



The effect of temperature on ceramic bricks with replacing waste glass in low volume ratio

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Abstract

This paper provides a review of research concerning the recycling of waste glass into eco-friendly ceramic bricks. The traditional methods for producing construction materials used natural resources. Besides, the industrial and urban management systems are generated solid waste and most often dumped them in open fields. These activities pose serious detrimental effects on environment. To safeguard the environment, many efforts are being made for the recycling of waste glass with a view of utilizing them in to production of various construction materials. In conclusion, the reuse of waste glass in brick production might be an environmental friendly method to manage them. In some cases it implies an enhancement of the brick properties as well as an advantage for brick producers.

Keywords: ceramic bricks, construction materials, fire clay, SEM, waste glass

1. Introduction

The industrial development of many countries brought it the problems of waste disposal contamination and health damages along. India produces about 70 MT of waste every year, 50% of which is not suitably dumped. Recycling is an important environmental and economical alternative for each material class, mainly for those that do not decompose easily. For example, glasses do not decomposed by microbial or atmospheric precipitation for thousands of years. For ordinary purposes glasses are usually prepared from silica, but currently technological application, such as in optical fiber, require other composition, like germanium ^[1] or fluoride glasses ^[2]. In India, glass contributes about 5% of the total waste.

Many researches worldwide are donated to obtain products of economical interest using glass waste. Japan, United state, Taiwan, Brasil and many European countries are attempting reuse waste glass to partly substitute considerable amount of ceramic building materials (porcelain material) ^[3]. The proper use of contaminated waste glass has also been suggested. Industrial waste metallurgical slag, fly-ash, etching refuse, coal ash and glass waste containing chemical combination has been used to produce glass-ceramic bricks ^[4]. Based on the usage of glass waste in the production of raw porcelain glass ^[5] had been researcher were made to estimate. The effect of up to 10% glass waste addition in porcelain stone ware tiles were researched ^[6]. Ceramic produced by addition of glass waste had been characterized to have higher strength in some composition with reduced sintering temperature ^[6-9]. It is proven that firing temperature influences the production cost as well the physical properties of the resulting ceramics. In addition the use of waste and by-product materials in infrastructure development is proven to be economically variable when environmental factors are considered and when

these materials meet appropriate performance standards.

The micro structural evolution of ceramic materials on firing has been exhaustively studied over the past fifty years. However because of the complex interplay between raw materials and firing kinetics, this type of ceramic materials continues to present many research challenges ^[10]. The introduction of waste glass as partial replacement for raw materials leads to the formation of high temperature phases which play an important role in the micro structural development on firing ceramic bricks, thus affecting its performance. It is important to determine the new phases formed and correlate their existence to the starting materials. The processing reaction between glaze and the near-surface materials and the glaze and kiln atmosphere have a significant effect on the bonding. Each raw material within the body formulation contributes differently to the final properties.

2. Materials and Method

The materials used for this study included dried glass waste powder, and porcelain composition (kaolin, quartz and feldspar). The equipment used for this study included furnace, mounting press, grinding mill, sieve shaker and set of sieves and compression strength tested. Dry glass waste and porcelain mixture obtained is from government ceramic Institute Vridhachalam, Cuddalore District, Tamilnadu, India. A mixture of porcelain composition made with various proposition of glass waste 0-20 wt % (S₀ - S₄) and various percentages. The resulting mixtures were then compacted in a mounting press to obtain rectangular bar shape samples. These samples were placed in furnace and fired at various temperatures ranging from 900-1100°C such that the additive powders burns off leaving some ash pores. A series of analyses were performed on the fired samples.

