

Article

Insights into Early-Age Interfacial Bonding Improvement of Polymer-Modified Sulfoaluminate Cement Mortar: Roles of Polymer Film Formation and Hydration Regulation

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Abstract

In this study, a polymer-modified fast-hardening and early-strength cement-based repair material was proposed, using the polymer cellulose ether (HPMC), polymer dispersible polymer powder (VAE) and silica fume. The optimal mixing ratio of the three polymers was designed by an orthogonal test by combinational evaluation of the fresh performance and mechanical properties, complemented by the observation of the changes in the macroscopic properties by XRD and SEM analysis. The results showed that the content of HPMC has a more significant effect on fluidity, consistency and water retention than other factors, and the content of silica fume has a more significant effect on setting time. The content of HPMC is the most important factor affecting the compressive strength and flexural strength. The content of HPMC and VAE has a more significant effect on the interfacial bonding performance, the 1-day interfacial bond strength was significantly higher than that without polymer. According to the SEM results, it was observed that, the porous structure of $\text{Ca}(\text{OH})_2$ induced by VAE, and the polymer film formed by HPMC covering the hydration product, the two polymers work together to significantly improve the interfacial bond strength of cement mortar. The optimal mixing ratio of the two interface bonding methods is the same, so the optimal mixing ratio of three factors is obtained: 0.2% of cellulose ether, 0.6% of redispersible polymer powder, and 10% of silica fume strong and fluid repair material.

Keywords: polymer; interface; orthogonal test; sulfoaluminate cement; bond strength



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1. Introduction

Cracks and erosion of building structures can be a result of long-term load and environmental factors, reducing the service life of buildings. To prolong the lifespan of buildings, a proper selection of repair materials is imperative [1]. Repair materials mainly include inorganic, organic, and organic modifications based on their compositions, of which organic modification materials can solve the issues of inorganic materials that have low bonding intensity and large shrinkage rates and organic materials that have poor compatibility with concrete-based materials and high cost. Among them, inorganic repair materials have problems such as low bonding intensity and large shrinkage rate; organic

repair materials have poor compatibility with cement concrete substrate materials and high cost; organic modified repair materials can just make up for the defects of the two. It can not only use polymers to improve the flexibility of repair materials and improve bond strength, but also ensure compatibility with cement concrete substrates because most of its composition is still cement [2].

Ordinary Portland cement (OPC), sulfoaluminate cement (SAC) and magnesium phosphate cement (MPC) are common inorganic repair materials for building structures, of which SAC has been considered an environmental-friendly cement material [3,4]. In addition to significantly less generation of greenhouse gas emissions during production than OPC, SAC has many excellent performances, including high early strength, micro-expansion and excellent durability. As a result of high early strength, SAC is often used as a material for crack repair. Organic modified repair materials are primarily modified using polymers such as cellulose ethers, redispersible polymer powders, and polyurethane. Among them, cellulose ether can adsorb onto the surface of cement particles through its long-chain molecular structure and form a highly viscous hydration layer, thereby improving the water retention capacity and cohesiveness of the fresh paste [5–8]. However, cellulose ether does not exert solely positive effects; excessive incorporation can significantly increase the viscosity of the paste, hinder ion diffusion and the continuous growth of hydration products, and may introduce additional pores, thereby reducing the compressive and flexural strength of the mortar [9,10]. Therefore, cellulose ether exhibits a dual effect in polymer-modified repair materials, namely “improving workability and interfacial wettability” while “weakening the load-bearing capacity of the matrix”, and its appropriate dosage plays a critical role in determining the overall performance of the material. Redispersible vinyl acetate-ethylene (VAE) copolymer powder is another commonly used polymer-modified component, whose polymer particles can form a continuous polymer film. This film can encapsulate hydration products such as AFt , $\text{Al}(\text{OH})_3$, and C-S-H gel, and bridge microcracks and pores between the old substrate and the new repair layer, thereby enhancing interfacial continuity and bonding performance. Wang et al. [11] reported that polymer powders not only provide water retention, water reduction, and air-entraining effects in both fresh and hardened states, but also improve the flexural strength and bond strength of mortars [11,12]. The strengthening mechanism is generally attributed to the gradual coalescence of polymer particles into a film during hydration and hardening, forming a continuous or semi-continuous organic phase between hydration products, aggregates, and the old substrate, thereby improving interfacial integrity and crack resistance [13]. Silva [14], Wang [15] and others observed a porous phase in EVA rubber powder-modified cement paste, speculating the phase to be a reaction product of hydroxyl and Ca^{2+} by forming a cross-linked network structure. The same phase has also been found by Wang [16] in other types of latex powder-modified cement pastes. However, the incorporation of VAE may also alter the pore structure and reduce matrix density, leading to a decrease in compressive strength. Therefore, the role of VAE in repair materials also exhibits a clear performance trade-off, being more beneficial for improving interfacial bonding and toughness rather than compressive strength.

In addition to polymers, silica fume (SF), as a highly reactive mineral admixture, is also widely used to improve the microstructure and mechanical properties of cement-based repair materials. Silica fume is characterized by its extremely small particle size and high reactive SiO_2 content. Through its micro-filling effect, it can fill the pores between cement particles and provide nucleation sites for hydration product precipitation, thereby promoting early hydration reactions and matrix densification [17]. Wang et al. [18] found that at a water-binder ratio of 0.3, the incorporation of 5–9% silica fume can enhance both the compressive and flexural strength of mortar; however, when the dosage exceeds a

certain range, the strength instead decreases. This indicates that excessive silica fume may reduce workability and disrupt the balance among hydration reactions, pore structure optimization, and polymer film formation. Almeida et al. [19,20] further demonstrated that the combined use of polymers and silica fume can significantly enhance mortar interfacial bond strength, suggesting a possible synergistic enhancement between organic polymer film formation and inorganic mineral filling/hydration regulation. In summary, HPMC, VAE, and silica fume can all improve the performance of cement-based repair materials to a certain extent; however, each component exhibits both beneficial and adverse effects. HPMC improves water retention and interfacial wettability but may reduce matrix strength; VAE enhances polymer film formation and interfacial bonding but may weaken compressive performance; silica fume refines pore structure and promotes hydration, but excessive incorporation impairs workability and affects polymer film formation [9]. Therefore, a single modifier is insufficient to simultaneously satisfy the comprehensive requirements of rapid-hardening repair materials in terms of workability, mechanical strength, and interfacial bonding performance.

At present, most existing studies have focused on ordinary Portland cement systems or single-polymer-modified systems. However, research on the ternary composite modification effects of hydroxypropyl methylcellulose (HPMC), redispersible vinyl acetate-ethylene (VAE) copolymer powder, and silica fume in sulfoaluminate cement-based rapid-hardening repair mortars remains limited. In particular, the dominant role of HPMC in regulating fresh-state properties and mechanical strength, the synergistic mechanisms between HPMC and VAE in enhancing early-age interfacial bonding performance, and the intrinsic relationships among polymer film formation, hydration product evolution, pore structure regulation, and macroscopic bonding performance have not yet been systematically elucidated. Based on this, the present study employs a sulfoaluminate cement-based rapid-hardening repair mortar as the matrix and adopts an $L_9(3^3)$ orthogonal experimental design to investigate the effects of HPMC, VAE, and silica fume on fresh properties, mechanical performance, and the interfacial bonding behavior between new and old mortars. Furthermore, scanning electron microscopy (SEM) and X-ray diffraction (XRD) analyses are conducted to reveal the underlying microstructural mechanisms, with the aim of providing a theoretical basis for mixture design and interfacial performance enhancement of rapid-hardening polymer-modified cement-based repair materials.

2. Materials and Methods

2.1. Materials

The cementitious materials used in this study include sulfoaluminate cement (SAC) and silica fume (SF), and their chemical compositions are summarized in Table 1. The SAC was supplied by Anhui Conch Group Co., Ltd., Wuhu, China, while the SF was obtained from Guangzhou Metallurgical Co., Ltd., Guangzhou, China. Hydroxypropyl methylcellulose (HPMC) was supplied by Nantong Dongfang Technology Co., Ltd., Nantong, China, with the model HPMC-10000; its main properties are listed in Table 2. The redispersible polymer powder was an ethylene-vinyl acetate (VAE) copolymer emulsion powder, manufactured by Shandong Science and Technology Co., Ltd., Jinan, China, with the model SWF-05; its main properties are presented in Table 3. The polycarboxylate superplasticizer, model HLX, was produced by Shandong Hongquan Construction Technology Co., Ltd., Jinan, China, with a water-reducing efficiency of 36.2%, an air content of 3%, and a bleeding ratio of 20%. The ISO standard sand was supplied by Xiamen ISO Standard Sand Co., Ltd., Xiamen, China, with a silicon dioxide (SiO_2) content of $\geq 98\%$. Tap water was used throughout the experiments.

