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Design and preparation of high-performance polymer mortars based on performance prediction model

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Abstract

This article had presented a systematic and adjustable method to forecast the performance of certain type of polymer mortars, most of all, using the performance prediction model to accurately control the dosages of water reducing agent (DWR), cellulose (DC), polypropylene fiber (DF), expansion agent (DEA), redispersible emulsion powder (DREP), and cement content (CC), according to the changes of properties of polymer mortars. The article had given a full verification supported by a series of orthogonal experiment results to demonstrate the effectiveness and the feedback loop between raw materials and properties of polymer mortars, which had revealed a big practical value and convenience for rapid construction in the job sites, especially for researchers and engineers in the field. Its effort to avoid, or at least control the damage of polymer powders to compressive strength of cement mortars. The mix proportion of high-performance polymer mortars (HPMs) was determined as follows, considering the working and mechanical properties: DWR was 0.85 g, DC was 1.05 g, DF was 1.15 g, CC was 42%, DEA was 10 g, and DREP was 20 g. Successfully created HPMs with pumping resistance of just 61.6N, compressive strength of 68.5MPa at 28d.

1. Introduction

Cement has exceptional gelling properties; it can hydate rapidly and bind sand, stone, and other elements to produce a solid structure. However, it is brittle, the mortar created by this approach is relatively inflexible [1, 2], and cracks tend to form when subjected to stress, reducing the structural stability [3]. There is a risk of cracking in high-performance cement mortar owing to its exceptionally low water-cement ratio and relatively low water content [4]. To satisfy the requirements of modern engineering applications, it is vital to design the material composition and structure of mortars and predict their performance [5, 6].

Polymer mortars (PMs) are a new cementitious materials consisting of cement, aggregates, and polymers that may be distributed in water [7–9]. PMs composite materials have prominent capabilities, including high flexibility [10] and mechanical strength [11], excellent acid-based corrosion resistance [12, 13] and waterproofing properties [14, 15], owing to the unique network structure of the mortar. Polymers are eco-friendly [16, 17] and cost-effective [18, 19]. Therefore, PMs can be widely used in surface plastering repair [20] and grouting [21, 22], which improves the fatigue performance of the structure under long-term loads and extreme environmental conditions [23–25]. Response surface methodology (RSM) is a combined design and analysis optimization technique. Through limited experiments, high-precision data can be acquired, and through optimization design, the influence of the interaction of multiple elements on the entire system can be evaluated [26–28]. The response surface approach is also useful for designing the mix proportion of mortar [29].

Based on the performance prediction model, a solution is provided to overcome the existing difficulties of polymer mortars, which can significantly improve mortar performance. A method for preparing high-
performance polymer mortars was shown to provide a novel concept for the performance enhancement and accurate prediction of polymer mortars.

2. Materials and methods

2.1. Materials
P.II 52.5 Portland cement was provided by China Cement Corp. (Nanjing, China). The chemical composition of the cement was listed in Table 1. The expansion agent, redispersible emulsion powder, cellulose, polypropylene fiber (PP fiber), and water reduction agent were provided by Jiangsu Sobute New Materials Co., Ltd (Nanjing, China). Moreover, the aggregate was 40–70 mesh river sand.

2.2. Preparation process and method
Figure 1 depicts the process used to mix and design the polymer mortars based on the performance prediction model, which consists mostly of ①. A performance prediction model of polymer mortars is constructed based on the orthogonal experiment method to clarify the impact of the interaction between various ingredients and the performance evolution of polymer mortars. ②. Optimization of the ratio parameters of polymer mortars based on the performance prediction model.

In the performance prediction model construction and ratio parameter selection experiment of polymer mortars, the water-binder ratio was 0.16:1. To anticipate the performance of polymer mortars, the response surface approach was utilized, and proportion parameters were chosen based on the performance prediction model. Taking the dosage of water reducing agent (D_{WR}), cellulose (D_{C}), and PP fiber (D_{P}), the cement content (C_{C}), the dosage of expansion agent (D_{EA}), and redispersible emulsion powder (D_{REP}) as factors, a three-factor and three-level test scheme was designed (see Table 2). Optimization of the preparation parameters for preparing the polymer mortar mix by response surface methodology was shown in Table 3. The characteristics of the polymer mortars were evaluated via room-temperature mixing. The polymer mortars were covered with plastic film and maintained at 20 ± 2°C for 24 h. Subsequently, the samples were removed from the mold and sealed until the specified age. Specimens with dimensions of 40 × 40 × 40 mm³ were used to test their compressive strength.

