

THE DEVELOPMENT OF CERAMICS CLAY BODY AND GLAZE FROM RECYCLING MATERIAL TO DECREASE PRODUCTION COSTS AND PRESERVING THE ENVIRONMENT

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Abstract

Waste recycling is one of the interesting topics in academia and industry. This research has 2 sections. The first section focuses on rice husk ash recycling to substitute as silica in the system of ceramic clay body. The second section focuses on defining the possibility of developing a recycling glaze from recycled glass. For the first part, 11-point Biaxial Blend were used to corroborate the composition of ceramics clay body between rice husk ash and Bannasan Surajathanee ball clay. All of the raw materials were ground for 1 h, forming by slip casting and sintered at 1,200 and 1,250°C for 1 h. The test pieces were physical properties tested for firing shrinkage and water absorption. Test piece No. 5 (40% rice husk ash and 60% ball clay) sintered at 1,200°C were the best properties at 30.4% of water absorption, 52.98 apparent porosity and 9.38 % firing shrinkage. For the second part, the result shows a complete melting point of recycling glass is 1,200°C. Kaolin, Ranong province and transparency bottles were used as starting raw materials in this research. The recycling glass was mixed with kaolin by 21-point biaxial bland, ground and sintered at 1,200°C. The results show samples No. 15-16 had the best appearance such as completely melting, good transparency, fine cracking, glossy and slightly down to cover a pattern and emphasis. It can save more than 60% of the commercial raw material. It is also beneficial to the ceramics industry in terms of cost savings and preserving the environment.

Keywords: Ceramics clay body; Glaze; Recycling Glass; Rice husk ash

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Introduction

Thailand is an agricultural country that cultivates a diverse range of economic crops, including rubber, rice, corn, cassava, and sugarcane, among others. Rice, in particular, stands out as the country's most significant agricultural product, and the country holds the position of the world's second-largest rice exporter. It is cultivated extensively across the nation, with a notable concentration in the lower northern and central regions. When rice is grown, after harvesting, we're left with rice straw and rice husks. Rice straws are handy for feeding livestock and for farming purposes. Rice husks, when burned, turn into rice husk charcoal, which is used in making soil mixes. Fresh rice husks are also used as fuel to generate electricity. Additionally, in traditional ceramic production like pottery and clay bricks, fresh rice husks mixed with deciduous wood are often used as fuel for firing kilns, among other applications (Jitchanwichai, 2010). When rice husks obtained from rice milling are burned, they produce rice husk ash, which consists primarily of silica or silicon dioxide (SiO_2) at approximately 15-20% by weight of the husk and accounts for about 95% by weight of the ash. The silica is obtained from rice husk ash that has been burned at temperatures below 800°C (Kaewkamnerd, 1998).

Ashes from different plants have traditionally served as raw materials for ceramic production. In certain Thai provinces, ashes from specific plants are utilized to create ceramic glazes. Notably, rice husk ash comprises over 85% silica and around 1-12% carbon. Silica serves as a fundamental material for synthesizing substances like silicon carbide, which results in robust mullite and silicon nitride. These materials boast corrosion resistance and can withstand high temperatures effectively (Sujirote *et al.*, 2002). Currently, most raw materials utilized in ceramic production are imported from overseas, including flint and quartz. Another raw material, known as Hanuman toothstone, yields silica, typically priced at no less than 8 baht per kg. Silica is an important raw material and is required in large quantities in coatings, and clay for ceramic production. However, it may not be possible to completely replace rice husk ashes, but it can still help reduce the use of quartz or Hanuman toothstones. And Flint can get off. Silica stands as a crucial raw material, essential in significant amounts for coatings and clay in ceramic manufacturing. Although completely substituting rice husk ashes might not be feasible, they can still aid in minimizing reliance on quartz, and even flint, to some extent. The key distinction lies in the fact that rice husk ash is derived from a product

of the agricultural sector-rice cultivation-which ensures a sustainable supply. In contrast, the minerals mentioned above are finite resources that may eventually be depleted with continuous usage.

Raw material from waste is intriguing. The use of waste causes clay body to be environmentally friendly in addition to sintering at the lowest temperature possible. Moreover, value-added waste is also gained.

The first of the interesting recycling raw materials is Rice husk ash. Thailand is an agricultural country. At the end of the harvest season, there will be a lot of rice husks from the rice mill. Most of them will be used as a source of bioenergy. There are many research studies on how to recycle rice husk ash. The remaining rice husk ash is used as a raw material in many industries. Rice husk ash gives about 99% silica in amorphous form. Silica is one of the important ingredients in the ceramics industry.

