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POSSIBILITIES TO RECYCLE AUTO GLASS WASTE IN BUILDING CERAMICS

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Abstract. Substantial amounts of glass waste end up in landfills because unlike ordinary glass, auto glass is unsuitable for recycling in the glass industry due to organic additives. However, it was determined that the additives make auto glass waste appropriate for the production of building ceramics, which results in improved physical-mechanical and structural properties. The optimum quantity of the additive contained in auto glass waste amounts to 15% of the formation mixture, and the most appropriate burning temperature is 1080 °C. The density of such ceramic chip is 2059 kg/m^3, the water absorption determined after 72 h is 1.2%, and the forecasted frost resistance at the beginning of decomposition is 1343 cycles, but amounts to 2188 cycles at the end of decomposition.

Keywords: pollution, auto glass waste, recycling, ceramics, physical-mechanical and structural properties, forecasted frost resistance.

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Introduction

Approx. 28.5 million tonnes of glass are recycled in the European Union annually (Uselytė *et al.* 2007). Substantial amounts of glass waste end up in landfills. Unlike ordinary glass, auto glass is unsuitable for recycling in the glass industry due to organic additives. The recycling of auto glass into building materials offers environmentally friendly solutions. In the middle layer between glass sheets, auto glass contains small amounts of polymer structures.

Glass can be recycled but not PVB. The main material used in glass lamination is polyvinyl butyral (PVB), which burns out at approximately 450 °C. It is a disposable by-product of the glass recycling industry available in quantity with no additional collection charges. Recycling by mechanical means is an alternative to landfilling. However, it is important to appreciate the variation in molecular structure of PVB and its effect on material properties and the end use (Dhaliwal, Hay 2002). The recycling problem was examined by Russian scientists (Gorokhovsky *et al.* 2005) focusing on the reuse of glass waste containing PVB. Tensile strength of PVB is 28–59.5 MPa, compressive strength is 80–140 MPa. PVB has good adhesion to different materials as well as good optical properties (colourlessness, resistance to light, translucence) and resistance to atmospheric agents. However, it disperses into water and butyraldehyde at a temperature higher than 160 °C (Kabanov *et al.* 1974).

Experiments (Jaw *et al.* 2001) proved that the binder system composite with polyvinyl butyral would show good thermal behaviour in ceramic processing and the binder system would burnout at a considerably low temperature. Moreover, this effect is advantageous for the binder burnout procedure and ensures easy removal of binder. According to researchers (Seo *et al.* 1997), PVB could be used as a binder in ceramic processing, because it burns out at 500 °C. Concentrations of hazardous substances are present in low concentrations; therefore, amounts emitted during the process of burning stay below the limits established by hygiene standards.

Recycling of various types of waste should be one of the most important contemporary issues. Many

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researchers describe possible uses of waste in the industry of building materials. The studies are carried out using the production waste of mineral or mullite wool, fly ash, crushed glass, waste of paper manufacturing, etc.

These works (Kourti *et al.* 2010; Roether *et al.* 2010) demonstrated, that air pollution control residues can be used as the raw material for the production of glass-ceramics or novel geopolymer–glass composites with excellent mechanical and physical properties.

Researchers (Chiang *et al.* 2010) investigated eco-bricks manufactured using water treatment plant (WTP) sludge and scrap glass. Materials containing from 20% of scrap glass and sintered at 1000 °C demonstrate low water absorption and relatively high compressive strength. These results confirm the feasibility of using scrap glass and WTP sludge to produce sintered construction eco-bricks.

Disposal of PC monitors and TV sets is a growing problem, as over 40% of their weight is comprised of waste glasses with high Pb or Ba–Sr concentrations. This makes them unsuitable for recycling and manufacturing of new glass. A possible way to reuse these types of glass is for manufacturing of clay bricks and roof tiles. No significant release of Pb, Ba, and Sr was observed during firing and leaching test for the carbonate-poor body; in contrast, some Pb volatilization during firing and Sr leaching was observed for the carbonate-rich body. According to authors (Dondi *et al.* 2009), the main constraint is that the glass must have a particle size below the limit of the pan mills used in brickmaking (< 1 mm).

