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Recycling Of Waste Glass As Aggregate For Clay Used In Ceramic Tile Production

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ABSTACT:*The need to convert waste to wealth (recycling) for economic benefit is of great concern to researchers. Investigation on the utilization of waste glass (cullet) as sintering aid in the production of ceramic tiles was carried out using clay as the base material in order to save energy during production. Large deposits of clay exist in the country which needed to be harnessed for proper use in the production of ceramic wares. Cullet is one of the most common silicate wastes and large amount of these waste are recycled by the glass industries while some are dumped in the immediate environment. In this research, soda-lime glass grinded passing through ranges of mesh φ_1 (0.088 to 0.125 mm) and φ_2 (0.037 to 0.088 mm) was prepared and added to the clay as aggregates in the following proportions (0, 5, 10 and 12%) in order to evaluate their effect on the sintering of the ceramic wares and these were characterized for various properties. The content of clay, sand and silt were also determined using the pipet method. Square tiles with standard cross section (2.5cm x 2.5cm) containing 0, 5, 10 and 12 % of glass cullet powder respectively were fired at five different temperatures (800, 900, 1000, 1100 and 1200^{o}C respectively) at a soaking time of 1hour at 10^{o}C/min. Cold crushing strength (CCS), water absorption, fired linear shrinkage, density and apparent porosity tests were conducted on the prepared square tiles. Test results show that shrinkage increases with the increase in glass content at 800^{o}C and all other properties aforementioned above improved. These changes were more exhibited at temperatures higher than 900°C, especially with 10 and 12% glass powder. 10% cullet composition which was fired at 1045°C had CCS value of 11MPa and no pinholes or cracks were recorded which is more appropriate for the mix ratio used. This study showed that, waste glass was not only utilized in clay products but also gainful for production of ceramic tiles by reducing firing temperatures. Addition of 10% percentage of waste glass to the clay reduced the firing temperature by 55^{o}C (about 3.9%) making it economical to use soda-lime-glass as sintering aid in the production of ceramic wall tiles.*

KEYWORDS: *ceramic tiles, waste glass, recycling, sintering, soaking time*

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I. INTRODUCTION

The ceramic industry is based mainly on the production of clayey ceramic materials for use in the civil construction, such as, bricks, ceramic blocks, wall tiles, roofing tiles and pipes (Santos, 1989). Common clays are basically raw materials used for the manufacture of ceramics. Ceramic tiles are widely used in homes as wall and floor tiles, WC, Jugs, plates, etc. The main constituent of clay is silica (SiO_2) and alumina (Al_2O_3) used in the production ceramic wares with the addition of other materials to attain the desire product standard. Other materials, such as, glass wastes which were sometimes being disposed in landfills have been used as substitutes of conventional raw materials in ceramic production (i.e. clay, sand and feldspar) for economic purposes [1]. This is aimed at converting waste to wealth leading to zero waste.

Zero Waste America, defines waste as "a resource that is not safely recycled back into the environment or the marketplace." This definition takes into account the value of waste as a resource, as well as the threat unsafe recycling can present to the environment and public health.

Glass can be recycled a million times over to produce bottles and jars of the same high quality every time. However, to keep producing the best end product, the recycled materials must be of a high quality. Minute amounts of some materials mixed with the glass during collections for recycling can cause contamination. Contamination of as little as five grams per ton can result in valuable glass going to landfill [2]. The most familiar type of glass used for centuries is Soda lime glass made up of about 75% Silica (SiO_2) plus about 15% Sodium Oxide (Na_2O),12% Calcium Oxide (CaO) and several minor additives [3].

Glass plays an essential role in science and technology. Their chemical, physical and optical properties make them suitable for various applications, such as, flat glass, container glass, optics and Optoelectronics material, laboratory equipment, thermal insulator (glass wool) reinforcement concrete and glass art (art glass, studio glass). It resists chemical interactions, does not leach chemicals like plastics and it can withstand heat and cold.Inpractice,therearetwo classes of waste broken glass, namely: a) Preconsumer glass; and b) Postconsumer g lass [3]. The Preconsumer glass waste is the glass that has defaults during production in the factory; while, Postconsumer glass waste is the one that has been produced and sold out for use before the defects.

