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Correlation of the Processing Parameters in the Formation of Granulated Ground Blast Furnace Slag Geopolymer

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Abstract: Geopolymers are inorganic materials with huge potential applications including building material, fire resistant materials, and agricultural construction materials. Various parameters influenced the final properties of these geopolymer concretes. This study developed the effects of several factors such as solid-to-liquid ratio, NaOH concentration, and Na2SiO3/NaOH ratio on the compressive strength of granulated ground blast furnace slag (GGBFS) by statistical design of experiment (DOE) approach. Analysis of the experimental results through ANOVA exhibited that the specimen with NaOH concentration of 10M, Na2SiO3/NaOH ratio equals to 2.5, and solid-to-liquid ratio of 3.0 curing at room temperatures for 28 days was potential of highest strength (168.705 MPa) in the considered procedure. Besides, the relationship between compressive strength and influential factors could be suitably by fraction factorial design method.

1. Introduction

Cement concrete is the main material used worldwide in building construction in the world due to the durability in various environment and appropriate mechanical properties. Contrary, the Portland cement clinker production is an energy- intensive process. Thus, the development of geopolymer materials is addressed by researchers as an alternative replacement to Portland cement [1-3]. The term 'Geopolymer' has been coined by Davidovits in 1970s, symbolizes the inorganic aluminosilicate based on geological materials which reacted with an alkaline solution to form binder through polycondensation reaction [4]. Geopolymer is an amorphous aluminosilicate structure consisting three-dimensional Si-O-Al polymeric network produced by the poly-condensation of the individual $(SiO)^{4-}$ and $(AlO_4)^{5-}$ tetrathedrals.

Geopolymer system can be established into two types of binding system which are silica-aluminum (Si+Al) with medium to high alkaline solution and silica-calcium (Si+Ca) with a mild alkaline solution [5]. Class F fly ash and metakaolin were the source material used in the (Si+Al) binding system due to silica and alumina content as the main composition. Meanwhile, ground granulated blast furnace slag (GGBFS) was used in the (Si+Ca) system due to the higher content of silica and calcium

in the composition. There are many potential benefits of using GGBFS as aluminosilicate sources material in geopolymer process include excellent mechanical strength [6], fast setting time [7], great thermal behavior [8], and fire resistant materials [9]. Practically, the mixture of sodium hydroxide (NaOH) and sodium silicate (Na_2SiO_3) are often used as an alkaline solution. The sodium silicate was acted as alkali activator, plasticizer, and binder [10]. Meanwhile, the alkali hydroxide appropriate for the dissolution of aluminosilicates sources [11]. In addition, alkaline solutions also promoted a certain quantity Si and Al atoms to dissolve the aluminosilicates sources, form monomers in solutions, and then polycondense to form rigid network [12]. Sodium silicate much preferable activating solutions owing to its soluble content, which leads to enhance the rate of polymerization reaction [13]. The properties of geopolymer are affected by the composition of raw materials, the type and the relative amount of alkali activator during the initial period of the geopolymerization reaction [13]. The solid-to-liquid (S/L) ratio and Na_2SiO_3/NaOH ratio have exceptional effects on mechanical properties [14]. Besides, these parameters affect the workability of the geopolymer slurry [15].

Design of Experiment (DOE) is a systematic approach to investigate of a system or process. A series of structured tests are designed in which planned changes are made to the input variables of a system or process. The effects of these changes on a pre-defined output are then assessed statistically. Subsequently, one of the outcomes of this statistical analysis is the ability to predict the optimum combination and level of variables in an experiment by examination of the response of a relatively small number of samples. Considering all the parameters in a single study may not be achievable. Thus, an appropriate experiment design can help in understanding the effect of various parameters at the same time by the minimum required works is performed in this study. The DOE studies were carried out to determine the effect of NaOH concentration, solid-to-liquid ratio, and Na_2SiO_3/NaOH ratio on the compressive strength of GGBFS. The factors were varied according to the fraction factorial design, devised using MINITAB. In addition, the results were analyzed by ANOVA (Analysis of Variance) to evaluate the compressive strength of GGBFS as a function of the above mentioned factors. The P-value was used to determine whether the results are statistically significant.