The use of waste was investigated by scientists from China (Lin *et al.* 2012). Their research involved the effect of solar panel waste glass on fired clay bricks. Brick samples were heated to temperatures varying from 700 to 1000 °C for 6 h. Solar panel waste glass brick contained 30% of solar panel waste glass and was sintered at 1000 °C; the addition of solar panel waste glass to the mixture reduced the degree of firing shrinkage. The salt crystallization test and wet–dry tests showed that the addition of solar panel waste glass had highly beneficial effects in increasing the durability of bricks. This indicates that solar panel waste glass is indeed suitable for partial replacement of clay in bricks.

In the researches on building ceramics, crushed glass made from windows and bottles is often used (Angjusheva 2011; Pavlushkina, Kisilenko 2011). It was determined that in terms of the beginning of decomposition, the expected frost resistance of samples containing ≤ 5% of glass waste without sintering additives increases to 548 cycles. Researchers (Mačiulaitis, Malaiškienė 2010, 2012) determined that the use of sawdust (10.4–15.6%) and crushed glass (6.4–9.6%) results in porous and extremely frost resistant ceramics with the total open porosity reaching 40%, and exploitative frost resistance of more than 300 cycles.

Researches conducted by Latvian scientists (Rozenstrauha *et al.* 2011) on the use of waste glass, mud and clay show that such samples have characteristics of sintered ceramics. In order to evaluate the suitable field of application, the following physical-mechanical properties of such samples were detected for novel glass-ceramic composites: bulk density, porosity and water uptake in the temperature range 1110–1140 °C. The composition (containing 20 wt. % waste glass, 20 wt. % of sewage sludge and 60 wt. % of clay) with bulk density of 2.19 g/cm^3, water uptake of 2.5%, porosity of 5%, and compressive strength of 60.3±5.1 MPa, obtained at the temperature range 1130–1140 °C appeared to be the most perspective.

Authors (Balkyavichus *et al.* 2003) determined that the use of very finely ground glass waste lowers the temperature of liquid phase appearance. This suggests that melting glass reacts more intensively with clay because of the increased surface area of the reaction zone and the maximum degree of glass phase can influence the formation of new heat-resistant products, extend the range of sintering and stabilise sintering structure. According to the findings, the optimum amount of ground waste glass is 10%. Then, the density of samples is approx. 2000 kg/m^3, compressive strength increases more than twice (about 40 MPa), and water absorption decreases to 6% or less. Scientists (Bernardo *et al.* 2006, 2007, 2010) analysed the influence of several different types of glass waste on the properties of ceramic samples Such types of waste decrease the sintering temperature from 1100 °C to 950 °C. It was determined that the use of such types of waste and firing of samples at a relatively low temperature (880–930 °C) for 1–3 h makes it possible to produce ceramic articles with compressive strength of 100 MPa and density of 2600 kg/m^3. Additionally, it has been determined that the chemical composition and other characteristics of glass itself have a big influence on the characteristics of finished ceramic products.

The manuscript (Mueller *et al.* 2012) analysed possibilities to recycle various types of glass waste. It revealed that auto glass was not usable in the industry.

This article aims to determine possibilities to recycle processed auto glass waste in building ceramics, which could reduce solid waste pollution.

1. Materials and research methods

Researches focused on the use of the mixture of clay, non-plastics additives and minced crushed auto glass. The chemical composition of clay is given in Table 1. Granulometric composition of such clay is presented in Table 2. The chemical and granulometric compositions of this clay were determined using standard methods (LST EN 725-5:2007; LST EN 1071-4:2002 and other). The clay was passed through a 0.63 mm sieve.

