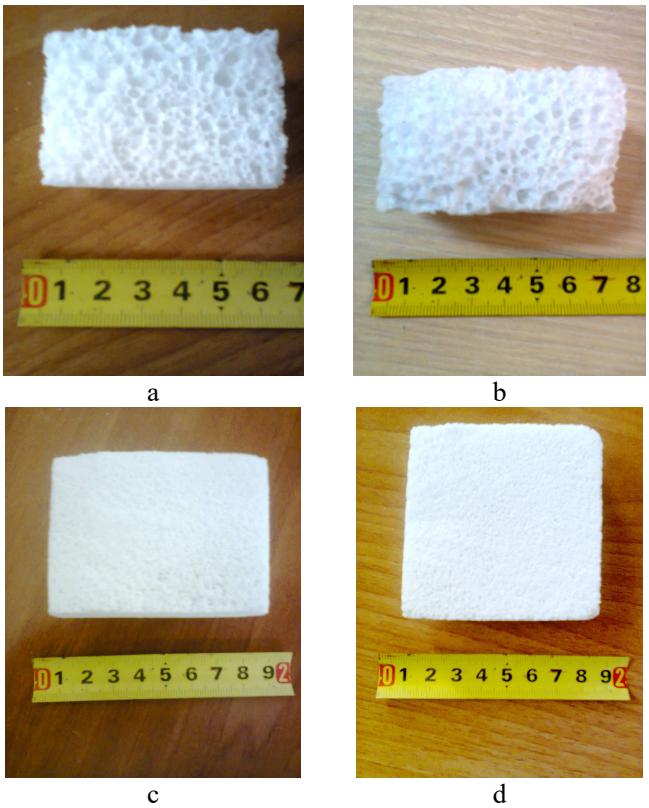



Fig. 2. Cross section of the glass foam samples
a – sample 1 heated at 826 ºC; b – sample 2 heated at 824 ºC;
c – sample 3 heated at 825 ºC; d – sample 4 heated at 827 ºC.

According to Figure 2, the appearance of the glass foam samples section differs clearly depending on the weight ratio of $CaCO_3$. The maximum ratio of 2.5 wt.% (sample 1) favored the formation of a higher pore size structure, while by reduction the $CaCO_3$ proportion up to 0.7 wt.% (sample 4) the pore size obviously decreased. The examination with the digital microscope of the microstructural configuration (Figure 3) allowed the pore size determination and their homogeneity distribution identification.

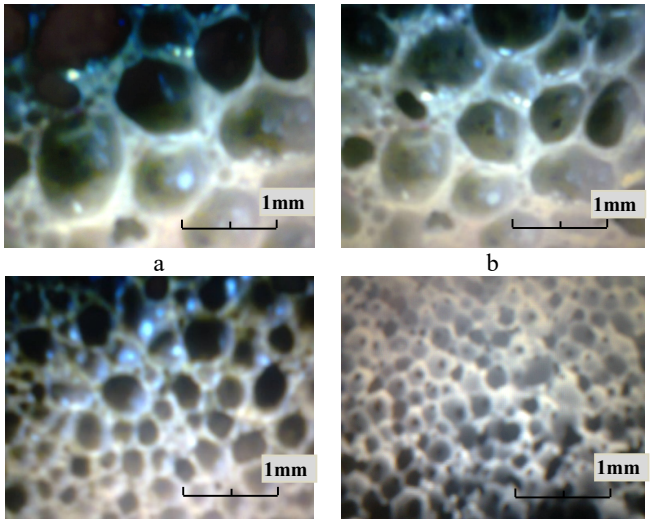



Fig. 3. Microstructural configuration of the glass foam samples
a – sample 1; b – sample 2; c – sample 3; d – sample 4.

The physical, thermal, mechanical and microstructural characteristics of the glass foam samples are presented in Table 4.

