### A Sustainable Cement Replacement Material From a Coal-Fired Power Plant Waste

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#### Abstract

Electrical energy is an important in a daily life. Coal is a primary fuel for electricity production. The waste from the coal-fired electricity generation is a fly ash. The numerous fly ash is a low quality fly ash (LQFA). LQFA is a low reactivity ash, and is a hazardous waste. Normally, LQFA is stored in the landfills. The heavy metals can leach from the stored LQFA into an environment, which is a serious problem. The sustainable ways to solve the problem is to utilize LQFA as a valuable material.

Although, cement is widely used in the construction work, it is an environmental unfriendly material. The high quantities of carbon dioxide gas are emitted from the cement producing process. Geopolymer is expected to be a sustainable cement replacement material due to its manufacturing process does not emit a carbon dioxide gas. Furthermore, geopolymer can be synthesized from the various types of waste materials

The aim of this work is to develop the geopolymer by using the LQFA and a calcined clay (CC) as a precursor. The results show that the compressive strength of LQFA-CC-geopolymer is higher than that of the commercial cement. The compressive strength of LQFA-CC-geopolymer is increased with increasing the concentration of NaOH and the ratio of Na<sub>2</sub>SiO<sub>3</sub> to NaOH solution. The knowledge of this work opens an opportunity to utilize the LQFA as a cement replacement material to reduce an environmental impact of the coal-fired power plant waste.

Keywords: Coal-fired electricity waste, Cement replacement, Geopolymer, Fly ash, Calcined clay

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#### Introduction

Portland Cement (PC) is one of the most used materials in the world. However, PC has negative environmental impacts because its production process requires high energy consumption and release a lot of carbon dioxide gas. The latter is the main cause of the global warming. Recently, several researchers have been investigating for a sustainable cement replacement material. Geopolymer is one of the most interested alternatives.

Geopolymer is an environmental friendly material, and, is expected to be used as the sustainable cement replacement materials. Because, the geopolymer production process consumes low energy and does not emit carbon dioxide gas to an environmental. Geopolymer can be synthesized by using the aluminosilicate materials mix with alkali hydroxide and/or alkali silicate. The aluminosilicate materials are the silica (SiO<sub>2</sub>) and alumina (Al<sub>2</sub>O<sub>3</sub>) rich materials. Variety of aluminosilicate materials are used for synthesized geopolymer such as calcined clay (CC), fly ash, bottom ash, and water treatment sludge (Ekkasit et al., 2016).

Fly ash is the waste from the coal-fired power plant, and is mainly used as the pozzolanic materials for the building applications. However, not all of fly ashes are suitable for construction applications. The properties of fly ashes depend on the fuel nature and the furnace used (Wesche, 1991). High quality fly ash is a highly desirable commodity in the construction industry, because, it is a high reactivity ash. High quality fly ash is obtained from high quality coal (anthracite and bituminous) with high temperature (around 1100 to1700 °C) combustion system. Morphology of the high quality fly ash is mainly vitreous spherical particles. However, the numerous fly ash is the low quality fly ash (LQFA). Wesche (1991) has described that LQFA is a low reactivity ash, and, has a high percentage of irregular crystalline particles. LQFA is a hazardous waste, it is undesirable for building applications. Normally, LOFA is stored in the landfills. The heavy metals can leach from the stored LQFA into an environment, which is a serious problem. In addition, the number of LOFA trend to significantly increase year by year. The sustainable ways to solve the problem is to treat LQFA as a valuable material, that is geopolymer. Although, several researchers study on the development of geopolymer from fly ash, there are a few work reports on the development of geopolymer from LQFA.

The aim of this work is to develop a geopolymer material from LQFA blends with a calcined clay (CC). The effect of LQFA/CC ratios, concentration of sodium hydroxide (NaOH) and the ratios of sodium silicate (Na<sub>2</sub>SiO<sub>3</sub>) to sodium hydroxide on compressive strength of the LQFA-CC geopolymer are examined. This research will enable LQFA traditionally destined for landfills to be used in a sustainable manner as a precursor in geopolymer, which is significant from engineering and environmental point of view.

