

Utilization of Soda Glass (Cullet) in the Manufacture of Wall and Floor Tiles

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Abstract

Ground Soda Glass (cullet) (SG) was utilized as an additive to a well blended mixture composed of orthoclase feldspar, quartz, grog and three types of local kaolins from three different sites in Egypt. All the used raw materials, including SG, were fully assessed chemically, thermally and mineralogically. SG was added in gradual proportions in order to study the effect of its addition on the physical properties of fired samples (water absorption, bulk density and apparent porosity). The samples were moulded under 20.7 MPa and fired at temperatures ranging from 950 to 1100°C, starting with 3 h as firing time, then decreased to 2 or even 1 h. Both XRD and microscopic investigations (either by polarizing microscope or scanning electron microscope) were used to identify the present phases and degree of vitrification. The results were interpreted due to the presence of liquid glassy phase. The composition containing 23% SG and fast fired at 1100°C for 3 h had the best mechanical strength. The composition containing 33.3% SG, and fast fired for 1 h at 1100°C was recommended for non-glazed floor tiles. Standard tests were performed on this composition in the form of tiles and the obtained results were compatible with standard values. Finally, the use of SG was recommended as an additive for its economical potential. © 1998 Elsevier Science Limited. All rights reserved

1 Introduction

Recently, both environment protection and energy saving have become very important concepts. This

means processes with little or no pollution and industrially utilized and/or recycled wastes. The aim of this work is to utilize the waste glass recycled garbage to reduce the firing temperature and time of the tile bodies due to easy melting and glassy phase formation which flows easily to block pores giving off good quality tiles with low porosity. Attempts to use glass powder from beer bottles¹ or a mixture including waste of scrap glass in a ratio of 18% in facade tiles and 5% in wall tiles, yielding a product of water absorption from 6–8% and lower firing temperature² but with an increase in the coefficient of linear thermal expansion, were carried on. The use of glassy waste containing large amounts of alkaline earth oxides and traditional and local polymineral clays, firing at 900–1000°C was also attempted in order to obtain densely sintered bodies.³

2 Experimental

A basic mixture (BM) for the tile bodies, composed of three types of local Egyptian clays from different locations in addition to orthoclase feldspar, quartz and grog in proper proportions was prepared. This body was completely investigated⁴ before adding the soda glass powder (SG) to it gradually. Table 1 summarizes the chemical, physical properties and X-ray identifications of the used raw materials. Thermal gravimetric analysis was carried on for clays, grog and glass. Samples of cylindrical shape of 1.65 cm diameter were moulded in dry pressing under 20.7 MPa. The specimen weight was 5 g. Fast firing technique was applied. Three firing temperatures 95°C, 1050°C, 1100°C, for 1, 2, 3 h firing time were used. Physical properties of fired samples were investigated. For comparison, two compositions were subjected to XRD. Evaluation by both polarizing

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Table 1. Chemical, physical properties and XRD results of raw materials.

	<i>Bahria Clay</i>	<i>Aswan Clay</i>	<i>Sinai Clay</i>	<i>Orthoclase</i>	<i>Feldspar</i>	<i>Quartz</i>	<i>Grog</i>	<i>Soda Glass</i>
1. Chemical analysis:								
SiO ₂	53.85	53.25	58.64	66.7	96.72	58.07	77.07	
Al ₂ O ₃	15.89	28.92	22.78	19.00	0.91	30.0	0.98	
Fe ₂ O ₃	14.30	3.22	3.69	0.99	1.02	7.0	0.86	
TiO ₂	0.67	0.69	0.55	—	—	Traces	—	
CaO	4.48	1.82	2.24	Nil	2.24	< 2.00	7.47	
Mgo	0.54	0.95	0.43	1.72	1.84	< 1.00	1.61	
Na ₂ O	0.45	0.43	0.33	2.42	0.17	< 1.00	5.20	
K ₂ O	0.87	0.93	0.61	10.96	0.16	< 3.00	4.65	
L.O.I	8.71	9.89	10.56	0.57	0.22	0.10	0.44	
2. Physical properties								
Liquid limit (L.L.) ⁵	24.5	24.5	1.8					
Plastic limit (P.L.) ⁵	13	16	13					
Plasticity index (P. I.) ⁵	11.5	8.2	5.1					
Shrinkage limit ⁶	20.00	18.5	16.28					
Shrinkage ratio	6"	1.79	1.88	1.833				
Volumetric shrinkage ⁶	41.09%	34.12%	26.62					
Specific gravity ⁶	2.79	2.58	2.59					
3. X-ray identification								
	Kaolinite	Kaolinite	Kaolinite	Orthoclase	Quartz			
	Quartz	Quartz	Quartz	Albite		Hematite		
	Hematite			Microcline				

