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Slag material's proportion optimised by polynomial regression

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Mixed activator agent for granulated blast furnace slag cementitious materials with potential hydraulic properties was prepared for tailings backfilling and optimised by the orthogonal test method and the Data Processing System analysis. The effects of lime, gypsum and cement clinker content on the 7 days compressive strength were investigated. Range and variance analysis, and quadratic polynomial step regression were used for analysing and optimising the formulation of the mixed activator agent. The optimised cementitious material contained 5% cement clinker, 15% lime and 4% gypsum. As a result, the maximum measured 7 days compressive strength was 2·79 MPa, which is 4·5 times higher than Portland cement (CEM I 32.5R). The difference between measured and predicted strength values was 4·5%, signifying good method applicability for slag cementing materials design.

1. Introduction

Resources and the environment are the two most important subjects that influence modern development. Each year, the mining industry generates significant amounts of mine tailings. The storage of these tailings requires large areas of land and leads to high environmental and ecological costs. In recent years backfill using cemented tailings has become increasingly popular not only for solving the tailings problems, but also for underground mining operations. Cement is used as a binding material for most tailings backfills, leading to increased mining cost and decreasing productivity; therefore, it is commonly used only in precious metals or non-ferrous mines (Benzaazoua and Marion, 2004; Benzaazoua *et al.*, 2002; Choi and Kim, 2009; Fall and Pokharel, 2010; Neto *et al.*, 2010; Zhang and Ahmaria, 2011). The low iron content of the ore causes the deficiency economic benefit, and choice of low-cost cementitious material as binding material for tailings backfills could enhance the economic performance of exploitation.

Granulated blast furnace slag (GBFS) is derived from the by-products of iron and steel processing, and is potentially hydraulic as its chemical composition is similar to Portland cement clinker (Lachemi *et al.*, 2010; Li *et al.*, 2002; Sadok *et al.*, 2011; Ye and Breugel, 2009; Zhao *et al.*, 2007). It was suggested that the cementitious material for tailings backfill referred to herein would be applied in some mines in Tang Shan, China. The Tang Shan area has many mineral resources including slag, gypsum and lime, and the prices of gypsum and lime are lower. Moreover, alkali and sulfate activation are the common processes in GBFS activation (Gong and Ye, 2009; Guneyisi and Gesoglu, 2008; Lin and Lin 2006; Yeau and Kim, 2005). Gypsum and lime were chosen as the activators of slag to be investigated in the present study.

The orthogonal experimental design is a popular approach for multi-factor and multi-level optimisation experiments. The feasibility of optimising the proportions of GBFS cementitious materials using the orthogonal experimental method has been demonstrated (Hu *et al.*, 2002; Nogales, 2006; Zhang *et al.*, 2008). The data generated from the orthogonal experimental design are usually evaluated with range and variance analyses, which cannot provide an exact relation between the composition of the cementitious material and its compressive strength. Quadratic polynomial step regression is, however, an advanced tool that is capable of utilising orthogonal experimental data to build a regression model, while avoiding instability in the regression coefficients due to multi-collinearity of the variables (Cheng *et al.*, 2005; Robeyst *et al.*, 2009; Wee *et al.*, 2000). Using quadratic polynomial step regression with the Data Processing System (Tang and Feng, 2007), all candidate materials would be analysed until the regression equation selects the significant variables. Moreover, the optimised

	CaO	MgO	SiO_2	Al_2O_3	Fe_2O_3	Cl^-	S_2^-	K_2O	Mn_2O_3	SO_3	Na_2O	IR*	LOI†
GBFS	38·16	10·10	33·35	16·23	0·62	0·03	0·17	–	0·44	0·07	–	0·56	0·27
Cement clinker	65·15	1·76	22·05	5·03	3·66	–	–	1·10	–	0·55	0·18	–	0·52
Mine tailings	5·44	3·11	73·58	8·70	6·19	–	–	–	–	–	–	–	2·98

*IR, insoluble residue; †LOI, loss of ignition.

Table 1. Cement clinker, GBFS and mine tailings chemical composition (wt.%)

cementitious material formula could be identified by fewer experiments and with increasing economic benefits for mining. Therefore, utilising the quadratic polynomial step regression to guide the experimental design has become a focus of modern cementitious materials development.

