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Effects of Parameters on the Setting Time of Fly Ash Based Geopolymers Using Taguchi Method

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Abstract

Geopolymers are the inorganic polymer materials possessing properties superior than the properties of conventional materials. Geopolymers are also environment friendly materials. This paper determines the effects of parameters such as silica to alumina (Si/Al) ratio, sodium to alumina (Na/Al) ratio, water to solid (W/S) ratio, and temperature on the setting of fly ash geopolymers. The experiments were designed using Taguchi model. The setting time was determined by Vicat Needle apparatus. The Si/Al - 2.2, Na/Al - 1.4, W/S - 0.30, and temperature of 40 °C increased the setting time and workability of the geopolymers. The Si/Al ratio of 1.8, Na/Al ratio of 1.0, W/S ratio of 0.20, and temperature of 80 ° C caused fast setting of geopolymers. The setting time of geopolymers can be optimized using Taguchi method for particular applications.

Keywords: Fly ash, Geopolymer, Setting time, Parameters, Taguchi method.

1. Introduction

Geopolymers are a class of alkali activated aluminosilicate materials produced by alkali activation of aluminosilicate source materials. Metakaolin, fly ash, and slag as aluminosilicate materials and sodium hydroxide and potassium hydroxide as alkali solutions and/or sodium and potassium silicates as source of silica are used for synthesis of geopolymers. Fly ash is a by-product of coal thermal power plants and it contains appreciable amounts of silica and alumina. The use of fly ash in synthesis of geopolymers results in the benefits of saving of the land reserved for its disposal as well as the economic benefits by the sale of by-product for the synthesis of valuable products [1]. Geopolymers are being used in a variety of applications due to their properties better than the properties of conventional materials. Fast setting and attaining high compressive strength, resistance against fire and acids, and low CO_2 production are the main properties of geopolymers.

Setting time of geopolymers is associated with its workability. It is the time available for transportation, placing, and compaction. Setting of geopolymers is linked with the initial formation of sodium aluminosilicate hydrate gel (NASH) [2, 3] while formation of

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Nomenclature

FAGP	Fly Ash Geopolymers	Na/Al	Sodium to alumina
W/S	Water to solid	Si/Al	Silica to alumina
CSH	Calcium Silicate Hydrate	CAH	Calcium Aluminate Hydrate

this gel at later stages is involved in strength development. Setting time of geopolymers depends on several factors such as composition of the alkaline solution, alkaline solution to fly ash ratio [4], silica modulus, and Na_2O content etc. Several studies on setting time of geopolymers have been conducted. Karakoc et al [5] determined the effect of silica modulus and Na_2O concentrations on setting of ferrochrome slag (FS) and found that lower the silica modulus in the paste higher the setting time. The increasing of the Na_2O content also decreased the initial and final setting times. Chindaprasirt et al [6] determined the effect of SiO_2 and Al_2O_3 on high calcium fly ash based geopolymers and observed that increasing SiO_2 and Al_2O_3 contents decreased the setting time. Silva et al [2] determined the effect of SiO_2/Al_2O_3 on metakaolin based geopolymers and noticed that increasing this ratio from 2.5 to 5.01 increased the setting time. Hanjitsuwan et al [7] determined the effect of NaOH concentrations on the setting of geopolymers and observed that increasing NaOH concentration from 8 – 18 M increased the initial and final setting times of the paste. All of the above studies determined the effects of different parameters on the setting of geopolymers by varying only one parameter at a time. However, no such study has been conducted in which all the parameters would have been varied at the same time. The experimental design technique such as Taguchi method provides this facility to determine the effects of different parameters by varying all the parameters at a time and also provides guidelines for analysis of the data. The application of Taguchi model to geopolymers is limited to only a few studies [8-11]. Olivia and Nikraz [8] used Taguchi method to optimize the mechanical properties and durability of fly ash geopolymer concrete. Riahi et al [9] used Taguchi method to determine the effects of parameters on the compressive strength of fly ash based geopolymers. Nazari et al [10] used Taguchi method to determine the effects of parameters on the compressive strength of geopolymers produced using Portland cement as a source of aluminosilicate material.