microscope* (universal—equipped with MC 63 photomicrographic camera) and scanning electron microscope† (JEOL, JSM-T₂O) of the recommended sample (BM + 50%SG, fired at 1100°C for 3 h) was performed. The chemical compositions are shown in Table 2. Finally, a sample having tile form of 5 cm × 5 cm was prepared of the recommended composition (BM + 50%SG, fired at 1100°C for 1 h). The powder had to be moistened by air jet to 5–6% humidity, which was necessary for workability.

3 Results and Discussion

3.1 Vitrification parameters

3.1.1 Water absorption

As shown in Fig. 1 a reasonable decrease of water absorption was observed for all samples as the

Table 2. Chemical composition of the two samples containing 20%, 50% soda glass additions (calculated)

Oxide	BM	BM + 20% SG	BM + 50% SG
SiO ₂	62.0	63.8	66.5
Al ₂ O ₃	21.0	17.5	14.2
Fe ₂ O ₃	4.0	4.7	4.0
TiO ₂	Traces	Traces	Traces
CaO	2.0	3.1	4.0
Mgo	< 1.0	1.0	1.1
Na ₂ O	0.50	< 1.5	2.1
K ₂ O	2.0	2.4	3.0
L.O.I.	6.5	6.0	4.8
	100.0	100.0	100.0

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percentage of added SG increases. This decrease was found to be more pronounced on firing at 1050°C and 1100°C. The best result was obtained for the sample containing an addition of 50%SG, fired at 1100°C for 3 h. During handling of this sample, vitrification (formation of glassy phase) was clear, and the edges slightly stuck to the carrying refractory. This result is expected because as the softening point of soda glass is around 720°C,⁷ so, the formation of a glassy phase at 1100°C is obvious. No more increase of soda glass in the composition was attempted.

3.1.2 Bulk density

Figure 2 shows that at low temperature (950°C), there is no marked effect above 10% addition. The effect is more pronounced for samples fired at 1050°C which all show an improvement in bulk density that increases with an increase in % SG. For a firing temperature of 1100°C, a slight increase is observed, then the density tends to decrease for more than 40% SG addition. As a result, it could be deduced that the addition of SG at relatively low temperatures densifies the mixture. At higher temperatures, bloating presumably occurs having for effect to decrease the bulk density as the % SG grows larger. As will be shown later, this result is assessed on examining the SEM photomicrographs.

3.1.3 Apparent porosity

It is shown in Fig. 3 that the curves have the same pattern as in Fig. 1. It is observed that, for temperature 950°C, the decrease in apparent porosity is reasonable for small percentages additions, but nearly stops over 30% SG addition. For the other two temperatures 1050°C and 1100°C, the relation

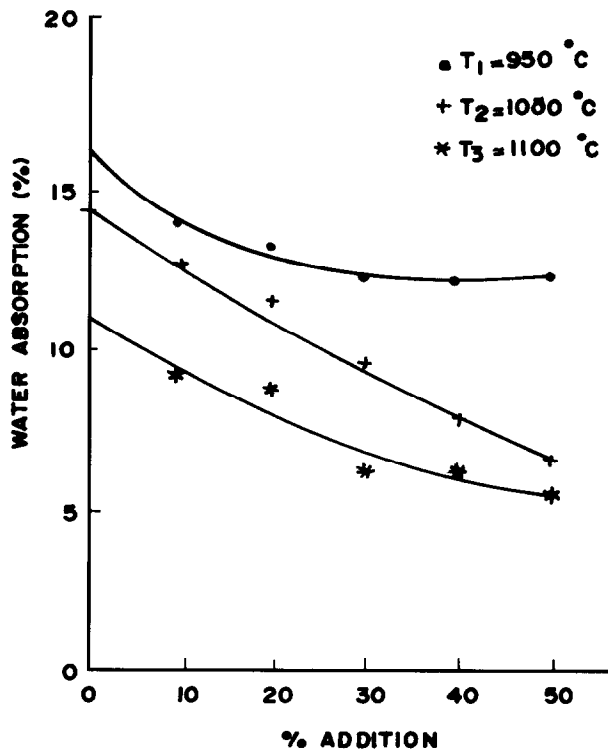


Fig. 1. Effect of firing temperatures on water absorption of different percentage addition of S. G. containing samples and fired for 3 h.

has almost a constant slope with a clear trend of much lower apparent porosity with increasing percentages of SG. This result is also expected due to formation of a glassy phase, especially on firing at 1100°C .

