



Research article

Investigations of the influence of various industrial waste materials containing rice husk ash, waste glass, and sediment soil for eco-friendly production of non-fired tiles

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Abstract: Attempting to reduce the energy consumption of various manufacturing processes is one of the alternative ways to conserve energy. Furthermore, an expanding industry has resulted in a large amount of solid wastes generation. Exploiting wastes as alternative materials for saving energy to produce valuable products is a great challenge. Therefore, this study aims to utilize waste materials, e.g., rice husk ash, brown glass cullet, and sediment soil for producing non-fired tiles. Twenty-four formulas were conducted and divided into three groups (A, B, and C). Each formula mixture was uniaxially pressed at 10 MPa and cured for 7 and 28 days. This research work examined the use of brown glass cullet as a replacement for river sand in group A. The best formula of group A with the highest modulus of rupture had been selected for further research in group B. After that, formulas of group B were replaced laterite soil with sediment soil and local clay, which the best formula of group B was also taken by adding 5% and 10% of rice husk ash in group C. The result showed that the best formula was B1 containing 15% replacement of river sand with brown glass cullet and 5% replacement of laterite soil with local clay at 28 days of curing. Formula B1 has the modulus of rupture of 15.95 MPa and water absorption of 12.87% that can meet the Thai Industrial Standard 2508–2555 type BIII of wall tiles. Its energy consumption cost in the non-firing process could reduce 5.04 USD/m² when compared with that of the fired tile at 950 °C. Besides, an extensive experimental program was carried out including scanning electron microscopy (SEM), X-ray diffraction (XRD), and colorimetric CIELAB method. It can be summarized that developing non-fired tiles by utilizing waste materials is feasible to reduce energy consumption and waste disposal costs.

Keywords: industrial waste materials; rice husk ash; brown glass cullet; sediment soil; local clay; eco-friendly production; energy consumption; non-firing process; non-fired tiles

1. Introduction

The expansion of many industries has resulted in significantly increasing the volume of wastes, which will be increased to 19 billion tons in 2025 [1]. It is the responsibility of entrepreneurs for conserving the environment and paying the disposal costs. Therefore, utilizing industrial wastes should be investigated for value-added products. Wastes from the agricultural industry are also recognized. In Thailand, rice cultivation had a significant portion with ranked 6th of production after China, India, Indonesia, Bangalore, and Vietnam [2]. Rice husk (RH) is a by-product of the rice milling industry. RH can be used as boiler fuel in biomass power plants. After burning process, about 17–25% of rice husk ash (RHA) were generated [3]. RHA also contains about 95% of silica (SiO_2) as a suitable silica resource to be used in the ceramic industries [4]. The utilization of RHA has great applicability as an alternative material for many industries, e.g., ceramics, construction, and chemicals. However, the firing process of ceramic products, it consumes significant amounts of fossil fuels and also leads to high production costs [5]. Hence, reducing energy consumption is one of the interesting issues.

Considering the glass sector, glass containers are used as one-way or non-returnable packaging. As a high consumption of glass packaging, it has been manufactured from industries for responding to consumers' needs. However, after the glass bottles are used for containing beverages. After the end of the used period, glass bottles are discarded. From the Thailand Pollution Control Department (PCD) reported, approximately 2.5 million tons of waste glass was generated and about 25% of waste glass was unused [6]. Therefore, utilizing waste glass as an alternative material to develop valuable products is a challenging work. Moreover, the main chemical compositions of glass cullet are similar to ceramic materials.

For the water treatment sector, supplying water to people in the metropolitan area is the responsibility of the Metropolitan Waterworks Authority (MWA). As the population growth, it leads to an increase in the production of MWA to serve an increasing demand. The water is treated at four water treatment plants; Bang Khen, Sam Sen, Thonburi, and Maha Sawat with the capacities 4, 0.55, 0.12, and 1.6 million cubic meters/day, respectively. After treatment process, a large amount of sediment soil is generated approximately 200–250 tons/day from all water treatment plants [7]. It is considered as by-products or wastes from the treatment process. This leads to problems of the MWA for disposing and managing this waste. Furthermore, its chemical composition is similar to ceramic materials and can be employed as alternative materials in this study [8].

2. Literature review

From the previous studies, there are many researchers interested in utilizing wastes; rice husk ash, waste glass, and sediment soil for producing ceramic products.

Many studies were attempting to determine the firing temperatures above 1000 °C. Chiang et al. concluded that lightweight bricks containing 15% RHA indicated higher compressive strength, fired at 1100 °C [9]. Tonayopas et al. suggested that utilizing 30% RHA with clayey soil fired at 1050 °C

can pass the Thai Industrial Standard (TIS) for producing lightweight clay bricks [10]. Sultana et al. discussed that the properties of red clay with 15% RHA and 15% fly ash can produce a wide range of low-cost ceramic products after firing at 1050 °C [11]. Monteiro et al. evaluated that the use of 10% waste sludge in red ceramics led to a slight increase in the water absorption, resulting in decreased flexural rupture strength of samples after firing at 1100 °C [12]. Teixeira et al. indicated that the addition of 10% sludge into ceramic bodies fired at 1000 °C can achieve the Brazilian tile standards [13]. Gomes et al. concluded that the addition of up to 10% sludge sintered above 1000 °C to produce ceramic bricks, leading to increased compressive strength due to high iron-based sludge (Fe content) [14]. Costa et al. studied the effects of glass cullet for producing roof clay tile fired at 1100–1200 °C. It was concluded that utilizing 10% glass cullet can improve the flexural strength of specimens [15]. Maschio et al. indicated for use with 60% paper sludge and 40% glass cullet by the addition of 30% red clay. When a fast-firing temperature above 1120 °C, it can meet the Italian requirement for industrial red stoneware production [16]. Furlani et al. investigated that consisting of paper mill sludge and glass cullet in a ratio of 60/40 mixed with red clay, yellow clay, and kaolin. The results showed that the best properties were to mix red or yellow clay fired at a temperature above 1080 °C [17].

