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Response Surface Optimization of Compressive Strength for Fluorogypsum-Based Cementitious Materials under Multi-Factor Interactive Effects

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Abstract: The massive accumulation of fluorogypsum (FG) poses severe environmental challenges. To address the limitations of previous single-factor studies, this research employed the Box-Behnken design and response surface methodology (BBD-RSM) to investigate the interactive effects of FG, ground granulated blast-furnace slag (GGBS), and sodium sulfate (Na_2SO_4) on the compressive strength of a novel solid waste-based cementitious material. Analysis of variance confirmed the high fitting accuracy of the established quadratic regression model. Results indicated that the significance of factors on compressive strength ranks as: GGBS > FG > Na_2SO_4. The optimal mix proportion for maximizing compressive strength was determined as 65.39% FG, 15% GGBS, 14.61% fly ash (FA), and 5% ordinary Portland cement (OPC), with an external addition of 1.47% Na_2SO_4. Under this formulation, the measured 28-day compressive strength reached 36.51 MPa, yielding a relative error of only 1.4% compared to the predicted value. This study provides robust theoretical guidance for the large-scale application of FG in green building materials.

Keywords: Fluorogypsum; Response surface methodology; Compressive strength.

1. Introduction

The production of ordinary Portland cement (OPC) is accompanied by massive energy consumption and enormous carbon dioxide emissions[1].To reduce the carbon footprint of the construction industry and promote the resource utilization of bulk industrial solid wastes, the development of solid-waste-based, low-carbon alternative cementitious materials has become a prominent research hotspot[2]. Fluorogypsum (FG), a major by-product of the fluorine chemical industry, consists primarily of anhydrous calcium sulfate. Because FG can be utilized directly as a raw material for cementitious materials at ambient temperature—eliminating the need for energy-intensive high-temperature calcination—it exhibits significant low-carbon and eco-friendly advantages. Furthermore, the micro-expansion characteristics accompanying its hydration process can effectively compensate for the volume shrinkage of the hardened paste, indicating broad application potential in the field of green building materials.

However, pure FG cementitious materials often exhibit limited compressive strength and a relatively loose internal structure due to the slow hydration rate of anhydrous calcium sulfate[3].. These inherent deficiencies lead to a fragile hardened matrix, which severely restricts their high-load-bearing engineering applications. To overcome this bottleneck, it is generally necessary to introduce supplementary cementitious materials possessing latent hydraulic or pozzolanic activity, such as ground granulated blast-furnace slag (GGBS) and fly ash (FA)[4]. Under the action of activators, these reactive materials undergo secondary hydration reactions to generate calcium silicate hydrate (C-S-H) gels and ettringite (AFt). These hydration products effectively fill the internal pores and significantly refine the microstructure of the system, thereby substantially enhancing the mechanical properties and structural density of the composite material.

In recent years, researchers worldwide have conducted extensive explorations into the activation of FG. He et al.[5] confirmed that potassium carbonate and potassium bicarbonate can effectively accelerate the hydration process of FG. Similarly, Liu et al.[6]investigated the activation mechanism of FG-based materials and demonstrated that the incorporation of sodium sulfate as a chemical activator effectively promotes the hydration process, leading to a substantial enhancement in the early and late-age compressive strength. However, most existing studies rely heavily on single-factor experiments or orthogonal experimental designs. These traditional methodologies struggle to comprehensively evaluate the nonlinear interactive effects among multiple variables and fail to accurately identify the global optimal mix proportions of the system.

In light of this, the present study introduces Response Surface Methodology (RSM) to systematically analyze the interactive effects of cementitious material proportions and Na2SO4 activator dosage on the performance of FG-based cementitious materials by constructing a multiple quadratic polynomial regression model. With compressive strength selected as the core response variable, an RSM-based experimental design was formulated and executed. Through model fitting and analysis of variance, the contributions of individual factors and their coupling effects on the macroscopic properties of the materials were quantitatively evaluated.

2. Materials and methods

(1) Materials

The fluorogypsum used in this study was sourced from Henan Do-Fluoride Chemicals Co., Ltd., and was subjected to a neutralization treatment using carbide slag prior to use. The GGBS and FA were supplied by Great Wall Co., Ltd., while the cement was provided by Jiaozuo Qianye Cement Co., Ltd. Analytical-grade anhydrous Na2SO4 obtained from Tianjin Chemical Reagent Co., Ltd., was employed as the chemical activator. The main chemical compositions of these raw materials are presented in Table 1.