Table 1. Average chemical composition of clay, wt. %

Average chemical composition of clay, wt. %						
SiO_2	Al_2O_3+TiO_2	Fe_2O_3	CaO	MgO	K_2O	L.O.I.
66.33	15.80	6.42	1.80	2.72	1.63	05.30

Table 2. Granulometric composition of clay, wt. %

Average elemental chemical composition of glass, wt. %								
C	O_2	Na	Ca	Mg	K	Al	Si	Fe
2.72	52.06	6.85	6.45	1.76	0.38	0.48	28.14	1.16

Table 3. Average chemical composition of glass, wt. %

Average elemental chemical composition of glass, wt. %								
C	O_2	Na	Ca	Mg	K	Al	Si	Fe
2.72	52.06	6.85	6.45	1.76	0.38	0.48	28.14	1.16

Ground auto glass was used in the researches as the additive fluxing the clay mass.

This glass has a distinguishing characteristic, namely, it contains a sintering component. The elemental chemical composition of glass is given in Table 3 and the X-ray structural analysis is shown in Fig. 1. According to the data in Table 3, no hazardous substances were found in the chemical composition of auto glass. The data in Fig. 1 show that the main mineral of used glass is Quartz *Q* (0.335 nm); also, there is some amorphous phase.

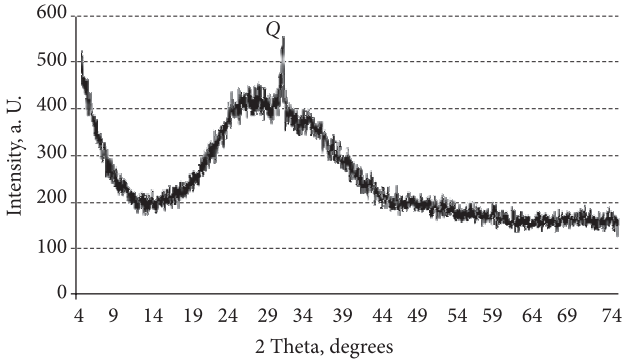
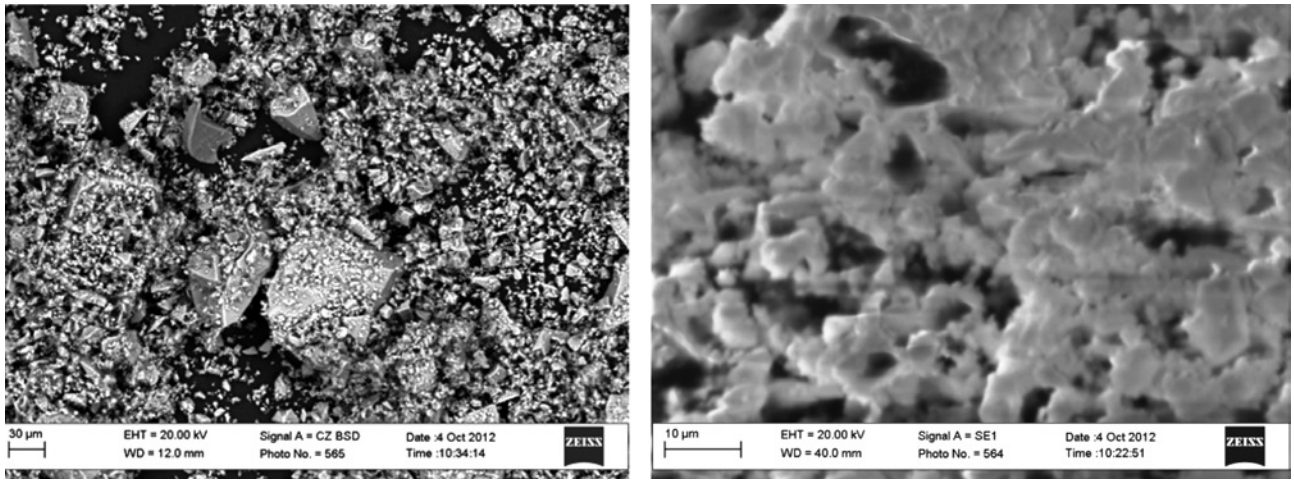


Fig. 1. X-ray diagram of auto glass

The image of used ground glass waste microstructure is presented in Fig. 2.

