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Research article

Determination of the Firing Temperature and Optimum Ratio of Valorizing Rice Husk Ash, Clear Glass Cullet and Ranong Clay for Developing Ecological Wall Tiles

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Abstract

Keywords

eco-friendly;

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rice husk ash;

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waste utilization

The purposes of this study were to investigate the effect of firing temperature and utilizing residue materials on the physical properties and microstructure of the ecological wall tiles. Rice husk ash (RHA), clear glass cullet (CGC), and Ranong clay (RNC) were utilized. Eleven formulations were constructed and divided into two groups with different contents of RHA, at 10% and 20%. After molding by uniaxial pressing at 10 MPa, the tested specimens were fired at temperatures of 950 and 1050ºC, with heating rate of 100ºC/h, and soaked for 1 h. The bending strength of specimens fired at 1050ºC were enhanced by up to 205% compared with specimens fired at 950ºC. Formula B1, fired at 1050ºC was optimal for further commercial production. It utilized 20 wt% RHA, 50 wt% CGC, and 30 wt% RNC which met the criteria of the Thai Industrial Standard (TIS) 2508-2555 type BIII. Moreover, the microstructure of specimens was characterized by scanning electron microscopy (SEM). It revealed that a glassy phase had developed in the high bending strength specimens under the influence of high firing temperature and CGC content. In addition, RHA residue material promoted a greyish color in ceramic bodies. Finally, it can be concluded that the valorization of RHA, CGC, and RNC in the development of ecological wall tiles with white-greyish texture was feasible.

1. Introduction

Ceramic tiles are one of the construction products that are available in the Thailand market. Inorganic and non-metallic materials are used as the main materials for producing ceramic products. Explosion booms in mining across the mountains for supplying minerals for the ceramics industry has caused degradation of the environment. Therefore, environmentally friendly manufacturing for

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ceramic products has become an important issue. Using waste from the industry sector as alternative materials for developing ceramic items is the challenge work. Thailand is ranked 6th in the world for rice production [1]. Rice husks generated during the first stage of rice milling have been used in biomass power plants as fuel for boiling water to generate steam and produce electricity. Rice husk ash is a by-product generated from biomass power plants. It was noted that silica (SiO_2) is a chemical component of rice husk ash that can be utilized for producing ceramic tiles [2]. Another waste material in Thailand, glass cullet, is left unutilized (32%) [3]. As it contains the oxides of potassium, sodium, and calcium, it can be recycled in ceramic tiles for reducing firing temperature [4]. Ranong clay or kaolin is a local clay that is mainly used in the ceramic tile industry in Thailand. It promotes the white texture of ceramic tiles and also contains a relatively high amount of silica and alumina (SiO_2-Al_2O_3) [5].

There are many researchers studying ways of exploiting wastes. The physical properties of ceramic products that incorporated rice husk ash were investigated. It was found that a sample consisted of 10% rice husk ash, 40% Ranong clay sintered at 1200ºC was optimal [6]. Utilizing local clay, white clay, sand and wood sawdust in Thailand for developing refractory brick has been performed. The optimal portion of sawdust up to 20 wt% and with firing at 1300ºC was proposed [7]. The firing behavior of three natural products; red quartzitic clay, kaolin, and yellow quartzitic clay was studied. Materials containing kaolin yielded the best overall behaviour independently of the kaolin quantity. The optimal sintering temperature that produced the best properties of the materials using red or yellow clay was above 1080ºC [8]. Feldspar was replaced by glass cullet to reduce the sintering temperature of ceramic floor tiles from kaolin and quartz. The study showed that cullet could reduce firing temperature of samples, and was fired at lower than 1150ºC [9]. For fast firing, the sintering behavior of tiles containing 40 wt % glass cullet, 60 wt% paper mill sludge, was examined. The results indicated that these materials added with 30 wt% red clay could be employed to develop high strength products for producing industrial tiles [10]. The microstructure of cullet-clay ceramics that had been made by slip casting and fired at 900ºC was investigated. It was found that the ceramics had a high content of Na, Al, and Si while the phase of SiO_2 and Al_2SiO_5 was a minor phase [11].