Table 1. Chemical composition of cementitious materials/%.

Material	CaO	Al ₂ O ₃	SiO ₂	SO ₃	Fe ₂ O ₃	MgO	Loss
Cement	46.35	25.47	12.52	9.72	2.36	1.82	1.64
Silica fume	1.63	0.97	85.04	-	1.04	0.32	10.00

Table 2. Chemical properties of HPMC cellulose ether.

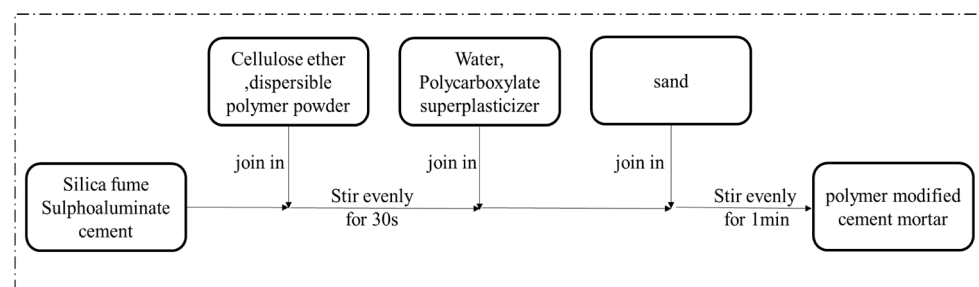
Material	Hydroxypropyl/%	Methoxy/%	Ash	Moisture	Viscosity (2% Aqueous Solution)
HPMC	8.4	20.8	3.03	4.5	13,332.0

Table 3. Chemical and physical properties of VAE redispersible polymer powder.

Item	Value
Polymer type	Vinyl acetate-ethylene copolymer
Protective colloid	Polyvinyl alcohol
Appearance	White powder
Bulk density	500.0 g/L
Non-volatile content	98.0%
Ash content	26.5%
pH value	7.0
Minimum film-forming temperature	0 °C

2.2. Methods

The preparation of repair mortar samples and tests of mortar performance (i.e., setting time, fluidity, and consistency and water retention tests) were carried out at $(20 \pm 2) ^\circ\text{C}$ and $(60 \pm 5)\%$ relative humidity. The preparation process of polymer-modified cement mortar is shown in Figure 1.


Figure 1. Preparation process of polymer-modified cement mortar.

The setting time tests refer to the standard GBT 1346-2011 cement standard consistency water consumption, setting time and stability test method [21]; the fluidity tests refer to the standard GBT 2419-2005 cement mortar fluidity measurement method [22]; the consistency and water retention tests follow the standard JGJ 70-2009 test method for the basic properties of building mortar [23].

The molding and curing of the mortar mechanical properties test's specimens were carried out at $(20 \pm 2) ^\circ\text{C}$ and $(90 \pm 5)\%$ relative humidity. The test standards for compressive strength and flexural strength refer to the standard GBT 17671-199 cement mortar strength test method (ISO method) [24].

The bond strength tests of repair mortar contain two test methods. (1) The bonding strength of the repair material is characterized by the flexural strength of the old and new

mortar specimens. A test piece with a size of 40 mm × 40 mm × 160 mm was prepared as a standard test piece. (2) The bond strength of the repair material is characterized by the splitting tensile strength of a cube mortar test block with a side length of 70.7 mm [25]. The shapes and dimensions of the corresponding test blocks are shown in Figure 2. The old mortar test block was prepared by using 42.5 grade ordinary Portland cement with a water-binder ratio of 0.5 and a mortar-sand ratio of 1:3. After 28 days of curing in the standard curing room, the mortar was cut from the middle with a cutting machine. After placing the half old mortar test block in the mold, new repair mortar material was poured to prepare a bonded test piece with a new and old mortar interface. Three test pieces were prepared for each group, and the test time included 1 day, 7 days and 28 days. The new and old mortar bonded specimens and test equipment are shown in Figure 3.

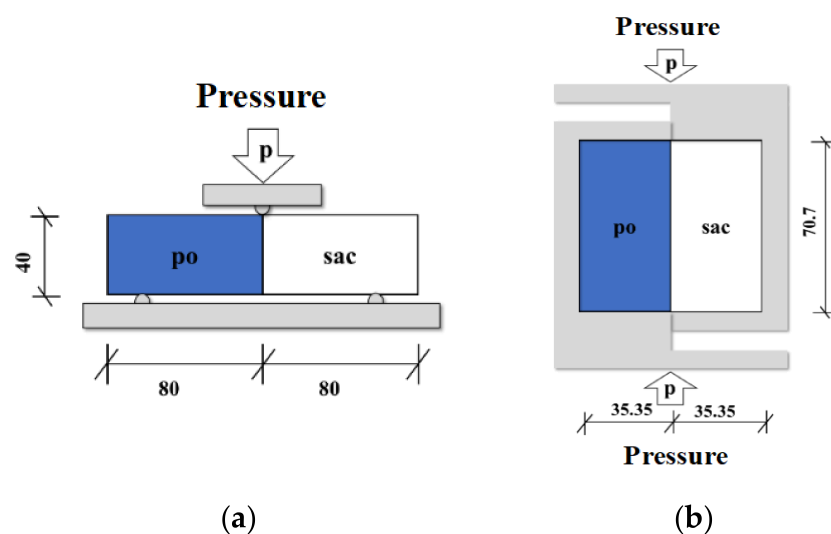


Figure 2. Geometry and size of specimens for different bond strength tests (unit: mm). (a) Interface bending tensile strength; (b) splitting tensile strength.



Figure 3. New and old mortar bonding specimens and field test equipment.

The microstructure of the interfacial region was observed using a eiss Gemini 300 scanning electron microscope (Carl Zeiss Microscopy GmbH, Jena, Germany). Before observation, the selected samples were immersed in absolute ethanol for 48 h to stop hydration and then dried in a vacuum oven (DZF-6020, Shanghai Yiheng Scientific Instrument Co., Ltd., Shanghai, China) at 40 °C to constant mass. The dried samples were sputter-coated with a thin gold layer to improve electrical conductivity. SEM observations were conducted under an accelerating voltage of 5–15 kV, and representative images were collected at magnifications ranging from 500× to 5000×, according to the observed microstructural features.

The mineralogical phases of the samples were characterized using an X'Pert PRO MPD X-ray diffractometer (PANalytical B.V., Almelo, The Netherlands) equipped with Cu K α radiation. The tube voltage and tube current were 40 kV and 40 mA, respectively. The diffraction data were collected over a 2 θ range of 5–60°, with a step size of 0.02° and a counting time of 0.5 s per step. Each sample was scanned once under the same testing conditions to ensure comparability among different mixtures.

2.3. Experimental Protocol

The orthogonal experimental design had three variable factors, i.e., the content of cellulose ether, redispersible latex powder, and silica fume, and three variable levels were set for each variable factor. The percentages of each factor were weighed separately, summarized in Table 4. A water-cement ratio of 0.3, a lime-sand ratio of 1:1, and a polycarboxylate water reducer of 0.2% of the cementitious material, were designated.

Table 4. Orthogonal test variation factors and variation levels.

Variation Factor		Level 1	Level 2	Level 3
Cellulose ether/%	A	0	0.2	0.3
Redispersible latex powder/%	B	0	0.3	0.6
Silica fume/%	C	0	5.0	10.0

For the three-factor and three-level test, the L9(3³) orthogonal table has been adopted, and the mixing ratio of the orthogonal test is shown in Table 5 below.

Table 5. Orthogonal test mix ratio.