In addition, attempting to reduce the sintering temperatures below 1000 °C has been studied by many researchers. Andreola et al. showed that utilizing RHA up to 5% had an effect on lightweight bricks after firing at 960 °C. It leads to increase pore-forming and reduces the strength of brick bodies [18]. Wangrakdiskul and Pirunjareunporn found that the use of 80% brown glass cullet, 10% RHA, and 10% local plastic clay had been expanded the ceramic bodies for firing at 950 °C, resulting in increased porosity and decreased bending strength of fired clay wall tiles [19]. Wangrakdiskul et al. summarized that the formula contained 60% green glass cullet, 30% local white clay, and 10% RHA that can pass the Thai Industrial Standard 2508–2555 of ceramic tiles fired at 950 °C. It had developed the needle-shaped mullite for promoting the strength of ceramic bodies due to the effect of fluxing agents of waste glass [20]. Kaewtabut et al. summarized that using 20% RHA and 20% waste sludge from cutting glass in Anghong pottery commercial product fired at 900 °C had increased water absorption, but no effect on the mechanical strength of the product [21]. Although attempting to reduce the firing temperature of ceramic products from many studies, it still consumes fossil fuel for firing ceramic bodies.

For conserving energy consumption, non-fired clay tiles have been investigated. Developing by using spent bleaching earth from the palm oil industry was proposed for producing non-fired wall tiles by Wangrakdiskul et al. The results showed that 3.435% replacement of laterite soil with spent bleaching earth can promote bending strength with 30 days of the curing period. However, the property of water absorption was not improved [22]. Wangrakdiskul and Neumrat investigated the effects of replacement of laterite soil and fluvial sand with sediment soil during aging for 60 days. The results revealed that non-fired wall tiles can improve bending strength and reduce porosity with the influence of the hydration process among raw materials and Portland cement [8].

According to many of the researchers, it can be concluded that three waste materials, e.g., rice husk ash, brown glass cullet, and sediment soil have not been combined to produce non-fired tiles, leading to an investigation in this study. The basic formula consisted of ordinary Portland cement, laterite soil, and river sand, as derived from the previous study [8]. It has been used in this works to mix waste materials, e.g., brown glass cullet, rice husk ash, and sediment soil for investigating the alternative waste materials of non-fired tiles. The physical properties are investigated regarding;

modulus of rupture (MOR), water absorption, linear shrinkage, bulk density, X-ray fluorescence spectroscopy (XRF), scanning electron microscopy (SEM), and X-ray diffraction (XRD). The colorimetric analysis has investigated the colors of samples by using the CIELAB colorimetry technique. In addition, non-consuming of fossil fuel for producing non-fired ceramics is proposed. Therefore, the energy consumption cost of non-fired tile is calculated to compare with that of the fired tile at 950 °C.

3. Materials and methods

3.1. Materials

All raw materials used are available in Thailand. The chemical analysis of the raw materials was determined by X-ray fluorescence analysis (XRF), as shown in Table 1. Rice husk ash (RHA) was obtained from a biomass power plant. Using finely ground RHA had played a role as a binder to promote the strength of cement due to its high pozzolanic activity and high silica content [23]. Ordinary Portland cement (OPC) type I was used to act as a binder for all constituent materials. Laterite soil (LS) with OPC stabilization to be used as base material. Local clay (LC) was taken from the Angthong brick factory for promoting plasticity and facilitating molding of specimens [24]. Sediment soil (SS) was taken from the Metropolitan Waterworks Authority. It contains silica-alumina ($\text{SiO}_2\text{-Al}_2\text{O}_3$) which is similar to the composition of laterite soil. River sand (RS) were used to reduce the risk of the appearance of cracks and a stabilizer to control the strength of ceramic bodies [20,22]. Brown glass cullet (BGC) was received from a recycling factory. It was also to act as a pozzolan material for an alternative aggregate of non-fired tiles.

Table 1. Chemical composition of raw materials used by XRF analysis.

Chemical composition (%)	Raw materials used (%)						
	RHA	OPC	LS	LC	SS	RS	BGC
SiO ₂	92.67	18.13	71.00	64.95	58.15	78.05	72.10
K ₂ O	1.96	0.77	1.16	2.46	2.76	6.35	0.20
CaO	1.18	66.63	0.24	0.59	0.94	1.35	10.60
P ₂ O ₅	1.50	0.08	0.09	0.17	0.23	-	-
MgO	0.78	1.11	0.43	1.43	1.37	0.44	2.40
Fe ₂ O ₃	0.54	3.36	4.45	6.45	7.17	1.44	0.10
Al ₂ O ₃	0.56	4.6	21.79	21.95	27.84	10.96	1.60
Cl	0.16	-	-	0.02	-	-	-
SO ₃	0.34	4.65	0.03	0.23	0.15	0.03	0.15
MnO	0.13	-	0.01	0.13	0.19	0.02	-
Na ₂ O	0.07	0.22	0.09	0.41	0.14	0.95	12.80
TiO ₂	0.03	0.24	0.64	0.99	0.86	0.23	0.04
BaO	-	-	0.02	0.12	0.06	0.09	-

Under the requirements of ASTM C 618 class N [25], the specified chemical requirements of waste materials, e.g., RHA, BGC, and SS can be categorized as natural pozzolan materials in this study, as shown in Table 2. They have potential for acceptable function as a partial cement replacement [26].

Table 2. Chemical requirements of waste materials under the requirements of ASTM C 618 class N.

Chemical compositions	Chemical requirements type of pozzolan; class N (%)	Waste materials used (%)		
		RHA	BGC	SS
SiO ₂ + Al ₂ O ₃ + Fe ₂ O ₃	>70.0	93.77	73.8	93.16
Sulfur trioxide (SO ₃)	<4.0	0.34	0.15	0.15

3.2. Methods

All materials were dried at 200 °C for 2 h except for OPC. Then, they were pulverized in a ball mill. Material powders were then sieved by 20 mesh (841 μm). The mixtures were conducted for twenty-four formulas which the basic formula was obtained from Wangrakdiskul and Neamlut [8]. Mixture proportions of the experiment are shown in Table 3. Firstly, the basic formula was name-calling of the formula A1 of group A. After that, RS was replaced with BGC by weight at 5%, 10%, 15% of formulas A2, A3, and A4, respectively. Secondly, the best formula of group A has been used as the basic formula of group B with the replacement of LS by LC and SS. Finally, the best formula of group B is used as the basic formula of group C for adding 5% and 10% RHA. Each formula mixture was uniaxially pressed at 10 MPa in a rectangular mold (50 × 100 × 7 mm). After forming, all specimens were sprayed with water and covered with a plastic sheet for curing at room temperature at 7 and 28 days. Finally, all specimens were dried at 200 °C for 2 h and were cooled down at room temperature.