3. Results and Discussion

3.1 Chemical Analysis

Table 1 shows the chemical and the normative mineralogical composition of the additive (waste glass) sample. The chemical data correlate well with the mineralogical composition as the silica and alumina contents agree with the quartz and kaolinite contents (Fig.1). The main oxides are SiO_2 , Al_2O_3 , CaO and Fe_2O_3 whereas BaO , K_2O , MgO , SO_3 and TiO_2 are present only in small amounts. The oxides value for Na_2O is higher in waste glass (13.74%). The relative abundance of SiO_2 indicates a rather high content of quartz, whereas Al_2O_3 can be correlated with clay minerals and feldspar. Varying amounts of quartz influence the plasticity and drying behavior of the clays. Relatively high iron oxide (Fe_2O_3) content provides a characteristic reddish – brown color to the fired body. However, Fe_2O_3 is not the only factor responsible for the coloring of ceramic wares [11]. Other constituents such as CaO , MgO and TiO_2 can appreciably modify the color of the fired body. The temperature of firing, relative amounts of Al_2O_3 , and the furnace atmosphere all play an important role in the development of color in the fired ceramic products [12]. The main effects of alkali in clay are to reduce their refractory temperature and therefore, they are called fluxes [13].

Lime melts silica in burning and binds the particle of brick together. It also reduces the shrinkage of brick during drying. Excess amount of lime cause the brick to fuse and the shape will be lost. Alkali and organic matter of small quantity assists burning of brick clay. They also reduce the fusion point. If excess amount of such unburned organic matter is present and not burned properly the brick would be porous based. Finally in chemical composition SiO_2 was the most abundant component, followed by Al_2O_3 , CaO and Fe_2O_3 contents, which was reaching up to 83.46% of waste glass. The main differences of high CaO (11.75%) of waste glass and lower Al_2O_3 (2.080%) present. These differences suggest that highest amount of CaO in the firing step can give rise to higher porosity in the brick specimen prepared to waste glass, which is also suggested their higher thermal loss [14]. From the chemical composition shown in (Table 1) the sum values of Fe_2O_3 and TiO_2 are 0.373%. Many studies have described the influence of these minerals in enhancing the process of sintering of clay matrix as the formation of mullite [15, 16]. Ti^{4+} and Fe^{3+} play an important role either substituting Al^{3+} or by their integration into the structure intensities of the matrix.

3.2 Mineralogy of the Unfired Materials (Samples S_0 - S_4)

Fig.2 displays the XRD patterns of without waste glass addition S_0 and with different levels of (0-20 %) of waste glass replacement ceramic brick samples respectively. The base (S_0) material used in the present study consisted mainly with the crystalline phase of predominantly quartz and some peaks from kaolinite, hematite and feldspar (Fig.2). Quartz phases could be detected at 2θ values of 4.26° , 3.34° , 2.13° , 1.81° and 1.54° , whereas kaolinite phases could be detected at 2θ values of 3.58° , 2.34° and 1.37° , hematite phases could be detected 2θ values of 1.49° and feldspar phases could be detected at 3.21° . The base material (S_0) used in the present study consisted mainly of crystalline phase with large amount of silica, which agrees with the composition shown in Table 2.

Alcantara *et al.*, [14] have studied the characterization of ceramic tiles using XRD and identified the clay minerals and non-clay minerals kaolinite, quartz, hematite and feldspar present in them. From these results the observed minerals were kaolinite, quartz, hematite and feldspar.

Table 1: Chemical composition of the raw material (Waste Glass)

S. No	Element composition	Waste Glass
1	SiO_2	69.320
2	Al_2O_3	2.080
3	BaO	0.110
4	CaO	11.750
5	Fe_2O_3	0.317
6	K_2O	0.928
7	MgO	1.370
8	Na_2O	13.740
9	SO_3	0.166
10	TiO_2	0.056

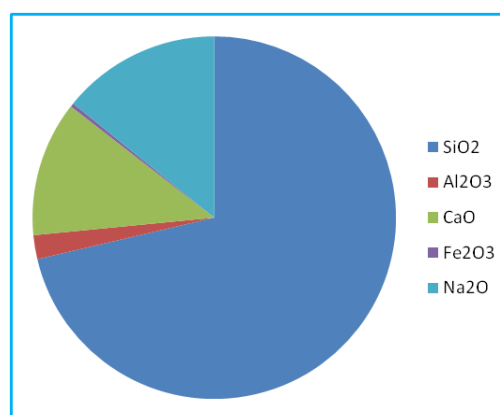


Fig 1: Major Chemical compositions of the raw material (Waste glass)

3.2.1 Mineralogy of the Sample (S_3 -15 wt %) addition of Waste Glass fired at different temperatures (800, 900, 1000 and 1100°C)

The X-ray diffraction pattern is modified by changes in the clay structure on heating and the nature of the modification may be used for diagnostic purposes.