In this research, the post-consumer glass waste was used with clay for the production of ceramic wares. In recycling, glass waste should be separated by chemical composition which depends on the end use and the processing capabilities. It is also very important to separate the glass wastes into different colors, since glass retains its original color after recycling. The glass component in municipal waste is usually made up of bottles, broken glass ware, light bubbles and other items made of glass [4]. Glass recycling uses less energy than manufacturing it from the raw materials (sand, lime and soda). Every metric ton of waste glass recycled into new items saves about 315 additional kilogram of carbon dioxide from being released into the atmosphere during the manufacturing of new glass [4].

More than 95% of all manufactured glass is made from sodium oxide, calcium oxide, and silicon dioxide, commonly referred to as a soda-lime-silica glass [5]. Soda lime glasses are vitreous silicates which are formed during the maturation of clay bodies (during firing) and these serve as fluxes, reducing clay body maturation temperature. The glass wastes can be used to substitute the conventional flux materials, such as, feldspar and feldspathoid rocks used in the composition of ceramic masses [6]. The different coloured glass wastes collected (brown, green, etc.) when crushed into fine particles can be used as a sand-substitute for sandblasting, water filtration, aggregates, bricks and tiles production. These saves natural resources (such as sand, soda ash and limestone); saves energy, since recycled glass melts at a lower temperature reducing the maturation temperature of the clay body and reduces pollution caused by emissions of gases like CO_2 into the atmosphere. There are no by-products and wastes generated during the process [7]. Recycling of the materials to be added to clay for the production of ceramic tiles may lead to economic growth and employment opportunity for unemployed. Recycling of broken glasses is a complex processing which include; washing, milling, sieving, mixing, drying, firing, etc., [6].

Previous studies have shown that the densification of the Ba (Mg1/3Ta2/3)O3 (BMT) ceramics perform well at lower temperatures of 1300–1350 °C, this could be achieved with the addition of 3-6 wt.% MCAS ($MgO–CaO–Al_2O_3–SiO_2$) glass [8]. Studies using soda-lime glass in replacement of soda feldspar in typical porcelain stoneware bodies (up to 10 wt.%) that underwent a laboratory simulation of tile making process, shows that, soda-lime glass had no significant effect on semi-finished products, but it influenced remarkably on the firing behavior of the ceramic products. The final stage of sintering are optimized via residual porosity and grain size and are believed to control mechanical characteristics with the aid of temperature and time of sintering [9]. Elimination of residual porosity at final stage of sintering may be accompanied by excessive grain growth and this may take place as a result of secondary recrystallization during solid state sintering [9]. Promising results have also been obtained using glass cullet waste to substitute the conventional flux materials[6] , as well as cathode ray tube (CRT) glass[10], different industrial residues [11] and various volcanic rocks has been used [12]. Research has also shown that 5–10 wt.% of cullet or CRT gave the optimal amount of the replacement with clay [10]. The crystalline proportion of the final ceramic products may be increased by using glass-ceramic frits or compositions with high crystallization trends [13]. As a result, several studies related to the substitution of conventional raw materials in tile-making (i.e. clays, sands and feldspars) with other natural resources or industrial wastes were carried out during the last decade[1, 14].

Due to re-crystallization processes during the heat treatment and as the amount of residual amorphous phase decreases while increasing the crystalline proportion, a positive effect on the mechanical properties of ceramic products is achieved [15]. Glass addition to other types of ceramic products has also contributed to their sintering aid as seen in the test with two commercially available glasses, $PbO–B_2O_3–SiO_2$ (GA-9) and $ZnO–B_2O_3–SiO_2$ (GP-032) which have been found to improve the sintering property of $La_4Ti_9O_{24}$ ceramics by lowering the sintering temperature of $La_4Ti_9O_{24}$ ceramics to 1000 °C instead of 1350 °C [16]. The present study was directed towards determining the possibility of introducing waste glass from urban areas into a ceramic tile body mix, as partial substitute of the fluxing agents.

II. MATERIALS AND METHOD

The materials used for this study include: clay materials and broken bottles (cullet). The clay materials used was obtained at a depth of 2.0m using digger and shovel from Nkwo-Alaike (NK) deposit in Ehime Mbano local government area, Imo State. The ceramic mass was air dried and then milled in a ball milling machine.