Number	Water	Cement	Sand	Cellulose Ether (HPMC)	Redispersible Latex Powder (VAE)	Silica Fume	Superplasticizer
1	135	450.0	450	0	0	0	0.9
2	135	405.0	450	0	1.35	45.0	0.9
3	135	427.5	450	0	2.70	22.5	0.9
4	135	405.0	450	0.90	0	45.0	0.9
5	135	427.5	450	0.90	1.35	22.5	0.9
6	135	450.0	450	0.90	2.70	0	0.9
7	135	427.5	450	1.35	0	22.5	0.9
8	135	450	450	1.35	1.35	0	0.9
9	135	405	450	1.35	2.70	45.0	0.9

3. Results and Discussions

3.1. Fresh Performance of the Mortar

The fluidity, consistency, water retention rate, and setting time of the repair mortar were measured by the orthogonal test group, shown in Table 6. The fluidity and consistency of the mortar gradually decrease with the increasing HPMC, mainly classified into three gradients. When no HPMC (No. 1–3) is present, the fluidity is greater than 220 mm, and

the consistency is about 110 mm; when 0.9 kg/m³ HPMC (No. 4–6) is present, the fluidity is about 190 mm, and the consistency is about 80 mm; when 1.35 kg/m³ HPMC (No. 7–9) is present, the fluidity is about 170 mm, and the consistency is about 65 mm. Compared with those samples with HPMC, the samples without HPMC are much higher regarding both fluidity and consistency, indicating that the incorporation of HPMC in the mortar will have a significant impact on the fluidity and consistency. The water retention rate of the mortar in the table was found to be significantly related to the level of HPMC. In the nine groups of tests, the water retention rate of the repair mortar mixed with HPMC was above 99%, and the water retention rate of the specimens without HPMC was significantly lower than that of the test piece mixed with HPMC. The water retention rate of the mortar will gradually increase with the increase in HPMC content. In contrast, the presence of HPMC increases the water retention to up to nearly 100% when the HPMC reached 0.9 kg/m³. Comparing with the sample (No. 1) without any additional materials, a decrease occurs in each of the remaining samples (No. 2–9) regarding both initial and final sets, suggesting that the additions of any three materials may speed up the setting time.

Table 6. Table of results of fresh mixing performance test (All data are averages of the results).

Number	Cellulose Ether (HPMC)	Redispersible Latex Powder (VAE)	Silica Fume	Liquidity/mm	Consistency/mm	Water Retention/%	Clotting Time/min	
							Initial Setting	Final Set
1	1	1	1	295	114	95.21	29	32
2	1	2	3	220	102	96.69	25	30
3	1	3	2	255	110	97.36	22	26
4	2	1	3	184	71	99.94	22	28
5	2	2	2	190	90	99.84	22	30
6	2	3	1	190	84	99.78	25	33
7	3	1	2	174	70	99.60	24	31
8	3	2	1	170	63	99.80	23	33
9	3	3	3	166	65	99.68	20	26

The factor-effect curves and optimal ratios of mortar fluidity, consistency, and water retention rate are shown in Figure 4. The fluidity of the mortar gradually decreases with the increase in HPMC and silica fume content, and decreases first and then increases with the increase in VAE content. The results of range analysis show that the order of the significance of each factor on fluidity is cellulose ether (R1) > redispersible latex powder (R2) > silica fume (R3). The content of HPMC has a significant effect on the fluidity, while VAE and silica fume have little effect on the fluidity. According to the maximum and optimal mortar fluidity, the level combination of each influencing factor is selected as A1B1C1. Consistency will decrease with increasing HPMC. Variations in VAE content had little effect on consistency. Silica fume has little effect on the consistency, showing a trend of increasing first and then decreasing. The significance order of each factor's influence on consistency is R1 > R3 > R2. The best ratio of mortar consistency is A1B3C2. The water retention rate will gradually increase with the increase in HPMC and VAE content, and first increase and then decrease with the increase in silica fume content. The results of range analysis showed that the order of significance of the influence of each factor on water retention rate was R1 > R2 > R3. The content of HPMC had a significant effect on the water retention rate, while the content of VAE and silica fume had little effect on the water retention rate. The best ratio of water retention is A2B3C2. The strong influence of HPMC on fluidity, consistency, and water retention can be attributed to its high molecular viscosity and hydrophilic functional groups. After being dissolved in the mixing water, HPMC increases the viscosity of the liquid phase and forms a water-retaining network, which restricts free-water migration and improves the stability of fresh mortar. Consequently, the fluidity and consistency decrease with increasing HPMC content, whereas the water

retention rate increases markedly. This explains why HPMC showed the highest range value for fluidity and water retention in the orthogonal analysis [26].

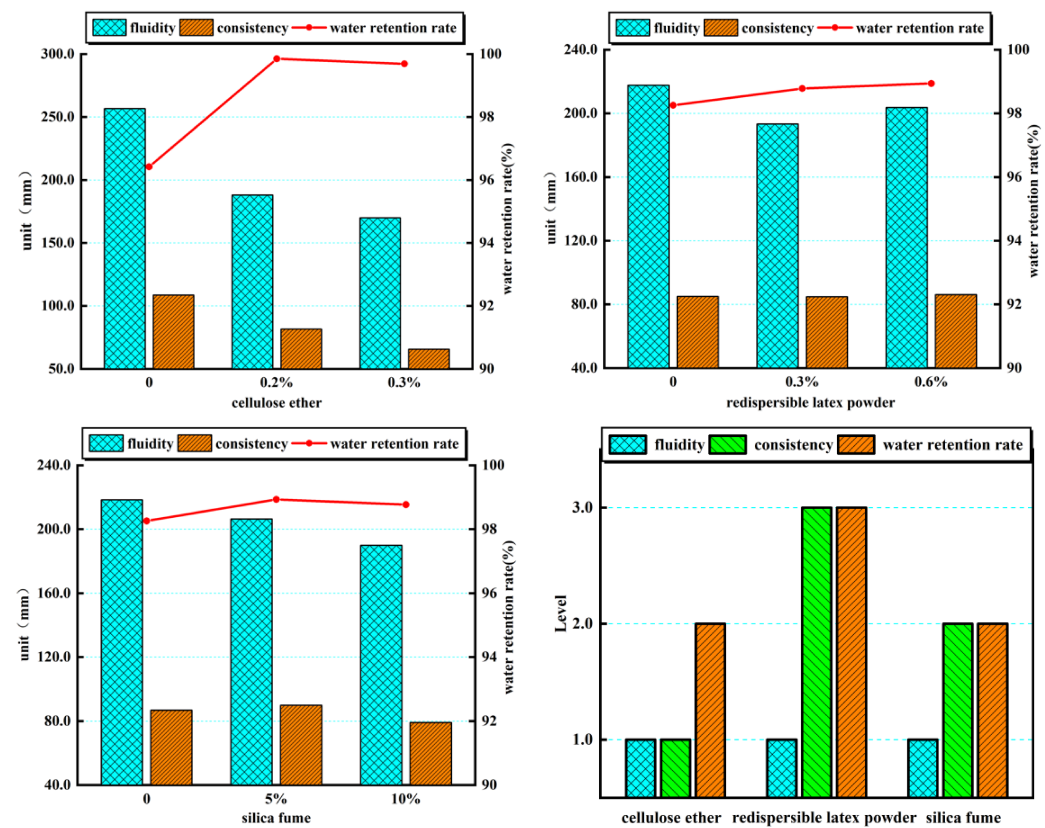


Figure 4. Effects of HPMC, VAE, and silica fume contents on the fluidity, consistency, and water retention of polymer-modified sulfoaluminate cement mortar.

The factor-effect curve and optimal ratio of setting time are shown in Figure 5, and the initial setting time will decrease with the increase in HPMC, VAE and silica fume content. The order of significance of the influence of each factor on initial setting time is $R3 > R1 > R2$, and the best ratio of initial setting time is A1B1C1. The final coagulation time was positively correlated with HPMC, and the final coagulation time increased with the increase in HPMC level. VAE had little effect on final setting time, showing a trend of increasing first and then decreasing. Silica fume has a significant effect on final setting time. The order of significance of each factor affecting final setting time was $R3 > R2 > R1$, and the best ratio of final setting time was A2B2C1. Silica fume mainly affects setting behavior. Its fine particles provide additional nucleation sites for hydration products and accelerate the precipitation of early hydration phases in the sulfoaluminate cement system. Therefore, the incorporation of silica fume shortens the initial and final setting time. VAE has a relatively limited effect on setting time because its main function is not hydration acceleration, but polymer particle dispersion and subsequent film formation [17,27]. According to research, cellulose ether can delay the hydration of ordinary Portland cement mortar [28], but for rapid-hardening sulphoaluminate cement, cellulose ether hardly affects its early hydration heat release [8].

