Experimental Study on Proportioning Optimized Ultra-Fine Aggregate Cementing Materials with Full Gold Tailings Using Response Surface Method

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Abstract. In order to prepare the ultra-fine full gold tailing aggregate cementing materials, the gold tailing powders were mixed with cement under different proportions. Then the effect of sand quantity, water quantity and dosage of water-reducing agent on the compressive strength of the full tailing aggregate cementing materials was studied using response surface method in Design-Expert software. Based on the regression analysis of test results, a visual model was provided. The research results show that when the relative mass dosage of cement is 1, the amount of sand is greater than 1.1, the amount of water is greater than 0.24, and the amount of water reducing agent is 0.01. The 7d compressive strength of tailings concrete shows a significant negative linear correlation with the amount of sand and water. That implays the concrete strength decreases with the increase of the amount of sand and water. When the relative mass of cement is 1, the maximum 7d strength of concrete appears at the relative mass of sand 1.0, the amount of water is 0.22, and the amount of water reducer is 0.01. The maximum 7d compressive strength is about 75.43MPa.

Keywords. Gold tailings, full tailing aggregate, cementing material, response surface method, strength test

1. Introduction

Concrete is one of the primary materials used in every construction. With the progress of China construction projects, the demand of concrete is growing day by day [1]. Sand and gravel aggregate are essential raw materials for preparing concrete. Over-exploitation of sand and gravel aggregates over the years has caused not only visual pollution, noise pollution, water pollution, air dust pollution and solid waste pollution but also caused serious soil erosion, vegetation destruction and ecological environment deterioration [2-3]. For this reason, in accordance with the requirements of environmental protection, the exploitation of natural sand and gravel aggregates in many areas have been restricted. This issue has led to a sharp increase in the price of required aggregates in concrete projects and the cost of construction projects. There is a

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shortage of aggregates in many places, which affects the smooth progress of infrastructure projects [4].

Gold tailings are a kind of industrial waste produced by gold mine beneficiation processes. Many years of mining development have created an enormous number of tailings.

The accumulation of tailings not only causes waste of mineral resources and occupation of arable land, but also leads to environmental pollution, soil erosion and vegetation destruction. The tailings pond accident can simply cause natural disasters such as collapse, landslide and mudslide [5-8]. However, from the resources and environmental science perspective, gold tailings are a potential secondary resource. Therefore, the development and utilisation of tailings not only make rational use of mineral resources, but also solve the environmental and safety problems caused by tailings. If tailings sand can be used instead of natural sand and gravel as the aggregate in concrete, it can make up the gap of raw materials for concrete production and also turn waste into treasure. But up to now, there has not been a large-scale application of materials of this size as all aggregates of concrete in the concrete industry. Regression analysis is a statistical analysis method to determine the quantitative relationship between two or more variables. However, the classical regression analysis method can only processes the existing experimental data passively, and does not put any requirements on the arrangement of the experiment. There are few studies showed how the accuracy of the regression equation can be improved, so that the multi-factor experiment problem often fails to achieve the experimental purpose, also, sometimes it is impossible to test the suitability of the model [9]. A new optimisation method called the response surface method (RSM) can be applied to solve this problem. RSM, also known as the regression design method, is an optimisation method that is integrated by experimental design, regression analysis and other mathematical techniques. It actively collects data based on multiple linear regression to obtain a regression equation (response function) of better properties. This method can be used to establish continuous variable surface models, find out the quantitative relationship between test results and various factors, and evaluate the factors that affect the change process and their interactions, and determine the optimal level range. Because the multi-dimensional space surface, it builds closer to the actual situation, and the number of required test groups is relatively small, saving manpower and material resources. This method has been successfully applied to various engineering process optimisation [10].