In the present study, lime and gypsum were used as the chemical activators of the mixed agent. In addition a small quantity of cement clinker was added as the chemical activator component to enhance the early strength of the GBFS cementitious materials for use as a substitute for cement in the tailings backfill. The orthogonal experiment was designed to optimise the proportions of the cementitious materials. The methods adopted to analyse the orthogonal experimental data were range analysis, analysis of variance and especially the combination of quadratic polynomial step regression with the Data Processing System. Using rational and effectiveness analysis, the regression equation was verified and a theoretical foundation for evaluating the proportions of the cementitious materials was defined.

2. Experimental details

2.1 Cement clinker

Cement clinker was provided by Jidong Cement Company, Ltd, Tang Shan, China. It was added to previously prepared cementitious materials, which had 7 and 28 days compressive strength of at least 28 and 58 MPa, respectively. The chemical composition of the cement clinker is given in Table 1.

2.2 Granulated blast furnace slag

The GBFS used in the experiments was supplied by Tanglong Materials Co., Ltd, Tang Shan, China. It was classified as S95 grade GBFS used in cement and concrete according to GB/T18046-2000 (CNS, 2000). Its Blaine specific surface was over 400 cm^2/g, and its 7 and 28 days activity coefficients were at least 75 and 95%, respectively. The chemical composition of the GBFS is given in Table 1.

2.3 Mine tailings

The mine tailings used for this study were obtained from Hebei Iron and Steel Co., Ltd, Shijiazhuang, China. Table 1 shows the chemical composition of the mine tailings and Figure 1 shows the particle size distribution curves. It can be seen that the tailings consisted mainly of silica and alumina with substantial amounts of calcium and iron.

2.4 Cement

Type 32.5R Portland cement was used for comparative tests. The cement met the requirements of the European Standard UNE-EN 196-1 (BSI, 2005).

2.5 Experimental programme

The $L_9(3^4)$ orthogonal array program was applied to obtain the optimal proportion of cementitious materials, for which the 7 days compressive strength was defined as the measurable indicator for verification of the feasibility of the proportions. The contents of cement clinker (*A*), lime (*B*) and gypsum (*C*) added were selected as the three necessary factors for

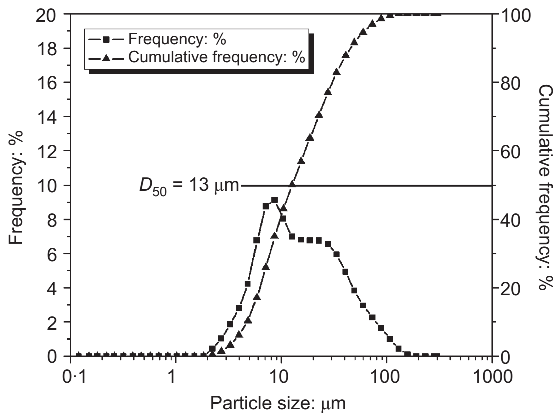

Figure 1. Particle size distribution of the mine tailings from Hebei iron and steel Co., Ltd, Shijiazhuang, China

proportion optimisation. The details of the factors and levels are presented in Table 2.

GBFS was homogenised with the activator, and mixed with tailings in the proportion 1 : 7. The mixture was blended with water so that the concentration of sand in the mortar reached 68%. It was poured into standard 70·7 mm × 70·7 mm × 70·7 mm steel moulds, and then all of the specimens were placed in the curing box with simulating standard conditions (20 ± 2℃ and 90 ± 5% relative humidity). After 24 h the specimens were removed from the moulds and continued curing. The 7 days compressive strength of the prepared cementitious materials was tested according to GB/T1767-1999 (CNS, 1999).

3. Results and discussion

3.1. Range analysis of orthogonal experimental results

Range analysis was employed to compare compressive strength among other factors. The results of the experiment and range analysis are shown in Table 3, where k_1, k_2 and k_3 express the average 7 days compressive strength at different factor levels. R represents the extreme difference of the 7 days compressive strength. Figure 2 presents the change of the 7 days compressive strength with factors and factor levels. It can be seen in Figure 2 that the amplitude of R variation in factor B was the largest among the three curves, whereas factor A was the smallest. The 7 days compressive strength gradually increased as the lime content changed from 5 to 10%, and from 10 to 15%, with the rate of the change between 10 and 15% being much higher than that between 5 and 10%. However, the 7 days compressive strength dropped to a minimum as the content of gypsum was increased from 4 to 8%, and further strength increase with higher gypsum content. The 7 days compressive strength also increased as the cement clinker content increased. As a result, for the 7 days compressive strength, the order of the effects of the three factors was: $B > C > A$.

