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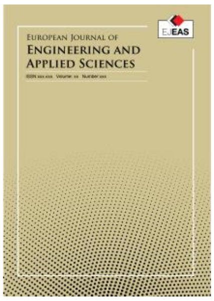

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Research Article

Cullet To Glaze: Converting Waste For Ceramic Artistic, Economic And Environmental Development

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Abstract: Glazes are usually derived from wood ashes and rock materials accomplished at high temperatures. On the hypothesis that cullet, which is non-biodegradable and in the Nigerian context unutilized, indiscriminately discarded, hazardous and constitutes serious solid waste disposal challenge, contains poly-chemical oxides of glass former and should fire at low temperature, this study is production of cullet-derived glazes. Samples were collected from dumps, cleaned, heated, pulverized, sieved, washed, dried, elementally analysed and formed into two glaze batches of cullet with ball clay and cullet with potash and ball clay; milled, applied on ceramic tiles; and fired in a kiln at 1100oC.Findings revealed that cullet has the potentials to produce pure compound of silica for glazing. The study has far reaching implications for the environment in solid waste disposal, economic and industrial development, conservation of energy, reduction of deforestation and anthropogenic carbon emission and ultimately contributes towards a cleaner earth.

Keywords: *Alcoholic Drink Bottle (ADB), Ceramic, Cullet, Economic Development, Glaze.*

Atık Camdan Cama: Atıkların Seramik Sanatı, Ekonomi ve Çevresel Kalkinma İçin Dönüştürülmesi

Özet: Cam genellikle yüksek sıcaklıklarda elde edilen odun küllerinden ve kaya malzemelerinden elde edilir. Bu çalışma, biyobozunur olmayan ve Nijerya bağlamında kullanılmayan, ayrım gözetmeksizin atılan, tehlikeli ve ciddi katı atık bertaraf etme zorluğu oluşturan cam kırıntılarının poli-kimyasal cam oluşturucu oksitler içerdiği ve düşük sıcaklıkta ateşlenmesi gerekmesi sebebiyle kırıntılardan cam elde edilmesi hipotezi üzerine kurulmuştur. Örnekler atık depolama sahalarından toplanmış, temizlenmiş, ısıtılmış, toz haline getirilmiş, elenmiş, yıkanmış, kurutulmuş, temel olarak analiz edilmiş, bilyeli kil ile cam kırıntısı ve potas ve bilyeli kil şeklinde iki cam kümesi halinde oluşturulmuştur. Cam kümeleri, öğütülmüş, seramik karolara uygulanmış ve 1100^{o}C'de fırında pişirilmiştir. Bulgular, cam kırıntısının cam eldesi için saf silika bileşiği üretme potansiyeline sahip olduğunu ortaya çıkarmıştır. Çalışma, katı atık bertarafı, ekonomi ve endüstriyel kalkınma, enerjinin korunması, ormansızlaşmanın azaltılması ve antropojenik karbon emisyonu konularında çevre için geniş kapsamlı etkilere sahiptir ve nihayetinde daha temiz bir dünyaya katkıda bulunmaktadır.

Anahtar kelimeler: *İçki şişesi, seramik, hurda cam, ekonomik kalkınma, sır.*

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1. Introduction

Glaze aesthetic and functional significance is mainly to encrust earthen stoneware with glossy or matt surface. [1] Defines glazes as cooled liquids formed by vitreous substances of high viscosity at melting temperature. Glazes are mixture of crystalline materials capable of erosion resistance and suitable compatibility with alumino-silicate substrates [2]. Glazes are usually formulated with glassy materials, some of which are formed from mass of fine-grained rock in which fossils, crystals and gems are embedded. Glazes contain high percentage of crystalline matter and are applied on clay-based ceramic products to provide a shiny and smooth surface and to seal the clay [3]. The compositions of glazes are based on alumino-silicate glass with combinations of natural and synthetic materials. Nonetheless, glaze is a pre-requisite to all finished ceramic wares [4]. [5] Explains that glaze formulation hold its artistic mystique even in the face of comprehensive scientific study and understanding and maintained its aura of heightened value in ceramic production.

