Available Online at www.e-iph.co.uk
Indexed in Clarivate Analytics WoS, and ScienceOPEN

E-B
Environment - Behaviour
Proceedings Journal

CSSR2022
https://cssr.uitm.edu.my/2022/index.php
9th International Conference on Science & Social Research 2022
Universiti Teknologi MARA (UiTM), Shah Alam, Malaysia (Online), 14-15 Dec 2022
Organizer: Office of Deputy Vice-Chancellor (Research & Innovation), UiTM, Malaysia

e-IPH
e-International Publishing House Ltd., United Kingdom

Composition of Various Percentages of Terracotta Clay as Colourant Glaze

Siti Norhashimah Suman[1,] Rusmadiah Anwar[2*], Nor Nazida Awang[3], Salwa Ayob[1]
**Corresponding Author*

[1*] College of Creative Arts, Universiti Teknologi MARA, 32610 Seri Isandar Campus Perak, Malaysia
[2] Nation Design Centre of Creative Arts, OrganisationUniversiti Teknologi MARA, 40450 Shah Alam Selangor, Malaysia
[3] GreenSafe Cities (GreSAFE), OrganisationFaculty of Architecture, Planning, & Surveying, Universiti Teknologi MARA, 32610 Seri Iskandar Campus, Perak, Malaysia

sitin185@uitm.edu.my, rusma9352uitm.edu.my, nazida803@uitm.edu.my, salwa948@uitm.edu.my
Tel *:+6012-6190057

Abstract
Terracotta is a type of clay that is highly valued for its mineral content. The minerals found in terracotta are commonly used to create pigments that are used to color ceramic bodies and glazes. Terracotta's natural minerals make it unique when compared to commercially available colors. In this study, researchers explored the possibility of using locally sourced terracotta clay in transparent glaze formulations. The team used a variety of experimental methods, including examining the percentage of clay powder, X-ray diffraction, X-ray fluorescence tests, and scanning electron microscope analysis (SEM) to study the clay's morphology and structure. By adding terracotta clay powder in different proportions (ranging from 20 to 50%), the glaze became translucent. The resulting colors were glossy, smooth, and creamy, with no defects, as confirmed by visual inspection after the samples were applied to porcelain and fired at gloss temperatures (1200°C). Keywords: Terracotta, colorant, glaze, percentage

Keywords: terracotta; colourant; glaze; percentage

DOI: https://doi.org/10.21834/e-bpj.v8iSI16.5228

1.0 Introduction

The exploration of ceramic materials has the potential to unlock new possibilities for future artistic design needs (Malek et al., 2022). Several reports in Malaysia indicate that the local ceramics have stable body characteristics that are yet to be discovered (Anwar, 2016). Terracotta, a type of earthenware made from clay, can be used glazed or unglazed, making it an attractive option for pottery enthusiasts. Its porous nature makes it easy to work with, and its iron content reacts with oxygen to give it a range of hues from reds, oranges, yellows, and pinks. The range of colors available in terracotta is not limited to a single shade, but rather to a family of dyes that resemble fired clay. When glazed, bright colors are often applied to complement the low firing temperature of terracotta. Using terracotta results in lower energy costs, less kiln wear and tear, and the ability to fire ware on stilts, which allows for glazed bottoms (Hansen, 2017). The exploration of ceramic materials has the potential to unlock new possibilities for future artistic design needs (Malek et al., 2022). Several reports in Malaysia indicate that the local ceramics have stable body characteristics that are yet to be discovered (Anwar, 2016). Terracotta, a type of earthenware made from clay, can be used glazed or unglazed, making it an attractive option for pottery enthusiasts. Its porous nature makes it easy to work with, and its iron content reacts with oxygen to give it a range of hues from reds, oranges, yellows, and pinks. The range of colors available in terracotta is not limited to a single shade, but rather to a family of dyes that resemble fired clay. When glazed, bright colors are often applied to complement the low firing temperature of terracotta. Using terracotta results in lower energy costs, less kiln wear and tear, and the ability to fire ware on stilts, which allows for glazed bottoms (Hansen, 2017).

