Using glass wastes and bentonite to produce a new ceramic tile

Uporaba steklenih odpadkov in bentonita za izdelavo novih keramičnih ploščic

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Abstract

This paper discusses the recycling of glass waste as a compound in the production of ceramic tiles. The present study aims to investigate the effects of glass waste (with two different granulometries) on the physical and mechanical properties of ceramic tiles, in order to demonstrate their suitability for the production of ceramic materials. A series of ceramic tiles was prepared based on bentonite (B) by adding 70, 65, and 60 wt% glass wastes (GW1 and GW2 with two different granulometries) into the batch composition. The ceramic tiles were sintered at a constant temperature of 900 °C following the same production protocol. The physicalmechanical properties and chemical durability of all ceramic tiles produced were evaluated. The results show that ceramic tiles can be produced from a basic mixture of 35% bentonite and 65% glass waste, with good physical, mechanical, and thermal properties (the local Algerian bentonite is considered a clay binder and has excellent plasticity).

Keywords: glass waste, bentonite, clay, ceramic tiles, physical and mechanical properties

Introduction

The glass industry continues to develop and the outstanding properties of this material give it importance in the industrial sector. In

Povzetek

Članek obravnava recikliranje odpadnega stekla kot snovi pri proizvodnji keramičnih ploščic. Namen pričujoče študije je raziskati učinke steklenih odpadkov (dveh različnih porazdelitev velikosti delcev) na fizikalne in mehanske lastnosti keramičnih ploščic z namenom prikaza njihove primernost za proizvodnjo keramičnih materialov. Serija keramičnih ploščic je bila pripravljena na osnovi bentonita (B) z dodajanjem 70, 65 in 60 mas.% odpadnega stekla (GW1 in GW2 z dvema različnima porazdelitvama velikosti delcev). Keramične ploščice so bile sintrane pri konstantni temperaturi 900 °C po istem proizvodnem postopku. Ocenjene so bile fizikalno-mehanske lastnosti in kemijska obstojnost vseh izdelanih keramičnih ploščic. Rezultati kažejo, da je mogoče iz osnovne mešanice 35 % bentonita in 65 % steklenih odpadkov proizvesti keramične ploščice z dobrimi fizikalnimi, mehanskimi in toplotnimi lastnostmi (lokalni alžirski bentonit velja za glineno vezivo in ima izvrstno plastičnost).

Ključne besede: stekleni odpadki, bentonit, glina, keramične ploščice, fizikalne in mehanske lastnosti

recent times, the use of waste glass in industry is very high because of its great physical and mechanical properties. For the interest of eliminating environmental pollution, the glass waste is used to develop new material

Received: February 05, 2023 Accepted: April 06, 2023 (glass foam, ceramic ...) [1, 2]. The waste glass must, however, avoid excessive volumes of waste. Glass is an important constitution of ceramic parts because it is formed during the vitreous phase with the sintering reaction. In this context, studies have been conducted about the possibility of using recycled glass as a raw material in the manufacturing of ceramic products [3]. Glass waste can also be used to prepare glass-ceramics materials through controlled crystallization of the glass phase [4]. The major crystalline phases of these materials are calcite, muscovite, and quartz [5]. These ceramics exhibit good physical and mechanical properties, thus are normally used for construction [6-7]. However, not many attempts to compose new ceramics based on the mixture of glass waste and clay have been reported. In our earlier studies [8–9], it has been shown that these new ceramics can be formed with higher density, higher hardness, lower drying shrinkage, less water absorption, and other good physical qualities [7–10].

Glass waste is a major contribution to the municipal solid waste collection. As has been demonstrated by other researchers [11-13], it can be used as a fluxing agent in ceramic production such as stoneware, tiles, bricks, concrete, and cement. It can also reduce the sintering temperature when mixing with clay [14]. Consequently, it appears to be a suitable raw material for ceramic bodies since it contains SiO2, CaO, Na2O, and Al2O3 [15]. Due to their distinctive characteristics, these properties are mainly due to their covalently bonded structure [16]. With close to ideal frictional behaviour, excellent wear resistance, high hardness, and outstanding durability, ceramics are materials with a wide range of engineering applications. At high temperatures, ceramic products are divided into inorganic solid and non-metallic states. This industry can provide a viable substitute for the inclusion of glass waste as a replacement for the natural resources currently used in ceramic production, as ceramic compounds are produced at high temperatures for use in various applications. The main problem with this application is that it consumes a lot of energy and is therefore not environmentally friendly. Although the use of waste glass is possible in different areas of ceramic production, the use of this type of waste in the production

of ceramics can excellently improve the quality of ceramics [17].