No industrial process can exist without emission of some waste materials and environmental damage is enhanced by these waste materials. Over the last decade, various researches were carried out on alternative usage of waste materials. Over the course of such explorations, different materials were explored by scholars for glaze possibilities. Results derived from the experimentation were ashes glazes from vegetable and fruit peelings [6] [7], carbon exhaust ash of electric power generating station [8] [9], blast furnace slag of granite rock [10] and glass waste [11] [12] [13] [14][15].Exploration of these waste materials has improved ceramic glaze products like electrical insulators and some components of certain machines [16] [17].

Glass is made from silica, the silicates or analogous glass forming compounds. Glass products become waste at the end of certain consumer products life cycle and in a number of cases post-consumer usage. Glass wastes are technically referred to as cullet. Cullets are one of the fastest growing solid wastes in Nigeria. In this context, there are three problems noteworthy of scholarly attention. First is that cullets are non-biodegradable. The second is that they are also not industrially utilized for economic benefit in the ceramics production. Third is improper disposal of cullets as the common practices are to landfill and which constitute environmental degradation and serious health hazard. In another context are two other problems. First is that the majority of the extant glazes are ash glaze types having significant short comings of poor resistance to scratches and time-bound loss of surface qualities despite the fact that they are achieved at high temperature. Second is that the other forms of glazes are produced from rock which consists of aluminum silicates of potassium, sodium and calcium. These types of glazes are mostly melted at high temperature. Contrariwise, cullet contains elements that are capable of low temperature glaze. It is a secondary raw material which has been pre-melted. Furthermore, the formation of glazes on clay depends on components like glass former, flux for the formulation, thermal degree, heating period and coating thickness. Hence, cullet had already contained poly-chemical oxides of glass former and fluxes of certain percentages. Harnessing such materials for industrial development will lessen environmental degradation and disaster [4]. Recycling cullet for glaze production on ceramic wares may therefore provide a lot of energy saving potential. Researches on the possibilities of use of cullets for glaze derivation are nonetheless still evidently few.

Against the foregoing backgrounds, this paper studied the possibilities and processes of using cullets for glazes. The aim of the study is to convert cullet for artistic and economic glaze production. The main objectives of the study are to evaluate the physical behaviour of cullet as substitution to glaze glass former; to scientifically analyze elemental and chemical properties of cullets; and to lessen glass waste from the environment and harness their elemental and chemical properties for industrial development and economic growth.

The common practice is to represent a glaze recipe in 100% batch form of glassy, flux and adhesive agents with absorbent aluminum silicate clay. However, it is hypothetically assumed that it is possible to attain glossy, transparent and opalescent glazes from cullets. The study therefore experimented with borosilicate glass, a type of glass composed with silica and boron oxide [18] [19]. The types of glass under borosilicate glass are biotechnology glass, photovoltaic glass, window, industrial optics glass, alcoholic drinks bottles, pharmaceutical glass, pre-filled syringes and dental cartridges. The borosilicate glass type for the experimentation is alcoholic drinks bottle (ADB). The rationale for the selection was based on the cullets availability.

2. Materials and Methods

The research design for the study involved laboratory analyses and studio artistic practice in order to obtain both quantitative and qualitative data. Collected cullets were sorted based on colour typology. The four selected colours are amber green, dark brown, blue and brown (Plates 1-4).Qualitative and quantitative chemical analytical data were obtained through laboratory processes .The samples' elemental compositions were established after laboratory evaluation for relative oxide properties analysis.