Table 3. Mixture proportions of the experiment.

No.	Compositions (%)						
	RHA	OPC	LS	LC	SS	RS	BGC
A1	0	22.5	62.5	0	0	15	0
A2	0	22.5	62.5	0	0	10	5
A3	0	22.5	62.5	0	0	5	10
A4	0	22.5	62.5	0	0	0	15
B1	0	22.5	57.5	5	0	The best	The best
B2	0	22.5	57.5	0	5	ratio of RS	ratio of
B3	0	22.5	52.5	5	5	of group A	BGC of
B4	0	22.5	47.5	10	5	(0–15%)	group A
B5	0	22.5	47.5	5	10		(0–15%)
B6	0	22.5	42.5	10	10		
B7	0	22.5	37.5	15	10		
B8	0	22.5	37.5	10	15		
B9	0	22.5	32.5	15	15		
B10	0	22.5	27.5	20	15		
B11	0	22.5	27.5	15	20		
B12	0	22.5	22.5	20	20		
B13	0	22.5	17.5	25	20		
B14	0	22.5	17.5	20	25		

Continued on next page

No.	Compositions (%)						
	RHA	OPC	LS	LC	SS	RS	BGC
B15	0	22.5	12.5	25	25	The best	The best
B16	0	22.5	7.5	30	25	ratio of RS	ratio of
B17	0	22.5	7.5	25	30	of group A	BGC of
B18	0	22.5	2.5	30	30	(0–15%)	group A
C1	5	22.5	The best	The best	The best		(0–15%)
C2	10	22.5	ratio of LS of group B (2.5–57.5%)	ratio of LC of group B	ratio of SS of group B (0–30%)		

In addition, three-point bending tests were performed for the determination of modulus of rupture (MOR). Water absorption was measured the difference of dry mass and wet mass by immersing in boiling water for 2 h, remain immersed in water to cool naturally for 4 h. The excess moisture on the sample surfaces was removed with chamois leather and wet mass was measured. Modulus of rupture and water absorption were tested according to the conditions established by the Thai Industrial Standard 2508–2555. The linear shrinkage was obtained by measuring the length of the samples before and after the curing process. Bulk density was also determined based on Archimedes' method using standard densimeter (Alfa Mirage, EW-300SG). Therefore, physical properties of specimens were examined regarding; modulus of rupture, water absorption, linear shrinkage, and bulk density. Fifteen samples of each formula were tested for the physical properties and calculated for the average values and standard deviation. Furthermore, X-ray fluorescence (XRF) technique was carried out to determine the chemical compositions of powder materials by using wavelength-dispersive X-ray fluorescence spectrometry; Model: S8 TIGER, Bruker, Germany. Besides, the microstructure of four samples was selected to investigate for morphologically surface evaluation by scanning electron microscope (SEM Hitachi, SU3500) at voltage set on 10 kV for 5000× magnification. Moreover, X-ray diffraction pattern (XRD) of the powder samples was carried out by using Bruker D8 advance diffractometer with Cu K α radiation. A scan rate of 0.01°·s⁻¹ was determined to observe the pattern in the 2 θ of 5–80°. Commission Internationale l'Eclairage (CIE) Lab, a three-dimensional (L*a*b*) color space method was used to measure the color of specimens. It was carried out on the UV-vis-NIR spectrophotometer using a Shimadzu UV-3600plus and MPC-603, in the 300–700 nm range.

4. Results and discussion

4.1. Physical properties

The results of mean and standard deviation (SD) of the experiment are presented in three groups (A, B, and C), as shown in Table 4. The standard deviation of all physical properties is less than 10% of the sample means at 7 and 28 days of curing. For easier interpretation, these results will be described as follows.

Table 4. Mean and standard deviation (SD) of physical properties of group A, B, C at 7 and 28 days of curing.

