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Effect of Processing Parameters on Compressive Strength of Metakaolinite Based Geopolymers: Using DOE Approach

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Abstract

Geopolymers are inorganic materials with a wide variety of potential applications such as fire resistant materials, building materials, cements and concrete, high-tech composites aircraft interior, and etc. Various parameters affect the properties of these materials. This article describes the effects of several factors such as molar ratios of SiO2/Al2O3, Na2O/SiO2, and H2O/Na2O and curing temperature on the compressive strength of metakaolinite based geopolymer bodies (MGBs) by statistical design of experiments (DOE) approach. It was found that the above-mentioned agents play an important role in MGBs' properties. Also, a model to present the relationship between compressive strength and influential factors was obtained.

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Keywords: geopolymers; inorganic material; metakaolonite; compressive strength; design of experiment.

Nomenclature

A Temperature
B Molar ratio of SiO_2/Al_2O_3
C Molar ratio of $Na2O/SiO_2$
D Molar ratio of H_2O/Na_2O
R Response for compressive strength
X_c The actual values

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X_r	The coded values
a	The average of upper and lower actual values of the factors
b	The difference of upper and lower actual values of the factors

1. Introduction

Inorganic polymers (geopolymers) are novel X-ray amorphous and are a group of alkali-activated materials revealing superior engineering properties compared to Portland cement, Kani and Allahverdi (2009). These materials have low apparent porosity and nano size pores, Roy (1999). The geopolymerisation is a reaction between amorphous silica and alumina rich solids with a high alkaline solution to form amorphous to semi-crystalline aluminosilicate inorganic polymers, Verdolotti et al. (2008). The development of geopolymer concrete is an important step towards the production of environmentally friendly concretes because the global cement industry contributes about 7% of the total man-made greenhouse gas emissions to the earth's atmosphere, Djwantoro Hardjito et al. (2004). Generally, the advantages of geopolymers over ordinary Portland cement may include: (a) remarkably less CO2 production; (b) longer life and better durability; (c) better defense against chemical attack (e.g. chlorides, sulfates); (d) rapid strength gain; (e) better performance in marine environments; and (f) use of industrial waste, Moseson et al. (2012).

A variety of systematic methods have been used to understand and control performance of geopolymer, but much about these materials is still not well understood. The properties of geopolymers depend on parameters such as the chemical and composition of the raw material, its dissolution properties, the composition and amount of alkali activator, and the curing conditions. Among these parameters, the chemical composition of both raw material and activating solution is playing an important role in geopolymer properties, Davidovits (1991), Duxson et al. (2007), Steveson and Sagoe-Crentsil (2005).

Considering all of the parameters in a single work may not be possible. However, a suitable experiment design can help to study the effects of more parameters at the same time by the minimum required experiments. This work presents the effect of molar ratio of SiO_2/Al_2O_3, Na_2O/SiO_2, H_2O/Na_2O and curing temperature on the compressive strength of metakaolinite based geopolymer bodies (MGBs) using a full factorial experiment design. Finally, the results were analyzed by ANOVA (ANalysis Of VAriance) to evaluate the compressive strength of MGBs as a function of the above mentioned parameters.

2. Experimental

2.1. Materials

In this work, metakaolinite obtained from calcining kaolin at 750^{oC} for 24 h. Table 1 shows the chemical composition of the metakaolinite, as determined by X-Ray Fluorescence (XRF) analysis. Sodium hydroxide in flake form (NaOH with 98% purity), and sodium silicate solution (Na_2O = 8.2%, SiO_2 = 27% and water = 64.8% by mass), were used as the alkaline regents. Deionized water was used throughout the experiments.

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

SiO_2	Al_2O_3	Fe_2O_3	CaO	Na_2O	K_2O	MgO	TiO_2	MnO	P_2O_5	S	L.O.I
53.68	42.68	0.34	0.12	0.02	0.12	0.08	1.395	0.009	0.020	0.011	1.32

2.2. Sample preparation and testing

Experiment design and analysis was performed on Minitab 16. A full factorial design was used to determine required experimental procedures. The accepted ranges for parameters are listed in table 2, Davidovits (2008), Van Jaarsveld et al. (2002) and 19 suggested experiments in table 3. To study the effect of the SiO_2/Al_2O_3, Na_2O/SiO_2 and H_2O/Na_2O molar ratios on the compressive strength of the geopolymer samples, sodium silicate, sodium hydroxide and deionized water were introduced in different amounts into the metakaolinite. The design included 16 model points and 3 additional center points. Totally 19 series of geopolymer samples with the mixture proportion illustrated in table 3 were prepared. The mixtures were cast in polyethylene cylinders molds (The ratio of height to diameter was 2) and vibrated for 20 min to remove entrapped air. The molds sealed with the lead then all samples cured at different temperatures according to table 3 for 24h.

