

DESIGN OF EXPERIMENT OF GEOPOLYMER FROM METAKAOLIN BLENDED WITH FLY ASH

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Abstract

Portland cement is one of the most widely used materials. However, the production process of this material releases carbon dioxide gas, which is the main cause of the global warming effect. Nowadays, many researchers have been developing a sustainable cement replacement material. Geopolymer is the outstanding candidate. It is an inorganic polymeric material which is known as an environmentally sustainable material. In this paper, a 2^k full factorial design of experiment (DOE) was studied and used to find the response or compressive strength of the geopolymer. The DOE factors were the oven curing temperature and metakaolin to fly ash ratio. The results of the compressive strength of the oven curing temperature at 30°C were better than oven curing at a temperature of 60°C. By utilizing analysis of variance, these factors have been optimized to determine the compressive strength. The results of the significant factors indicated that the most important factor was the metakaolin to fly ash ratio. Moreover, this study further tested at an oven curing temperature 30°C different metakaolin to fly ash ratios: 0%, 10%, 30%, 50%, 70%, and 100%. The most outstanding result was the 70% of metakaolin to fly ash ratio. This research presents the design of the experiment for utilizing hazardous waste, i.e, fly ash as a valuable material.

Keywords: Design of experiment, geopolymer, metakaolin, fly ash

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Introduction

Portland cement (PC) is widely used as a construction material around the world. However, PC is an environmentally unfriendly material because the PC production process consumes high energy and emits high quantities of carbon dioxide gas (Kong and Sanjayan, 2010). The required temperature to produce cement is around 1,450°C. The carbon dioxide gas released into the atmosphere is around 1 ton for the production of 1 ton of ordinary PC (Imbabi *et al.*, 2012). As is well known, carbon dioxide gas is the main cause of global warming. Recently, geopolymers have been investigated by several researchers as a sustainable cement replacement material.

Geopolymer is an inorganic and environmentally friendly material. Geopolymer can be synthesized by mixing aluminosilicate materials (silica- (SiO_2) and alumina- (Al_2O_3) rich materials) with an alkali hydroxide and/or alkali silicate solution. After curing, the obtained strength of the geopolymer is sufficient for construction applications. The production process of geopolymer consumes low energy and releases 80% less carbon dioxide than the PC production process (Kim and Kim, 2017). In addition, geopolymer can be synthesized from various kinds of waste materials such as slag, water treatment sludge (Nimwinya *et al.*, 2016), and fly ash.

Fly ash is the waste from a coal-fired power plant. Normally fly ash is used as a pozzolanic material. However, only highly reactive fly ashes are suitable for construction applications. High quality fly ash (HQFA) is obtained from high quality coal (anthracite and bituminous) with a high temperature (over 1,100°C) combustion system. HQFA-based geopolymer usually has a high compressive strength comparable with PC (Nasvi *et al.*, 2012). Skvara *et al.* (2005) have reported that HQFA-based geopolymer has an excellent durability in the corrosive environment of salt solution. However, much of the fly ash is low quality fly ash (LQFA) which is undesirable for building applications, because LQFA has low pozzolanic reactivity. Normally, LQFA is stored in landfills.

There are many factors that have an effect on the properties of geopolymer such as the concentration of NaOH, the ratio of Na_2SiO_3 to NaOH, and the precursor powder to an alkali solution ratio. Time and a lot of money are required to do an experiment of all the parameters. Design of experiment (DOE) is an effective way to determine the significant factors for reducing the research time and the research budget. Although numerous researchers have studied the development of geopolymer from fly ash (Ramesh and Joy, 2017; Zhuang, *et al.*, 2016; Al Bakri, *et al.*, 2011), few research works have reported on the development of geopolymer from LQFA. In addition, the significant parameters that have an influence on the compressive strength of LQFA-based geopolymer have never been revealed. The aim of this research is to develop geopolymer material from LQFA blended with calcined clay for use as a sustainable cement replacement material. The significant parameters that influence the properties of LQFA-based geopolymer have been evaluated by using the experimental design methods.

