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Research article

Combination of rice husk ash, bagasse ash, and calcium carbonate for developing unglazed fired clay tile

Witsanu Loetchantharangkun* and Ubolrat Wangrakdiskul

Department of Production Engineering, Faculty of Engineering, King Mongkut's University of Technology North Bangkok, Thailand

* **Correspondence:** Email: s6001021810052@email.kmutnb.ac.th; Tel: +66902264388.

Abstract: This research aims at developing unglazed fired clay tiles by utilizing industrial wastes; rice husk ash (RHA), bagasse ash (BGA), calcium carbonate (CC), and fly ash (FA). Brown glass cullet (BGC) has been mixed with these waste materials for reducing firing temperature. In addition, local clay is also used for facilitating specimens' plasticity. Work pieces are molded by uniaxial pressing at 100 bars with dimensions 50 × 100 × 7 mm and fired at 850 and 950 ºC. Formulation mixtures of the experiment are divided into 4 groups. Calcium carbonate (CC), residue from sugar mill plant mixing with local materials, LBC and LWC (Local ball clay and local white clay) have been utilized in group 1. After testing the physical properties of fired specimens, a high bending strength of formula in group 1 has been selected. It is further employed as the basic formula of the next three groups by mixing RHA, BGA, and FA, respectively. The results found that the optimal ratios containing 5% RHA, 5–10% BGA, and 5% FA of group 2, 3, and 4 which fired at 950 ºC can achieve Thai Industrial Standard (TIS 2508-2555 type BIII) in terms of bending strength and water absorption. Clarifying the color of selected formulas is determined by CIELAB color coordinate. In addition, analyzing the microstructure of selected specimens by scanning electron microscopy (SEM) and X-ray diffraction (XRD) has been conducted. Glassy phase and wollastonite crystal are found in the specimens providing high bending strength.

Keywords: brown glass cullet; rice husk ash; bagasse ash; $CaCO_3$; fly ash; unglazed wall tiles

1. Introduction

Currently, industrial wastes are intensively increased as the growth of many industries. For the non-hazard waste materials, reutilizing them as alternative materials has been attempted by many researchers [1]. Manufacturing related on agricultural sectors in Thailand has been investigated in this study focusing on sugar, palm oil mill, biomass power plant, and including glass sector.

For highly cultivation the sugarcane crop, Thailand is ranked third for production accounting for 131 million tons in 2016 [2]. It is the main source of raw materials for sugar production. There are 58 sugar refineries plants in Thailand [3]. After the extracting process, 250–300 kg of bagasse is generated from one-ton of sugarcane. It is recycled as fuel to generate steam in factories [4]. After the boiling process, it is transformed to bagasse ash (BGA) which leads the problem to manufacturers for elimination. In addition, milk lime (CaO) has been used for purifying sugar. After this process, calcium carbonate ($CaCO_3$; CC) has been generated. Recycling this waste is also crucial and highly concerned.

For the other crop in Thailand, the oil palm plant is also ranked third for production capacity after Indonesia and Malaysia [2]. It is the main raw materials for production edible palm oil. There are a lot of crude palm oil factories in Thailand which are 173 plants [3]. However, palm oil processing has generated a huge number of wastes, such as empty fruit bunch, palm fiber, and palm shell. The empty fruit bunch and palm fiber have been recycled in the plants as the boiler fuel for steaming fresh fruit bunch. After the boiling process, fly ash (FA) of empty fruit bunch and palm fiber is generated which also leads the problem to manufacturers. This work intends to utilize fly ash (FA) as the alternative raw material.

As recognition of fossil fuel depletion, the biomass power plants have been setting up as Thai government supporting. There are biomass power plants for 218 plants in Thailand [5]. The present work focuses on the biomass power plant which utilizing rice husk as fuel for generating electricity. After the burning process, the rice husk has been transformed to rice husk ash (RHA). The manufacturer also faces the problem for disposing these residues. Reutilizing rice husk ash (RHA) as the alternative materials is also the objective of this study.

In 2017, the Pollution Control Department of Thailand reported that there were solid wastes such as glass cullet amounting to 2.5 million tons. They had been recycled for 73% [6]. However, 27% of wastes has not been recycled. It should be reutilized as a combination mixture for producing valued products. Brown glass cullet (BGC) exploited in this work is derived from the glass manufacturing in Thailand.

This paper aims to utilize all waste materials as mentioned above. Due to containing with high silica similarly to ceramic materials, they are intended to be utilized as alternative materials in ceramic manufacturing. However, the relevant studies have been investigated, which will be described as follows.

Recognition on wastes from sugarcane plants, there are many researchers investigating the effect of exploiting these residues on construction manufacturing. Saleem et al. studied about bagasse and rice husk ash as replacement materials in clay brick fired at 800 ºC. The results showed that adding wastes had reduced mechanical properties of clay brick. In addition, modulus of rupture of specimens using 5% proportion of bagasse and rice husk ash can achieve ASTM C 67 [7]. Bagasse ash was also used as replacement materials in floor tile, which studied by Schettino et al. The proportion up to 2.5% bagasse ash can increase flexural strength when fired at 1190 ºC [8].

Furthermore, Schettino et al. had studied about utilizing bagasse ash in porcelain stoneware tile which fired at 1230 ºC. They found that ratio of bagasse ash up to 2.5% can provide the highest flexural strength [9]. Phonphuak and Chindaprasirt reported that utilizing 2.5% bagasse ash fired at 1000 ºC and 2.5%, 5.0%, and 7.5% bagasse ash fired at 1100 ºC can achieve the required strength of ASTM C62-13a for clay brick [10]. Utilizing calcium carbonate is also recognized. Kim et al. have prepared glass ceramics reinforced with wollastonite mixing with fluorescent waste glass and calcium carbonate. They found that firing temperature at 800 and 900 ºC can developed good chemical resistance phase in test specimens [11]. Serra et al. studied the effect of talc, spodumene and calcium carbonate playing the role as fluxes in triaxial ceramic properties. The results found that spodumene and calcium carbonate can enhance the sintering ability of specimen which fired at 1200 ºC. In addition, effect of talc and spodumene obtained the higher bending strength than that of calcium carbonate [12]. Furthermore, effects of calcium carbonate for firing shrinkage and moisture expansion were investigated by Lira et al. Proportion of calcium carbonate up to 10% can reduce firing shrinkage and moisture expansion of ceramic bodies fired at 1200 ºC [13].