Table 2. The introduced range for each factor in full factorial design.

Factors	Low	High
Temperature (*℃*)	55	85
SiO_2/Al_2O_3	2.90	4.40
Na_2O/SiO_2	0.20	0.30
H_2O/Na_2O	13.75	21.25

Compressive tests were performed according ASTM C39. Three samples of each formulation were tested and the average data were reported. A universal test machine was used for the measurements. The loading was displacement-controlled at a constant rate of 0.5 mm/min for all the tests. Finally, to generate the model with the highest statistical significance, ANOVA and other diagnostic tools were used.

Table 3. Suggested tests from full factorial method for four factors and compressive strength.

Run Order	Center point	T (**℃**)	SiO_2/Al_2O_3	Na_2O/SiO_2	H_2O/Na_2O	compressive strength (*MPa*)
1	1	55	2.90	0.20	13.75	53.24
2	1	85	2.90	0.20	13.75	65.86
3	1	55	4.40	0.20	13.75	33.37
4	1	85	4.40	0.20	13.75	4.46
5	1	55	2.90	0.30	13.75	22.49
6	1	85	2.90	0.30	13.75	15.15
7	1	55	4.40	0.30	13.75	24.89
8	1	85	4.40	0.30	13.75	20.29
9	1	55	2.90	0.20	21.25	9.52
10	1	85	2.90	0.20	21.25	9.59
11	1	55	4.40	0.20	21.25	0.00
12	1	85	4.40	0.20	21.25	0.00
13	1	55	2.90	0.30	21.25	0.94
14	1	85	2.90	0.30	21.25	1.39
15	1	55	4.40	0.30	21.25	0.66
16	1	85	4.40	0.30	21.25	0.41
17	0	70	3.65	0.25	17.50	9.90
18	0	70	3.65	0.25	17.50	9.91
19	0	70	3.65	0.25	17.50	10.33

3. Results and discussion

The results of compressive strength, Effect index and p value are shown in table 3 and 4. As it is shown in table 4, the molar ratio of H2O/Na2O and the interaction of SiO2/Al2O3 and Na2O/Si2O molar ratios have the highest negative and positive effect on compressive strength, respectively. The right hand side column of table 4 shows the P value of each term. The p value in fact shows the degree of effectiveness of each term. Each factor with a p value less than 0.05 is considered as an effective factor and those with p values larger than 0.05 are regarded as ineffective ones.

Table 4. Effect and p- value of the terms.

Term	Effect	P	Term	Effect	P
Constant		0.000	SiO_2/Al_2O_3*Na_2O/SiO_2	13.33	0.000
T	-3.49	0.001	SiO_2/Al_2O_3*H_2O/Na_2O	6.67	0.000
SiO_2/Al_2O_3	-11.76	0.000	Na_2O/SiO_2*H_2O/Na_2O	7.30	0.000
Na_2O/SiO_2	-11.23	0.000	T*SiO_2/Al_2O_3*Na_2O/SiO_2	5.45	0.000
H_2O/Na_2O	-27.15	0.000	T*SiO_2/Al_2O_3*H_2O/Na_2O	4.75	0.000
T*SiO_2/Al_2O_3	-4.94	0.000	SiO_2/Al_2O_3*Na_2O/SiO_2*H_2O/Na_2O	-8.87	0.000
T*H_2O/Na_2O	3.56	0.001	T*SiO_2/Al_2O_3*Na_2O/SiO_2*H_2O/Na_2O	-5.61	0.000

3.1. The effect of processing parameters

Main effects of the four factors include: curing temperature and molar ratios of SiO_2/Al_2O_3, Na_2O/SiO_2 and H_2O/Na_2O are shown in fig. 1. As it is shown in figure, by increasing all of the mentioned factors, mean of the compressive strength decreased and no significant effect was observed with regard to the curing temperatures. The same behaviour can be seen in table 4.