2.1. Sample treatment and pulverisation

Collected samples were treated in two phases to ensure thorough purification of the material. Noteworthy is the possibility of contaminants and impurities from other waste materials that may stain the cullet collected from dump site. Likewise, cullets disposed on dump sites were likely to be soiled with dirt or grime. The samples were therefore first submerged with sublimate water and castile soap for four hours (4 h) to cause the impurities materials to become an emulsion (Plates 5-6). The samples were thereafter washed to remove unwanted materials. The washing was done manually with sponges and long neck multiple mouth brushes for the inner portion of the samples. The samples were dried after thorough washing (Plate 7) before being subjected to the second phase of the treatment; combustion and calcinations.

Plate 1 Plate 2 Plate 3 Plate 4

Figure 1. Plates of alcoholic drink bottles (left to right: Plate 1. Alcoholic drink bottles, dark brown; Plate 2. Alcoholic drink bottles, amber green, Plate 3. Alcoholic drink bottles, brown and dark brown, Plate 4. Alcoholic drink bottles, blue, Photograph by Ajadi Michael 2017)

The combustion was to thaw extraneous oily materials from the samples. After this stage, the samples were treated in separate batches based on four selected colours and similar processes repeated for each of the sample batches..

The cullets were heated in the kiln at low temperature. However, the heat and flame in the kiln did not affect the colours of the cullets during combustion.

Plate 5 Plate 6 Plate 7

Figure 2. Plate 5 (Cullet Samples, Multi-colour and Multi-Structure), Plate 6(Drenching of Cullet Samples, Multi-colour and Multi-Structure), Plate 7 (Cullet Collections After Lavation, *Multi-colour and Multi-Structure),* Photograph by Ajadi Michael 2018

The machine and equipment used for the hammer-milling and processing of the cullets are jaw crusher, pulveriser machine, jaw mill and sieve (Plates 8-9). The samples were crushed with jaw crusher based on the selected colour of the cullets. The jaw crushed samples were soaked in transparent buckets and washed several times before being desiccated under the sunlight. After thorough drying, the samples were pulverised again with Rocklabs Pulveriser Machine (model CRC 3E). The machine designedly operated at three (3) minutes per operation. The regulating minutes of operation however was contingent on hardness not the sample and particle size. For this study, cullets were regulated for one (1) minutes on account of material composition of high siliceous oxide content. The additive materials used with the sample were regulated at thirty (30) seconds. The pulverised samples were further washed to remove the remaining impurities. The thoroughly washed samples were entangled in 350 nm fine mesh, for about seven hours to drain extra water before being disentangled from the mesh for finally drying with electric oven at 60^{o}C.

Plate 8 **Plate 9**

Figure 3. Plate 8 (Pulveriser machine, Rocklabs (model CRC 3E, (*CERD), OAU, Ile-Ife; Plate 9 (*Jar Mill, G90, GE: 1880913, *Ceramic Studio LAUTECH, Ogbomoso)*

The electric oven operational procedures were replicated for all the four (4) sample batches to ensure thorough desiccation of the samples. Thereafter, the desiccated pulverised residues were packed in transparent plastic can and stored in a dry cool place for further processing. The pulverized samples were statistically weighted before and after hammer-milling, after pulverization, and after washing (Table 1).

Table 1: Statistical Weight Changes of Collected Sample in Kilogram (kg)

S/N	Glass Type	Colour Types	Actual Weight (kg)	Weight After Hammer milling (kg)	Weight After Grinding (kg)	Weight After Washing (kg)	Weight after sieving (kg)
1	**Alcoholic DrinksBottles (ADB)**	Amber Green	11.06	10.89	8.99	8.78	8.77
		Dark Brown	10.02	9.84	7.12	7.02	7.02
		Blue	13.75	13.55	10.24	9.04	9.04
		Amber Brown	7.93	7.72	5.72	5.50	5.50