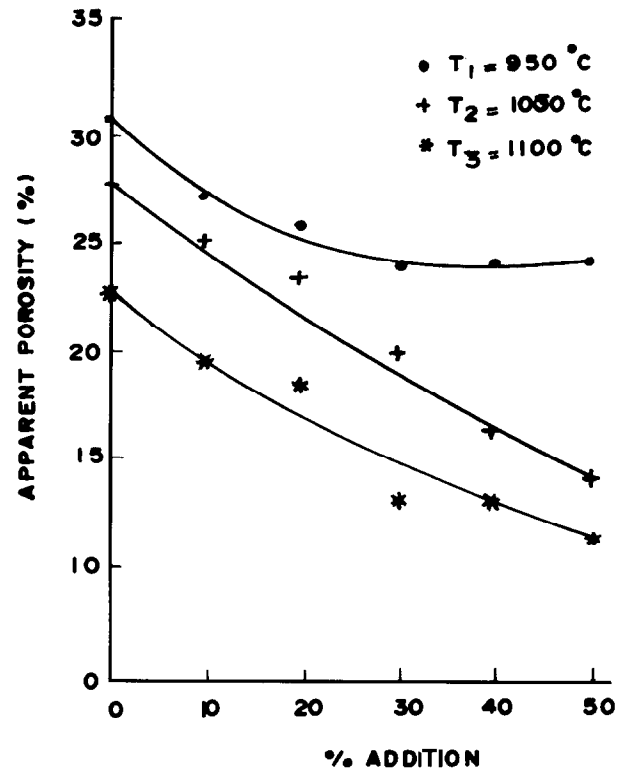


Fig. 3. Effect of firing temperatures on apparent porosity of different percentage addition of S.G. containing samples and fired for 3 h.

In general, the addition of soda glass (cullet) shows a great improvement in the vitrification properties, with an increase of soda glass limited to 50%, to avoid flow under gravity on firing.

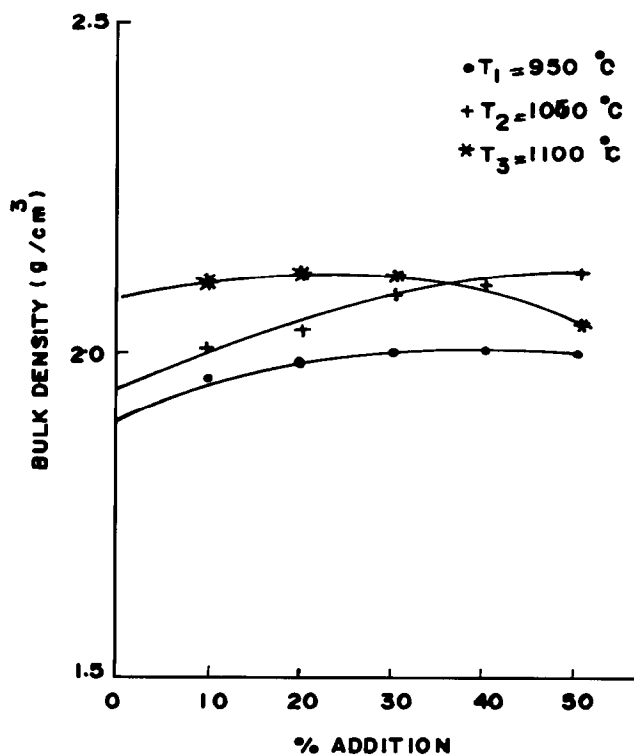


Fig. 2. Effect of firing temperatures on bulk density of different percentage addition of S. G. containing samples and fired for 3 h.