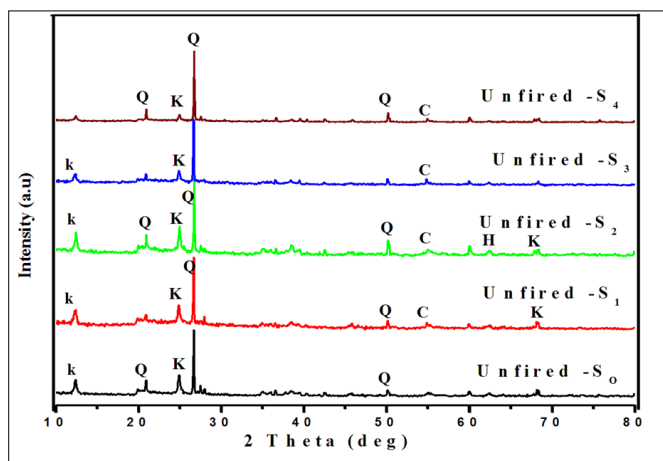


Fig 2: XRD spectra of ceramic brick with different percentage of waste glass ($S_0 - S_4$) Samples (unfired)

Each clay mineral undergoes structural changes at different temperatures. The removal of absorbed water in smectites, vermiculites and hydrated halloysite occurs at temperatures below 300°C. All clay minerals exhibit structural changes at higher temperatures (>500°C) when the removal of combined water occurs. The ultimate effect of heat treatment is for the X-ray pattern to be replaced by that of the fired product which is often amorphous to X-rays (kaolinite). With kaolin, X-ray diffraction of specimens after heat treatment is not necessary before a positive identification of a clay mineral assemblage can be made. However, knowledge of the X-ray diffraction pattern of clays that have been heated to different temperatures may be useful in identifying clay raw materials that have undergone some heat treatment during processing.

Table 3 shows that above 900°C, the clay minerals (illite) peak breaks down (illite peaks not seen). The densification behavior of the ceramic brick mixture is influenced by the sources of flux materials such as K₂O, Na₂O and Fe₂O₃, which favour the formation of vitreous phase. Above >1000°C, the phases present are quartz (SiO₂), hematite (Fe₂O₃) and anorthite (CaO.Al₂O₃.2SiO₂). It can be observed that the characteristics peaks of kaolinite are disappeared.

The hematite that appears is higher than that in the unfired material, suggesting that there is a formation of iron oxides from the recrystallization of iron after the breakdown of the phyllosilicates. The thermal transformation of illite to mullite was described by previous authors [17, 18]. Metakaolinite decomposes into spinel and an amorphous phase by firing.

Table 2: XRD Diffraction data for samples (So-S₄) group-I unfired

d spacing (Å)						
S. No	S ₀	S ₁	S ₂	S ₃	S ₄	Name of the Minerals
1	-	4.46420	-	-	4.40329	Quartz
2	4.26188	4.23186	4.23879	4.25332	4.24184	
3	3.34830	3.34360	3.33587	3.34462	3.33538	
4	2.45919	2.45692	2.45297	2.45665	2.45141	
5	2.13047	-	2.12472	-	2.12385	
6	1.81947	1.81805	1.81595	1.81793	1.81552	
7	1.54384	1.54223	1.54020	1.54113	1.54004	
8	7.18830	7.17967	7.12854	7.19030	7.13556	
9	3.58026	3.57688	3.56677	-	3.56718	Kaolinite
10	2.34313	2.33622	2.33749	2.34317	2.33853	
11	1.37302	-	1.37445	1.37431	1.37275	
12	-	-	-	-	1.28668	
13	-	-	2.29293	2.28276	2.94265	Hematite
14	1.49339	-	1.48812	1.49003	1.48883	
15	-	-	-	-	1.45247	Muscovite
16	2.56037	2.53177	2.55697	2.51976	2.56088	
17	-	3.21240	3.23519	-	3.23687	Feldspar
18	-	1.98213	1.98636	1.98655	1.97741	
19	1.67325	1.66742	1.66440	1.67208	1.66935	Calcite
20	-	-	-	-	1.25500	