The samples were subjected to elemental and chemical analyses through Tandem Pelletron using Particle Induce X-ray Emission (PIXE) technique. The studio artistic practice aspect of this study entailed seven steps namely: kneading of clay; casting of tiles; biscuit firing; formulation of glaze batches; batch-milling; application of glazes and glaze firing. Each selected cullet sample was experimented in two (2) cullet-ceramic-glaze phases with additive materials These materials were ball clay and potash as flux in batches. milling composition on biscuit ceramic wares. These materials were potash (P) as flux to reduce the melting temperature of the cullet (ADB) and ball clay (B) for the cullet-ceramic-glaze to stay on the earthen-wares. The batch trial samples, namely batches 1 to 8 were highlighted in simple addition ratio of hundred percent formula (100%) in (Table 2) below. Subscripts in each of the composition represent the cullet colour for visual analysis after subjected to kiln firing.

Table 2: Batch constituents by percentage

Sample Batch Compositions	Amber green		Dark brown		Blue		Brown	
	1 $ADB_{ag}B$	2 $ADB_{ag}BP$	3 $ADB_{db}B$	4 $ADB_{db}BP$	5 $ADB_{bl}B$	6 $ADB_{bl}BP$	7 $ADB_{br}B$	8 $ADB_{br}BP$
Pulverized sample	90	70	90	70	90	70	90	70
Ball clay	10	10	10	10	10	10	10	10
Potash	-	20	-	20	-	20	-	20
Total	**100**	**100**	**100**	**100**	**100**	**100**	**100**	**100**

Four centimeters by six centimeters (4cm x 6cm) rectangular tiles with low alpha-numeric inscriptions of one to eight (1-8) and A-B at the back were moulded (Plates 10 and11). The thoroughly exsiccated tiles were stacked into wood kiln and the kiln was sealed immediately for pre-heating. The pre-heated pieces were observed through spy hole and chimney vapour for two hours and thereafter, the fire was raised for actual firing until it is fired to 750^{o}C which took ten hours thirty-two minutes (10 h 32 mins). The pieces were offloaded fifteen hours (15 h) after firing. Sequentially, the cullet-ceramic-glaze compositions were applied as glaze recipes on biscuit ceramic tiles (Plate 12) and were fired at 1100^{o}C.

Plate 10 **Plate 11** **Plate 12**

Figure 4. Tiles (Plate 10:Back side tiles with Inscription, Ceramic Studio, COE, Lanlate; Plate 11: Front side of slab-tiles, Ceramic Studio, COE, Lanlate; Plate 12: Glaze coated biscuit tiles, Ceramic Studio, LAUTECH), Photograph by Adeleke Ibukun 2018.

The coated tiles were exposed to dry in open air. The dehydrating period of each glaze composition varied. Some dried quickly while some took several hours. A gas kiln was used for firing. Coated cullet glaze tiles were stacked in kiln with cone beside the spy-hole for easy examination of cone reactions to the heat. The kiln was sealed off with fired bricks leaving two spy-holes as monitoring channels to the kiln chamber. The firing which commenced at 5:00 pm was for almost seven hours and was stopped at 11:48 pm of the same day. The stoppage was based on the observed bent of the pyrometric cone for temperature accuracy. After the firing, the kiln was allowed to cool for about ten hours (10 h) before the wares were off-loaded.

3. Results and Discussion

The oxide analysis results of the selected four alcoholic drink bottles, potash and ball clay were presented in Table 3 below.The concentration of the oxide components were analysed in percentage

.

Table 3: Chemical composition of pulverised alcoholic drinks bottles (ADB), potash and ball clay

