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DEVELOPMENT OF A SUSTAINABLE MID-TEMPERATURE GLAZE FOR CERAMICS TABLE WARES USING BOROSILICATE GLASS AS A MAJOR FLUX

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ABSTRACT

Based on the experiment carried out in this research and the results obtained from the research, the research did justice to the key points in this research. The three key points that need to be stressed are sustainability, mid-temperature, and food safety. In this research 20 glaze sample was formulated using computer glaze software ("GlazeMaster"). While using the glaze software, three component was consider in the glaze formulation; Alumina silica ratio, Level of alkaline and Percentage of flux in the glaze recipe. 15 glaze recipes were first formulated and fired, at temperature range of 1080ºc, 1110ºc, and 1140ºc. After the firing only glaze sample 10 which compose of (Ball clay 15, Borosilicate 50, Feldspar 11, Whiting 20 and Zinc Oxide 10) gave a glossy surface at temperature of 1080ºc while sample 13, 5, 6,14,7,12,9 and 8, gave a matte surface at temperature of 1140ºc. Glaze sample 11, 15,4,3,2 and 1 did not melt between the temperature ranges of 1080ºc to 1140ºc. Another 5 glaze sample was formulated after the result gotten from the first 15 glaze recipe was not satisfactory. The new 5 glaze recipe was named N1, N2, N3, N4 and N5. The glaze was first fired to 1110ºc, after the first glaze firing glaze sample N1. N2, N3, N4 and N5 all gave matte surface. The second glaze firing was taken to 1140ºc, after the glaze firing glaze sample N1, N2 and N3 gave a glossy surface while glaze sample N4 and N5 gave clear matte. After all the glaze firing glaze sample 1, N1, N2, and N3 gave the best result between temperature range of 1080ºc and 1140ºc. All the glaze sample craze on clay body, of ratio of Ball clay to Kaolin, ratio 1:1, while the glaze sample did not craze on clay body, of ratio of Ball clay to Kaolin, ratio 2:1 and 3:1.

KEYWORDS: Glaze, Sustainability, Mid-Temperature, Food Safety, Borosilicate glass, Glaze Software, and Glaze firing.

1.0 INTRODUCTION

Ceramic glazes are glassy coatings applied to ceramics. It serves as a decorative coating, a means of keeping liquids out, and a means of preventing pollution from adhering to items. The surface is also more durable. Stoneware and porcelain are also coated with glaze. Additionally to their functionality, glazes can be applied to surfaces with different finishes, including glossy or matte finishes and different colors. Inscriptions, carvings, or painted designs can also be enhanced by glazes. Although ceramic glaze akin to glass, it actually can dissolve over time when in contact with liquids if its chemistry is not balanced and stable. The significance of glaze application in the finishing of traditional ceramics cannot be underestimated. Aside from functional and mechanical qualities, a well-finished glaze lends aesthetic values which in turn add to the perceptual value of the clayware. (Adelabu, Ologunwa & Akinbogun 2013).

In addition to the general classification of glazes fired at low, medium, and high temperatures, glazes were also classified by the type of ware, the dominant fluxes, and characteristics such as transparency or matte (Brain, 2005). There are two categories of mid-temperature glaze range, low mid-fire range temperature ranges from 1110^{o}c to 1145^{o}c, and mid-range temperature ranges from 1165^{o}c to 1210^{o}c.

According to Toluwalope and Oluwatuase (2015), Borosilicate glass is a specific type of glass that is primarily made from silica (70-80%) and boric oxide (7-13%). It also contains smaller amounts of alkalis such as sodium and potassium oxides, as well as aluminum oxide. This category of glass contains relatively low amounts of alkalis; it is chemically durable as well as thermal shock resistant (it does not break under rapid temperature changes). As a result, it is widely used in the chemical industry for laboratory equipment, pharmaceutical containers, and ampoules. The source of borosilicate glass in this research will be processed empty injection vials which can also be called cullet. Cullet is a term used to refer to waste glass that contains the main materials needed for preparing glaze.

Fluxes are substances, usually oxides, used in glasses, glazes, and ceramic bodies to lower their high melting point. They function by promoting partial or complete liquefaction of the main glass-forming constituents, usually silica and alumina (Richard &. Eppler 2005). Getting the right flux to lower the fusion temperature of glaze has been one of the major challenges in glaze-making in Nigeria.

Several studies have examined the potential of using cullet as a replacement for natural fluxes, including Tarvornpanich (2006). Cullet recycling is therefore of paramount importance. When waste materials are used to make ceramic glazes, it not only turns trash into treasure but also saves time and money that would have been spent finding other materials. Additionally, glass has a range of different types and properties that can be utilized in this process. Using cullet as a flux in glaze recipes will be the focus of this research, to achieve a glaze that is sustainable. In this direction, using a cullet for total or partial replacement of fluxes in ceramic glazes as be a promising initiative as it will contribute strongly to the sustainable development of ceramics industries in Nigeria, and also contribute to ecological sustainability.