2. Experimental

2.1. Material
In this study, the fineness of particle size of GGBFS is below 150 µm. The raw materials of GGBFS are drying in an oven at 100°C in order to evaporate the moisture content. The GGBFS are supplied by Ann Joo Integrated Steel Sdn. Bhd, Penang, Malaysia. Table 1 shows the chemical composition of the GGBFS as determined by X-Ray Fluorescence (XRF) analysis.

Table 1. Chemical composition of GGBFS (wt.%).

Compound	CaO	SiO_2	Al_2O_3	MgO	Fe_2O_3	TiO_2	ZrO_2	MnO_2
Wt %	50.37	30.4	10.5	3.2	0.53	0.98	0.05	0.71

NaOH in pellet form with 99% purity, and Na_2SiO_3 consists of Na_2O = 9.4%, SiO_2 = 30.1% and H_2O = 60.5% with weight ratio SiO_2/Na_2O = 3.20-3.30 and specific gravity at 20°C = 1.4 gm/cc, were used as a mixture of alkaline activator.

2.2. Sample preparation and testing
Experiment design and analysis was approach by Minitab 17. A factorial design was used to obtain required experimental procedures. The acceptance ranges for parameters are listed in table 2 [16-18] and 25 suggested experiments in table 3. To study the effect of the NaOH concentration, solid-to-liquid ratio, Na_2SiO_3/NaOH ratio on the compressive strength of the GGBFS geopolymer samples, sodium silicate, and sodium hydroxide were introduced in various amounts into GGBFS. The design

included 24 model points and 1 additional center points. Total of 25 series of GGBFS geopolymer samples with mixture was prepared. The mixture of geopolymer was cast in acrylic plastic cube molds (50mm x 50mm x 50mm). The molds sealed with plastic wrap and all samples cured in room temperature for 24 hours before undergo analysis.

Table 2. Range for each factor in factorial design (low level indicates the lowest range factor in this studies and high level for the optimum strength achieved).

Factors	Low	High
NaOH concentration (M)	6	10
Solid-to-liquid ratio	1.0	3.0
Na_2SiO_3/NaOH ratio	1.5	2.5

Compressive test was performed according to ASTM C109. Three samples of each formulation were tested and the average data were obtained. A universal Instron machine series 5569 was used for the measurement. The loading was displacement controlled at a constant rate of 0.5mm/min for all the tests. Then, to generate the model with the optimum statistical significance, ANOVA and prediction and were used.

Table 3. Full factorial design method for three factors (NaOH ratio, solid-to-liquid ratio, and Na_2SiO_3/NaOH ratio) affecting compressive strength of GGBFS.

Run Order	NaOH	S/L	Na_2SiO_3/NaOH (SS/NaOH)	Compressive strength (MPa)
1	6	1	1.5	47.9
2	10	1	1.5	31.8
3	6	3	1.5	109.3
4	10	3	1.5	80.4
5	6	1	2.5	60.9
6	10	1	2.5	53.1
7	6	3	2.5	109.5
8	10	3	2.5	161.7
9	6	1	1.5	34.8
10	10	1	1.5	31.6
11	6	3	1.5	115.4
12	10	3	1.5	97.5
13	6	1	2.5	61.4
14	10	1	2.5	65.3
15	6	3	2.5	115.4
16	10	3	2.5	168.7
17	6	1	1.5	60.9
18	10	1	1.5	37.1
19	6	3	1.5	103.3
20	10	3	1.5	76.8
21	6	1	2.5	62.3
22	10	1	2.5	62.2
23	6	3	2.5	103.3
24	10	3	2.5	155.3
25	8	2	2.0	101.9

3. Result and Discussion

Table 4 showed the overall main affects for GGBFS compressive strength found to be statistically significant as indicated by the F-value of 141.31 where the corresponding P-value of 0.00 as the result obtained from Minitab. The solid-to-liquid ratio exhibited highest positive effect on compressive strength, respectively. Contrary, NaOH concentration gives the lowest effect on compressive strength. Moreover, as the P-value for NaOH concentration is 0.391, it will give less effect (3.08) on the compressive strength of GGBFS as response in these studies. Hence, with the P-value unequal to 0.00 gives no significant effects on the compressive strength of GGBFS.

