

Izabela Marchewka^{1*}, Ewa Downar-Zapolska¹, Waldemar Pichór²

¹Lakma SAT, Cieszyn, Poland

²AGH University of Science and Technology, Faculty of Materials Science and Ceramics, al. A. Mickiewicza 30, 30-059 Krakow, Poland

*Corresponding author. E-mail: izabela.marchewka@lakma.com

Received (Otrzymano) 30.04.2022

THERMAL, MECHANICAL AND MOISTURE PROPERTIES OF POLYMER PLASTER MODIFIED WITH LIGHTWEIGHT FILLERS

Thermally insulating lightweight plasters are urgently needed for the development of sustainable energy-saving buildings. The creation of such a material is possible because of the use of lightweight fillers, which are of interest to many researchers. As a result of their porous structure, it is possible to lower the density and thermal conductivity of various materials. The aim of this work is to analyze the influence of hollow glass microspheres in two granulations and expanded glass on the parameters of polymer plaster. Composites containing various amounts of lightweight fillers were prepared. Then, their parameters were compared in terms of the mechanical properties, thermal conductivity, vapor permeability, and water absorption. All three lightweight fillers successfully reduced the density of the polymer mass and improved plaster insulation. The best effect was achieved with both types of hollow glass microspheres. The weight of the polymer binder was reduced by up to 80%. In the case of expanded glass, the density was only reduced by 38%. At the same time, the composites were studied in terms of their mechanical and moisture properties. The specimens with the highest amount of lightweight filler showed significant deterioration in elasticity due to disturbance of the pigment:binder ratio. They cracked at lower deflection angles. By not exceeding 20% of the filler, no effect on flexibility was observed in any of the three investigated fillers. Interpretation of the research results indicates that the studied lightweight fillers allow a plaster mass to be obtained with an extremely low thermal conductivity coefficient, less than 0.086 W/mK. However, while ensuring the optimal proportion between fillers and polymer binder, it is possible to reduce the coefficient to 0.261 W/mK without noticeable deterioration of the mechanical properties.

Keywords: lightweight fillers, polymer plaster, thermal insulating, hollow glass microspheres, expanded glass

INTRODUCTION

The intensive development of the economy causes a constantly increasing demand for energy. According to some forecasts, global energy consumption will grow by another 50% by 2030 [1]. For this reason, the greatest challenges of the 21st century include both the search for new energy sources and the effective use of existing ones. An important element of this strategy is to increase the energy efficiency of buildings. According to recent studies, the building sector consumes 40% of the energy generated and approximately 60% of the total energy used in buildings is absorbed by heating and cooling the space [2]. Thus, a large proportion of the energy used in buildings depends on the employed insulation strategy [3]. One of the challenges is to reduce heat loss through the facade.

The demand for materials with low thermal conductivity is increasing to improve the thermal insulation of the building and decrease energy consumption. Commonly used building insulation methods are often not a fully satisfactory solution. The aim is also to minimize heat transfer through the building materials themselves. This can be achieved, for example, by incorporating lightweight fillers into the structure of the material.

Lightweight fillers possess a high content of air voids within the structure, creating a lightweight and thermally insulating material [4]. The thermal conductivity of air is lower than that of conventional materials, and thus a higher porosity would lead to reduced thermal conductivity. However, it could be seen that increased porosity of the structure also affects other composite parameters [5]. Research that has already been published focuses on the use of different types of fillers. The application of expanded glass [6], glass beads [7], expanded perlite [8, 9], expanded polystyrene [10], expanded clay [11], aerogel [12, 13], silica nanospheres [14], fly ash cenospheres [13, 15], pumice, diatomite [16], glass bubbles [17] and many more in cement composites was studied. The addition of all of the fillers mentioned above to cement composites resulted in a decrease in the density of the material, which translated into improvement in its thermal insulation properties.