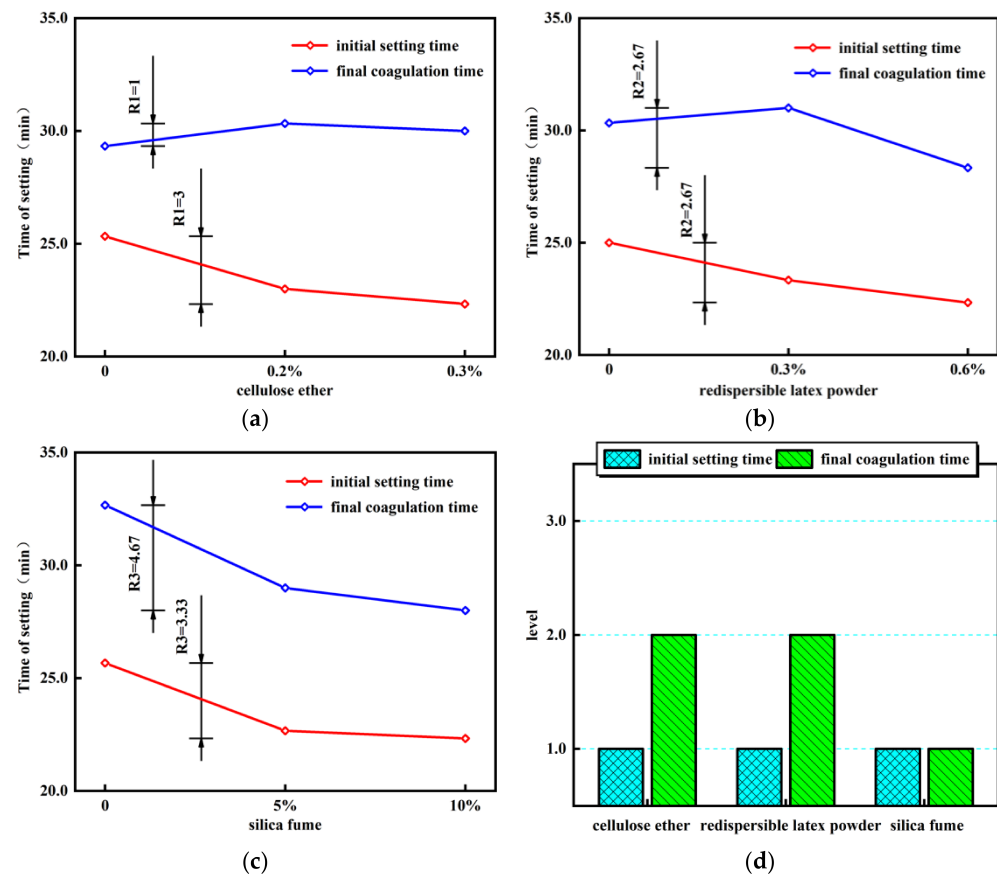


Figure 5. Effects of different factors on the setting behavior of polymer-modified sulfoaluminate cement mortar: (a) HPMC content; (b) VAE content; (c) silica fume content; (d) Comparison of factors.

3.2. Mechanical Properties

Table 7 presents the test results of compressive strength, flexural strength, interfacial flexural tensile strength, and splitting tensile strength of the repair mortar. In terms of mechanical performance, HPMC is the dominant factor affecting the compressive and flexural strengths at 1 d, 7 d, and 28 d, with the influence order being $R1 > R3 > R2$. As the HPMC content increases, both compressive and flexural strengths generally decrease. This is attributed to the fact that although the thickening and water retention effects of HPMC are beneficial for interfacial wetting and construction stability, excessive HPMC can hinder ion diffusion and the continuous growth of hydration products, and may introduce additional closed pores or weak interfacial films, thereby reducing the load-bearing capacity of the matrix. Therefore, HPMC exhibits a dual effect of “interfacial improvement” and “strength reduction”, which is the fundamental reason for its role as the dominant component. In contrast, VAE has a relatively minor effect on compressive strength, but it enhances interfacial bonding through its film-forming capability. During hydration and drying, dispersed VAE particles gradually coalesce into polymer films, which connect hydration products and fill local discontinuities at the interface between old and new mortar. Silica fume mainly contributes through a filling effect and pozzolanic reaction, refining the pore structure and promoting the formation of a denser hydration product matrix. Therefore, the mechanical performance of the repair mortar is governed by the balance between matrix strength and interfacial toughness.

The interface bonding strength is an important indicator of repair mortar, and excellent bonding performance can bring good results for engineering crack repair. It can be clearly seen from the table that when the bond strength is 1 d, the bond strength of the test group mixed with polymer is greater than that of No. 1. It is worth noting that the

maximum strength of No. 4 is 4.30 MPa, which is an increase of 138.9% compared with No. 1's 1.80 MPa. No. 2 has a minimum value of 2.30 MPa excluding No. 1, but it is still an increase of 27.8% compared with No. 1. The interface flexural tensile strength of 1 d and 7 d ages increases with the increase in age. For the interface flexural tensile strength of 28 d age, the interface flexural tensile strength of samples numbered 1–3 will increase with the increase in age. Overall, the samples numbered 4–9 will appear as a shrinkage phenomenon. The splitting tensile strength of the mortar is similar to the interfacial bending tensile strength.

Table 7. Mechanical properties of mortar (all data are averages of the results).

Number	Compressive Strength/Flexural Strength (MPa)			Interfacial Bond Strength (MPa)					
				Interface Bending Tensile Strength			Splitting Tensile Strength		
	1 d	7 d	28 d	1 d	7 d	28 d	1 d	7 d	28 d
1	49.77/9.93	58.43/9.66	77.75/9.47	1.80	4.10	5.60	0.87	1.94	3.12
2	42.41/7.89	53.21/7.30	69.54/8.54	2.30	3.90	4.10	1.48	2.08	2.10
3	44.04/7.55	51.10/7.22	61.61/7.90	2.80	4.40	4.90	1.93	2.32	2.24
4	27.73/5.45	33.26/4.81	37.55/6.02	4.30	4.20	3.80	2.36	2.50	2.16
5	31.01/5.90	35.98/5.34	42.68/6.10	4.20	4.90	4.50	1.92	2.64	2.87
6	32.54/5.76	40.55/5.95	44.95/6.75	3.50	4.70	4.20	2.65	2.98	2.93
7	28.40/5.22	34.11/4.72	38.16/5.72	2.80	4.30	4.00	2.07	2.78	2.58
8	27.75/4.98	36.34/4.50	40.43/5.61	3.40	4.60	4.10	2.10	2.57	2.02
9	26.32/5.40	27.13/4.93	32.79/5.10	3.80	4.10	3.90	2.27	2.71	2.48

3.2.1. Compressive Strength of Mortar

The compressive strength factor-effect curves and optimal ratio of mortar at 1 d, 7 d and 28 d are shown in Figure 6. The content of HPMC has a significant impact on the compressive strengths of 1 d, 7 d and 28 d, and the content of VAE and silica fume has little effect on the compressive strengths of each age. At any age, the significance order of each factor's influence on compressive strength is $R1 > R3 > R2$. It is found in the figure that the three factors have a gradually decreasing trend on the compressive strength of the mortar at different ages. According to research [9–12], HPMC will reduce the compressive strength and flexural strength of cement mortar, and VAE will reduce the compressive strength of the cement mortar, but is conducive to the improvement of flexural strength. Therefore, taking the maximum compressive strength as an index, the best mix ratios for 1 d, 7 d and 28 d compressive strengths are all A1B1C1.

3.2.2. Flexural Strength of Mortar

Figure 7 shows the effect curves and optimal ratio of the flexural strength factors of mortar 1 d, 7 d, and 28 d. Similar to the compressive strength, the content of HPMC also has a significant impact on the flexural strength of 1 d, 7 d, and 28 d. The trend of flexural strength decreases with the increase in HPMC content. But it is worth noting that the flexural strength of each factor 1 d will be greater than the flexural strength of 7 d, and less than the flexural strength of 28 d, resulting in the phenomenon of strength shrinkage. The content of VAE and silica fume has little effect on the flexural strength of each age. In the flexural strength of 7 d, 0.6% VAE is higher than 0.3%, which is not reflected in 1 d and 28 d. It may be because the content of VAE is not large, so the effect of improving the flexural strength is not obvious enough. At any age, the significance order of each factor affecting the flexural strength is $R1 > R3 > R2$. Taking the maximum flexural strength as an index, the best mix ratios of 1 d, 7 d and 28 d flexural strength are all A1B1C1.