Symbols	Elements	Amber green	Dark brown	Blue	Brown	Potash	Ball Clay
		Oxides Concentration (%)	Oxides Concentration (%)	Oxides Concentration (%)	Oxides Concentration (%)	Oxides Concentration (%)	Oxides Concentration (%)
Na	Sodium	12.65	14.04	12.80	12.93	29.08	1.06
Mg	Magnesium	0.99	1.64	1.04	0.45	0.20	1.48
Al	Aluminum	0.45	0.55	0.50	0.35	0.16	6.04
Si	Silicon	77.86	76.05	76.88	77.75	6.61	81.46
P	Phosphorus	0.09	0.07	0.11	0.11	0.33	0.18
S	Sulphur	0.00	0.00	0.00	0.00	0.00	0.00
Cl	Chlorine	0.00	0.00	0.00	0.00	3.65	0.00
K	Potassium	1.08	0.47	1.07	0.55	58.16	1.74
Ca	Calcium	6.95	6.60	6.88	7.17	0.33	0.49
Ti	Titanium	0.05	0.07	0.07	0.09	0.02	0.53
Fe	Iron	0.29	0.30	0.40	0.32	0.12	6.45
Zn	Zinc	0.01	0.01	0.01	0.01	0.00	0.01
Zr	Zirconium	0.01	0.02	0.01	0.04	0.00	0.09
Mo	Molybdenum	0.00	0.00	0.00	0.02	0.00	0.00
Sb	Antimony	0.00	0.00	0.17	0.21	0.04	0.42
Rb	Rubidium	0.00	0.00	0.01	0.01	0.00	0.03
Sr	Strontium	0.02	0.01	0.02	0.04	0.00	0.03
Ba	Barium	0.00	0.17	0.00	0.00	0.00	0.00
Pb	Lead	0.01	0.01	0.04	0.01	1.31	0.00
	Grand total	**100.00**	**100.00**	**100.00**	**100.00**	**100.00**	**100.00**

The major oxides in the six samples are (Na2O, MgO, Al2O3, SiO2, CaO, TiO2, PbO, Cl and FeO) while (P2O5, SO3, K2O, ZnO, ZrO2, Sb2O5, Rb2O, SrO and BaO) were minor and introuvable in some samples. In colour representation, iron oxide (FeO) gives a bluish green, barium (BaO) gives reddish-blue, sulphur (S) plus iron sulphide give dark amber, and titanium (Ti) is also capable of colourant for glass but mostly used for brightening other colourant oxides. Cobalt (Co) is the strongest oxide for blue glass and chromium (Cr) for dark green glass production. Combination of sulphur (S) with iron (Fe) gives amber glass and shade of reddish brown. Glass with high proportion of boric oxide gives pure blue colour when mixed with sulphur (S).The intensities of colour on glass are determined by oxides concentration of iron (Fe), barium (Ba), sulphur (S), titanium (Ti), cobalt (Co), antimony (Sb), molybdenum (Mo), calcium (Ca), phosphorous (P), chrominum (Cr), selenium (Se) and Tellurium (Te) [18] [20] [21] and [22].The visual results of each batch after kiln firing temperature arepresented in tables 4 to 5 below.

Table 4. Plates and physical analysis of trial batches 1-4

Batch Number	1	2	3	4
Visual result	 **Plate 13** **Batch 1**:$ADB_{ab}B$ Photograph by Ajadi Michael 2018	 **Plate 14** **Batch 2**:$ADB_{ab}PB$ Photograph by Ajadi Michael 2018	 **Plate 15** **Batch 3**:$ADB_{ag}B$ Photograph by Ajadi Michael 2018	 **Plate 16** **Batch 4**:$ADB_{ag}PB$ Photograph by Ajadi Michael 2018
Recipe compositions	**$ADB_{ab}B$**	**$ADB_{ab}PB$**	**$ADB_{ag}B$**	**$ADB_{ag}PB$**
Test Wares	Tiles	Tiles	Tiles	Tiles
Temperature 1100^{o}C	Matt	Melted and crazed	Matt and peel	Melted and glossy
Output colour	Greenish-brown	Greenish-brown	Greenish-brown	Greenish-brown
Matured surface quality	Glossy	Glossy	Glossy	Glossy
Modification	Required flux	Not needed	Required flux	Not needed
Composition evaluation	Fair	Good	Poor	Good
Flow resistance	Thick	Thick	Thick	Thick
Kiln type and heat source	Downdraught/gas	Downdraught/gas	Downdraught/gas	Downdraught/gas