*Table 4*

**Main physical, thermal, mechanical and microstructural characteristics of glass foam samples**

| Variant | Apparent density ($g/cm^3$) | Porosity (%) | Thermal conductivity (W/m·K) | Compressive strength (MPa) | Water absorption (vol.%) | Pore size (mm) |
|---|---|---|---|---|---|---|
| 1 | 0.17 | 91.8 | 0.049 | 1.32 | 0.93 | 0.60-1.00 |
| 2 | 0.19 | 90.9 | 0.053 | 1.51 | 1.25 | 0.50-0.80 |
| 3 | 0.22 | 89.4 | 0.060 | 1.83 | 1.16 | 0.20-0.50 |
| 4 | 0.23 | 88.9 | 0.063 | 2.02 | 1.84 | 0.10-0.25 |

According to the data in Table 4, the main characteristics of glass foam samples show excellent thermal insulating properties. The apparent density values are very low between 0.17-0.23 $g/cm^3$ and the thermal conductivity is also very low (0.049-0.063 W/m·K) determining the thermal insulating properties. The porosity values are high, the range being between 88.9-91.8 %. The glass foam samples are waterproof materials, the water absorption being below 1.84 vol.%. According to the pictures in Figure 3, the pore size is between 0.60-1.00 mm for sample 1 and between 0.10-0.25 mm for sample 4. Compared to the glass foams industrially manufactured by conventional heating techniques, the characteristics of the experimental glass foams produced by the unconventional technique of microwave heating are similar being suitable for using as thermal insulating building material.

The compressive strength of glass foam (1.32-2.02 MPa) was significantly improved by the addition of borax in the composition of the raw material mixture.

Generally, a foamed material prepared with only $CaCO_3$ as a foaming agent has a compressive strength less than 1.3 MPa [13]. By decreasing the $CaCO_3$/borax ratio from 1.67 (variant 1) to 0.47 (variant 4) the compressive strength value increased by 1.5 time.

Analyzing the glass foam characteristics it is found that all four experimental variants correspond in terms of quality and energy to the requirements of an adequate thermal insulation material. However, the authors adopted as the optimal variant, variant 3 produced with 1.1 wt.% $CaCO_3$ and 1.5 wt.% borax mixed with 97.4 wt.% colorless glass waste and a water addition of 9 wt.% as a binder. Its apparent density was 0.22 g/cm$^3$ and the thermal conductivity had the value of 0.060 W/m·K ensuring very good thermal insulation properties. The compressive strength of 1.83 MPa is high enough for the material to have excellent mechanical strength. The specific energy consumption was very low (0.66 kWh/kg).

## 4. Conclusion

The paper aimed to improve in terms of energy the process of manufacturing glass foam from glass waste by applying an unconventional technique of predominantly direct microwave heating, unlike the conventional techniques industrially used in the world. The heating method is original, being tested by authors in various variants in the last four years. The experimentally manufactured glass foam with $CaCO_3$ as a foaming agent and borax as a fluxing agent has characteristics similar to those industrially produced by conventional techniques (apparent density of 0.22 g/cm$^3$, thermal conductivity of 0.060 W/m·K and compressive strength of 1.83 MPa). The specific energy consumption of the experimental process was very low (0.66 kWh/kg) below the level of energy consumptions of industrial producers.

## References

[1] *** Glass: Material-Specific Data, Environmental Protection Agency of the United States, November 2020. https://www.epa.gov/facts-and-figures-about-materials-waste-and-recycling/glass-material-specific-data

[2] C. Bina, „World Oil and Crisis of Globalization" in A Prelude to the Foundation of Political Economy, New York, 2013, pp. 17-43. https://doi.org/10.1057/9781137106971_2

[3] G. Scarinci, G. Brusatin, E. Bernardo, „Glass Foams" in Cellular Ceramics: Structure, Manufacturing, Properties and Applications, Wiley-VCH GmbH & KgaA, M. Scheffler, P. Colombo eds., Weinheim, Germania, 2005, pp. 158-176.

[4] J. Hurley, "Glass-Research and Development", Final Report, A UK market survey for foam glass, The Waste and Resources Action Programme Publication, Banbury-Oxon, Great Britain, 2003.

