Effect of Utilizing Different Colors Waste Glass and Calcium Carbonate Waste on Properties of Ecological Ceramic Clay Tiles

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Abstract

This research aims at investigating the effect of utilizing different colors waste glass and calcium carbonate on properties of ecological ceramic clay tiles. The wastes materials used in this study is consisted of Calcium carbonate (CC), Green glass cullet (GGC), Clear glass cullet (CGC), Brown glass cullet (BGC). In addition, Local ball clay (LBC), and local white clay (LWC) have been combined in the mixtures. The process of producing such specimens starts from sintering the materials at 850 ºC and 950 ºC. The properties of the specimens are then tested, in which the test results can be grouped into group A, B, and C. Group A, which contains portions of green glass cullet, has the lowest water absorption value of 13.35 % and 11.06 % and the highest bending strength of 10.44 and 17.19 MPa when firing at 850 ºC and 950 ºC. With utilizing clear glass cullet of group B, it has the lowest water absorption value of 14.25 % and 11.16 %, and the highest bending strength of 14.66 and 19.27 MPa of firing at two temperatures. **Group C**, containing the portions of brown glass cullet, has the lowest water absorption value of 12.59 % and 10.02 % and the highest bending strength of 14.69 and 27.7 MPa. of firing at two temperatures. It is found that sintering specimens at high temperature resulting in higher bending strength values and low water absorption. Furthermore, group C has higher bending strength than that of Group A and B resulting in an increase of product strength. As utilizing these wastes for producing environmentally friendly wall tiles., this study can alleviate the problem of degradation environment from industrial wastes and reduce its disposal cost.
Keywords: Calcium Carbonate, Green Glass Cullet, Clear Glass Cullet, Brown Glass Cullet, Ecological Wall Tiles

1. Introduction

Today in Thailand, there are many types of industries generating wastes that can potentially harm the environment. The Pollution Control Department reported about the use of recycled material in the industrial sector in 2013 – 2017, it showed that the amount of waste generated from this sector is increasing every year. However, the amount of reused and recycled material is still lower than the waste generated. In the recycled and reused segment, total glass cullet waste generated were 2,550,000 tons, meanwhile only 1,600,000 tons were recycled and 250,000 tons were reused, 700,000 tons were left [1].

In agricultural sector in Thailand, sugar cane is cultivated the most comparing to agricultural areas [2]. It is used as the main raw material for the sugar mill plant. During the process, CaO has been used for purifying sugar juice and after precipitation

Calcium Carbonate ($CaCO_3$) is generated. It has been disposed and can harm the environment.