The second interesting recycling raw material is recycling glass. Recycled glass bottles in Thailand are cheaper when compared to other countries in Southeast Asia. The price is between 30-50 US Dollars per ton yet varies by quality. Glass contains silica, calcium oxide and sodium oxide as the main elements which have chemical properties similar to glaze.

This research focused on the properties of ceramic clay bodies from Rice husk ash and the application of recycling glass in producing glaze on ceramic. The research aimed to identify the factors in the production process that affected ceramic product properties when glass was applied as the ceramic glaze. In terms of cost savings and preserving the environment.

Experimental Procedure

The majority of the raw materials are sourced from C.K. Ceramics Co., Ltd., which has been engaged in managing products for over 1,000 industrial ceramic items, including glassware and general chemicals. They have evolved into importing chemicals from various countries worldwide for distribution to leading controllers and companies in Thailand. Researchers conducted fieldwork in Nakhon Pathom province, collecting samples of rice husks that had undergone the burning process and were agricultural waste. These samples were then analyzed for chemical composition using X-ray fluorescence (XRF) and mineralogical components using X-ray diffraction (XRD) techniques.

Table 1. Chemical composition of raw materials (wt%)

Oxide	Al ₂ O ₃	SiO ₂	CaO	MgO	Na ₂ O	K ₂ O	Fe ₂ O ₃	TiO ₂	L.O.I.
Rh	0.12	95.0	1.19	0.46	0.10	1.44	0.84	0.01	-
Ball Clay	23	51.5	0.37	0.12	0.22	0.92	1.42	0.63	19.6

The researchers analyzed rice husk ash obtained from agricultural burning processes in Nakhon Pathom province. They examined the particle size and post-burning color of the rice husk ash, as well as recycled glass, specifically recycled glassware collected from household waste or leftover materials, which could be repurposed for new uses. These materials were then transformed into raw materials for the production process, incorporating white soil from Ban Sompian Beach, Ranong province, and black soil from Nasa Sub-district, Surat Thani province. The researchers subjected the rice husk ash to screening and conducted analyses of the resulting narratives and mineralogical history using XRD techniques.

For the testing of ceramic materials, the process involved mixing at a ratio that had been ground for 1 h and then subjected to firing at temperatures of 1,200°C and 1,250°C. This was followed by testing on specimens sized 5 cm × 7.5 cm × 0.7 cm for physical properties such as water absorption, flexural strength, and chemical composition analysis using XRF technique and wavelength dispersive X-ray fluorescence (WD-XRF) with Rh radiation in a Bruker S4 Pioneer instrument. Structural analysis was conducted using XRD with CuKα radiation in a Bruker D8 Advance Diffractometer. Particle size distribution analysis was performed using a Laser Scattering Particle Size Analyzer LA-950. The physical properties tested included porosity according to ASTM C373-88 (Reapproved 1999) standard.

For coating testing, after the separation of glass bottle waste, it undergoes a recycling process that begins with a cleaning step. The procedure includes the following steps: Testing the melting point between 800-1,200°C by mixing with white soil using the Biaxial Blend (Line Blend) technique. Various formulas obtained from the mixing ratios are combined and finely ground. Then, a coating with a thickness of 1-1.5 mm is applied to the test pieces. Physical analysis is conducted on the products to examine the following properties: (1) Melting ability; (2) Texture and color; (3) Transparency and density; (4) Separation of coating material and product; and (5) Coating flow smoothness. The final two steps include testing for suitable colors and applying the coating color on the products.

Results and Discussions

Chemical Compositions

In this research, we utilized gray rice husk ash, one of the three types of rice husk ash distinguished by color, alongside white and black variants. The chemical compositions of gray rice husk ash and ball clay are shown in Table 1.

The physical property study using test pieces to determine the mixing ratios can be conducted by combining the materials used in the experiment, which include rice husk ash, Nakhon Ratchasima white clay, and black soil. These materials are then baked at a temperature of 110°C for 5 h to ensure uniform moisture content. Subsequently, the materials are divided for particle size distribution testing using a particle size distribution analyzer. Afterwards, prepare the rice husk ash by washing and baking fresh rice husks at 110°C for 5 h. Then, burn them at a temperature of °C for 6 h. Finally, wash the burned rice husk ash using the washing method described by Nakbua (1993).