No.	Compositions (%)							Modulus of rupture (MPa)		Water absorption (%)		Linear shrinkage (%)		Bulk density (g/cm ³)	
	RHA	OPC	LS	LC	SS	RS	BGC	7 days	28 days	7 days	28 days	7 days	28 days	7 days	28 days
A1	0	22.5	62.5	0	0	15	0	5.36 ± 0.11	8.30 ± 0.16	10.51 ± 0.34	8.92 ± 0.41	0.11 ± 0.01	0.14 ± 0.01	2.52 ± 0.01	2.46 ± 0.02
A2	0	22.5	62.5	0	0	10	5	6.02 ± 0.38	9.50 ± 0.42	11.82 ± 0.23	11.32 ± 0.2	0.09 ± 0	0.19 ± 0.01	2.51 ± 0.03	2.40 ± 0.01
A3	0	22.5	62.5	0	0	5	10	6.14 ± 0.32	10.22 ± 0.33	13.44 ± 0.34	12.34 ± 0.15	0.09 ± 0	0.24 ± 0.01	2.43 ± 0.03	2.38 ± 0.02
A4*	0	22.5	62.5	0	0	0	15	7.12 ± 0.36	11.58 ± 0.52	14.69 ± 0.2	12.85 ± 0.22	0.09 ± 0	0.33 ± 0.02	2.41 ± 0.02	2.34 ± 0.03
B1**	0	22.5	57.5	5	0	0	15	8.55 ± 0.32	15.95 ± 0.49	14.61 ± 0.26	12.87 ± 0.18	0.38 ± 0.03	0.71 ± 0.04	2.52 ± 0.01	2.25 ± 0.01
B2	0	22.5	57.5	0	5	0	15	5.68 ± 0.54	12.79 ± 0.46	15.49 ± 0.31	15.15 ± 0.28	0.38 ± 0.02	0.65 ± 0.04	2.38 ± 0.03	2.19 ± 0.02
B3	0	22.5	52.5	5	5	0	15	5.54 ± 0.28	12.21 ± 0.53	16.42 ± 0.39	15.67 ± 0.21	0.36 ± 0.02	0.62 ± 0.05	2.35 ± 0.03	2.18 ± 0.03
B4	0	22.5	47.5	10	5	0	15	5.92 ± 0.35	13.27 ± 0.63	15.84 ± 0.44	15.37 ± 0.25	0.38 ± 0.02	0.65 ± 0.03	2.30 ± 0.04	2.10 ± 0.03
B5	0	22.5	47.5	5	10	0	15	3.37 ± 0.23	11.33 ± 0.24	17.28 ± 0.19	16.82 ± 0.3	0.34 ± 0.02	0.60 ± 0.03	2.21 ± 0.03	2.08 ± 0.05
B6	0	22.5	42.5	10	10	0	15	2.90 ± 0.18	8.68 ± 0.26	18.05 ± 0.12	17.64 ± 0.18	0.33 ± 0.03	0.52 ± 0.03	2.18 ± 0.05	2.05 ± 0.05
B7	0	22.5	37.5	15	10	0	15	2.02 ± 0.08	8.03 ± 0.23	18.45 ± 0.22	18.33 ± 0.35	0.31 ± 0.03	0.41 ± 0.03	2.18 ± 0.04	2.05 ± 0.03
B8	0	22.5	37.5	10	15	0	15	1.25 ± 0.07	7.11 ± 0.52	19.45 ± 0.14	19.37 ± 0.13	0.30 ± 0.02	0.40 ± 0.04	2.20 ± 0.04	2.04 ± 0.04
B9	0	22.5	32.5	15	15	0	15	1.22 ± 0.09	4.37 ± 0.37	19.25 ± 0.21	18.63 ± 0.29	0.28 ± 0.02	0.38 ± 0.02	2.22 ± 0.02	2.06 ± 0.04
B10	0	22.5	27.5	20	15	0	15	1.13 ± 0.08	2.89 ± 0.26	19.85 ± 0.27	19.50 ± 0.11	0.26 ± 0.01	0.36 ± 0.03	2.22 ± 0.03	2.10 ± 0.02
B11	0	22.5	27.5	15	20	0	15	1.05 ± 0.08	2.37 ± 0.19	20.83 ± 0.44	20.26 ± 0.28	0.24 ± 0.02	0.35 ± 0.02	2.24 ± 0.02	2.18 ± 0.03
B12	0	22.5	22.5	20	20	0	15	1.02 ± 0.05	2.40 ± 0.22	20.98 ± 0.25	20.90 ± 0.15	0.18 ± 0.01	0.32 ± 0.02	2.39 ± 0.01	2.22 ± 0.04
B13	0	22.5	17.5	25	20	0	15	0.48 ± 0.03	2.26 ± 0.18	20.76 ± 0.24	20.56 ± 0.21	0.14 ± 0.01	0.31 ± 0.02	2.34 ± 0.02	2.20 ± 0.02
B14	0	22.5	17.5	20	25	0	15	0.36 ± 0.02	1.48 ± 0.11	21.65 ± 0.19	21.35 ± 0.12	0.12 ± 0.01	0.24 ± 0.02	2.29 ± 0.04	2.19 ± 0.03
B15	0	22.5	12.5	25	25	0	15	0.23 ± 0.01	1.46 ± 0.12	21.97 ± 0.25	21.79 ± 0.24	0.11 ± 0.01	0.19 ± 0.01	2.29 ± 0.03	2.17 ± 0.04
B16	0	22.5	7.5	30	25	0	15	0.20 ± 0.01	1.26 ± 0.12	22.37 ± 0.17	21.94 ± 0.26	0.10 ± 0.01	0.16 ± 0.01	2.26 ± 0.03	2.14 ± 0.02
B17	0	22.5	7.5	25	30	0	15	0.12 ± 0.01	0.64 ± 0.03	23.88 ± 0.17	23.41 ± 0.19	0.09 ± 0	0.15 ± 0.01	2.27 ± 0.02	2.17 ± 0.05
B18	0	22.5	2.5	30	30	0	15	0.00 ± 0	0.33 ± 0.03	23.48 ± 0.17	23.25 ± 0.22	0.07 ± 0	0.12 ± 0.01	2.28 ± 0.03	2.19 ± 0.04
C1	5	22.5	57.5	5	0	0	15	10.36 ± 0.33	13.06 ± 0.52	16.53 ± 0.24	16.64 ± 0.09	0.05 ± 0	0.06 ± 0	2.30 ± 0.05	2.24 ± 0.02
C2	10	22.5	57.5	5	0	0	15	10.23 ± 0.39	12.04 ± 0.7	17.14 ± 0.16	17.44 ± 0.24	0.18 ± 0	0.25 ± 0.01	2.26 ± 0.04	2.22 ± 0.03

*Note: Formula A4 is the best formula of group A and the basic formula in group B for the replacement of LS by LC and SS. Formula B1 is the best formula of group B and the basic formula in group C for adding RHA.

4.1.1. Group A with the replacement of RS by BGC

The aim of group A is to utilize BGC effectively as a replacement for RS of non-fired tiles. The basic formula containing 22.5% OPC, 62.5% LS, and 15% RS was obtained from Wangrakdiskul and Neamlut [8]. The results show the replacement of RS with BGC by weight at 5%, 10%, and 15% at 7 and 28 days of curing, as shown in Table 4. The effect of increasing the BGC ratio can enhance MOR of specimens. Hence, formula A4 utilizing 15% BGC as a replacement for RS is the best formula of group A at 7 and 28 days of curing due to the highest MOR value. This is the influence of the high pozzolanic reaction of BGC content of the curing process, which is a similar result to Younes et al. [27]. It has been increased water absorption when a high replacing ratio of RS by BGC at 7 and 28 days of curing. Note that, a longer curing period at 28 days of curing can increase MOR and reduce water absorption of specimens as compared with 7 days of curing. Linear shrinkage of all formulas is slightly different from 0.09–0.11% and 0.14–0.33% at 7 and 28 days of curing, respectively. It is summarized that linear shrinkage is lower than 1% due to the non-fired process. Moreover, the bulk density is within the range of 2.34 to 2.52 g/cm³ at 7 and 28 days of curing. The experimental results clearly show the low bulk density of BGC content, leading to a decrease in mass per unit volume. This is a similar result to Habeeb and Mahmud [28].