Design of Experiment

DOE is an experiment in which the input variables or factors are changed to consider the causal factors involved with changes in responses in the process of interest (Montgomery, 2005). Sir Ronald Fisher (1960) first developed this method in the 1920s. Input variables can be divided into the following 2 types: controllable variables and uncontrollable variables. Experimental designs are usually done in a certain order. The first experiment is conducted to screen the variables (the screening experiment) and find key variables from all the controllable variables in the experiment. The next experiment is aimed at determining a suitable level of variables affecting the outcome nearest to the target. A factorial experiment is an experiment capable of studying multiple factors simultaneously

with the main objective of studying co-effects between the factors or interactions (Sammy, 2002). Factorial experiments can be divided into 2 types, namely, full factorial experiments and fractional factorial experiments. The impact analysis of the main factors and interactions that is commonly used is analysis of variance (ANOVA) by beginning with consideration of the interactions. If the interactions are significant, the main factors affected by the interactions will not be determined. Many researchers have applied DOE for selecting suitable parameters in manufacturing industries (Tanco *et al.*, 2007) such as hot press forming process parameters (Kandar and Akil, 2016), laser cutting (Huehnlein *et al.*, 2010), laser surface processes (Sotnikov *et al.*, 2010), and micro injection molding processes (Packianather *et al.*, 2013). An experimental study of the geopolymer concrete process with glass fibers was undertaken by Rani and Anusha (2016).

The 2^k full factorial DOE was studied and used to find the response or compressive strength of geopolymer because 2^k factorial designs are especially useful in the early stage of experimental work when there are many

factors to be investigated. The DOE factors were the oven curing temperature and metakaolin to fly ash ratio. The first factor was divided into 2 levels, 30°C and 60°C. The second factor was divided into 3 levels, 0, 30, and 100. The experimental design needed 6 runs with 3 replicates, making a total of 18 experiments. The order of the experiment was randomized by using the “create factorial design” order in the Minitab Release 15 program. The experimental design and the results of the compressive strength in each set of the experiment are shown in Table 1.

Experimental Procedure

The experimental procedure is shown in Figure 1. The details are as follows:

Precursor Preparation

The LQFA was obtained from Siam Kraft Industry Co., Ltd., Bangkok, Thailand. To remove moisture and the impurities, the LQFA was oven-dried at 100°C for 24 h and was passed through a 120-mesh sieve. Calcined clay (CC) was prepared by calcining kaolinite clay (from

Table 1. The 2^k full factorial experimental design

StdOrder	RunOrder	Curing Temp.	Ratio of metakaolin to fly ash	Compressive Strength (MPa)
1	6	30	0	15.452
2	13	30	30	14.678
3	18	30	100	4.654
4	3	60	0	13.991
5	17	60	30	12.300
6	5	60	100	7.337
7	15	30	0	14.188
8	8	30	30	15.954
9	12	30	100	14.180
10	2	60	0	13.552
11	1	60	30	13.210
12	16	60	100	6.110
13	7	30	0	17.254
14	11	30	30	14.804
15	9	30	100	7.762
16	4	60	0	14.404
17	10	60	30	7.722
18	14	60	100	8.543

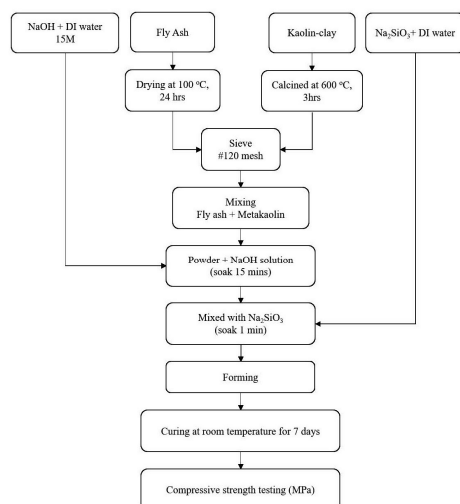


Figure 1. Experimental procedure

Ranong province, Thailand) at 600°C for 3 h. After that, the CC was passed through a 120-mesh sieve.

Alkali Solution Preparation

Sodium hydroxide (NaOH) pellets and distilled water were mixed to obtain a concentration of 15 M, and then allowed to cool down at room temperature. A sodium silicate (Na_2SiO_3) solution was mixed with distilled water to reduce the viscosity. The used ratio of Na_2SiO_3 to distilled water is 17:3 by weight.

Sample Preparation

To obtain enough information for analysis by the DOE technique, the interval of 20% of

fly ash replaced by metakaolin was designed. The LQFA and CC were blended together in the LQFA:CC ratios of 100:0, 90:10, 70:30, 50:50, 30:70, and 0:100 by weight.

The blended powder was mixed with the NaOH solution (15 M) for 15 min, and then added to the Na_2SiO_3 solution and mixed for 1 min. The used ratio of NaOH: Na_2SiO_3 was 1:2 by weight. After that, the paste was poured into a 50 mm × 50 mm × 50 mm steel mold and compacted as described in ASTM C109 (ASTM, 2002). The samples were sealed with a vinyl sheet to prevent moisture evaporation before curing at room temperature for 7 days.