Studying wastes from biomass power plants i.e., rice husk ash, Sobrosa et al investigated about using rice husk ash replacement in kaolin clay for producing refractory fired at 1300 ºC. They indicated that 10% replacement kaolin clay by rice husk silica can improve the mechanical strength and maintain the thermal shock resistance [14]. Abeid et al. had reported that ceramic bodies fired at 1180 ºC made from blending 20% vermiculite and 5% RHA by weight had the flexural strength closing to the requirement of ceramic tiles [15]. Rice husk ash was utilized as green material in concrete brick by Muhamad et al. The results showed that up to 10% rice husk ash can be used and achieve the Malaysian standard in terms of compressive strength and water absorption [16].

Fly ash from crude palm oil industries also are concerned. Many researchers have investigated the influence of utilizing this residue in civil products. Jamo et al. had studied about fly ash and rice husk ash which were utilized in porcelain ceramic. It was found that specimens containing 12% rice husk and 8% fly ash fired at 1200 ºC have the highest flexural strength [17]. In addition, More et al. also used these wastes; fly ash and rice husk ash for producing fired clay brick. The results indicated that mixing of 12.5% fly ash and 12.5% rice husk ash has the highest compressive strength [18]. Castellanos et al. attempted to investigate the effect of using fly ash for developing high impact strength roofing tile fired at 1060 ºC. They reported that mixing of clay, filler (grog), and fly ash showed higher peak forces and lower degree of damage than that mixing of clay and fly ash bodies [19].

Concerning on energy saving for all over the world is still a crucial issue. Ceramic processing is one of manufacturing that highly consuming energy in the firing process. Exploring the alternative materials for reducing energy consumption is the challenge work. Glass cullet is one of alternative materials that can reduce energy consuming by lowering firing temperature of ceramic bodies. There are many studied utilizing glass cullet in ceramic products. Pena et al. investigated the effect of particle size and proportion weight of glass cullet on physical properties of red clay brick. The results showed that 30% by weight and 212 micrometer glass cullet have the highest compressive strength [20]. For the other ceramic products, Karayannis et al. proposed fired ceramics 100% from lignite fly ash and glass cullet. They found that mixture of 15% glass cullet and 85% fly ash fired at 900 ºC can decrease porosity and improve the mechanical strength of ceramic bodies [21]. Loryuenyong et al. tried to improve the mechanical and physical properties of ceramic bricks with glass cullet. The results showed that compressive strength as high as 26–41 MPa and water absorption as low as 2–3% were achieved for bricks containing 15–30 wt% of glass content and fired at 1100 ºC [22]. Furthermore, Demir studied the effect of milled glass cullet adding in ceramic brick. It was found that up to 10% glass cullet and fired at 950, and 1050 ºC can obtain the highest compressive strength and achieve TS EN 771-1 standard [23]. Wangrakdiskul and Loetchantharangkun also utilized green glass cullet for lowering firing temperature and developing new texture color. They indicated that using 60% green glass cullet and fired at 950 ºC can achieve the Thai Industrial Standard (TIS 2508-2555). In addition, greenish brown color was developed [24].

This proposed research aims at utilizing various industrial wastes i.e., calcium carbonate, brown glass cullet, rice husk ash, bagasse ash, and fly ash for lowering sintering temperature of ceramic bodies. Further, for facilitating of molding specimens, the local clays consisting of ball clay and white clay from Prachinburi province in Thailand have been exploited.

2. Materials and methods

2.1. Materials

Materials used in this study are divided into two main types which are waste materials and local clay materials. Waste materials consist of calcium carbonate (CC), brown glass cullet (BGC), rice husk ash (RHA), bagasse ash (BGA), fly ash (FA). For local clay materials, local ball clay (LBC), local white clay (LWC), are used for improving the plasticity of ceramic bodies and easily forming of specimens. Chemical composition of all materials is analyzed by X-ray fluorescence (XRF) technique that is shown in Table 1. The X-ray fluorescence (XRF) was performed by a wavelength-dispersive XRF spectrometer with Bruker model S8 Tiger.

Table 1. Chemical composition of material used.

Compound	Concentration (%)						
	CC	BGC	RHA	BGA	FA	LBC	LWC
SiO_2	0.58	72.1	92.67	73.02	59.01	66.71	58.38
K_2O	0.02	0.2	1.96	3.36	-	1.85	0.19
CaO	54.75	2.4	1.18	5.44	11.08	0.09	0.04
P_2O_5	0.08	-	1.5	1.44	-	0.09	0.06
MgO	0.73	0.05	0.78	1.85	3.55	0.94	0.13
Fe_2O_3	0.08	0.1	0.54	3.53	7.44	3.89	2.11
Al_2O_3	0.19	1.6	0.56	10.91	1.15	24.65	37.87
Cl	0.07	-	0.16	0.1	-	-	-
SO_3	0.52	0.15	0.34	0.54	2.59	0.55	0.09
MnO	-	-	0.13	0.09	-	0.02	-
Na_2O	-	12.9	0.07	0.22	-	0.2	-
TiO2	-	-	0.03	0.41	0.1	0.86	1.06

Analyzed by X-ray fluorescence (XRF).