3.2 Effect of firing time

Figure 4 shows the effect of three firing times (1, 2 and 3 h) on water absorption of the compositions containing BM + 40% SG and BM + 50% SG compared to the basic mixture itself, fired at 1100°C . The firing times 2 and 3 h give better effects than 1 h firing on the composition of 40% SG addition. They show no effect on the composition of 50% SG addition which fulfilled the target of water absorption less than 6%⁸ with clear visual vitrification. So, it can be recommended to use the composition of 40% SG addition, fired for 1 h, for normal-duty glazed floor tiles. Meanwhile, the composition of 50% SG addition fired for 1 or 2 or 3 h is recommended for normal duty non-glazed floor tiles.

On the other hand, Fig. 5 shows the effect of the same firing times on the bulk density of the same two compositions, fired at 1100°C . Differences from the basic mixture bulk density are small. Firing for 1 and 2 h gives very close bulk densities of the two mixtures containing SG. In case of BM + 50% SG, fired for 3 h, the bulk density of the 50% addition mixture is lower than both the basic mixture itself and the composition containing 40% SG addition.

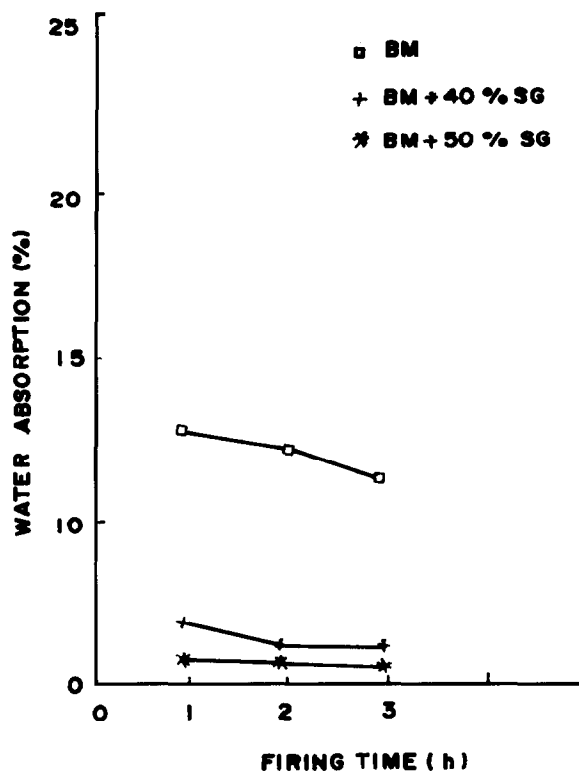


Fig. 4. Effect of firing time on water absorption of samples containing 40% and 50% SG, fired at 1100°C.

Finally, Fig. 6 shows how firing time affects the apparent porosity of these compositions, fired at 1100°C. For the composition of 40% SG addition, there is a difference between 1 and 2 h firing, but no difference was observed between 2 and 3 h. For the composition of 50% SG addition, the firing times

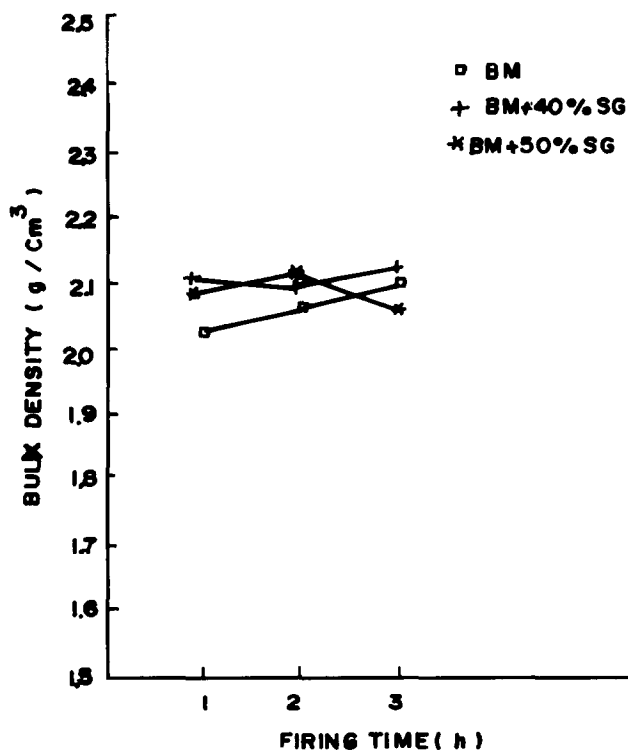


Fig. 5. Effect of firing time on bulk density of samples containing 40% and 50% SG, fired at 1100°C.

show no difference. In general the apparent porosity of these two compositions shows a remarkable difference towards improvement on increasing firing time. These two compositions were recommended for use in view of their low water absorption.