The purpose of this study is to investigate the effect of firing temperature on the physical properties and microstructure of ceramic bodies. In addition, the effects of utilizing rice husk ash and clear glass cullet waste on properties and texture of ecological wall tiles were also studied.

2. Materials and Methods

The chemical composition of raw materials was determined by X-ray fluorescence analysis (XRF), as shown in Table 1. Rice husk ash (RHA) was obtained from burnt rice husk from the biomass power plant in Suphan Buri province, Thailand. Clear glass cullet (CGC) was obtained from a recycling factory in Ayutthaya province, Thailand. Ranong clay (RNC) is a kaolinite mineral from Ranong province, Thailand. It is a local clay that is utilized for promoting plasticity of specimens.

Table 1. Chemical composition analysis of materials with X-ray fluorescence (XRF)

Materials	Chemical composition (%)										
	SiO_2	K2O3	CaO	P_2O_5	MgO	Fe_2O_3	Al_2O_3	Cl	SO_3	MnO	Na_2O
RHA	92.67	1.96	1.18	1.50	0.78	0.54	0.56	0.16	0.34	0.13	0.07
CGC	71.08	0.18	10.11	-	2.48	0.12	1.69	0.03	0.12	-	14.11
RNC	50.84	1.27	-	-	-	0.88	46.79	-	0.02	0.06	-

All materials were dried at temperature 200ºC for 2 h. Then, they were filled into a ball mill to be ground into fine particles. The milling time of RHA and RNC was 30 min, and it was 1 h for CGC. The materials were then sieved with 50 mesh (297 µm). All 11 mixture formulations were prepared, as shown in Table 2. The rectangular specimens were molded with dimensions of 50 x 100 x 7 mm by uniaxial pressing at 100 MPa. All specimens were dried at temperature of 200ºC for 2 h, fired at temperatures of 950 and 1050ºC with heating rate of 100ºC/h, and then soaked for 1 h. Because 1050ºC is typically used in the fired clay industry, 1050ºC was used in this experiment. In addition, a lower firing temperature (950ºC) was also used in this study. After the firing process, the fired specimens were cooled down naturally to room temperature in the electric kiln.

Table 2. Mixture formulations of this experiment

		Formula No										
		A1	A2	A3	A4	A5	A6	B1	B2	B3	B4	B5
(%) Raw materials	RHA	10	10	10	10	10	10	20	20	20	20	20
	CGC	60	50	40	30	20	10	50	40	30	20	10
	RNC	30	40	50	60	70	80	30	40	50	60	70

Bending strength was evaluated by three-point bending. The parameters related to the calculation of bending strength are described as follows. P is the maximum force applied (N). L is the support span (mm). The a and b are thickness and width of each tested piece (mm), respectively.

The calculation the bending strength of specimens is shown in equation (1).

$$\text{Bending strength} = \frac{3PL}{2\text{a}^2 b} \tag{1}$$

Water absorption parameters are calculated as shown in equation (2). W_d in gram (g) is the dry weight of specimens before boiling. W_w is the wet weight of specimens after boiling (g) in water for 2 h and being immersed in water for 24 h.

$$\%\text{Water absorption} = \frac{w_w - w_d}{w_d} \text{x}100 \tag{2}$$

Bending strength and water absorption properties were tested and compared with the criteria of the Thai Industrial Standard 2508-2555 type BIII [12]. X-ray fluorescence (XRF) is an X-ray technique used for the chemical analysis of powder materials using a WD-XRF spectrometer (Model S8 TIGER, Bruker, Germany). The microstructures of fried specimens were examined with a scanning electron microscope (SEM Hitachi, SU3500) at voltage set on 10 kV for 1000x, 3000x, and 5000x magnification.

3. Results and Discussion

As the properties of fired specimens have been mentioned in the previous section, their results will be described below.

3.1 Fluxing agent

Before describing the physical properties and microstructures of fired specimens, the components of the fluxing oxide, Na_2O, K_2O, and CaO, are considered. They play the important role of considerably reducing sintering temperature and developing the strength of ceramic bodies [13]. Therefore, the fluxing agent makeup of each formula was calculated. It was evaluated from chemical compositions consisting of K_2O, Na_2O, and CaO, as shown in Table 3. The largest proportion of the fluxing agent, K_2O, CaO, and Na_2O were from CGC. Therefore, formula A1, which contained the highest content of CGC (60 wt%.), provided the highest content of fluxing agent.