Statistically it can be observed that all interaction (three way and two-way) was found to be significant effect on the compressive strength of GGBFS geopolymer due to the P-value equal to 0.00 and the effect value is higher (1533.3 and 4259.7) compared to others one-way interaction factors. Meanwhile, parametric study of the combination factors affecting the GGBFS geopolymer compressive strength are strongly recommended first before conducting further experimental related to mechanical properties and other application of the geopolymer concrete [19, 20]. Furthermore, this statistical study generated the information which enhanced the manufacturing of GGBFS properties those benefits for further researches.

**Table 4.** ANOVA values of the models for compressive strength.

Source	Degree of freedom	Effect	Adjusted mean square	F-value	P-value	Significance
Main effect	3	31051.4	10350.5	141.31	0.000	Yes
NaOH	1	3.08	56.8	0.78	0.391	No
S/L	1	65.61	25826.9	352.6	0.000	Yes
SS/NaOH	1	29.35	5167.7	70.55	0.000	Yes
2-way interaction	3	4259.7	1419.9	19.39	0.000	Yes
NaOH*S/L	1	10.96	720.7	9.84	0.006	Yes
NaOH*SS/NaOH	1	22.48	3031.4	41.39	0.000	Yes
S/L*SS/NaOH	1	9.20	507.6	6.93	0.017	Yes
3-way interaction	1	1533.3	1533.3	20.93	0.000	Yes
NaOH*S/L*SS/NaOH	1	15.99	1533.3	20.93	0.000	Yes
Residual error	17	1245.2	73.2	-	-	-
Curvature	1	321.1	321.1	5.56	0.031	No
Pure error	16	924.1	57.8	-	-	-

3.1. The effect of various parameters

Main effects of the three factors include: NaOH concentration, solid-to-liquid ratio, $Na_2SiO_3/NaOH$ ratio are shown in Figure 1. The solid-to-liquid ratio (S/L) has a huge effect than NaOH concentration and $Na_2SiO_3/NaOH$ ratio. The length of solid-to-liquid ratio from the low level (1.0) to the high level (3.0) was immense compared to NaOH concentration and $Na_2SiO_3/NaOH$ ratio effects. Thus, this proved that the solid-to-liquid ratio had a significant effect on the compressive strength of GGBFS.

In figure 2, the Pareto chart exhibited all factors; solid-to-liquid (S/L) ratio and $Na_2SiO_3/NaOH$ (SS/NaOH) ratio except the concentration of NaOH factors alone have significant effect on the compressive strength of GGBFS geopolymer. All factors except NaOH concentration extends beyond the reference lines (2.11) demonstrated potentially important to the compressive strength of GGBFS as shown in figure 2 [21]. Following are the interpretation of the main effect plot for GGBFS compressive strength:

i. Solid to Liquid ratio (S/L)	:	Runs with 3.0 S/L ratio had higher compressive strength than runs at 1.0 S/L ratio
ii. Concentration of NaOH (NaOH)	:	Runs at higher and lower concentration of NaOH had comparable compressive strength of GGBS geopolymer.
iii. Na_2SiO_3/NaOH ratio (SS/NaOH)	:	Runs at higher and lower SS/NaOH ratio had comparable compressive strength of GGBS geopolymer.
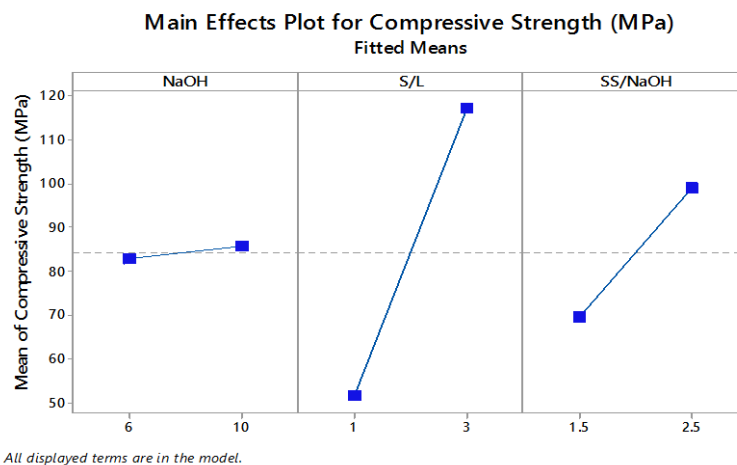
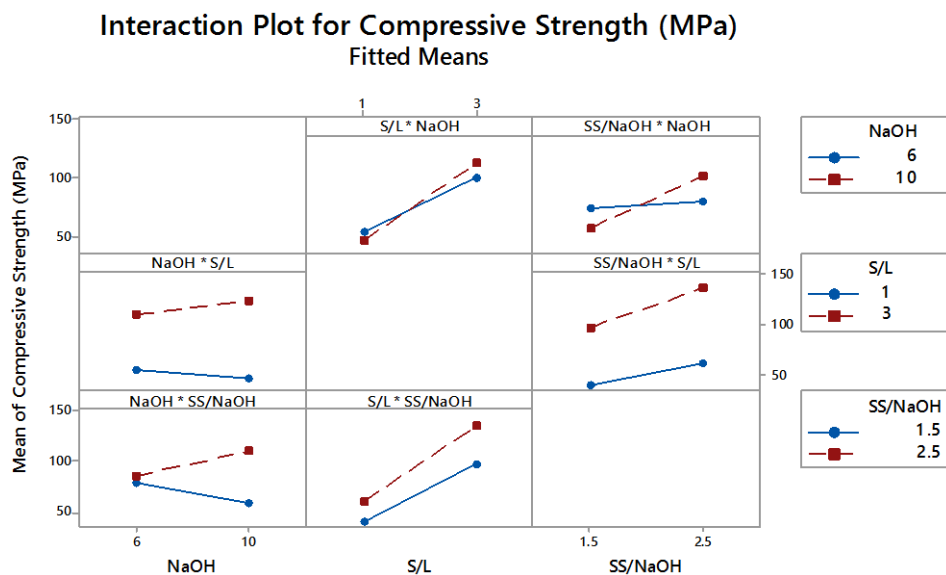**Figure 1.** Main effect plots for compressive strength of GGBS geopolymer.
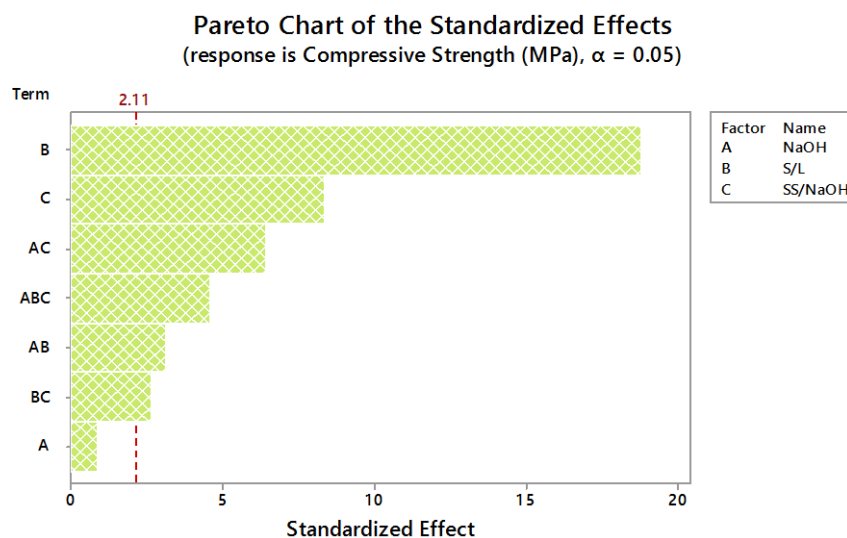
Figure 2. The effects of concentration of NaOH, solid to liquid ratio, activator ratio, on compressive strength of GGBFS geopolymer.