2.0 Revisit terracotta as the potential substance for ceramic glaze

Terracotta is not typically made from pure clay but is instead found mixed with other minerals that are rich in flux and iron oxide. It is a

DOI: https://doi.org/10.21834/e-bpj.v8iSI16.5228

clay-like ceramic that can be glazed or left unglazed. Terracotta is commonly used for flower pots, water and sewage pipes, bricks, and sculptures. It can be made from any type of organic clay, but earthenware clay is the most common and produces the brown-orange color that is known as terra cotta. Terra-cotta products are fired at low temperatures, resulting in a more porous and penetrable surface. Terracotta clay is very plastic and versatile, containing a high iron content between 5% and 8%. When fired between 1000°C and 1150°C, the clay produces a rich brown color. Terracotta clay is a low-grade fire clay that can be used to manufacture large terracotta pieces.

Sayong in Perak was chosen for its rich natural resources and easy accessibility to terracotta clay. This clay is not only cheap but also of good quality for ceramic and pottery making. With its potential for development and expansion in usage and functionality, such as ceramic colorants and glazes, as well as decoration techniques, terracotta clay has been used for generations. The term "glaze" refers to a mixture of materials that can be a powder or a suspension in water, ready for application to ceramic ware by dipping or spraying. After suitable heat treatment, this powdered mixture vitrifies and develops specific properties of a glass layer appropriate to the intended use of the glaze on the ceramic body. The choice of raw materials determines the properties of the glaze, and studies have shown that the raw materials used can optimize the end product of the glaze. Different compositions result in different properties, such as different melting points and viscosities. Additionally, the choice and formulation of glaze composition depend on factors such as the type of substrate, firing methods (gas, electric), firing cycles, and kiln atmosphere.

Iron oxides, especially magnetite ($Fe3O4$) and hematite (α-$Fe2O3$) are key minerals for tracking the thermal transformations undergone by burnt clay minerals. The composition of a glaze is chosen to ensure certain well-defined properties, such as adhesion to the substrate, correspondence of thermal expansion, transparency or opacity, surface texture, and resistance to chemical attack. Minerals are the earth's cooling and subsequent weathering, breaking down and recombining. Glaze raw materials are broadly classified as a glass former, a flux, and clay. The flux is used to lower the firing temperature. The stabilizer is used to extend the melting range of a glaze and to stiffen the glaze, while the glass former is the main ingredient of a glaze. However, colorants or opacifying oxides might be added to the basic recipe.

The final color and surface effects, such as brilliance and opacity, also depend on the method of glaze application, the composition of the ceramic body, the firing process, the pre-processing of the materials, and the amount and nature of the colorants. The combinations of elements have distinctive physical and chemical properties and, in the present environment, a degree of solidity.

3.0 Method

Before the clay powder process, samples of local terracotta clay go through the sedimentation process (Anwar et al., 2011). The local terracotta clay was soaked in distilled water for 24 hours, respectively. This sedimentation process is performed to separate impurities such as rough silica, twigs, dry leaves, and terracotta clay. After the sedimentation process, the terracotta clay went through the drying process by removing the water from the plaster batt. Furthermore, raw material, local terracotta clay, was crushed and ground using agate mortar to get fine particles and continued for XRF, XRD, and SEM testing. The elemental composition components in the raw material terracotta clay are determined using an XRF test (Anwar et al., 2015). The X-ray diffraction (XRD) analysis to determine the phase structure of terracotta clay. SEM testing was performed on raw material terracotta clay for phase scanning on the surface with a focused beam of electrons.

This study aims to explore the potential use of locally sourced terracotta clay in creating a transparent glaze. Suman et al. (2022) reveal that the addition of terracotta clay powder produces a transparent glaze that requires only one firing profile at 1200°C, with a 60-minute soaking period. Figure 1 depicts the firing profile, highlighting the gradual cooling process employed. Table 1 presents the weight percentage formula for the colorant glaze formulation used in the study. The research delves into the colorant properties of locally sourced terracotta clay powder in a transparent glaze. The study adds clay powder in varying quantities, from 0% to 50%, as a substitute for conventional colorants. The weight percentages used were determined based on previous studies.

To achieve the desired color, the terracotta clay found in the area is mixed with a transparent glaze in varying proportions. It is important to make precise calculations for the percentage of additions needed for the color to develop during the glaze firing process. The percentage additions can range from 20% to 50%, with increments of 10%.