2.0 MATERIAL AND METHODS

All the glaze raw materials (Borosilicate glass, ball clay, kaolin, whiting, feldspar, Zinc oxide, and flint) that were used for this research were locally sourced within the southwest and South-south Nigeria. Borosilicate glass was sourced from the Federal University of Technology Akure Health Centre. The borosilicate glass was a major flux in this research. Ball clay and Kaolin were sourced from Auchi Edo State, The ball clay and kaolin were a source of Alumina (Al) and Silica (SiO_2) in the glaze that was formulated. Feldspar was sourced from Ijero Eikit State and will serve as a flux in the glaze that was formulated. Whiting was sourced from Auchi Edo State; the Whiting was a source of calcium oxide (CaO) in the glaze that was formulated. Dolomite was sourced from Auchi Edo State, and the dolomite was a source of magnesium oxide(MgO). Flint was sourced from a Ceramics Store in Akure and it served as the major source of silica (SiO_2) in the glaze that will be formulated. Zinc oxide was sourced from a ceramic store in Akure and it served as flux.

2.1 MATERIALS PROCESSING

The materials that were used in developing the glaze in this research were Borosilicate Glass which will be obtained from empty injection vials, Kaolin, Flint, whiting, feldspar, and ball clay. Kaolin, flint, whiting, and feldspar were soured in a processed state. To make Borosilicate glass and ball clay, suitable for glaze formulation, both were made to undergo some beneficiation process. The beneficiation process that was used for borosilicate glass includes Washing and drying the empty injection vials, Crushing the empty injection vials, Pulverizing the crushed empty injection vials, and Ball milling of the pulverized empty injection vials. All this process is important because the smaller the particle sizes of the material the lower the eutectic point due to the smaller surface area. This also aids the glaze glossier when it melts on the glazed body as stated by Ologunwa, Akinbogun, and Kashim (2012).

2.3 Glaze Recipe Formulation

Among the factors affecting glaze composition is the method of preparing and applying glazes, as outlined by Akinbogun and Fadiro (2009).GlazeMaster software was used to formulate the glaze recipe. GlazeMaster software is a formula calculation package and glaze storage system. Using this software, 20 glaze recipes were developed. Before the glaze formulation was done the result of the chemical analysis of Kaolin, Ball clay, Flint, Borosilicate glass, and Feldspar was entered into the glaze application.

Figure 2.1: Interface for glaze formulation on Glaze

3.0: Result and Discussion

3.1 Analysis of raw material used

The raw materials that were selected are Ball clay, Flint, Milled Borosilicate glass, Kaolin, and Feldspar. The method employed in this analysis is the energy-dispersive analysis (ED-XRF) and the machine model used to determine the basic chemical composition of the sample is the Varian EDX-8000VF. The result of the analysis is shown in the table below;

Sample	SiO_2 (%)	AL_2O_3	Fe_2O_3	MgO	Cao (%)	Na_2O	K_2O	MnO	TiO_2	P_2O_3	LOI (%)s

		(%)	(%)	(%)		(%)	(%)	(%)	(%)	(%)	
Ball clay	61.14	23.03	5.11	0.89	0.61	0.18	2.52	0.16	0.09	0.13	6.03
Borosilicate glass	66.58	1.32	0.73	3.48	11.11	8.42	0.52	0.01	0.02	0.20	4.10
Kaolin	58.84	17.12	5.76	0.42	2.01	0.73	1.78	0.03	0.09	0.08	12.61
Feldspar	61.41	18.22	0.11	0.10	2.00	5.96	9.77	0.03	0.01	0.04	2.56
Flint	68.72	0.46	0.05	0.19	3.96	6.01	9.52	-	0.01	4.02	1.56

Table 3.1: Result of XRF Analysis

Sample	BaO	B_2O_3	ZnO
Borosilicate Glass	2.52	7.50	-
Flint	1.02	1.10	2.74

Table 3.2 Result of XRF Analysis, showing chemicals that are only present in borosilicate glass and flint.

3.2 Develop the glaze recipe using computer glaze software.

Before the glaze formulation was done. The result of the chemical analysis of Kaolin, Ball clay, Flint, Borosilicate glass, and Feldspar, which were obtained from the Xray Florescent , was input into the data base of the "Glazemaster" software. 15 glaze recipes were first formulated using this glaze software, but after the third firing test, just one of the glazes was glossy. So to achieve glossy glaze 5 glaze recipe was formulated using the "Glazemaster" application. The parameters that are considered in the glaze formulation are as follows;

The ratio of silica to alumina: This determines if the glaze is to come out matt or glossy.

