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UTILIZATION OF THE GLASS WASTE AS A SINTERING AGENT AND SILICA SOURCE TO PRODUCE CERAMIC PRODUCTS

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ABSTRACT

In this study, waste glass was utilized as a sintering agent and silica source in red ceramic products and investigated its effect on the properties of these products. The sample composition was prepared from waste glass, kaolin clay, and natural sand. Formulations of samples were contained (0- 40) % of glass waste. After preparation of the samples by the powder technology technique, some properties such as apparent porosity, bulk density, linear shrinkage, fracture strength, hardness, SEM, and XRD were examined. From the results of this investigation, the highest bulk density of the tested specimens was 2.19 g/cm^3, the lowest apparent porosity was 3.2%, the highest strength was 59 MPa and the highest hardness was 219 MPa. From results of XRD analysis for the specimens, it can be indicated that the similarity in crystalline patterns confirms that the improvement mechanism is physical (liquid phase sintering) and not chemical (formation of new compounds), which is a positive aspect that enhances the simplicity of industrial application. The SEM image for the specimen with 40% of glass waste shows that the glassy phase content considerably increases, while the pores significantly reduce. Therefore, the utilization of glass waste improves mechanical properties and reduces the large quantity of the inherent imperfections in the microstructures of ceramic products. Thus, industrial economic and environmental benefits can be obtained by this replacement from the side of avoiding landfilling with these wastes, saving energy, and reducing material consumption.

KEYWORDS

Waste glass, Kaolin clay, Natural sand, Mullite, Quartz.

1. INTRODUCTION

In recent years, the reuse of some material waste in the production of ceramics has been largely investigated to avoid landfilling with these wastes as well as to economically vindicate the high costs associated with its manufacturing (Antunes MLP et al. 2019; Salman MM et al. 2020; Salman MM and Nhabih HT., 2020). In recent years, it has been shown that the industrial solid waste generation has mostly increased around the world. This increase in solid waste has as chief causes the population growth and the consequent increase in the generation of waste (Parthan SR et al. 2012); the industrialization processes (Hu D et al. 1998; Nhabih HT et al. 2020; Nhabih HT et al. 2018; Al-Amiri and Al-Akaishi, 2025); urbanization process which means the numbers increase for the inhabitants in a big city and that leads to increase in waste generation (Mrayyan B and Hamdi MR. 2006); and the number increase of unsuitable positions for the final wastes discarding in dumps (Parthan SR et al. 2012, Mrayyan B and Hamdi MR 2006), Hussein AM et al. 2020), that further worsens these environmental problems. Therefore, the necessity to find methods to reuse the solid waste is very important (Nhabih HT et al., 2024; Ozturk ZB et al., 2023; Nhabih HT et al., 2025). Also, the present work investigates the alternative, which is the reuse of wastes, such as waste glass, in the industry of red ceramics, where the wastes can play various functions in the production of ceramics (Luo Y et al. 2021). More important wastes from an industrial viewpoint are glass wastes. The reusing of these wastes in other applications has been studied in many research studies. Most of the waste glass is manufactured from soda-lime silica glass (SLS) coming from flat and container glass (Rawlings RD et al. 2006). SLS glass is the least costly and most common glass form. It usually consists of (12-18%) soda, (5-12%) lime, and (60-75%) silica. It has low resistance to sudden changes in temperature and high temperatures, only fair resistance to corrosive chemical materials, a melting temperature of about 1000°C, and a glass transition temperature of approximately 500-600°C (Sutatip T et al., 2010). SLS glass waste can be reused in glass production again and in other applications. Where it can be reused as an additive material in the construction aggregates (Wang X et al. 2013, Sasui S et al. 2021), in the ceramic tiles production (Farid L et al. 2023), and in metallurgical processes (Lee CT 2013, Scheffler M and Colombo P 2005, Darweesh HHM 2019). Also, the reuse of glass waste was studied as a total and partial substitution for the quartz sand in the sand-lime products. The effect of recycled glass on the mechanical and physical properties of the produced samples was investigated. From the results of this study, a lower density and higher compressive strength were obtained in comparison with the reference sample after use of the waste glass with sand and lime (Katarzyna B et al., 2020). In addition, the glass wastes were studied as a total and partial substitute for feldspar in porcelain. Where different percentages of waste glass, kaolin, quartz, and feldspar were used to form the samples using the powder technology technique. From the results of this study, good mechanical, physical, and thermal properties were obtained by using 75% waste glass of feldspar (Salman MM and Nhabih HT 2020). Also, the reuse of glass wastes was studied as a partial replacement of natural fluxes into whiteware formulations (Darweesh HHM 2021, Khaled B et al. 2019, Tarnkamol T and Guilherme PS 2014, Adediran A et al. 2021). In addition, the glass-ceramics can be made from the waste glass specifically (Angjusheva B et al. 2016). The current work evaluates a way to reuse the waste glass in useful ceramic products by using these wastes as a sintering agent and silica source in red ceramic products, because this approach leads to reducing the solid wastes (the waste glass), saving the energy (by using a sintering agent) and conserving the resources (the silica source). Consequently, it investigates environmental and economic benefits. The studied compositions are suitable for the red ceramic products, such as roof tiles, blocks, and bricks used for wall and base fillings, and ceramic tubes. Although numerous published studies have explored the reuse of glass waste, it is necessary to analyze new methodologies that enable the actual application of the waste in ceramics. When the current study is compared with other investigations already carried out on the topic, it can be shown that the present study was made with different compositions of raw materials, the effect of high replacement ratios (up to 40%) in red ceramics while maintaining a constant clay content has not been sufficiently explored, particularly concerning the accompanying economic and environmental analysis, and used different parameters of the fabrication, such as pressure and temperature. Thus, industrial economic and environmental benefits can be obtained by this replacement.