The present study aims to investigate the effect of glass waste (with two different granulometries) on the physic-mechanical properties and chemical durability of ceramic tiles to demonstrate its suitability for ceramic materials production. A series of ceramic tiles was prepared based on bentonite (B) by adding 30, 35, and 40 wt% glass wastes (GW1 and GW2 with two different granulometries) into the batch compositions. The ceramic tile sintering was performed at kept temperature 900 °C while applying the same production protocol. The physic-mechanical properties and chemical durability of all obtained ceramic tiles of were evaluated.

Experimental Study

Raw material characterisation

To produce ceramic tiles, the raw materials used were bentonite (B), Glass Waste 1, and Glass Waste 2. The chemical composition and granulometries and thermal analyses (Thermogravimetric Analysis and Differential Scanning Calorimetry) are represented respectively in the tables and figures.

The chemical composition of raw materials

As for the chemical composition of the bentonite obtained (Table 1), the silica content is very high (about 55.94%), resulting in a mass ratio SiO2/Al2O3 = 4.12; this high value indicates the presence of a large amount of free silica in the clay fraction. The content of alumina Al₂O₂, which gives stability and hardness to its structure, is also important. The bentonite also contains a medium amount of CaO and MgO, and the magnesium and calcium may be part of the structure in the intermediate layers of montmorillonite (the main component of bentonite). The CaO and MgO contents also indicate the presence of calcite CaCo3 and dolomite $MgCa(CO_{a})$, as the CaO contents are more important than the Na2O contents, and it can be said that the bentonite used is a calcium type (less puffed than the sodium bentonite) [18]. The Fe2O content is 3.14%< 6%, so this clay is intended for fine ceramics [19].

For glass waste, the SiO2 content is over 70% for the types used. However, the Na2O content is 11% to 13% and the CaO content is 8% to 11%, so it can be assumed that our glass waste is sodium-calcium glass [20].

Particle size analysis (Laser granulometric analysis)

The results of laser analysis of particle sizes are shown in (Figure 1). From the granulometric curve, it can be seen that the GW2 curve has smaller diameters than the GW1 curve. It was also found that the maximum diameter corresponding to a cumulative volume of all materials used is 10% D (0.1), 50% D (0.5) and 90% D (0.9). The results of the maximum diameter are shown in Table 2 below:

From the results of Table 2 and Figure 1, it was noted that the glass waste 2 (GW2) shows a higher amount at D (0.5), which suggests a

Table 1: Chemical composition of raw material

strong agglomeration of this particles which we will consider to be the consequence of poor dispersion of the suspension.

The bentonite grains present a diameter superior to $87 \mu m$. Thus, the addition of bentonite increases the pores and defect sizes and promotes diffusion.

Thermogravimetric analysis and differential scanning calorimetry

The Differential Scanning Calorimetry curve (see Figure 2) shows an endothermic peak between 600 °C and 800 °C, located at 768.3 °C, indicating the decomposition of the glass waste and the raw material consisting of silicates, which can be confirmed by the lower mass of about 38.59% in this temperature interval.

The mass loss between 100 °C and 300 °C indicates evaporation of water from the surface (4.92%), which is due to the elimination of

Chemical composition %	Fire loss	SiO ₂	Fe ₂ 0 ₃	Al ₂ 0 ₃	Ca0	Mg0	SO ₃	K ₂ 0	Na ₂ 0	P ₂ O ₅	Ti
Bentonite	14.16	55.94	3.14	13.57	5.91	1.74	0.41	2.19	2.47	0.15	0.33
Glass waste	2.67	70.26	0.99	0.44	11.79	1.68	0.21	0.52	11.43	0.01	0.02



Figure 1: Granulometric distribution of used materials (bentonite, Glass Waste 1 and Glass Waste 2).

water from the clay mineral (see Figure 2). The endothermic peak between 400 °C and 700 °C, associated with a mass loss of about (6.96%), is related to the dihydroxylation of phyllosilicates. The exothermic peak at 1082 °C with an energy release of 2.716 mw/mg and a mass loss of 11.87% corresponds to the fusion of bentonite.