3.3 X-ray results

Two compositions of 20% SG and 50% SG additions, fired at relatively low temperature (950°C) for 3 h, were exposed to XRD. Their identifications are compared. The existing phases are the same. They include quartz, cristobalite, plagioclase and hematite. These minerals were also identified for the same compositions fired at 1100°C for 3 h. Table 2 shows the calculated chemical compositions of these samples.

It was not possible to interpret the X-ray results using the relevant phase diagram since the fluxing oxides K_2O , Na_2O , Fe_2O_3 , CaO and MgO are present in comparable ratios and any attempt to simplify the situation to a ternary or even a quaternary system did not give pertinent results. However, the presence of a liquid glassy phase is expected to be present in large amounts due to the presence of large proportions of fluxes. This will be apparent on discussing SEM micrographs.

3.4 Microscopic investigations

3.4.1 Polarizing microscope

The polished surface photomicrograph shows that the glassy material is abundant (Fig. 7) and the

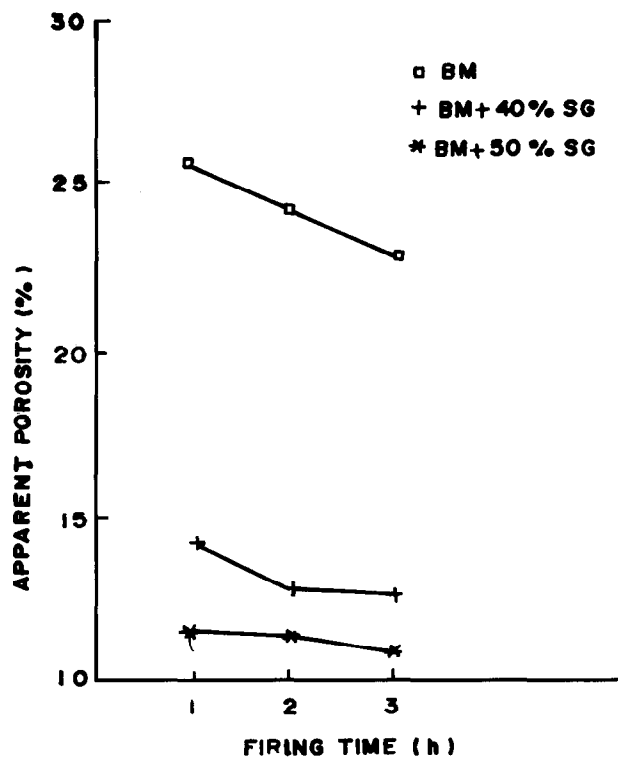


Fig. 6. Effect of firing time on apparent porosity of samples containing 40% and 50% SG, fired at 1100°C.

iron oxide phases are rarely encountered and poorly crystallized. The thin section (Fig. 8) shows that the glassy material (most probably calcedony) is abundant and the outer boundaries of grog are diffused within the glassy groundmass.

3.4.2 Scanning electron microscope (SEM)

The SEM photomicrographs (Figs 9 and 10) show that at least two different types of crystals are seen to be present in a vitrified matrix. Quartz is abundant and the other phase is presumably feldspar. Large bubbles appear due to vitrification in which some crystals have sunk and the vitrous phase is evident.

The phenomenon of bloating is assessed by scanning electron microscope.

3.5 Compressive strength results

Figure 11 shows three curves fitted for the data of compressive strength (in MPa). For the samples fired at 950°C, the curve shows an initial increase

in strength as SG is added. The value rises from 20 MPa to about 39 MPa as 10% SG is added. This is in accordance with Fig. 3 where the value of porosity drops to an almost constant value over 20% SG addition. This means that the increase in compressive strength at that low firing temperature is mainly due to reduction in porosity.

