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The study of preparation of fly ash geopolymer based on optimization for response surface methodology

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ABSTRACT

In order to solve the optimization problem of parameters and conditions in fly ash geopolymer preparation effectively, a series of research experiments were designed and conducted in the paper using response surface methodology (RSM). Firstly, Single factor gradient analysis was adopted to determine the reasonable level of various factors in the response surface analysis, the paper investigated the 28-day compressive strength development with water glass modulus, NaOH content, water /fly ash ratio, curing temperature and curing time. Secondly, the preparation conditions were optimized to improve the 28-day compressive strength of the materials based on the single factor analysis by using RSM, and high strength geopolymer materials was prepared by this method. The study also proved the effectiveness of RSM to optimize the preparation conditions of geopolymer through validated test.

KEYWORDS

Response surface methodology; Fly ash; Geopolymer; Strength; Optimization design.

INTRODUCTION

Geopolymer has not only excellent composite properties but also lower energy consumption and pollution during its production compare with the cement. Especially geopolymer discharges no or only a small amount of CO_2 emissions in basic production process[1]. Therefore, geopolymer is considered to be a very promising green material in the 21st Century[2].

Relevant scholars have done a lot of work on research for preparation conditions of geopolymer and many valuable research results were obtained. The literature[3-5] studied the influence of Na_2SiO_3/NaOH ratio on material properties, literature[6] studied the effect of NaOH concentration on material properties, literature[7] adopted water glass modulus to reflect the composition of alkaline activator. Different influential factors such as liquid/solid ratio[4],[6], Liquid/fly ash ratio[5],[6],[8],and FA/clay ratio[5] on the strength development of geopolymer were studied. In the aspect of material curing regime, literature[6-7], [9-11] studied influence of curing regime including curing temperature, curing humidity, curing time and curing methods on the formation process and the influence of the comprehensive performance of geopolymer. Preparation of geopolymer cured at ambient temperature has been studied by different groups of authors recently[12],[13].

The most of the experimental design methods above-mentioned were through single-variable or simple multi-factor combination experiment and orthogonal design to find out the optimum value. These researches only focus on a few isolated and scattered points and unable to find the optimal preparation conditions within the entire value region of influencing factors and the interaction among various factors cannot be analyzed by these methods. RSM is to use the results of the experimental data obtained by scientific experiment design to conduct regression and establish the continuous response surface model which can reflect the relationship between the factors and the response value. Within the range of independent variable values, RSM can find the optimal combination and achieve the purpose of optimizing the experimental program or product formulation. RSM has advantages of a relatively small number of test groups, reducing costs, improving efficiency and so on, it is widely used in biological, chemical, machinery, agriculture, etc.. But the application of RSM is unusual in the field of civil engineering particularly in the research of building materials preparation conditions. The paper uses RSM to design experiments, and establishes the regression model between the influence parameters and the strength of materials. The paper gets the materials optimal preparation conditions by optimization analysis, and verifies the optimization results through the verification test.

MATERIALS AND METHODS

Materials

Fly ash of Fuxin power plant was used in this study, its specific gravity was 2.12g/cm^3 and its chemical composition is shown in TABLE1. The analytical grade NaOH was in pellet form with 96% purity, the commercial water glass was in liquid form with 38% concentration, modulus of 3.4, 8.97% Na_2O and 29.16% SiO_2. ISO standard sand was adopted as fine aggregate.

TABLE 1 : Chemical composition of fly ash (%)

SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	K_2O	Loss
53.75	29.37	5.64	3.68	1.08	1.29	0.68	3.58

Experiment methods

Compressive strength test of geopolymer

The compressive strength test referred to GB/ T17671-1999, the test materials according to the preliminary mix proportions design were mixed fully in a cement mortar mixer before casting in 40mm×40mm×160mm steel moulds, then the specimens were vibrated to remove the air bubbles, and cured in predesigned conditions. After that, the specimens were demoulded and stored at standard curing box until the age of 28 days, and then compressive strength tests were performed.

Univariate analysis

In order to optimize the preparation conditions effectively by using RSM, experimental points area should include the best experimental conditions, so univariate analysis must be done to determine the reasonable affecting factors and experimental value interval firstly. combining of the existing research results, water glass modulus, NaOH content (take account of fly ash mass percentage, the same below), water /fly ash ratio, curing temperature and curing time were chose as examine factors, this study conducted univariate analysis by using the index of 28-day compressive strength of geopolymer.