4.1.2. Group B with the replacement of LS by SS and LC

The purpose of group B is to investigate the combination of LC (local clay from Angthong Province) and SS (wastes from the water supply treatment process) as a partial replacement for the LS of non-fired tiles. After obtaining the best formula of group A, the formula A4 has been selected as the basic formula in group B for further research in group B with the replacement of LS by SS and LC, as expressed in Table 4. For the 7 days curing period, the effect of replacement of LS with LC and SS has decreased MOR of all specimens as compared with formula A4, except for formula B1. From the results obtained at 28 days of curing period, the replacement of 5% LS with LC of formula B1 is the best formula of group B. It has 15.95 MPa of MOR and 12.87% of water absorption. This is due to a highly dense structure of non-fired ceramic bodies, which is confirmed by SEM analysis. Moreover, it can achieve the Thai Industrial Standard of ceramic tiles, TIS 2508–2555 in terms of MOR and water absorption. In addition, the effect of MOR on the 28 days curing period of formula B2 by replacing 5% LS with SS can be increased more than formula A4. However, MOR and water absorption values of formula B2 to B18 cannot pass the TIS 2508–2555 type BIII. Note that, the replacement higher than 5% of SS has contributed to an adverse effect of the pozzolanic activity resulting in decreased MOR of specimens. This corroborates from Yagüe et al. [29]. In addition, linear shrinkage is within the range of 0.07–0.71%. It leads to reduce the risk appearance of cracks of specimens. A longer curing period can slightly increase linear shrinkage and can reduce the porosity and water absorption to improve the strength and dense structure of non-fired tiles. The bulk density values of all formulas are slightly different from 2.04–2.52 g/cm³ at 7 and 28 days of curing time.

4.1.3. Group C by adding 5% and 10% RHA content

The aim of group C is to examine the effect of adding RHA in non-fired tiles. The best formula of group B is formula B1 as the basic formula in group C that has been drawn to add 5% and 10% RHA, as shown in Table 4. The formulas of C1 and C2 at 7 days of curing found a high MOR than formulas A4 and B1 due to the pozzolanic activity demonstrate high pozzolanic activity of produced rice husk ash with a short period of curing. A long curing period at 28 days, the formulas C1 and C2 are higher MOR than formula A4. However, the MOR values of these formulas are less than formula B1 (without RHA addition) due to the influence of RHA content increases. This is in agreement with the theory on the slow evolution of the pozzolanic action with longer ageing [30]. However, water absorption of formulas C1 and C2 can pass TIS 2508–2555 type BIII, but MOR cannot achieve the TIS 2508–2555. Both formulas obtain low linear shrinkage that are slightly different from 0.05–0.25% at 7 and 28 days of curing time. Likewise, the bulk density is no significant difference from 2.22–2.30 g/cm³. Note that, low water absorption has been related to a low porosity of specimens. For increasing MOR value of 5% RHA addition, extending the curing period of formula C1 should be investigated in future work.

4.2. Comparison of non-fired wall tiles with the Thai Industrial Standard 2508–2555

Four formulas have been compared with the Thai Industrial Standard of ceramic tiles, TIS 2508–2555 type BIII in terms of modulus of rupture (MOR > 15 MPa) and water absorption (10% < E ≤ 20%) [31]. The best formula of each group which formulas A4, B1, and C1 had been compared with the formula A1 as the basic formula, as shown in Table 5. For water absorption property, all formulas can achieve TIS 2508–2555 type BIII. However, formula A1 can classify water absorption requirement (6% < E ≤ 10%) type BII_b with an average value of 8.92%, but it cannot pass the MOR requirement type BII_b. Therefore, it can be classified in the water absorption type BIII with TIS 2508–2555 as well. Moreover, formulas A4, B1, and C1 are higher than MOR that of formula A1. It can be concluded the formula B1 that can pass only one formula in terms of MOR and water absorption with TIS 2508–2555 type BIII to define the type to wall tile product. It was claimed that the non-fired tile with the eco-friendly concept has been successfully developed.

Table 5. Comparison physical properties of non-fired wall tiles with the Thai Industrial Standard 2508–2555 type BIII at 28 days of curing.

No.	Compositions (%)							Modulus of rupture		Water absorption		Linear shrinkage (%)	Bulk density (g/cm ³)
	RHA	OPC	LS	LC	SS	RS	BGC	Require	(MPa)	Require	(%)		
A1	0	22.5	62.5	0	0	15	0	×	8.30	✓	8.92	0.14	2.46
A4	0	22.5	62.5	0	0	0	15	×	11.58	✓	12.85	0.33	2.34
B1	0	22.5	57.5	5	0	0	15	✓	15.95	✓	12.87	0.71	2.25
C1	5	22.5	57.5	5	0	0	15	×	13.06	✓	16.64	0.06	2.24

4.3. CIELAB colorimetric coordinates

Instrumental color measurement with CIELAB has the advantage of obviating the subjective

aspects of color assessment. The development of the CIELAB colorimetric system is an accepted method for differential color measurement [32]. The instruments measure color and express in terms of three coordinate values (L^* , a^* , and b^*), which locate the object in a three-dimensional color space. The L^* coordinate represents the brightness of an object ($L^* = 0$ indicates black and $L^* = 100$ indicates diffuse white), the a^* value represents the red or green color (negative values indicate green while positive values indicate red), and the b^* value represents the yellow or blue color (negative values indicate blue and positive values indicate yellow) [33]. In this study, two formulas compare and classify colors with CIELAB colorimetric, e.g., formula A1 as basic formula and formula B1 as formula can pass the Thai Industrial Standard 2508–2555. The color results of samples as illustrated in Table 6. They can be analyzed as follows, L^* (41.3–48.18), a^* (11.69–13.43), and b^* (11.09–15.24), as shown in Figure 1a. The visual evaluation of the color of the samples showed a similar color. However, the brightness of formula B1 has slightly increased than formula A1 due to the effect of BGC and LC increases. The formula A1 has indicated higher red and yellow tones than formula B1. Textures of non-fired tiles are also shown in Figure 1b,c.

Table 6. CIELAB color of samples at 28 days of curing.