Hollow glass microspheres (HGMS) are among the most promising lightweight fillers because of their small size and closed-shell structure. They are made of sodium borosilicate glass and are available on the commercial market in many types with different densi-

ties and particle sizes. Zhuge et al. [18] investigated the effect of a 10% cement replacement by HGMs in a fiber-reinforced composite. A reduction in density of 6% was achieved with a simultaneous only slight deterioration of the compressive strength. In a similar work, Aslani and Wang [19] obtained a 10% reduction in density and only a 3% loss in compressive strength. Further work was also able to demonstrate a positive effect on the Young modulus and fire resistance [20]. Only when the proportion of HGM in cement exceeds 20%, the strength of the mortar begins to deteriorate markedly. Nevertheless, with the addition of 40%, it was possible to reduce the thermal conductivity by 45% [21]. Therefore, it can be assumed that the addition of microspheres up to a certain limit does not significantly affect the compressive strength.

Another kind of lightweight filler is expanded glass (EG), which is post-consumer recycled glass. The expansion process produces granules with fine pores filled with air [22]. In recent years, researchers have also been trying to use expanded glass in building materials. In Yu's research [23], the addition of expanded glass made it possible to obtain ultralight concrete with a density of 750 kg/m^3 and a thermal conductivity of approximately 0.165 W/mK . Furthermore, compared to other lightweight concretes of the same density, it had significantly higher compressive strength (about 16 MPa). A relationship was also observed between the size of the expanded glass granules and the strength of the concrete. The finer the filler, the less strength loss was observed [24].

The results obtained for composites containing light fillers and mineral building materials are promising. However, classic mineral plaster, which is the final layer of a building partition, is increasingly being replaced by polymer plasters. They are more durable, flexible and provide protection against weather conditions. In the present study, three types of lightweight fillers: expanded glass, $30 \text{ }\mu\text{m}$ glass microspheres, and $66 \text{ }\mu\text{m}$ glass microspheres, were introduced to polymer plaster. The aim is to reduce the thermal conductivity coefficient and to analyze the influence of light fillers on other parameters of the plaster.

The binder presented in the work is part of the polymer plaster; it constitutes approximately 60% by weight of the finished plaster mass. Therefore, a significant reduction in its thermal conductivity coefficient will translate into improvement in the thermal insulation properties of the entire material. In addition, thin-layer plasters in most cases function in conjunction with thermal insulation systems, being part of ETICS (external thermal insulation composite system). Hence, the material presented in the work may be one of the elements of a building partition. The thermal resistance of the partition is the sum of the thermal resistance of the individual layers of materials. By improving the thermal insulation properties of one of them, even as thin as facade plaster, the thermal conductivity coeffi-

cient of the entire partition will be reduced. As a result, the energy losses through the facade will decrease.

MATERIALS AND METHODS

Characteristics of materials and preparation of composites

The basis of the experiment was commercial polymer plaster containing a styrene-acrylic matrix. Except for the binder, the plaster consisted of fine inorganic fillers, functional additives, and a diluting medium (in this case water). Nonetheless, the plasters were deprived of fracturing aggregates. Plaster composites with various amounts of three different kinds of lightweight fillers were prepared. Lightweight fillers were used instead of part of carbonate filler. In the case of the specimens with the addition of lightweight filler above 22.7 vol.%, part of the diluting medium was also changed. The fillers were added to the whole plaster mass during the dispersing process and then were mixed until homogeneous. Two types of hollow glass spheres and fine granulated expanded glass were used as the lightweight filler. The parameters of all the mentioned fillers are given in Table 1.

TABLE 1. Characteristics of lightweight fillers

	HGM-I	HGM-II	EG
Mean particle size [μm]	30	66	100
Effective density [g/cm^3]	0.28	0.19	1.15
Crush strength [MPa]	21	3.4	9
Thermal conductivity coefficient [W/mK]	0.1	0.12	no data available

The composition of the plaster components was various for different specimens. The plasters were prepared in two series. At first, the lightweight fillers were introduced to the mass as much as the wettability allowed. Subsequently, specimens were prepared with half of the prescribed maximum amount of filler. The second series was designed so that the volume of each lightweight filler was at a similar level in the corresponding specimens.