This research focuses on recycling industrial waste, as material for developing ecological wall tiles. Furthermore, this can decrease the disposal of unused industrial waste altogether. Ponce Peña et al. experimentally added Green Glass fragments to clay bricks. The sizes of the fragments are less than 500 µm, 300 µm, and 212 µm. The result of the smallest particles is the strongest, which can reduce energy consumption of firing process and enhance the properties of clay bricks [3]. Ogunro et al. added glass fragments of 0, 5, 10, and 12 % in concentration, sintered at 800, 900, 1000, 1100, and 1200 °C. The specimen with 10% concentration yielded lesser tile porosity, enhanced the properties of the tiles, decreased the temperature and duration required during sintering process [4]. Karayannis et al. added glass fragments of 0 – 15 % concentration and then sintered at 900 °C. By adding the glass fragments, the tile porosity was reduced by 12 %, the strength of the tiles was increased, and the sintering temperature was decreased [5]. Theerapapvisetpong et al. added glass fragments to roof tiles resulting in less water absorption property, increase in tile strength, and was able to pass the TIS 158 – 2518 standards [6]. Mozo et al. added 10 % and 20 % for reducing cracks, deformations, and increase compressive strength of the bricks [7]. Chidiac et al. experimented with adding glass fragments to clay bricks in ratio of 0, 2.5, 5, and 10 % according to ASTM, C67 1986 standards, then sintered at 850, 950, and 1050 °C. It was found that adding glass fragments and sintering at 950 and 1050 °C produced the strongest specimens [8]. Lira Serra et al. studied the mixture of Calcium Carbonate at 5, 10, and 15% in ceramic body then sintered at 1050, 1125, and 1200 ºC. The result had shown that the mixture of CC can reduce moisture expansion and firing shrinkage of ceramic test pieces [9]. Serra et al. studied the mechanical properties and flux influence by conducting an experiment at 800 – 1200 ºC. Clay, quartz, and feldspar mixture, the industrial standard mixture was used as the comparison standard. Talc, Spodumene, and Calcium Carbonate were added to the experimental mixture for replacement of feldspar. The best mixture from the experiment consisted of Clay 60 %, Quartz 15 %, Feldspar 10 %, Talc 15 %, Spodumene 15 %, and Calcium Carbonate 15 % [10]. Kim et al. prepared wollastonite reinforced ceramic glass by using fluorescent glass and calcium carbonate as the precursor material, then sintered at 800, 900, and 1000 °C. It was found that when ceramic glass was fired at 800 and 900 °C, the chemical resistance had increased due to crystalline phase [11]. Bakar et al. found that adding 5 % Calcium Carbonate (CC) and comparing to unused CC, the specimen with 5 % CC has better microstructure, density, porosity, shrinkage, and bending strength [12]. Loetchantharangkun and Wangrakdiskul studied the use of CC by adding up to 10 % mixture. If adding CC higher than 10 %, it will be the result in decreasing of bending strength [13].

This research aims to recycle industrial waste which are 3 types of glass waste from recycling plant and Calcium Carbonate waste from sugar mill factory. It is the expected benefit for reducing waste disposal, adding value of wastes for developing ecological ceramic clay tile.

2. Method and Experiment

The materials used in this research consist of 6 types, which are calcium carbonate (CC), green glass cullet (GGC), clear glass cullet (CGC), brown glass cullet (BGC), local ball clay (LBC), and local white clay (LWC). The chemical composition of each material is analyzed at the National Institute of Nuclear Technology by X-ray Fluorescence (XRF) technique. The results are shown in Table 1.

Table 1 Chemical composition of material used in the experiment

Compound	Concentration (%)					
	CC	GGC	CGC	BGC	LBC	LWC
SiO_2	0.58	70.24	69.96	69.19	66.7	58.4
K_2O	0.02	0.32	0.14	0.21	1.85	0.19
CaO	54.8	11.45	10.9	10.89	0.09	0.04
P_2O_5	0.08	-	-	-	0.09	0.04
MgO	0.73	1.10	2.35	2.71	0.94	0.13
Fe_2O_3	0.08	0.45	0.14	0.35	3.89	2.11
Al_2O_3	0.19	1.79	1.93	2.26	24.7	37.9
Cl	0.07	0.04	0.04	0.02	-	-
SO_3	0.52	0.09	0.19	0.08	0.55	0.09
Na_2O	-	13.98	14.26	14.11	0.2	-
TiO_2	-	0.09	0.06	0.1	0.86	1.06
Cr_2O_3	-	0.33	-	0.03	0.01	0.01
ZrO_2	-	0.03	0.02	0.03	0.03	0.06

Remark Analyzed by X-ray Fluorescence (XRF)

Calcium carbonate (CC), green glass cullet (GGC), clear glass cullet (CGC), brown glass cullet (BGC), local ball clay (LBC), and local white clay (LWC) were sizing by 50 mesh (297µm.) sieve. The mixture formulars were divided into 3 groups consisting of 21 formulars. The ratio of CC was varied from 0 – 30 % and the ratio of glass fragment was varied from 30 – 60 %. Group A, B, and C used GGC, CGC, and BGC as the glass fragment mixture respectively as shown in Table 2.