No.	Compositions (%)							CIELAB		
	RHA	OPC	LS	LC	SS	RS	BGC	L^*	a^*	b^*
A1	0	22.5	62.5	0	0	15	0	41.3	13.43	15.24
B1	0	22.5	57.5	5	0	0	15	48.18	11.69	11.09

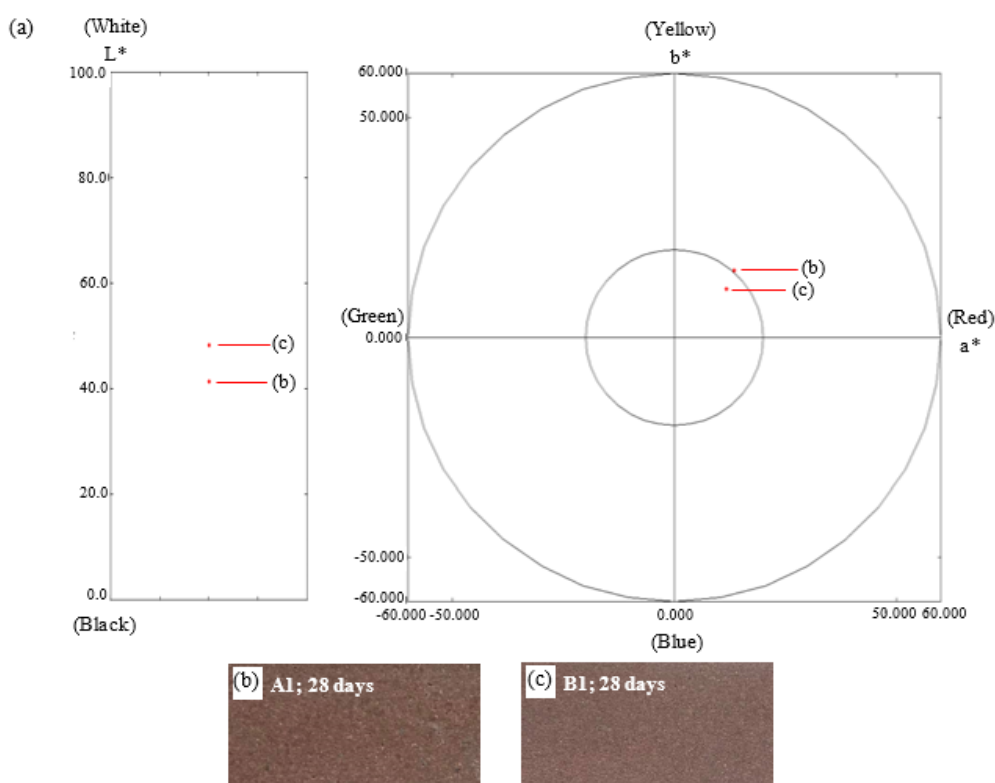


Figure 1. (a) CIELAB graphics of non-fired tiles. Textures of non-fired tiles at 28 days of curing (b) formula A1 and (c) formula B1.

4.4. Scanning electron microscopy (SEM) analyzing

Scanning electron microscopy (SEM) has been conducted to investigate the microstructure of specimens with a focused beam of electrons [34]. After curing at 28 days, the highest and lowest MOR of group A (formulas A1 and A4) and B (formulas B1 and B18) are selected the samples to analyze the microstructure at 5000 \times magnification. The results have been depicted in Figure 2. A random distribution of pores structure is found in formula A1, leading to a low MOR of specimens (Figure 2a) [35]. Replacement of RS with 15% BGC of formula A4 has contributed to the pozzolanic reaction which can contribute to the MOR of samples (Figure 2b) [36]. Meanwhile, formula B1 with 5% LC as a replacement for LS and without SS content has been found kaolinite (hexagonal kaolinite plates) and calcite in the continuous matrix. These phases and the pozzolanic reaction of BGC content can promote a highly dense structure of non-fired ceramic bodies (Figure 2c) [37]. Finally, formula B18 containing a ratio of 30% LC and 30% SS has found illite content as flakes. This phenomenon leads to increase porosity and decrease MOR and increase water absorption (WA) of samples (Figure 2d) [38].