Table 3. Fluxing agent content of all formulas

%Oxide	Formula no.										
	A1	**A2**	**A3**	**A4**	**A5**	**A6**	**B1**	**B2**	**B3**	**B4**	**B5**
K_2O	0.68	0.79	0.90	1.01	1.12	1.2	0.86	0.97	1.08	1.19	1.29
Na_2O	8.47	7.06	5.65	4.24	2.82	1.41	7.06	5.65	4.24	2.83	1.42
CaO	6.18	5.17	4.16	3.15	2.14	1.12	5.29	4.28	3.26	2.25	1.24
Total	15.34	13.02	10.71	8.40	6.09	3.77	13.22	10.91	8.59	6.28	3.97

3.2 Bending strength

The bending strengths of specimens in groups A and B fired at 950 and 1050ºC were compared, and are illustrated in Table 4. The bending strength of the formulas of groups A and B varied between 26.6-158.8% and 37.9-205.6%, respectively. The best formulas of groups A and B were formulas A1 and B1, which had been fried at 1050ºC. The bending strength of them had improved by 120% and 205%, respectively, when compared with specimens fired at 950ºC. It means that the higher firing temperature significantly enhanced the bending strength of tested pieces.

It should be noted that the maximum content of CGC utilized in this study was 60 wt%, which provided the highest bending strength of fired specimens. This was similar to the findings of Esposito *et al*. [14]. It corresponded with the high content of fluxing agent.

Table 4. Comparison of the bending strength of specimens fired at 950 and 1050 ºC

		Group A						Group B				
	Formula no.	**A1**	**A2**	**A3**	**A4**	**A5**	**A6**	**B1**	**B2**	**B3**	**B4**	**B5**
Mixture formulation (%)	RHA	10	10	10	10	10	10	20	20	20	20	20
	CGC	60	50	40	30	20	10	50	40	30	20	10
	RNC	30	40	50	60	70	80	30	40	50	60	70
Bending strength (MPa)	950ºC	10.9	7.4	5.5	5.1	4.5	3.8	5.9	3.6	3.3	2.9	2.5
	1050ºC	24.1	19.2	11.1	9.7	6.7	4.9	18	10.8	6.2	4.6	3.4
	Increased (%)	120.4	158.8	101.6	89.8	50.1	26.6	205.6	204.5	86.6	57.8	37.9

3.3 Water absorption

A comparison of the water absorption of two formulas group fired at 950 and 1050ºC has been depicted in Table 5. The firing temperature at 1050ºC could reduce the water absorption of ceramic bodies compared to specimens fired at 950ºC. It decreased the water absorption by 4.4-36.7% and 5-33.9% of groups A and B, respectively. The higher firing temperature promoted the removal of porosity in ceramic bodies.

In addition, a decrease of CGC content increased the water absorption of the tested pieces, with the same result being seen for groups A and B. It can be concluded that higher CGC content and firing temperature can decrease the water absorption of specimens. The fluxing agent in CGC played the role of melting oxides and filled in the pores of specimens. This was in agreement with the conclusions of Demir [15]. On the contrary, the higher content of RHA and RNC had the effect of increasing the water absorption of fired specimens in the two groups.

Table 5. Comparison of the water absorption of specimens fired at 950 and 1050ºC

		Formula of group A						Formula of group B				
		A1	**A2**	**A3**	**A4**	**A5**	**A6**	**B1**	**B2**	**B3**	**B4**	**B5**
(%) Raw material	RHA	10	10	10	10	10	10	20	20	20	20	20
	CGC	60	50	40	30	20	10	50	40	30	20	10
	RNC	30	40	50	60	70	80	30	40	50	60	70
Water absorption (%)	950ºC	19.6	20.2	22.5	24.2	26.4	28.8	25.7	27.4	28.6	31	33.5
	1050ºC	12.4	13.4	18.2	19.8	23.6	27.6	17	20.4	25	28.2	31.9
	Decreased (%)	36.7	33.8	19.2	18.2	10.5	4.4	33.9	25.5	12.7	9.2	5