Factors	Levels 1	2	3
A (cement clinker content)	0	0·025	0·05
B (lime content)	0·05	0·10	0·15
C (gypsum content)	0·04	0·08	0·12

Table 2. Factors and levels of the experiment

Experiment number	*A*	*B*	*C*	Compressive strength: MPa
1	1	1	1	1·38
2	1	2	2	2·30
3	1	3	3	2·27
4	2	1	2	1·02
5	2	2	3	2·2
6	2	3	1	2·76
7	3	1	3	1·94
8	3	2	1	2·65
9	3	3	2	2·36
k1	1·983	1·447	2·263	
k2	1·993	2·383	1·893	
k3	2·317	2·463	2·137	
Range	0·334	1·016	0·370	

Table 3. Response data from experiments and range analysis

3.2. Analysis of variance of orthogonal experimental results

Analysis of variance with the Data Processing System was conducted to analyse the orthogonal experiment results, since the range analysis cannot provide accurate significance factors for comparison with experimental results. It was found that the *F*-ratio of all factors was not significant, assuming the error was only produced in the blank column. This is because the degree of freedom was too small for the experimental error, while the threshold of the significant *F*-ratio was too high. In order to increase the sensitivity of the analysis, the *A* factor with small *F*-ratio was also hypothesised to generate experimental errors.

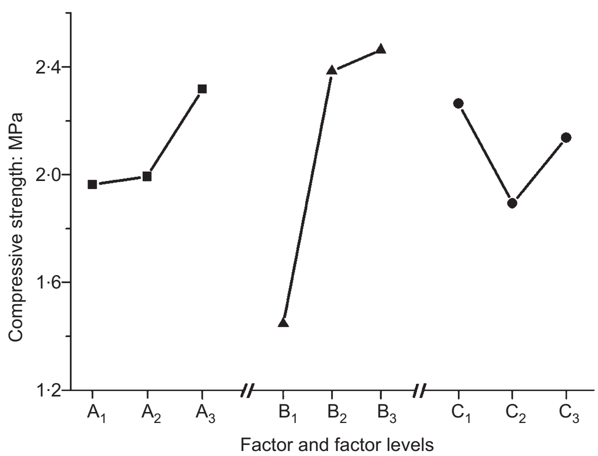

Figure 2. Range analysis results of the orthogonal experiment. The order of the raw material contents' effect is: lime (*B*) > gypsum (*C*) > cement clinker (*A*)

The analysis of variance results are listed in Table 4, which shows that the added content of lime (B) and gypsum (C) in GBFS cementitious material had significant effects on the compressive strength when their P-values were below 0·05. Usually, the inclusion of cement clinker improves the early strength of the GBFS cementitious material (Atis *et al.*, 2009), but the content of cement (A) did not play an important role in increasing early strength in the present experiments, as shown in Table 4. This could be explained by the addition of lime improving the early strength better than clinker. In conclusion, the optimal factors' assembly was $A_3B_3C_1$, obtained by connecting the analysis of variance and regular analysis.

3.3 Quadratic polynomial step regression with the Data Processing System

The quadratic polynomial step regression algorithm in the Data Processing System was used to establish a regression model, which could describe the relationship between the compressive strength of cementitious materials and the content of additives. The specific calculation process is now described. First, the Data Processing System constantly introduced every independent variable, according to its significance on the dependent variable Y. Then the originally introduced independent variable was replaced by the next most significant independent one. Meanwhile, the variance ratio was calculated during every step to guarantee the statistical significance of the introduced variables. Thus, only independent variables that were obviously impacting Y were included in the quadratic polynomial step regression model. Finally, the test proceeded iteratively until the independent variables significantly impacting Y were left in the model. The stability and validity of the regression models were tested through a variety of statistical methods. As a result, the final predictive model for lime, gypsum and cement clinker is:

Factors	Sum of squares	Degrees of freedom	F-ratio	P*
A	0·2158	2		
B	1·9174	2	8·2388	0·0382
C	0·2122	2	4·3627	0·0483
blank column	0·2497	2		
error	0·4654	4		

P*, probability for F-ratio test.