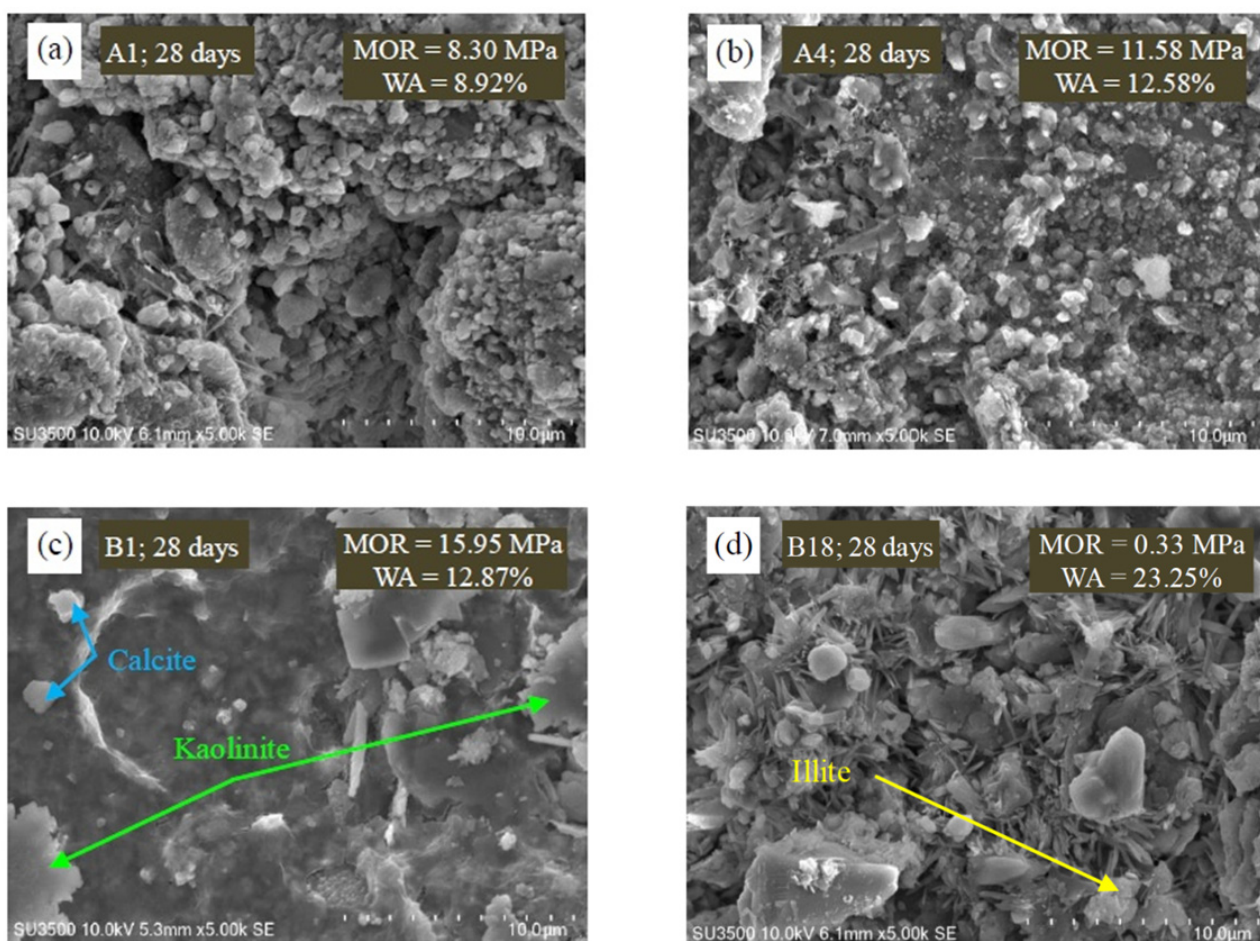


Figure 2. SEM images at 5000 \times magnification of samples at 28 days of curing (a) formula A1, (b) formula A4, (c) formula B1, and (d) formula B18.

4.5. X-ray diffraction (XRD) analyzing

The same formulas have been analyzed by SEM that are also taken to investigate crystalline phases by XRD pattern. Crystalline phases of all formulas are found in quartz (SiO_2 , ICDD 00-046-1045), kaolinite-1A ($\text{Al}_2(\text{Si}_2\text{O}_5)(\text{OH})_4$, ICDD 01-080-0885), calcite (CaCO_3 , ICDD 00-005-0586), calcium silicate (α' - Ca_2SiO_4 , ICDD 00-033-0303), calcium silicate (Ca_3SiO_5 , ICDD 00-049-0442), and albite low ($\text{Na}(\text{AlSi}_3\text{O}_8)$, ICDD 01-076-0899), except for formula A4 cannot be found albite low phase, as shown in Figure 3. Moreover, the illite-1M ($\text{KAl}_2(\text{Si}_3\text{AlO}_{10})(\text{OH})_2$, ICDD 00-002-0462) has been only found in formula B18, leading to a decrease in the MOR of specimens as shown in Figure 3d. Similarly, it leads to the lowest MOR and the highest water absorption due to the flake shape by confirming the result with SEM analysis (Figure 2d).

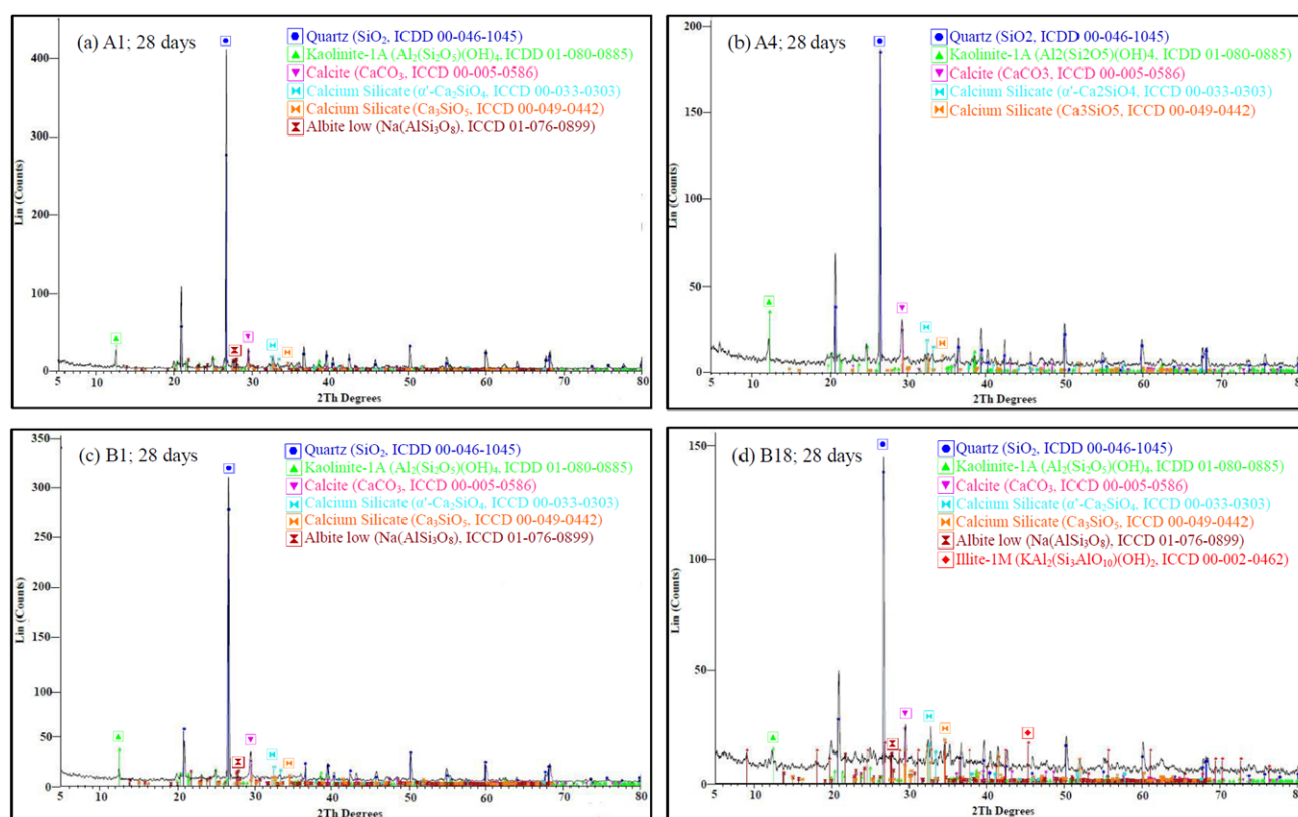


Figure 3. XRD pattern of power samples at 28 days of curing (a) formula A1, (b) formula A4, (c) formula B1, and (d) formula B18.