Table 1. Transparent formulation

	Raw material (wt%)				
Sample code	Potash feldspar	Silica	Zinc oxide	Calcium carbonate	Colorant
A1	33	32	15	20	20
A2	33	32	15	20	30
A3	33	32	15	20	40
A4	33	32	15	20	50

Based on the transparent formulation specified in Table 1, four samples for the terracotta glaze recipe (A1–A4) were noted. All materials used are of the same percentage, except that the conventional local terracotta clay was added in the range of 20% to 50%. Potash feldspar at 33%, silica at 32%, zinc oxide at 15%, calcium carbonate at 20%, and terracotta as a colorant. Its purpose is to compare the effects of the varying percentage of terracotta in the transparent glaze. The raw materials used weight percentage using an electronic balancing apparatus to obtain a 100g dry weight of the powder, then added with water (120 ml) and milled in a ball-mill machine for grinding and consistent mixing. The grinding process takes one hour and then is left for 24 hours for the aging process. The bisque test pieces are cleaned first with a damp sponge to remove dust, and the dipping technique for the application process.

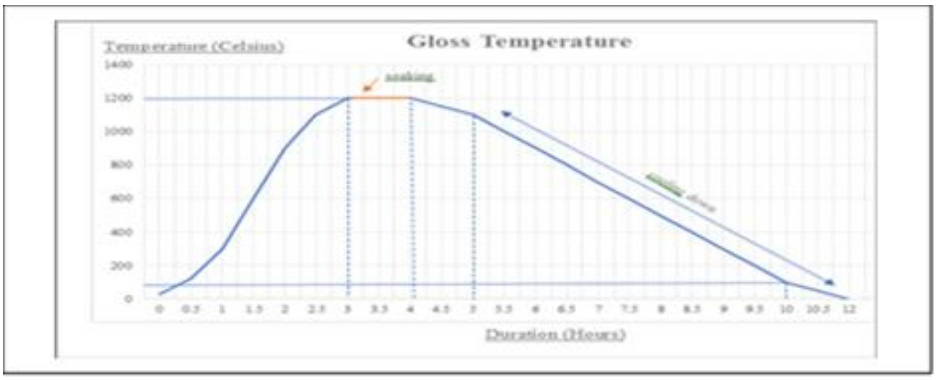

Fig.1. The firing profile at 1200 °C

4.0 Result and Discussion

The study focused on the use of local terracotta clay powder and the identification of oxide phases. X-ray diffraction to identify the raw material utilized in the cleansing process of the local terracotta clay powder. Figure 2(a) in this section displays the phases obtained from the local terracotta clay after the cleansing process. The second peak on the chart indicates the presence of Kaolinite 1A, with a low level of quartz (SiO2) and Zeolite. The sample showed high intensities of Al2O9 and SiO2, with peak intensities visible between 10 and 30 theta position. Each peak corresponds to the intensity of silica content. According to Rhodes (2015), the composition of the molten material, different pressure conditions, and different rates can affect numerous minerals.

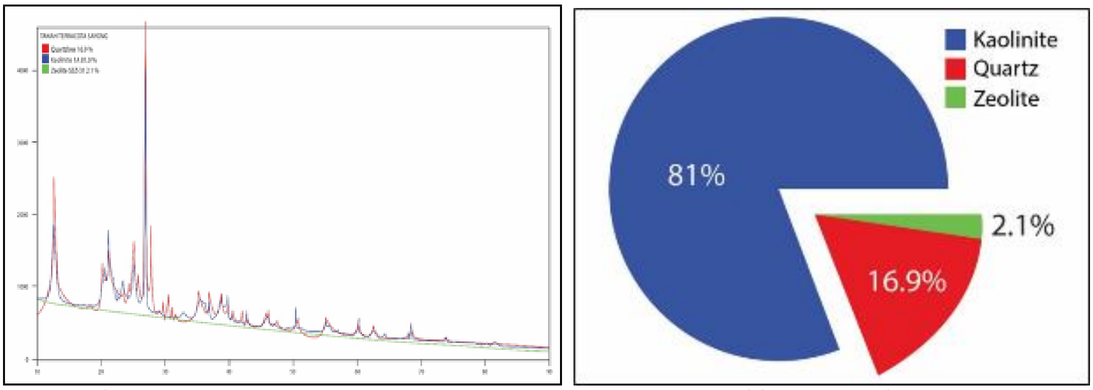

Fig. 2. (a) XRD results for kaolinite, quartz low, and zeolite phase in local terracotta clay powder; (b) XRD results for kaolinite, quartz low, and zeolite phase in local terracotta clay powder.

Figure 2 (b), shows the pie chart of XRD results of kaolinite, quartz low, and zeolite phase in local terracotta clay powder. Moreover, the result showed the highest peak and optimal appearance of Kaolinite at about 81%. Referring to the graph, 16.9% of the appearance phase for quartz is at second high temperatures, and the result for Zeolite Is about 2.1%.