### Materials and methods

#### **Precursor preparation**

The LQFA was passed through a sieve (number 120 mesh) and oven-dried at 100 °C for 24 hours to remove the impurities in the LQFA. Kaolinite clay from Ranong province, Thailand was milled by mortar and passed through a sieve (number 120 mesh) before being calcined at 600 °C for 3 hours to obtain CC powder.

### Sample preparation

Sodium hydroxide (NaOH) pellets and distilled water were mixed to obtain a concentration of 5, 10 and 15 M, then allowed to cool down at a room temperature. The alkali activator solution was prepared by mixing sodium silicate (Na<sub>2</sub>SiO<sub>3</sub>) solution with NaOH solution. The ratios of Na<sub>2</sub>SiO<sub>3</sub> to NaOH were 0:1, 0.5:1, 1:1, 1.5:1, 2:1 and 1:0 by weight. The mixed solutions were stored for 24 hours prior to use. Chemical composition of Na<sub>2</sub>SiO<sub>3</sub> solution consists of Na<sub>2</sub>O = 16.3 %, SiO<sub>2</sub> = 34.2 % and H<sub>2</sub>O = 49.5 %.

LQFA and CC powder were mixed in the various LQFA/CC ratios of 100:0, 50:50, 40:60, 30:70, 0:100 by weight. The mixed powder was mixed with an alkali activator solution by a mortar at a solid to liquid ratio of 1.0. Then, the slurry mixture was poured into a 50 mm x 50 mm x 50 mm steel mold and compacted as described in ASTM C109 (2002). The samples along with the molds were sealed with vinyl sheet to prevent moisture evaporation during curing at 60 °C for 7 days.

# **Characterization techniques**

Morphology and mineral compositions of LQFA were evaluated by scanning electron microscope (SEM, JOEL JSM-6010LV) and X-ray diffraction (XRD, Bruker D5005) with CuK<sub> $\alpha$ </sub> radiation, respectively. X-ray fluorescence (XRF, HORIBA XGT-5200) was used to analyze chemical compositions of LQFA and CC. Density and compressive strength of the 7 days cured samples were measured following ASTM C138 (2009) and ASTM C109 (2002), respectively.

# **Results and Discussion**

Chemical compositions of LQFA and CC are seen in table 1. Main chemical compositions of LQFA and CC are  $SiO_2$  and  $Al_2O_3$ .

Table 1. Chemiear compositions of EQTA and CC.								
Raw	Chemical compositions (weight%)							
materials	$Al_2O_3$	SiO <sub>2</sub>	K <sub>2</sub> O	Na <sub>2</sub> O	CaO	MgO	Fe <sub>2</sub> O <sub>3</sub>	Etc.
LQFA	19.68	33.19	2.29	2.40	11.93	4.43	10.64	15.44
CC	48.48	49.40	0.99	0.08	0.01	0.12	0.67	0.25

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The X-ray diffractograms of LQFA is seen in Figure 1, crystalline phase of  $SiO_2$  and  $CaSO_4$  are the main compositions. Figure 2 shows SEM micrograph of LQFA. Almost all particles are irregular in shape. The XRD and SEM results demonstrate that the used fly ash is in the type of LQFA.



Figure 1 X-ray diffraction (XRD) spectra of LQFA



Figure 2 SEM image of LQFA; almost all particles are irregular in shape.

Figure 3 illustrates that by using LQFA as a precursor alone, geopolymer has low strength. Because, LQFA has low reactivity with the alkali activator solution. Compressive strength of LQFA-geopolymer is around 10 MPa, is lower than the minimum requirement for PC of 19 MPa (ASTM C150, 2003). Compressive strength of CC-geopolymer is around 34 MPa, which is higher than that of LQFA-geopolymer. Because, CC has high reactivity with the alkali activator solution (Zuhua et al., 2014). Therefore, compressive strength of LQFA-geopolymer is improved via replace LQFA by CC. The maximum strength is obtained at LQFA/CC ratio of 50:50 by weigth, and is around 42 MPa. However, strength of LQFA-CC-geopolymer is reduced when LQFA is replaced by CC over 50 wt%. The role of the LQFA/CC ratio on the developed strength is explained in the term of the Si/Al ratios. Table 2 presents Si/Al ratios for various LQFA/CC ratios, which were calculated from the chemical compositions of the LQFA and CC (Table 1), as well as the Na<sub>2</sub>SiO<sub>3</sub> solution. The Si/Al ratio is an importance factor for the developed strength of geopolymer.

suitable Si/Al ratios for the cement replacement materials were reported as approximately 3 - 4 (Chindaprasirt et al., 2012 and Abdullah et al., 2011). Although, CC has high reactivity with the alkali activator solution, Si/Al ratio is reduced with increasing of CC replacement.





