

Effect of Composition on Characteristics of Powder Coating Sludge (PCS)-Metakaolin-Based Geopolymer

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Abstract

The aim of this research is to experimentally study the application of powder coating sludge (PCS) and metakaolin as geopolymer. The PCS is a lot of waste which is released after the powder coating production process. While metakaolin comprises SiO_2 and Al_2O_3 which can be used to produce geopolymer. Therefore, this study aimed to determine a suitable geopolymer mix ratio, using PCS as part of the mixture. This research also investigated the effect of metakaolin/PCS ratio, sodium hydroxide (NaOH)/sodium silicate (Na_2SiO_3) ratio, curing time, NaOH concentration and solid/liquid ratio on compressive strength of geopolymer. The coefficient of thermal conductivity was tested by using a suitable composition. Kaolin in this experiment was calcined at 700 °C for 2 h. Alkaline activators for molding were NaOH and Na_2SiO_3 . Samples were hardened in an electric oven at 80 °C for 4 h before being removed from the moulds and cured at room temperature within 7, 14 and 28 days. Metakaolin/PCS ratios of 90:10, 80:20, 70:30 and 60:40, and solid/liquid ratios of 0.8 and 0.6 were performed. Concentration of NaOH were 6, 8, 10, 12 and 14 molars and NaOH/ Na_2SiO_3 ratios were 1:1.5, 1:2 and 1:2.5, respectively. The experiments were conducted in triplicate. Results of 3 PCS tests showed that the highest composition of Al_2O_3 and SiO_2 were 3.37 and 1.99 % w/w, respectively. The sample which is based on 60 % metakaolin and 40 % PCS, 10 molars NaOH, NaOH/ Na_2SiO_3 of 1:1.25, solid/liquid of 0.6 and curing time of 28 days had a maximum compressive strength of 81 kg/cm², while the coefficient of thermal conductivity was 0.1766 W/(m·k). Therefore, it can be concluded that the PCS has the potential to partially replace metakaolin as geopolymers material.

Keywords: Powder coating sludge, Metakaolin, Compressive strength, Geopolymer, X-ray fluorescence

Introduction

In Thailand, Ordinary Portland Cement (OPC) is the most extensively used building material. This observation agrees with the findings of Lin *et al.* [1] who found that the OPC was the most used building material worldwide with 1.6 billion tons produced annually [2]. However, Elumalai *et al.* [3] found that the production of OPC released carbon dioxide and the cement industry globally produced around 2.8 billion tons of the green gases emissions annually, approximately 7 % of the petrol emission to the earth's atmosphere. Moreover, OPC production needs large amounts of energy consumption [4,5]. It is necessary to find alternatives for suitable concrete which is more environmentally friendly and to reduce energy consumption. Geopolymer mainly comprises alumina (Al_2O_3) and silica (SiO_2). Therefore, it has excellent mechanical properties and chemical resistance and producing geopolymeric source material consumes a little energy [1,6-8]. Metakaolin is a common industrial mineral and its degradation is evaluated to emit 80 - 90 % less CO_2 than the OPC; hence, it is more environmentally friendly [9,10]. Kaolinite was used to partially substitute for fly ash to study the effect of composition and temperature on properties of geopolymer which showed that the maximum compressive strength was achieved when metakaolinite was calcined between 400 and 600 °C [11]. Khale and Chaudhary [12] reviewed that the

usual range of calcining kaolinite was about 600 - 800 °C and Elimbi *et al.* [13] found that the most convenient temperature for the calcination of kaolinite clays in view of producing geopolymer cement was around 700 °C. Powder coating is commonly used in a variety of applications, including furniture, air conditioners, electrical appliances, automotive parts and architectural work. Thus, powder coating production has continuously increased in Thailand [14,15]. However, there are a lot of PCS left after the production process. More than 180 tons of the PCS were left each year per production line and a lot of PCS seriously affected the environment as pollution. Moreover, companies have to deal with the elimination of PCS, in both quantity and methods, which can be difficult to manage in the future. To solve the problem, this study aims to explore chemical properties of PCS, then adapt and use it to partially replace metakaolin in order to produce geopolymer. The experiments will examine and seek for appropriate metakaolin/PCS ratio and concentration of activators or solution to form specimens. Then, compressive strength and coefficient of thermal conductivity will be tested by the standard.