The silica (SiO2) in terracotta clay powder was identified by X-ray diffraction and was higher form other material compositions. The X-ray fluorescence allows obtaining the elemental composition of the sample, while the X-ray diffraction technique permits obtaining the composition of the soil minerals. In this section, the phases of structure composition and all oxide contents in the local terracotta clay are shown in Table 2. Hence the result shows that the raw local terracotta clay powder has 55.56% Silica. The result showed the highest peak and optimal appearance of SiO2. The glassy phase appears at 700 °C, but its intense formation begins at a temperature of 850 °C to 900 °C (Wisniewska K, 2021). Clearly, with increasing temperature the SiO2 content in Terracotta clay increased due to the continued condensation of Silica ions, and eventually formed Si-O-Si bonds (Hamer, 2004; Taylor & Bull, 1986). Aluminium Oxide (Al_2O_3). is found at 35.88%. Alumina is known as an intermediate oxide because it helps build strong chemical links between fluxes and SiO_2 to help keep the materials in suspension.

Next is iron oxide (Fe_2O_3) which is found at 4.57%, in which the content of Hematite in the Iron Oxide helps produce Red Ochre color. Hematite is a dry iron oxide, which means it has little to no chemical water in its crystalline structure. This ochre is responsible for strong reds historically used for paints and stains because of its quick drying time and resistance to sun damage. Ochres are considered a specific group or family of earth pigments and essentially variations of ferric oxides mixed with varying degrees of clay and sand or a calcium-rich, chalky consistency. Ochres are non-toxic and can be quickly introduced into slips and glazes where iron reds are desired. In ceramic, Iron oxide compounds are the most common coloring agent. However, zinc oxide has modified the iron oxide properties and gives some striking variegated effects. The firing temperature of terracotta clay can affect the color of the resulting pottery. A lower temperature can produce pottery with reddish hues due to the presence of iron oxide. The clay also contains 3.07% potassium oxide (K_2O), which acts as a fluxing agent and can create a glassy surface at lower temperatures. When added to glaze formulations, terracotta clay can produce a glassy surface even at temperatures as low as 1140°C to 1200°C.

The clay also contains 0.42% titanium dioxide (TiO_2), which can be reduced to create a lustrous silver transition metal with low density and high strength. Calcium oxide (CaO) and titanium can encourage the formation of decorative colors and effects on glossy glazes. The clay also contains 0.09% rubidium oxide (Rb_2O), a yellow-colored solid material. Finally, the clay contains 0.06% magnesium oxide (MgO), commonly used in crushable ceramics due to its excellent thermal conductivity and electrical resistance, even at high temperatures.

Table 2. XRF analysis results for oxide composition of local terracotta clay powder

Oxide composition	**SiO_2**	**Al_2O_3**	**Fe_2O_3**	**K_2O**	**TiO_2**	**CaO**	**Rb_2**	**MgO**	
Weight percentage	55.56	35.88	4.57	3.07	0.421	0.09	0.09	0.06	

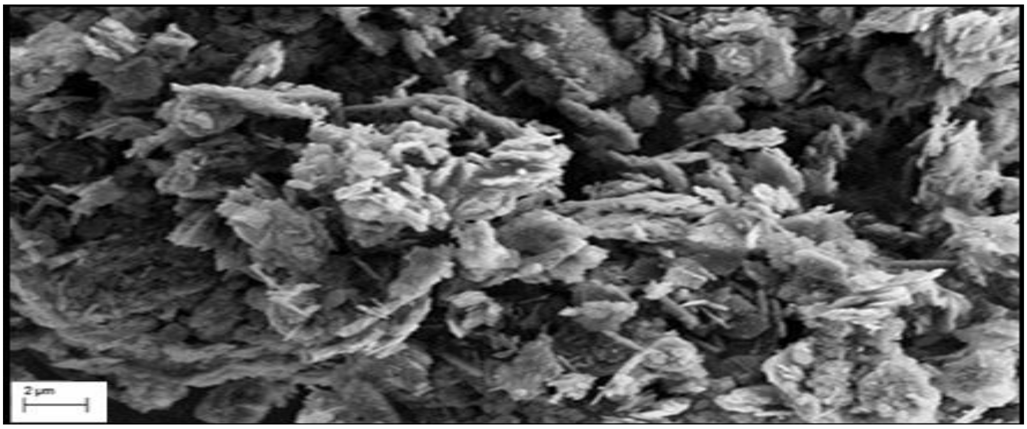

Fig. 3: Illustration SEM results of terracotta clay (Mag 5.00KX), (Signal A SE2),(Wd 5.0mm),(Bht 5.00kv)

The sample of local terracotta clay powder results of the SEM testing shows the characteristic depth of field of SEM micrographs. SEM is a type of electron microscope that produces local terracotta clay powder images by scanning the surface with a forced beam of electrons. Mirco analysis of terracotta clay powder is shown in Figure 4, which shows the main phase of the different coating samples.

