

Lime-Barium and Lime-Zinc Raw Glazes with Raw Materials from Tanzania and Cameroon

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ABSTRACT

In the paper, the lime-barium and lime-zinc glazes used in Southern and Eastern Africa Mineral Center (SEAMIC) ceramic unit in Tanzania were investigated for the production of high temperature raw glazes in Cameroon. The base Seger formula used was 0.3KNaO, 0.3CaO, 0.4ZnO or BaO; xAl_2O_3 and $ySiO_2$. Different Al_2O_3/SiO_2 formula ratios for glossy and matt glazes were tested with raw materials from Cameroon and Tanzania. In the Cameroonian glazes, a local pegmatite (quartz, microcline, albite) with low coloured oxides replaced an albitic feldspar in the Tanzanian glazes. The other materials were kaolin, limestone, silica and barium carbonate or zinc oxide. The glazes were tested at Seger Cone 7 (1240°C) on stoneware slabs (made up of the same feldspathic glaze material: 18%, silica sand or quartz 27%, ball clay 40% and kaolin 15%) from both Tanzania and Cameroon materials. From results obtained, zinc oxide acted as a more vigorous flux than barium carbonate at cone 7, which is obvious in the glazes with pegmatite (with high quartz content). At a Seger formula of Al_2O_3/SiO_2 1:10, transparent glazes were obtained. In these glazes the alumina content was lower and when increased to higher Seger ratios 1:4.7 - 8.7, the gloss was lowered to matt-luster due to fine crystal formation.

Keywords: Lime-Zinc Glazes, Lime-Barium Glazes, Raw Glazes, Seger Cone 7

1. Introduction

Glazes constitute a fundamental material to the success of the ceramic industry. During recent years, the demands of shorter firing cycles and easy control of surface microstructure have called for the use of fritted compositions. Raw glazes have to a large extend been replaced by fritted glazes. However, in the case of high temperature ceramic products like tiles, stonewares and sanitarywares fired at temperatures above 1200°C, raw glazes are still a competitive alternative to fritted formulations [1]. Glaze is similar to glass but with a more complex system because it deals with more components; 7 or 8 oxides in significant amounts are common [1-6]. These components are divided into the glass network former (SiO₂ or B₂O₃), stabilizer (generally Al₂O₃) and flux or glass network modifier or melter (R₂O or RO, where R is an alkaline, alkaline earth or fluxing metal). The complication with glazes is accentuated by the fact that a thin layer is applied to a ceramic body and the glaze body interaction may result in faults. Such faults like cracks (at times voluntary) can affect the aesthetics and technical properties of the product. A glaze is a mixture of materials, powder or suspension in water which when applied as a coating on a body and fired to an appropriate temperature, vitrifies and develops specific properties. For every given body composition, it is important to test the ideal glaze based on aesthetics and properties envisaged.

This research was carried out to produce stoneware glazes using Cameroonian raw materials, to replace costly imported stoneware and porcelain glazes commonly used in Cameroon. The studies were carried out at SEAMIC in Tanzania, investigating the base glaze compositions used on its decorative stoneware products. These high temperature glazes are of lime-barium (barium glazes) and lime-zinc systems and are raw, not using fritted materials or lead. Barium glazes are recommended for their ability to produce brilliancy and hardness [7].

The base Seger formula for the glazes was 0.3KNaO, 0.3CaO, 0.4ZnO or BaO; x Al₂O₃ and y SiO₂ at cone 7 (1240°C). The x/y (Al₂O₃/SiO₂) ratio has been reported to have a great influence on the glaze surface as indicated in **Figure 1** [2,3,8,9].



SiO₂ Figure 1. Variation of x (Al₂O₃) and y (SiO₂) molecular formula for glaze preparation.

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2. Experimental

2.1. Raw Materials

Raw materials used in Tanzanian glazes (TG) were available in SEAMIC and are given in **Table 1**. Included are those for Cameroonian (CG) glazes, indicating their sources. Chemical compositions were analysed by x-ray florescence, fused bead method with a SIEMENS SRS 3000 spectrometer. Mineralogical analyses were carried out with a SIEMENS D 5005 diffractometer.