The microstructure shows that the size of auto waste glass particles varies from 1 to 70 μm, the particles are irregularly shaped and the finest particles are stuck together. However, mixed with clay and non-plastics additives, the particles separate and evenly distribute in the mixture

As presented in Table 4, compositions of ceramic mixtures from these raw materials were selected. The amount of waste was selected from other research results as well as results of researches on various types of waste



a) b)

Fig. 2. Image of ground glass waste microstructure: a – (magnified 400 times); b – (magnified 2500 times)

conducted by the article authors (Malaiškienė *et al.* 2011). The amount of waste was not busted, because of small glass particle formation on the surface of ceramics. Increasing the amount of waste to 20%, a significant amount of glass grains are produced on the surface, which is undesirable.

Table 4. Compositions of ceramic mixtures

Mixture	Mixture of clay and non-plastic additive, %	Auto glass waste, %
S1	100	0
S2	95	5
S3	90	10
S4	85	15

The non-plastic additive is quartz sand (10%), fraction 0/1.

Formed samples (50×50×50 mm) were dried for 3 days under normal laboratory conditions. Later, they were dried to constant weight in the laboratory oven at 105 °C. After drying, the samples were burnt at three different temperatures: 1000 °C (temperature rise rate is 1.04 °C/min, maintaining at the maximum temperature for 4 h, the temperature decrease rate is 1.11 °C/min), 1050 °C (temperature rise rate is 1.09 °C/min, maintaining at the maximum temperature for 4 h, the temperature decrease rate is 1.17 °C/min) and 1080 °C (temperature rise rate of is 1.13 °C/min, maintaining at the maximum temperature for 4 h, the temperature decrease rate is 1.12 °C/min). Temperature decreases to 400 °C in accordance with the set program. Later, the decrease of temperature slows down. The burning temperature was chosen according to the literature and earlier results. The burning temperature was not higher; samples lost their form because of a higher temperature.

The sintered and cooled samples were used to determine the physical-mechanical and structural parameters and forecasted frost resistance according to Table 5 (Mačiulaitis, Malaiškienė 2012). The analysis of glass powder was performed with the help of XRD. The main instrument used was the diffractometer DRON–7; a cobalt anode was used (the wavelength of $\lambda = 0.1792$ nm). The micro-structural analysis of car glass waste was performed using the microscope Carl Zeis Evo LS 25. The parameters of net dry density and water absorption of ceramic samples were determined in accordance with standards LST EN 772-13:2003 and LST EN 771-1+A1, and compressive strength – LST EN 772-1.

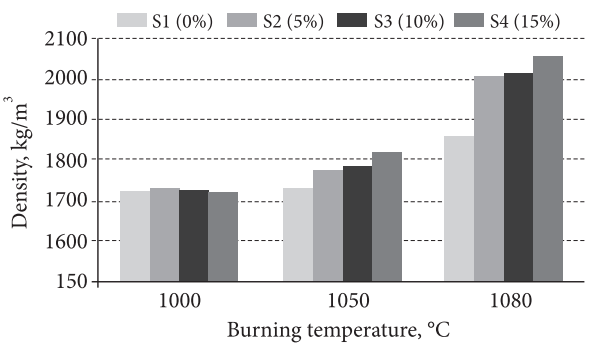


Fig. 3. Density of the samples

To determine whether the change in compressive strength was only determined by the change in density, the strength was recalculated based on the constant density selected from literature (Laukaitis, Sinica 2006):

$$R_{gn} = \frac{R_{gneks} \cdot \rho_{sk}^2}{\rho_{eks}^2}, \quad (1)$$

where: R_{gn} – compressive strength, when density is 1850 kg/m^3, MPa; R_{gneks} – experimental compressive strength, MPa; ρ_{eks} – experimental density, kg/m^3, ρ_{sk} – estimated ceramic density 1850 kg/m^3.

2. Research results and analysis

The average values of parameters are presented in the discussion of obtained results. The obtained density results are presented in Fig. 3.

