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The Effect of the Addition of Construction & Demolition Waste on the Properties of Clay-based Ceramics

Biljana Angjusheva[1*)], Vilma Ducman[2], Emilija Fidanchevski[1]
[1]Ss. Cyril and Methodius University in Skopje, Faculty of Technology and Metallurgy, Skopje, Republic of North Macedonia
[2]Slovenian National Building and Civil Engineering Institute, Ljubljana, Slovenia

Abstract:
Waste glass and reclaimed brick are types of construction and demolition waste (C&DW) that could potentially be used as secondary raw materials in the production of ceramics. Ceramics based on clay, waste demolished brick (5-15 wt.%) and waste glass (5-20 wt.%) were produced by pressing (P = 68 MPa) and subsequently sintered at 900, 950, 1000, and 1050 ^{o}C for one hour. The physical and mechanical properties of the ceramics obtained were evaluated. The addition of demolished brick decreased the density and mechanical properties of the clay specimens and increased the water absorption. The incorporation of waste glass improved the sintering behavior and its mechanical properties. The addition of 20 wt.% waste glass and 10 wt.% waste demolished brick into the clay matrix improved the flexural strength by up to 20.6 % and decreased the water absorption by up to 22 %. The approach presented promotes an opportunity to recycle construction and demolition waste into alternative resource materials, and represents a positive contribution to the environment.
***Keywords**: Recycling; Mechanical properties; Sintering; Microstructure.*

1. Introduction

Construction and demolition waste (C&DW) is defined as unnecessary and damaged products and materials that result from construction, renovation, demolition and other construction activities [1]. According to Eurostat, around 30 % of all waste generated in the EU 27 in 2018 could be attributed to construction and demolition waste. It consists of various materials including concrete, brick, wood, metals, plastic, glass, asbestos, and gypsum, with recycling levels varying across member states from 10-90 % [2]. With an estimated 550 thousand tons each year, CD&W is one of the largest sources of waste in the Republic of North Macedonia [3], but almost all of this is landfilled or ends up in wild dumps. It currently stands in the top five waste materials by quantity, and even more concerning is the fact that this annual quantity is expected to rise further in line with the economic development of the country.

When meeting recycling targets the management of construction and demolition waste (CDW) plays a crucial role for two reasons. On the one hand, there is a huge difference between the material composition of buildings (tiles, bricks, mortars, plaster board etc.) and civil infrastructure (e.g. roads, pipe networks), making potential recycling rates difficult. On the other hand, the waste is heavy and has a large volume, making it difficult or expensive to transport, so it is necessary to investigate every possible application. Researchers have reported for the possible quantification and management of construction waste [4-6], and

[*)] **Corresponding author:** biljana@tmf.ukim.edu.mk

solutions of waste utilization [7-9]. Substantial research has been carried out regarding the use of C&DW with cements and lime as binding agents [10,11], mortars [12,13] and aggregates [14,15]. Experiments conducted with three ceramic wastes as supplementary cementitious materials in Portland Blended Cements, in quantities of 8, 24 and 40 % by mass, showed that ceramic wastes from the construction and demolition process have pozzolanic activity, and it has been indicated that they should be classified as slow pozzolana [16]. Baroso and Pala [17] investigated the effect of incorporating construction and demolition waste into clay–based ceramic materials and showed that a mixture containing 20 % C&DW had the best chemical and mechanical properties. The authors also concluded from the semi-industrial trial of ceramic brick fabrication that no significant changes to the industrial procedure are needed, making construction and demolition waste a potential raw material.

Furthermore, the process undertaken to obtain glass is energy intensive, so it therefore needs to be recycled and reused. The varying chemical composition of glass is the main reason why most waste glass (WG) cannot always be remanufactured into glass products. According to Demir [18], waste glass in quantities of 2.5, 5 and 10 % can be added to clay bricks obtained by extrusion. It was concluded that the amorphous nature of waste glass enhanced the sintering action and significantly improved the compressive strength of the fired samples. Similarly, Shishkin et al. [19] also observed the beneficial influence of the addition of glass to illitic-type clay. The addition of glass (up to 15 %) reduced the sintering temperature from 900 to 860^{o}C, and more than doubled the compressive strength.

Another advantage of using inorganic glasses is that they can incorporate large amounts of heavy metal ions, chemically bonding them inside their inorganic amorphous network [20]. Various glassy systems have been shown to be suitable for producing glass ceramic products that are thermally and mechanically stable and exhibit good chemical durability [21].

The aim of this work is to analyze the possibility of producing ceramics (tiles) that utilize waste demolished brick and waste glass. Ceramics with different ratios of clay, demolished brick and waste glass were sintered at different temperatures. The results and discussion of this research open up potential for the production of ceramics (tiles) with properties comparable to those commercially produced.