Table 1. Chemical composition (wt.%) of raw materials

Samples	CaO	SO3	Al2O3	SiO2	MgO	Fe2O3	MnO	P2O5	Others
FG	41.43	54.35	0.29	1.17	0.38	0.23	0.01	---	2.14
FA	4.44	0.40	19.75	58.63	2.07	9.55	---	---	0.3
GGBS	44.70	---	13.78	29.51	7.87	1.03	1.53	0.71	0.87
OPC	63.34	2.42	4.40	20.82	2.88	3.27	---	---	2.87

(2) Specimen preparation

To ensure the scientific validity of the response surface methodology design, the specific dosage ranges for both FG and the externally added Na2SO4 activator were determined based on the optimal results from preliminary single-factor experiments. Meanwhile, taking economic cost control in practical engineering applications into consideration, the investigated dosage range for GGBS was set between 0% and 15%. Within the composite cementitious system (with the total mass defined as 100%), OPC content was fixed at 5%, while FA was incorporated as the balance component (i.e., FA % = 100% - FG % - GGBS % - 5%). It should be particularly noted that the Na2SO4 activator was added externally based on the total mass of the composite cementitious materials.

The dry raw materials were accurately weighed according to the mix proportions designed by the Response Surface Methodology. Initially, all the dry powders were introduced into a planetary mixer and dry-mixed at a low speed for 1–2 minutes to ensure homogeneous dispersion. Subsequently, water was added at a fixed water-to-binder (W/B) ratio of 0.35, followed by wet-mixing for an additional 2–3 minutes until a uniform, lump-free paste was formed. After mixing, the fresh paste was immediately cast into 40 mm × 40 mm × 40 mm stainless steel cubic molds and compacted on a vibrating table to eliminate entrapped air bubbles. The surfaces of the molded specimens were covered with plastic film to prevent rapid moisture evaporation and then placed in a standard curing room (temperature: 20±2 °C, relative humidity: ≥95%). After curing for 24 hours, the specimens were demolded and further cured under the same standard conditions until the designated testing ages for subsequent evaluations of mechanical properties.

Guided by prior exploratory tests, the addition rates of FG, GGBS, and the Na2SO4 activator were determined as the three independent factors. The study employed a 3-factor, 3-level design methodology, where the parameter levels were assigned codes of +1, 0, and −1. The precise values for these experimental levels are summarized in Table 2.

Table 2. Factor levels and coding for the Box-Behnken design.

Factor	Code	Unit	Level		
			-1	0	1
FG	A	%	60	65	70
GGBS	B	%	5	10	15
Na2SO4	C	%	1	1.5	2

(3) Compressive strength test

The experiments were performed according to the GB/T 17671-2021 standard. Testing ages were set at 3, 7, and 28 days for the specimens. The results represent the average of three specimens.

3. Results

(1) Response surface experiment optimization

A total of 17 experimental runs were generated using Design-Expert 13.0 software. This experimental matrix consisted of 12 factorial points to evaluate the interactive effects among the factors, and 5 center points to estimate the experimental error and detect the curvature of the model.

Table 3. Design and results of the Box-Behnken experiments

Sample	Factor			Compressive strength (MPa)
	FG (%)	GGBS (%)	Na2SO4 (%)	
1	60	5	1.5	20.6
2	70	5	1.5	19.4
3	60	15	1.5	34.1
4	70	15	1.5	32
5	60	10	1	25.2
6	70	10	1	24.5
7	60	10	2	25.1
8	70	10	2	24.7
9	65	5	1	22.8
10	65	15	1	35.1
11	65	5	2	22.4
12	65	15	2	33.4
13	65	10	1.5	29.5
14	65	10	1.5	30.8
15	65	10	1.5	29.4
16	65	10	1.5	30.2
17	65	10	1.5	28.4

The acquired experimental data were subjected to quadratic polynomial regression fitting using Design-Expert 13.0 software to establish mathematical models for the compressive strength. Subsequently, analysis of variance (ANOVA) and significance testing were conducted to evaluate the statistical adequacy of the developed regression models.