The chemical composition structure or pattern of enamel matrix is rich in plate-like structure which are probably clay mineral ones. Different minerals of different sizes are blended and distributed randomly in the materials. This analysis shows that plates of the clay scale 2um appear to be composed of much smaller particles. The unfired sample revealed particles of different shapes and sizes the presence of coarse and porous particles is also observed. The terracotta clay powder is rich in Kaolinite platelets are visible. SEM image of coarse fraction, separated from clay, shows the presence of some large particles of quartz and kaolinite. Quartz particle at higher magnification shows spongy-like surface. Indicates that the unfired kaolinite shows isolated quartz low and Zeolite. The samples are composed of clay particles deposited in a face-to-face manner with small voids which show that the clay contains iron impurity. According to Lahuta, clay mineralogy is an important factor controlling the physical, chemical, and mechanical properties (Lahuta H., 2021).

The basic formulation has been confirmed to give a stable result of glaze. Next, this research continued with the investigation of adding raw terracotta clay powder in glaze formulation as shown in Table 1. Based on the transparent formulation specified in Table 1, it has four samples for terracotta glaze recipe results (A1–A4). All materials used are the same percentage, except for conventional local terracotta clay which was added in the range of 20% to 50%. The material is 33% potash feldspar, 32% silica, 15% zinc oxide, 20% calcium carbonate, and terracotta as a coloring agent. Its purpose is to compare the effects of varying the percentage of terracotta in the transparent glaze.

Table 3, shows the result of colored terracotta clay samples fired at 1200 °C of gloss temperature with soaking for one hour at 1200 °C of glaze temperature. Samples A1 and A4 were perfect, and the glaze produced a glossy surface with the color of terracotta clay. The color sample A1 local terracotta, which accounts for approximately 20% of the findings, has a very light cream-coloured surface. The color for sample A2, which included 30% local terracotta clay, had a glossy surface, smooth melt, and was cream in color, which Is reflected in the sample form. The following sample A3, with 40% terracotta powder with the same glaze formulation, shows a completely colored surface on the layer of the sample, which shows the melting effect of color. Finally, sample A4 with 50% terracotta clay powder added had more colorization occurring on the surface layer of the glaze. Previous research has shown that despite having such a high glaze, compositions based on this system will result in a low gloss when compared to the gloss typically obtained with frits.

Table 3. Glazing 1200˚C temperature transparent glaze on porcelain sample

	Raw material (wt%)			
Sample code	A1	A2	A3	**A4**
				
Colorant %	**20%**	**30%**	**40%**	**50%**

*The samples were prepared according to the testing clay body porcelain on terracotta clay as a colorant glaze. The sample varied in terms of percentage (20%, 30%,40%,50%) for the local terracotta clay with a temperature of 1200°C

5.0 Conclusion

After conducting several experiments using terracotta clay as a colorant for the glaze, it was applied to the final product to determine the suitability of the glaze formulation with the addition of terracotta clay powder to all samples. The results showed that terracotta clay as a colorant glaze produced a cream-like color and gave a good result at 1200°C. Various tones at different percentages produced colors ranging from bright to medium tones.

The stability of materials, composition, and temperature during glaze firing is critical for this research. It ensures that the glaze does not flow or melt while still producing a good appearance on the glaze basic mixture as an alternative to the conventional colorant. The flux material influences the glaze stability process, including the reduction of the firing temperature's maturity on the porcelain body. Additionally, the firing process can also optimize the sample, making it free from defects such as bubbles, crazing, peeling, speaking, crawling, or tearing.

In summary, the local terracotta clay offers a range of natural colors for ceramic production while also being cost-effective and maintaining glaze stability. This could potentially provide a new source of colorant for the ceramics industry. The investigations conducted have yielded valuable information that can be used in future studies on the use of raw clay residues in pottery, as well as their behaviors and impacts on the final product.