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2.2. Experimental Procedure

Al₂O₃

2.2.1. Glaze Formulation

Molecular ratios of Al_2O_3 and SiO_2 were varied to produce glossy glazes with single glass phase glossy (zone A) and matt glazes with fine crystals (zone B) using **Figure 1**. Molecular Al_2O_3/SiO_2 ratios 1:10 for A zone and 1:4.6 - 8.7 for the B zone were tested. **Tables 2** and **3** indicates different ratios of x and y tested for Tanzanian glazes and Cameroonian glazes. The glaze recipes were calculated with these assumptions: that limestone, kaolin, and silica sand (flint) were pure, of respective composition CaCO₃, Al_2O_3 ·2SiO₂·2H₂O and SiO₂; and that feldspathic materials (feldspar and pegmatite) were mixed feldspar of composition KNaO·Al₂O₃·6SiO₂.

2.2.2. Glaze Preparation and Application

As received materials (flint, quartz, feldspar, limestone

and pegmatite) were crushed in a DIETZ jaw crusher to <400 μ m and dried at 105°C for 24 hours. The glaze recipes were composed using a Sartorius V3600S (max load 3640 g and sensibility 0.1 g) scale. 45% water was added to the raw glaze mixture which was then ball milled until all passed a 75 μ m sieve. Densities were adjusted to about 1550 g/L using a Baume hydrometer.

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Table 1. Raw materials and their source.

Materials	Source				
Pegmatite	Mont Nlonako, Littoral Region, Cameroon				
Kaolin C	Mayoum, Foumban, West Region, Cameroon				
Quartz	Mvan, Yaoundé, Center region, Cameroon				
Clay C1	Mvan, Yaoundé, Center Region, Cameroon				
Clay C2	Bambili, North-West Region, Cameroon				
Feldspar	SEAMIC, Tanzania				
Kaolin T	SEAMIC, Tanzania				
Flint T	SEAMIC, Tanzania				
Limestone T	SEAMIC, Tanzania				
Clay T	SEAMIC, Tanzania				
Zinc oxide	SEAMIC, Tanzania				
Barium carbonate	SEAMIC, Tanzania				

				Recipe (wt%)*						Class sustant
Glaze x	У	AI_2O_3/SIO_2 (Seger) –	F	K (T)	L	Fl	Zn	Ba	- Glaze system	
TG1	0.30	3.0	1:10	54.6	n.u.	10.1	24.3	11.0	n.u.	Ca-Zn
TG2	0.30	3.0	1:10	47.2	n.u	8.8	21.0	n.u.	23.0	Ca-Ba
TG3	0.35	3.5	1:10	42.4	3.4	7.9	25.3	20.0	20.8	Ca-Ba
TG4	0.40	4.0	1:10	38.9	6.2	7.2	28.8	n.u.	18.9	Ca-Ba
TG5	0.65	3.0	1:4.6	47.0	26.2	8.7	8.7	9.4	n.u	Ca-Zn
TG6	0.65	3.0	1:4.6	41.4	23.1	7.7	7.7	n.u.	20.2	Ca-Ba
TG7	0.45	3.0	1:6.7	44.6	10.6	8.3	14.9	n.u.	21.7	Ca-Ba
TG8	0.65	5.0	1:7.7	31.7	17.7	5.9	29.4	n.u.	15.4	Ca-Ba

Table 2. Tanzanian glaze (TG) recipes and Al₂O₃/SiO₂ molecular ratios.

*F = Feldspar; K(T) = Kaolin; L = Limestone; Fl = Flint (silica sand); Zn = ZnO, Ba = BaCO₃; n.u. = not used in recipe.

Table 3. Cameroonian glaze (CG) recipes and Al_2O_3/SiO_2 molecular ratios.