2. Materials and Experimental Procedures

Commercial clay (C) from the eastern part of North Macedonia (the Vinica region) was used as a matrix for the production of ceramics with waste demolished brick (WDB) and waste flat glass (WG) incorporated into the mix. Demolished bricks and waste window glass from a local demolition site were used in this study.

A schematic depicting the process for the production of ceramics from clay and waste materials is presented in Fig. 1.

Firstly, the bricks were collected, manually separated from the mortar, hammer crushed into 2-3 cm sized-chunks, and then dry milled for 30 min. using a planetary mill. Waste glass was crushed and dry milled for 30 minutes in the planetary mill. The particle density of the materials was determined by the pycnometer method [22]. The particle size distribution of the investigated materials was determined on dry samples by laser granulometry (Laser Cilas 1090). Classical chemical analysis was used to estimate the chemical composition of the materials.

The morphology of the particles was recorded by scanning electron images using a JEOL JSM 5500LV.

Compression of the powders was performed by uniaxial pressing at a pressure of 68 MPa, using (2 %) water as a binder. Sintering of the pressed composites took place under the following conditions:

- T= 900, 950, 1000 or 1050°C,
- heating rate-10 °/min,
- isothermal treatment at maximal temperature for 1h, in an air atmosphere,
- cooling within the furnace.

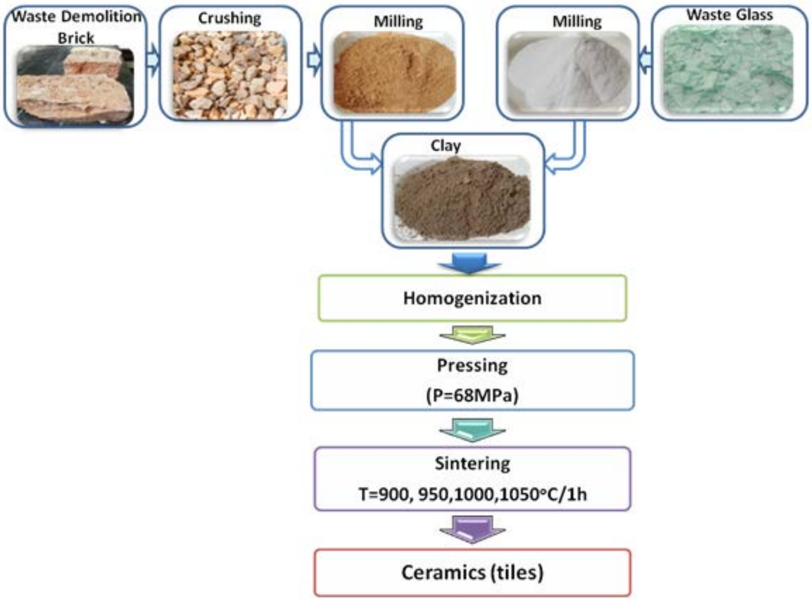

Fig. 1. Schematic outlining the production of ceramics (tiles).

The ceramics obtained were characterized with regard to their physical and mechanical properties. The density (D) of the ceramics (tiles) was measured by the immersion method, according to Equation (1). The apparent porosity (AP) was calculated according to Equation (2). The water absorption (WA) of the tiles was performed according to the method described in [23] and calculated according to Equation (3).

$$D = \frac{m_d}{(m_w - m_i)} \left[\frac{g}{cm^3}\right] \tag{1}$$

$$AP = \frac{(m_w - m_d)x100}{(m_w - m_d)} [\%] \tag{2}$$

$$WA = \frac{(m_w - m_d)x100}{m_d} [\%] \tag{3}$$

where m_w is the wet mass, m_d is the dry mass and m_i is the mass immersed in water.
Linear shrinkage ($\Delta L/L$) was calculated according to Equation (4), where l_d is the length of the dried test samples (mm) and l_f is the length of the sintered test sample (mm).

$$\frac{\Delta L}{L} = \frac{(l_f - l_d)x100}{l_d} [\%] \tag{4}$$

The flexural strength and E-modulus were estimated by a 3 point bending strength tester, model Netzsch 401/3, with a 30 mm span and 0.5 mm/min loading rate, using samples with dimensions of 5x5x50mm. For these mechanical tests at least three samples were evaluated for each type mentioned and the results of all the trials were averaged.

The mineralogical composition of selected sintered ceramic samples was determined by X-ray diffraction in a PANalytical Empyrean X-ray diffractometer equipped with CuKα radiation at λ = 1.54 A. Data were collected at 45 kV and a current of 40 mA, over the 2θ range from 4° to 70°.