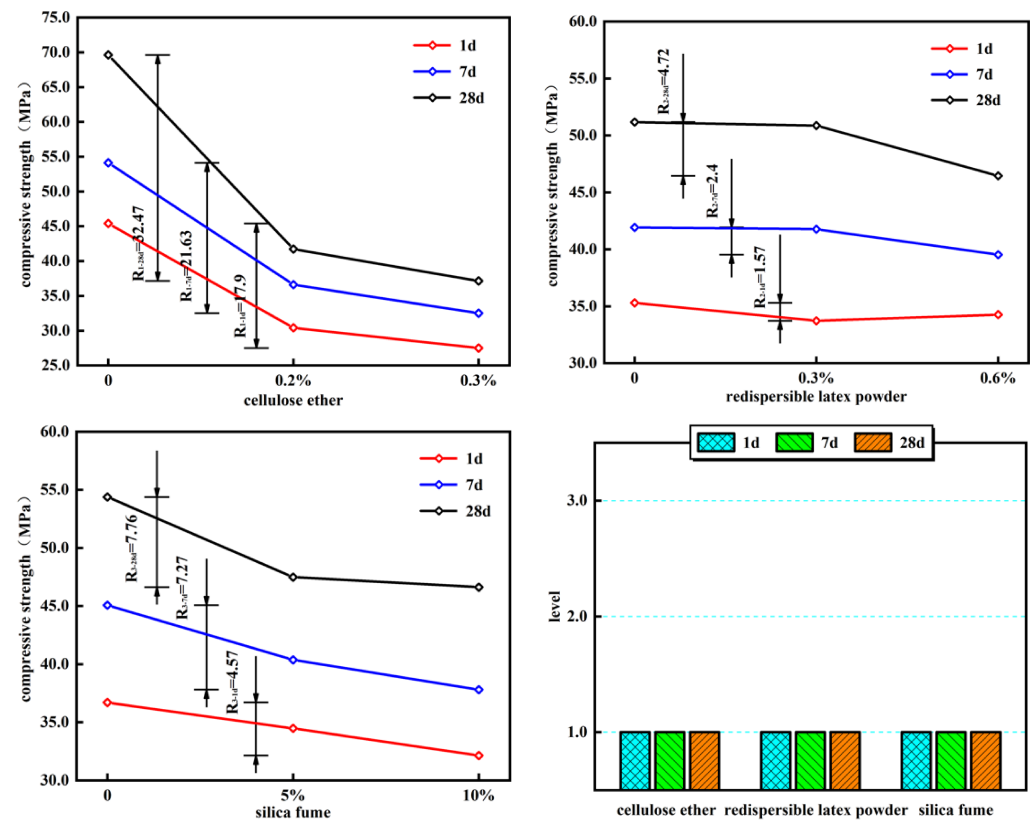


Figure 6. Factor-effect curves of compressive strengths of polymer-modified sulfoaluminate cement mortar at 1, 7, and 28 days.

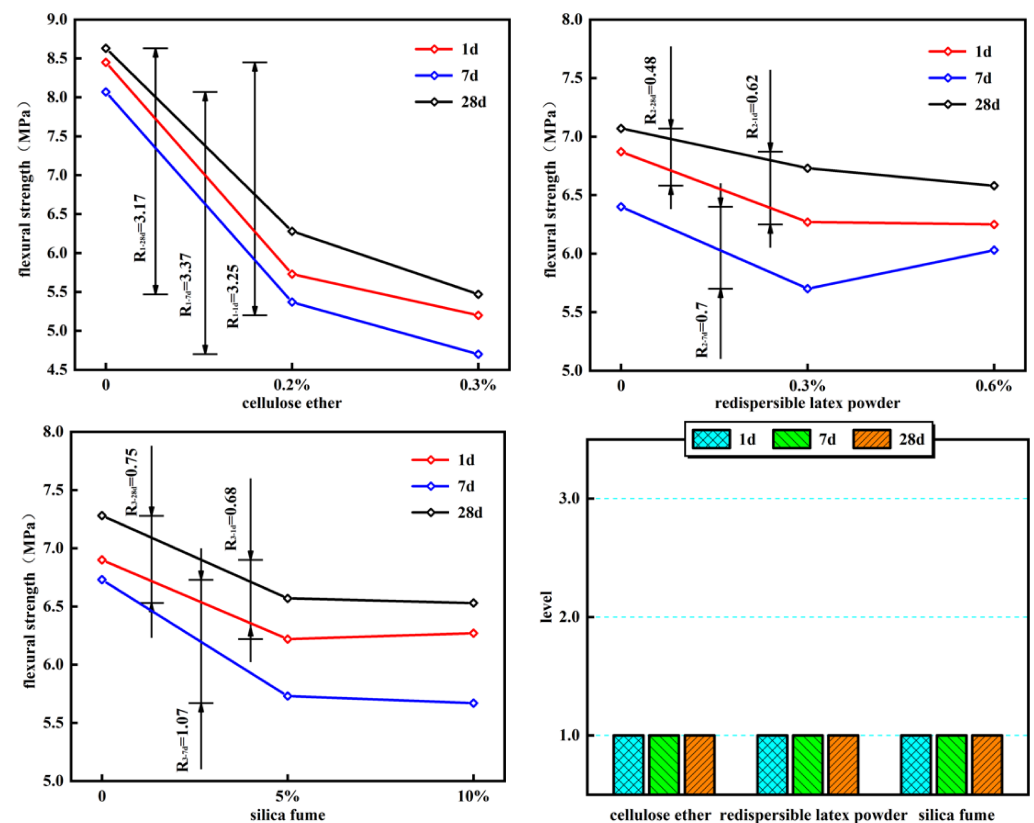


Figure 7. Factor-effect curves of flexural strength of polymer-modified sulfoaluminate cement mortar at 1, 7, and 28 days.

3.2.3. Interface Bending Tensile Strength

Figure 8 shows the interface bending and tensile strength factor-effect curves and optimal ratios of mortars at 1 d, 7 d and 28 d. From the bonding strength of mortar at 1 d, HPMC has a significant impact on the bonding strength, and the bonding strength reaches the maximum value when the content is 0.2%, while VAE and silica fume have little impact on the bonding strength, but the bonding strength increases with the increase in their respective levels. From the point of view of the bond strength of mortar at 7 d, there is no shrinkage phenomenon, but a trend of substantial improvement. The trend corresponding to HPMC and VAE is similar to the bond strength at 1 d. The bond strength corresponding to 5% silica fume content is the largest, and decreases when the content is 10%. From the point of view of the bond strength of mortar at 28 d, the bond strength is the highest when the content of the three variable factors is 0, and the bond strength at 28 d is in a downward trend after the variable factors are added. Because the focus of this study is on the rapid-hardening and early-strength cement mortar, the bond strength index at 1 d of age is more important. The analysis of range results shows that the ranking order of the significance of each factor on the 1 d bond strength is $R_1 > R_3 > R_2$, and the best mix ratio for the 1 d bond strength of mortar is A2B3C3.

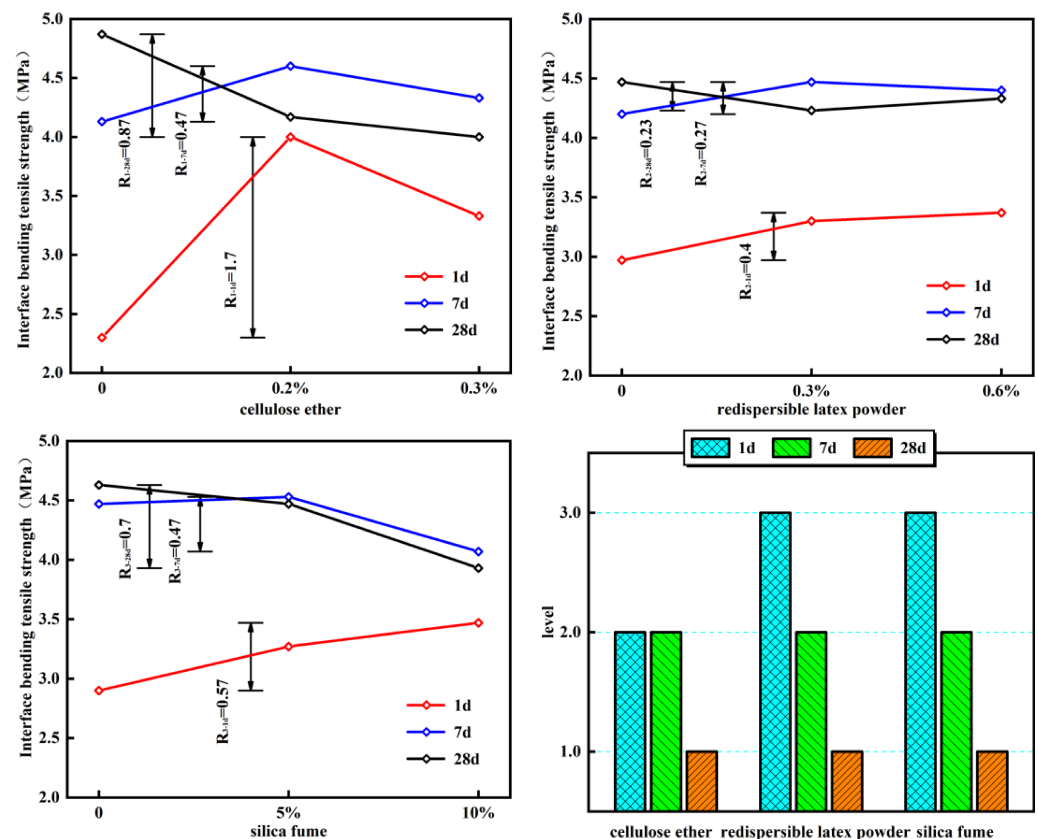


Figure 8. Effects of HPMC, VAE and silica fume content on the interfacial flexural strength between old mortar substrate and repair mortar at 1, 7 and 28 days.