The reaction between the two phases produces mullite. The amorphous phase plays an important role in the densification process. Mullite can also play an active role in the process. Different authors have demonstrated that mullitization can be increased by ions such as Fe³⁺. This metallic ion helps mullite formation by replacing the Al³⁺ ion in the glass structure during firing [19, 20]. At 1100°C, the amount of residual quartz decreases with higher temperature due to its dissolution and due to the conversion of a part of a SiO₂ in

crystalite. The anorthite peak intensities begin to increase and quartz peak begins to decrease (Fig.3).

The XRD peaks of the residual clay phases remain with similar intensity at this temperature range, whereas mullite XRD peaks begin to emerge between 1000 and 1100°C the background of the crystalline clay reflections as the temperature reaches 1000°C the intensity of the appearance of hematite. Quartz decomposes gradually from 800-1100°C, diminishing drastically at 1100°C.

Table 3: XRD Diffraction Data for WG additive clay brick sample (S₃) unfired and sintered at different temperatures

d spacing (Å)						
S. No	Unfired	800°C	900°C	1000°C	1100°C	Name of the Minerals
1	4.25332	4.25377	4.25496	4.25302	4.25360	Quartz
2	-	-	-	3.7595	-	
3	3.34462	3.34188	3.34310	3.34205	3.34351	
4	-	2.12702	2.12463	2.12517	2.12590	
5	1.98655	1.97919	-	1.98268	-	
6	1.81793	1.81748	1.82009	1.81773	1.81837	
7	1.54113	1.54080	1.54082	1.54167	1.54396	
8	-	-	-	1.52762	1.52809	
9	-	-	-	-	1.3381	

10	7.19030	-	-	-	-	Kaolinite
11	3.5739	-	-	-	-	
12	2.34317	-	-	-	-	
13	-	1.38030	1.37959	-	-	Metakaoline
14	-	-	-	-	-	
15	-	-	2.07912	-	-	
16	-	-	-	-	2.6994	Hematite
17	2.51976	-	-	-	-	
18	2.28276	2.28353	2.28561	-	2.29110	
19	-	-	-	-	1.44606	
20	-	3.19768	3.19302	-	-	
21	-	1.38030	1.37959	1.37794	1.37609	
22	2.45665	2.45614	2.4550	2.45568	2.45873	Anorthite
23	-	-	-	-	2.54946	Muscovite
24	-	-	2.0791	-	-	Calcite
25	1.67208	-	-	-	1.60294	
26	-	-	-	-	5.9127	Mullite
27	-	-	-	2.2087	2.2114	
28	-	-	-	1.5276	1.5281	
29	-	-	-	1.3794	1.3760	
30	-	-	-	-	4.0483	Cristobalite

The formation of primary mullite is between the temperatures of 900-1000°C and 1000-1100°C [21]. The characteristic peaks also became sharp, showing a highly crystalline phase. Similar results have been reported in previous studies [22-23]. With increasing temperature, the complete disappearance of some mineral phases and the diminishing of the remains are observed, as suggested by the decrease in the intensity of the diffraction maximum or rising of the back ground noise, suggests the presence of an amorphous phase similar to the previous report [24]. At this step the clay body would essentially be an amorphous material with some residual compounds, and newly formed mullite and hematite. XRD

data reveal that decomposed and disappearing phases all contribute to the formation of an amorphous phases above 1000°C, besides mullite (at 1000°C) and hematite formation of new crystalline phases appeared. The diffractograms of the sample S_3 formulation presented mullite, anorthite, hematite in crystalline phases but due to the amount of crystalline components present in the waste (WG 15 wt. %) the presence of several other phases was seen. It was observed that incorporation of 15 Wt. % waste glass provides an increasing effect on the mullite content. As a result, it is suggested that waste glass added sample (S_3) can be used as waste glass addition up to 15 wt%.