The microstructure of selected sintered ceramic samples was assessed using scanning electron microscopy (SEM, Leica S440I) on samples with gold-coated surfaces.

Porosity and pore size distribution in the pore size range from 360 μm to 0.003 μm was determined by mercury intrusion porosimetry (MIP; Micromeritics) by applying pressure up to 414 MPa.

The identification labels and compositions of the samples investigated are presented in Tab. I.

Tab. I Composition and identification labels of the clay matrix (C) samples with waste demolished brick (WDB) and waste glass (WG).

Sample	C [wt.%]	WDB [wt.%]	WG [wt.%]
A	90	5	5
B	70	15	15
E	70	10	20

3. Results and Discussion

Chemical composition of the investigated materials

The chemical compositions of the clay (C), waste demolished brick (WDB) and waste glass (WG) are presented in Tab. II.

Tab. II Chemical compositions of clay (C), waste demolished brick (WDB) and waste glass (WG).

Material	**Oxide, [wt.%]**								
	SiO_2	**Al_2O_3**	**Fe_2O_3**	**CaO**	**K_2O**	**Na_2O**	**SO_3**	**MgO**	**LOI**
C	58.79	13.60	9.28	3.59	2.51	3.83	0.18	1.72	5.79
WDB	61.12	17.58	8.45	7.82	0.29	0.33	/	3.01	0.86
WG	71.50	0.31	0.31	8.96	0.19	9.50	0.37	3.62	/

The clay is rich in SiO_2 and Al_2O_3 (with a lower content of CaO, MgO, Na_2O, K_2O), accompanied by 9.28 wt.% Fe_2O_3. The alkaline and earth alkaline oxides are considered fluxes, which can influence the densification behavior of ceramic materials during firing [24]. The content of iron (III) oxide is also an important factor which, due to the bloating mechanism at higher sintering temperatures, could influence the physico-mechanical properties of the sintered ceramics [25]. In terms of oxides, the WDB basically consists of SiO_2, CaO, $Al_2O_3$and Fe_2O_3, with minor contents of MgO, K_2O and Na_2O. The relatively high CaO content (7.82 wt.%) could be explained by the presence of cement in the waste material. It has been reported that separation of the clay bricks from the mortar is preferred, since the cement can affect the mechanical properties of the final ceramic product [26].

Granulometry and morphology of the investigated materials

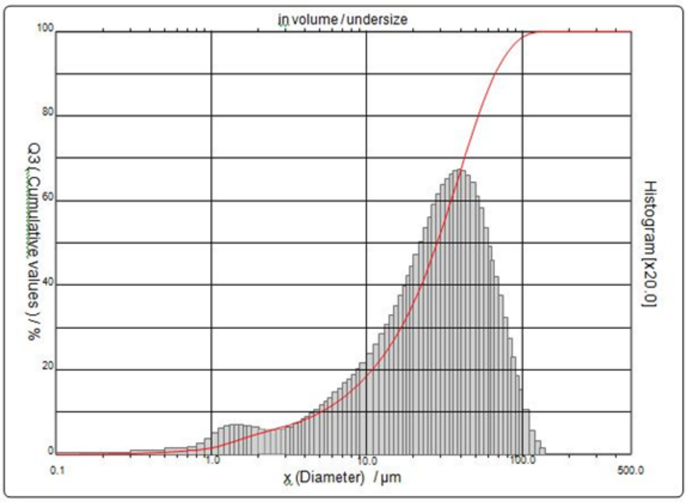

Fig. 2. Granulometric composition of (a) clay, (b) WDB, and (c) WG.

The granulometric compositions of clay, waste demolished brick and waste glass (Fig. 2) show that the clay and WDB both have a bimodal particle size distribution, while WG has an almost unimodal distribution. In clay the first interval of the particle size distribution is from 0.1 to 5µm, and the second from 5 to 110 µm. In WDB it ranges from 0.1 to 5µm in the first interval and from 5 to 90 µm in the second. The range for WG is 0.3 to 105 µm. The specific gravities and the characteristic diameters at 10, 50, and 90 wt.% are presented in Table III. Specific gravities of the raw materials correspond to values presented in the literature [26].

Tab. III Diameter of the particles at 10, 50, and 90 wt.% and specific gravity of the raw materials.

Sample	**D_{10} [µm]**	**D_{50}[µm]**	**D_{90}[µm]**	**Specific Gravity [g/cm^3]**
C	1.89	18.11	58.75	2.70
WDB	1.43	12.33	41.43	2.65
G	5.01	28.30	66.86	2.66

Fig. 3 shows the typical ilite morphology of clay. The presence of round grains and particles with irregular geometry are evident, as well as small grains of aggregate. The morphology of the WDB and WG reveals sharp grain edges with varying geometry and dimensions, noticeable especially in the WG.