Materials and methods

Kaolin, metakaolin and PCS

Table 1 shows the percentage of chemical composition of kaolin from Uttaradit province, Thailand [16]. The table shows that the kaolin comprises mainly Al_2O_3 and SiO_2 therefore it can produce geopolymer [1,6-8]. **Figure 1(A)** shows the original kaolin from Uttaradit province which was used for this study. Literature indicates that in order to produce geopolymer materials, many authors have calcined kaolin at various temperatures [1,12,13]. Therefore, in this study, the kaolin was calcined at a temperature of 700 °C for 2 h and then the kaolin became metakaolin (**Figure 1(B)**). Three sets of powder coating sludge (PCS) were obtained from a powder coating manufacturing plant in Thailand (**Figure 1(C)**).

Table 1 Percentage of chemical composition of kaolin from Uttaradit province, Thailand [16].

SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	TiO ₂	CaO	K ₂ O	Na ₂ O	MgO	Rb ₂ O	MnO	ZrO ₂	CuO	BaO	Other
59.5	17.3	1.78	0.18	0.13	4.81	0.01	0.87	0.03	0.02	0.03	0.02	0.06	15.26

Specimen preparation

Granizo *et al.* [17] studied Alkali activation of metakaolins: Parameters affecting mechanical, structural and microstructural properties. They performed the Na (OH) as activators of 6, 8, 10, 12, 15, 18 and 20 M. The result was found that mechanical strength of metakaolin with sodium silicate in combination with NaOH as the alkali activation was higher than the alkali activation with NaOH alone. Lin *et al.* [1] tested their work by using the alkali activation with sodium hydroxide/sodium silicate mole ratio of 0.8 to 2, solid/liquid ratio of 0.4, 0.6, 0.8 and 1 with curing time of 1, 7, 28 and 60 days, while NaOH/ Na₂SiO₃ of 1.2 was tested by Kong *et al.* [18]. Therefore, all parameters of this study are shown in **Table 2**. **Figures 2(A) - 2(C)** show the preparation of geopolymer samples. Slurry after was mixed in the specified proportions of the metakaolin, PCS and activator solution as shown in **Figure 2(A)**. Then the slurry was cast in 5×5×5 cm³ cubic moulds (**Figure 2(B)**) and vibrated for about 10 min to remove trapped air. These samples were sealed (**Figure 2(C)**) and left at room temperature for an hour before being hardened in an electric oven at 80 °C for 4 h, removed from the moulds (**Figure 3(A)**) and cured at room temperature with 7, 14 and 28 days, respectively. Each experiment was tested in duplicate therefore 3 samples were prepared in each batch.

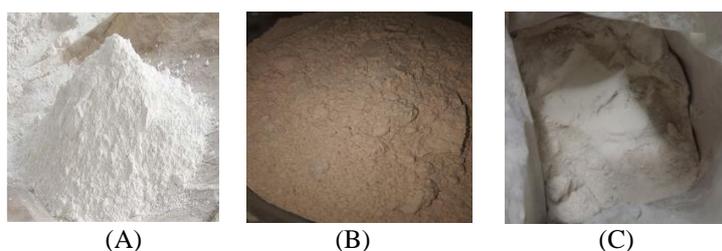


Figure 1 The materials: (A) Kaolin from Uttaradit, (B) Metakaolin and (C) PCS.

Table 2 The test parameters of this study according to previous research.

solid/liquid = 0.6		
Metakaolin/PCS (wt %)	Concentration of NaOH (molar)	NaOH/ Na ₂ SiO ₃
90:10, 80:20, 70:30, 60:40	6, 8, 10, 12, 14	1:1.5, 1:2, 1:2.5

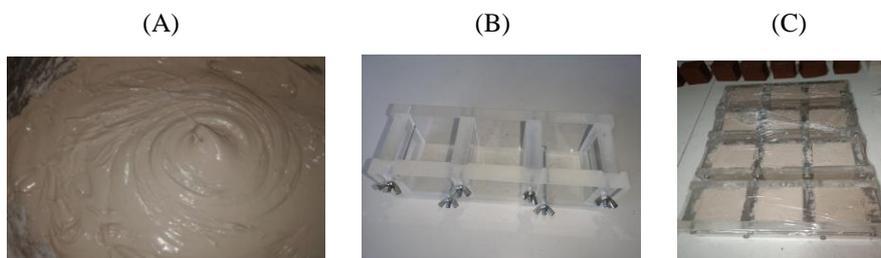


Figure 2 Specimen preparation: (A) Slurry after was mixed by metakaolin, PCS and activator solution, (B) Cubic moulds for cast the slurry and (C) Samples were sealed and left at room temperature.