The data of Fig. 3 show that the increase of density in ceramics is highly dependent on the burning temperature. Due to the ongoing sintering processes, the higher is the temperature, the thicker are the samples. The temperature of 1080 °C is sufficient in order to get the density higher than 2000 kg/m^3. The samples of batches S2–S4 had a much higher density compared to the control samples without the additive of glass. Therefore, the additive of ground auto glass acts as a flux (which stimulates sintering) and increases the density of samples. Subsequent to analysis of the samples burnt at the temperature of 1080 °C, a large difference in density was determined between samples that did not have the fluxing additive and contained 15% of ground auto glass, accordingly 1860 kg/m^3 and 2060 kg/m^3. Such an increase in density is associated with melting of ground auto glass particles at a higher temperature, which results in thicker structure and reduced volume of pores in the sample.

The results regarding the general contraction of ceramics are given in Fig. 4. According to the data, the general contraction increases with the increasing amount of glass waste. Besides, as the general contraction increases, the burning temperature becomes higher. Such effect could be explained by a higher sintering rate. Higher burning temperature and milled glass additive promotes sintering processes.

One of the most important characteristics of ceramics is water absorption. Water absorption, effective and

Table 5. The methodology for forecasting frost resistance according to structural parameters (Mačiulaitis, Malaiškienė 2012)

Description of the basic parameter and units of measurement	Physical meaning of parameters	Calculation formulas	Description of partial values and units of measurement
Reserve of pore volume R, %	Reserve of pore volume defines the quantity of reserve pores and capillaries, to which water or plastic ice penetrate very hardly. The bigger is the reserve of pore volume, the bigger is the exploitation frost resistance of ceramics.	$R = \left(1 - \frac{W_e}{W_p}\right) \cdot 100$	W_e – effective porosity from water absorption after 72 h, %; W_p – total open porosity from water absorption in the vacuum process (special regime), %.
Effective porosity of ceramic body W_e, %	Effective porosity of ceramic body defines the amount of effectively working pores and capillaries.	$W_e = \rho \frac{m_1 - m}{m} \cdot 100$	m – mass of the sample dried up to the constant weight, g; m_1 – mass of the saturated sample, g; ρ – density of the sample, g/cm^3.
Total open porosity W_p, %	Total open porosity defines the total open porous space of ceramic body in the macrostructure and microstructure dimension.	$W_p = \rho \frac{m_2 - m}{m} \cdot 100$	m_2 – mass of the sample saturated using the vacuum process in air, g; m – mass of the sample dried up to the constant weight, g; ρ – density of the sample, g/cm^3.
Qualified thickness of the wall of pores and capillaries D, %	This parameter defines qualified thickness of the wall of pores and capillaries. The greater is the thickness of the wall of pores and capillaries, the greater is the exploitation frost resistance of products.	$D = \frac{100 - W_p}{W_p}$, %	W_p – total open porosity from water absorption in the vacuum process (special regime), %.
Degree of structural inhomogeneity $N_{H,}$ units	Degree of structural inhomogeneity enables to evaluate the structural inhomogeneity of effective capillaries according to their equivalent length.	$N_H = \frac{H_{\max} - H_{\min}}{H_{\min}}$	H_{max}, H_{min} – rates of capillary wetting front (after 5 min), mm.
Capillary rate of mass flow g_1, g/cm^2	Capillary rate of mass flow indicates the equivalent diameter of capillaries. These parameters detail the anisotropy of pores and capillaries.	$g_1 = \frac{m_3 - m}{S}$	m_3 – mass of the sample saturated by the capillary suction process (after 30 min), g; m – mass of the sample dried up to the constant weight, g; S – surface area of the sample, cm^2.
Capillary rate of mass flow in a vacuum in the direction of freezing G_1, g/cm^2		$G_1 = \frac{m_4 - m}{S}$	m_4 – mass of the sample which is being saturated for 10 min by capillary suction in a vacuum as saturation goes through the plane which is being frozen under exploitation, g; m – mass of the sample dried up to the constant weight, g.
Capillary rate of mass flow in a vacuum in a vertical direction to freezing G_2, g/cm^2		$G_2 = \frac{m_5 - m}{S}$	m_5 – mass of the sample which is being saturated for 10 min by capillary suction in a vacuum as saturation goes through the plane which is vertical to the freezing plane under exploitation, g; m – mass of the sample dried up to the constant weight, g.
The beginning of destruction, cycles		$F_{RE1} = 0.231 \frac{R^{1.068} D^{1.345} G_1^{0.275} G_2^{0.663}}{N^{0.285} g_1^{0.830}}$	
The end of destruction, cycles		$F_{RE2} = 0.223 \frac{R^{1.465} D^{0.759} G_1^{0.383} G_2^{0.852}}{N^{0.168} g_1^{1.034}}$	

total open porosity of the samples determined during the researches are presented in Figs 5–7.