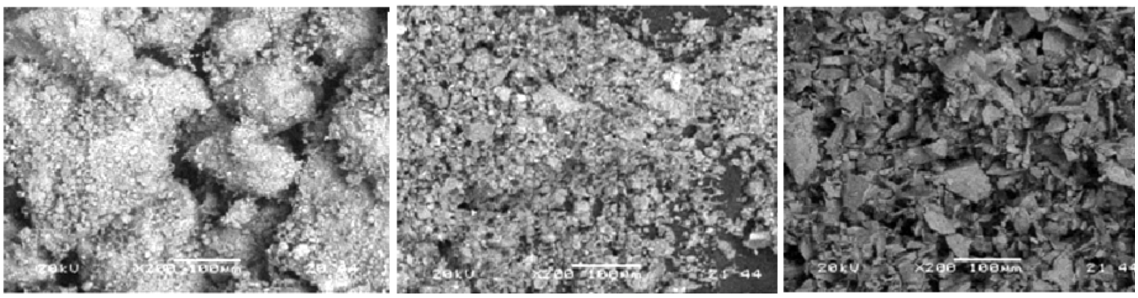

Fig. 3. SEM micrographs of a) clay, b) WDB, and c) WG.

Physical and mechanical properties of the ceramic tiles

The dependence of the density, porosity, water absorption and linear shrinkage of the ceramics on the WDB and WG content and the sintering temperature is shown in Figs. 4-7.

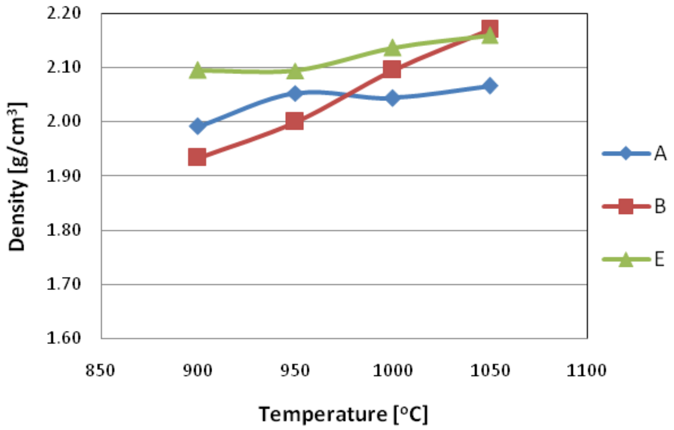

Fig. 4. Density of ceramics sintered at defined temperatures.

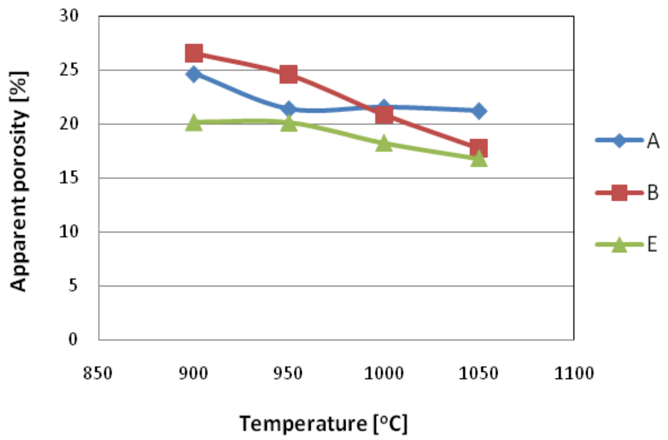

Fig. 5. Apparent porosity of ceramics sintered at defined temperatures.

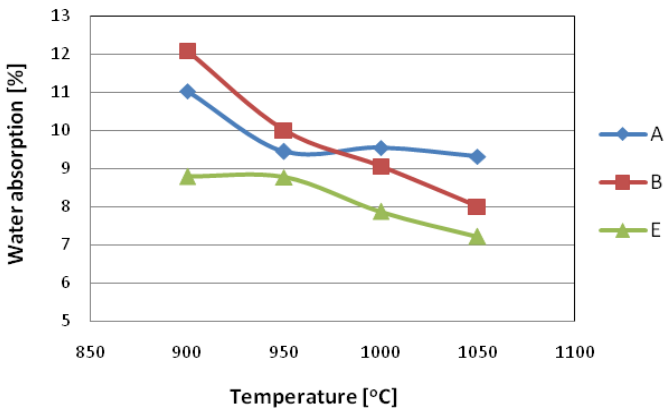

Fig. 6. Water absorption of ceramics sintered at defined temperatures.