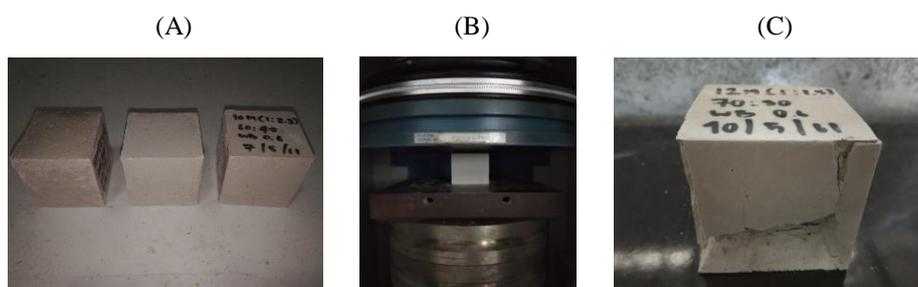


Figure 3 (A) Specimens were cured at the room temperature of 7, 14 and 28 days, (B) The compressive testing machine (ADR-Auto) and (C) The specimen after tested following procedure ASTM 109.

Analysis methodology

The composition of PCS, metakaolin and geopolymer samples were performed by X-ray fluorescence (XRF) analysis (**Figure 4**). The compressive strength of the samples was tested after the curing time of 7, 14 and 28 days following the standard test of ASTM 109 (**Figures 3(B)** and **3(C)**). The coefficient of thermal conductivity was tested using a sample with 20×20×2.5 cm³ following the standard test of ASTM C177.

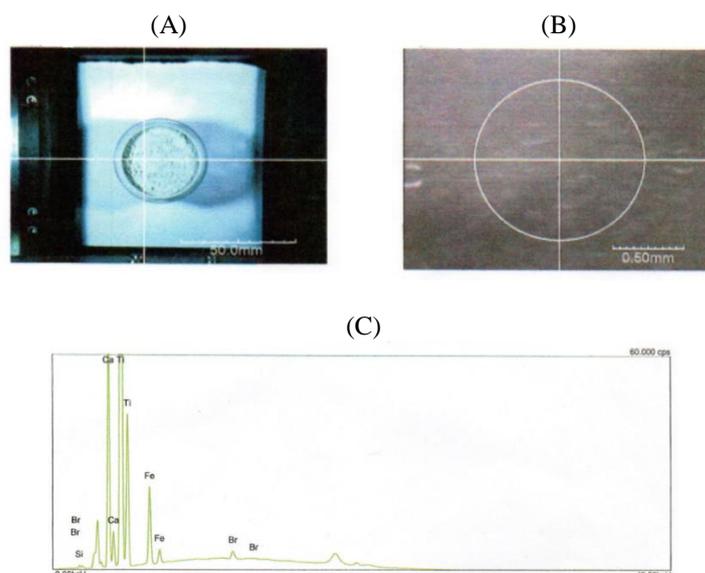


Figure 4 X-ray fluorescence (XRF) analysis: (A) Whole image, (B) Detail image and (C) Sample testing result.

Results and discussion

Composition of PCS, metakaolin and geopolymer

Chemical composition of 4 sets of PCS and metakaolin were tested by X-ray fluorescence (XRF) analyzer (**Tables 3 and 4**). Referring to **Table 3**, the result showed that all sets of PCS comprised Al_2O_3 and SiO_2 . The PCS No.4 had the highest composition of Al_2O_3 and SiO_2 , 3.37 and 1.99 % w/w, respectively therefore the PCS No.4 was used for this study. While the Al_2O_3 and SiO_2 of metakaolin were about 16.99 and 66.85 % w/w, respectively (**Table 4**). The results can be confirmed that the PCS and metakaolin can be used to produce geopolymer. Moreover, the Ti content of PCS No.1 was approximately 42.21 % w/w (**Table 3**), which can provide good compressive strength. This observation agrees with the findings of Naskar and Chakraborty [19]. Their experiments observed that 1 % TiO_2 addition provides good strength at both of 7 and 28 days.

Table 3 Composition of PCS.

Composition	No.1 (% w/w)	No.2 (% w/w)	No.3 (% w/w)
Al_2O_3	3.37	3.26	1.59
SiO_2	1.99	1.37	1.13
SO_3	11.55	12.28	12.55
TiO_2	48.99	47.78	47.79
SrO	0.56	0.62	0.66
BaO	33.55	34.70	36.27

Table 4 Composition of metakaolin and geopolymer.