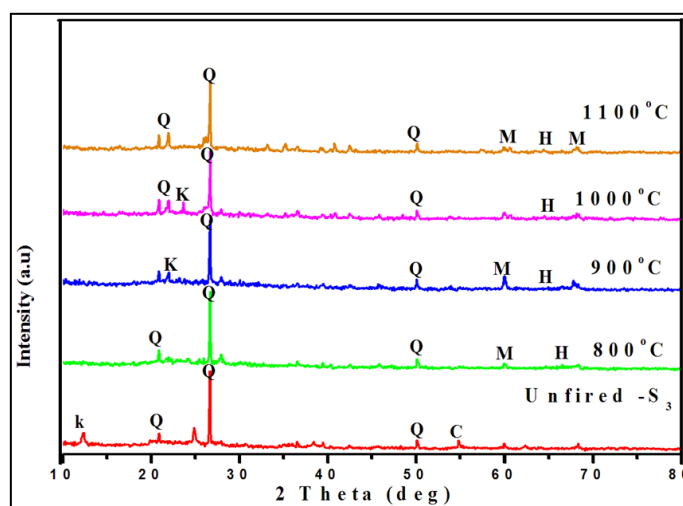


Fig 3: XRD spectra of samples (S_3) unfired and sintered at different temperatures

3.3 Morphological Analysis

A FESEM fitted with an EDAX analysis was used to identify minerals and study their morphology changes at various temperatures (800, 900, 1000 and 1100°C) in ceramic bricks added 15 Wt% glass waste (S_3). The morphological structures and EDAX spectra are shown in Figs. 4 (a-d) to 8 (a-d) and

their elemental concentration are provided in Table 4. Fig.4 (a-c) shows how the FESEM can solve the problem of determining the morphology of a ceramic brick powder. Fig.4 (a) is a highly magnified view of unfired ceramic brick (S_3) sample. This Fig shows that large plates of the clay appear to be composed of much smaller particles. The unfired sample

(S3) revealed the particle of different shapes and sizes the presence of coarse and porous particles is also observed (Fig.4a-d). It can be seen that the clay sample S₃ is rich in

plate like structure which are probably clay mineral ones. Different minerals of different sizes are blended and distribute randomly in the materials.