2. MATERIALS AND METHODS

In this study, waste from soda-lime-silica glass (SLS) was utilized as a sintering agent and silica source in red ceramic products. The preparation of samples included the utilization of raw materials from waste glass, kaolin clay, and natural sand. The waste glass was fragmented into a fine powder by using the ball mill and sieved to obtain the desired particle size. The average particle size for the powder of waste glass was 37.6 μm. The chemical composition of raw materials (% wt..) is shown in Table 1. Formulations of the prepared samples were contained (0, 10, 20, 30, 40) % of glass waste as shown in Table 2; therefore, the produced samples were termed W0, W10, W20, W30, W40. To obtain a homogeneous mixture of materials, an electrical mixer was used for a period of 4 hours. The average particle sizes for the materials' powders were 33.5 μm for kaolin clay and 74.3 μm for natural sand. From the changes observed in the compositions resulting from the addition of glass waste, the color of the samples changed from reddish to gray due to the presence of a percentage of iron oxide in the natural sand composition.

Table 1. The chemical composition of raw materials (% wt.).

Composition	SiO_2	Al_2O_3	Fe_2O_3	TiO_2	K_2O	MgO	Na_2O	CaO	LOI
Kaolin clay	47.4	38.5	0.5	0.1	0.6	0.3	0.3	0.3	12.0
Natural sand	81.26	11.9	1.2	0.47	1.5	0.62	0.84	0.51	1.7
Waste glass	72.58	1.82	0.55	0.01	0.36	2.53	6.88	15.54	0.00

LOI: loss on ignition.

Table 2. The proportions of mixture for the samples (% wt.).

Constituents	Formulations of the ceramic samples				
	W0	W10	W20	W30	W40
Kaolin clay	60	60	60	60	60
Natural sand	40	30	20	10	0
Waste glass	0	10	20	30	40

The samples were shaped by a steel die with a diameter of 13 mm, pressing pressure of 70 MPa, and a hydraulic pressing machine. A semi-dry pressing method was utilized to bond the powders of the materials by use polyvinyl alcohol PVA as a plasticizer. After the formation of specimens, a temperature of 105°C was utilized to dry them to eliminate the moisture, and a temperature of 1000°C was used to sinter them in an electric furnace with 2-hour soaking time and a 7°C/min heating rate. The samples shaped in this study are shown in Fig. 1.

Fig. 1 Schematic representation of tested specimens. The samples in this study: sample A (W0), sample B (W10), sample C (W20), sample D (W30), sample E (W40).

After the formation of samples, some physical and mechanical properties of the specimens were measured, such as apparent porosity, bulk density, linear shrinkage, hardness, and fracture strength. Additionally, SEM and XRD analyses were performed on the produced samples. These tests were done as follows:

The apparent porosity and bulk density of specimens were measured depending on Archimedes' procedure and ASTM C373-88 standard, Eqs. 1 and 2 were used for calculation the apparent porosity (P_o, %) and the bulk density (D, g/cm^3) of specimens respectively (ASTM C373 (1988)).

$$P_o\,\% = \frac{M_S - M_D}{M_S - M_P} .100 \tag{1}$$

$$D = \frac{M_D * D_{water}}{M_S - M_P} \tag{2}$$

where, D_{water}: the water density (g/cm^3); M_S: the water-saturated mass of specimen (g); M_D: the dry mass of specimen (g); M_P: the suspended mass of specimen (g).