The interaction plot exhibited the relationship between one categorical factor and continuous response depends on the value of the second categorical factors.

The interactions between the combinations factors on compressive strength for GGBFS geopolymer are illustrated in figure 3. The line in the interaction plot represent the interaction affected the relationship between the factors and the response. In addition, the more nonparallel the lines are, the greater the strength of the interaction [22].

This interaction effect indicated that the relationship between NaOH concentration and compressive strength of GGBFS depends on the value of SS/NaOH ratio. The SS/NaOH ratio is equal to 1.5 and the NaOH concentration at 6M is associated with the highest mean compressive strength of GGBFS. However, when the SS/NaOH ratio is equal to 2.5 and the NaOH concentration at 10M is associated with the highest mean compressive strength. Thus, the combination (SS/NaOH ratio*NaOH) has a significant effect compared to other combination of factors due to nonparallel lines in the interaction plots as obtained in figure 3.

Additionally, increasing the sodium silicate to alkaline solution ratio has enhanced the compressive strength of the geopolymer [23]. Sodium silicate promotes the compressive strength of GGBFS based geopolymer paste in two ways; (1) the sodium silicate solution improves the dissolution rate of Si and Al; (2) then, the Al-O bonds are weaker than Si-O bonds in raw material, Al dissolve rapidly in alkali solution [2]. Thus, if Si ion is provided prior to being available through dissolution of raw material, it can improve the degree of geopolymerization and enhanced the mechanical properties [24]. Otherwise, the dissolution and formation of Si-O-Si oligomers as a strong bond can be enhance due to a simultaneous increase in Na and Si ions [25]. Besides, the hydration degree of GGBFS increase due to ability of OH- ions from NaOH solution then create intermediary reactants which last precipitate into C-S-H gel and other final product [26].

Figure 3. Interaction plots for compressive strength of GGBS geopolymer.

3.2. Prediction of compressive strength

The model equation for the GGBFS geopolymer can be used to mathematically estimate the compressive strength as the response for any combination of factor levels within the design space. The equation predicts the compressive strength of GGBFS based on the concentration of NaOH, solid-to-liquid ratio, alkaline activator ratio and other combination factors (NaOH*S/L, NaOH*SS/NaOH, S/L*NaOH, S/L*NaOH). Thus, in order the get the optimum compressive strength of GGBFS by using the fixed S/L ratio and alkaline activator ratio, the concentration of NaOH can be predict. The model represents the data of the present study are written as follows:

Compressive strength   =   -41.5 + 4.78 (NaOH) + 120.4 (S/L) + 48.9 (SS/NaOH) -13.25 (NaOH*S/L) – 4.75 (NaOH*SS/NaOH) - 54.7 (S/L*NaOH) + 7.99 (c)

Where; NaOH = concentration of NaOH, S/L = solid-to-liquid ratio, SS/NaOH = alkaline activator ratio.

The equation gives the benefits to evaluate the manufacturing of GGBFS compressive strength in order required minimum experiment. Also, by using this prediction equation, the researchers can focus on the further application such as fire resistant material by using the GGBFS as the raw precursor material

4. Conclusions

In this study, the effect of three parameters, namely, NaOH concentration, solid-to-liquid (S/L) ratio, and Na_2SiO_3/NaOH ratio on compressive strength was investigated. In summary, the following summarized the most important outputs derived statistically from DOE analysis on GGBFS geopolymer.

i.   Increasing the concentration of NaOH from 6M to 10M leads to an increase in compressive strength of GGBFS geopolymer.
ii. The concentration of NaOH at 10M, solid-to-liquid ratio (S/L) of 3.0, and Na_2SiO_3/NaOH ratio equal to 2.5 exhibited an optimum compressive strength of GGBFS geopolymer of 168.7 MPa.
iii. ANOVA data analysis obtained that ratio of the solid-to-liquid (S/L) was the highest influenced than NaOH concentration and Na_2SiO_3/NaOH ratio.

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