Within the theoretical framework of Response Surface Methodology, the statistical validity of regression models is comprehensively evaluated through significance testing and goodness-of-fit assessments. Generally, the overall significance of a model is measured by its F-value and P-value. A P-value of less than 0.05 indicates that the regression equation is statistically significant. Concurrently, the P-value for the "Lack of Fit" must be greater than 0.05 (i.e., insignificant) to confirm that the residuals originate primarily from pure random error rather than structural defects within the model.

Regarding fitting accuracy, the coefficient of determination (R2) is employed to quantify the correlation between the experimentally observed values and the models predicted trajectories; a value closer to 1 signifies a stronger explanatory capacity. Correspondingly, the adjusted coefficient of determination (Adj R^2) reflects the true performance of the model by eliminating the false inflation of goodness-of-fit caused by the incorporation of multiple independent variables. Furthermore, the difference between the Adj R^2 and the predicted coefficient of determination

(Pred R^2) should be less than 0.2 to ensure a robust generalization capability and effectively prevent overfitting.

Additionally, the models ability to suppress background noise is characterized by the signal-to-noise ratio, termed "Adequate Precision." This value must be maintained above 4 to ensure effective navigation within the designated design space. Combined with a low coefficient of variation (C.V. < 10%), these metrics collectively constitute the core criteria for determining whether the mathematical model possesses high fidelity and robustness in describing the complex evolutionary relationships between the variables and the responses.

By fitting the experimental data shown in Table 3, the response surface regression equation for compressive strength was established as follows:

$$Y= -556.7 + 17.31A + 1.84B + 16.16C - 0.009AB + 0.03AC - 0.13BC - 0.1337A2 + 0.0083B2 - 5.77C2$$

As summarized in Table 4 , the ANOVA results for the quadratic model of compressive strength yield an F-value of 52.22 with a corresponding P-value of less than 0.0001, indicating that the regression model is highly significant. The P-value for the Lack of Fit is 0.5180, which is non-significant (P>0.05) relative to the pure error. This confirms the adequacy of the model and suggests that the regression equation is suitable for predicting the compressive strength within the experimental range.

Table 4. ANOVA for the quadratic model of compressive strength.

	Sum of Squares		Mean Square	F-Value	P- Value	Significant
Model	366.73	9	40.75	52.22	<0.0001	significant
A:FG	2.42	1	2.42	3.10	0.1216	
B:GGBS	305.05	1	305.05	390.94	<0.0001	
C: Na2SO4	0.5	1	0.5	0.6408	0.4497	
AB	0.2025	1	0.2025	0.2595	0.6261	
AC	0.0225	1	0.0225	0.0288	0.8700	
BC	0.4225	1	0.4225	0.5415	0.4857	
A2	47.04	1	47.04	60.29	0.0001	
B2	0.1813	1	0.1813	0.2323	0.6445	
C2	8.76	1	8.76	11.23	0.0122	
Residual	5.46	7	0.7803			
Lack of Fit	2.19	3	0.7300	0.8924	0.5180	Not significant
Pure Error	3.27	4	0.8180			
Total	372.19	16				

Table 5. Model Fit Statistics

Std. Dev.	Mean	C.V. (%)	R2	Adj R2	Pred R2	Adeq Precision
0.8833	27.51	3.21	0.9853	0.9665	0.8921	22.36

Table 5 summarizes the statistical reliability indices of the response model for compressive strength. The results indicate that the coefficient of determination (R^2 = 0.9853) is in high agreement with the adjusted R^2 (Adj R^2 = 0.9665). Furthermore, the difference between the adjusted R^2 and the predicted R^2 (Pred R^2 = 0.8921) is within the threshold of 0.2, confirming the excellent goodness-of-fit and predictive stability of the regression equation within the design space. The low coefficient of variation (C.V. = 3.21%) and high adequate precision (22.36) significantly exceed the statistical requirements, demonstrating that the model possesses high discriminative power for precise prediction and optimization.

To ensure the statistical validity of the developed quadratic model, diagnostic assessments were performed to evaluate the adequacy and reliability of the regression results. The comprehensive diagnostic evaluation for compressive strength is presented in Fig. 1.