In this paper, the RSM is used to carry out an experimental study on the strength of all tailings aggregate concrete. In the case that the relative mass of cement is 1, the influence of sand, water and water reducer on the compressive strength of concrete is studied to provide a reference for the application of tailings sand as concrete aggregates.
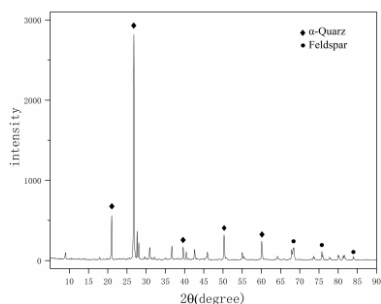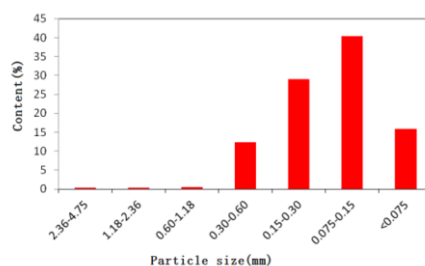
2. Design of Experiments

2.1. Materials

A gold mine in Shaanxi, China produces the tailing sand used in the concrete as the flotation tailing sand. It is in the form of loose silt, with a specific surface area of 5.35 m^2/kg and a density of 2.96 g/cm^3. The chemical composition was analysed by Shimadzu's LAB CENTER XRF-1800 X-ray fluorescence spectrometer, and the test results are shown in table 1.

Table 1. Chemical composition of gold tailing/%.

SiO$_2$	TiO$_2$	Al$_2$O$_3$	Fe$_2$O$_3$	MnO	MgO	CaO	Na$_2$O	K$_2$O	P$_2$O$_5$	LOI	TOTAL
68.76	0.43	13.59	2.99	0.06	1.42	3.13	2.48	3.35	0.23	3.01	99.45

The D8 Advance X-ray diffractometer of the German Bruker Company was used to analyse the mineral composition of the tailing samples. The results are depicted in figure 1. It can be seen from figure 1, the main mineral components of gold tailings are quartz and feldspar. Refer to the national standard "GB/T14684-2011 Sand for Construction" to determine the particle gradation and fineness modulus of gold tailings. The screening results are presented in figure 2. After calculation, the fineness modulus of tailings sand is 0.58, which does not meet the fineness requirement (>0.7) of the national standard for construction sand superfine sand. Therefore, it can be concluded that tailings sand belongs to ultra-fine aggregate.

Figure 1. XRD pattern of gold tailing

Figure 2. Particle size distribution of gold tailing.

The cement used is P.O.42.5 ordinary Portland cement produced by Shaanxi Shengwei Group Co., Ltd., and the quality meets the requirements of 'General Portland Cement Standard GB175-2008'. The considered water-reducing agent, is a commercially available high-efficiency polycarboxylic acid water-reducing agent PCE, with a solid content of 38.5%, and the quality conforms to the 'Concrete Admixture GB8076-2008'. The water used is tap water.

2.2. Methods

According to the 'GB-T 50081-2019 Standard for Test Methods of Concrete Physical and Mechanical Properties', the gold tailings sand was used to carry out a non-coarse aggregate concrete preparation test for all aggregates. The RSM was conducted to analyse a 7d compressive concrete strength, and to verify the experiment results.

3. Results and Discussion

3.1. Initial Regression Orthogonal Design

The purpose of the initial regression orthogonal test design is to find the steepest ascent direction of the strength extreme value and lay the foundation for the next rapid ascent test. The initial test examines the influence of sand-cement ratio (x_1), water-cement

ratio (x_2) and water reducer (x_3) on the 7d compressive strength of full tailings aggregate concrete.

Equations (1) indicates the relationship between 7d compressive concrete strength y and the sand-cement ratio $x1$, the water-cement ratio $x2$ and the water-cement ratio $x3$:

$$y_a = \beta_0 + \sum_{j=1}^{3} \beta_j x_{aj} + \varepsilon_a \quad (a=1,2,...,N) \tag{1}$$

Where, y_a is the 7d compressive of the concrete strength in a test, MPa; x_{aj} is the independent variable, j=1~3, which are the values of the sand-to-cement ratio, water-cement ratio and the amount of water-reducing agent in a test; β_j is the independent variable coefficient, j=0~3 to be determined. ε_a is the random error of the *ath* trial, and it is assumed that it follows a normal distribution with a mean value of 0 and a variance of σ^2.

The orthogonal regression test uses the factor code value instead of the factor level value to perform the statistical analysis of the test results after the coding transformation, which significantly simplifies the statistical analysis. The coding of each factor x_i in this article is represented by capital X_i. The codes and levels of research factors used in the experiment are shown in table 2. In this table, Δ shows the interval changes in the test level. $L_8(2^7)$ orthogonal table is used to arrange the test, the number of tests is n=8. Table 3 provides the test results.