		Recipe (wt%)*								
Glaze	X	У	AI_2O_3/SIO_2 (Seger)	Р	K (C)	L	Q	Zn	Ba	- Glaze system
CG1	0.30	3.0	1:10	54.7	n.u.	10.1	24.3	10.9	n.u.	Ca-Zn
CG2	0.40	4.0	1:10	38.9	6.2	7.2	28.8	n.u.	18.9	Ca-Ba
CG3	0.40	4.0	1:10	44.0	7.0	8.0	32.0	9.0	n.u.	Ca-Zn
CG4	0.35	3.0	1:8.6	46.3	3.7	8.6	18.9	n.u.	22.5	Ca Ba
CG5	0.45	3.0	1:6.7	44.5	10.6	8.3	14.9	n.u.	21.7	Ca Ba
CG6	0.50	3.0	1:6.0	43.7	13.9	8.1	13.0	n.u.	21.3	Ca Ba
CG7	0.50	3.0	1:6.0	48.0	15.0	9.0	18.0	10.0	n.u.	Ca Zn
CG8	0.35	3.5	1:10	56.7**	n.u.	8.7	24.9	9.6	n.u.	Ca - Zn

*P = Pegmatite; K = Kaolin; L = Limestone; Q = Quartz; Zn = ZnO; Ba = BaCO₃; n.u. = not used; **Pegmatite was replaced by the fusible clay C2.

Slips were applied by dipping method on unfired stoneware body.

2.2.3. Stoneware Body Composition

The Tanzanian glazes were applied on a body using raw materials from Tanzania. Similarly, Cameroonian glazes were applied on a body made up of local materials from Cameroon. The Tanzanian ceramic body was made up of 18% feldspar T, 27% flint T, 40% clay T and 15% kaolin T and that of Cameroonian 18% pegmatite C, 27% quartz C, 40% clay C1 and 15% kaolin C. The stoneware body test pieces were produced by crushing the raw materials with water in a ball mill and filter pressed to remove excess water. The paste obtained was flattened on a roller, and slabs cut for glaze trials.

2.2.4. Firing Schedules

The glazed test pieces were single fired on Seger cone 7: 1240°C, in a Linn Electrotherm electric kiln. From am-

bient temperature to 650°C, the firing rate was 2.71°C/ min, and from 650°C to 1240°C, 2.46°C/min. The test pieces were soaked at 1240°C for 2 hours and were allowed to cool in the kiln to ambient temperature before removal.

3. Results and Discussion

3.1. Chemical and Mineralogical Compositions of Raw Materials

The results of chemical composition determinations on Tanzanian and Cameroonian raw materials are given in **Tables 4** and **5** respectively. The feldspar is more sodic and the pegmatite more potassic. The percentage of non-feldspathic components in these materials is quite low. The high SiO_2 content of pegmatite is expected to alter the melting behaviour as compared to feldspar, when the recipe contains the same amount of feldspathic

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Oxide	Silica sand	Feldspar T	Kaolin T	Limestone T	Clay T
SiO ₂	97.58	65.18	49.60	5.25	51.28
Al ₂ O ₃	1.41	20.70	35.76	2.03	30.14
Fe ₂ O ₃	0.19	0.10	1.07	0.99	1.69
MnO	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
TiO ₂	0.31	< 0.01	0.40	< 0.01	0.31
MgO	0.05	0.12	0.11	0.41	0.41
CaO	0.18	1.40	0.09	49.70	041
Na ₂ O	0.06	8.70	0.17	0.40	0.61
K_2O	< 0.01	2.61	0.19	< 0.01	2.00
P_2O_5	< 0.01	< 0.01	<0.01	< 0.01	< 0.01
L. O. I.	0.20	0.41	12.58	40.70	13.04
Total	99.98	99.22	99.97	99.48	99.89

Table 4. Chemical composition (wt%) of Tanzanian raw materials.

Table 5. Chemical composition (wt%) of Cameroonian raw materials.