Part of the sample was assayed for particle size distribution to determine the content of the clay, sand and silt, using the pipet method [16]. The rest was passed through a set of screens to determine particle size distribution to help in the preparation of the mixtures for the specimen. The brown beer bottles (350 ml long neck type) collected were washed with detergents to remove contaminant, air dried, broken down into pieces and pulverized in a ball milling machine for 6 h. The powder was passed through sieves of 0.037, 0.088 and 0.125 mm to obtain two different particle sizes: (φ_1) 0.088 to 0.125 mm and (φ_2) 0.037 to 0.088 mm. These particle sizes were chosen considering the particle size distribution obtained for the ceramic mass. The NK clay and glass powder were dried in an oven for 24 h at 110^{o}C. The mixtures of NK clay and glass powder were homogenized in a ball milling machine for 6 h. Four mixtures were prepared containing 0, 5, 10 and 12% by weight of glass powder and 15wt% of water was added to each sample mix for two particle sizes; $\varphi_1$and φ_2. Six specimens of approximately 20g were made from each percentage sample of the glass, with dimensions of (2.5cm x 2.5cm). A hydraulic press whose compactive strength varied between 10 to 50MPa was used for pressing the specimen to produce the tiles. The specimen were cleaned, labeled, measured and weighed. After marking and measuring (length and weight) of the specimen, they were placed in an oven at 110°C for 24 h to dry. They were placed in a desiccator until the specimen reached the temperature 25^{o}C. The specimen were weighed again and measured. The oven-dried specimens were fired in a kiln at 800, 900, 1000, 1100^{o}C and 1200^{o}C to produce the biscuit wares at 10°C/min for 2 h. The biscuit wares were allowed to cool to ~ 60°C; and placed in a desiccator until they reached the room temperature. Their dimensions and weight were then determined again using caliper and analytical balance. Afterward, the fired linear shrinkage (FLS) and loss on ignition (LOI) of the biscuit were calculated. Cold crushing strength (CCS) using a compressive tester (Testometric M-500-25KN) was determined after firing the specimen. Apparent specific weight (ASW), water absorption capacity (WAC) and apparent porosity (AP) were determined after firing, using the Archimedes method (hydrostatic balance) [17]. Other test carried out includes; bulk density (BD), as well as the chemical analysis using Atomic Absorption Spectrophotometer (AAS, PG 990 AFG).

III. DISCUSSION OF RESULTS

A typical average chemical content of the oxides in the cullet is as shown in Table 1 below:

Table 1: Chemical Analysis of Glass Sample (Soda-lime glass)

S/N	Parameters	Level Detected (%)	Level Detected (%) {BREF, 2009} Standard
1.	Silicon oxide ($Si0_2$)	72.08	71-75
2.	Aluminum oxide (Al_20_3)	1.15	-
3.	Magnesium oxide (Mg0)	0.86	-
4.	Calcium oxide (Ca0)	12.09	10-15
5.	Zinc oxide (Zn0)	0.03	-
6.	Sodium dioxide (Na_20)	15.02	12-16
7.	Potassium oxide (K_20)	0.03	-

The clay sample studied in this research had the following chemical composition (oxide %): SiO_2 (56.70), Al_2O_3 (26.55), Fe_2O_3 (1.76), CaO (0.21), K_2O (0.08), TiO_2 (1.68) and MnO (0.11) which indicated that it is a typical kaolinitic composition [1, 14]. Therefore, the presence of alkaline and the alkaline earth oxides in the glass composition will act as fluxing agents helping the sintering process of the ceramic material with addition of glass powder [14].

3.1 Particle size distribution (PSD)

The PSD was obtained in triplicate without extraction of organic matter (OM) and the results presented as the average of the three measurements for each fraction as shown in Table 2. The clay fraction found in the sample was higher than the recommended range for the production of ceramic wall tiles, allowing it to be mixed with non-plastic material, such as, glass is required [18].

Table 2: Particle Size distribution for tile mass

Fraction	Concentration (%)	Recommended (%)*
Sand	23.10	20-40
Silt	13.01	20-50
Clay	51.10	30-40

* Pracidelli and Melchiades, (1997)

3.2 Loss on ignition (LOI)

As observed from Figure 1, LOI increased with sintering temperature for all the mixtures. At 1000°C and higher, the loss of mass was constant, indicating that the reaction (dehydration, burning-off of the impurities, etc.) occurred at the lower temperatures and was almost completed. Previous studies have shown that

Kaolinitic clays have greater loss of mass at temperatures below 600°C in which there is a loss of moisture; structural water (hydroxyls) in clays, hydroxides and organic matter [20]. Increasing the percentage of cullet in the mixtures φ_1 and φ_2 reduces loss of mass since it substitutes for the clay as can be observed in Figure 1. The moisture content of the clay sample was 9.5% which was within the recommended range of 8-10%, an indication that a moderate addition of water to the mixtures of φ_1 and φ_2 was required during beneficiation.