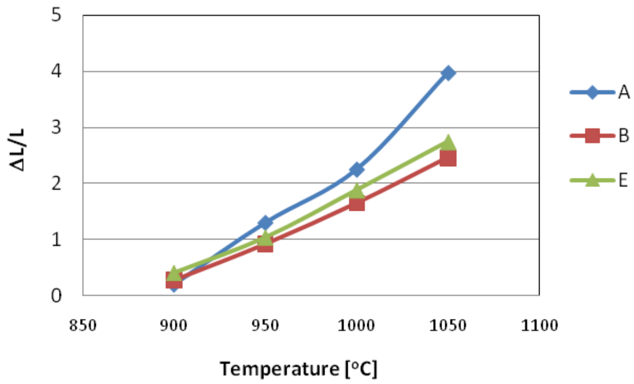

Fig. 7. Linear shrinkage of ceramics sintered at defined temperatures.

The graph showing the density of the ceramics investigated (Fig.4) shows that the influence of sintering temperature is comparable across all the ceramic samples, regardless of their composition. It can, however, be concluded that the E ceramics have the highest density (2.09÷2.16 g/cm^3) at all sintering temperatures. It is clear that the addition of WDB increases the apparent porosity at lower sintering temperatures (Fig. 5). In the B ceramic tiles the construction waste content is 30 wt.%, while in tiles A the content is 10 wt.%, with an equal ratio of WDB and WG. In both the E and B tiles the content of demolition waste is 30 wt.%, but the ratio of waste demolished brick (WDB) to waste glass (WG) is different. It can be seen that the addition of glass decreases the apparent porosity and water absorption of ceramics tiles at all sintering temperatures. The reduction in water absorption in line with an increased sintering temperature is related to the reduction of open porosity in the fired bodies. In Figure 6 it can be seen that at 950^{o}C the water absorption values varied from 12 % in tile sample B to 8.5 % in tile sample E, while at 1050^{o}C it ranged from 9.2 % in ceramic tile A to 7.2 % in tile sample E. This proves that the addition of glass (20 wt.% in tile E) decreases water absorption by around 22 % (22.7 % at the lowest sintering temperature and 21.7 % at the highest). Pontikes et al. [27] reported that using 30 % glass as a material substitution in a traditional roofing tile mixture improves the water absorption and strength of ceramics. An investigation into linear shrinkage (Fig. 7) shows that all compositions display relatively similar shrinkage behavior at lower sintering temperatures. As the sintering temperature rises, so does the shrinkage, in all compositions investigated. The highest linear shrinkage (4 %), is seen in ceramic A at a sintering temperature of 1050^{o}C, which relates to the higher content of clay (90 wt.%), which has high plasticity. Sintered ceramics B and E show similar shrinkage behavior at all sintering temperatures, which can be attributed to the non-plastic character of the pre-sintered materials, since WDB act as inert materials.

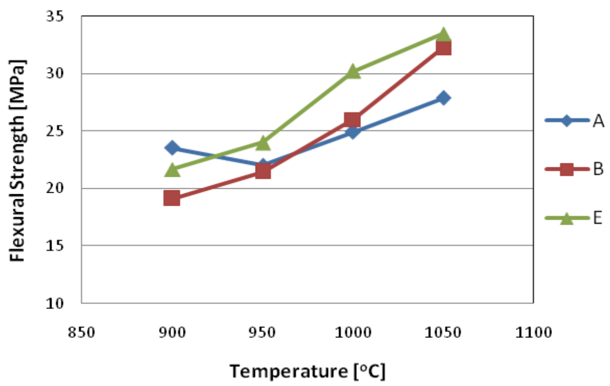

Fig. 8. Flexural strength of ceramics sintered at defined temperatures.

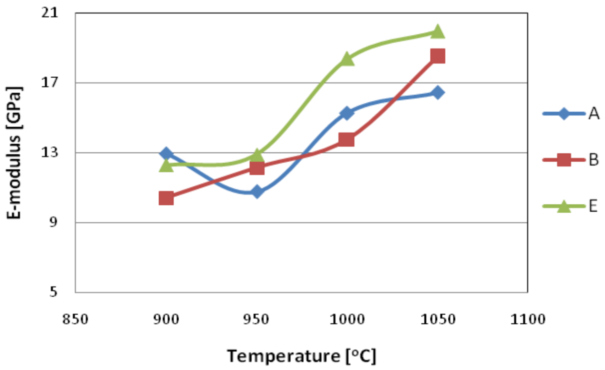

Fig. 9. E-modulus of ceramics sintered at defined temperatures.