Oxide	Quartz	Pegmatite	Kaolin C	Clay C1	Clay C2
SiO ₂	99.02	82.55	45.12	60.02	71.71
Al_2O_3	0.40	8.93	34.59	27.83	12.86
Fe_2O_3	0.22	0.52	1.25	2.17	2.86
MnO	< 0.01	0.03	< 0.01	< 0.01	0.03
TiO_2	< 0.01	0.03	4.76	1.15	0.19
MgO	0.15	0.28	0.35	0.56	0.26
CaO	< 0.01	0.38	< 0.01	< 0.01	0.35
Na ₂ O	0.17	2.05	0.15	0.22	0.74
K ₂ O	< 0.01	4.85	0.13	0.39	3.90
P_2O_5	< 0.01	< 0.01	0.33	0.39	< 0.01
L. O. I.	0.03	0.08	13.15	7.08	6.83
Total	99.99	99.70	99.83	99.81	99.73

material. Both kaolin T and C contain about 35% Al₂O₃ and iron content within the stoneware composition. The presence of 4.76% TiO₂ in kaolin C resulted in stoneware with a darker colour. The limestone contain 89% CaCO₃, with main impurities SiO₂ and Al₂O₃ and low MgO. The quartz stones are of very high purity.

From x-ray analyses, pegmatite contained quartz, albite and microcline; clay C1 quartz and kaolinite; and clay C2 a very fusible clay (fusion point 1200°C): kaolinite, montmorillonite, quartz and microcline. Kaolin C, from previous analysis [10] is made up essentially of kaolinite and quartz with traces of illite, anatase and anorthite. Clays T and C1 are ball clays for stoneware bodies.

3.2. Glazed Test Pieces

The visual appearance of the glazed test pieces are described in **Tables 6** to **8** in which the glaze systems, SiO_2 and Al_2O_3 contents, are included.

TG1-TG4 gave glossy transparent glazes (**Table 6**). The Al_2O_3/SiO_2 molecular ratio is 1:10. TG1 is a Bristol glaze and is colourless, whereas the barium glazes TG2-

		ALO wt%		Surface structure			
Glaze	SIO ₂ wt%	AI_2O_3 wt 70	Giaze system -	Colour	Appearance		
TG1	59.9	11.3	Ca-Zn	colourless	high gloss, transparent, smooth feel, no craze		
TG2	51.8	9.8	Ca-Ba	light khaki	high gloss, coloured, smooth, crazed		
TG3	54.8	5.5	Ca-Ba	light khaki	high gloss, coloured, smooth, crazed		
TG4	45.9	10.5	Ca-Ba	light khaki	glossy, coloured, no crazing, fine bubbles, rough feel		
TG5	52.3	19.1	Ca-Zn	white	high gloss, opaque, no crazing		
TG6	46.1	16.8	Ca-Ba	pastel beige	opaque, matt, no crazing		

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Table 7. Effect of colorants on Tanzanian glazes.

Claza Description		Class system	Surface structure			
Glaze	Description	Glaze system	Colour	Appearance		
TG7	$TG7 + 5\% \ MnO_2$	Ca-Ba	purple + whitish specks	low gloss, opaque, no crazing, smooth feel		
TG8	$TG8 + 5\% \ MnO_2$	Ca-Ba	yellow-brown	glossy ,opaque, no crazing		
TG9	$TG6 + 5\% MnO_2$	Ca-Ba	dark brown	opaque, matt, no crazing		
TG10	$TG6 + 5\% Fe_2O_3$	Ca-Ba	chocolate brown	opaque, matt, no crazing		
TG11	TG6 + 1% CoO	Ca-Ba	blue	opaque, matt, no crazing		
TG12	TG6 + 2% NiO	Ca-Ba	pale pink	opaque, matt, no crazing		
TG13	$TG6 + 2\% \ Cr_2O_3$	Ca-Ba	dark brown	opaque, matt, no crazing		
TG14	$TG1 + 2\% Fe_2O_3$	Ca-Zn	brown	glossy, opaque, no crazing		
TG15	TG1 +1% CoO	Ca-Zn	deep marine blue	high gloss, opaque, no crazing		

Table 8. Visual appearance of Cameroonian glazes and their silica + alumina contents.