This study determines the effects of parameters such as silica to alumina (Si/Al) ratio, sodium to alumina (Na/Al) ratio, water to solid (W/S) ratio, and temperature on setting time using Taguchi method. The setting time of geopolymers was determined using Vicat Needle. The setting time data was analyzed using response index based on signal to noise ratio (S/N) ratio principle [12] developed by Taguchi model.

2. Materials and methods

2.1. Materials

Fly ash (obtained from local thermal power plant) was used for synthesis of geopolymers. Fly ash contains Al_2O_3 - 43.25%, SiO_2 - 20.58%, Fe_2O_3 - 12.41, and CaO - 11.11%. Detailed composition of fly ash is given in another paper [13]. AR grade NaOH and Na_2SiO_3 (SiO_2 - 37.79% and Na_2O - 16.36%) purchased from R & M Chemicals, Malaysia were used for synthesis of geopolymers.

2.2. Methods

2.2.1. Designing of experiments

Taguchi experimental design method was used for designing of experiments. Four parameters such as silica to alumina (Si/Al) ratio, sodium to alumina (Na/Al) ratio, water to solid (W/S) ratio, and temperature were selected for this study. The parameters and their levels are shown in Table.1. The ranges and levels of the parameters were selected in a way to represent the best possible compositions based on the presented compositions in the literature [14-18]. We used one of the orthogonal arrays such as L9 (4^3) developed by Taguchi method to represent all the factor or parameter levels. Using Taguchi design expert software a total of 9 trials or samples were obtained. Chemical compositions of the samples are shown in Table.2.

Table.1: Factors and levels

Factors	Level 1	Level 2	Level 3
Na/Al ratio	1	1.2	1.4
Si/Al ratio	1.8	2.2	2.6

W/S ratio	0.20	0.25	0.30
Curing Temperature	40 °C	60 °C	80 °C

Table.2. Suggested experimental series by Taguchi method

Trail Number	Sample Name	Si/Al	Na/Al	W/S	Temp.° C
T1	FAGP-1	1.8	1	0.20	40
T2	FAGP-2	1.8	1.2	0.25	60
T3	FAGP-3	1.8	1.4	0.30	80
T4	FAGP-4	2.2	1	0.25	80
T5	FAGP-5	2.2	1.2	0.30	40
T6	FAGP-6	2.2	1.4	0.20	60
T7	FAGP-7	2.6	1	0.30	60
T8	FAGP-8	2.6	1.2	0.20	80
T9	FAGP-9	2.6	1.4	0.25	40

2.2.2. Geopolymer synthesis:

Alkaline solution was prepared by dissolving sodium hydroxide pellets in distilled water and gently mixing the solution until the pellets completely dissolved. The solution was left for 2 hours at room temperature to cool to normal temperature. The sodium silicate was in solution form and used whenever required. The geopolymer samples were prepared following the design of mixtures suggested by Taguchi method (Table 2). The alkaline solution was poured into the fly ash and both were mixed for 8 minutes using a hand mixer. The geopolymer paste was poured into the mould and put in an electric oven at the desired temperature for curing. Geopolymer samples were cured at three different temperatures of 40 ˚C, 60 ˚C, and 80 ˚C.

2.2.3. Setting Time

The Vicat Needle ASTM C187/C191, BS 4550 model NL 3012X/ 002 was used for determining the setting time. The setting time measurement of geopolymer samples started after 15 minutes of curing the samples in the oven and continued with an interval of every 10 minutes up to the final setting time. Initial setting time is the time at which the penetration of needle is less than 39.5 mm while the final setting time is the time at which the penetration of needle is less than 0.5 mm.

The data obtained from the setting time was analyzed using response index principle based on signal to noise (S/N) ratio. The signal to noise (S/N) ratio is the performance of the experiment in terms of power. The higher the signal to noise ratio the better the result. The effect of different levels of the synthesis parameters on the setting time was determined by averaging the setting time of the geopolymer samples which contain that level of the parameter to be analyzed. For example, the response index for setting time at Si/Al ratio of 1.8 was calculated by first determining the signal to noise (S/N) ratio of the samples which possess Si/Al ratio of 1.8 (FAGP-1, FAGP-2, and FAGP-3) and then the response index at Si/Al ratio of 1.8 was calculated by averaging the signal to noise ratios (S/N) of the samples at Si/Al ratio of 1.8. This result is represented as response index at Si/Al ratio of 1.8.