Table 2. Factors and level coding for initial test.

Factor code	x_1	x_2	x_3
−1	3.0	0.62	0.009
0	2.9	0.61	0.010
+1	2.8	0.60	0.011
Δ	0.1	0.01	0.001

Table 3. Primary orthogonal test arrangement and result.

No.	X1	X2	X3	Y (7d compressive strength, MPa)
1	−1	−1	−1	21.2
2	−1	−1	1	20.9
3	−1	1	−1	21.3
4	−1	1	1	21.4
5	1	−1	−1	22.8
6	1	−1	1	23.2
7	1	1	−1	23.5
8	1	1	1	23.6
$\Sigma X_j y_j$	8.3	1.7	0.3	
$\beta_j = \Sigma X_j y_j / n$	1.0375	0.2125	0.0375	

Based on the experimental results presented in table 2, the analysis of variance of the initial mathematical model and the significance test of the regression coefficient was carried out. Table 4 illustrates the results.

Table 4. Significance test of primary regression results.

Source of variation S_j, sum of squares f_j, Degree of freedom			$F=(S_j/f_j)/(S_{rest}/f_{rest})$	$F_\alpha(f_j, f_{rest})$	Significance
X_1	8.61125	1	222.2258065	$F_{0.05}(1,4)=7.709$	*
X_2	0.36125	1	9.322580645	$F_{0.05}(1,4)=7.709$	*
X_3	0.01125	1	0.290322581	$F_{0.05}(1,4)=7.709$	
return	8.98375	3	77.27956989	$F_{0.01}(3,4)=16.694$	**
rest	0.155	4			

From the results of the significance test, it can be seen that the linear regression equation is highly significant (significance level $\alpha=0.01$), the coefficients of X_1 and X_2 are both significant ($\alpha=0.05$). In contrast, the coefficient of X_3 is not significant. Because the first-order regression orthogonal design is orthogonal and the coefficients of each factor are independent to each other, the variable X_3 has no significant effect and can be directly discarded in the model. The response function is obtained as a linear regression equation and can be expressedas follow:

$$y=22.2375+1.03755X_1+0.2125X_2 \tag{2}$$

3.2. Steepest Ascent Method Test Design (Rapid Ascend Method)

According to the regression equation (2), set the step length factor to -0.2, and use the steepest ascent method for the second round of experiments. Since the amount of water-reducing agent x_3 has no significant effect, in this round of tests, x_3 is fixed to 0.010, and the sand-to-cement ratio (x_1) and water-cement ratio (x_2) are changed to study its effect on the 7d strength of tailings sand concrete. The test design and test results are shown in table 5.

Table 5. Climb-quickly methods test arrangements and results.

	x_1	x_2	y
Original test	3.0	0.62	21.2
β_j	1.0375	0.2125	
Order/step factor	-0.2	-0.2	
1	2.79	0.58	21.4
2	2.59	0.54	31.3
3	2.38	0.49	37.9
4	2.17	0.45	43.4
5	1.96	0.41	51.8
6	1.76	0.36	55.6
7	1.55	0.32	58.7
8	1.34	0.28	64.3
9	1.13	0.24	67.4
10	0.92	0.19	65.1

In the first 9 tests; the concrete strength value increases when sand amount $x1$ and water amount x_2 changes. According to the statistics analyses obtained from the first 9 test results, it can also be seen when the sand content $x1$ is greater than 1.1 and the water content x_2 is greater than 0.24, the 7d strength of concrete has a significant negative linear correlation with the sand content x_1 and the water content x_2. The correlation coefficients are $R_1 = -0.987$ and $R_2 = -0.988$; which means as the amount of sand and water increases, the strength of concrete decreases.

Then the linear regression equation of concrete 7d strength, sand amount $x1$ and water amount $x2$ when sand amount x_1 is greater than 1.1 and water amount $x2$ is greater than 0.24 can be calculated as below:

$$y=101.8+28.6x_1-269.9x_2 \tag{3}$$

The calculated coefficient of determination is R2=0.978. After testing, the linear regression equation is highly significant, and the confidence probability is >99%.