				Surface structure			
Glaze	SIO ₂ wt%	Al ₂ O ₃ wt70	Glaze system	Colour*	Appearance		
CG1	74.0	8.4	Ca-Zn	whitish	incomplete fusion		
CG2	65.7	5.7	Ca-Ba	colourless	Glossy, transparent, no crazing		
CG3	71.4	6.3	Ca-Zn	colourless	Glossy, transparent, no crazing		
CG4	58.7	5.4	Ca-Ba	colourless	Opaque, matt, no crazing		
CG5	56.4	7.7	Ca-Ba	whitish	Opaque, matt, fine crazes		
CG6	55.3	8.71	Ca-Ba	colourless	Glossy, transparent, no crazing		
CG7	64.4	9.5	Ca-Zn	colourless	Glossy, transparent, no crazing		
CG8	65.5	7.3	Ca-Zn	yellow	Opaque, matt, no crazing		

*The stoneware body fired to beige, consequently mild coloration could not be perceived.

TG4 possess a light khaki tint. Decreasing $BaCO_3$, a fluxing agent, reduces the glossiness. In the BaO-CaO-SiO₂ ternary phase diagram, a liquid phase exists at as

low as 1268°C [10]. This eutectic contributes to the low temperature phases obtained at cone 7. Omar *et al.* [11], observed that to introduce glass-ceramics into industrial

applications, crystallizing agents had to be added to Baaluminosilicate glasses. With high BaCO₃ contents: 23% in TG2 and 21% in TG3, the glazes showed mild crazing, probably due to the high dilatation coefficient of BaO, in addition to the high amount of alkaline feldspar. The dilatation coefficient of BaO is 3.0×10^{-7} /°C in fritted glazes but can attain 6.0×10^{-7} C in raw glazes [12]. The thermal expansion in these glazes could be lowered by reducing the alkali content, that is, the feldspar content [2,8,13]. ZnO showed a vigorous fluxing action (TG1, 11.0% ZnO) when compared to BaO (TG2, 16% BaO) for the same Al₂O₃/SiO₂ molecular ratio, 1:10. In the ZnO-Al₂O₃-SiO₂ ternary phase diagram, low temperature compounds were located at the poor Al₂O₃ and high (above 50%) SiO₂ ends [10]. In TG5 Bristol glaze, a white opacification occurred. ZnO which forms the basis of Bristol glazes has been noted to increase whiteness, especially in zircon glazes. At low Al₂O₃/SiO₂ molecular ratio 1:4.6 (high amount of Al_2O_3), opacity is probably due to very fine crystals of zinc compound(s), such as gahnite, ZnO·Al₂O₃.

The barium glaze TG6, with Al₂O₃/SiO₂ molecular ratio 1:4.6, is the base matt glaze in SEAMIC's ceramic unit. It has been reported that low barium release (non toxic) could be obtained with up to 20% barium, when the Al₂O₃ + SiO₂ content is high, *i.e.* above 75% [14]. In this matt lime-barium glaze, the barium content was 14% and the Al₂O₃ + SiO₂ 63%. For the other barium glazes tested, the barium contents were between 10.7% and 16.0% with Al₂O₃ + SiO₂ content between 60% and 56%. It is important to study barium release for these glazes when use in dinner ware industry is envisaged.

Comparing the Al₂O₃/SiO₂ molecular ratios of the glazes, for Bristol glazes, 1:10 ratio (TG1) gave transparent glaze and opacity developed when varied to 1:4.6 (TG5). This confirms the widely accepted Figure 1 and observations that the x-y ratio has a great influence on glaze surface. In **Table 6**, it is noted that this is directly related to the Al₂O₃ content; 11.3% Al₂O₃ in TG1 and 19.1% in TG5. The same is true for the barium glazes; TG2-TG4 of 1:10 molecular ratio are transparent with < 10.5% Al₂O₃ and a matt glaze is obtained with formula 1:4.6, containing 16.8% Al₂O₃. Transition metal oxides, when added raw, give colour upon firing of ceramic glazes. The colours obtained with MnO₂, Fe₂O₃, CoO, NiO and Cr₂O₃ are indicated in **Table 7**. TG7, TG8 and TG9 (base TG6) all barium glazes contain 5% MnO₂. TG7 is matt and purple; TG8 is of low gloss, fine bubbles and brown, while TG9 is of low gloss, and dark brown. MnO₂ generally gives brown but in high alkali glazes, fires to purple [8]. TG7 with the highest feldspar content (45%) fired to purple, while TG9 (base TG6) and TG8 containing 42% and 32% feldspar respectively,

gave brown hues. 5% Fe_2O_3 gave a matt chocolate-brown on barium base glaze TG6, *i.e.* TG10; while 2% Fe_2O_3 on Bristol base glaze TG1 (*i.e.* TG14) gave a high gloss pale brown glaze.