The difference in porosity of the samples burnt at the same temperature is insignificant. Burning temperature has the main influence on water absorption and porosity. The lowest water absorption (determined after 72 h) of 1.2%, the effective and total open porosity were characteristic to samples of the batch S4 (the amount of glass waste in formation mixture was 15%), which were burnt at the highest temperature of 1080 °C. This was affected not only by the highest sintering temperature, but also by ground auto glass, as this additive melted during the burning process resulting in the structure of more closed pores and capillaries.

The results of the compressive strength of ceramics are presented in Fig. 8.

According to the results of compressive strength obtained through the research, the increase in the burning temperature results in the increase of the compressive strength due to the decrease in the porosity of the sample. However, investigations on the influence of the amount of waste produced some very interesting results. Once the samples were burnt at temperatures of 1000 °C and 1050 °C, it was determined that the compressive strength of samples with 5 and 10% glass waste was lower compared to samples without this additive. Addition of 15% of ground auto glass to the formation mixture increased the compressive strength to 9–14% in comparison to the control samples. This phenomenon can be explained by small amounts of glass and low burning temperature, which is insufficient for the glass to start melting and forming new combinations. A larger amount of glass reacts with clay particles faster and forms new combinations.

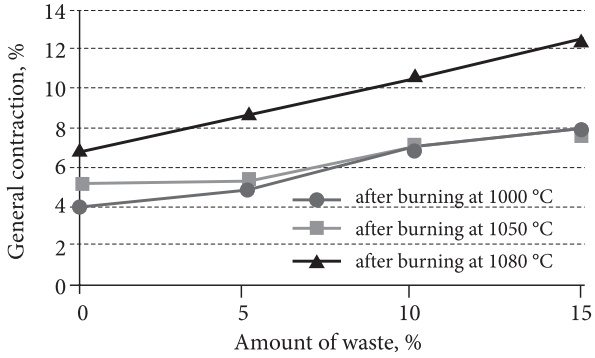


Fig. 4. General contraction of the samples

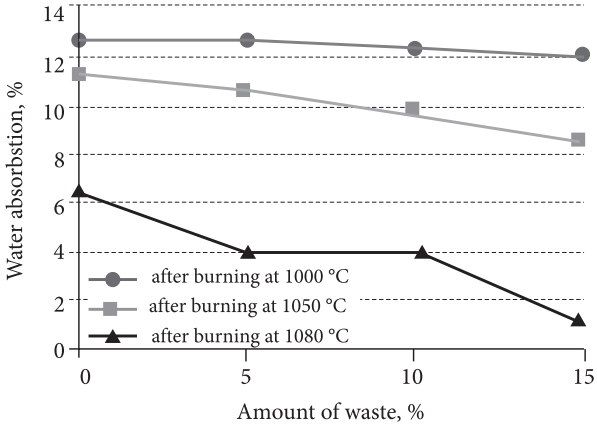