3.3. Quadratic Regression Orthogonal Rotation Combination Design

The strength value of the 10th test in table 4 is reversed, and the straight bar graph of the change of the test strength value is shown in figure 3. Therefore, we redesigned the experiment around the highest point (1.13, 0.24) of the experimental results in table 4. Since the response function in the area near the highest point does not conform to the law of linear change, a quadratic design and regression test are planned. The design method of the quadratic regression orthogonal rotation combination is adopted. The regression equation model is:

$$y_a = \beta_0 + \sum_{j=1}^{2}\beta_j x_{aj} + \sum_{j=1}^{2}\beta_{ij} x_{ai}x_{aj} + \sum_{j=1}^{2}\beta_{jj} x_{aj}^2 + \varepsilon_a \quad (a=1, 2, ..., N, \ i, j=1, 2) \tag{4}$$

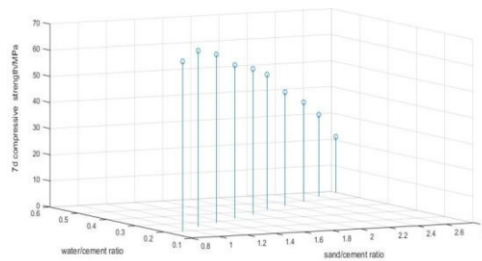The meaning of each variable was addressed before.

Figure 3. Straight bar graph of the relationship between 7d compressive strength of concrete and sand-cement ratio and water-cement ratio.

The codes and levels of research factors used in the test are shown in table 6. In table 6, r is the radius of the test rotating sphere from the test centre point; Δ has the same meaning as before. Similar to a regression orthogonal experiment, after coding transformation, the factor code value is substituted for the factor level value to perform the test results' statistics. However, in the quadratic regression orthogonal rotation

combination design, due to the correlation between the X_0 column and the X_j^2 column, to eliminate this correlation, this paper focuses on the square term X_j^2, as

$$Xa_j' = Xa_j^2 - 1/n * \Sigma Xa_j^2.$$

Table 6. Factors and level coding for the third round of test.

x_j	x_1	x_2	Sand-to-cement ratio	Water-cement ratio
-r	−1.414	−1.414	0.79	0.19
−1	−1	−1	0.85	0.20
0	0	0	1.00	0.22
+1	1	1	1.15	0.24
r	1.414	1.414	1.21	0.25
Δ	1	1	0.15	0.02

The test arrangement and results are shown in table 7.

Table 7. Quadratic orthogonal rotation experiment design and test result.

No.	X_0	X_1	X_2	Equivalent sand	Equivalent water	X_1*X_2	X_1'	X_2'	Y
1	1	−1	−1	0.85	0.20	1	0.385	0.385	66.8
2	1	−1	1	0.85	0.24	−1	0.385	0.385	63.3
3	1	1	−1	1.15	0.20	−1	0.385	0.385	64.1
4	1	1	1	1.15	0.24	1	0.385	0.385	63.2
5	1	1.414	0	1.21	0.22	0	1.385	−0.615	63.9
6	1	−1.414	0	0.79	0.22	0	1.385	−0.615	66.2
7	1	0	1.414	1.00	0.25	0	−0.615	1.385	65.3
8	1	0	−1.414	1.00	0.19	0	−0.615	1.385	63.2
9	1	0	0	1.00	0.22	0	−0.615	−0.615	69.7
10	1	0	0	1.00	0.22	0	−0.615	−0.615	70.4
11	1	0	0	1.00	0.22	0	−0.615	−0.615	69.7
12	1	0	0	1.00	0.22	0	−0.615	−0.615	71.9
13	1	0	0	1.00	0.22	0	−0.615	−0.615	70.3
bi	72.33	−0.057	0.921			0.65	−2.292	−2.744	

After calculation and analysis, Equations (5) presents the preliminary regression:

$$Y = 72.33 - 0.057X_1 + 0.921X_2 + 0.65X_1X_2 - 2.292X_1' - 2.744X_2' \tag{5}$$

The results obtained from the analysis of variance are summarised in table 8.

It can be seen from the results analysis of variance presented in table 8, the regression equation has a significant impact, but the effects of X_1, X_2 and X_1X_2 are not significant. Therefore, discarding the X_1, X_2, and X_1X_2 terms from the initial regression equation (5) is changed to the following equation :

$$Y = 72.33 - 2.292X_1' - 2.744X_2' \tag{6}$$

Restore the centralisation transformation to X_j^2,

$$Y = 75.43 - 2.292X_1^2 - 2.744X_2^2 \qquad (7)$$

Table 8. Quadratic regression orthogonal rotation test design variance analysis table.