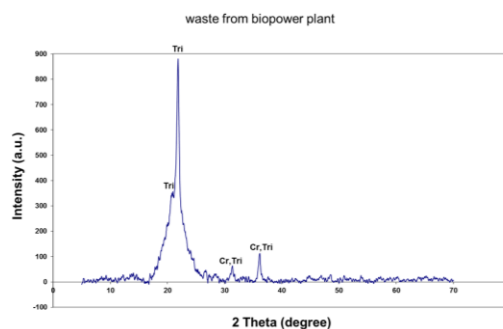


Figure 1. XRD pattern of the rice husk ash and the standard peak positions of Tridymite (Tri), and Cristobalite (Cr)

Figure 1 displays the XRD pattern of rice husk ash, revealing the presence of Tridymite and Cristobalite phases. The median particle size and calculated specific surface area of the raw materials are presented in Table 1.

In Figure 2, water samples mixed with particles were tested to determine particle size using a Laser Scattering Particle Size Analyzer, LA-950. The sample preparation procedure was as follows:



Figure 2. Precipitation of rice husk ash (4% by weight) suspended in water 1, 5, 60, and 180 min

-Dispersant: water-Circulation Speed: 3-Ultrasonic speed: 5 (20 sec) The refractive index of silica used in the analysis was 1.459-0.000i. The testing was conducted by allowing a suspension of rice husk ash (4% by weight) to settle in water, with observation intervals at 1, 5, 60, and 180 min.

Physical properties were examined using primary raw materials comprising rice husk ash and black soil obtained after the burning process. Each type of prepared raw material was mixed in various ratios and ground in a 5.00 kg grinder for 30 min. Rice husk ash powder was obtained through grinding processes lasting 1, 2, 3, 4, and 5 h. The resulting samples, mixed with water, were subjected to particle size analysis using the Laser Scattering Particle Size Distribution Analyzer LA-950. The sample preparation included using water as a dispersant, with a circulation speed of 3 and an ultrasonic speed of 5 (20 sec). The refractive index of silica used in the analysis was 1.459-0.000i. The test results are depicted in Figure 3.

Figure 3 shows the comparison of rice husk ash in the time between 1-5 h. The grey, green, blue, red,

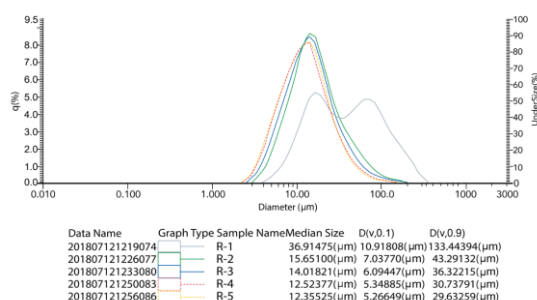


Figure 3. Particle size distributions of rice husk ash

and yellow lines represent the particle of rice husk ash through grinded in 1, 2, 3, 4, and 5 h, respectively.

Physical Properties of Clay Body

Figures 4 and 5 show the linear shrinkage and the water absorption, respectively, after firing of samples 1-5 at 1,200°C and 1,250°C. The sample 5 after firing at 1,200°C got the lowest shrinkage

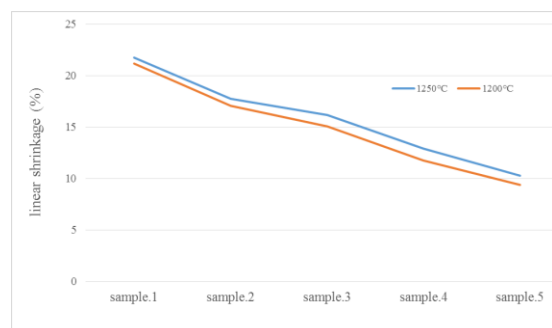


Figure 4. The linear firing shrinkage of the samples after firing at 1,200°C and 1,250°C

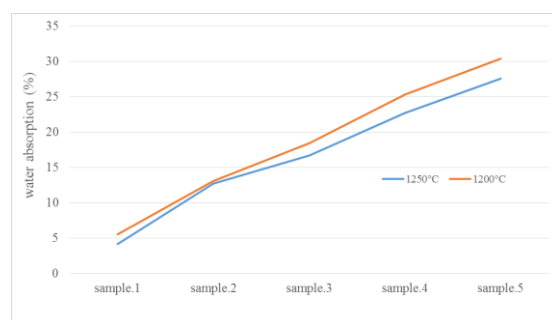
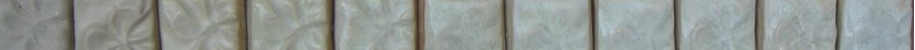