Fig. 5. Influence of the amount of waste on water absorption

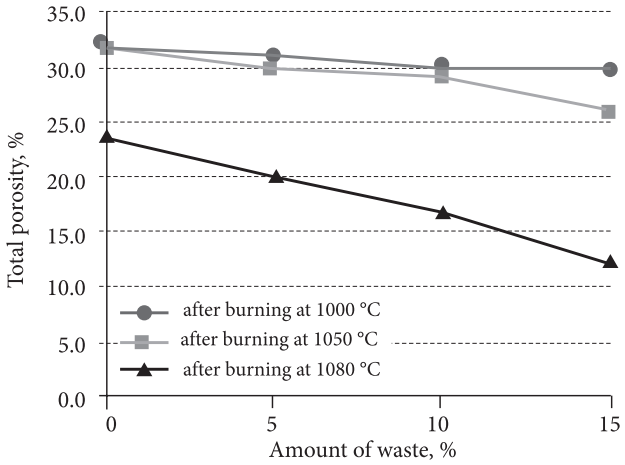


Fig. 7. Influence of the amount of waste on the total open porosity

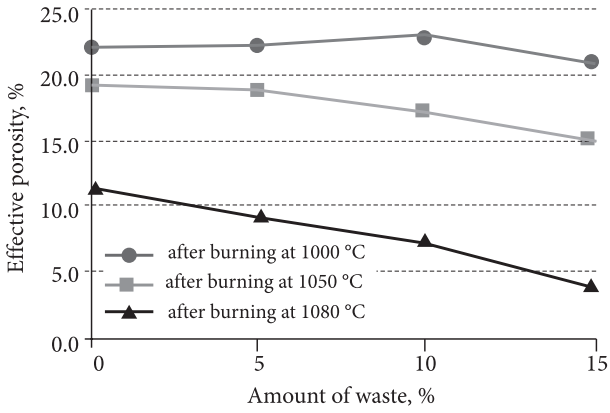


Fig. 6. Influence of the amount of waste on effective porosity

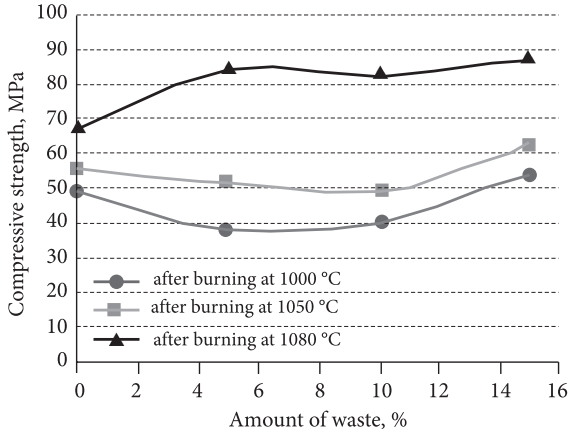


Fig. 8. The results of compressive strength

Results recalculated by the same density (1850 kg/m^3) compressive strength (Laukaitis, Sinica 2006) are given in Fig. 9.

Figs 8 and 9 demonstrate similar persisting variation tendencies of compressive strength, which varied up to 20%. Consequently, the change of the compressive strength not only depends on change in density, but also on the amount of waste.

Sufficiently high compressive strength values were determined in small size samples of 50×50×50 mm, as such samples resulted in fewer defects.

The values of examined structural parameters, by which frost resistance is forecasted, are presented in Table 6. The capillary mass flow rate of the samples decreases with the increase in the burning temperature and the amount of ground auto glass, while the reserve of pore space and the relative thickness of pores and capillaries increase significantly. According to the results of structural parameters, it is possible to assume that in this case, samples with 15% of auto glass waste burnt at the temperature of 1080 °C would have the highest frost resistance.

The forecasted frost resistance of the samples is presented in Table 7.

According to calculation results, the maximum frost resistance (when the beginning of destruction reaches 1343 cycles and the end of decomposition is at 2188 cycles) is particular to samples that contain 15% of ground auto glass waste in the formation mixture and burnt at the temperature of 1080 °C.