3.2.4. Splitting Tensile Strength

Figure 9 shows the splitting tensile strength factor-effect curve and optimal ratio of mortar at 1 d, 7 d and 28 d. Judging from the splitting tensile strength of mortar at 1 d and 7 d, the influence of various factors on the strength is similar to that of the interfacial bending and tensile strength. HPMC has a significant effect on the bond strength. When the content is 0.2%, the bond strengths at 1 d and 7 d are both at maximum. The influence of silica fume on the bonding strength is small, the strength at 1 d increases with the

increase in content, and the strength at 7 d reaches the maximum when the content is 5%. VAE has little effect on bond strength, and the strength at 1 d and 7 d increases with the increase in content. Judging from the bond strength of mortar 28 d, the influence trend of VAE and silica fume on mortar is similar to that of interface bending and tensile strength. The difference lies in the influence of HPMC on the strength. The maximum value is when the HPMC content is 0.2%, which may be due to the difference caused by the influence of curing conditions. The analysis of range results shows that the ranking order of the significance of each factor on the 1 d bond strength is $R_1 > R_2 > R_3$, and the best mix ratio for the 1 d bond strength of the mortar is A2B3C3.

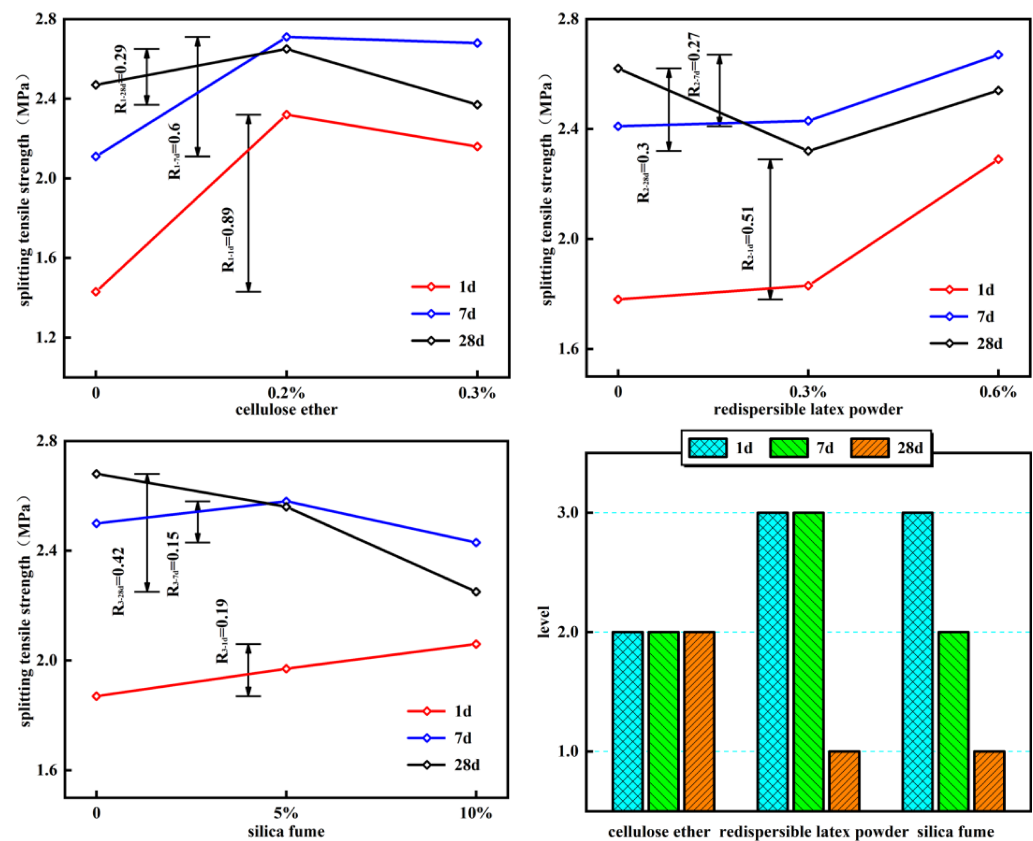


Figure 9. Effects of HPMC, VAE and silica fume content on splitting tensile strength between old mortar substrate and repair mortar at 1, 7 and 28 days.

According to the 1 d strength data measured by the two methods of interfacial bond strength and the range analysis, it is obtained that their optimal mix ratio is A2B3C3. This is because the synergistic effect of HPMC and VAE is primarily manifested in the interfacial zone. HPMC enhances the adhesion and water retention of the paste, providing a stable moisture environment that facilitates the uniform dispersion of VAE particles at the interface and their subsequent film formation. During water evaporation and ongoing hydration, VAE particles gradually coalesce into continuous or semi-continuous polymer films, which bridge the hydration products and the surface of the old mortar, thereby improving interfacial continuity. Silica fume, on the other hand, promotes the deposition of hydration products through micro-filling and nucleation effects, enabling the polymer film and inorganic hydration products to jointly form an organic-inorganic composite interfacial layer.

3.3. Microstructural Analysis

3.3.1. XRD Analysis

Figure 10 presents the X-ray diffraction (XRD) patterns of polymer-modified sulfoaluminate cement mortar for mixtures No. 1, No. 4, and No. 6 at the curing ages of 1 and 7 days. The main clinker phases of sulfoaluminate cement include ye'elimite (C_4A_3S) and dicalcium silicate (C_2S), which govern the early hydration behavior of the system [29]. Compared with the control mixture (No. 1), the intensities of quartz-related reflections in No. 4 and No. 6 are slightly higher, while the reflections associated with $CaSO_4$, $CaCO_3$, ye'elimite, C_2S , and gypsum show a slight reduction. This phenomenon is mainly attributed to the incorporation of silica fume in the modified systems, which contributes additional crystalline and amorphous silica phases rather than being generated through polymer-related reactions. Notably, the No. 4 mixture exhibits significantly stronger quartz reflections at both 1 day and 7 days compared with other groups, indicating a higher content of siliceous components and more pronounced micro-filling effects. Meanwhile, the gypsum reflections in No. 4 show a decreasing trend with curing time, suggesting a more efficient consumption of sulfate phases during early hydration and a faster formation of hydration products such as ettringite (AFt).