Characterization Techniques

The compressive strength value was measured by using a compression test machine (a universal testing machine).

Results and Discussion

The data obtained from the experiment were analyzed using the Minitab Release 15 program. A factor with a p-value <0.05 was considered statistically significant with a 95% confidence interval. The ANOVA test showed that the most significant factor was represented by the metakaolin to fly ash ratio. The table of the ANOVA test is shown in Table 2.

The results from main effect plot and interaction plot are shown in Figure 2. The interaction plot represented that the 30°C curing temperature achieved a better result of compressive strength than the 60°C curing temperature. For the main effect plot, the 30°C curing temperature obtained a better result of

Table 2. Analysis of variance for compressive strength, using adjusted SS for tests

Source	DF	Seq. SS	Adjust SS	Adjust MS	F	P
Curing Temp.	1	26.298	26.298	26.298	4.29	0.061
Ratio of metakaolin to fly ash	2	146.049	146.049	73.024	11.91	0.001
Curing Temp.*Ratio of metakaolin to fly ash	2	6.139	6.139	3.070	0.50	0.618
Error	12	73.560	73.560	6.130		
Total	7	252.047				

compressive strength than the 60°C curing temperature. Moreover, the interaction plot represented that the 30°C curing temperature is appropriate for further use in the experiment.

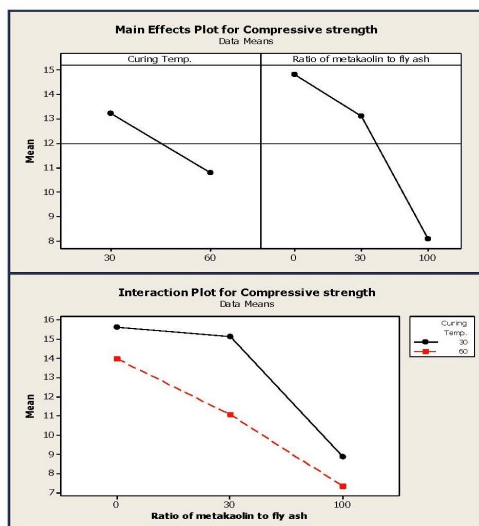


Figure 2. The main effect plot and interaction plot for compressive strength

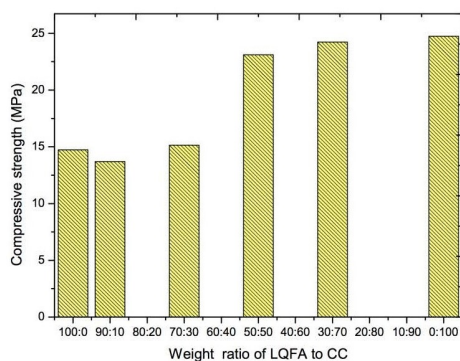


Figure 3. Compressive strength of the geopolymers with different ratios of LQFA/CC

The results of the further testing at the curing temperature of 30°C with different percentages of the metakaolin to fly ash ratio are shown in Figure 3. The most outstanding result was with the 70% of metakaolin to fly ash ratio. Its compressive strength is around 25 MPa which is higher than the minimum

requirement for PC of 19 MPa in ASTM Standard C150 (ASTM, 2003). Compared with PC, the developed geopolymer in the present work has a lower production cost and is more environmentally friendly.

The cost of the developed geopolymer mainly depends on the amount of the metakaolin used due to metakaolin's required energy for synthesis. Although, the 70% of metakaolin to fly ash ratio has a high compressive strength, the 50% of metakaolin to fly ash ratio is the optimized ratio because its compressive strength meets the minimum requirement of PC and it has a lower production cost than the 70% of metakaolin to fly ash ratio.

Conclusions

In this study, a sustainable cement replacement material, i. e., geopolymer, was developed by using low quality fly ash blended with calcined clay as a precursor. Design of experiment (DOE) was used to determine the significant factors which have an effect on the compressive strength of the geopolymer. The DOE factors were the oven curing temperature and metakaolin to fly ash ratio. The results of the compressive strength of the oven curing temperature at 30°C were better than at the 60°C temperature. By applying analysis of variance (ANOVA), these factors have been optimized to determine the compressive strength. The results of the significant factors indicated that the most important factor was the metakaolin to fly ash ratio. Moreover, this study further tested oven curing at a temperature of 30°C with different metakaolin to fly ash ratios: 0%, 10%, 30%, 50%, 70%, and 100%. The most outstanding result was the 70% of metakaolin to fly ash ratio.

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