Table 2 Mixture formulation of the experiment

	Concentration (%)	CC	GGC	CGC	BGC	LBC	LWC
Group A	A1	0	60			30	10
	A2	5	55			30	10
	A3	10	50			30	10
	A4	15	45			30	10
	A5	20	40			30	10
	A6	25	35			30	10
	A7	30	30			30	10
Group B	B1	0		60		30	10
	B2	5		55		30	10
	B3	10		50		30	10
	B4	15		45		30	10
	B5	20		40		30	10
	B6	25		35		30	10
	B7	30		30		30	10
Group C	C1	0			60	30	10
	C2	5			55	30	10
	C3	10			50	30	10
	C4	15			45	30	10
	C5	20			40	30	10
	C6	25			35	30	10
	C7	30			30	30	10

*A1 is the basic formula derived from Wangrakdiskul and Loetchantharangkun

The materials of each formula were prepared by dry mixing process for 30 minutes, then sprayed with 10 % of water by the total weight and sized by the 20 mesh (841 µm.) sieve. The mixture was then compression molded at 100 bars for producing test piece with the dimension of 50 x 100 x 7 mm. The test pieces then were dried at 200 °C for 2 hours, and sintered at heating rate of 100 °C / hour and maintained at the maximum temperatures of 850 and 950 °C for 1 hour. After that, the properties of fired specimens were tested and analyzed consisting of firing shrinkage, weight Loss, bending strength, density, and water absorption.

3. Results

3.1 Fluxing agent analysis

Before describing the test properties of specimens, fluxing agent analyzing will be identified. Na_2O, K_2O, CaO [14] are the fluxing oxide which can reduce the sintering temperature of ceramic bodies. Considering the quantity of fluxing oxide of 3 glass cullet types, the fluxing agent value of BGC is lower than that of GGC and CGC. However, ratio of fluxing agent compared with SiO_2 of BGC is higher than that of GGC and CGC, as shown in table 3. This means that an effect of BGC for lowering the sintering of specimens is higher than the other types of glass cullet.

Table 3 Comparison fluxing oxide of 3 glass cullet types and ratio of fluxing oxide by SiO_2

Cullet Type	Compound (%)												
	CaO			K_2O			Na_2O						
	%Content	Wt. ratio	%Wt.ratio	%Content	Wt. ratio	%Wt.ratio	%Content	Wt. ratio	%Wt.ratio	Total	Total/ 8.5	%SiO2%Content	$(CaO+K_2O+Na_2O)/SiO_2$
GGC	11.45	1	11.45	0.32	2.5	0.8	13.98	5	69.9	82.15	9.66	70.24	0.1376
CGG	10.9	1	10.9	0.14	2.5	0.35	14.26	5	71.3	82.55	9.71	69.96	0.1388
BGC	10.89	1	10.89	0.21	2.5	0.525	14.11	5	70.55	81.965	9.64	69.19	0.1394

Remark Total ratio of $CaO+K_2O+Na_2O$ = 8.5

3.2 Physical properties

3.2.1 Bending strength (B) and firing shrinkage (%S) of test pieces

- They have the similar trend as shown in table 4. The effect of increasing CC content has decreased bending strength and firing shrinkage. It is the same result of group A, B, and C.
- However, bending strength (B) and firing shrinkage (%S) of group C has the highest value. It is the impact of high ratio fluxing oxide by Al_2O_3 of BGC in group C as mentioned in the previous sub section.
- In addition, higher firing temperature can promote the higher B and %S of specimens as indicated in table 4. Therefore, firing temperature at 950 ºC has the higher effect than that of 850 ºC.