2.3. Methods
A vernier calliper is used to measure the thickness of the paste.

The pumping resistance of the freshly mixed polymer mortars was evaluated using a pumping resistance device (Figure 2).
The mechanical properties of the mortar were determined according to GB/T 17671-2020 [30]. The compressive strengths of the specimens were tested at 3d and 28d using a YAW-2000 microcomputer-controlled electro-hydraulic servo pressure tester with a loading rate of 0.8 MPa/s.
3. Results

3.1. Optimization of polymer mortar ratio parameters based on working performance prediction model

3.1.1. Experimental test results of the optimized design

The paste thickness and pumping resistance of the 17 sets of optimally designed polymer mortar specimens are listed in Table 4. Changes in the dosage of the water-reducing agent, cellulose, and PP fiber can have a substantial impact on the fluidity of PMs. This leads to fluctuations in paste thickness and pumping resistance.

3.1.2. Second-order response surface model and significance test

(1) Second-order response-surface model

The Design-Expert software was used to model the experimental results obtained according to equation (1) [31]: A mathematical model of the relationship between the experimental factors, levels, and response values was established. In equation (1), Y was the predicted value; \( \beta_0, \beta_i, \beta_{ii}, \beta_{ij} \) were fixed constants and regression coefficients, respectively; \( x_i \) and \( x_j \) were the factor variables; and \( e \) was the systematic error.

\[
Y = \beta_0 + \sum_{i=1}^{k} \beta_i x_i + \sum_{i=1}^{k} \beta_{ii} x_i^2 + \sum_{i<j}^{k} \beta_{ij} x_i x_j + e(x_1, x_2, \ldots, x_k)
\]  

Second-order response surface models of the thickness \( R_1 \), pumping resistance \( R_2 \), dosage of the water-reducing agent \( D_{WR} \), dosage of cellulose \( D_C \), and PP fiber \( D_F \) were established. The models were as follows:

\[
R_1 = 15.00 - 2.37 A + 1.75 B - 0.8500 \times C + 2.00 \times A \times B - 1.25 \times A \times C + 1.00 \times B \times C - 0.3750 \times A^2 - 0.1250 \times B^2 - 0.3750 \times C^2
\]

\[
R_2 = 69.50 - 9.16 \times A + 10.61 \times B - 0.1750 \times C - 1.08 \times A \times B - 10.20 \times A \times C - 7.10 \times B \times C + 9.61 \times A^2 + 7.06 \times B^2 - 5.96 \times C^2
\]

(2) Significance tests

Table 5 presents the results of the analysis of variance of the second-order response surface model. The influence of various factors and their interactions on the thickness and pumping resistance of the sample were analyzed and were indicated by the correlation coefficient \( R^2 \) and \( P\text{-value} \).

The correlation coefficients of the paste thickness and pumping resistance of the 17 sets were 0.9561 and 0.8561, respectively. The \( P\text{-value} \) is less than 0.05, indicating that the surface model was significant and may be used to forecast the working performance of polymer mortars.

3.2. The effect of interaction on the working performance of polymer mortars

3.2.1. Interaction between the molar ratio of the water-reducing agent and cellulose dosage

Figure 3 shows the model of the working performance of polymer mortars under the interaction of the water-reducing agent and cellulose dosage. Influenced by the interaction between \( D_{WR} \) and \( D_C \), the thickness and pumping resistance surface model of the freshly blended polymer mortars exhibit a clear convex tendency. Under a constant dosage of cellulose, the operational performance of the polymer mortars improves with increasing dosage of the water-reducing agent; it means that the thickness and pumping resistance were decreased. A suitable amount of a water-reducing agent was applied to the polymer mortar while stirring. The molecules of the water-reducing agent break down in water into macromolecular anions and adsorb on the...
surface of the hydration products of the cement particles. Then, the electrostatic repulsion between particles increases, which damages and prevents flocculation in the system’s structure and increases the amount of free water in the system, thereby increasing the paste microviscosity [32]. The addition of a polycarboxylic acid water-reducing agent can simultaneously generate an irregular sheet structure in the C-S-H structure, improve the pore structure of mortar, and increase the mortar’s water resistance and impermeability [33, 34].