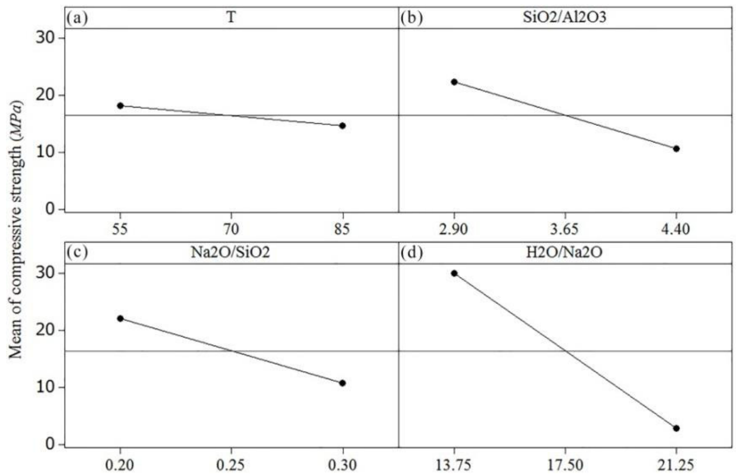

Fig. 1. Main Effects plot for mean Compressive Strength for (a) Temperature; (b) SiO_2/Al_2O_3; (c) Na_2O/SiO_2; and (d) H_2O/Na_2O.

Figure 1a shows that temperature has low effect on compressive strength which relates to the selected temperature range. This is due to the fact that this is a short range and hence does not have a significant effect on compressive strength. According to Fig. 1b and 1c it can be concluded that molar ratios of SiO_2/ Al_2O_3 and Na_2O/SiO_2 have negative effect on Compressive strength. While these two molar ratios increase, the system will be saturated with such ions as Na+, which will prevent the formation of Oligomers and subsequently the geopolymerisation process, Khale and Chaudhary (2007). Fig. 1d shows that molar ratio of H2O/ Na2O has significant negative effect on strength. The negative effect relates to high porosity introduced into the system due to water withdrawal. The high effect of this ratio can be due to the prevention of alumina silicate from dissolving, which will result in reduction of geopolymerisation and prevention of forming strong bonds.

3.2. Modelling of compressive strength

One of the outstanding advantages of DOE plans compared to the conventional planning is that DOE reveals the interaction effects of the influential parameters. Effect index of interactions of parameters are presented in table 4. As mentioned before, the interaction of molar ratios of SiO_2/ Al_2O_3 and Na_2O/SiO_2 has the highest positive effect on compressive strength. This result can be attributed to a simultaneous increase in Na and Si ions, which will result in an increase in dissolution and formation of Si-O-Si Oligomers that is considered as a strong bond, Al Bakri et al. (2012).

In should be mentioned, if a factor individually has no significant effect on the response but causes a meaningful interaction on conjunction with other factors, it should be considered in the model, regardless of its individual ineffectiveness, Barrentine (1999). The model which represents the data of the present study can be written in its coded state as follows:

$$R(MPa) = -1.75T - 5.88B - 5.61C - 13.58D - 2.47A.B + 1.78A.D + 6.67B.C + 3.34B.D + 3.65C.D + 2.73A.B.C +$$

$$2.38A.B.D - 4.43B.C.D - 2.81A.B.C.D + 16.39$$

The R-Sq. value of the above model is 99.96 which indicate a very high accuracy (table 4). For converting the actual values to their corresponding coded ones, the following relationship can be used, Cleraux et al. (2012).

$$X_c = (X_r - a)/b$$

Where a is the average of upper and lower actual values of any factor and b is the difference of these two values.

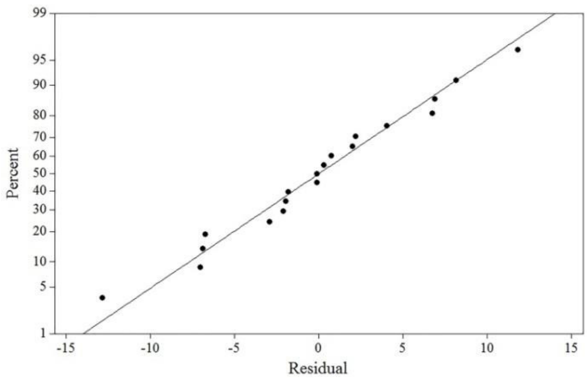

Fig. 2. Normal probability plot of the residuals for compressive strength.