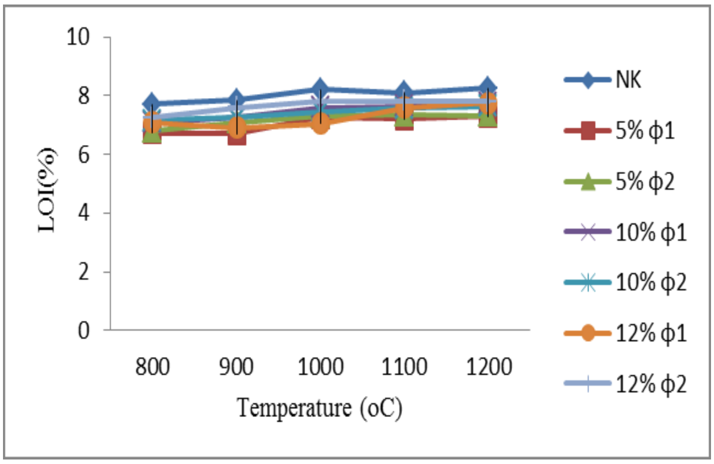

Figure 1: Loss of mass on ignition to ceramic mass with glass powders (φ_1 and φ_2).

The effect of particle size of the glass powder was found to be in the range 0.16- 0.87%; which is less than 1% in loss of mass between the φ_1 and φ_2 mixtures.

3.3 Fired Linear Shrinkage (FLS)

From Figures 2, at 800°C, for all concentrations of glass powder φ_1, 10 and 12% for glass powder φ_2, the FLS was not altered. For φ_2 with 5% glass powder, the FLS decreased slightly. At 900 and 1000°C, φ_1 with glass powder of 5% in the specimen, the FLS decreased. At 1000°C and higher, there was a tendency for linear shrinkage to increase for samples with glass powder (φ_1 and φ_2) due to the presence of fluxing oxides. The softening warped the samples, mainly those samples sintered at 1200°C, impairing the measurements used in the determination of FLS, resulting in errors in the measurements. Between 950 and 1225°C, vitrification occurs with kaolinitic samples, due to the release of silicon\oxide (SiO_2) which reacts with free oxides, mainly alkaline, alkaline earth and iron oxides, forming glass [17]. Some of these oxides are present in clays; some are released in the breaking of the structure of clay minerals, and others, mainly alkaline oxides that are present in glass powder. Even for samples fired at 1200°C, FLS was below the recommended maximum limit of 6% [19]. From Figure 2**(**a, b**)**, FLS was in the range of 0.15-3.4% at temperatures < 1000°C which indicated a small influence on FLS. The φ_2 glass powder which was finer, had FLS values that was higher than φ_1 at temperatures <

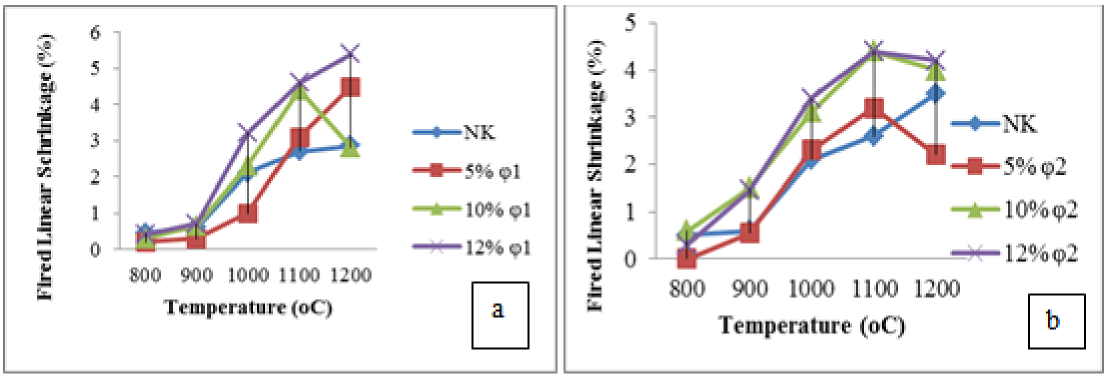

Figure 2(a,b): Linear firing shrinkage as a function of firing temperature for ceramic probes containing 0, 5, 10 and 12% glass powder, φ_1 and φ_2