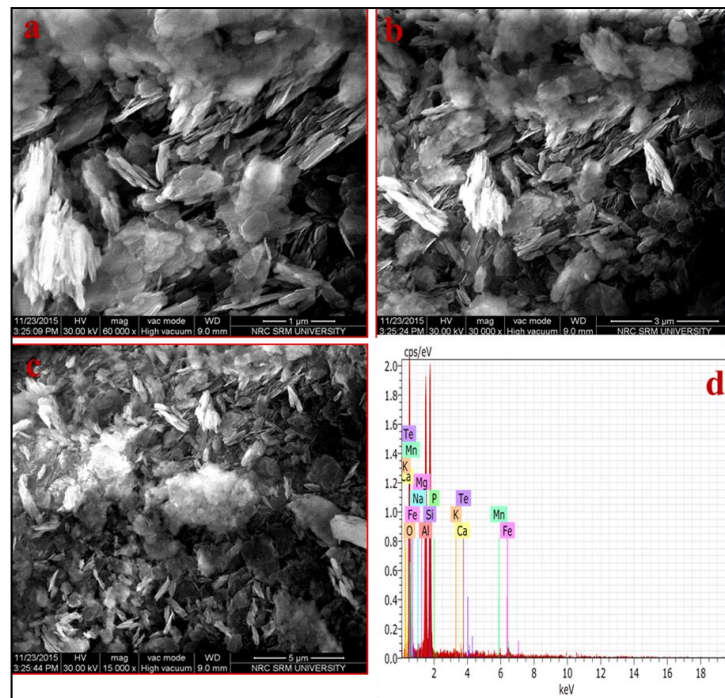


Fig 4: FESEM images of unfired (S₃) and corresponding EDX

Kaolinite platelets are clearly visible, and this is comparatively free from patches (Fig.4c). SEM image of coarse fraction, separated from clay, shows presence of some large particles of quartz. Quartz particle at higher magnification shows spongy like surface. Fig.4 (a-c) indicates that the unfired kaolinite shows isolated quartz crystal and amorphous Fe hydroxides. The samples S₃ are composed of clay particles deposited in a face to face manner with small voids, which show that the clay contains iron impurity, Later they are converted into hematite during calcination. The EDX analysis of sample S₃ (Table 4) shows the dominant elements in the unfired samples are oxygen, silicon and aluminum.

Table 4: Elemental concentration of ceramic body unfired (S₃) and different temperatures

Elements	Atomic weight (%)				
	unfired	800°C	900°C	1000°C	1100°C
O	69.90	67.63	60.35	64.75	64.66
Si	14.24	20.76	20.35	16.00	18.55
Al	14.15	9.05	15.33	15.01	12.90
Fe	0.52	0.48	0.61	0.64	0.41
Na	0.52	1.32	1.57	2.00	2.10
Mg	0.35	0.30	0.17	0.38	0.23
Mn	0.13	0.11	0.07	0.00	0.00
K	0.13	0.34	0.87	0.61	0.63
Ca	0.05	0.01	0.52	0.48	0.38
P	0.01	-	0.02	0.02	0.00
Te	0.00	0.00	0.14	0.10	0.13
C	-	-	-	0.00	-

Table 4 exhibits the presence of oxygen (69.04%), aluminium

(15.04%) and silicon (14.08%) they are exhibited in major portion and iron (0.64%), sodium (Na) (0.26%), magnesium (0.13%) and phosphorous (0.01%) in smaller amount. The alkaline oxides, K₂O and Na₂O, are essential to liquid phase formation. The earth alkaline oxides CaO and MgO, must be maintained in low amount as shown in Table 4, due to the possibilities of excessive fluxing as well as undesired variation on the ceramic color.

3.4 15% Waste Glass added Ceramic Bricks (S₃) Samples Sintered at different temperatures (800, 900, 1000 and 1100°C)

The relationship between mineralogy of the raw materials and phase changes taking place during their sintering under different condition has been examined. The sample with glass content 15 wt. % (Figs.5 to 8) features the presence of a number of large pores. By increasing the temperature from 800 to 1000°C, the pores become smaller and progressively decrease in their lower and higher temperature as shown by Fig.5 and Fig.7, corresponding to sample fired at 800, 900, 1000 and 1100°C respectively. For 900°C, the secondary electrons images of all samples tested; Fig.6 present a similar microstructure, typical for 800°C, with pre-dominantly open and interconnection porosity and clearly distinguished quartz grains. No notable differences are observed among the 800 and 900°C fired samples. Between 900 and 1000°C sintering process takes place which consists in the aggregate composition of particles. This process is not complete. So the ceramic brick bodies are still quite porous. When firing temperature was higher than 500°C, the dehydroxylation of kaolinite begins. Its structure starts to deform, at the same

time, the crystal structure transforms. At higher temperatures, 1000 °C the microstructure is different from the previous ones. Fig.7 shows both open and closed pores and vitrification taken place. It reveals less porous with a higher percentage of closed pores. The formation of glassy phase is enhanced compared to 800 and 900°C, as a continuous vitrified matrix has been developed. FE-SEM image of ceramic bricks (S_3) sintered at 1000°C is presented in Fig.7. The micrographs (Fig.7a-c) indicate a denser more sintered microstructure. When compared with Figs.5&6, the sintered sample S_3 at (1000°C) shows a dense matrix containing some

isolated small voids and very few pores can be seen in the microstructure. Between 900 and 1000°C a considerable decrease in the porosity occurs coinciding with the beginning of vitrification. The little increase in the average pore size as firing was increased from 1000-1100°C is difficult to explain. Although it could be attributed to production, in this interval of temperatures, volumetric changes subsequent to polymorphous change which readjust to available spaces. We also tried to account for this behavior by attributing it to the lack of homogeneity in the raw materials.