Weight of the flux inside the composition: This helps to predict the temperature at which the temperature will. For mid-temperature glazes the weight of the flux most not be less than 30% according to Digtalfire (2023).

Level of the Alkali: This helps to determine if the glaze will craze or not. Because if the level of the alkali is to high the glaze will craze.

Glaze raw materials/ Glaze sample	Ball clay	Kaolin	Dolomite	Flint	Feldspar	Zinc oxide	Whiting	Borosilicate glass
1	19	-	-	1	10	2	19	50
2	14	-	-	8	18	-	12	50
3	21	-	-	10	-	3	16	50
4	18	-	-	-	16	-	21	60
5	14	-	-	-	8	7	14	60
6	20	-	-	-	-	8	10	52
7	17	-	-	-	6	-	10	62
8	14	-	-	-	-	6	20	55
9	16	-	-	7	-	2	-	61
10	15	-	-	-	11	10	20	50
11	17	-	-	2	10	-	21	50
12	12	-	-	6	-	6	25	68

13	12	-	8	10	-	-	18	52
14	6	-	-	12	27	-	5	49
15	10	-	-	6	16	-	18	50

Table 3.2: 15 Glaze Recipes That Was First Formulated.

3.3 First Glaze Firing

After the first firing only the glaze sample 10 melted. The glaze was fired under an oxides firing. The glaze is glossy and transparent with a light greenish color. All these can be visually observed in plate 4.2. The glaze did not run but craze, the crazing was suspected to be based on the high level of alkali in the glaze recipe, which was from Sodium oxide 5.21 grams and potassium oxide 1.82 grams, making a total of 7.03 grams of alkali. The level of the flux in this glaze is 37.22 grams, the level of silica is 52.46 grams and the level of alumina is 6.52 grams.

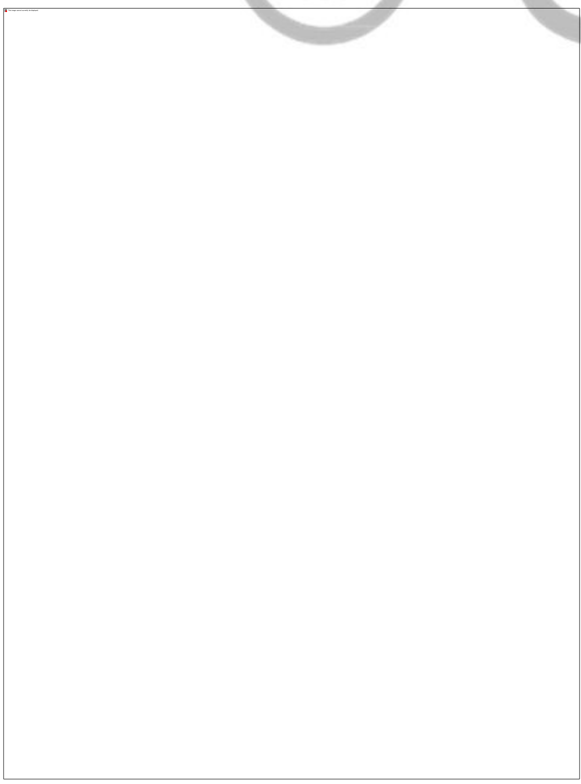

Plate 3.1: Glaze Sample 10 on Test Tile.

3.4 Second glaze firing

The second firing was taken to 1110ºc which is a 30ºc increase. The firing was done in an electric kiln. After the glaze firing it was observed that glaze samples 11, 15.4,3,2, and 1 did not melt while glaze samples 13, 5, 6,14,7,12,9 and 8 melted but formed a matt surface, which can visual observed visually in plates 4.3. The result can be used on decorative ceramic wares.

Plate 3.2: Glaze Sample 5,6,7 8,9,12,13 And 14, On Test Tiles.

3.5 Third glaze firing

Third firing was taken to the highest temperature range set for this study 1140ºc, which means 30ºc was added to 1110ºc. The firing took about 7 hours in an electric kiln. After the third glaze firing, glaze samples 13, 5, 6,14,7,12,9, and 8 melted but formed a matt surface while glaze samples 11, 15,4,3,2, and 1 did not melt, this can be observed in plate 4.4. The result of glaze samples 11.15.4.3.2 and 1, can be related to the level of flux present in each of the glaze recipes, which is listed in the table below;

Glaze sample	Level of flux
11	29.18%
15	27.83%

4	28.53%
3	29.63%
2	23.60%
1	29.65%

Table 3.3: level of flux in glaze samples 11,15,4,3,2 and 1

It can be observed in the table above that the level of flux present in each of the glazes is lower than 30%. Each of the glaze samples that melt in this study has a flux level that is higher or equal to 30%, except glaze sample 14 which has a flux level of 20.63% but it is complemented by the alkaline level of 9.41%, which aids it to melt.