Experimental mixtures are constructed and divided into 4 groups. The basic formula No. 1_0 is derived from Wangrakdiskul and Loetchantharangkun [24], but using brown glass cullet (BGC) instead of green glass cullet (GGC). The first group is constructed by adding $CaCO_3$ residue to the basic formula by varying ratios from 0%, 5%, 10%, 15%, 20%, 25%, and 30%. After obtaining the optimal formula with a high bending strength of group 1, as compared of bending strength and water absorption with the Thai Industrial Standard 2508-2555 type BIII. it is used as the basic formula of groups 2, 3, and 4. They are constructed by exploiting RHA, BGA, and FA with the ratios 0%, 5%, 10%, 15%, 20%, 25%, and 30% to the basic formula of groups 2, 3, and 4, respectively, which can be described in Table 2.

Table 2. Experimental mixtures of 4 group-formulas.

Group	Formula No.	Composition (%)						
		CC	BGC	RHA	BGA	FA	LBC	LWC
1	*1_0	0	60	-	-	-	30	10
	1_1	5	55	-	-	-	30	10
	1_2	10	50	-	-	-	30	10
	1_3	15	45	-	-	-	30	10
	1_4	20	40	-	-	-	30	10
	1_5	25	35	-	-	-	30	10
	1_6	30	30	-	-	-	30	10
2	2_1	10	50	5	-	-	30	10
	2_2	10	50	10	-	-	30	10
	2_3	10	50	15	-	-	30	10
	2_4	10	50	20	-	-	30	10
	2_5	10	50	25	-	-	30	10
	2_6	10	50	30	-	-	30	10
3	3_1	10	50	-	5	-	30	10
	3_2	10	50	-	10	-	30	10
	3_3	10	50	-	15	-	30	10
	3_4	10	50	-	20	-	30	10
	3_5	10	50	-	25	-	30	10
	3_6	10	50	-	30	-	30	10
4	4_1	10	50	-	-	5	30	10
	4_2	10	50	-	-	10	30	10
	4_3	10	50	-	-	15	30	10
	4_4	10	50	-	-	20	30	10
	4_5	10	50	-	-	25	30	10
	4_6	10	50	-	-	30	30	10

*Note: 1_0 is the basic formula derived from Wangrakdiskul and Loetchantharangkun.

2.2. Methods

All materials are dried by the electric oven and milled by a ball mill. After sieved under 50 mesh (297 microns), they all mixed as the constructed mixtures by dry mixing. Then, they are molded by uniaxial pressing at 100 bars with dimension 50 × 100 × 7 millimeters. All specimens are fired by the electric kiln at 850 and 950 ºC with 100 ºC/h of heating rate and soaking for 1 h. Physical properties of specimens are examined consisting of linear firing shrinkage, weight loss, bulk density, bending strength, and water absorption. The bending strength and water absorption are carried out and compared with Thai Industrial Standard 2508-2555 (TIS 2508-2555 type BIII) [25]. Its requirement is ≥15 MPa of bending strength and 10–20% of water absorption. The microstructure of test samples is characterized by scanning electron microscope (SEM) and X-ray diffractometer (XRD). Scanning electron microscope has been operated by using a scanning electron microscope Hitachi SU3500 at an acceleration voltage of 10 kV with 5000× magnification. X-ray diffraction (XRD) analysis was carried out by a Bruker D8 diffractometer (Cu Kα radiation) with a step size of 0.01° and step time of 1 s over an angular range of 5–80°. In addition, the color measurement of specimens is employed by CIELAB color coordinate which using a UV–vis–NIR spectrometer (UV-3600 plus, Shimadzu).

The technological test of samples is examined by the methods and equations which will be explained as follows (Eqs 1–5).

2.2.1. Linear firing shrinkage was determined by measuring the length before (L_0) and after firing (L)

$$\%\text{Shrinkage} = \frac{L_0 - L}{L_0} \times 100 \tag{1}$$

2.2.2. Weight loss was determined a difference in weight of specimens before (W1) and after firing (W2)

$$\%\text{Weight loss} = \frac{w_1 - w_2}{w_1} \times 100 \tag{2}$$

2.2.3. Bulk density with Archimedes principle can be verified by determining the mass (m) and total volume (v)

$$p = \frac{m}{v} \tag{3}$$

2.2.4. Bending strength is performed by the three-point bending test, which P is the load (Force) at the fracture point (N), length of the support span (L), a is the thickness of specimens, and b is the width of specimens

$$\text{Bending strength} = \frac{3PL}{2a^2 b} \tag{4}$$

2.2.5. Water absorption is to determine difference of wet weight (Ww) and dry weight (Wd) after boiled specimens in water for 2 h at 110 ± 5 ºC and leaving them underwater for 4 h

$$\%\text{Water absorption} = \frac{w_w - w_d}{w_d} \times 100 \tag{5}$$

3. Results and discussion

3.1. Fluxing agents and physical properties

Fluxing agent (Na_2O, K_2O, and CaO) is considered as material reducing the sintering temperature of ceramic bodies [24]. Fluxing agent of materials used (BGC, RHA, BGA, FA, LBC, and LWC) are analyzed. For all formulas, fluxing agent has been expressed in Table 3. For calculating fluxing agent of these 3 oxides, different weight is considered with 1, 2.5, and 3 weight ratios of K_2O, CaO, and Na_2O.

Table 3. Fluxing agent of all materials used (BGC, RHA, BGA, FA, LBC, and LWC).

Compound	Concentration (%)					
	BGC	RHA	BGA	FA	LBC	LWC
K_2O	0.2	1.96	3.36	-	1.85	0.19
CaO	2.4	1.18	5.44	11.08	0.09	0.04
Na_2O	12.9	0.07	0.22	-	0.2	-

Considering on physical properties of group 1 as expressed in Table 4, it indicates that high value of fluxing agent has an effect on the high bending strength and high linear firing shrinkage, which is a similar result to Bragança and Bergmann [26]. Also, higher firing temperature can increase bending strength and linear firing shrinkage. Effect of high $CaCO_3$ (CC) content in each formula leads to high weight loss and water absorption. Comparing between 850 and 950 ºC, higher firing temperature and ≥20% $CaCO_3$ (CC) content has an effect on lowering shrinkage and increasing water absorption of specimens. However, bulk density of all formulas is not significantly different. Evidently, the formula No. 1_2 utilizing 10% CC and firing at 950 ºC have the high bending strength (14.69 MPa) and nearly achieve the TIS 2508–2555 (B ≥ 15 MPa). This formula will be further used as the basic formula for the next experiment group; groups 2, 3, and 4.