Basic characteristics of specimens

The basic parameters determined for the prepared specimens were dry density, viscosity, and the pigment:binder ratio (P:B). Before the viscosity test, the specimens were cooled to 20°C . The viscosity was determined using a Ford cup with a diameter of 10 mm. The viscosity is expressed by the time 50 ml of mass flows out of the Ford cup. Before the dry density measurements, the mass was applied to a round form of known volume and dried at room temperature to

a constant weight. Dry specimens were weighed, and the dry density was calculated according to the known volume. The P:B ratio was calculated from the composition of the recipe.

Mechanical properties

The elasticity of the composite was tested by bending a thin disc of dried mass. The plaster mass was applied in special forms with a thickness of 4 mm and a diameter of 80 mm. The form was removed after 20 minutes. The discs were allowed to dry and then seasoned for 14 days under 50% humidity and a temperature of $20\pm 2^\circ\text{C}$. The flexibility of the plaster was determined by the angle at which the mass fractured when bending a disc around a metal roller with a diameter of 7 mm. The test was repeated three times.

Thermal properties

The thermal conductivity study was carried out on specimens in the form of discs with a thickness of 10 mm. Plaster was applied to plastic molds in three layers with two-day intervals between them. The next specimens were seasoned for 14 days under 50% humidity and a temperature of $20\pm 2^\circ\text{C}$. After being completely dried, the specimens were ground to obtain a smooth parallel surface. The thermal conductivity measurements were carried out using the stationary method with a TA Instruments FOX 50 apparatus.

Moisture properties

Two parameters were measured – the water absorption and vapor permeability of the coating. The specimens for both tests were prepared in the same way as the specimens for the mechanical properties test. For the vapor permeability measurements, plastic cups with a capacity of 200 ml were used, with an opening with a surface and shape corresponding to the previously prepared discs. The containers were filled with water to a height of 1 cm from the upper border, and then the cup was covered with a previously prepared plaster disc. The place of connection of the disc to the edge of the cup was sealed with a plastic mass. The specimens prepared this way were weighed and then placed in a chamber that kept the temperature at 40°C and relative humidity of the air at about 40%. The specimens were weighed every 24 hours. On the basis of the weight loss of the specimen, the water vapor permeability was calculated.

After conditioning the specimens, they were weighed and then placed in a container with water at room temperature ($23\pm 2^\circ\text{C}$). The discs were arranged in such a way as to allow free contact with water on both sides. After 24 hours, the specimens were removed from the water container, dried with a damp cloth, and then weighed. On the basis of the difference in mass and the knowledge of the contact surface of the

specimen with water, the water absorption was calculated.

RESULTS AND DISCUSSION

Basic characterization of composites

The basic properties of the prepared plaster specimens are shown in Table 2. The maximum amount of lightweight filler that could be incorporated into the mass depended on the type of filler. Expanded glass was successfully introduced in the smallest amount, only 22.77 vol.%. Hollow glass microspheres exceeded this value twice: the smaller microspheres were added in the amount of 46.24 vol.% and the larger ones in the amount of 49.17 vol.%. This difference is due to the different surface development values that are related to wettability.

The density of the composites with all the kinds of lightweight extenders are lower than those of the reference specimen. The obtained density results fluctuate in the range from 1.54 to 0.37 g/cm^3 . The lowest value was achieved for the specimen with 49 vol.% of HGM-I and is 80% lower than the reference specimen. When the same amount of different fillers was used, the composites with the expanded glass had the highest density. The plasters with the same amount of both types of glass microspheres showed similar density results.

An increase in the viscosity of the mass could be observed with large amounts of introduced lightweight extenders. With a filler content greater than 22.77 vol.%, the consistency of the mass was no longer measurable by the selected method. The plasters with the light filler additions were also characterized by an increased value of the P:B ratio. This is due to the increased proportion of fillers in the volume to the acrylic binder in the plaster formulation.