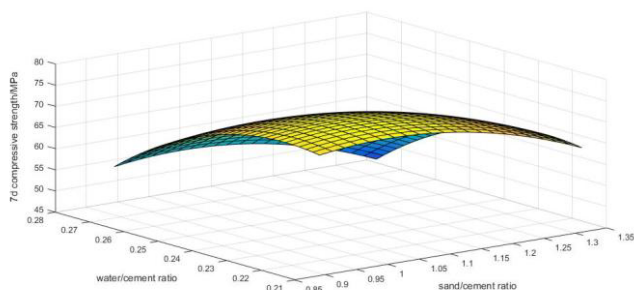| Source of variation S		Degree of freedom Mean square F			$F_{0.05}$	Significance
X_1	0.0256	1	0.0256	0.007	5.5914	
X_2	6.7885	1	6.7885	1.8608	5.5914	
X_1X_2	1.69	1	1.69	0.4632	5.5914	
X_1'	37.1755	1	37.1755	10.1901	5.5914	*
X_2'	53.291	1	53.291	14.6075	5.5914	*
return	98.9706	5	19.7941	5.4257	3.9715	*
rest	25.5371	7	3.6482			
deviation	3.24	4	0.81			
Lack of fit	22.2971	3	7.4324	2.0373	4.3468	

Therefore: $S_{tatal} = S_{return} + S_{rest} = 98.9706 + 25.5371 = 124.5077$. The sum of variances of X_1' and X_2 is $S_R = 37.1755 + 53.291 = 90.4665$ decisive factor as $R^2 = S_R/S$ 总 $= 90.4665/124.5077 = 0.7266$, Correlation coefficient as $R = 0.8524$.

In the case of a significance level of 0.01, the correlation coefficient's critical value is 0.7078, so the confidence probability of the regression equation (7) is >99%.

Substituting $Xj = (xj - xj_0)/\Delta j$ into equation (7),

$$y = 75.43 - 2.292(\frac{x_1 - 1.0}{0.15})^2 - 2.744(\frac{x_2 - 0.22}{0.02})^2 \qquad (8)$$

According to this, changing surface of concrete 7d compressive strength and sand volume and water volume can be compared in figure 4. According to formula (8) and figure 4, among the parameters studied, the maximum 7d strength of concrete appears at (sand, water) = (1.0, 0.22), and the maximum strength is 75.43MPa.

Figure 4. Variation of concrete strength with sand and water content at the top of the function.

3.4. Concrete Strength Verification Test

Based on the 'GBT50080-2016 Standard for Test Methods for the Performance of Ordinary Concrete Mixtures' and 'GB-T 50081-2019 Standard for Test Methods for the Physical and Mechanical Properties of Concrete', the preparation test of all tailings aggregate concrete was carried out. The test ratio and three repetition results are shown in table 9.

Table 9. Test ratio and result table.

No.	Raw material quality ratio				Expansion ,mm	Compressive strength, MPa		Flexural strength, MPa	
	cement	Tailing sand	Water	Water-reducing agent		7d	28d	7d	28d
1	1	1.0	0.22	0.01	180	73.8	82.6	11.9	13.4
2	1	1.0	0.22	0.01	185	76.3	87.5	12.4	14.6
3	1	1.0	0.22	0.01	182	74.6	83.7	12.6	13.8
average	1	1.0	0.22	0.01	182	74.9	86.6	12.3	14.1

It can be seen from table 9, the 7d average compressive strength value of tailings concrete obtained by repeated experiments is 74.9 MPa, which has only 0.7% error from the maximum strength value of 75.43 MPa in the optimisation analysis. Good reproducibility.

4. Results

(1) When the relative mass amount of cement is 1, the sand amount is greater than 1.1, the water amount is greater than 0.24, and the amount of water-reducing agent is 0.01, the 7d compressive strength of tailings concrete has a significant negative linear relationship with the amount of sand and water. It can be concluded that while the amount of sand and water increases, the strength of concrete decreases.

(2) When the relative mass of cement is 1, the maximum 7d strength of concrete appears at the relative mass of sand 1.0, the amount of water is 0.22, and the amount of water reducer is 0.01. The maximum 7d compressive strength is about 75.43MPa.

Acknowladgements

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