Table 5. Plates and physical analysis of trial batches 5-8

Batch Number	5	6	7	8
Visual result	 **Plate 17** **Batch 5**:$ADB_{bl}B$ Photograph by Ajadi Michael 2018	 **Plate 18** **Batch 6**:$ADB_{bl}PB$ Photograph by Ajadi Michael 2018	 **Plate 19** **Batch 7**:$ADB_{db}B$ Photograph by Ajadi Michael 2018	 **Plate 20** **Batch 8**:$ADB_{db}PB$ Photograph by Ajadi Michael 2018
Recipe compositions	**$ADB_{bl}B$**	**$ADB_{bl}PB$**	**$ADB_{db}B$**	**$ADB_{db}PB$**
Test Wares	Tiles	Tiles	Tiles	Tiles
Temperature 1100^{o}C	Matt	Melted and crazed	Matt	Matt and pores
Output colour	Greenish-brown	Greenish-brown	Greenish-brown	Greenish-brown
Matured surface quality	Glossy	Glossy	Glossy	Glossy
Modification	Required flux	Not needed	Required flux	Not needed
Composition evaluation	Fair	Good	Poor	Good
Flow resistance	Thick	Thick	Thick	Thick
Kiln type and heat source	Downdraught/gas	Downdraught/gas	Downdraught/gas	Downdraught/gas

The plates in tables 4 and 5 present the visual result of glass-ceramic-glaze batches formulated from amber brown alcoholic drink bottles (ADBab). Batch 1ADBabB was formulated with ball clay(B) and batch 2 ADBabPB was formulated with addition of potash(P) and ball clay(B). The result from the batch with addition of potash as flux showed increase in their viscosity and fitted well with smooth flow on ceramic earthwares surface at selected temperature (1100oC). Differences in the degrees of heat on each batch of this selected cullet colour can easily be visualized through the visual results. The batch with addition potash gave quick melting results in terms of glossiness and flow resistance. Batches 3 and 4 show the visual result of glass-ceramic-glaze formulated with amber green alcoholic drinks bottles (ADBag). The batch 4 with additive flux of potash gave the highest glossiness after the firing.

Plate 17 of ADBblB is the acronym for composition of blue alcoholic drink bottles and ball clay. The composition matt without addition of potash at 1100oC and gave and opalescent light green colour effect. This batch required addition of chemical flux for economic benefit in terms of melting temperature. The result from the addition of potash (Plate 18) gave quick melting temperature and good glossy surface output. Plates 19 and 20 revealed the differences in the surface morphology of the batches 7 and 8. The physical features in batches 7 and 8 showed differences in ratio of cracks and glossiness. The reactions of additive potash flux in batch 8 were also disparately distinct from batch 7 without the chemical flux. The result of the batches (2,4, 6 and 8) with additive flux of potash gave assurance that glass-ceramic-glaze can be made from alcoholic cullets at low temperature without cracks and pores.

4. Conclusion

The research has indicated that cullet has the potential to produce pure compound of silica for ceramic glaze components. It is also evident that cullet produced essential glossy qualities in glass glaze formulation. Cullets as glaze materials are also probably save in terms of health and malignant neoplasm. The usage of cullets in this regards will drastically reduce the glass waste in the environment, especially in Nigeria and similar contexts where solid waste disposal is a serious and significant problem. Waste disposal at individual and municipal levels however need to be systematic to allow for separation and collection of cullets from other waste materials. Furthermore, the study has provided good information on both the physical and elemental properties of a wide range of cullets. Significantly, the study has demonstrated the suitability and importance of cullets in forming low temperature ceramic glazes as alternative to other high temperature glazes. This has far reaching implications for the environment as it will ultimately conserve energy and firing resources and consequently reduces deforestation, cut heat and anthropogenic carbon emission and contribute towards a cleaner earth and its atmosphere.

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