3.2.2. Interaction between the molar ratio of the water-reducing agent and PP fiber dosage

Figure 4 shows the model of the working performance of polymer mortars under the interaction of the water-reducing agent and PP fiber dosage. When there was a low dosage of the water-reducing agent, the working performance of freshly mixed polymer mortars decreased as the fiber dosage increased, and the thickness and pumping resistance increased. Under a high dosage of the water-reducing agent, the working performance of the freshly mixed polymer mortars improved as the PP fiber dosage increased, and the thickness and pumping resistance decreased. Extreme thickness and pumping resistance values were achieved when the dosage of the water-reducing agent was 0.50 g and the dosage of the PP fiber was 1.50 g. The addition of fiber drastically impairs the fluidity of the mortar and impedes the flow of aggregates in the mortar [35, 36]. However, under the circumstance of a high dosage of the water-reducing agent, the addition of PP fibers greatly lessens the obstructive impact of the mortar. Because water-reducing chemicals can be adsorbed on the surface of cement particles, they can harm or prohibit cement flocculation via space steric hindrance and electrostatic repulsion. Subsequently, the cement particle spacing expands, releasing a substantial amount of free water and increasing

![Figure 3. Model of polymer mortars under the interaction of the water-reducing agent and cellulose dosage.](image)

![Table 5. Analysis of variance of response surface model.](image)

| Source         | Thickness F value | Thickness P-value | Pumping resistance F value | Pumping resistance P-value |
|----------------|-------------------|-------------------|---------------------------|---------------------------|
| Model          | 16.93             | 0.0006            | 4.63                      | 0.0279                    |
| X1-X1          | 66.50             | <0.0001           | 9.53                      | 0.0176                    |
| X2-X2          | 36.11             | 0.0005            | 12.79                     | 0.0090                    |
| X1-X3          | 9.03              | 0.0198            | 0.0035                    | 0.9546                    |
| X2-X3          | 23.38             | 0.0018            | 0.0656                    | 0.8052                    |
| X1^2           | 9.21              | 0.0190            | 5.91                      | 0.0454                    |
| X2^2           | 5.89              | 0.0456            | 2.86                      | 0.1345                    |
| X3^2           | 0.8726            | 0.3813            | 5.52                      | 0.0511                    |
| X1^2-X1        | 0.0970            | 0.7646            | 2.98                      | 0.1279                    |
| X2^2-X2        | 0.8726            | 0.3813            | 2.12                      | 0.1883                    |
| X3^2-X3        | 0.9561            | 0.8561            |                           |                           |

Note: P-value ≤ 0.01, highly significant.
P-value ≤ 0.05, significant.
P-value > 0.05, not significant.
the paste flowability. The water-reducing agent and PP fiber form competition adsorption on the surface of cement particles within the mortar, and in the case of a high dosage of water-reducing agent, its effect on the improvement of mortar liquidity was greater than the blocking effect of PP fiber on mortar liquidity, which increases the mortar liquidity. However, excessive quantities of PP fiber may reduce the fluidity of the polymer mortars.

3.2.3. Interaction between the molar ratio of the cellulose and PP fiber dosage

Figure 5 shows the model of the working performance of polymer mortars under the interaction of the cellulose and PP fiber dosage. Under a constant dosage of PP fiber, the working performance of freshly mixed polymer mortars decreased as the dosage of cellulose increased, indicating that the thickness and pumping resistance increased. The hydroxypropyl group of hydroxypropyl methylcellulose has a hydrophilic -OH group, and the proportion of hydroxypropyl groups in the molecular chain ranges from 7 to 12 percent. Therefore, when hydroxyl -OH or -OR comes into contact with water, it forms hydrogen bonds with the water molecules and absorbs a substantial amount of water into its molecular chain. Under agitation, the cellulose helical structure was straightened. Many such molecular chains crisscrossed, overlapped, or overlapped with other particles in the system to form a three-dimensional network structure that binds water molecules in the paste and functions as water retention [37]. The uniform distribution of cellulose in mortar can build a supporting system, block the
surface water extraction and aggregate settlement of mortar, minimize mortar bleeding, reduce the mortar’s bleeding channel, and substantially reduce the porosity [38, 39].