According to the ASTM C1407 standard, the linear shrinkage (LS, %) of sintered samples was measured using Eq. 3 below.

$$LS = \frac{D_b - D_f}{D_b} . 100 \qquad (3)$$

where, D_b: the specimen diameter before the firing process; D_f: the specimen diameter after the firing process.

According to the ASTM C773-88 standard, the fracture strength for samples (σ_f, MPa) was calculated using Eq. 4 below (ASTM C773 1988).

$$\sigma_f = \frac{F}{C_a} \qquad (4)$$

where, F: the applied load up to fracture (N); C_a: the cross-section area of sample (mm^2).

Depending on the ASTM C1327- 90 standard, the sample's microhardness was measured with the Vickers hardness method. 10s is the time selected to apply an indentation load of 90 N on the sample surface. Eq. 5 below was utilized to calculate the values of Vickers hardness for samples (HV, MPa) (Roy M 2010, Salman MM et al. 2025).

$$HV = 1.854. \ \frac{D}{A^2} \qquad (5)$$

where D: the indentation load (N); A: the indentation diagonal on the specimen surface (mm).

To detect the crystalline phase developed in the sample, XRD analysis was performed on the sample's powders. The sample was powdered to produce a fine powder, which was employed in XRD analysis. The SHIMADZU XRD–6000 machine was used to obtain XRD patterns for the samples by utilizing the continuous scan mode with a scan speed of 7˚/min, a θ-2θ range of 0-70˚, and a step size of 2θ = 0.02˚.

In addition, scanning electron microscopy SEM was employed to observe the microstructures and surface morphology of the produced samples.

3. RESULTS AND DISCUSSION

The results of the physical tests on the produced specimens are presented in Table 3, which includes the bulk density, linear shrinkage upon firing, and apparent porosity. It can be demonstrated that as the glass waste content increases in the composition of the samples, the linear shrinkage and bulk density of the produced samples also increase, while the apparent porosity decreases. This occurs due to the chemical composition of glass waste, which is rich in alkalis, such as CaO, Na_2O, and MgO. These alkalis work to form the liquid phase, which is considered the main sintering mechanism for ceramics (Conconi MS et al. 2019). Where sintering occurs with the presence of a liquid phase using fluxes, this process is known as liquid-phase sintering (Jamo HU et al., 2022; Salman MM and Nhabih HT, 2025). The liquid phase approaches the particles of the sample by the influence of the surface energy force (Salman MM and Nhabih HT 2025). Thus, an increase in the formation of the liquid phase results in a reduction in the open porosity of the produced samples. Similar results were detected in prior research (Salman MM and Nhabih HT 2020, Ozturk ZB and Ay N 2012) and results detected in (Darweesh HHM 2019) with a firing temperature of 1000°C only. Also, the bulk density and firing shrinkage of the produced samples increased with the increase in sintering ability.

Table 3. The results of physical tests for the samples.

Sample No.	Linear shrinkage %	Apparent porosity %	Bulk density (g/cm^3)
W0	2.6 ± 0.4	20.2 ± 1.2	1.56 ± 0.13
W10	4.3 ± 0.51	15.4 ± 0.8	1.84 ± 0.1
W20	5.4 ± 0.49	8.6 ± 0.9	2.03 ± 0.11
W30	6.7 ± 0.53	5.4 ±0.5	2.09 ± 0.1
W40	9.1 ± 0.58	3.2 ±0.6	2.19 ± 0.09

Additionally, the results of the mechanical tests on the produced specimens are presented in Figs. 2 and 3, which represent the fracture strength and hardness, respectively. It can be demonstrated that as the glass waste content in the sample composition increases, the fracture strength and hardness of the produced samples also increase. This occurs because of the thermal reaction that takes place among the ceramic batch constituents during firing. Where the chemical composition of glass wastes is rich in the alkalis, such as CaO, Na_2O, and MgO. These alkalis form the liquid phase during firing. The formed liquid phase bonds the sample particles and reduces the pores in the sample structure. Thus, a highly rigid network is created in the sample structure. Besides, a reduction in the content of the free quartz crystals in the sample composition resulting from a replacement of natural sand by waste glass enhances the mechanical strength of the produced sample. In addition, an effect of the particle size distribution of the raw materials. Where a replacement of material with big particle size (74.3 μm for natural sand) by material with a small particle size (37.6 μm for waste glass) improves the mechanical strength (Hashim AM and Nhabih HT, 2018; Hassan et al. 2025). That leads to enhancing the mechanical properties of the produced samples. Similar results were detected in prior research (Salman MM and Nhabih HT, 2020; Ozturk ZB and Ay N, 2012; Costa FB et al. 2009; Martín-Márquez J et al. 2010; Jiaad et al. 2025) and results detected in (Darweesh HHM 2019) with a firing temperature of 1000°C only.