Response surface experiment design

Experiments were designed in the paper using Box-Behnken Design(BBD) based on univariate analysis, Design-Expert software V8.0.6.1 was used to make response surface regression analysis for data of experiment results. Functional relationship was fitted using a secondary polynomial.

RESULTS AND DISCUSSION

Single factor effect analysis

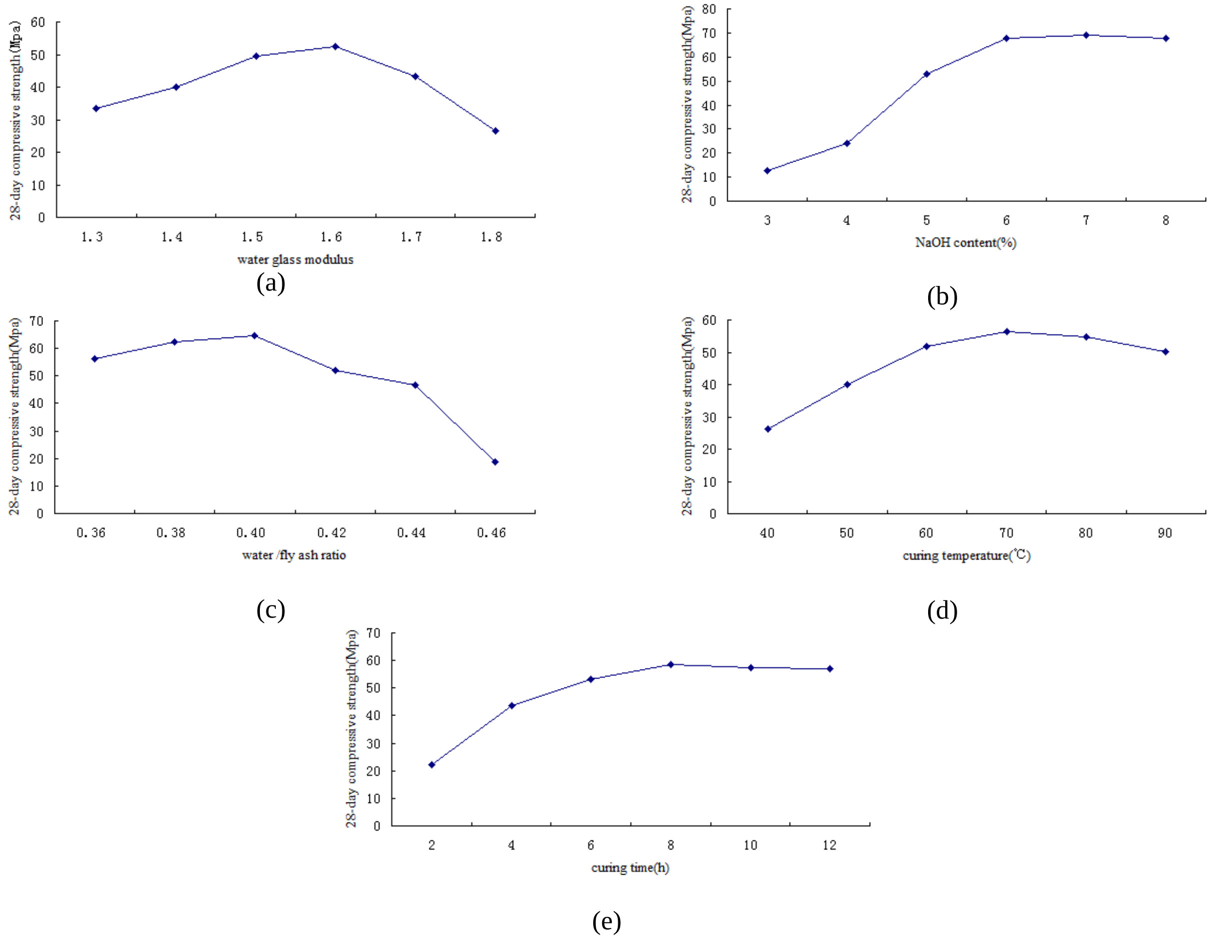


Figure 1 : Influence regular of changes of various factors in the 28d compressive strength

The influence of water glass modulus

The preparation conditions were set for curing temperature is 60℃, curing time is 6h, NaOH content is 5%, water /fly ash ratio is 0.42, the modulus of water glass is1.3,1.4,1.5,1.6,1.7,1.8, respectively. 28-day compressive strength variation is shown in Figure 1 (a), the 28-day compressive strength degree increases at first and then decreases with the water glass modulus increasing, strength reaches the maximum value when the modulus of water glass is 1.6, the strength was significantly decreased when modulus of water glass is more than 1.7. The large number of low silicate polymers contained in water glass liquid plays an important role in promoting the aluminum silicate polymerization reaction. Low silicate polymers concentration is relatively low when the modulus of water glass is low, therefore, geopolymer strength is low. Concentration of low silicate polymers in solution is improved after water glass modulus increases, which can promote the polymerization reaction and the strength of geopolymer is also increased. But, if the modulus of water glass is too large, silicate in solution is mainly for the occurrence as high polymers form which causes the proportion of low silicate polymers in solution decrease, so strength of geopolymer decreases obviously.