Table 4. Fluxing agent, bending strength, linear shrinkage, weight loss, bulk density, and water absorption of group 1 formulas fired at 850 and 950 ºC.

Group	Formula No.	Compositions (%)							%FA	B (MPa)		%S		%WL		D (g/cc)		%WA	
		CC	BGC	RHA	BGA	FA	LBC	LWC		850 ºC	950 ºC	850 ºC	950 ºC	850 ºC	950 ºC	850 ºC	950 ºC	850 ºC	950 ºC
1	1_0	0	60	-	-	-	30	10	5.28	14.69	27.70	3.27	3.61	4.35	3.70	2.29	2.21	12.59	10.02
	1_1	5	55	-	-	-	30	10	4.85	11.98	22.12	3.00	3.50	6.38	6.22	2.31	2.22	13.58	11.93
	1_2	10	50	-	-	-	30	10	4.43	11.40	18.52	2.91	3.24	8.68	8.07	2.34	2.23	14.33	13.89
	1_3	15	45	-	-	-	30	10	4.01	9.53	14.64	2.73	2.97	9.54	10.63	2.34	2.24	16.69	16.20
	1_4	20	40	-	-	-	30	10	3.59	8.48	10.06	2.70	2.38	12.21	12.52	2.36	2.28	19.20	21.09
	1_5	25	35	-	-	-	30	10	3.17	6.15	6.59	2.54	1.74	13.43	15.21	2.39	2.33	21.93	24.48
	1_6	30	30	-	-	-	30	10	2.75	4.05	4.24	2.48	1.64	15.06	18.14	2.39	2.34	24.61	29.69

FA: Fluxing agent; B: Bending; S: Shrinkage; WL: Weight loss; D: Density; WA: Water absorption.

For agricultural residue; RHA has been added to a basic formula (No. 1_2) varying from 5–30% by increased 5% in each formula of group 2. Likewise, BGA and FA are also used by adding in a basic formula for 5–30% by increased 5% in each formula of groups 3 and 4, respectively. The results are demonstrated in Table 5. Formula No. 2_1 of group 2 utilizing 5% RHA and firing at 950 ºC has the highest bending strength and lowest water absorption. This is the same result of group 3 (formula No. 3_1) and 4 (formula No. 4_1) which utilizing 5% BCG and 5% FA, respectively. They have the highest bending strength and lowest water absorption comparing within their group. These formulas can pass TIS 2508-2555 with bending strength and water absorption properties. However, firing specimens at 850 ºC unable to achieve TIS standard.

Table 5. Fluxing agent and physical properties of group 2, 3, and 4.

Group	Formula No.	Compositions (%)							%FA	B (MPa)		%S		%WL		D (g/cc)		%WA	
		CC	BGC	RHA	BGA	FA	LBC	LWC		850 ºC	950 ºC	850 ºC	950 ºC	850 ºC	950 ºC	850 ºC	950 ºC	850 ºC	950 ºC
2	2_1	10	50	5	-	-	30	10	4.06	11.42	16.61	2.62	3.02	7.97	8.17	2.31	2.21	17.65	16.70
	2_2	10	50	10	-	-	30	10	3.91	9.53	13.26	2.45	2.62	8.37	8.50	2.23	2.19	19.91	19.47
	2_3	10	50	15	-	-	30	10	3.77	8.06	10.03	2.27	2.20	8.73	8.93	2.24	2.19	22.40	22.92
	2_4	10	50	20	-	-	30	10	3.65	5.41	8.31	1.96	2.07	8.89	9.31	2.22	2.18	24.93	24.88
	2_5	10	50	25	-	-	30	10	3.53	3.59	5.58	1.63	1.94	9.01	9.48	2.18	2.18	27.07	27.01
	2_6	10	50	30	-	-	30	10	3.42	2.61	4.42	1.35	1.76	9.10	9.69	2.19	2.15	29.53	29.03
3	3_1	10	50	-	5	-	30	10	4.31	11.42	17.50	3.28	3.61	15.03	8.43	2.30	2.25	16.14	15.05
	3_2	10	50	-	10	-	30	10	4.20	10.03	15.25	3.37	3.63	15.07	8.72	2.29	2.25	17.97	17.54
	3_3	10	50	-	15	-	30	10	4.10	8.45	12.50	3.40	3.64	15.21	8.76	2.26	2.24	19.53	19.02
	3_4	10	50	-	20	-	30	10	4.01	6.70	9.73	3.34	3.47	15.25	9.14	2.26	2.24	21.06	20.76
	3_5	10	50	-	25	-	30	10	3.92	4.91	8.00	3.05	3.52	15.47	9.25	2.25	2.24	22.93	22.41
	3_6	10	50	-	30	-	30	10	3.84	2.94	7.09	2.87	3.48	15.67	9.40	2.24	2.23	24.80	23.47
4	4_1	10	50	-	-	5	30	10	4.29	10.60	16.64	3.04	3.47	8.16	8.25	2.30	2.28	16.85	16.50
	4_2	10	50	-	-	10	30	10	4.16	7.91	13.56	2.94	3.43	8.46	8.61	2.22	2.24	19.51	19.44
	4_3	10	50	-	-	15	30	10	4.04	7.57	11.46	3.03	3.40	8.80	9.02	2.29	2.23	21.36	21.20
	4_4	10	50	-	-	20	30	10	3.93	5.98	9.63	2.98	3.22	9.29	9.36	2.28	2.23	23.16	23.01
	4_5	10	50	-	-	25	30	10	3.82	4.84	7.52	2.94	3.23	9.38	9.77	2.25	2.20	24.87	24.46
	4_6	10	50	-	-	30	30	10	3.73	2.43	6.73	2.60	3.24	9.84	10.27	2.22	2.20	27.39	26.03

FA: Fluxing agent; B: Bending; S: Shrinkage; WL: Weight loss; D: Density; WA: Water absorption.