Fig. 1(a) shows the normal probability plot of residuals for compressive strength. The experimental data points closely align with the 45° red baseline, confirming that the experimental errors strictly follow a normal distribution. This indicates that no significant non-linear terms were omitted during the model derivation, thereby satisfying the prerequisite assumption for the analysis of variance.

Fig. 1(b) shows the predicted versus actual value plot for compressive strength. All sample points are tightly clustered along the black diagonal line (the line of perfect prediction) without any significant deviation. This visually demonstrates the exceptionally high predictive accuracy of the quadratic polynomial regression model, proving its capability to authentically characterize the evolutionary relationship between the mix proportion parameters and the strength indicator.

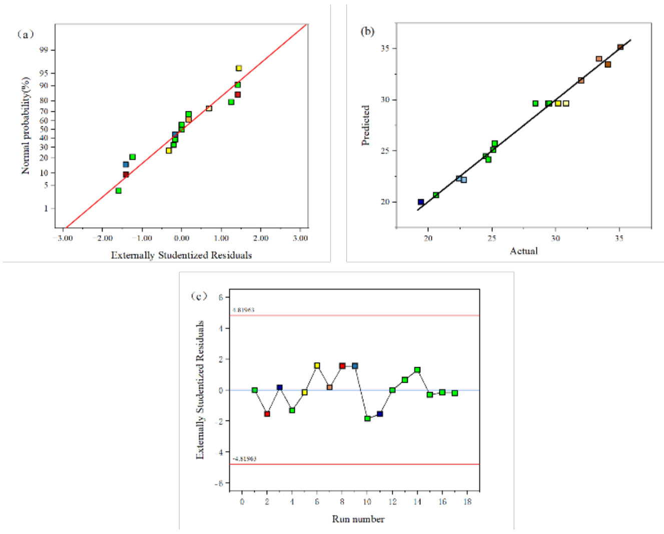

Fig. 1 Model diagnostic plots for compressive strength: (a) Normal probability plot of externally studentized residuals; (b) Predicted versus actual values; (c) Externally studentized residuals versus run number.

Fig. 1(c) displays the externally studentized residuals plot for compressive strength. The residual values of all 17 experimental runs fall strictly within the upper and lower control limit thresholds (the red horizontal lines). This indicates excellent data consistency, confirming the absence of significant outliers in the experimental system and thereby entirely eliminating the potential impact of isolated interference points on the robustness of the regression equation.

In summary, the comprehensive diagnostic analysis confirms that the established quadratic regression model fulfills all necessary statistical assumptions. The high degree of linearity in the probability plot, the tight clustering of predicted versus actual values, and the absence of any outliers beyond the control limits collectively demonstrate the high fidelity and robustness of the model.

To visually elucidate the synergistic mechanisms and the intensity of interactions among the design variables, 3D response surface plots and their corresponding 2D contour maps were generated, as illustrated in Fig. 2.

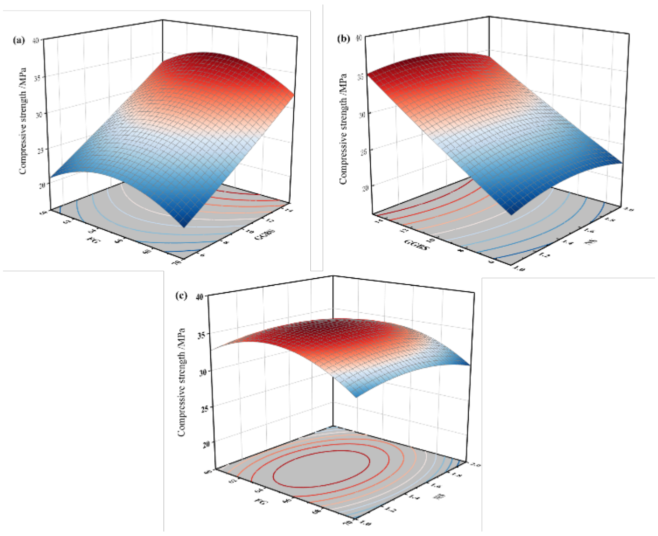

Fig. 2 Effects of interactive factors on compressive strength: (a) FG and GGBS contents; (b) Na2SO4 and GGBS contents; (c) FG and Na2SO4 contents.