3.3. Optimization of polymer mortar ratio parameters based on mechanical properties prediction model

3.3.1. Experimental test results of the optimized design

The 3d and 28d compressive strength of 17 sets of optimally designed polymer mortar specimens were listed in table 6. Changes in the cement content, dosage of the expansion agent, and redispersible emulsion powder can significantly affect the development of the internal ‘locking’ structure and the overall compactness of PMs, causing the strength of the mortar to fluctuate at different ages.

3.3.2. Second-order response surface model and significance test

(1) Second-order response-surface model

The Design-Expert software was used to model the experimental results obtained according to equation (1): A mathematical model of the relationship between the experimental factors, levels, and response values was established.

Second-order response surface models of compressive strength $R_{3d}$, cement content $C_C$, dosage of expansion agent $D_{EA}$, and redispersible emulsion powder $D_{REP}$ were established. The models were as follows:

$$ R_{3d} = 30.44 + 10.61 \times A - 0.87 \times B - 4.61 \times C + 0.65 \times A \times B - 1.03 \times A \times C + 1.85 \times B \times C - 0.43 \times A^2 - 0.16 \times B^2 + 0.22 \times C^2 $$

$$ R_{28d} = 54.20 + 13.72 \times A - 1.06 \times B - 3.76 \times C + 1.45 \times A \times B - 0.65 \times A \times C + 1.47 \times B \times C - 3.01 \times A^2 - 2.04 \times B^2 - 0.7875 \times C^2 $$

(2) Significance tests

Table 7 presents the results of the analysis of variance of the second-order response surface model. The influence of various factors and their interactions on the 3d and 28d compressive strength of the sample were analyzed and were indicated by the correlation coefficient $R^2$ and $P$-value.

The correlation coefficients of the 3d and 28d compressive strength of the 17 sets were 0.9884 and 0.9757, respectively. The $P$-value is less than 0.01, indicating that the surface model was highly significant and may be used to forecast the mechanical properties of polymer mortars.

3.4. The effect of interaction on the mechanical properties of polymer mortars

3.4.1. Interaction between the molar ratio of cement content and the expansion agent dosage

Figure 6 shows a model of the mechanical properties of polymer mortars under the interaction of the cement content and the expansion agent dosage. As a result of the interaction between $C_C$ and $D_{EA}$, the surface models of the bending and compressive strength of polymer mortars at 3d and 28d exhibit a clear convexity trend. Under conditions of constant cement content, the polymer mortar strength decreased as the expansion agent dosage increased. Adding a suitable amount of expansion agent can improve the pore structure of the mortar and the interface between the skeleton components and paste, particularly during the early stages of hydration. Because the hydration reaction of the expansion agent is quicker, the hydration product is denser and can fill and

| Group | 3d  | 28d | Group | 3d  | 28d |
|-------|-----|-----|-------|-----|-----|
| B1    | 19.5| 38.0| B10   | 31.2| 52.7|
| B2    | 40.0| 61.7| B11   | 26.1| 47.1|
| B3    | 18.4| 33.7| B12   | 26.1| 47.2|
| B4    | 41.5| 63.2| B13   | 29.3| 56.8|
| B5    | 23.7| 38.9| B14   | 29.4| 53.7|
| B6    | 46.4| 68.5| B15   | 31.2| 57.8|
| B7    | 16.1| 33.6| B16   | 30.8| 52.4|
| B8    | 34.7| 60.6| B17   | 31.5| 50.3|
| B9    | 58.6| 58.5|       |     |     |
compress the C-S-H system. Although this impact reduces the early strength of the mortar, it contributes to its later strength and resistance to cracking [40]. However, suppose that the amount of the expansion agent is excessive or improperly combined. In such a circumstance, expansion products such as Ca(OH)$_2$ will also combine with the transition zone of the aggregate and paste interface, resulting in excessive local expansion, early damage (microcracks), decreased mortar strength, and increased creep [41].