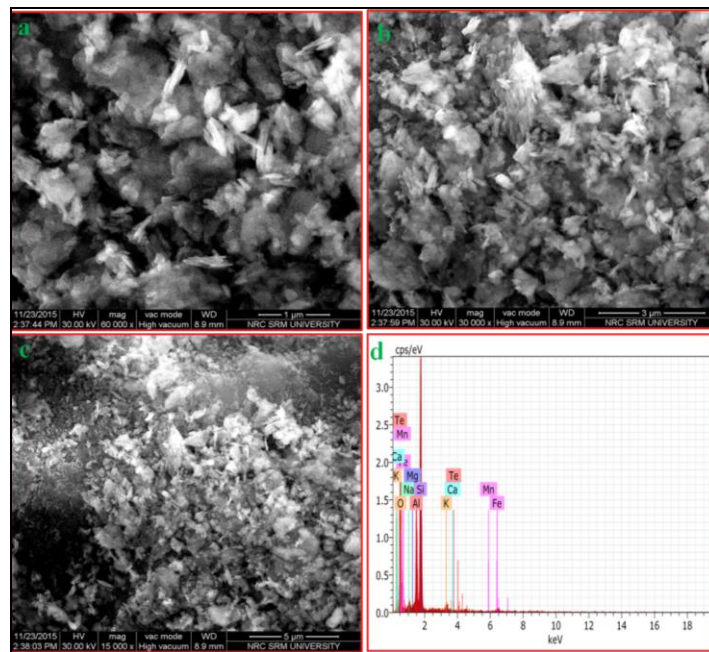


Fig 5: FESEM images of ceramic body (S_3) fired at 800°C with corresponding EDX spectrum

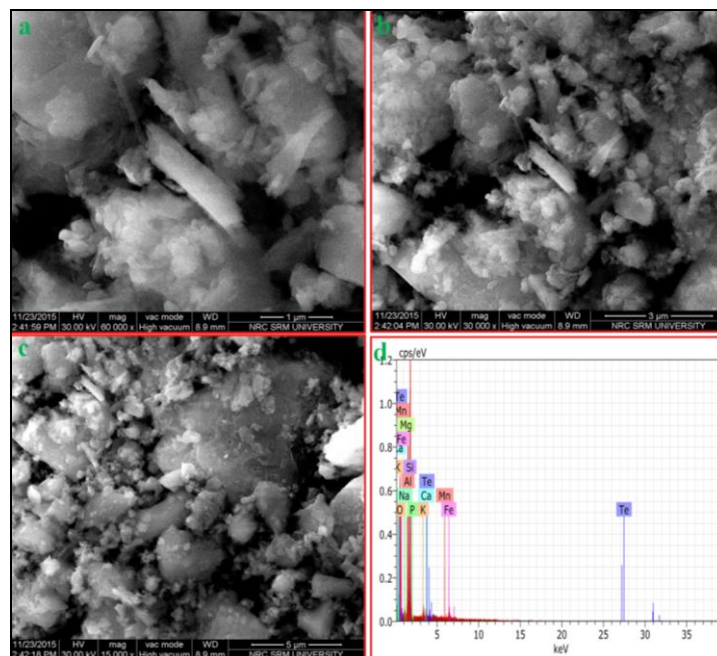


Fig 6: FESEM images of ceramic body (S_3) fired at 900°C with corresponding EDX spectrum