The influence of NaOH content

The preparation conditions were set for water glass modulus is1.6, curing temperature is 60℃,curing time is 6h, water /fly ash ratio is 0.42, NaOH content is 3%,4%,5%,6%,7%,8%, respectively. 28-day compressive strength variation is shown in Figure 1 (b). NaOH can provide large amounts of OH^-, which can make the paste in strong alkaline environment and glass phase composition of fly ash dissolve, then resulting in the dissolution of active silicon-aluminium ingredient in fly ash and lead to formation of various aluminosilicate oligomers, then polycondensation reacts to form three-dimensional network aluminosilicate gels that making materials with high strength. It is shown in Figure 1 (b) that strength of geopolymer is low with the less dosage of NaOH, dissolution rate of fly ash will be increased with the increase of NaOH dosage, and then strength of geopolymer increased obviously. But when NaOH content is too large, liquidity of paste decreases obviously, and the paste appears quick- setting phenomenon, which causes vibrating difficultly when pouring, and compactness descending because of pore increasing in the block, in turn, results in the decrease of strength.

The Influence of water /fly ash ratio

The preparation conditions were set for water glass modulus is1.6, curing temperature is 60℃, curing time is 6h, NaOH content is 5%, the water /fly ash ratio is 0.36, 0.38, 0.4, 0.42, 0.44, 0.46, respectively. 28-day compressive strength variation is shown in Figure 1 (c). If the water /fly ash ratio is too small, the paste will be quite dry or even looks like powder without fluidity which causes vibrating difficultly, so the strength decreases. After improvement of water /fly ash ratio, the fluidity of materials, the degree of compaction casting in mould and homogeneity of the material improves evidently. But the exaggerated fluidity, low speed of condensing and phenomenon of eduction, bleeding in paste appear when the water /fly ash ratio become too high, numbers of holes left after water evaporation which lead to much tiny cracks when block under loads and then breaking finally, so strength increases at first and then decreases with the water /fly ash ratio increasing, and the strength decreases significantly when the water /fly ash ratio is more than 0.4.

The influence of curing temperature

The preparation conditions were set for water glass modulus is1.6, curing time is 6h, NaOH content is 5%, water /fly ash ratio is 0.42, the curing temperature is 40℃, 50℃, 60℃, 70℃, 80℃, 90℃, respectively. 28-day compressive strength variation is shown in Figure 1 (d). It can be seen that increasing curing temperature has a positive effect on increasing the strength, the 28-day compressive strength gradually increased with increasing of temperature. But the strength decreased when the temperature is too high, the reason is that when the temperature is too high, water of mixtures evaporates too fast and resulting in its reaction medium is weakened, at the same time reactant on surface of fly ash particles generates rapidly which hinder the further reaction.

The influence of curing time

The preparation conditions were set for water glass modulus is1.6, curing temperature is 60℃, NaOH content is 5%, water /fly ash ratio is 0.42, curing time is 2h, 3h, 6h, 8h, 10h, 12h, respectively. 28-day compressive strength variation is shown in Figure1(e). When the curing time is 2 hours, the excitation effect has not developed, strength is the lowest. When the curing time is 4 or 6 hours, the effect of potential thermal activation has played out, the strength increases obviously. When the curing time is 8 hours later, the alkali activated reaction of paste has entered a relatively gentle stage.

Experimental results and analysis of the response surface

The 28-day compressive strength variation is shown in single factor analysis above-mentioned, the strength reaches the maximum value when the modulus of water glass is 1.6, so take water glass modulus from 1.5 to 1.7 as the scope of response surface analysis. 28-day compressive strength gradually increased with increasing of NaOH content on the whole, but the paste liquidity reduced significantly when NaOH content is too much, so take NaOH content from 5% to 7% as the scope of response surface analysis. 28 day compressive strength degree reaches the maximum value when water /fly ash ratio is 0.4, so take water /fly ash ratio from 0.38 to 0.42 as the scope of response surface analysis. As for curing regime, although strength gradually increased with increasing of temperature, the strength kept within a certain range when temperature is 60-90℃ except slight lower in 40℃ or 50 ℃, the extent of change is limited, the strength increases with the curing time increasing also, but when the curing time is 8 hours later, the strength increases slowly. Therefore, through comparative analysis and considering the factor of energy consumption, the curing temperature keeps 60℃ and the curing time keeps 8h in the follow-up study.