At 1050°C there is a gradual increase in strength from 42 MPa (0% SG) to 57 MPa (50% SG addition) in harmony with the decrease in porosity observed in Fig. 3, although, as the % SG addition reaches 50%, there is a tendency for the compressive strength to reach a fixed value. This may be due to the effect of the low strength of the glassy phase formed as its amount increases. This effect is more pronounced for the sample fired at 1100°C which reaches a peak in strength at 30% SG addition followed by a gradual decrease. This result agrees with Abdrakhimov^{9,10} on firing at 1100°C of such mixtures. In view of porosity results of Fig. 3, it is clear that as the % glassy phase largely increases (at 1100°C), the reduction in porosity

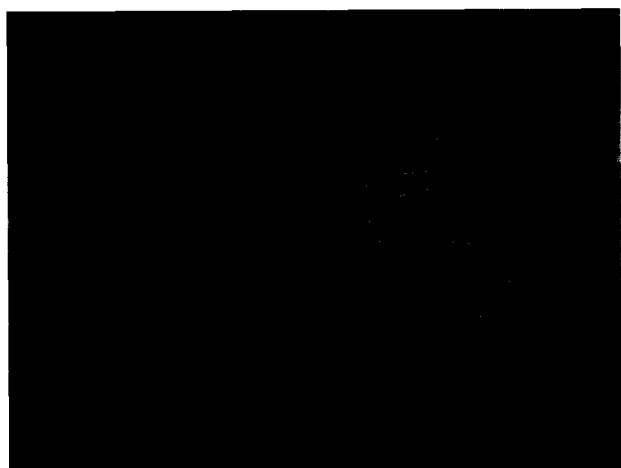


Fig. 7. Photomicrograph of polished surface showing poorly crystallized hematite phase highly diffused in the glassy groundmass (X500).

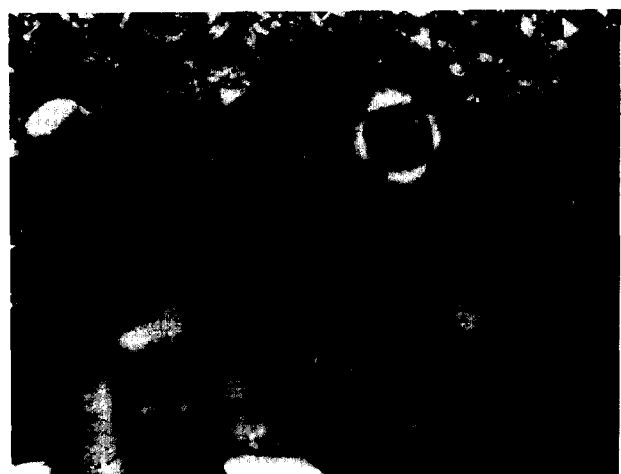


Fig. 8. Photomicrograph of thin section showing a glassy silica phase (most probably calcedony) developed in the groundmass (X 125).



Fig. 9. Scanning Electron Microscope photomicrograph (X500).



Fig. 10. Scanning Electron Microscope photomicrograph (X1000).

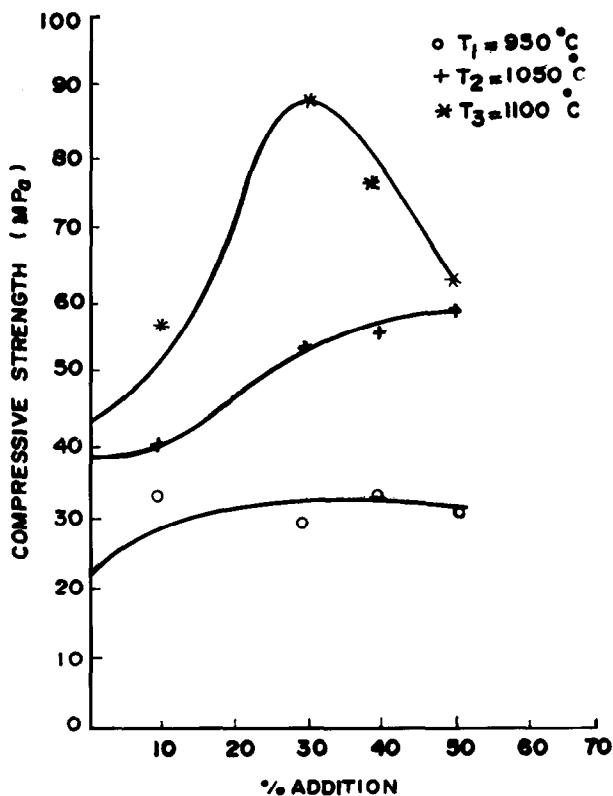


Fig. 11. Effect of firing temperatures on the compressive strength of mixtures containing S.G. and fired for 3 h.

does not play a marked role in sustaining high values of strength.