The Thermogravimetric Analysis indicates that a liquid phase formed, contributing to the fusion of the clay (bentonite). We hypothesise that this change is related to a sintering mechanism in the liquid phase. This mechanism involves the dissolution of the solid material in the liquid phase and the growth of the crystal

Table 2: Maximum diameter of raw material

Diameter Max (µm)	D (0.1)	D (0.5)	D (0.9)
Bentonite	17.2	84.454	278.142
Glass Waste 1	0.536	3.919	142.64
Glass Waste 2	5.735	53.713	168.7

phases. The closed pores are formed by the trapped gas and the micropores are formed by the crystallisation process.

Elaborations of the ceramic tile samples

Mixing of the glass waste and bentonite to form the tiles into rectangles (100 mm \times 100 mm \times 10 mm) was carried out at a mould humidity of 6% and a pressure of 100 bar, followed by drying and thermal treatment at 900 °C for 20 minutes. The physical-mechanical properties were determined using five samples for each mixture to obtain the average of the data. Bulk density, water adsorption and porosity, linear shrinkage, and flexural strength were quantified according to ASTM standard methods [21–23]. Four-point flexural strength was measured using a universal testing machine according to the standard procedure [24].

Ceramic tile mixing details: The chemical composition of the ceramic tile samples is shown in the following table.



Figure 2: Thermogravimetric Analysis and Differential Scanning Calorimetry Curves of raw materials (Bentonite and Glass Wastes).

Symbol	Glass waste Type	Glass waste (% by Mass)	Bentonite (% by Mass)	Shaping Humidity (% by Mass)
GB ₃₀	1	70	30	6
GB_{35}	1	65	35	6
GB_{40}	1	60	40	6
$G'B_{30}$	2	70	30	6
$G'B_{35}$	2	65	35	6
$G'B_{40}$	2	60	40	6
B_{100}	-	00	100	8

Table 3: Initial parameters of the prepared ceramic tiles.

Results and Discussion

Manufacturing of all studied ceramic tiles

Two series of ceramic tiles based on waste glass (GW) with bentonite content of 30%, 35%, and 40% were produced. GW-based ceramics were used in two different particle sizes (GW1 and GW2) with 70wt%, 65wt% and 60 wt% by substitution of bentonite (30%, 35% and 40 %) (see Table 3).

Figure 3 shows the macroscopic aspect of the ceramic tiles produced on the basis of GW1 (tile samples GB30, GB35 and GB40). The second series of ceramic tiles based on GW2 is shown in Figure 4. GW1 was replaced by GW2, and we changed the granulometry of the glass waste and followed the same steps to produce the tiles. Starting from 30% bentonite, we obtained the tile samples G'30, G'35 and G'40 (see Figure 4). To compare the effect of the glass wastes on the properties of ceramic tiles, another batch at 100% bentonite called B was elaborated and later characterized (see Figure 5).

The temperature required to produce a tile of 100% bentonite is about 1100 °C, which is about 200 °C higher than for tiles produced of 30, 35 and 40% bentonite, due to the rigid structure of bentonite and the covalent bonds between the structural units of the network of silica and alumina. This requires a high sintering temperature, and in this case, it shows that the addition of glass waste can produce ceramic tiles with lower time and temperature requirements. The glass waste plays the role of the vitreous phase during sintering, and we combine it with bentonite, which plays the role of the binder. In this way we can obtain a good ceramic at a low temperature of 900 °C.



Figure 3: Ceramic tiles obtained from 30% (a), 35% (b) and 40% (c) of bentonite content with Glass Waste 1 (GW1).

Characterisation of the elaborated ceramic samples

Determination of certain properties of the obtained tiles corresponding to the composition of 30, 35 and 40 mass percent bentonite in the mixture of glass waste-bentonite was



Figure 4: Ceramic tiles obtained from 30% (a), 35% (b) and 40% (c) of bentonite content with Glass Waste 2 (GW2).

prepared with two different particle sizes of glass waste (GW1 and GW2). The same work was carried out for the tile made of 100% bentonite with the designation B.

The results of the characterization of the tiles made from GW1 and GW2 are presented in the following Tables 4 and 5 respectively.