3. Results and Discussion:

3.1. Effect of parameters on setting time

The final setting time results were analyzed using response index principle. The effects of parameters on setting time are discussed in the following section.

3.1.1. Effect of Si/Al ratio

The effect of Si/Al ratios on response index (setting time) is shown in Fig.1 (a). The response index increases by increasing the Si/Al ratio from level 1 to level 2 (1.8 to 2.2) and further increasing this ratio to level 3 (2.6) the response index decreases. The higher response index was obtained at Si/Al ratio of 2.2 (level 2).

The Si/Al ratio of 1.8 causes low geopolymerization reaction due to lower amount of silica available for the reaction and geopolymerization takes less time to complete its major portion of dissolution reaction resulting in the lower setting time of geopolymer. Increasing the Si/Al ratio from 1.8 to 2.2 increases the setting time due to high amount of soluble silica. The presence of high amount of soluble silica in the alkaline solution enhances the geopolymerization and the reaction takes more time to set and harden increasing the setting time of geopolymer. Further increasing the Si/Al ratio from 2.2 to 2.6 decreases the setting time of geopolymer due to higher content of silica. Increasing the Si/Al ratio increases the silica and Na_2O contents and both of these compounds enhance the geopolymerization reaction and it takes less time to complete its major portion of dissolution reaction before setting of the paste causing compaction of the paste very quickly resulting in the decrease of setting time [19]. The higher amount of silica also decreases the geopolymer formation due to very fast compaction of the paste resulting in the decrease of geopolymer synthesis.

3.1.2. Effect of Na/Al ratio

The effect of Na/Al ratios on the response index (setting time) is shown in Fig.1 (b). The response index increases by increasing the Na/Al ratio from level 1 to level 3 (1.0 to 1.40). The higher response index was obtained at Na/Al ratio of 1.4.

The Na/Al ratio is related with the concentration of sodium hydroxide (NaOH). The lower the Na/Al ratio the lower the concentration of NaOH is and vice versa. The Na/Al ratio of 1.0 and 1.2 causes less dissolution of fly ash due to lower content of NaOH resulting in lower geopolymerization reaction and the paste solidifies quickly causing decrease of setting time. At low NaOH concentration, the dissolution of Ca^{2+} in the solution was not hindered which results in the formation of calcium silicate hydrate (CSH) and the calcium aluminate hydrate (CAH) gels, which causes faster setting of the paste and setting time reduced [6, 20]. Increasing the Na/Al ratio to 1.4 increases the NaOH concentration which causes enhancement in dissolution of fly ash. The Na/Al ratio of 1.4 causes higher dissolution of fly ash releasing more silica and alumina and less calcium in the solution which causes enhancement in geopolymerization or gel formation and the reaction takes more time to complete the early stage reaction before setting of the paste resulting in the increase of setting time [6, 20].

3.1.3. Effect of W/S ratio

The effect of water to solid (W/S) ratio is shown in Fig.1(c). The response index (setting time) increases by increasing the water to solid ratio from level 1 to level 3 (0.20 to 0.30). Higher response index was obtained at W/S ratio of 0.3.

Water plays a very important role in geopolymerization reaction and it takes part in dissolution of aluminosilicate and polycondensation of geopolymers [21]. The lower water to solid ratio of 0.20 and 0.25 may not be sufficient for dissolution of fly ash resulting in the rapid setting of the paste. By increasing the water to solid ratio from 0.20 to 0.30, the dissolution of fly ash increases due to higher water content resulting in slow setting of the paste. Therefore, the water to solid ratio is related to the dissolution of fly ash and the workability of the paste and both of these properties affects the setting of geopolymers.