3.4 Water Absorption Capacity (WAC)

From Figure 3(a, b), it was observed that WAC diminished with increase in concentration of glass and firing temperature of the samples. Glass powder reduced WAC for the biscuits, all of the biscuit samples showed a WAC of < 17%. Therefore, at the firing temperatures (800 to 950°C) all samples have WAC in according with limit values of 18% [20]. At 1100°C, WAC was less than 10%, and at 1200°C, the samples with 10% glass powder (φ_1 and φ_2) showed a WAC of < 7.5%. All these values are below the recommended maximum value of 18% for the production of wall tiles [20]. It can be observed from the Figure 3(a, b), that the blended biscuit samples had lower WAC in the range of 5-15.2% while the biscuit without glass powder was in the range of 10.2-16% at 800-1200°C respectively. An indication that the addition of glass powder at controlled proportion will drastically reduce the firing temperature and consequently the time of firing.

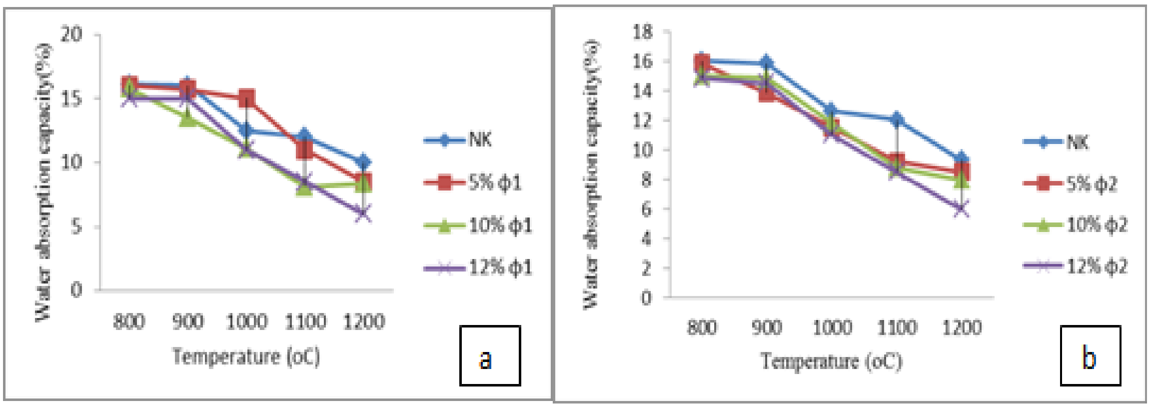

Figure 3(a,b): Water absorption capacity (WAC) of ceramic mass with 0, 5, 10 and 12% glass powder, φ_1 and φ, as a function of firing temperature.

3.5 Apparent porosity (AP)

AP tended to decrease since a greater densification of the sample occurred as the firing temperatures increases. The apparent porosity of the biscuit was approximately 30% at 800 and 900°C. At these temperatures, the addition of the two glass powders (φ_1 and φ_2) showed a tendency for a small decrease in the AP value when compared with the value of the unblended biscuit. At 1000°C and higher, there was a continuous fall in AP for the two glass powders, up to 1200°C, where there appears to be a difference between percentage of glass incorporated into the sample. At 1200°C, a decrease in AP (AP < 15%) was observed for 12% glass powder (φ_1 and φ_2) due to the burning off of some impurities. Low percentage of AP enhances the entrapping of gases in the ceramic products which may be responsible for some pinholes on the glazed tiles and this phenomenon will adversely affects the life span of the tiles when in use [1].

3.6 Apparent specific weight (ASW)

From Figure 4(a, b), it shows that, there was an increase in ASW as the firing temperature was increased and it improved with addition of 10 and 12% of the powder glass [19]. All the samples showed ASW values greater than the recommended minimum value (1.7g/cm^3) an indication that the addition of glass powder improved the ASW.