TABLE 2. Basic characteristic of specimens

Specimen	Lightweight filler [vol.%]	Density [g/cm^3]	Viscosity [s]	P:B
R	0	1.86	17	3.13
EG-A	7.67	1.54	19	3.77
EG-B	14.17	1.31	27	4.42
EG-C	22.77	1.15	im	6.94
HGM-I-A	14.17	1.10	13	4.42
HGM-I-B	18.47	0.94	32	5.68
HGM-I-C	22.77	0.75	im	6.94
HGM-I-D	28.39	0.77	im	7.27
HGM-I-E	46.24	0.50	im	11.45
HGM-II-A	14.17	1.1	15	4.42
HGM-II-B	18.47	0.95	180	5.68
HGM-II-C	22.77	0.81	im	6.94
HGM-II-D	41.84	0.63	im	9.48
HGM-II-E	49.17	0.37	im	15.85

im - immeasurable

Mechanical properties

The results of the flexibility test are shown in Table 3. All the specimens with a P:B coefficient not differing by more than 122% from that of the reference specimen showed no deterioration in terms of elasticity relative to that of the reference specimen. None of the plasters with a P:B ratio below 6.94 was damaged during the test. The composites with higher contents of light filler were characterized by smaller bending resistance. Nevertheless, the test results did not depend on the type of employed filler, but the amount of filler was significant. The loss of flexibility is due to the reduced binder content, which is responsible for the mechanical properties of the coating. The flexibility of the composite containing the same amounts of different fillers was similar, within the measuring deviation. The results confirmed that the content of the polymer binder is largely responsible for the mechanical strength of the polymer coatings. The role of the binder is to hold all the components of the mass together. The amount of polymer binder must be sufficient to wet all the pigments and fillers present in the mass. When a certain limit of dry components is exceeded, the parameters of the coating change, including the mechanical properties.

TABLE 3. Results of flexibility test

Specimen	Damage angle	Standard deviation
R	nd	-
EG-A	nd	-
EG-B	nd	-
EG-C	111°	6.66°
HGM-I-A	nd	-
HGM-I-B	nd	-
HGM-I-C	122°	25.66°
HGM-I-D	61°	5.29°
HGM-I-E	45°	5°
HGM-II-A	nd	-
HGM-II-B	nd	-
HGM-II-C	118°	11.50°
HGM-II-D	51°	7.94°
HGM-II-E	45°	6.08°

nd – no damage at angle of 180°

Thermal conductivity coefficient

The thermal conductivity coefficient values are given in Figure 1. The coefficients of the thermal conductivity of the plasters with the lightweight filler are substantially lower than that of the reference specimen. The measurement results for the prepared composites ranged from 0.517 to 0.086 W/mK.

The obtained effect of lowering the thermal conductivity depended on the type of filler used. On the other hand, in direct comparison of the fillers, the composite with the addition of expanded glass obtained the lowest reduction effect of the thermal conductivity coefficient.

In this way, it was possible to reduce heat conduction by 61%. Much higher results were obtained for the composites containing hollow glass microspheres. At the same dosing level, the larger and smaller achieved, respectively, a reduction of 72 and 77%.

An additional advantage of the microspheres was the possibility of introducing them into the mass in a much greater amount than the expanded glass. With a dosage of microspheres in an amount close to 50%, it was possible to reduce the thermal conductivity by up to 88%, thus obtaining an extremely low level of thermal conductivity. When compared directly, the best insulation properties were found for the materials with the smaller microspheres, while the larger ones could be incorporated into the plaster in greater amounts. Therefore, the lowest thermal conductivity coefficient was obtained for the specimen with the addition of larger microspheres because of the higher possible dosage.