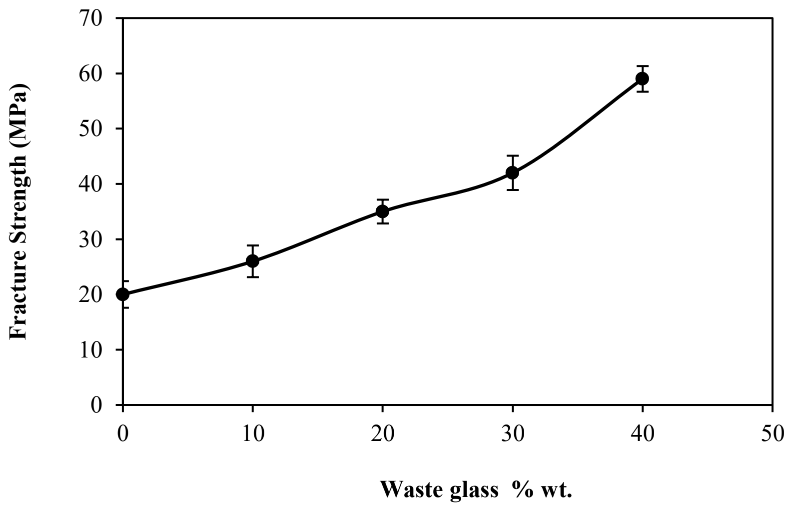


Fig. 2 Effect of waste glass % wt. on the fracture strength of the produced samples.

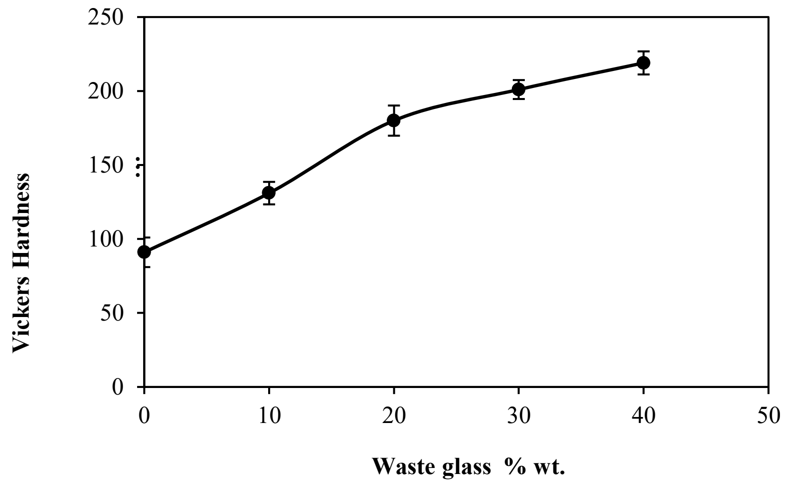


Fig. 3 Effect of waste glass % wt. on the hardness of the produced samples.

The results of XRD analysis for the specimens with the compositions W0, W20, and W40 are shown in Fig. 4. XRD patterns displayed the presence of quartz phase (ICDD 046-1045) and mullite phase (ICDD 074-4143) as the two main phases in all the specimens. It can be seen that the number of quartz phase peaks noticeably reduced for the specimen with the composition W40, due to the reduction in the percentage of natural sand in the composition of this specimen. Additionally, the mullite phase forms during the firing process, while the quartz phase remains as a residual phase in the fired specimens at the temperature used in the current study, which is 1000°C. These identified phases, which are mullite and quartz phases, influence the microstructural and mechanical characteristics of the produced specimens. It can be indicated that the similarity in crystalline patterns confirms that the improvement mechanism is physical (liquid phase sintering) and not chemical (formation of new compounds), which is a positive aspect that enhances the simplicity of industrial application.