Consequently, water glass modulus, NaOH content and water /fly ash ratio are chosen as influence factors, and response surface analysis tests of three-factor-three-level are designed using Box-Behnken Design(BBD) method. The response variable (*Y*) represents 28-day compressive strength. Factors and code levels of RSM are shown in TABLE 2, experimental design obtained by Design-Expert software V8.0.6.1 and experimental results are shown in TABLE 3.

TABLE 2 : Factors and code levels of RSM

Factor	Code	Levels of code		
		-1	0	1
Water glass modulus	X_1	1.5	1.6	1.7
NaOH content	X_2	5	6	7
Water /fly ash ratio	X_3	0.38	0.4	0.42

Design-Expert software V8.0.6.1 is used to make secondary polynomial regression for data in TABLE 3, relationship between the response variable (*Y*), representing 28-day compressive strength, and influence factors such as X_1,X_2,X_3 was fitted using a second-order regression equation given as

$$\begin{aligned} Y = & -2646.073 + 1026.618X_1 + 64.819X_2 + 8589.625X_3 + 14.125X_1X_2 \\ & + 222.5X_1X_3 - 102X_2X_3 - 381.025X_1^2 - 3.135X_2^2 - 10606.875X_3^2 \end{aligned}, \quad (1)$$

TABLE 3 : Experimental design of RSM

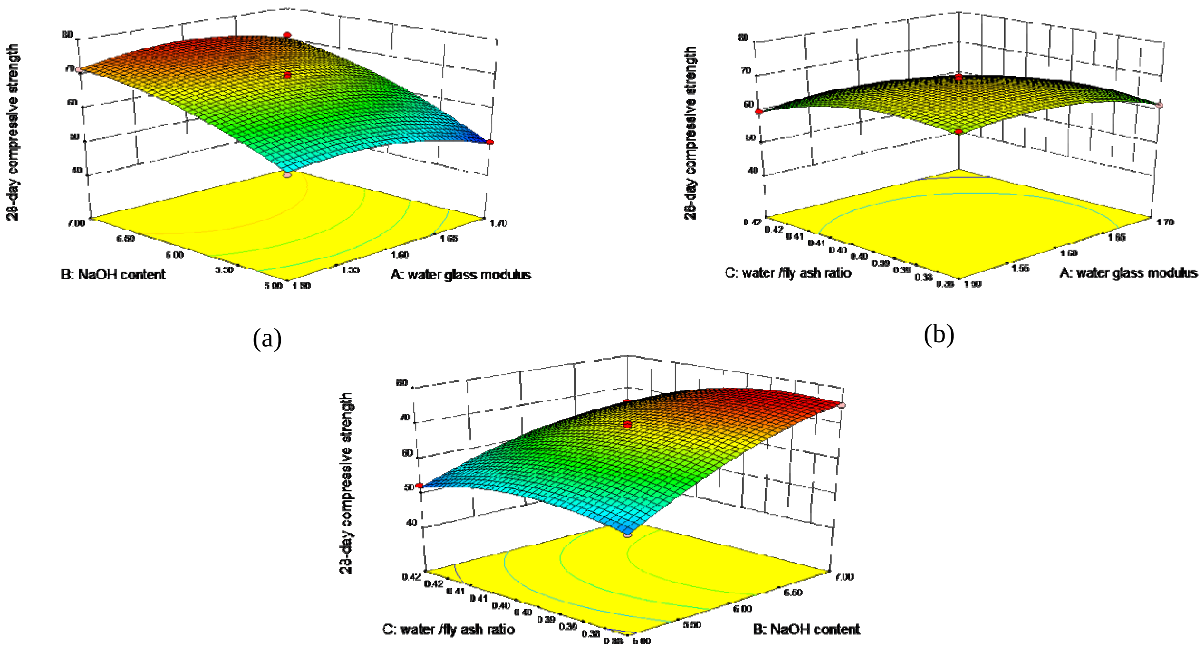
Test number	Design of tests			28-day compressive strength (MPa)	Test number	Design of tests			28-day compressive strength (MPa)
	water glass modulus	NaOH content (%)	water /fly ash ratio			water glass modulus	NaOH content (%)	water /fly ash ratio	
1	1.5	5	0.4	55.65	10	1.6	7	0.38	75.15
2	1.7	5	0.4	50.11	11	1.6	5	0.42	52.35
3	1.5	7	0.4	71.35	12	1.6	7	0.42	65.73
4	1.7	7	0.4	71.46	13	1.6	6	0.4	69.63
5	1.5	6	0.38	67.31	14	1.6	6	0.4	70.18
6	1.7	6	0.38	61.57	15	1.6	6	0.4	69.39
7	1.5	6	0.42	59.61	16	1.6	6	0.4	68.26
8	1.7	6	0.42	55.65	17	1.6	6	0.4	67.98
9	1.6	5	0.38	53.61	-	-	-	-	-