From Figs 8 and 9 it is evident that the flexural strength and E-modulus increased as the sintering temperature increased. At lower sintering temperatures (900°C), it is observed that ceramics tiles A have slightly higher values of flexural strength, which may be due to the inert nature of the added waste. At temperatures of 950°C and above, the flexural strength and E-modulus were affected by the addition of glass; a significant improvement was evident in tiles E compared to tiles A, due to the presence of fluxing agents (CaO and Na_2O) in the waste, as well as the presence of a liquid phase formed at higher temperatures. At 1050°C, the improvement in flexural strength and E-modulus is 20.6 and 25 %, respectively. This is higher than the values reported by Acchar et al. [28] for clay-based ceramics containing various quantities of C&DW.

European standard EN14411 [23] establishes the classification of tiles: Ceramic tiles are divided into groups according to their method of manufacture and their water absorption. Type A represents extruded tiles, while type B are dry-pressed tiles. There are three main groups in relation to water absorption: tiles with low water absorption (Group I), WA $\leq$ 3 %, tiles with medium water absorption (Group II), 3 % $<$ WA $\leq$ 10 %, and tiles with high water absorption (Group III), WA $>$ 10 %. Relevant to the present study are tiles of type BIIb, which require a WA of 6-10 % and a flexural strength of 18 MPa, and type BIII, which require $\geq$ 10 % WA and a flexural strength of 15 MPa. According to the data obtained in the actual investigation, ceramics E (WA: 7-9 % and flexural strength of 21-34 MPa) obtained at all applied sintering temperatures can be classified as BIIb; ceramics B sintered at 1000 and 1050°C can also be classified as BIIb (WA: 8 and 9%; flexural strength: Greater than the prescribed requirement of 18 MPa), while the samples sintered at 900 and 950°C can be classified as BIII (WA: 12 and 10 %; flexural strength:18 and 22 MPa). Ceramics A sintered at 950, 1000 and 1050°C belong to class BIIb (WA: <10%; flexural strength: 21-34 MPa),while the samples sintered at 900°C are classed as BIII (WA: 11 %; flexural strength: 24 MPa).

Ceramics E satisfy the criteria for BIIb, thus allowing sintering at a lower temperature, which would reduce the energy requirement to produce the tiles, while at the same time incorporating a significant amount of waste materials.

Sample E, which was recognized as a product with potentially optimal properties in terms of water absorption, was additionally analyzed for its microstructural properties. Namely, the addition of waste influences the sintering and mineralogy of samples fired at different temperatures, which is reflected in its bulk properties. Fig. 10 presents the XRD diffraction patterns for samples E sintered at 950 and 1050°C, as well as for each additive. As expected, waste glass (WG) contains no minerals, only amorphous phase, while quartz (SiO_2) and anorthite ($CaAl_2Si_2O_8$) are detected in the WDB sample. WDB, which represents fired bricks, is expected to resemble sample E, which is based on fired clay with the addition of WG and WDB. In sample E, fired at 950°C, the main phases identified are quartz (SiO_2), anorthite ($CaAl_2Si_2O_8$), nepheline ($Na_3KAl_4Si_4O_{16}$) and gehlenite (($Ca_2Al[AlSiO_7]$). The same phases are also present after firing at 1050 ^{o}C, but after firing at a higher temperature the main peak corresponding to quartz (Q) starts to decrease, which, one part of the reason, could be ascribed to the onset of the eutectic melting of quartz in the presence of waste glass (WG), and on the other to the increased formation of anorthite (A), whose peaks increased noticeably.