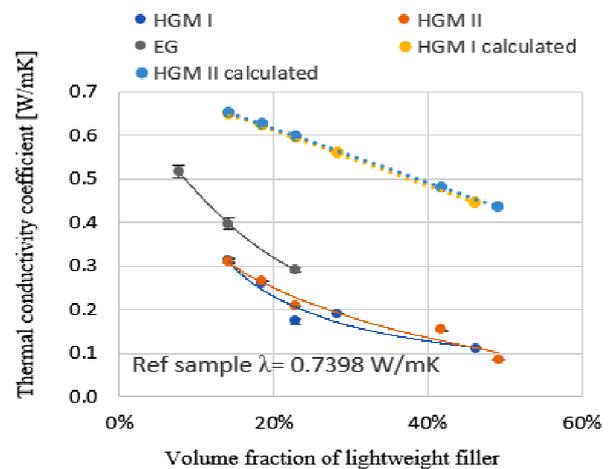


Fig. 1. Thermal conductivity of composite vs. volume fraction of lightweight filler

The thermal conductivity of all the specimens with HGM I and HGM II was significantly lower than the value calculated with the assumption of additivity. The differences may be due to the fact that the calculations used the value of thermal conductivity coefficient of lightweight fillers given by the manufacturers in the technical specification. Therefore, the method of its determination is unknown.

Moisture properties

The vapor permeability and water absorption of the prepared composites were tested. The obtained results are presented in Figures 2 and 3. All the three lightweight fillers changed the vapor permeability of the plaster coating. With an addition below 20%, the change was hardly noticeable; the results are within the measuring error of the reference specimen. Minimal fluctuations could be caused by replacing the carbonate filler with a filler with a different granulation. A significant increase in vapor permeability was

observed for the composites with a light filler content greater than 20%. These specimens were characterized by a generally high filler content, which resulted in a disturbance of the P:B ratio. The reduction of the volume fraction of the binder increased the porosity of the coating, leading to higher water vapor permeability. The highest vapor permeability was observed in the specimens with the addition of HGM-I.

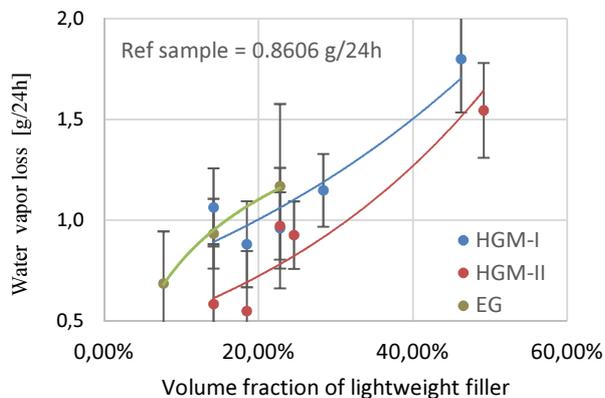


Fig. 2. Water vapor loss vs. volume fraction of lightweight filler

The specimens with the addition of the lightweight filler exhibited higher water absorption than the reference specimen. The amount of water absorbed increased with the amount of introduced additive. The most intensive growth was observed for the composites with the addition of expanded glass. This phenomenon could have been influenced by the much greater surface development obtained in the expansion process than in the case of microspheres with an ideal spherical shape. The open porosity of the expanded glass increased the water absorption capacity of the plaster mass. However, the results obtained for both the HGM sizes were very similar.

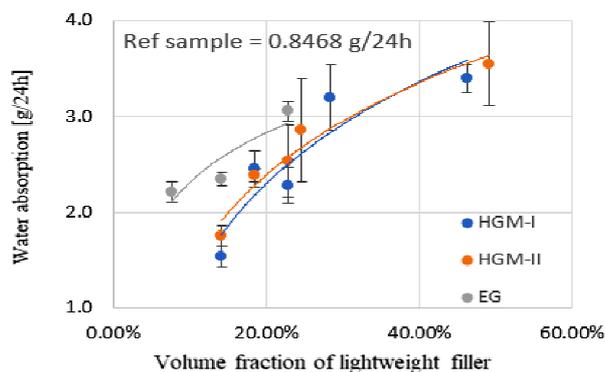


Fig. 3. Water absorption vs. volume fraction of lightweight filler

CONCLUSIONS

The results of the experiments confirmed the assumptions made in the paper. The addition of a lightweight filler allows the plaster density to be reduced, and thus a reduction in the thermal conductivity.

This confirmed the effectiveness of the use of lightweight fillers to improve the thermal insulation properties of polymer plaster. The material obtained in the work can be used as one of the elements of a building partition with low thermal conductivity.