Composition	metakaolin (% w/w)	geopolymer (% w/w)
Al_2O_3	16.99	10.84
SiO_2	66.85	53.44
K_2O	10.26	6.33
CaO	-	5.04
TiO_2	0.31	16.80
Fe_2O_3	5.43	7.31
Rb_2O	0.10	0.12
ZrO_2	0.06	0.13

Effect of metakaolin/PCS ratios on compressive strength

The effect of the metakaolin/PCS ratios on compressive strength with NaOH/Na₂SiO₃ ratios of 1:2.5, sodium hydroxide concentration of 10 molar and curing time of 28 days as shown in **Figure 5(A)**. The figure shows that the compressive strength with the metakaolin/PCS ratios of 90:10, 80:20, 70:30 and 60:40 were 34.7, 52.2, 55.2 and 81 kg/cm², respectively. It can be seen that when the amount of PCS increases, resulting in increased content of Ti, and the increasing of Ti content is related to increasing of the compressive strength [19]. The same figure also shows the maximum compressive strength of approximately 81 kg/cm² with the metakaolin/PCS ratios of 60:40, NaOH/ Na₂SiO₃ ratios of 1:2.5 and curing time of 28 days.

Effect of sodium hydroxide concentration on compressive strength

Figure 5(B) shows the effect of sodium hydroxide concentration on compressive strength with metakaolin/PCS ratios of 60:40, NaOH/ Na₂SiO₃ ratios of 1:2.5 and curing time of 28 days. The same figure shows that the compressive strength with the sodium hydroxide concentration of 6, 8, 10, 12 and 14 molar was 75.7, 74.2, 81, 65.1 and 62.4 kg/cm², respectively. It can be seen that when sodium hydroxide concentration was increased from 6 to 10 molar, the compressive strength was increased and reached the maximum at the concentration of 10 molar. On the other hand, when the sodium hydroxide concentration was increased from 10 to 14 molar, the compressive strength was decreased and reached the minimum at the concentration of 14 molar. Therefore, increasing of the activating solution volume in the 1st period (before 10 molar) affects a rising in structure of Si and Na and this result causes increasing of compressive strength. The activating solution volume was then increased in the 2nd period (after 10 molar), as were the liquid and porosity volumes, and these results significantly affected the decrease in compressive strength [17]. The maximum compressive strength occurred at 10 molar of the sodium hydroxide concentration, this agrees with Ridditirud [20].