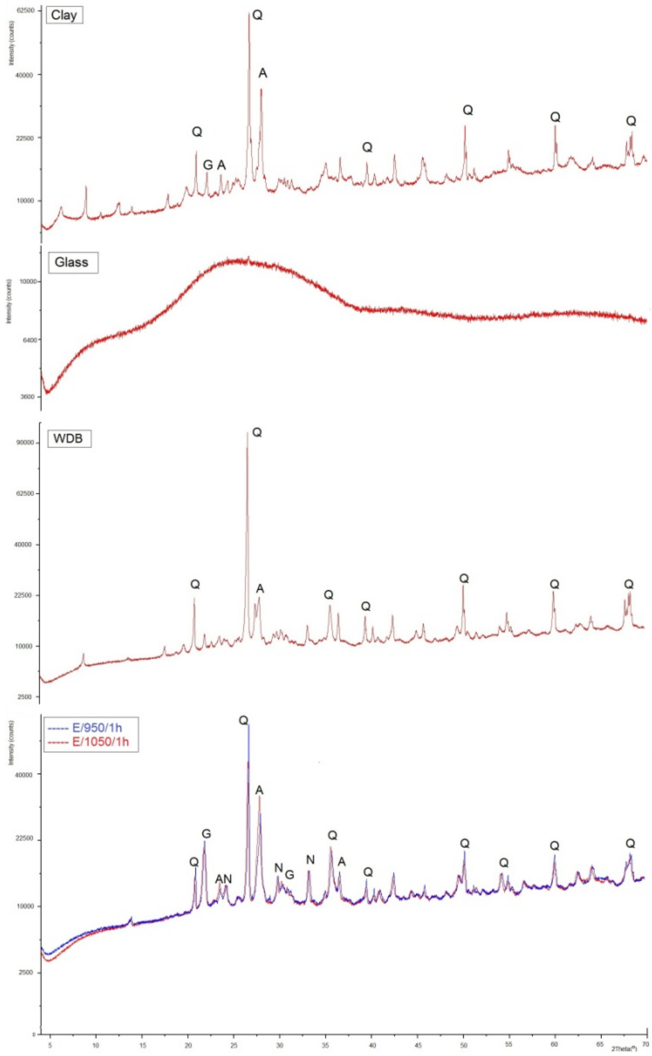

Fig. 10. XRD diffractograms of the raw materials and samples E/950º/1h and E/1050º/1h (Q-Quartz, A-Anorthite, N- nepheline, G-Gehlenite).

Microstructure of the ceramic tiles

Fig. 11 shows the microstructure of the polished surfaces of ceramic samples E sintered at 950 and 1050^{o}C. The grains of waste material in the WDB (with a size of up to around 100 µm) are clearly seen, and are relatively well dispersed in the clay matrix after firing at 950^{o}C. The microstructure of sample E sintered at 1050^{o}C is smoother and more homogeneous, with a lower percentage of pores (15 % porosity) compared to a sample of the same composition sintered at 950^{o}C (20 % porosity). Larger magnifications more clearly show the presence of the liquid phase from the glass in the ceramic tiles.

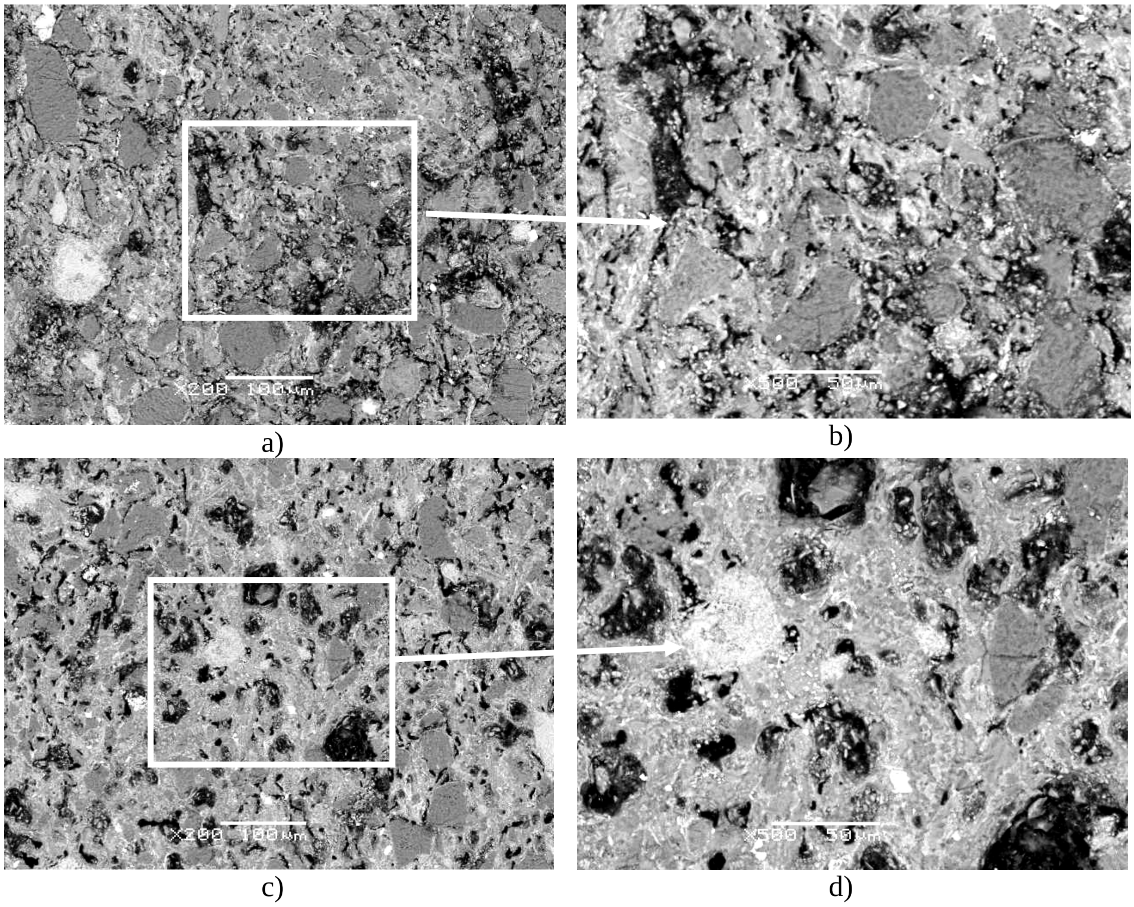

Fig. 11. SEM micrographs of E/950º/1h (a and b) and E/1050º/1h (c and d).