This could be a starting point for the researcher or other researchers to perform or view this study from a specific point of view and explore alternate ways to reach a more positive conclusion from this research. Terracotta has the potential to replace traditional colors in ceramic glazes, particularly when low-cost components are utilized. Because color glazes often incorporate flux agents, the glaze maturation temperature is 1200°C.

Future studies could investigate glaze formulation and temperature to learn more about color development through a wide variety of colors and natural glazes. With their impact on color and texture, slip glazes, which are simple to create using generally available and affordable materials, can open up new possibilities for ceramic manufacturers and ceramic artists.

(1)

Acknowledgments

We would like to acknowledge the financial support provided by Universiti Teknologi MARA under PYBP and express our gratitude to the interaction designers for their generous participation in the research. We also appreciate the support and resources provided by the International Collaborator,Conference on Science & Social Research and National Design Centre, UiTM.

References

Anantakarn, K., Sornchomkaew, P., Phothong, T. (2019). Improve Quality of Global DEM for Topographic Mapping: Case Study of Petchaburi Province, Thailand. International Transaction Journal of Engineering, Management, & Applied Sciences & Technologies. 10(9), 10A09H, 1-9.

Anwar, R. (2016) Characterizing a syntactic pattern of form giving in design thinking process Universiti Teknologi MARA

Anwar, R., Abidin, S.Z. and Hassan, O. H. (2015). A framework of empirical study through design practice for industrial ceramic sanitary ware design. International Colloquium of Art and Design Education Research (i-CADER 2014) Springer Singapore. Pp. 683-694

Anwar, R., Salleh, M. R. Vermol, V. V., Zakaria, Z. and Hassan, M. R. (2015). Hard ceramic porcelain physical test through potential formulation parameter. Conference Proceedings of the International Symposium on Research of Arts, Design and Humanities (ISRADH 2014) Pp 323-332 Springer Singapore

Anwar, R., Kamarun, Vermol, V. V., and Hassan, O. H. (2011) Marble dust incorporated in standard local ceramic bodies as enhancement in sanitary ware products.2011 IEEE Colloquium on humanities, science, and engineering. pp 355-357. IEEE.

Genc, Oztoprak.B, S. (2016). Composition analysis of medieval ceramics by laser-induced breakdown spectroscopy(LIBS). Applied Physics A, 1-19.

Raif, D. M, Ibrahim, N. S., Vermol, V. V. and Anwar, R. (2015). The potential of Coldstream bidor clay (CBC) as a replacement for porcelain body. Conference Proceedings of the International Symposium on Research of Arts, Design and Humanities (ISRADH 2014) Pp. 313-321. Springer Singapore.

Hansen, T. (2017). Digitalfire.com. Retrieved Nov 24, 2017, from Digitalfire Web site: http://www.digitalfire.com

Ibrahim, N. S., Raif, D. M., Vermol, V. V., Anwar. R. (2015). Reformulating glaze defect recipe to recycled as ceramic surface treatment. Proceedings of the International Symposium on Research of Arts, Design and Humanities (ISRADH 2014) Pp. 89-100. Springer Singapore.

Jordanova,N., Jordanova, D., Barron, V., Lesigyarski, D., & Kostadinova-Avramova, M.(2019). Roack-magnetic and color characteristics of archaeological and Anthropological Sciences, II (7), 3595-3612.

Lahuta H., P. L. (2021). Research of Behavior of Clay Materials with Double Porosity. MDPI, 13,3219.

Malek, M. F. K. A., Anwar, R. and Hashim, H. Z. (2022). Emerging Practices of Design and Economics as Studio-Ceramic Entrepreneurial Concepts Journal Environment-Behaviour Proceedings Journal Vol. 7 Special Issue 7 Pp. 191-197

Olazabal, A. A, O. L. (2022). Glaze Characterization Of the Glazed Pottery From The Medieval Workshop of Vega (Burgos, Spain). Raman Spectroscopy, 1204-1213.

Rhodes, D. (2015). Clay and Glazes for Potter., Krause Publications

Suman, S. N., Anwar, R., Awang, N. N. and Ayob (2022). Case Study of Ceramic Firing Profile for Terracotta-Based Glaze. Journal Environment-Behaviour Proceedings Vol. 7 Special Issue 9 P. 123-130

Williams, H., (2006). Glaze Chemistry Primer, Second Edition. www.hamiltonwilliamsclayworks.com/glazeprimer.pdf

Wisniewska.K., P. (2021). Influence of Firing Temperature on Phase Composition and Color Properties of Ceramic Tile Bodies. Material 2021, 1-19.