Physical properties

Humidity: The level of humidity remains almost the same for all tiles produced with GW1 and is estimated to be around 6% (see Figure 6). For GW2, it varies depending on the amount of bentonite added; it increases with each addition of bentonite and is significantly higher for sample B (with 100% bentonite), which requires more moisture for its moulding.

Linear shrinkage: The dilation of sample GB30 is greater the more the content of glass waste slightly favours the expansion of the vitreous phase. It can be observed that this dilation is better for the GB30 variant (see Figure 7). For the GB35 and GB40 ceramic samples, it is observed that the shrinkage decreases with increasing glass waste content. The decrease in thermal shrinkage can be attributed to devitrification of the waste glass, which occurs with increasing temperature. This process causes all particles to move toward the centre, reducing the size of the samples [25]. In GW2, the tiles shrank, and a reduction in shrinkage was observed several times with the addition of bentonite. This is due to the plastic nature of bentonite. The optimum shrinkage value is 4.78% for composition B with 100% bentonite.



Figure 5: Ceramic tile samples at 100% bentonite (B) content at 1100 °C: (a) Face 1 and (b) Face 2.

Proprieties/tiles	GB30	GB35	GB40	B100
Humidity (%)	6.83582	6.89445	6.57	11.7428
Shrinking (%)	3.916	-7.182	-5.8492	-4.78
Mass loss (%)	15.417	4.475	4.78	8.4946
Density (g/cm ³)	1.542	2.078	2.02	2.04
Mass specific (g/cm ³)	2.33278	2.2003	2.3371	2.34
Absorption (%)	22.182	2.6598	6.57	6.18
Porosity (%)	33.904	5.525	13.3	12.62
Resistance flexion (MPa)	19.2444	32.6762	24.6616	14.2101
Breaking strength (N)	714	729	1062	500
Hardness (HV)	90	188	153	208

 Table 4: Properties of ceramic tile samples – Glass Waste 1 and bentonite (GB).

 Table 5: Properties of ceramic tile samples - Glass Waste 2 and bentonite (G'B)

Properties/tiles	G'B30	G'B35	G'B40	B100
Humidity (%)	6.1809	5.79	7.4779	11.7428
Shrinking (%)	-7.08	-6.097	-5.335	-4.78
Mass loss (%)	4.3195	4.64	5.042	8.4946
Density (g/cm3)	2.1325	2.09	2.065	2.04
Mass specific (g/cm3)	2.1605	2.29	2.36	2.34
Absorption (%)	6.5625	4.29	6.07	6.18
Porosity (%)	13.975	8.9625	12.53	12.62
Resistance flexion (MPa)	29.9578	28.844	31.734	14.2101
Breaking strength (N)	553	742	743	500
Hardness (HV)	159	186	167	208



Figure 6: Variation of humidity as a function of bentonite content (G: Glass Waste 1; G': Glass Waste 2).



Figure 7: Variation of shrinking as a function of bentonite content (G: Glass Waste 1; G': Glass Waste 2).



Figure 8: Weight loss of elaborated ceramic tiles as a function of bentonite content (G: Glass Waste 1; G': Glass Waste 2).

Weight loss: There is a mass loss proportional to the bentonite content (see Figure 8). This is due to the elimination of water formed by the clay, which is significantly higher with each addition of bentonite. However, it decreases to a minimum from variant GB35 and then increases slightly with variant GB40. The combination of the two vitreous and crystalline phases reinforces the structure. This is also true for GW2. The mass loss is greater for variant (B), which is due to the higher water mass in the bentonite, which disappears after baking.

Bulk density and specific mass: The value of density and specific mass increases with the addition of bentonite, since a filling of the structural units is favoured by the modifying ions produced by the clay when Glass Waste 1 (GW1) with a fine granulometry is used, and for GW2 the density and specific mass decreased when we used a finer glass waste (GW1), so we



Figure 9: Variation of bulk density (a) and specific mass (b) of elaborated ceramic tiles as a function of bentonite content (G: Glass Waste 1; G': Glass Waste 2).

can say that the size of GW2 has an influence on the compaction of the tiles (see Figure 9). This was less important because the larger glass waste requires a longer baking time during sintering. Also, more voids remain inside the produced tiles.

Porosity and water absorption: For GW1, the porosity and water absorption have decreased due to the compaction of the tiles and the recrystallization of the processed products.