3.2. Characterization microstructure of test samples

Analyzing microstructure of fired specimens has been carried out by X-ray diffractometer (XRD) and Scanning Electron Microscope (SEM). With the highest bending strength, basic formula (No. 1_2) fired at 950 ºC has been selected for investigation. Due to the second highest bending strength, formula No. 3_1 fired at 950 ºC is also selected. For the lowest bending strength, formula No. 2_6 fired at 950 ºC has been selected. In Table 6, the results of XRD analysis have been illustrated. It found that high content of nepheline, wollastonite phase are developed in formula No. 1_2 and 3_1, which can improve the high bending strength of ceramic bodies [27]. However, lower content of nepheline, wollastonite phase in formula No. 2_6 has an effect on lowering bending strength. Furthermore, XRD patterns of selected formulas are illustrated in Figure 1.

Table 6. XRD analyzing of selected formulas specimens fired at 950 ºC.

Formula No.	% phase content						Bending strength (MPa)
	Cristobalite low	Quartz	Nepheline	Wollastonite	Calcium silicate	Tridymite	
1_2	18.45	22.48	26.46	28.05	0.89	3.67	27.7
3_1	20.1	18.86	27.55	29.24	0.76	3.48	16.64
2_6	42.09	9.34	19.28	22.46	0.84	5.98	4.42

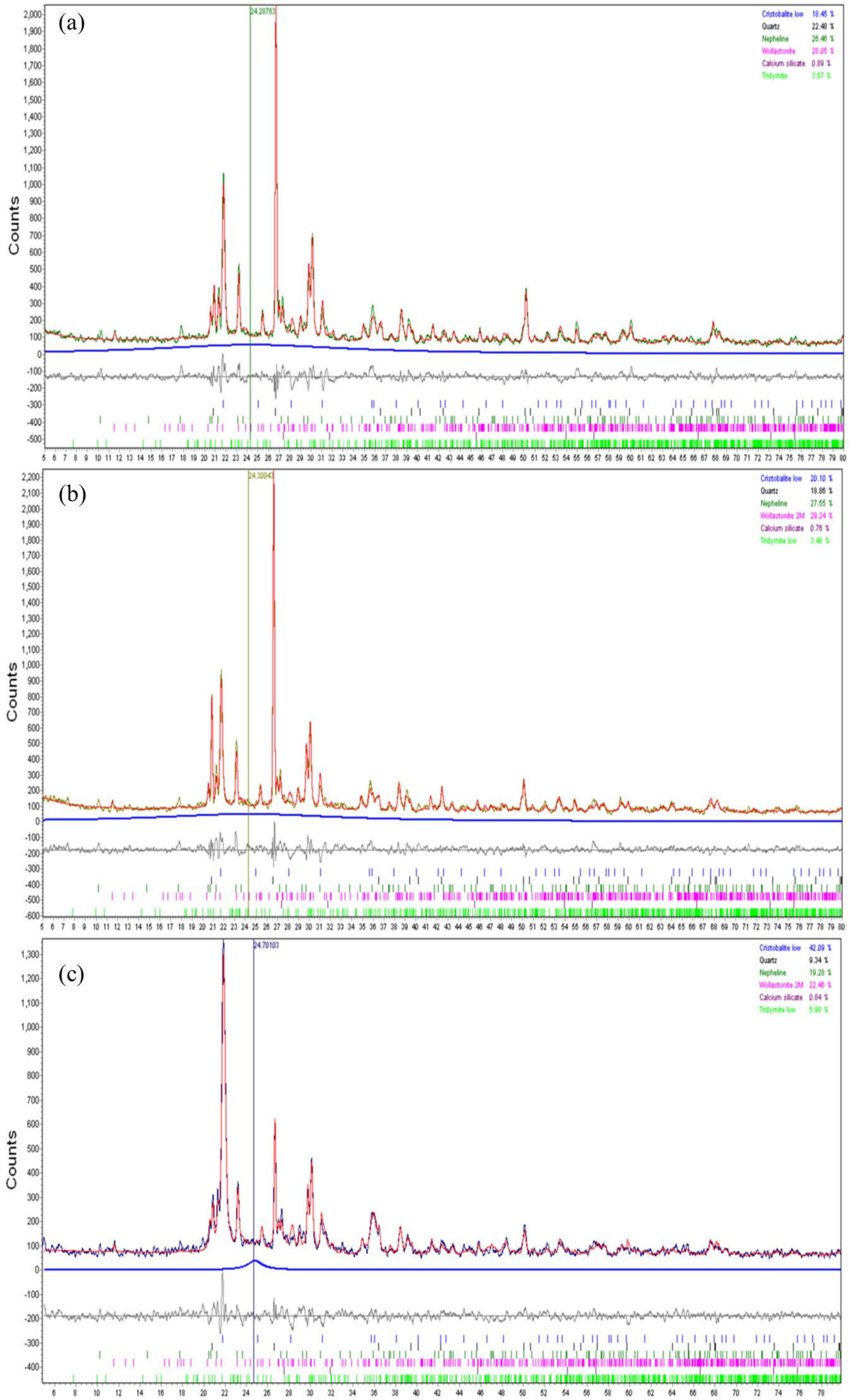

Figure 1. XRD pattern of selected formulas (a) formula No. 1_2, (b) formula No. 3_1, (c) formula No. 2_6.

The microstructure of selected formulas (formula No. 1_2, 3_1, and 2_6) are also characterized by SEM technique. They are carried out with a magnification of 3000×, and 5000× (Figure 2). They indicate that formula No.1_2, 3_1 represent the high dense body more than that of the formula No. 2_6. Due to formula No. 2_6 has a high RHA content which leads to the low strength of sample [28]. It also occurs the glassy phase in formula No.1_2 (Figure 2a,b) due to the high content of BGC [29]. High porous structure is found in formula No. 2_6 (Figure 2e,f), leading to low bending strength, as corroborates to Liu et al. [30].