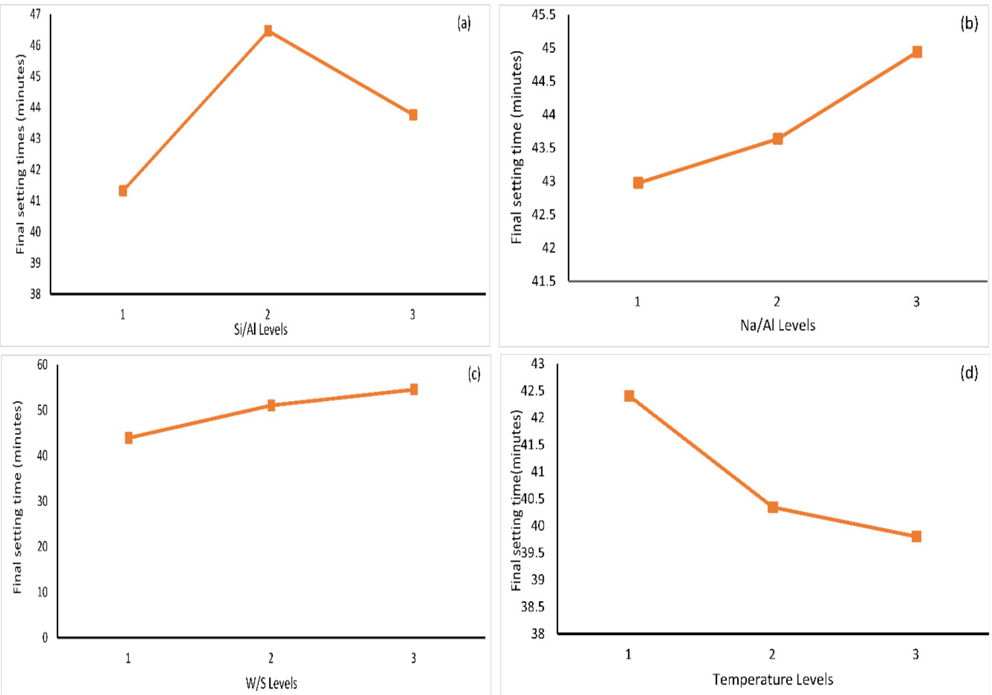

Figure 1: Effect of parameters on final setting time, (a) Si/Al ratio, (b) Na/Al ratio, (c) W/S ratio, and (d) and temperature

3.1.4. Effect of temperature

The effect of temperature on the response index (setting time) is shown in Fig.1 (d). The response index decreases by increasing the temperature from 40 °C to 80 °C (level 1 to level 3). The higher response index was obtained at the temperature of 40 °C.

At lower temperature of 40 °C, the solubility of aluminosilicate is less which causes slow geopolymerization reaction resulting in slow setting of the paste. At higher temperatures of 60 °C and 80 °C (level 2 and level 3), the geopolymerization reaction is accelerated due to higher temperature producing higher amounts of silica and alumina to participate in the geopolymerization resulting in the rapid setting of the paste.

During the setting period, major dissolution of fly ash takes place due to the presence of alkali and silicate solutions. These dissolved species present in the mix are needed for nucleation, gel formation, and growth [22]. After setting of the paste, dissolution of the precursors is replaced by polycondensation, which causes the release of water from the solidified gel matrix. Though it is believed that dissolution and polycondensation occur simultaneously but after setting of geopolymer paste, major reaction shifts from dissolution to polycondensation and dissolution also occurs but in a less prominent way.

The overall effect of parameters on final setting time is shown in Fig.2. Water to solid (W/S) ratio has the highest effect while Na/Al ratio has the lowest effect on final setting time.

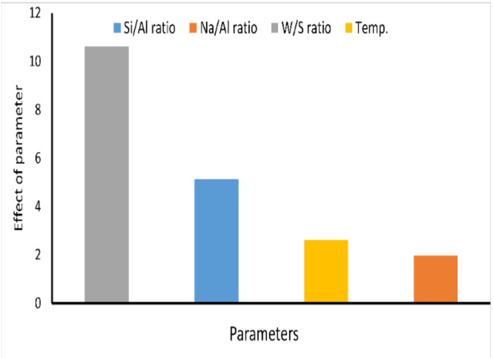

Figure 2: Overall effect of parameters on final setting time

4. Conclusions

Effects of parameters on the setting time designed by Taguchi method were successfully determined. The Si/Al ratio of 2.2, Na/Al ratio of 1.4, W/S ratio of 0.30, and temperature of 40 °C produced higher setting time geopolymers. The Si/Al ratio of 1.8, Na/Al ratio of 1, W/S ratio of 0.20 and curing temperature of 80 ˚C produced fast setting geopolymers. The overall effect of W/S ratio was higher while Na/Al ratio was lower on the setting of geopolymer.

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