Both expanded glass and HGMs effectively reduced the thermal conductivity coefficient. Nevertheless, a greater effect was obtained for microspheres than for expanded glass, which was successfully introduced in much smaller amounts. Moreover, the effect of improving the insulation obtained for HGM-I and HGM-II was very similar. In the case of the content of microspheres not exceeding 18.47 vol.%, the thermal conductivity coefficient determined for the microspheres of various sizes differed by a maximum of 1.6%. For larger amounts of filler, the differences were more noticeable; however, in direct comparison the specimens with the smaller microspheres were always characterized by lower values of thermal conductivity.

The research showed that the addition of microspheres does not adversely affect the mechanical properties, provided that the volume proportions between the binder and fillers are kept unchanged. The specimens with the addition of lightweight fillers at the level of 18.77 vol.% and below showed no loss of flexibility of the plaster coating. With larger amounts, clear deterioration of this parameter was observed.

The influence of lightweight fillers on the moisture parameters of the coating was also observed. The water absorption of the coating increased with the amount of introduced fillers. The effects of HGM I and HGM II were similar, significantly higher water absorption was obtained for expanded glass. Nonetheless, no significant changes in vapor permeability were observed at low amounts of the additive. The increase was only observed above 20 vol.% light filler.

REFERENCES

- [1] Ong H.C., Mahlia T.M.I., Masjuki H.H., A review on energy scenario and sustainable energy in Malaysia, *Renewable and Sustainable Energy Reviews* 2011, January, 15, 1, 639-647, DOI: 10.1016/j.rser.2010.09.043.
- [2] Ozel M., Thermal performance and optimum insulation thickness of building walls with different structure materials, *Applied Thermal Engineering* 2011, December, 31, 17, 3854-3863, DOI: 10.1016/j.applthermaleng.2011.07.033.
- [3] Aditya L., Mahlia T.M.I., Rismanchi B., Ng H.M., Hasan M.H., Metselaar H.S.C., Muraza O., Aditya H.B., A review on insulation materials for energy conservation in buildings, *Renewable and Sustainable Energy Reviews* 2017, June, 73, 1352-1365, DOI: 10.1016/j.rser.2017.02.034.
- [4] Al-Homoud D.M.S., Performance characteristics and practical applications of common building thermal insulation materials, *Building and Environment* 2005, March, 40, 3, 353-366, DOI: 10.1016/j.buildenv.2004.05.013.
- [5] Hanif A., Lu Z., Cheng Y., Diao S., Effects of different lightweight functional fillers for use in cementitious composites, *International Journal of Concrete Structures and Materials* 2017, March, 11, 1, 99-113, DOI: 10.1007/s40069-016-0184-1.