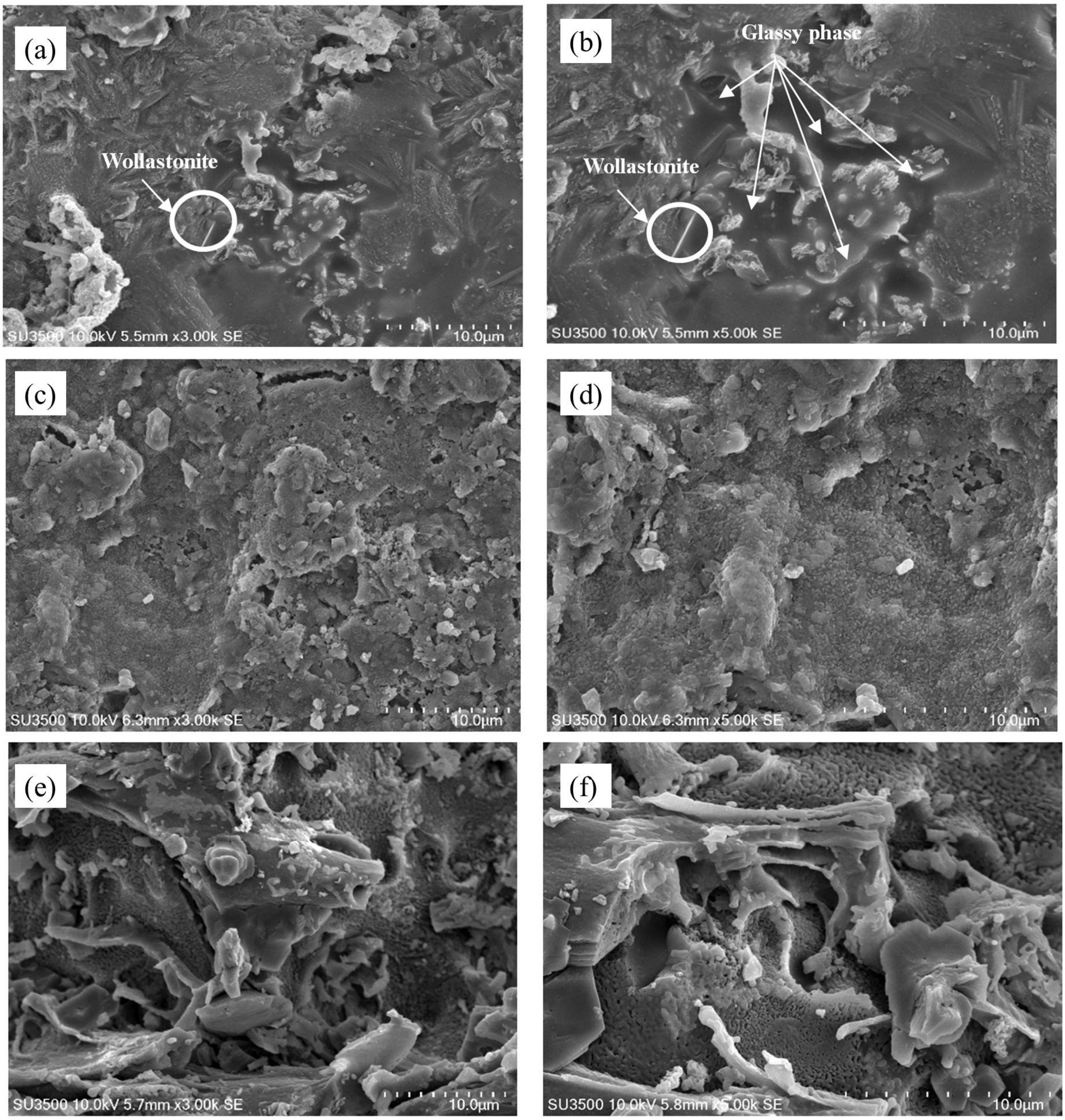

Figure 2. SEM pattern (a) No. 1_2 (×3000), (b) No. 1_2 (×5000), (c) No. 3_1 (×3000), (d) No. 3_1 (×5000), (e) No. 2_6 (×3000), (f) No. 2_6 (×5000).

3.3. Color measurement of 7 selected formulas

There are several methods for color measurements. However, the CIELAB method is usually accepted to specify the color of a product in ceramic practice. Its system has provided quantitative representation of color, which has been extensively applied in ceramic industry to study the existing products with the most immediately noticeable. This method measures the visible region to obtain the three parameters L*, a*, and b*, measuring brightness, red/green, and yellow/blue color intensities, respectively [31]. Color measurement by CIELAB test of selected formula's specimens is given in Table 7. The brightness coordinates L* (up to 68), color coordinates a* (up to 11.13) and b* (up to 21.05) are detected. The experiment shows that the dark brown color is occurred with the high content of BGC (60 wt%) of formula No. 1_0 which indicated in lower L* coordinates value (58.83). For the high coordinates L* (63.4–68) of formula 1_1, 1_2, 2_1, 3_1, 3_2, and 4_1, they have more lightness than that of formula 1_0. In addition, the CIELAB graphics of 7 formulas are illustrated in Figure 3.

Table 7. The color measurement of specimen fired at 950 ºC.