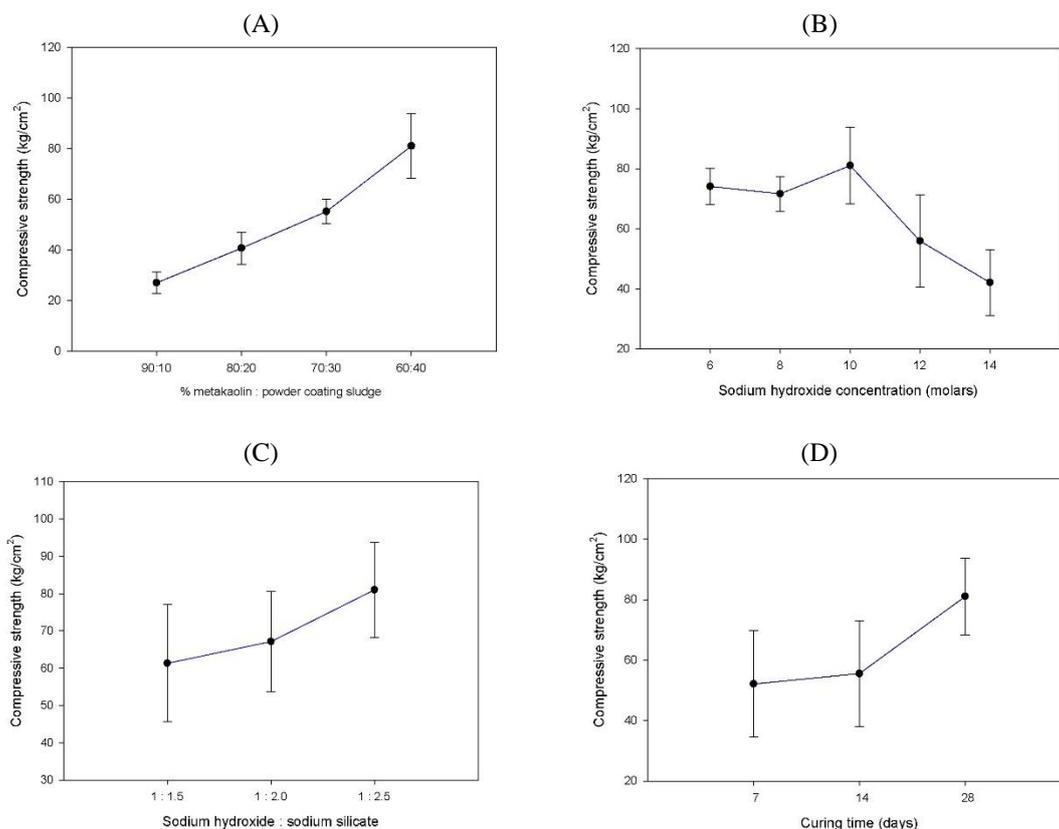


Figure 5 Results: (A) Effect of metakaolin/PCS ratios on compressive strength, (B) Effect of sodium hydroxide concentration on compressive strength, (C) Effect of NaOH/Na₂SiO₃ ratios on compressive strength and (D) Effect of curing time on compressive strength.

Effect of NaOH/ Na₂SiO₃ ratios on compressive strength

When the NaOH/Na₂SiO₃ ratios were slightly increased, the compressive strength was significantly increased as shown in **Figure 5(C)**. This observation agrees with Granizo *et al.* [17]. From the same figure, the compressive strength of the sample with the NaOH/Na₂SiO₃ ratios of 1:1.5, 1:2 and 1:2.5 when the metakaolin/PCS ratios of 60:40, sodium hydroxide concentration of 10 molars and the curing time of 28 days were 61.3, 67.2 and 81 kg/cm², respectively. The results show that the amount of NaOH and Na₂SiO₃ affects the compressive strength of geopolymer. Moreover, the degree of polymerization is significantly affected the compressive strength of geopolymer [1].

Effect of curing time on compressive strength

Figure 5(D) shows that when the curing time was significantly increased, it caused the compressive strength of the samples to increase, which agrees with the study of Lin *et al.* [1]. The compressive strength of the sample with curing time of 7, 14 and 28 days when the metakaolin/PCS ratios of 60:40, NaOH/Na₂SiO₃ ratios of 1:2.5, and sodium hydroxide concentration of 10 molars were about 52.1, 55.5 and 81 kg/cm², respectively. **Figure 5(D)** also shows that when the curing time was increased from 7 to 14 days, the compressive strength was slightly increased. The compressive strength was, then, sharply increased after the curing time of 14 days, which agrees with Ridtirud [20].

Composition of geopolymer

To validate of geopolymer of this research, the composition of the sample with metakaolin/PCS ratios of 60:40, NaOH/Na₂SiO₃ ratios of 1:2.5, sodium hydroxide concentration of 10 molars and curing time of 28 days was tested by the XRF method. **Table 4** shows that the composition of Al₂O₃ and SiO₂ of the sample was about 10.84 and 53.44 % w/w, respectively. Therefore, it can be confirmed that the sample of this research is geopolymer. Moreover, the composition of approximately 21.79 % Ti ensures that the geopolymer has a high compressive strength.

Coefficient of thermal conductivity

The coefficient of thermal conductivity of the sample with metakaolin/PCS ratios of 60:40, NaOH/Na₂SiO₃ ratios of 1:2.5, sodium hydroxide concentration of 10 molars, and curing time of 28 days was performed according to the standard procedure of ASTM C177. The result showed that the thermal conductivity coefficient of the geopolymer in this study was 0.1766 W/(m·k). While the coefficient of thermal conductivity of brick (common) and concrete blocks were 0.72 and 0.77 W/(m·k), respectively [21]. When the result of this study was compared with brick (common) and concrete blocks, it was found that the thermal conductivity coefficient of this study was lower than brick (common) and concrete blocks by about 75.5 and 77.1 %, respectively. Therefore, it can be concluded that this geopolymer is a good insulation material.

Conclusions

From this research, it can be concluded that the PCS can be reused and can replace Portland. The maximum compressive strength was 81 kg/cm² with metakaolin/PCS ratios of 60:40, NaOH/ Na₂SiO₃ ratios of 1:2.5, sodium hydroxide concentration of 10 molars and curing time of 28 days. Moreover, this geopolymer has a high Ti, the strength was increased. Finally, the geopolymer of this research has a low thermal conductivity coefficient therefore it can be a good insulating material.

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