With 2% NiO, the barium base TG6 glaze gave pale pink (TG12). 2% Cr_2O_3 produced a brown coloration on barium base TG6 glaze. The refractoriness of Cr_2O_3 was obvious as this glaze TG13 did not fuse completely. 1% CoO on both barium and Bristol base TG6 and TG1 glazes gave respectively a matt blue TG11 and high gloss and strong ultramarine coloration, TG15. Cobalt is a very powerful colorant which easily dissolves in glaze matrix (with fluxing action) and enters the structure [8]. Cobalt colours tend to be brighter in Bristol glazes whereas iron colours are duller [4].

Cameroonian glazes CG2 and CG3, Table 8, respecttively for barium and Bristol glazes with Al₂O₃/SiO₂ molecular ratio 1:10 were uncoloured, transparent and of high gloss. These surface structures were similar to Tanzanian glazes with same ratio. These glazes can serve as base glazes for transparent stoneware using Cameroonian local materials. Barium glaze CG6 and Bristol CG7 of ratio 1:6 were of mild gloss and showed some opacification. This was probably due to the formation of very fine barium or zinc aluminosilicate compounds. CG1 of Bristol glaze series presented an incomplete fusion at cone 7. The insufficient fluxing action of the 11% ZnO was probably due to the high quartz content: 82% SiO₂ in pegmatite (54.7% in the recipe). An increase in temperature to a higher cone, >1240°C is required. It is worth observing that with 54.6% feldspar in the Tanzanian glaze TG1, a single glass phase was obtained at cone 7. This implies that ZnO had a higher fluxing action on the feldspar (Tanzanian) than on the pegmatite (Cameroonian). At lower pegmatite content, 44% in the recipe, the fluxing action of 9.0% ZnO is evident in CG3. Some opacification gave a mid matt glaze in CG4 barium glaze (Al₂O₃/SiO₂ molecular ratio 1:8.6). A matt glaze was obtained with CG5 of ratio 1:6.7. Both CG4 and CG5 are recommended as base glazes for coloured Cameroonian matt glazes. Clay C2 of high fusibility (melts at 1200°C) replaced pegmatite in glaze CG8. A yellow opacification was obtained on a matt surface. This clay could be explored in the production of glazes in Cameroon. On the recommended base glazes for transparent (CG2 and CG3) and matt glazes (CG4 and CG5), detail investigation of the effect of colorant transition metal oxides should be carried out when required by exploring the results on the Tanzanian glazes.

4. Conclusions

Cameroonian materials (pegmatite, kaolin, quartz and limestone) can be used in raw lime-barium and lime-zinc

(Bristol) glazes at cone 7 on stoneware when fritting is not possible. The base Seger or molecular formula was 0.3KNaO, 0.3CaO and 0.4ZnO or BaO; x Al₂O₃ and y SiO₂. The influence of Al₂O₃/SiO₂ molecular formula that is x-y variation on the glaze surface structure was evident on both the Tanzanian and Cameroonian glazes. Ratio 1:10 gave transparent glazes and 1:4.6 - 1:8.7 mid matt to matt glazes. The latter, a result of very fine crystal formation. Transparent glazes contained lower amounts of Al₂O₃ than matt glazes, alumina therefore influenced crystallization.

Different hues were obtained upon addition of metal oxides MnO_2 , Fe_2O_3 , Cr_2O_3 , NiO and CoO on the base glazes for both transparent and matt glazes. The glaze recipe and system (lime-barium or lime zinc) influenced the colour and the surface structure for the same colorant metal oxide.

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