Formula No.	Compositions (%)							CIELAB test		
	$CaCO_3$	BGC	RHA	BGA	FA	LBC	LWC	L*	a*	b*
1_0	0	60	-	-	-	30	10	58.83	11.13	19.58
1_1	5	55	-	-	-	30	10	63.4	11.62	21.05
1_2	10	50	-	-	-	30	10	65.63	10.75	19.5
2_1	10	50	5	-	-	30	10	64.7	8.17	16.71
3_1	10	50	-	5	-	30	10	66.52	9.57	17.49
3_2	10	50	-	10	-	30	10	67.51	8.6	16.76
4_1	10	50	-	-	5	30	10	68	8.01	15.64

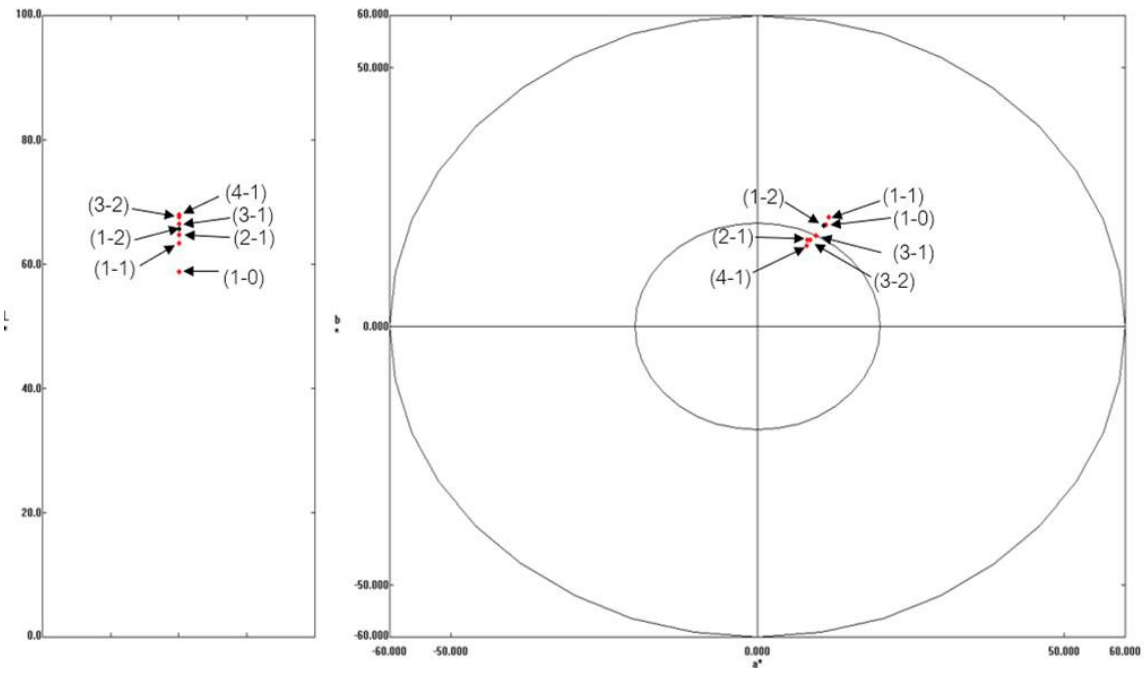

Figure 3. The CIELAB of 7 formulas fired at 950 ºC.

3.4. Comparison the physical properties of test samples with TIS 2508-2555

Four groups of this experiment have been summarized by comparing with TIS 2508-2555 focused on bending strength and water absorption [25]. There are 7 formulas (No. 1_0, 1_1, 1_2, 2_1, 3_1, 3_2, 4_1) that can achieve TIS 2508-2555 type BIII. For type BIII, it means that test samples have the high-water absorption but not exceeding 20%. They are all expressed in Table 8.

Table 8. Comparison bending strength and water absorption of the 7 formulas with TIS 2508-2555.

Formula No.	Compositions (%)							TIS 2508-2555 (BIII)	
	CC	BGC	RHA	BGA	FA	LBC	LWC	Bending strength (≥15, MPa)	%Water absorption (>10%, ≤ 20%)
1_0	0	60	-	-	-	30	10	27.7	10.02
1_1	5	55	-	-	-	30	10	22.12	11.93
1_2	10	50	-	-	-	30	10	18.52	13.89
2_1	10	50	5	-	-	30	10	16.61	16.7
3_1	10	50	-	5	-	30	10	17.5	15.05
3_2	10	50	-	10	-	30	10	15.25	17.54
4_1	10	50	-	-	5	30	10	16.64	16.5

3.5. Discussion for utilizing wastes on physical properties and texture of fired specimens

The results of this study can be concluded that utilization waste materials for producing eco-friendly ceramic tiles is feasible. They can achieve TIS 2508-2555 type BIII in terms of bending strength and water absorption. Increasing the replacement of CC in BGC, it leads to lower bending strength and higher water absorption of ceramic bodies. For utilizing RHA, BGA, and FA in the selected formula of group 1 (No. 1_2), they promote the similar effect of the formula which increasing content of CC. Moreover, utilizing BGA (group 3) provide the higher effect of bending strength than that of RHA (group 1) and FA (group 2), it corresponds to Hariharan et al. [32]. The color of specimens can pass TIS 2508-2555, as shown in Figure 4.