Plate 3.3: Glaze Sample 1, 2, 3,4,11, and 15 on Test Tiles.

3.6 Fourth glaze firing

With the knowledge gained from the first 15-glaze recipe that was formulated, a new 5-glaze recipe was formulated. Before formulating the new 5 glaze recipe, two criteria were set:

The first criterion was to increase the level of flux above 30%

The Second criterion was to reduce the level of alkane to below 8%.

The first criterion will aid the glaze melt at a temperature range between 1080ºc and 1140ºc. The second criterion will reduce or eliminate crazing. The new glaze sample was named N1, N2, N3, N4 and N5. The glaze is listed in the table below;

Glaze material Glaze sample	Borosilicate glass	Feldspar	Whiting	Ball clay	Flint	Kaolin	Zinc oxide
N1	65	15	5	5	10	-	-
N2	50	11	20	10	-	10	15
N3	50	7	20	-	13	15	15
N4	45	11	20	-	-	20	10
N5	54	5	20	-	-	17	10

Table 3.4: Glaze Formulation Glaze Sample N1, N2,N3,N4 And N5

Glaze sample	Flux level	Alkaline level
N1	36.60	6.37
N2	38.67	6.64
N3	39.07	7.34
N4	36.10	6.61
N5	36.03	5.55

Table 3.5: Level Of Flux And Alkaline In Glaze Sample N1, N2, N3, N4 And N5

Glaze samples “N1”, “N2”, “N3”, “N4” and “N5” were weighted, composed, applied on a test tile and fired in an electric kiln. The firing was first taken to 1100^{o}c, after the first firing all the glaze samples give a matt surface. After the first firing the second firing was taken to 1140^{o}c. After the Second firing, glaze sample “N4” and “N5” was matt while glaze sample “N1”, “N2”, and “N3”, give a glossy surface. Glaze sample “N1” was glossy and transparent. Glaze sample “N2” was glossy with a tone of red color. Glaze sample “N3” was glossy and transparent. All this can be visually observed in plate 3.4

Plate 3.4: Glaze Sample N1, N2, N3, N4 and N5 on Test Tiles.

3.7 Leaching test

In this research lemon juice was used to test for leaching. Lemon juice is a natural base solution. The lemon juice was poured into the glazed wears and left for 48 hours. After 48 hours the lemon juice was poured out of the glazed wears and washed with water. After the glaze had dried the color of the lemon juice and the surface of the glaze wear were observed. It was observed that both the color of the lemon juice and the surface texture of the glaze wear did not change. All this can be observed in plate 3.5. 3.6 and 3.7.

Plate 3.5: leaching test for glaze sample N3.

Plate 3.6: leaching test for glaze sample N2.

Plate 3.7: Leaching Test For Glaze Sample N1.

4.0 Conclusion

With the experiment carried out in this research and the result that was obtained from the research, the research did justice to key points in this research. The key points are sustainability, mid-temperature, and food safety. In this research 20 glaze sample was formulated using computer glaze software ("GlazeMaster"). While using the glaze software, three component was considered in the glaze formulation; Alumina silica ratio, Level of alkaline, and Percentage of flux in the glaze recipe. 15 glaze recipes were first formulated and fired, at a temperature range of 1080ºc, 1110ºc, and 1140ºc. After the firing only glaze sample 10 which is composed of (Ball clay 15, Borosilicate 50, Feldspar 11, Whiting 20, and Zinc Oxide 10) gave a glossy surface at temperatures of 1080ºc while samples 13, 5, 6,14,7,12,9, and 8, gave a matte surface at a temperature of 1140ºc. Glaze samples 11, 15,4,3,2, and 1 did not melt between the temperature ranges of 1080ºc to 1140ºc. Another 5 glaze sample was formulated after the result from the first 15 glaze recipe was not satisfactory. The new 5-glaze recipe was named N1, N2, N3, N4 and N5. The glaze was first fired to 1110ºc after the first glaze firing glaze sample N1. N2, N3, N4, and N5 all gave matte surfaces. The second glaze firing was taken to 1140ºc, after the glaze firing glaze samples N1, N2, and N3 gave a glossy surface while glaze samples N4 and N5 gave a clear matte. After all the glaze firing glaze samples 1, N1, N2, and N3 gave the best result between the temperature range of 1080ºc and 1140ºc. All the glaze samples craze on the clay body, with a ratio of Ball clay to Kaolin, a ratio of 1:1, while the glaze sample did not craze on the clay body, with a ratio of Ball clay to Kaolin, a ratio of 2:1 and 3:1.

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