[5] Oxana Kharissova, B.I. Kharissov, J.J. Ruiz Valdés, "Review: The Use of Microwave Irradiation in the Processing of Glasses and their Composites", Industrial & Engineering Chemistry Research, vol. **49**, no. 4, 2010, pp. 1457-1466.

[6] M. Knox, G. Copley, "Use the Microwave Radiation for the Processing of Glass", Glass Technology, vol. **38**, no. 3, 1997, pp. 91-96.

[7] R.R. Menezes, P.M. Souto, R.H.G.A. Kiminami, "Microwave Fast Sintering of Ceramic Materials" in Sintering of Ceramics-New Emerging Techniques, 2012. https://www.doi.org/10.5772/34181
[8] M.N. Rahaman, "Sintering of ceramics, CRC Press, Taylor & Francis Group, Boca Raton, London, New York, 2007. https://books.google.ro
[9] U. Kolberg, M. Roemer, "Reacting of Glass", Ceramic Transaction, vol. **111**, 2001, pp. 517-523.
[10] L. Paunescu, B.T. Grigoras, M.F. Dragoescu, S.M. Axinte, A. Fiti, "Foam Glass Produced by Microwave Heating Technique", Bulletin of Romanian Chemical Engineering Society, vol. **4**, no. 1, 2017, pp. 98-108.
[11] D.A. Jones, T.P. Lelyveld, S.D. Mavrofidis, S.W. Kingman, N.J. Miles, "Microwave Heating Applications in Environmental Engineering-A Review", Resources, Conservation and Recycling, vol. **34**, no. 2, 2002, pp. 75-90.
[12] L. Paunescu, S.M. Axinte, B.T. Grigoras, M.F. Dragoescu, A. Fiti, "Testing the Use of Microwave Energy to Produce Foam Glass", European Journal of Engineering and Technology, vol. **5**, no. 4, 2017, pp. 8-17.
[13] S.M. Axinte, L. Paunescu, M.F. Dragoescu, Ana C. Sebe, "Manufacture of Glass Foam by Predominantly Direct Microwave Heating of Recycled Glass Waste", Transactions on Networks and Communications, vol. **7**, no. 4, 2019, pp. 37-45. https://doi.org/10.14738/tnc.74.7214
[14] Helen J. Kitchen, S.R. Vallance, Jennifer L. Kennedy, Nuria Tapia-Ruiz, Lucia Carassiti, "Modern Microwave Methods in Solid-State Inorganic Materials Chemistry: From Fundamentals to Manufacturing", Chemical Reviews, vol. **114**, no. 2, 2014, pp. 1170-1206.
[15] K.S.P. Karunadasa, C.H. Manoratne, H.M.T.G.A. Pitawala, R.M.G. Rappakse, "Thermal Decomposition of Calcium Carbonate (Calcite Polymorph) as Examined by In-Situ High-Temperature X-Ray Powder Diffraction", Journal of Physics and Chemistry of Solids, vol. **134**, 2019, pp. 21-28.
[16] T. Koizumi, K. Kido, T. Nakamura, "Foaming Agent for Powder Metallurgy Production of Aluminum Foam", Materials Transactions, vol. **52**, no. 4, 2011, pp. 728-733. https://www.doi.org/10.2320/MATERTRANS.M2010401
[17] H.F. Miller, "Borax, Borates and Other Boron-Carrying Compounds", Agronomy Notes, no. 16, University of Kentucky, 1964.
[18] *** "Manual of Weighing Applications", Part 1, Density, 1999. http://www.docplayer.net/21731890-Manual-of-weighing-applications-part-1-density_html
[19] L.M. Anovitz, D.R. Cole, "Characterization and Analysis of Porosity and Pore Structures", Reviews in Mineralogy and Geochemistry, vol. **80**, 2005, pp. 61-164.
[20] *** Foam Glass Manufacturing, Energocell Foam Glass, Debrecin, Hungary, 2014. https://www.energocell.hu/en/foamglass-manufacturing/