The results of SEM analysis for the specimens with the compositions W0, W20, and W40 are respectively shown in Fig. 5 (a), (b), and (c). It can be seen from the SEM image for the specimen with the composition W0, which is shown in Fig. 5 (a), the presence of a notable amount of undissolved free quartz and a noteworthy number of pores in the microstructure for this specimen. The SEM image of the specimen with the composition W20, shown in Fig. 5b, reveals the presence of a glassy phase and a reduction in pore size within the microstructure of this specimen. Additionally, the SEM image of the specimen with the composition W40, shown in Fig. 5 (c), reveals a significant amount of the glassy phase and a reduction in the pore and quartz crystal content within the microstructure of this specimen. Similar results were detected in prior research (Angjusheva B et al. 2016, Ozturk ZB and Ay N 2012). This substitution can improve the mechanical properties and reduce a large quantity of the inherent imperfections in the microstructures of red ceramic products. Thus, economic and environmental benefits can be achieved by this replacement.

From the economic and logistical analysis, it was found that the cost of pre-treatment (collection, sorting and grinding) of the glass waste used in this study is less than the costs of raw materials, energy and landfill fees, which provides financial savings in industrial applications.

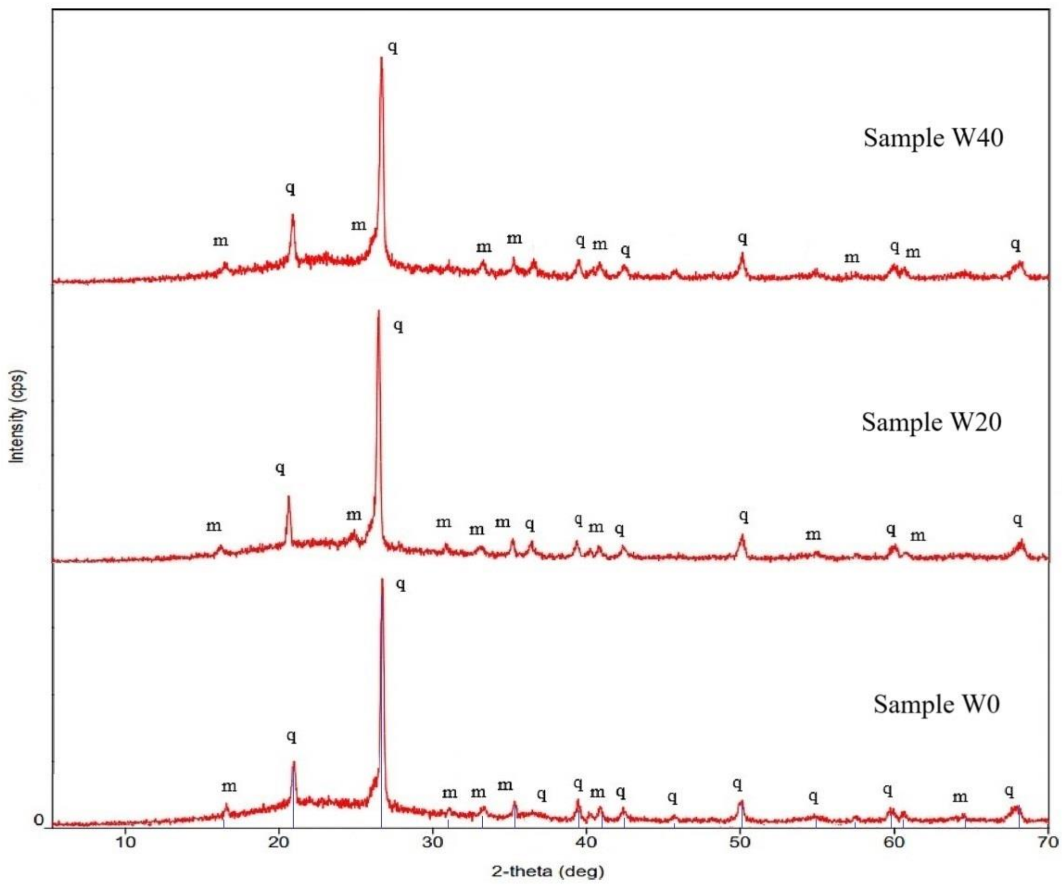
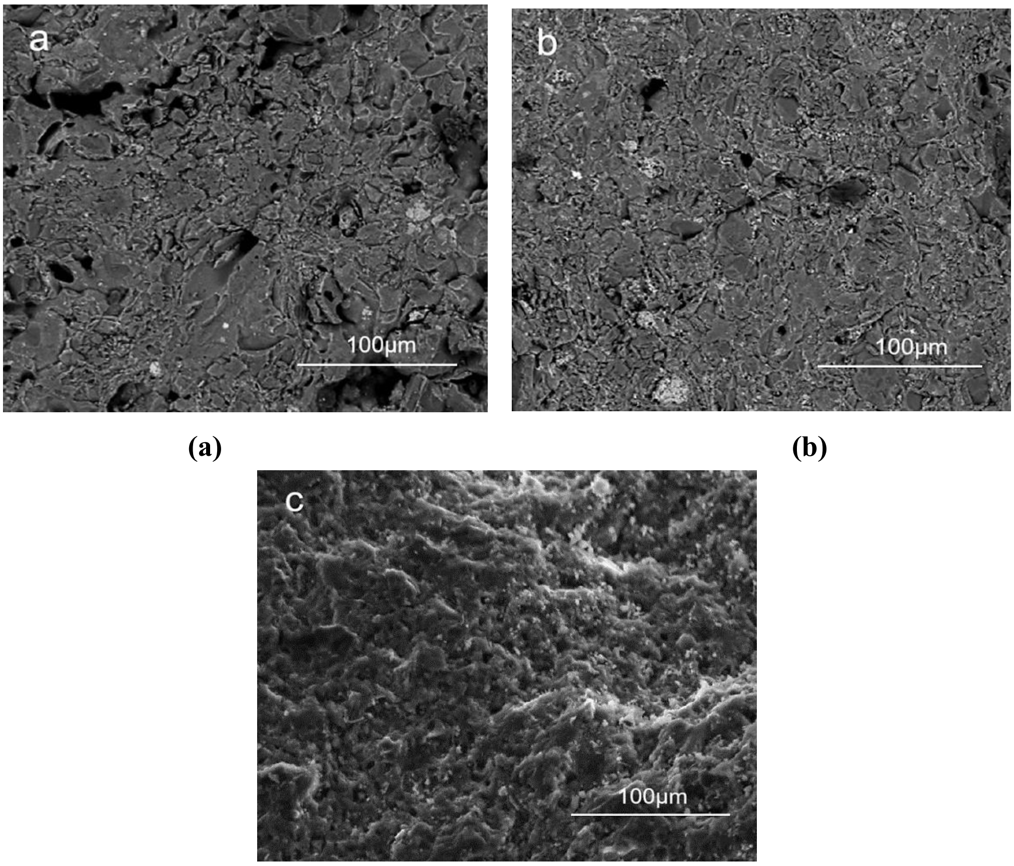