The optimum values are obtained for the GB35 variant (5.525% porosity and 2.625% absorption) (see Figure 10). However, porosity and water absorption increased for GW2 compared to the ceramic tiles produced with GW1. For the sample with 100% bentonite, the porosity is 12.6% and the water absorption is 6.18%. The pores become narrower with decreasing content of glass waste. This phenomenon is caused by the formation of the vitreous phase, which



Figure 10: Variation of porosity (a) and water absorption (b) of elaborated ceramic tiles as a function of bentonite content (G: Glass Waste 1; G': Glass Waste 2).

causes progressive filling of the pores and contraction of the ceramic material [26].

Mechanical proprieties

Flexural resistance and breaking strength: The value of the mechanical resistance to flexion increases with the addition of bentonite and reaches the optimum value of 32.71 MPa for variant GB35 (see Figure 11), after which it decreases. This decrease correlates with the structural strengthening due to the combination of the two structures, vitreous and crystalline, where the alumina introduced by the clay improves the mechanical strength of the mixture. The same behaviour is observed for the required breaking strength when GW2 is used. A decrease in flexural strength is observed with the addition of bentonite. The same observation applies to the breaking strength, which is due to the large amount of glass waste forming a porous structure and reducing the mechanical resistance to flexion. For the ceramic tile samples with 100% bentonite, the flexural strength is estimated to be 14.21 MPa and the breaking strength is estimated to be about 500 N, which are weaker values than



Figure 11: Flexural (a) and breaking strength (b) of elaborated ceramic tiles as a function of bentonite content.

those of GB35. Due to the high silica content of the material used, it can react with the calcium hydroxide formed during hydration to produce a calcium silicate hydrate (C-S-H) gel. The C-S-H content has an important effect on the strength of the ceramic [27, 28].

Hardness: It was found that Vickers hardness improves with the addition of bentonite, which contains a high proportion of silica and alumina, increasing the hardness of the glass waste-bentonite mixture, with variant GB35 having the optimum value 188KgF/2 (see Figure 12). The hardness of variant B is 200 kgF/2, the highest of all the variants obtained, which is due to the higher bentonite and alumina content in GW2. It' is the same variant as GW1. Thus, the particle sizes of the

glass waste didn't affect the hardness of the produced tiles. The hardness increases with increasing proportion of glass waste. This shows that the ceramic becomes more compact when the proportion of glass waste is increased. This is the case because at this stage the glass phase in the ceramic mixture is vitrified [29–30].

Structure al aspects by optical microscopic With the help of metallurgical microscope OPTIKA B-500MET, we have observed various produced tiles in different scales. It can be observed that the coloration of the tiles becomes darker with the addition of bentonite (the coloration becomes brick red). As can be seen in Figure 13, this is due to the amount of iron oxide added to the bentonite. We also



Figure 12: Variation of hardness of ceramic samples as a function of bentonite content.



Figure 13: Observation under microscope at 500 µm.

compare tiles with the same chemical composition but produced with different types of glass waste. It can be seen that the tiles produced with GW1 are higher than those produced with GW2. Compared with the appearance of the grains with the same composition of the tiles, but produced with different glass waste, it was found that in the tiles made with GW2, the grains were slightly larger due to the larger granulometry. The gradual addition of bentonite makes the surface smoother and harder. This can be explained by the addition of alumina, which is contained in the bentonite.

Conclusion

It is known that ceramic tiles are usually produced at 1100 °C. They are expensive and environmentally harmful compared to the ceramic tiles discussed in this study. The aim of this work was to produce new ceramic tiles based on waste glass while reducing the thermal energy for treatment. Ceramic tiles made from waste glass (65% by mass) and bentonite (35% by mass) can not only solve the problems associated with the storage of waste glass, but also save energy. Waste glass is environmentally friendly, which means it can be ground and used to produce the proposed ceramic tiles at 900 °C with lower energy consumption, and it is very easy to mould. In addition, the bentonite content in these tiles is lower and allows the production of ceramic tiles with acceptable physical and mechanical resistance. This increases sustainability and resistance to various factors. According to the results, the properties of the ceramic tiles produced are good and meet the standards. The process is still

in its infancy, but with further development it can be used in various industries and reduce energy and raw material consumption.

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