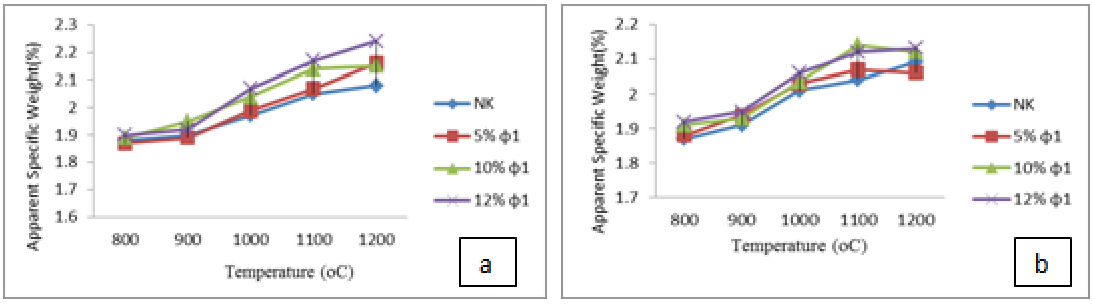

Figure 4(a, b): Apparent specific weight (ASW) of the ceramic mass containing 0, 5, 10 and 12% glass powder, φ_1 and φ_2, as a function of firing temperature.

3.7 Cold Crushing Strength (CCS)

As observed in Figure 5(a, b), the average CCS determined was greater than 5 MPa, for all the samples., thus, the result of the analysis is favorable. Firing at 900°C and higher; all the biscuit samples showed a CCS greater than 6.5MPa, the minimum limit values for ceramic tiles [17]. Up to 1000°C, the graphs show that, the values are equal for the biscuits with and without glass powder. From 1100 to 1200°C, an increase in

CCS is seen. At 1200°C, the samples with glass powder φ_1 show a CCS greater than that of samples with glass powder φ_2, and the addition of 12% glass powder resulted in a higher CCS value (in the order of 17MPa). 10% cullet composition which was fired at 1045°C had CCS value of 11MPa and no pinholes or cracks were recorded.

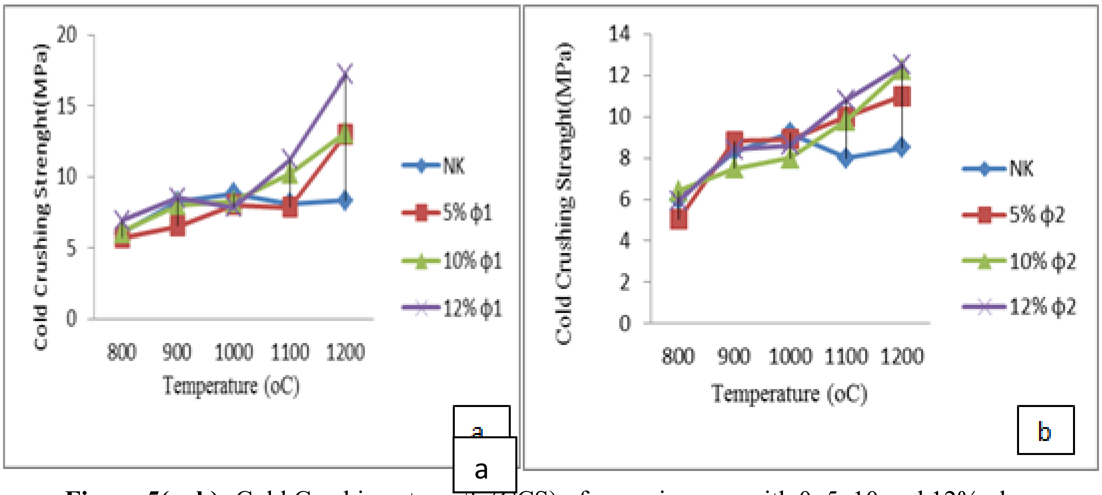

Figure 5(a, b): Cold Crushing strength (CCS) of ceramic mass with 0, 5, 10 and 12% glass powder, φ_1 and φ_2, as a function of firing temperature.

IV. CONCLUSION

This study showed that, waste glass was not only utilized in clay products but also gainful for production by reducing firing temperatures. Addition of 10% of waste glass can reduce the clay firing temperature by 55^{o}C. Biscuit firing of φ_1 and φ_2 without cullet addition came out without cracks at 1100^{o}C. Blending with cullet showed fewer pinholes compared with the biscuit without cullet addition, especially, for biscuit fired at 1045^{o}C as against the 1100^{o}C for the unblended biscuit however, the addition of 10% gave the best result without pinholes. Glass powder can be used for improving the properties of wall tiles by the ceramic industry. The addition of 12 and 10% glass powder, in general, showed better results compared to 5%. The variation in the particle size of the glass powder does not change significantly the ceramic properties. The addition of cullet to clay for the production of ceramic tiles had economic value because the firing temperature of the biscuits was reduced by about 3.9%, an indication that the addition of cullet to clay at controlled proportion will drastically reduce the firing temperature and consequently the time of firing.

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