Fig. 2(a) illustrates the interactive effect of FG and GGBS contents on compressive strength. The response surface exhibits a distinctly convex shape. The compressive strength shows a clear monotonic upward trend with the increase of GGBS content, whereas it exhibits a non-linear pattern—initially increasing and then decreasing—with the increase of FG content, reaching its peak within the FG range of 64%–66%. By comparison, the surface slope on the GGBS side is steeper and the contour lines are denser, indicating that the influence of GGBS on compressive strength is significantly greater than that of FG.

Fig. 2(b) demonstrates the interactive effect of GGBS and Na2SO4 contents on compressive strength. The compressive strength presents a steep, monotonic upward trend with the increase of GGBS content, showing extremely high response sensitivity. Conversely, its response to the increasing Na2SO4 content follows a parabolic pattern of an initial rise followed by a decline, with relatively gentle surface fluctuations. The bottom contour distribution reveals that the lines on the GGBS side are much denser than those on the Na2SO4 side, further confirming that the enhancing effect of GGBS on compressive strength far exceeds that of Na2SO4.

Fig. 2(c) presents the interactive effect of FG and Na2SO4 contents on compressive strength. The response surface displays a typical umbrella-like structure, indicating that the compressive strength has clear peak responses (initially increasing and then decreasing) to the variations of both factors. The closed elliptical contour lines at the bottom reveal that the interactive effect is most significant when the FG content is in the range of 63%–67% and the Na2SO4 content is between 1.2% and 1.6%. Furthermore, the steeper slope and denser contour lines on the FG side suggest that the impact of FG on compressive strength is greater than that of Na2SO4.

Comprehensive Conclusion: Based on the surface slopes and contour line densities, the order of significance regarding the impact of each factor on compressive strength is: GGBS content > FG content > Na2SO4 content. Among these, the enhancing effect of GGBS is the most prominent, and the evolutionary trend for the system to achieve high compressive strength clearly points toward a higher dosage of GGBS.

To verify the reliability of the established response surface model, numerical optimization was performed using Design-Expert 13.0 software with the explicit objective of maximizing the compressive strength. The optimal mix proportion was determined to be 65.39% FG, 15% GGBS, 14.61% FA, and 5% OPC, with 1.47% Na2SO4 added externally. Subsequently, triple independent laboratory validation experiments were conducted utilizing this optimal combination, and the average value was recorded for comparison. As presented in Table 6, the predicted values and the actual experimental results are in excellent agreement, with the relative errors strictly controlled within 5%. This comprehensively demonstrates that the developed regression model possesses outstanding predictive accuracy and can provide reliable theoretical guidance for the mix design of fluorogypsum-based cementitious materials.

Table 6. Experimental validation results of the optimized response surface model.

	28d compressive strength (MPa)
Predicted value	35.99
Actual value	36.51
Relative error (%)	1.4

4. Conclusions

In this study, the Box-Behnken design combined with response surface methodology was employed to systematically investigate the interactive effects of FG, GGBS, and Na2SO4 activator on the compressive strength of a novel solid waste-based cementitious material. The main conclusions are drawn as follows:

(1) A quadratic polynomial regression model targeting compressive strength was successfully established and validated. Analysis of variance and residual diagnostics confirmed that the model errors strictly follow a normal distribution. The model exhibits exceptionally high fitting accuracy and statistical reliability.

(2) Based on the comprehensive analysis of response surface slopes and contour line densities, the significance of the effects of various factors on compressive strength is ranked as follows: GGBS content > FG content > Na_2SO_4 content. Among them, GGBS, acting as the core reactive component, plays an absolutely dominant and monotonically promoting role in the steady enhancement of the systems strength.

(3) Through numerical optimization, the optimal theoretical mix proportion for maximizing compressive strength was determined as 65.39% FG, 15% GGBS, 14.61% FA, and 5% OPC, with an external addition of 1.47% Na2SO4. The measured 28-day compressive strength under this optimal formulation reached 36.51 MPa, yielding a relative error of only 1.4% compared to the models predicted value

(35.99 MPa). This remarkably high agreement confirms the high fidelity of the RSM optimization method and provides robust theoretical guidance for the large-scale and high-value-added utilization of the bulk industrial solid waste, fluorogypsum, in green building materials.

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