3.4.2. Interaction between the molar ratio of cement content and the redispersible emulsion powder dosage

Figure 7 shows the model of the mechanical properties of the polymer mortars under the interaction of the cement content and the redispersible emulsion powder dosage. The fluctuations in $C_e$ and $D_{REP}$ significantly altered the bending and strength of the polymer mortars. The 3d and 28d compressive strength surfaces indicate that increasing the cement content from 34% to 50% substantially enhances the bending and strength. Changes in the dosage of the redispersible emulsion powder had no discernible impact on the strength when the cement content was low. In the group with a high cement content, the 28d strength model indicates that the strength of the polymer mortars increases dramatically as the dosage of the redispersible emulsion powder increases. Under a constant dosage of redispersible emulsion powder, increasing the cement content enhanced the compressive strength and brittleness. When the cement content increased, the hydration degree of the system improved in the polymer mortar system, and more active cement particles reacted to generate a denser structure and higher crosslinking degree, thereby increasing the polymer mortar strength [42]. When the cement content is 50%, the polymer mortars compressive strength reaches its maximum of 68.5MPa at 28d.

### Table 7. Analysis of variance of response surface model.

| Group               | Source   | 3d        | 28d        |
|---------------------|----------|-----------|------------|
|                     | F value  | P-value   | F value    | P-value   |
| Compressive strength| Model    | 66.51     | <0.0001    | 31.23     | <0.0001   |
|                     | $X_1X_2$ | 491.22    | <0.0001    | 247.47    | <0.0001   |
|                     | $X_2X_3$ | 3.34      | 0.1104     | 1.48      | 0.2628    |
|                     | $X_1X_3$ | 92.79     | <0.0001    | 18.60     | 0.0035    |
|                     | $X_1X_2$ | 0.92      | 0.3691     | 1.38      | 0.2783    |
|                     | $X_2X_3$ | 2.29      | 0.1739     | 0.2775    | 0.6146    |
|                     | $X_1X_3$ | 7.46      | 0.0293     | 1.43      | 0.2708    |
|                     | $X_1^2$  | 0.43      | 0.5332     | 6.27      | 0.0407    |
|                     | $X_2^2$  | 0.057     | 0.8182     | 2.87      | 0.1341    |
|                     | $X_3^2$  | 0.11      | 0.7514     | 0.4288    | 0.5335    |
| Lack of Fit         | 2.73     | 0.1781    | 0.1460     | 0.9271    |
| $R^2$               | 0.9884   | 0.9757    |            |           |

Note: $P$-value $\leq 0.01$, highly significant.

$P$-value $\leq 0.05$, significant.

$P$-value $> 0.05$, not significant.
3.4.3. Interaction between the molar ratio of the expansion agent and redispersible emulsion powder dosage

Figure 8 shows a model of the mechanical properties of polymer mortars under the interaction of the dosage of the expansion agent and the dosage of the redispersible emulsion powder. Changes in the dosage of the redispersible emulsion powder had no discernible effect on the strength when the dosage of the expansion agent was constant, and the strength decreased to some degree. The redispersible emulsion powder has a high bonding strength. When introduced into inorganic materials, the inorganic material particles can form tight bonds. The most significant drawback of inorganic materials is their high stiffnesses.

When a redispersible emulsion powder with good flexibility was applied, the particles were filled with inorganic particles, increasing the scalability between inorganic particles, which can significantly improve the flexibility, and resistance to cracking of the material. The hydrophobicity of a polymer can reduce the water absorption rate of the material, enhancing its resistance to freezing, weather resistance, durability, and wear resistance. When combined with cement mortar, polymer particles spread uniformly throughout the cement paste and were deposited on gels and unhydrated cement particles. In the hydration reaction, water was continuously consumed, hydration products grow, and polymer particles progressively accumulate in the pores, generating a compact accumulation horizon on the surface of the gels and unhydrated cement particles. The densely packed polymer particles on the gel and in the spaces condensed into a continuous film, forming an interpenetrating matrix with the paste and bonding the hydration products to the aggregate. As the hydration products with polymers create a covering film at the interface and the polymer condenses into a film in the interfacial transition zone gap, the internal stress was released, the stress concentration decreases, and the occurrence of microcracks decreases.
4. Discussion