- [6] Bumanis G., Bajare D., Korjakins A., Mechanical and thermal properties of lightweight concrete made from expanded glass, *Journal of Sustainable Architecture and Civil Engineering* 2013, June, 2, 3, 26-32, DOI: 10.5755/jol.sace.2.3.2790.
- [7] Chung S.Y., Han T.S., Kim S.Y., Kim J.H.J., Youm K.S., Lim J.H., Evaluation of effect of glass beads on thermal conductivity of insulating concrete using micro CT images and probability functions, *Cement and Concrete Composites* 2016, January, 65, 150-162, DOI: 10.1016/j.cemconcomp.2015.10.011.
- [8] Kramar D., Bindiganavile V., Mechanical properties and size effects in lightweight mortars containing expanded perlite aggregate, *Materials and Structures* 2010, September, 44, 4, 735-748, DOI: 10.1617/s11527-010-9662-0.
- [9] Sengul O., Azizi S., Karaosmanoglu F., Tasdemir M.A., Effect of expanded perlite on the mechanical properties and thermal conductivity of lightweight concrete, *Energy and Buildings* 2011, February-March, 43, 2-3, 671-676, DOI: 10.1016/j.enbuild.2010.11.008.
- [10] Sayadi A.A., Tapia J.V., Neitzert T.R., Clifton G.C., Effects of expanded polystyrene (EPS) particles on fire resistance, thermal conductivity and compressive strength of foamed concrete, *Construction and Building Materials* 2016, June, 112, 716-724, DOI: 10.1016/j.conbuildmat.2016.02.218.
- [11] Devecioglu A.G., Biçer Y., The effects of tragacanth addition on the thermal and mechanical properties of lightweight concretes mixed with expanded clay, *Periodica Polytechnica Civil Engineering* 2015, January, 60, 1, 45-50, DOI: 10.3311/PPci.7984.
- [12] Gao T., Jelle B.P., Gustavsen A., Jacobsen S., Aerogel-incorporated concrete: An experimental study, *Construction and Building Materials* 2014, February, 52, 130-136, DOI: 10.1016/j.conbuildmat.2013.10.100.
- [13] Hanif A., Diao S., Lu Z., Fan T., Li Z., Green lightweight cementitious composite incorporating aerogels and fly ash cenospheres – Mechanical and thermal insulating properties, *Construction and Building Materials* 2016, July, 116, 422-430, DOI: 10.1016/j.conbuildmat.2016.04.134.
- [14] Gao T., Jelle B.P., Sandberg L.I.C., Gustavsen A., Mono-disperse hollow silica nanospheres for nano insulation materials: synthesis, characterization, and life cycle assessment, *ASC Applied Materials & Interfaces* 2013, January, 5, 3, 761-767, DOI: 10.1021/am302303b.
- [15] Hanif A., Lu Z., Li Z., Utilization of fly ash cenosphere as lightweight filler in cement-based composites – A review, *Construction and Building Materials* 2017, July, 144, 373-384, DOI: 10.1016/j.conbuildmat.2017.03.188.
- [16] Topçu I.B., Uygunoğlu T., Properties of autoclaved lightweight aggregate concrete, *Building and Environment* 2007, December, 42, 12, 4108-4116, DOI: 10.1016/j.buildenv.2006.11.024.
- [17] Yun T.S., Jeong Y.J., Han T.S., Youm K.S., Evaluation of thermal conductivity for thermally insulated concretes, *Energy and Buildings* 2013, June, 61, 125-32, DOI: 10.1016/j.enbuild.2013.01.043.
- [18] Zhuge Y., Shen C.J., Lu G.X., Hesse G., Chen S., Ruan D., Material properties and impact resistance of a new lightweight engineered cementitious composite, 23rd Australasian Conference on the Mechanics of Structures and Materials, Byron Bay, Australia, December 2014, ACMSSM 23, 9-12.
- [19] Aslani F., Wang L., Development of strain-hardening lightweight engineered cementitious composites using hollow glass microspheres, *Structural Concrete* 2019, June, 21, 20, 673-688, DOI: 10.1002/suco.201900096.
- [20] Aslani F., Wang L., Fabrication and characterization of an engineered cementitious composite with enhanced fire resistance performance, *Journal of Cleaner Production* 2019, June, 221, 202-214, DOI: 10.1016/j.jclepro.2019.02.241.
- [21] Kabay N., Kizilkanat A., Aktruk B., Kahraman Y., Lightweight cement-based composites incorporating hollow glass microspheres: Fresh and hardened state properties, *Teknik Dergi* 2022, January, 33, 1, 1-25, DOI: 10.18400/tekderg.677447.
- [22] <https://poraver.com/us/poraver/>
- [23] Yu R., Van Onna D.V., Spiesz P., Yu Q.L., Brouwers H.J.H., Development of ultra-lightweight fibre reinforced concrete applying expanded waste glass, *Journal of Cleaner Production* 2016, January, 112, 690-70, DOI: 10.1016/j.jclepro.2015.07.082.
- [24] Adhikary S.K., Rudzionis Z., Influence of expanded glass aggregate size, aerogel and binding materials volume on the properties of lightweight concrete, *Materials Today: Proceedings* 2020, April, 32, 4, 712-718, DOI: 10.1016/j.matpr.2020.03.323.