Table 4. Analysis of variance of experimental data

$$Y = 30{\cdot}67370891 - 44{\cdot}85106347x_3 - 31{\cdot}69395065x_4 - 79{\cdot}75347419x_1x_2 - 130{\cdot}64354028x_2x_2 + 0{\cdot}4130428745x_3x_2 - 198{\cdot}91165879x_1x_2 - 18{\cdot}699016682x_1x_3 + 4{\cdot}444304228x_1x_4 + 13{\cdot}649849113x_2x_4 + 16{\cdot}494771327x_3x_4 \quad (1)$$

where x_1, x_2, x_3 and x_4 stand for the content of cement clinker, lime, gypsum, and GBFS, respectively, in the cementitious materials.

The following three aspects should be checked in the regression equation: (1) when $P \leq 0{\cdot}05$, Y cannot be utilised in the analysis of variance or the regression equation; (2) whether the significance level of the partial correlation coefficients for all regression coefficients is less than 0·05 or not; (3) whether the Durbin-Watson statistic approaches 2 or not. It was found that the significance level of the regression equation was less than 0·05, and the Durbin-Watson statistic was 1·607. Thus all regression equations were valid, and the formula of forecasting peak surge was then derived using the MATLAB toolboxes as the following: $x_1 = 0{\cdot}02$, $x_2 = 0{\cdot}15$, $x_3 = 0{\cdot}04$, $x_4 = 0{\cdot}79$; $Y = 2{\cdot}67$ (Alberty *et al.*, 1999).

3.4 Verification test

Further verification testing in order to predict the precision of the regression models and the optimisation of the proportioning of cementitious materials was operated on the basis of previous experimental data treatment. The comparison between the predicted and measured values is shown in Table 5. By analysing the experimental error, it was found that the quadratic polynomial step regression with the Data Processing System is an ideal method to establish valid regression models, as the predicted and measured values showed little difference. The compressive strength of 2·79 MPa obtained by connecting analysis of variance and regular analysis (proportion 14) was lower than that obtained from the quadratic polynomial step regression (proportion 15) in Table 5. This further indicates the superiority of the quadratic polynomial step regression with Data Processing System when applied for optimisation of the proportion of GBFS in the cementitious materials. Therefore, the optimal proportions of cementitious materials, namely 5% cement clinker, 15% lime and 4% gypsum, and the model which had a higher prediction precision in comparison with traditional statistical analyses provided a theoretical foundation for defining the optimal proportions of cementitious materials. Under the same experimental conditions, the tailings backfill body with optimised GBFS content (proportion 15) had 4·5 times higher compressive strength in comparison with the traditional 32.5R Portland cement materials (proportion 16).

Experiment number	Measured value	Predicted value	Error: %
1	2·3	2·29	0·44
2	2·27	2·17	4·61
3	1·02	1·12	−8·92
4	2·2	2·08	5·77
5	2·76	2·5	10·4
6	2·65	2·5	6
7	2·36	2·15	9·77
8	2·46	2·51	−1·99
9	1·75	1·9	−7·89
10	1·98	2·16	−8·33
11	2·24	2·36	−5·08
12	1·67	1·67	0
13	2·25	2·47	−8·9
14*	2·24	2·36	−5·08
15†	2·79	2·67	4·49
16‡	0·62	–	–

*14, optimised result of connecting analysis of variance and regular analysis.
†15, optimised result of quadratic polynomial step regression.
‡16, result of 32.5R Portland cement backfill body under the same experimental conditions as experiment 15.

Table 5. Response data from further verification experiments

Thus the application of the obtained experimental results can lead to a decrease in mining costs, while increasing the strength of cementitious backfilling body.

The regression equation obtained here only applies to a particular set of materials. However, quadratic polynomial step regression is an ideal method for defining the proportions of cementing materials. Using this method the optimised formulation of cementitious materials can be obtained, based on a small number of experiments.

4. Conclusion

The quadratic polynomial step regression with Data Processing System is an ideal approach to establish a valid regression equation. The rationality and effectiveness of the regression equation was verified, the best optimised proportions and a theoretical foundation for defining the proportions of cementitious materials were defined which could assist in solving the solid waste recycling problem and increasing the economic benefits of mining. According to the regression equation, when the added contents of cement clinker, lime and gypsum were 5, 15 and 4% respectively; the mixed activator agent produced the best result in activating GBFS, and this was verified by experiments. As a result, the maximum measured value of 7 days compressive strength reached 2·79 MPa, which was 4·5 times higher than the value measured for 32·5R Portland cement. The difference between the measured strength and predicted strength of the regression model was 4·49%. A viable method for the design of cementitious materials is presented.

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