Figure 9 shows the influence of the combination of the three explainable features on the working performance of PMs. Red represents high thickness and pumping resistance, whereas blue indicates low thickness and pumping resistance. The dosages of PP fiber and cellulose ether significantly reduced the constructability of the polymer mortars. In contrast, the dosage of the water-reducing agent can effectively improve the operability of mortar, partially offsetting the negative effect of PP fibers and cellulose ether on mortar performance. The following quantities of raw materials are determined to satisfy the actual construction requirements: \( D_{WR} \) was 0.85 g, \( D_C \) was 1.05 g, and \( D_F \) was 1.15 g.

Figure 10 shows the influence of the combination of the three explainable features on the mechanical properties of the PMs. Increasing the cement content can substantially improve the compressive strength of mortar. The impact of the redispersible emulsion powder on the strength of the mortar was minimal. It may even decrease the mortar strength, which was reflected more in the mortar toughness and adhesion performance. The addition of an expansion agent can increase the strength of the mortar to a certain extent. However, if its concentration is too high, it also causes local expansion, resulting in microcracks that diminish the strength of the mortar and increase its creep. To meet the actual building requirements, the required quantity of raw materials was determined as follows: \( C_C \) was 42%, \( D_{EA} \) was 10 g, and \( D_{REP} \) was 20 g.

In order to verify the project applicability of the performance predictive model, the selection point was performed in the model through predictive performance, and then the measured measurement was performed according to the formula. Table 8 gives a polymer mortar performance prediction model verification. The measured performance and predictive performance error values of each group were below 0.01, and the part of the group is even approaching 0. Verification, the model was accurate, and according to the actual project needs,
the corresponding proportion parameters were pushed by polymer mortar working performance mechanical properties indicators, which had a significant saving ratio parameter adjustment time. Therefore, the performance prediction model can effectively solve the problem that the formula cannot be quickly adjusted according to the changes in demand (the performance prediction model construction and application diagram are shown in figure 11).

5. Conclusions

The performance prediction model was proven to be an effective approach for designing and preparing polymer mortars. By this method, a high-performance polymer mortars was successfully prepared. When the dosage of water reducing agent was 1.20 g, the pumping resistance of polymer mortars was the lowest at 61.1N. When the cement content was 50%, the 3d and 28d compressive strength of polymer mortars was the highest at 46.4 MPa and 68.5MPa, respectively. The key proportioning parameters included: the dosage of water reducing agent is 0.85 g, the dosage of cellulose is 1.05 g, and the dosage of polypropylene fiber is 1.15 g, cement content of 42%, expansion agent dosage is 10 g, and redispersible emulsion powder dosage is 20 g.

Compared to conventional techniques, the performance prediction model can accurately predict the performance of polymer mortars and clarify the effect of the interaction between various factors on the evolution of mortar performance. Using the performance prediction model, the mortar formulation can be modified rapidly, and its quality can be accurately predicted. It is possible to manufacture polymer mortars with exceptional qualities to fulfill all strength requirements and diverse technical purposes.

6. Patents

Author contributions: Conceptualization, Shuai Qi and Xingyao Wang; methodology, Xingyao Wang, Bo Li, Chang Liu, Xiao Zhang, and Shuai Qi; software, Xingyao Wang; validation, Xingyao Wang, Shuai Qi, and Dewen Sun; formal analysis, Xingyao Wang; investigation, Shuai Qi; resources, Shuai Qi; data curation, Xingyao Wang;...
writing-original draft, Xingyao Wang; writing-review and editing, Shuai Qi and Qianping Ran; visualization, Xingyao Wang; supervision, Shuai Qi and Qianping Ran; project administration, Shuai Qi. All the authors have read and agreed to the published version of the manuscript.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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Conflicts of interest

The authors declare no conflicts of interest.

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Figure 11. Schematic of the construction and application of the polymer mortars based on performance prediction model.
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