3.4 Microstructure analysis

The microstructures of the fired specimens were characterized by scanning electron microscope (SEM) at 10 kV for 1000x, 3000x, and 5000x magnification. The formulars with the lowest and highest bending strengths, both of which contained 10 wt% RHA, formulas A1 (Figure 1a, Figure 1b) and A6 (Figure 1c, Figure 1d) in group A, and both of which had been fired at 1050º C, were selected for analysis because the big difference in their bending strengths. Formula A1, which consisted of 60 wt% CGC, had developed a glassy phase and a dense structure, which led to it having the highest bending strength of the fired specimens, as illustrated in Figure 1, which was in line with previous studies [16-18]. Meanwhile, formula A6, which contained low CGC and high RNC, had a highly porous structure.

Next, considering group B specimens with 20% RHA content fired at 1050ºC, formulas B1 and B5 were selected as they had the high differences in bending strengths. Their microstructures are s illustrated in Figures 2a, 2b, 2c, and 2d. However, the dense structure and glassy phase of formula B1 are less than that of formula A1, due to the effect of the higher RHA and lower CGC content in the specimens, which is a similar result to De Silva and Surangi [19]. Therefore, formula B1 displayed decreased bending strength and increased water absorption when compared to formula A1. The pore distribution of formula B5 were greater than formula B1. This was in accordance with previous findings [20].

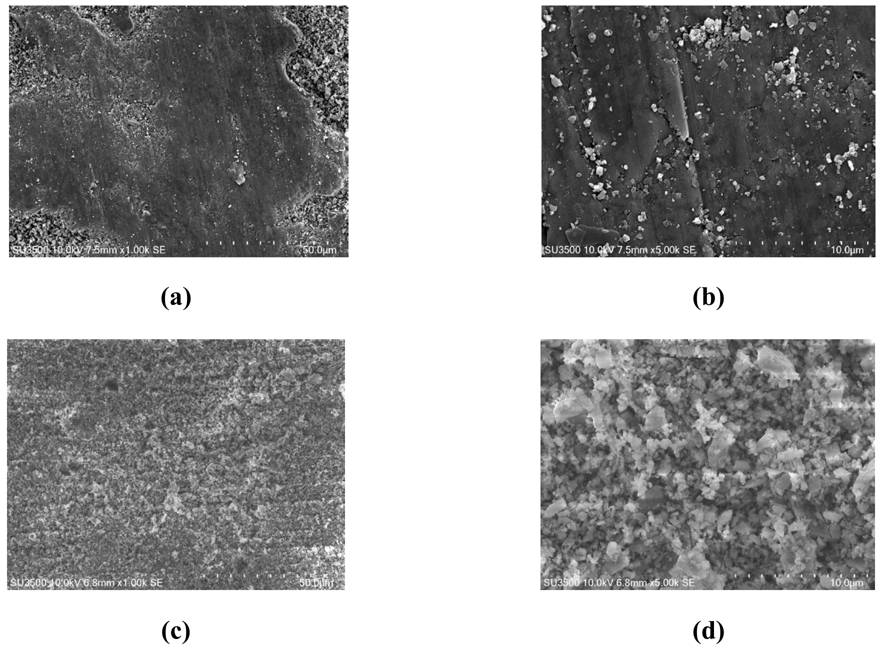

Figure 1. Comparison of the microstructures of specimens fired at 1050ºC (a) formula A1 (1000x), (b) formula A1 (5000x), (c) formula A6 (1,000x), (d) formula A6 (5000x)