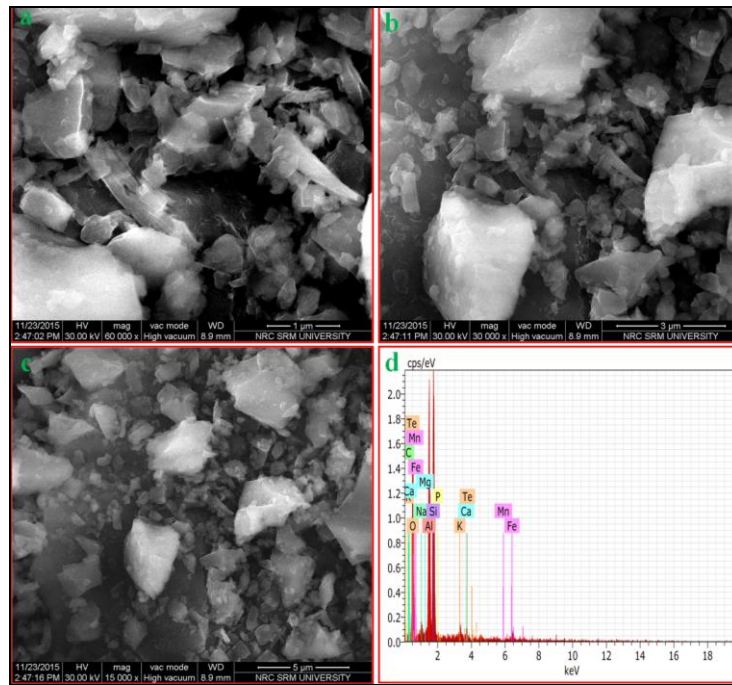


Fig 7: FESEM images of ceramic body (S_3) fired at 1000°C with corresponding EDX spectrum

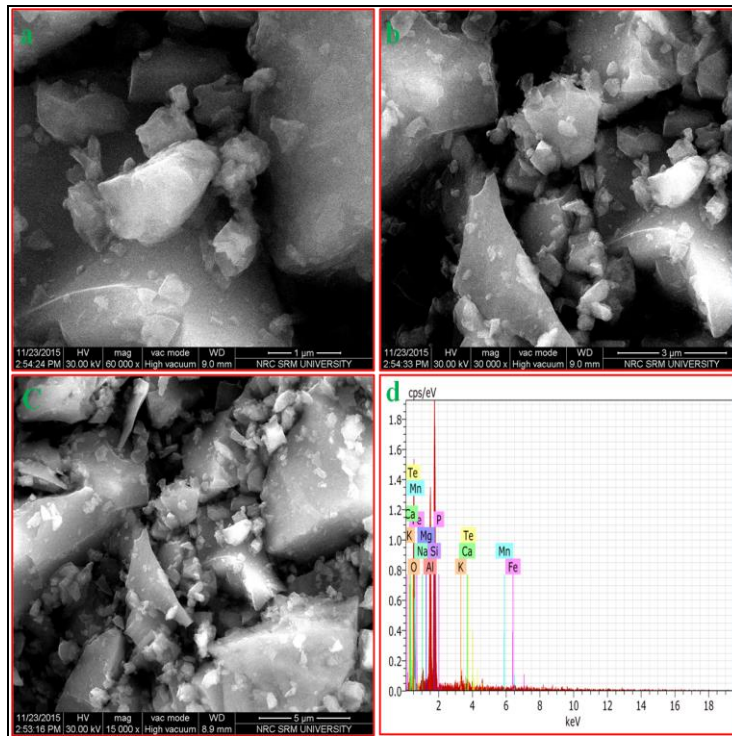


Fig 8: FESEM images of ceramic body (S_3) fired at 1100°C with corresponding EDX spectrum

4. Conclusions

This study has demonstrated the feasibility to use the glass wastes as a material for manufacture of ceramic materials with different compositions. The waste glass powder can be used as an efficient fluxing agent when it is used as an additive mixture. During firing waste glass powder accelerates the densification process, with some effect (lower porosity, water absorption) combined with higher firing shrinkage. Morphological analysis shows the fine interlocked and needle-

like crystals. Finally the physical and mechanical properties, glass wastes and ceramic industrial products are recommended as raw materials in the manufacture of ceramic products as industrial building bricks and elevated decorative materials, reducing the amounts disposed in landfills, the consumption of raw materials and energy costs.

5. References

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