Table 4 Physical properties: bending strength, firing shrinkage, weight loss, bulk density, and water absorption of the experimental formulas

No	B (MPa)		% S		%WL		D (g/cc)		%WA	
	850 °C	950 °C	850 °C	950 °C	850 °C	950 °C	850 °C	950 °C	850 °C	950 °C
A1	10.44	17.19	2.35	2.71	4.01	4.63	2.33	2.22	13.35	11.06
A2	10.18	15.78	2.3	2.62	6.01	6.23	2.34	2.23	14.04	12.45
A3	9.16	15.14	2.22	2.44	8.43	9.15	2.35	2.24	15.56	14.42
A4	7.91	11.81	2.15	2.21	10.73	10.97	2.36	2.25	17.89	17.42
A5	5.65	8.93	2.04	1.98	12.85	13.16	2.37	2.27	20.89	20.52
A6	4.92	6.1	1.94	1.52	14.64	15.08	2.38	2.31	23.98	24.68
A7	3.24	3.73	1.61	1.27	16.12	17.82	2.38	2.34	27.32	28.34
B1	14.66	19.27	3.13	2.99	3.89	4.51	2.3	2.23	14.25	11.16
B2	12.84	18.45	2.78	2.9	6.00	6.58	2.3	2.23	14.38	11.94
B3	11.83	17.44	2.71	2.77	8.23	9.13	2.31	2.24	15.31	14.22
B4	8.68	14.29	2.49	2.6	10.4	11.39	2.31	2.26	17.97	16.87
B5	7.68	10.52	2.4	2.2	12.52	13.72	2.31	2.29	21.05	20.06
B6	6.27	6.71	2.27	1.94	15.88	17.04	2.37	2.33	24.35	24.81
B7	3.52	3.87	2.13	1.82	18.26	18.69	2.38	2.34	28.17	29.31
C1	14.69	27.7	3.27	3.61	3.7	4.36	2.29	2.21	12.59	10.02
C2	11.98	22.12	3.00	3.5	6.04	6.37	2.31	2.22	13.58	11.93
C3	11.4	18.52	2.91	3.24	8.07	8.68	2.34	2.23	14.33	13.89
C4	9.53	14.64	2.73	2.97	9.54	10.63	2.34	2.24	16.69	16.2
C5	8.48	10.06	2.7	2.38	12.21	12.52	2.36	2.28	20.41	19.2
C6	6.15	6.59	2.54	1.74	13.43	15.21	2.39	2.33	21.93	24.48
C7	4.05	4.24	2.48	1.64	15.61	18.14	2.39	2.34	25.4	29.69

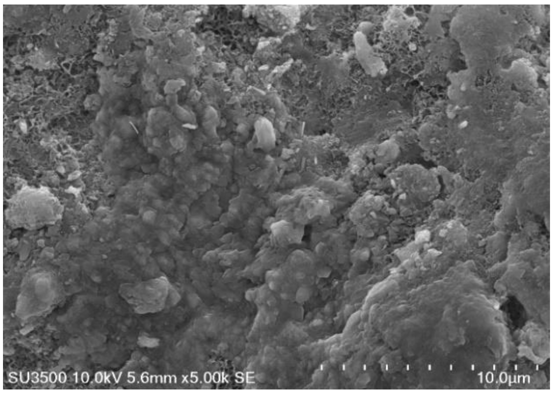
Note Fluxing agent = FA; bending = B; shrinkage = S; weight loss = WL; density = D; and water absorption = WA.

3.2.2 Weight loss, density, and water absorption

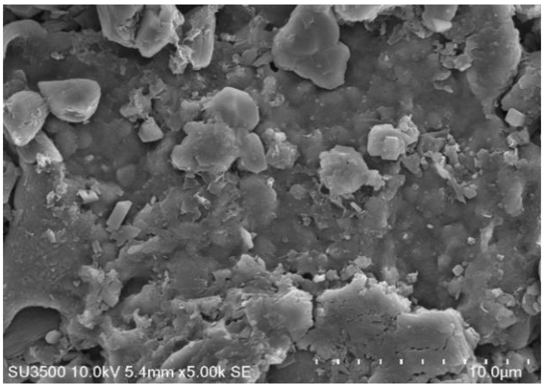
- In table 4, the result shows increasing trend of %weight loss (%WL), density (D) and %water absorption (%WA), when increasing CC content. It is the effect of CC has transformed to CaO and generated CO_2 form the specimens. WL and WA values are inversely proportional to B and S properties.
- When comparing between 950 and 850 °C, the higher sintering temperature can yield the higher WL and WA.
- Density value of all group formulas and two firing temperature is slightly difference. This means that varying materials and firing temperature have the slightly effect on fired specimens.