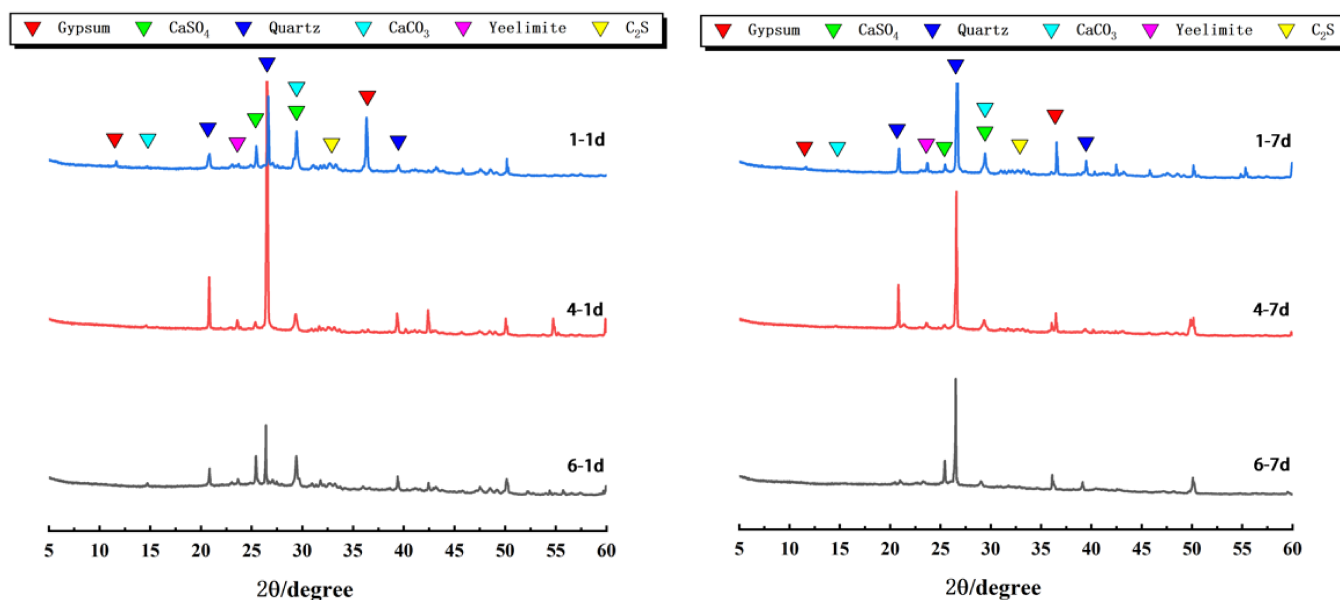


Figure 10. XRD patterns of groups at 1 d and 7 d, numbered 1, 4, and 6.

From a microstructural perspective, the enhancement in siliceous phase content and the accelerated hydration process contribute to the densification of the matrix and interfacial transition zone (ITZ). The refined microstructure reduces pore connectivity and promotes better mechanical interlocking between hydration products and polymer films, which is beneficial for stress transfer across the old-new mortar interface. At the macroscopic level, this microstructural evolution is consistent with the observed improvement in interfacial bonding performance. For the control mixture (No. 1), the interfacial flexural tensile strength and splitting tensile strength at 1 day are 1.8 MPa and 0.87 MPa, respectively, increasing to 4.1 MPa (+128%) and 1.94 MPa (+123%) at 7 days. This indicates that the improvement in early-age bonding performance is not caused by the formation of new SiO_2 crystals induced by polymer incorporation but rather results from the synergistic effects of silica fume-induced micro-filling, accelerated hydration, and polymer film formation.

3.3.2. SEM Analysis

Figure 11 shows the microstructure of a cement slurry without redispersible emulsion powder and cellulose ether (Figure 11(a1,a2)), a cement slurry with 0.2% cellulose ether (Figure 11(b11,b21,b12,b22)), and a cement slurry with 0.2% cellulose ether and 0.6% redispersible emulsion powder (Figure 11(c1,c2)) at 1 and 7 days of age. The AFt crystal in Figure 11(a1) was inserted into the cementification at an early stage to form a skeleton, thus providing high strength in the early stage of the hydration of sulfoaluminate cement [30]. Compared with cement pastes containing cellulose ethers and redispersible emulsions, the improvement in interfacial bond strength was smaller, which may be due to the uneven distribution of AFt crystals and the lack of polymer film coverage at the interface. Figure 11(a2) shows that, after 7 days, the cement slurry is more dense, the hydration products are densely coating on the cement interface, and the AFt crystals are wrapped by the hydration products of sulfoaluminate cement. The research [31–33] show that the main hydration products of cement paste are AFt, $\text{Al}(\text{OH})_3$, C-S-H, and $\text{Ca}(\text{OH})_2$, in which part of AFt is converted into sheet or layered crystals of monosulfate, thereby reducing the strength of the cement stone. Figure 11(b11,b12) shows that the HPMC cellulose ether polymer membrane structure is partially covering $\text{Ca}(\text{OH})_2$ and AFt. The AFt crystals in Figure 11(a1) are relatively slender and unevenly distributed, while the AFt crystals in Figure 11(b12,b22) are relatively small in length and diameter, showing a short rod shape and uniform distribution. According to research [22,34], ordinary cement paste reacts quickly when it meets water, so AFt crystals are relatively slender, while HPMC cellulose ether increases the viscosity of cement paste, reduces the binding rate of solution ions and delays the water reaching the surface of clinker particles, so the increasing trend of the aspect ratio of AFt is weak, and the morphology features are also short rods. Figure 11(c1) $\text{Ca}(\text{OH})_2$ surface micro-morphology observed a porous image. According to research [16], it may be that Ca^{2+} in $\text{Ca}(\text{OH})_2$ can react with $-\text{COOH}$ to generate calcium formate or calcium acetate. In Figure 11(c2), the AFt crystals appear as short rods and are uniformly distributed, and the polymer film can be clearly seen. Thus, compared to a, the microtopography is well improved due to the polymer films of b and c covering the interface, an observation that is consistent with the measured interfacial bond strength.

The SEM observations provide direct evidence for the improvement in interfacial bond strength. In the unmodified mortar, AFt crystals are relatively slender and unevenly distributed, and no continuous polymer film can be observed at the interface. Such a microstructure leads to local discontinuities and weak contact between old and new mortars, resulting in low 1 d bond strength. After the incorporation of HPMC, a membrane-like polymer phase partially covers $\text{Ca}(\text{OH})_2$ and AFt crystals, which improves the continuity of the interfacial region. When VAE is further introduced, the dispersed polymer particles gradually coalesce into a film, while the hydration products become more uniformly distributed.

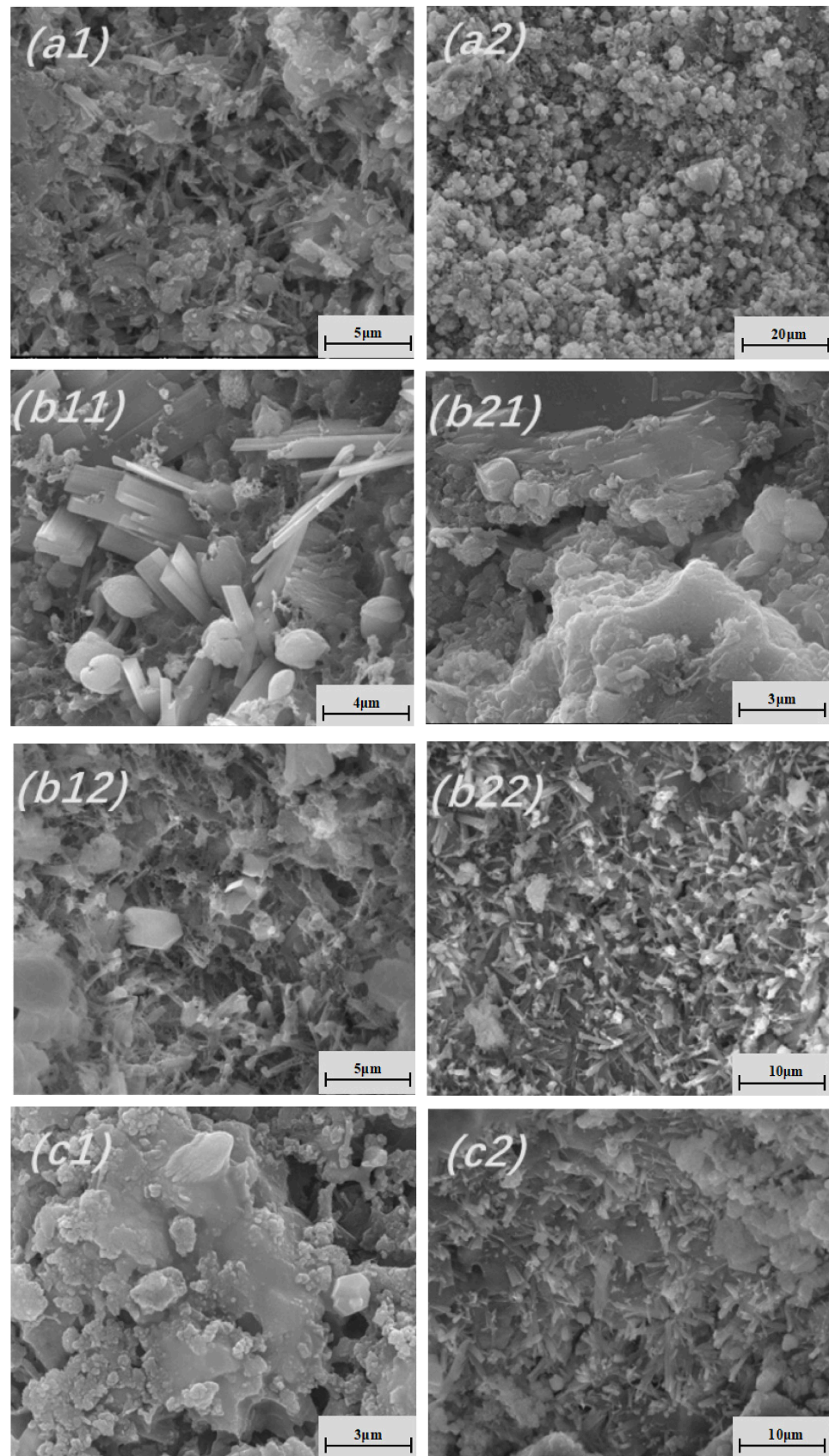


Figure 11. SEM images of the test part of the interfacial bond strength at 1 d and 7 d. (a1) 1–1 d, (a2) 1–7 d, (b11,b12) 4–1 d, (b21,b22) 4–7 d, (c1) 6–1 d, (c2) 6–7 d.