After the model has been obtained, it is necessary to evaluate its statistical reliability. From statistical viewpoint, one condition has to be satisfied by a model for its reliability becomes confirmed. This condition is normalized residuals. The term residual means the difference between an experimentally measured value and that obtained by the model. As seen in Fig. 2, residual values of all experiments of the present work locate on a straight line which is an indication of residual normality. Since the condition of model reliability has been satisfied by the model, its reliability is confirmed.

4. Conclusions

In this study, the effect of four parameters, namely, curing temperature, molar ratios of SiO_2/Al_2O_3, H_2O/Na_2O and H_2O/SiO_2 on compressive strength was investigated. According to the obtained results, all the above-mentioned parameters have a negative effect on compressive strength among which, temperature and molar ratio of H_2O/Na_2O have the lowest and highest effect on compressive strength, respectively. By means of analysis of variance, a linear model which shows the relationship between compressive strength four parameters was obtained, that was statistically confirmed.

References

Al Bakri, A.M.M., Kamarudin, H., Abdulkareem, O. a. K. a., Ruzaidi, C.M., Rafiza, A.R., Norazian, M.N., 2012. Optimization of Alkaline Activator/Fly ASH Ratio on the Compressive Strength of Manufacturing Fly ASH-BASED Geopolymer. Appl. Mech. Mater. 110-116, 734–739.

Barrentine, L. B., 1999. An introduction to design of experiments : a simplified approach.

Cleraux, C., Croarkin, C., Filliben, J., Guthrie, W., Heckert, A., Reeder, M., Ryan, T., Splett, J., Zhang, N.-F., 2012. NIST/SEMATECH e-Handbook of Statistical Methods.

Davidovits, J., 1991. Geopolymers. J. Therm. Anal. 37, 1633–1656.

Davidovits, J., 2008. Geopolymer : chemistry and applications, 2nd ed. Institute geopolymer, Saint-Quentin.

Djwantoro Hardjito, Steenie E Wallah, Dody M.J. Sumajouw, B.V Rangan, 2004. Factors Influencing the Compressive Strength of Fly Ash-Based Geopolymer Concrete. Civ. Eng. Dimens. 6, 88–93.

Duxson, P., Mallicoat, S.W., Lukey, G.C., Kriven, W.M., van Deventer, J.S.J., 2007. The effect of alkali and Si/Al ratio on the development of mechanical properties of metakaolin-based geopolymers. Colloids Surfaces A Physicochem. Eng. Asp. 292, 8–20.

Kani, E.N., Allahverdi, A., 2009. Effects of curing time and temperature on strength development of inorganic polymeric binder based on natural pozzolan. J. Mater. Sci. 44, 3088–3097.

Khale, D., Chaudhary, R., 2007. Mechanism of geopolymerization and factors influencing its development: A review. J. Mater. Sci. 42, 729–746.

Moseson, A.J., Moseson, D.E., Barsoum, M.W., 2012. High volume limestone alkali-activated cement developed by design of experiment. Cem. Concr. Compos. 34, 328–336.

Roy, D.M., 1999. Alkali-activated cements: Opportunities and challenges. Cem. Concr. Res. 29, 249–254.

Steveson, M., Sagoe-Crentsil, K., 2005. Relationships between composition, structure and strength of inorganic polymers : PPPart 2 Fly ash-derived inorganic polymers. J. Mater. Sci. 40, 4247–4259.

Van Jaarsveld, J.G.S., van Deventer, J.S.J., Lukey, G.C., 2002. The effect of composition and temperature on the properties of fly ash- and kaolinite-based geopolymers. Chem. Eng. J. 89, 63–73.

Verdolotti, L., Iannace, S., Lavorgna, M., Lamanna, R., 2008. Geopolymerization reaction to consolidate incoherent pozzolanic soil. J. Mater. Sci. 43, 865–873.