**Figure 3** compressive strength of the geopolymers with different LQFA/CC ratios; all samples used NaOH 15 M mixed with Na<sub>2</sub>SiO<sub>3</sub> (Na<sub>2</sub>SiO<sub>3</sub>/NaOH ratio = 2:1), as the alkali activator solution.

Compositions (weight %)		Si/Al ratios	
LQFA	CC	SI/AI Tatios	
100	0	2.4	
50	50	1.6	
40	60	1.5	
30	70	1.4	
0	100	1.3	

Table 2. Calculation of Si/Al ration in the different replacement of LQFA by CC

Figure 4 and Figure 5 demonstrate that the high compressive strength of LQFA-CCgeopolymer is obtained by using high concentration of NaOH (Figure 4) with high Na<sub>2</sub>SiO<sub>3</sub>/NaOH ratios (Figure 5). In addition, figure 5 shows that both of NaOH and Na<sub>2</sub>SiO<sub>3</sub> are required for obtaining the high strength LQFA-CC-geopolymer. By using NaOH or Na<sub>2</sub>SiO<sub>3</sub> as the alkali activator solution alone, LQFA-CC-geopolymer has low strength. Strength of geopolymer is developed via geopolymerization process. The role of NaOH and Na<sub>2</sub>SiO<sub>3</sub> in the geopolymerization process is the dissolvent and the binder, respectively. High concentration of NaOH promotes dissolution of fly ash, as well as, increases the concentration of Al<sup>3+</sup> and Si<sup>4+</sup> ions (Ubolluk and Prinya, 2009). The linkage between Al<sup>3+</sup> and Si<sup>4+</sup> ions in the geopolymerization process is enhanced by increasing amount of Na<sub>2</sub>SiO<sub>3</sub> which act as a binder. By using 15 M of NaOH with Na<sub>2</sub>SiO<sub>3</sub>/NaOH ratio is 2:1, compressive strength of LQFA-CCgeopolymer is around 42 MPa, which is higher than the minimum requirement of PC.



**Figure 4** compressive strength of the geopolymers with different concentration of NaOH. All samples have the ratio of LQFA/CC and Na<sub>2</sub>SiO<sub>3</sub>/NaOH is 50:50 and 1.5:1, respectively.



**Figure 5** compressive strength of LQFA-CC-geopolymer with different Na<sub>2</sub>SiO<sub>3</sub>/NaOH ratios; the LQFA/CC ratio of 50:50 and NaOH 15 M are used for all samples.

Density of material is important for a structure cost. Density of all samples in the present work are around 1.12 to 2.18, which are lower than that of PC. The density of PC is approximately 3.15 g/cm<sup>3</sup> (ASTM C138, 2009; ASTM C185, 2008; ASTM C595/C595M, 2013). Low density is an advantage of the LQFA-CC-geopolymer over PC, which can be used for development of the lightweight materials. The outcome of the present work opens an opportunity to utilize LQFA as the sustainable cement replacement materials.

#### Conclusions

In the present work, a sustainable cement replacement material was developed by using a coal-fired power plant waste, i.e., low quality fly ash (LQFA) blend with a calcined clay (CC), as the precursor. LQFA has low reactivity with the alkali activator solution. Reactivity of the precursor is enhanced by blend LQFA with CC. Strength of the LQFA-CC-geopolymer is higher than the minimum requirement of Portland cement (PC) around 2 times after curing at 60 °C for 7 days. By using NaOH or Na<sub>2</sub>SiO<sub>3</sub> as the alkali activator alone, LQFA-CC-geopolymer has low strength. The high concentration of NaOH solution and the sufficiency of Na<sub>2</sub>SiO<sub>3</sub> binder are required to obtain the high strength LQFA-CC-geopolymer. Density of the LQFA-CC-geopolymer is significantly lower than that of PC. The knowledge in this work opens an opportunity to convert the hazardous west to be the useful product, i.e., a sustainable cement replacement material.

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