In order to better analyze the porosity in terms of pore size distribution, MIP analysis was performed on the same samples that were analysed by SEM. The overall porosity in sample E, as determined by means of MIP, amounted to 26.7 and 24.2 % for samples fired at 950 and 1050^{o}C, respectively. The porosity determined by means of MIP is higher than that obtained by water absorption, because MIP can measure a pore size as low as 3 nm. Pore size distribution is presented in Fig. 12. In accordance with the sintering behavior expected (the densification and coarsening of grains and pores), the overall porosity decreased by firing at a higher temperature, while pore size distribution (Fig. 12) shifted towards higher values; while the peak for the sample fired at 950^{o}C is around 2 μm, it is around 4 μm in the sample fired at 1050^{o}C.

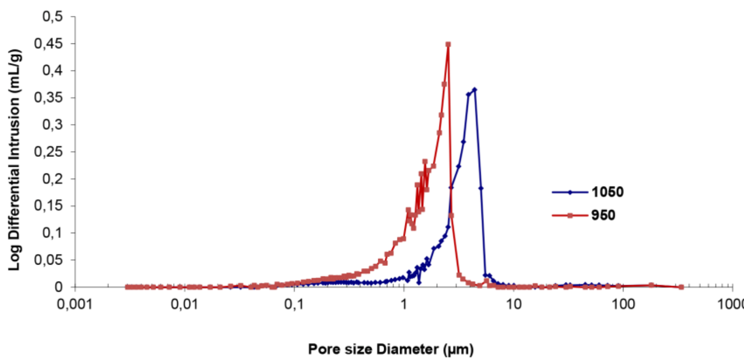

Fig. 12. Pore size distribution of samples a) E/950º/1h, and b) E/1050º/1h.

4. Conclusion

The investigation undertaken has demonstrated that C&DW can be successfully used in clay-based ceramics. Generally, the addition of waste demolished brick reduces the physical and mechanical properties of the ceramics investigated. The addition of waste glass has a beneficial role in the sintering behavior of ceramics i.e. sinterability was improved by increasing the glass content. Namely, the addition of glass decreases the porosity and water absorption of the tiles produced. As presented in the results, higher temperatures result in better sintering and consequently better properties: lower porosity and water absorption, and consequently improved mechanical properties. Linear shrinkage increases with temperature but decreases slightly with the addition of waste glass.

The results presented are in accordance with other studies related to the incorporation of C&DW into the ceramic matrix, which conclude that waste glass and demolished brick can be used as alternative raw materials in ceramic building materials, thus promoting an increase in the technological properties of the sintered materials. Considering all the results it can be concluded that the E ceramics, composed of 10 wt.% WDB and 20 wt.% waste glass, showed the highest density, lowest water absorption and highest mechanical properties at all temperatures investigated, thus satisfying the criteria for BIIb ceramic tiles. The ceramics produced from different contents of clay and C&DW can be classified as either BIIB or BIII tiles, according to the water absorption and flexural strength obtained.

The use of C&DW in practice to produce clay-based ceramics (tiles) might be an innovative approach to reduce the volume of C&DW at disposal sites, while also enabling the preservation of natural resources.

Acknowledgments

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Сажетак: *Отпадно стакло и регенерисана цигла су врсте грађевинског отпада и отпада од рушења се потенцијално могу користити као секундарне сировине у производњи керамика. Керамике на бази глине, регенерисане цигле (5-15 wt.%) и отпадног стакла (5-20 wt.%) су добијене пресовањем (P=68 MPa) и синтеровањем на 900, 950, 1000 и 1050°C током једног сата. Одређена су физичка и механичка својства добијених керамика. Додатак цигле је смањио густину и механичка својства узорака на бази глине и повећао абсорпцију воде. Уградња отпадног стакла је побољшала синтеровање и механичка својства. Додатак 20 wt.% отпадног стакла и 10 wt.% цигле у матрицу на бази глине побољшали су отпорност на савијање до 20.6 % и смањили абсорпцију воде до 22 %. Овакав приступ пружа прилику да се отпадни грађевински материјал користи као алтернативни ресурни материјал, и представља допринос у очувању околине.*
Кључне речи*: рециклажа, механичка својства, синтеровање, микроструктура.*