4. Discussion

The improvement in the early-age interfacial bonding of polymer-modified sulfoaluminate cement mortar can be explained by the coupled effects of polymer film formation, hydration regulation, and pore structure refinement. HPMC first dissolves in the mixing water and increases the viscosity of the liquid phase. This improves the water retention capacity of fresh mortar and reduces rapid water loss at the old-new mortar interface. A stable water environment is beneficial for the early hydration of sulfoaluminate cement and for the uniform distribution of VAE particles. During hydration and drying, VAE particles gradually coalesce into polymer films. These films cover hydration products and bridge micro-defects at the interface, thereby improving interfacial continuity. The polymer film also enhances the flexibility of the interface and reduces stress concentration caused by shrinkage or local defects. Therefore, VAE mainly contributes to bond strength rather than compressive strength. Silica fume contributes to pore refinement through its micro-filling and nucleation effects. Its fine particles fill voids between cement grains and provide nucleation sites for hydration products. Meanwhile, the active SiO_2 in silica fume reacts with calcium-containing hydration products to form additional gel phases, which densify the interfacial transition zone. The combined action of silica fume and polymer film formation produces a denser and more continuous organic-inorganic composite interfacial layer [27]. The detailed mechanism is illustrated in Figure 12.

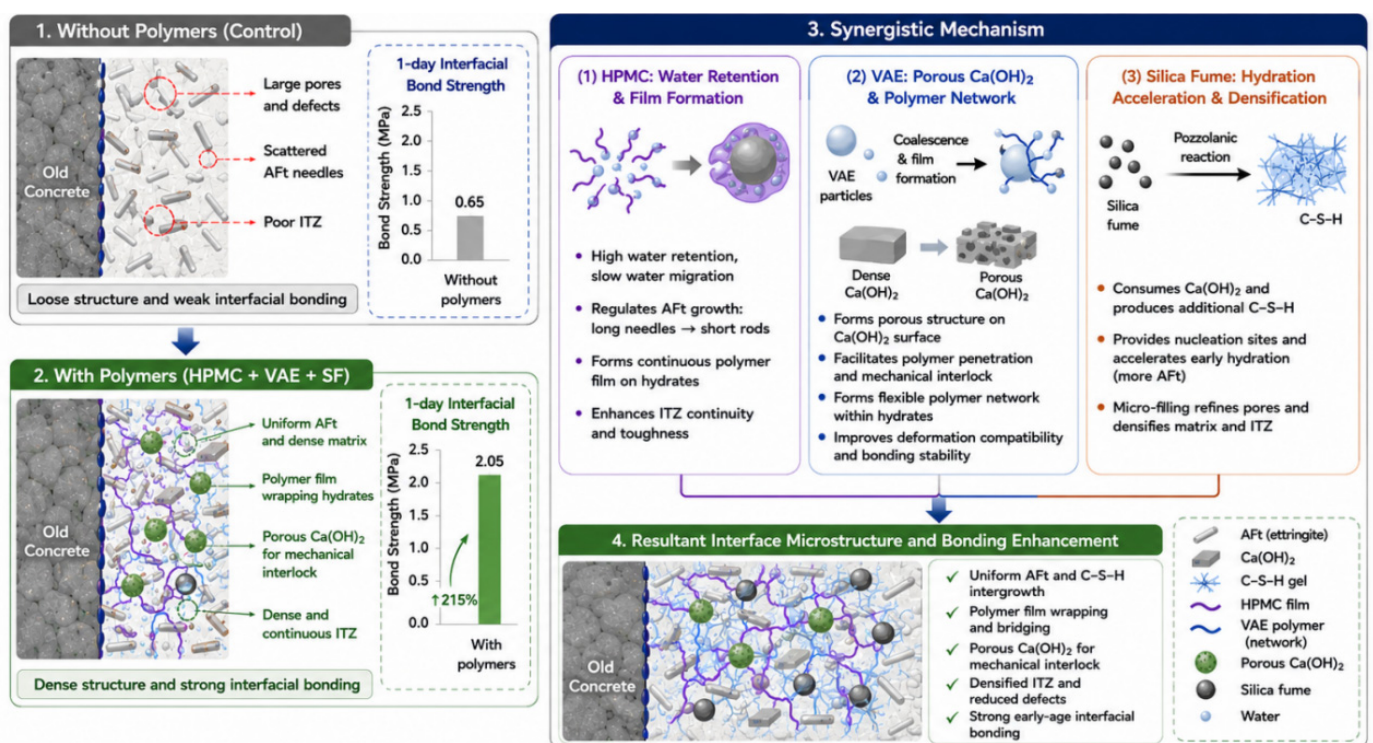


Figure 12. Analysis of the interfacial bonding mechanism of modified cement mortar.

Therefore, the optimized mixture A2B3C3 achieves superior early-age interfacial bonding because it balances three competing effects: HPMC provides water retention and adhesion, VAE forms a continuous polymer film, and silica fume refines the pore structure and promotes hydration. However, excessive HPMC may reduce matrix strength by increasing viscosity and hindering ion transport. This explains why HPMC is the dominant factor affecting mechanical strength, while the HPMC-VAE-silica fume combination is more effective in improving early-age interfacial bond performance [32].

5. Conclusions

This paper proposes a rapid-hardening and early-strength cement-based repair material modified by the polymer. Using polymer cellulose ether, polymer redispersible latex powder and silica fume as changing factors, the optimum formula of polymer-modified cement-based material was obtained by orthogonal experiment to optimize the mix ratio of repair materials. According to the test analysis, the main results are as follows:

- (1) HPMC is the dominant factor influencing both the workability and mechanical properties of the polymer-modified sulfoaluminate cement repair mortar. As the HPMC content increases, the fluidity and consistency of the paste decreases, whereas the water retention capacity increases significantly. This behavior is primarily attributed to its thickening and water retention effects, as well as the reduced ion migration rate.
- (2) Regarding compressive and flexural strength, the influence of HPMC is significantly greater than that of VAE and silica fume. An appropriate polymer dosage is beneficial for optimizing the interfacial structure; however, excessive HPMC may weaken the load-bearing capacity of the matrix by increasing paste viscosity and altering the pore structure.
- (3) HPMC and VAE exhibit a synergistic enhancement effect on early-age interfacial bonding performance. Orthogonal analysis indicates that the optimal mix proportion for 1-day interfacial bond strength is A2B3C3. At this proportion, the mortar exhibits a flowability of 200 mm, an initial setting time of 22 min, a final setting time of 33 min, and a 1-day interfacial bond strength of 4.5 MPa, thereby combining rapid hardening, early-strength development, and high flowability. This demonstrates that the incorporation of polymers significantly enhances the integrity and bonding capacity of the early-age repair interface.
- (4) XRD and SEM results indicate that polymer modification alters the distribution of hydration products and the interfacial microstructure of the sulfoaluminate cement system. SEM observations reveal polymer film coverage and ettringite (AFt) crystal refinement in the interfacial region, which may contribute to improved interfacial continuity and reduced sensitivity to local defects, thereby enhancing interfacial bonding performance.

Overall, the improved performance of the repair mortar does not arise from a single factor but rather from the combined effects of HPMC-induced water retention and thickening, VAE-induced polymer film formation, and silica fume-induced filling and nucleation. The synergy among these three components constructs a more homogeneous and denser interfacial transition zone, thereby significantly enhancing early-age interfacial bonding performance.

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