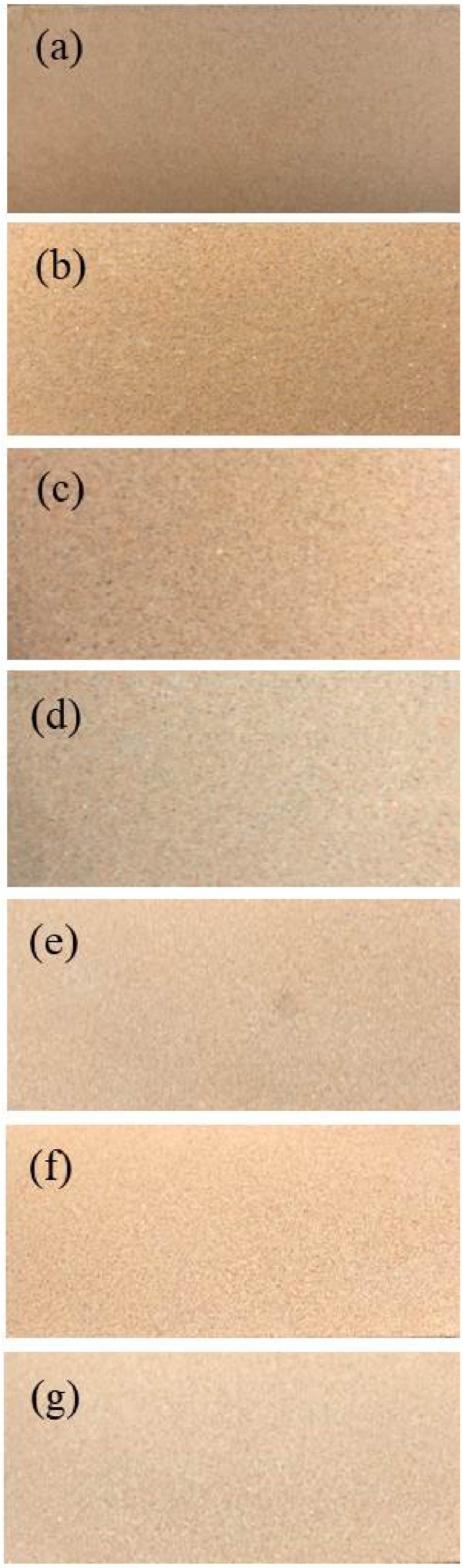

Figure 4. The color of specimens can pass TIS 2508-2555 (a) formula No. 1_0, (b) formula No. 1_1, (c) formula No. 1_2, (d) formula No. 2_1, (e) formula No. 3_1, (f) formula No. 3_2, (g) formula No. 4_1.

3.6. Cost considering of optimal formulas

The 7 formulas have been mentioned in the previous subsection are the optimal formulas. They can meet TIS 2508-2555 type BIII having high bending strength. Their production cost has been calculated as shown in Table 9. It consists of material, energy cost fired at 950 ºC. However, labor cost is not considered, due to this calculating is based on the lab scale. Total cost of optimal formulas is calculated by considering on 100 pieces of specimens. The lowest total cost is 356.13 THB/100 pcs of formula No. 1_2. However, total cost of all formulas is slightly difference which varying between 356.36–16.83 THB/100 pcs.

Table 9. Production cost estimation of 7 formulas.

List	*Cost	Unit	fn. 1_0		fn. 1_1		fn. 1_2		fn. 2_1		fn. 3_1		fn. 3_2		fn. 4_1	
			Qu	CT	Qu	CT	Qu	CT	Qu	CT	Qu	CT	Qu	CT	Qu	CT
CC	-	TK	-	0	0.38 K	0	0.77 K	0	0.73 K	0	0.73 K	0	0.70 K	0	0.73 K	0
BGC	2.2	TK	4.61 K	10.14	4.23 K	9.306	3.85 K	8.47	3.66 K	8.052	3.66 K	8.052	3.50 K	7.7	3.66 K	8.052
RHA	0.9	TK	-	0	-	0	-	0	0.37	0.33	-	0	-	0	-	0
BGA	-	TK	-	0	-	0	-	0	-	0	0.33 K	0	0.70 K	0	-	0
FA	0.72	TK	-	0	-	0	-	0	-	0	-	0	-	0	0.33 K	0.24
LBC	2	TK	2.31 K	4.62	2.31 K	4.62	2.31 K	4.62	2.2 K	4.4	2.2 K	4.4	2.3 K	4.6	2.2 K	4.4
LWC	2	TK	0.77 K	1.54	0.77 K	1.54	0.77 K	1.54	0.73 K	1.46	0.73 K	1.46	0.70 K	1.4	0.73 K	1.46
RM	12.3	Th	2.5 h	30.75	3 h	36.9	2.5 h	30.75	3 h	36.9	3 h	36.9	3 h	36.9	3 h	36.9
Dr	24.6	Th	2.5 h	61.5	2.5 h	61.5	2.5 h	61.5	2.5 h	61.5	2.5 h	61.5	2.5 h	61.5	2.5 h	61.5
Pr	8.2	Th	1 h	8.2	1 h	8.2	1 h	8.2	1 h	8.2	1 h	8.2	1 h	8.2	1 h	8.2
Firing	22.96	Th	10.5 h	241.1	10.5 h	241.1	10.5 h	241.1	10.5 h	241.1	10.5 h	241.1	10.5 h	241.1	10.5 h	241.1
Total cost of 100 pieces			-	358	-	363	-	356	-	362	-	362	-	361	-	362

*Note: Cost of the lab scale; RM: Raw material; Dr: Drying; Pr: Pressing; TK: THB/Kg; Th: THB/hr; K: Kg; h: hr; Qu: Quantity used; CT: Cost (THB); fn: formula No.

4. Conclusions

This study has used waste industrial materials; rice husk ash, bagasse ash, calcium carbonate, fly ash, and brown glass cullet in order to produce eco-friendly unglazed fired clay tiles. The seven formulas can achieve TIS 2508-2555 type BIII (No. 1_0, 1_1, 1_2, 2_1, 3_1, 3_2, 4_1) with fired at 950 ºC. The comparison of the properties including three types of ashes, the best formula uses 10% of bagasse ash that has the highest bending strength is 17.5 MPa and water absorption of 15.05%. The next group, the formula uses 5% of fly ash that has bending strength of 16.64 MPa and water absorption 16.5%. In the last group, the formula uses 5% of rice husk ash that has a bending strength of 16.61 MPa and water absorption of 16.7%. The color of the 7 formulas has slightly different tones. Values are in the range of between L* = 58.83–68, a* = 8.01–11.62, b* = 15.64–21.05.

The results of this research can alleviate the burden of manufacturers for disposing and managing their wastes. In addition, utilizing these wastes for producing the valued products can promote the value added for these wastes. However, increasing the amount of using them is the challenge work. It can be further carried out the darker color, increasing firing temperature or exploiting the other sintering material such as boric acid should be further investigated.

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Conflict of interest

All authors declare no conflicts of interest in this paper.

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