4.5.1. Comparison of the quantitative phase analysis with MOR and water absorption

The quantitative phase analysis of XRD data was performed by using the TOPAS program. The results are shown in Table 7. The best fit to the experimental pattern is obtained, the R_{wp} (R-weighted pattern) value is between 12.91–14.61, which means the misfits between the measured and computer model data [39]. In addition, the GOF (goodness of fit) value is between 1.19–1.42. This is sufficient for low misfits due to the vicinity of 1 value [33,40]. Therefore, the refinement of four formulas in this study is observed to be of acceptable quality.

Table 7 shows the comparison of phase with MOR and water absorption values, increasing of calcite and α' -Ca₂SiO₄ phases results in increased the MOR of samples. Hence, formula B1 has the highest calcite and α' -Ca₂SiO₄ contents, leading to the highest MOR as compared with formula A1, A4, and B18. On the contrary, the high Ca₃SiO₅ and illite-1M contents lead to low MOR of specimens, i.e., formula B18. Note that, the kaolinite-1A phase is no significant difference values. In addition, the quartz and albite low values are between 28.80–58.38% and 3.14–7.78%, respectively.

Table 7. Comparison phase content, Rietveld disagreement factors (R_{wp} and GOF), MOR, and water absorption.

No.	Phase content (%)							R_{wp}	GOF	MOR (MPa)	Water absorption (%)
	Quartz	Kaolinite- 1A	Calcite	α' -Ca ₂ SiO ₄	Ca ₃ SiO ₅	Albite low	Illite-1 M				
A1	58.38	20.56	6.76	2.69	8.48	3.14	-	14.61	1.19	8.30	8.92
A4	56.65	19.89	11.73	4.00	7.73	-	-	12.91	1.42	11.58	12.85
B1	48.68	19.64	14.02	4.82	6.03	6.81	-	13.19	1.19	15.95	12.87
B18	28.80	19.14	4.05	3.36	13.71	7.78	23.16	13.50	1.29	0.33	23.25

4.6. Comparison physical properties and energy costs of non-fired with fired wall tiles

Reducing energy consumption for producing wall tiles is one of the aims of this study. Comparing physical properties and energy consumption of non-fired tile (formula B1 at 28 days of curing) with fired tile (fired at 950 °C), as shown in Table 8. They can achieve the Thai Industrial Standard 2508–2555 type BIII in terms of MOR and water absorption. The power consumption of an electric kiln in a laboratory-scale of fired tile was 5 kW/h, usage time was 10.5 h from the maximum firing temperature for 950 °C, the heating rate of 100 °C /h, and soaking for 1 h [19]. Besides, the average price of electricity was 0.096 USD/kW refer to data from the Metropolitan Government in Thailand. It can be concluded that the non-fired tile from this study provides a higher MOR and lower water absorption than that of fired tile. The non-fired tile can also save the energy consumption cost of the firing process for 5.04 USD/m² as compared with the fired tile at 950 °C. Finally, the non-fired product can benefit for manufacturers to reduce the energy consumption of the firing process.

Table 8. Comparison of physical properties and energy costs with non-fired and fired tiles.

Types of tiles	Processes	MOR (MPa)	Water absorption (%)	Power consumption (kW/h)	Usage time (h)	Average price of electricity (USD/kW)	Energy cost of firing process (USD/m ²)
Non-fired tile	Cured 28 days	15.95	12.87	0	0	0.096	0
Fired tile [19]	Fried at 950 °C	15.18	16.17	5	10.5	0.096	5.04

*Note: energy cost (USD/m²) = power consumption (kW/h) × usage time (h) × average price of electricity for 0.096 (USD/kW) refer to data from the Metropolitan Government in Thailand.

5. Conclusions

The results of utilizing materials; brown glass cullet (BGC), sediment soil (SS), rice husk ash (RHA), and local clay (LC) for producing non-fired wall tiles are investigated in this study. The following conclusions could be drawn.

- (1) The influence of various industrial waste materials of three groups (A, B, and C) of this experiment will be described as follows.
 - (1.1) Group A, the results show that the effect of replacement of RS with up to 15% BGC can contribute to the strength due to the pozzolanic reaction of BGC content in non-fired ceramic bodies at 28 days of curing.
 - (1.2) Group B indicates the best formula B1 with the replacement of 5% LS with LC that has the modulus of rupture of 15.95 MPa and water absorption of 12.87% at 28 days of curing. It can achieve the Thai Industrial Standard 2508–2555 type BIII of non-fired wall tiles.
 - (1.3) Group C by adding 5% and 10% RHA of non-fired tiles at 28 days of curing, it has resulted in a decrease the MOR and increase water absorption due to RHA content increases. However, enhancing the MOR of specimens by utilizing RHA should be further investigated, which extending the curing period to 90 days is suggested in further works.
- (2) The microstructure by SEM at 28 days of curing, kaolinite and calcite are found to promote a highly dense structure due to the higher pozzolanic reaction of BGC. Besides, illite content as flakes leads to decrease the MOR due to the high porosity of samples. Similarly, the calcite phase can develop strength, but the illite-1M phase leads to decrease the MOR of samples by XRD analysis.
- (3) The longer curing period at 28 days of curing of non-fired ceramic bodies can improve the strength more than 7 days of curing.
- (4) Note that, the effect of high replacement of LS with SS has decreased the MOR of specimens.
- (5) Non-fired tile at 28 days of curing can also save the energy consumption cost of the firing process for 5.04 USD/m² as compared with fired tile (fired at 950 °C). Moreover, the physical properties of non-fired tile are similar to the fired tile.

As a result of the above, it can be summarized that eco-friendly non-fired tiles by utilizing waste materials can reduce energy consumption and waste disposal cost for entrepreneurs.

Acknowledgments

The authors wish to express their sincere gratitude by the Production Engineering Department, Faculty of Engineering, King Mongkut's University of Technology North Bangkok, Thailand.

Conflict of interest

All authors declare no conflicts of interest in this paper.

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