ANOVA and analysis of interaction

TABLE 4 : ANOVA of the model

Source	Freedom	Sum of squares	Mean square	F value	P value
Model	9	974.15	108.24	102.15	< 0.0001
X_1	1	28.61	28.61	27.00	0.0013
X_2	1	647.46	647.46	611.03	< 0.0001
X_3	1	73.81	73.81	69.66	< 0.0001
X_1X_2	1	7.98	7.98	7.53	0.0287
X_1X_3	1	0.79	0.79	0.75	0.4159
X_2X_3	1	16.65	16.65	15.71	0.0054
X_1^2	1	61.13	61.13	57.69	0.0001
X_2^2	1	41.39	41.39	39.06	0.0004
X_3^2	1	75.79	75.79	71.53	< 0.0001
Residual	7	7.42	1.06	-	-
Lack of Fit	3	3.93	1.31	1.50	0.3429
Pure Error	4	3.49	0.87	-	-
Cor Total	16	981.57	-	-	-

It can be seen from Analysis of variance(ANOVA) in TABLE 4, the response surface regression model reached highly significant level(P<0.0001), lack of fit item is not significant(P=0.3429>0.05). It is show that the response surface model fitting well with actual situation. It can also be seen from TABLE 4, the factors in the study have a significant influence on 28-day compressive strength, and the order of the influence is $X_2>X_3>X_1$,Interactive item X_1X_2 is significant, interactive item X_2X_3 is highly significant, interactive item X_1X_3 is not significant, It is shown that influence of interaction of water glass modulus and NaOH content and interaction of NaOH content and water /fly ash ratio on 28-day compressive strength are obvious, and influence of interaction of water glass modulus and water /fly ash ratio on 28-day compressive strength is not obvious. The degree of the influence of factors on 28-day compressive strength and interaction of factors were described in Figure 2 (a)-(c), The shape of 3D response surface and contour can reflect the degree of the interaction, 3D response surface in Figure 2 (a),(c) is steep, and contour is elliptical, it represents a significant interaction between two factors, 3D response surface in Figure 2 (b) is gentle, and contour is circular, it represents that there is no significant interaction between two factors, it is consistent with the results of variance analysis in TABLE 4.



(a) (b) (c)

Figure 2 : Interaction of factors on 28-day compressive strength

Determine the optimal preparation conditions and verification

Using automatic optimization function of Design-Expert software V8.0.6.1, the optimal values of the tested variables for the highest strength gain from calculation are water glass modulus 1.59, NaOH content 7%, water /fly ash ratio 0.39, curing temperature 60℃, and curing time 8h. Based on the optimum condition, the maximum strength is predict to be 76.5 MPa. This prediction was verified by three validation experiments under optimum condition, The mean value of 28-day compressive strength obtained is 77.35MPa, it is visible that the response surface model above has high accuracy and can predict the properties of geopolymer and optimize the preparation conditions better.

CONCLUSIONS

Through univariate analysis, the influence regulars of water glass modulus, NaOH content, water /fly ash ratio, curing temperature and curing time in 28-day compressive strength of geopolymer were studied. In order to study the preparation conditions of materials, experiments was designed using BBD-RSM and the experimental results were analyzed comprehensively. The order of the influence is NaOH content, water /fly ash ratio and water glass modulus, combined with experimental results, the 28-day compressive strength prediction model was established based on the RSM. The optimum condition were water glass modulus 1.59, NaOH content 7%, water /fly ash ratio 0.39, curing temperature 60℃, and curing time 8h. Based on the optimum condition, the maximum strength was predict to be 76.5 MPa, this prediction was verified by validation experiments, and geopolymer with 77.35 MPa was prepared and effectiveness of the optimization results of the response surface model was proved.

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