3.3 Microstructural Analysis

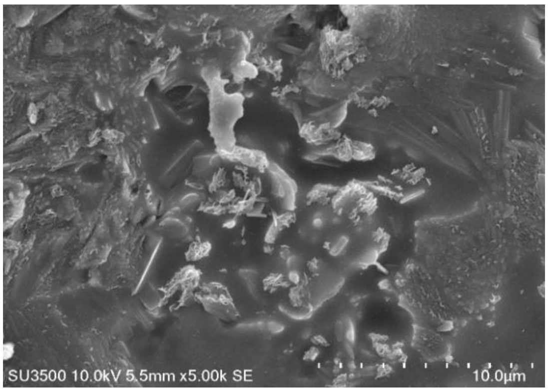
- Formula no. A3, B3, and C3 of group A, B, and C fired at 950 ºC have been selected. They all can pass TIS 2508 - 2555 type BIII and utilized CC upto10 %.
- The SEM micrograph with the 5000X magnification has been illustrated in Figure 1. Figure 1 (a) shows the microstructure of formula A3 with high porosity resulting in low bending strength [15], similarly the microstructure of formula B3 which can be observed in Figure 1 (b). Formula C3 are shown in Figure 1 (c). The glassy phase occurred and formed Mullite ($3Al_2O_3$ $2SiO_2$) which shows the needle-like shape in white circle. It indicates that the specimen is stronger than the specimens in Figure [16] 1 (a) and 1 (b).



(a)



(b)



(c)

Figure 1 SEM micrograph (a) formula no. A3 (5000X); (b) formula no. B3 (5000X) ; (c) formula no. C3 (5000X) fired at 950 °C

4. Conclusions and discussion

- The results from this experiment show that 9 mixture formulars can pass the TIS 2508 - 2555 type BIII. It is the high-water absorption type. They are expressed in table 5. Utilizing glass cullet is varied from 50 - 60 % wt., and 0 - 10 % of calcium carbonate content. They are the same content for all 3 groups; group A, B, and C.

Table 5 TIS 2508 – 2555 Qualified formulars sintered at 950 °C

Formula No.	Compositions						TIS 2508-2555 (B III)	
	CC	GGC	CGC	BGC	LBC	LWC	Bending Strength >=15 (MPa)	%Water absorption >10%, ≤ 20%
A1	0	60			30	10	17.19	11.06
A2	5	55			30	10	15.78	12.45
A3	10	50			30	10	15.14	14.42
B1	0		60		30	10	19.27	11.16
B2	5		55		30	10	18.45	11.94
B3	10		50		30	10	17.44	14.22
C1	0			60	30	10	27.7	10.02
C2	5			55	30	10	22.12	11.93
C3	10			50	30	10	18.52	13.89

- This research focuses on producing ecological wall tiles that qualifies TIS 2508 – 2555 BIII standard. It can alleviate the environmental problem from industrial waste such as Green Glass, Glass Cullet, Brown Glass, Calcium Carbonate.
- Sintering temperature has affected the strength of the tiles, sintering at 950 °C yielded stronger than 850 °C.
- Brown glass cullet has the higher impact for promoting high bending strength than that of green glass cullet and clear glass cullet.
- The higher content of calcium carbonate can be utilized, if the sintering temperature is increased.
- Wall tiles can be further developed and produced in small and medium enterprises for responding the demand of customers who are interested in environmentally friendly products.

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