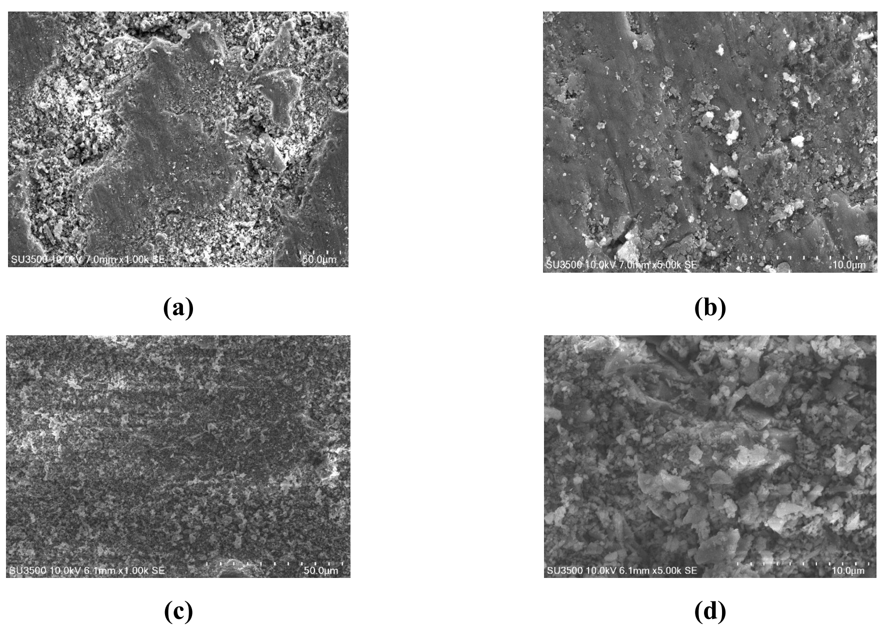

Figure 2. Comparison of the microstructures of specimens fired at 1050ºC (a) formula B1 (1000x), (b) formula B1 (5000x), (c) formula B5 (1,000x), (d) formula B5 (5000x)

3.5 Comparing physical properties of fired specimens with TIS 2508-2555

The bending strength and water absorption of specimens were evaluated and compared with the Thai Industrial Standard TIS (2508-2555) type BIII. The requirements for bending strength and water absorption of this standard are shown in Table 6. Formulas A1, A2, and B1, fired at 1050ºC were able to pass the TIS 2508-2555 standard.

Table 6. Properties of formulas meeting the Thai Industrial Standard 2508-2555

Formula no.	Mixture formulation (%)			Bending strength (MPa) Required >15 MPa	Water absorption (%) Required (10% < E < 20%)
	RHA	CGC	RNC		
A1	10	60	30	24.1	12.4
A2	10	50	40	19.2	13.4
B1	20	50	30	18	17

3.6 Texture and color of fired specimens of the proposed formulas

The proposed formulas, A1, A2, and A3, contributed the greyish white color in fired specimens. This is the influence of high Al_2O_3 content in RNC which promoted the white texture of test pieces. In addition, the effect of black color of RHA material contributed the greyish color of specimens. However, the effect of high content of CGC led to the darker texture of formula A1 than that of formulas A2 and B1, as shown in Figures 3(a), 3(b), and 3(c).

(a) **(b)** **(c)**

Figure 3. Color and texture of specimens fired at 1050ºC that met the TIS 2508-2555
(a) Formula A1, (b) Formula A2, (c) Formula B1

4. Conclusions

The results of this study which involved valorizing industrial wastes in order to produce ecological wall tiles can be summarized as follows.

1) The optimal firing temperature is 1050 ºC. It improves the bending strength of ceramic bodies to achieve the criteria of the Thai Industrial Standard 2508-2555 type BIII. Formulas A1, A2, and B1 fired at 1050 ºC meet this standard.

2) Valorizing RHA is possible for developing ecological wall tiles. Although utilizing 20% RHA in group B, which was fired at 950ºC and 1050ºC, had the effect of decreasing the bending strength and increasing the water absorption of ceramic bodies. The formula B1, containing

20 wt% RHA, 50 wt% CGC, and 30 wt%, has been suggested because it incorporates RHA waste at 20% which is higher than that of formulas A1 and A2.

3) Utilizing CGC provides a fluxing agent that can enhance the bending strength and reduce porosity of ceramic bodies. The 50 wt% of CGC is suggested as it can provide a high bending strength and decreased water absorption of ceramic tiles, characteristics that meet the Thai Industrial Standard.

4) The effect of higher firing temperature (1050ºC) is better development of glassy phase, which further contributes to the high bending strength of specimens. This was confirmed by SEM analysis.

An important contribution of this study is to show that waste materials from the industrial sector can be used as alternative materials for replacing commercial minerals. White-gray color wall tiles can be produced using RHA, CGC, and RNC. For a further study, it is recommended that local waste clays, for example, sediment soils from the water treatment process of industries, be tested in the production of ceramic tiles.

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