Fig. 4 XRD pattern of the produced samples with the compositions of W0, W20, and W40.



(a) (b)

(c)

Fig. 5 SEM images of the produced samples: (a) for the sample W0, (b) for the sample W20, and (c) for the sample W40.

4. CONCLUSIONS

As a result of the tests above, it can be concluded that the increase in glass waste content in the sample composition enhances their densification behavior, due to the formation of a larger liquid phase resulting from the chemical composition of the glass waste, which is rich in alkalis. Thus, it can enhance their physical properties. Also, the increase of glass waste content in the composition of samples improved their mechanical properties, due to strengthening the bond process among the sample particles and reducing the pores because of the formation of a liquid phase, which produced a higher vitrification in the produced samples, and also reduced the content of the free quartz crystals in the microstructure of these samples. Where the following results were obtained: the highest bulk density of the tested specimens was 2.19 g/cm^3, the lowest apparent porosity was 3.2%, the highest strength was 59 MPa and the highest hardness was 219 MPa. From results of XRD analysis for the specimens, it can be indicated that the similarity in crystalline patterns confirms that the improvement mechanism is physical (liquid phase sintering) and not chemical (formation of new compounds), which is a positive aspect that enhances the simplicity of industrial application. The SEM image for the specimen with 40% of glass waste shows that the glassy phase content considerably increases, while the pores significantly reduce. Therefore, the utilization of glass waste improves mechanical properties and reduces the large quantity of the inherent imperfections in the microstructures of ceramic products. Therefore, a highly rigid network was created in the produced samples, which provided an improvement in the properties. Hence, the reuse of glass waste in the red ceramic industry has a positive impact. Industrial, economic and environmental benefits can be achieved by this reuse from the side of avoiding landfilling with these wastes, saving energy required for sintering, and reducing raw material consumption.

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