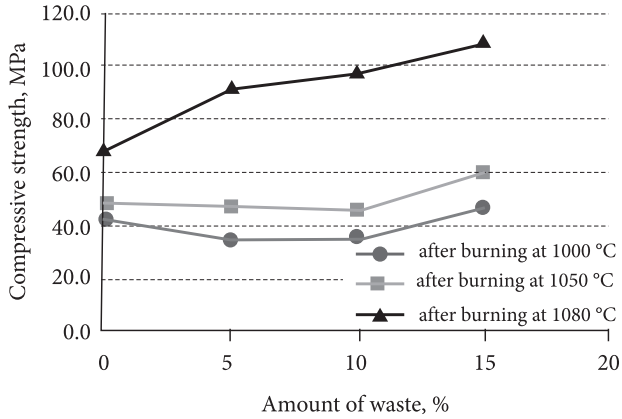


Fig. 9. The results of compressive strength, when density is 1850 kg/m3

Such significant increase in frost resistance as a parameter was predetermined by sufficient burning temperature and the maximum amount of fluxing additive. The glass melted at the temperature of 1080 °C, which decreased the amount of open pores and defects in the sample. As there is a little amount of burning additive in auto glass, it produces a large number of small pores, which is filled completely or partially with glass melted at sufficiently high burning temperature. Then, the relative thickness of pores and capillaries and the reserve of

Table 6. Average values of the derivatives of structural parameters

Mixture	*R*, %	*D*, %	G_1, g/cm^2	G_2, g/cm^2	*g*, g/cm^2	*N*, units
The highest burning temperature 1000 °C						
S1(0%)	28.61	2.2	0.59	0.52	0.98	0.250
S2(5%)	26.87	2.2	0.54	0.60	1.03	0.558
S3(10%)	27.64	2.1	0.67	0.59	1.01	0.260
S4(15%)	28.09	2.4	0.71	0.69	1.25	0.241
The highest burning temperature 1050 °C						
S1(0%)	38.32	2.2	0.522	0.550	0.78	0.485
S2(5%)	34.91	2.4	0.517	0.513	0.81	0.277
S3(10%)	42.14	2.4	0.486	0.546	0.68	0.265
S4(15%)	40.34	2.9	0.402	0.431	0.61	0.514
The highest burning temperature 1080 °C						
S1(0%)	49.22	3.6	0.174	0.239	0.24	0.579
S2(5%)	55.88	4.5	0.081	0.094	0.05	1.16
S3(10%)	54.45	4.9	0.187	0.245	0.05	0.692
S4(15%)	66.85	7.1	0.087	0.110	0.02	0.316

R – reserve of porous volume, %; *D* – relative thickness of the wall of pores and capillaries; G_1 – capillary mass flow rate in a vacuum in the direction of cooling, g/cm^2; G_2 – capillary mass flow rate in a vacuum perpendicular to the direction of cooling, g/cm^2; *g* – capillary mass flow rate determined under normal conditions, g/cm^2; *N* – structural inhomogeneity, units.

Table 7. The forecasted frost resistance of the samples

Mixture	The beginning of decomposition, in cycles	The end of decomposition, in cycles	The beginning of decomposition, in cycles	The end of decomposition, in cycles	The beginning of decomposition, in cycles	The end of decomposition, in cycles
	1000 °C		1050 °C		1080 °C	
S1(0%)	21	33	28	58	81	145
S (5%)	16	28	31	54	204	353
S3(10%)	20	34	47	90	203	406
S4(15%)	23	37	40	71	1343	2188

pore space increase, while open porosity, water absorption and capillary mass flow rate decrease. Such a change of structural parameters causes a very high frost resistance and compressive strength.

Conclusions

It was determined that ground auto glass waste can be successfully used in the production of sintered ceramics. The optimum amount of waste in formation mixture is 15%, the burning temperature is 1080 °C for the duration of 4 h. The water absorption of such ceramics amounts to 1.2%, and the forecasted frost resistance at the start of decomposition is 1343 cycles and at the end of decomposition it amounts to 2188.

1. Addition of 15% of ground glass into formation mixture and burning of samples at 1080 °C reduces the porosity of the samples by approx. 50% and increases the compressive strength by 14%, density by 10% and the reserve of pore space by 36%.

2. Compressive strength largely depends on the amount of ground auto glass in the formation mixture (R = 0.858–0.998). Burning samples at the temperature equal or below 1050 °C, compressive strength varies parabolically depending on the amount of waste used; burning of samples at the temperature of 1080 °C, the compressive strength starts to increase exponentially with the increasing amount of glass in the mixture.

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