On the other hand, since water absorption tests carried out at 1100°C (Fig. 1) did not show an appreciable difference when firing was effected at 1 or 2 or 3 h, it was necessary to study the effect of firing time on the compressive strength. Figure 12 shows that it is unnecessary to use prolonged firing time over 1 h since the values of compressive strength exceed those obtained after 2 h soaking time.

3.6 Firing shrinkage

The linear shrinkage ($\Delta L/L$) for samples in the form of tiles (50×50 mm) was measured. The obtained values for average shrinkage for the samples containing 50% SG addition and fired at 1100°C for 1 h was 3.78% compared to 5.1% for the basic mixture fired at 1200 for 3 h. In addition to improving firing shrinkage, this limit is within the safe limits for industrial production.

3.7 Simple economic assessment

From the results of the experimental work, recommended economic compositions utilizing the addition of soda glass were 40% SG addition (23% SG by the composition weight), fired at 1100°C for 1 h (6.9% water absorption)⁸ for glazed floor tiles and 50% SG addition (33.3% SG by the composition weight), fired at 1100°C for 1 h (5.6% water

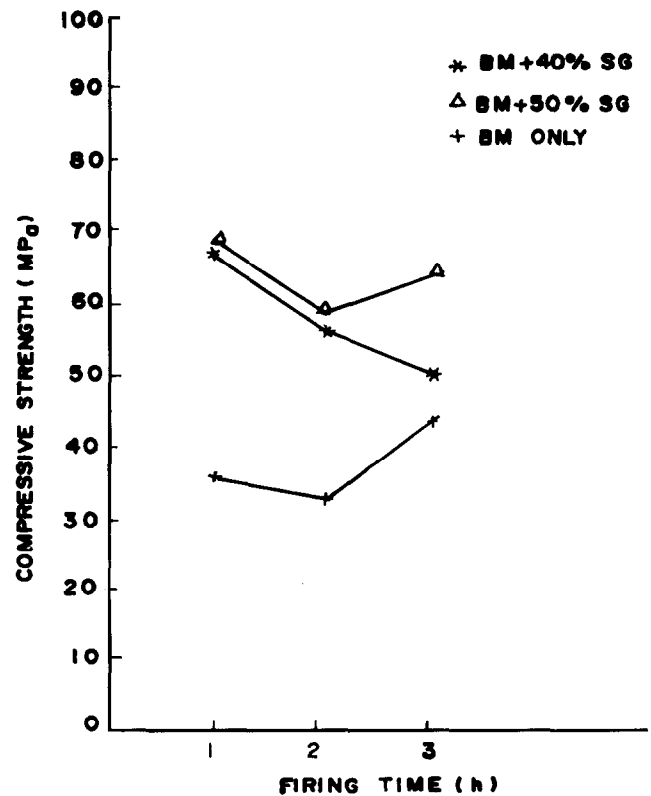


Fig. 12. Effect of firing time on the compressive strength of mixtures containing S.G. and fired at 1100°C.

absorption)⁸ for non-glazed floor tiles. Referring to Bakr,¹¹ where the raw materials of the body represents 17.8%, fuel represents 9.6% and electricity represents 17% of the total cost in case of fast firing, the economical utilization of SG depends on the fact that it saves fuel and electricity as it is considered as a strong flux costing less than feldspar.

3.8 Quality assessment

The composition of 50% SG addition fired at 1100°C for 1 h was tested in the form of non-glazed tiles (50×50 mm) according to ES 293.¹² The results compared with commercial tiles were encouraging to be used as glazed wall tiles and it showed good resistance to acids and alkalis. Visual examination of the samples prepared in tile form (50×50 mm) showed no bubbles on the surface.

4 Conclusions

The addition of soda glass is economically recommended in the existing tile manufacturing plants which use such clay compositions. The saving will be either in fuel consumption or in raw materials if expensive fluxes are used. The composition containing 33.3% by weight SG, gives tiles of reddish deep brown colour, and is recommended for non-glazed floor tiles.

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