Figure 5. The water absorption of the samples after firing at 1,200°C and 1,250°C

Sample No.	1	2	3	4	5	6	7	8	9	10
Kaolin	100	95	90	85	80	75	70	65	60	55
Glass	0	5	10	15	20	25	30	35	40	45
Test piece										

Sample No.	11	12	13	14	15	16	17	18	19	20	21
Kaolin	50	45	40	35	30	25	20	15	10	5	0
Glass	50	55	60	65	70	75	80	85	90	95	100
Test piece											

ground for 1 h, forming by slip casting and sintered at 1,200°C and 1,250°C for 1 h. The test pieces were physical properties tested for firing shrinkage and water absorption. Test piece No. 5 (40% rice husk ash and 60% ball clay) sintered at 1,200°C were the best properties at 30.4 % of water absorption, 52.98 apparent porosity and 9.38 % firing shrinkage.

For the second part, the result shows the complete melting point of recycling glass is at 1,200°C. Kaolin, Ranong province and transparency bottles were used as starting raw materials in this research. The recycling glass was mixed with kaolin by 21-point biaxial bland, ground and sintered at 1,200°C. The results show samples No. 15-16 had the best appearance such as completely melting, good transparency, fine cracking, glossy and a slightly down to cover on a pattern and emphasis. It can save more than 60% of the commercial raw material.

It also contributes to cost savings for the ceramics industry, with reductions of 40% for clay body and 60% for glaze, while simultaneously promoting environmental conservation. From experiments replacing silica with rice husk ash for ceramic production, it was found that the characteristics of rice husk ash as a raw material exhibited varying degrees of fineness. Rice husk ash had an average particle size of 106 µm, while white and black soils averaged 45 µm or higher. This aligns with the statement by Ingsiriwat (1998) which states that raw materials suitable for mixing with clay body and glaze or for use in ceramic production processes must have a high degree of fineness and be able to pass through a 100 mesh (150 µm) sieve. All types of raw materials used in this research had sizes no smaller than 95% passing through a 120 mesh (125 µm) sieve. Grinding the materials further with a grinder helps reduce their particle size. Upon examining the characteristics of rice husk glazes, it was found that the glossiness level of these glazes, especially those with a rice husk ash content exceeding 40 %, was not fully developed. According to Kosiyapan (1991), analyzing each type of rice husk ash reveals little variation in their compositions. Components

contributing to the glass formation include silica, fire-resistant aluminum, and fluxing agents. Rice husk ash contains a high silica content, up to 96%. According to Kato (2010), rice husk ash is primarily composed of silica, which is classified as an acidic material in ceramic work. When used in glaze mixing, silica helps increase the melting point of the glaze. Therefore, if used in excessive amounts, it can enhance the glaze's ability to withstand high temperatures. Conversely, an appropriate amount of flint and feldspar, which serve as fluxing agents, can lead to the formation of semi-matte and glossy glazes. Feldspar belongs to the group of fluxes and aids in the glaze's maturation.

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References

- ASTM C373-88. (Reapproved 1999). Standard Test Method for Water Absorption, Bulk Density, Apparent Porosity, and Apparent Specific Gravity of Fire Whiteware.
- Ingsiriwat, P. (1998). Ceramic Body. Bangkok: Odean Store, 177p. (in Thai)
- Jitchanwichai, T. (2010). An Experiment of Zircon Body for Ceramic Materials Grinding. VRU Research and Development Journal Science and Technology, 5(2):13.
- Kaewkamnerd, K. (1998). Fire-resistant Materials. Chiang Mai: Ceramics Technology Research Center, Thailand, 494p.
- Kato, E. (2010). The Fundamentals of the Glaze Preparation. Chulalongkorn University Press, Thailand, 211p.
- Kosiyapan, S. (1991). Water Glaze for Pottery. Thai Watthana Panich, Thailand, 106p.
- Nakbua, S. (1993). Coating with plant ashes. J. Film Process, Thailand, 220p.
- Sujirote, K., Saewong, P., and Thavorniti, P. (2002). The production of ceramics by pressing. National Metal and Materials Technology Center, Thailand, 90p.