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Preparation and Performance Study of Cementitious Capillary Crystalline Waterproof Materials

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Abstract: Cementitious capillary crystalline waterproof materials (CCCW for short) offer durability and excellent waterproofing properties, making them a popular option for building waterproofing. Some scholars have studied the proportioning of such materials. However, these studies lack the relationship between the impermeability pressure of mortar and the components, and the mechanism of action is somewhat debatable. Therefore, we adopted a two-step method in our experiments. Firstly, we screened out the components that significantly impact impermeability from a variety of active components by orthogonal test. We then optimized the design of the active group ratio using the simplex lattice method. Lastly, we conducted a performance test of the optimal ratio and explored the waterproofing mechanism of homemade CCCW.

Keywords: Cementitious penetration crystalline waterproof material; Impermeability; Mechanism analysis; Optimization design

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1. Introduction

Concrete is widely used in construction due to its high strength, ease of construction, and relatively low cost. Concrete is a porous material that contains several types of pores. These pores can be classified according to their sizes as gel pores (< 10 nm), transition pores (10–50 nm), capillary pores (50–100 nm), and macropores (> 100 nm), where macropores are usually regarded as hazardous pores, which have the greatest impact on the leakage of concrete [1]. In addition to pores, the permeability of concrete is also affected by factors such as microcracks [2]. Cracking occurs when concrete is subjected to external factors such as temperature changes, loading, and shrinkage [3], These cracks are usually unnoticeable in the initial stage. However, when concrete is in a watery environment, moisture can spread along these cracks and capillaries, allowing harmful chemicals to seep inside and cause serious leakage problems [4,5]. The poor performance of domestic waterproofing materials leads to leakage of building structures, which causes huge economic losses [6]. Therefore, the study of waterproof materials can ensure the integrity and stability of the building structure and reduce maintenance

costs, which is of great significance in engineering and construction [7]. Cementitious capillary crystalline waterproof (CCCW) materials were first invented by the German chemist Lauritz Jensen in 1942 to solve the problem of leakage in cement ships [8-10]. Its main ingredients are silicate cement and quartz sand, and it also contains some additives [11–14].

Although there has been some research output on CCCW materials [15-18,11,7], there are still many shortcomings to these materials. From the analysis above, it can be seen that many active components can be used in CCCW materials, and there are different opinions on which components play a more significant role in impermeability, so it is essential to identify these components and optimize them. This paper summarizes the relevant studies on CCCW materials [15,19] and identifies common effective components that can react with Ca^{2+} to form insoluble crystals; substances that can form complexes with calcium ions, sulfates, calcium ion additives, and substances that can cause the formation of calcium carbonate. Therefore, seven additives, namely sodium silicate, sodium carbonate, sodium sulfate, complexing agent, calcium ion compensator, retarding agent, and thickening agent, are discussed in this paper, in which retarding agent and thickening agent are used to improve the performance of CCCW without participating in chemical reactions. In this study, a two-step testing process was designed. First, key additives affecting impermeability were identified using an orthogonal experimental design. Next, the simple lattice method was employed to examine the interactions between these components. The importance of each component was evaluated based on test results, leading to the determination of optimized ratios. These optimized ratios underwent performance verification through various tests: impermeability, compression, and water absorption. Additionally, XRD analysis was conducted to further evaluate the optimized ratios. Finally, the waterproofing mechanisms were investigated and characterized using XRD, SEM, and NMR testing methods, summarizing the findings from the macroscopic experiments.

2. Experimental

2.1. Raw materials

The cement used was P-O 42.5 ordinary silicate cement, which met the requirements of General Silicate Cement (GB175-2007) and was produced by Jiaozuo Qianye Cement Co. The sand was ISO standard sand, produced by Xiamen Aisiou Standard Sand Co., Ltd. CCCW was primarily composed of cement, sand, and additives mixed in specific proportions. The water used was laboratory tap water, which complied with the national standards.

2.2. Experimental design

The proportioning test was carried out in two steps. First, seven additives were screened using the orthogonal test method with impermeability pressure as the indicator. The additives comprised 8% of the total CCCW, with a cement-to-sand ratio of 2:1. The total mass of additives was determined and then adjusted proportionally so that their combined total equaled 1 (**see Table 2**). This ensured that the total amount of additives in each group remained the same and that each additive's ratio stayed consistent with the pre-adjustment values. The factor level table for the orthogonal test is shown in **Table 1**. The 28-day impermeability pressure value for the blank group specimen was 0.3 MPa. Analysis of polar deviation identified four effective components. Based on these findings, an optimized CCCW design was proposed using the simple lattice mixing method. This resulted in an experimental scheme with 15 different ratio groups. To ensure the reliability of the results, five groups from the scheme (groups 1, 4, 6, 7, and 12) were repeated (see **Table 3** for mix ratios). Assuming the total amount of each component as unit "1," the dosage ranges were set as follows: sodium carbonate 69–77% (mass fraction), sodium silicate 8–16% (mass fraction), complexing agent 10%-18% (mass fraction), and calcium ion compensator 5–13% (mass fraction). The performance of CCCW was then tested according to GB 18445 to

verify its waterproof effectiveness.

Table 1. Factors and levels for orthogonal test

Sample no.	Mix proportion/ %						
	Sodium carbonate	Sodium sulfate	Sodium silicate	EDTA-4Na	CaO	Retarder	Thickener
E1	20	7	3	4	2	0.5	0.05
E2	20	7	3	7	4	1	0.1
E3	20	13	6	4	2	1	0.1
E4	20	13	6	7	4	0.5	0.05
E5	42	7	6	4	4	0.5	0.1
E6	42	7	6	7	2	1	0.05
E7	42	13	3	4	4	1	0.05
E8	42	13	3	7	2	0.5	0.1

2.3. Analysis and testing

In accordance with the provisions of "Cement-Based Penetration Crystalline Waterproofing Materials" (GB 18445-2012) and "Cement Sand Strength Test Method" (GB/T 17671-1999), quantitative amounts of water, cement, and sand were poured into a mortar mixer and thoroughly mixed. The mortar was then poured into truncated conical metal molds (upper diameter: 70 mm, lower diameter: 80 mm, height: 30 mm) and plastic test molds (40 mm × 40 mm × 160 mm) for molding. The specimens were moved to a standard curing room for 1 day, demolded, and then had CCCW applied to their surfaces in two layers, with a total dosage of 1.5 kg/m² and a water-to-material ratio of 0.4. After application, the specimens were cured in water for 28 days. Finally, the cured specimens were tested for impermeability, water absorption, and compressive strength. The samples were soaked in anhydrous ethanol for 48 h to terminate hydration, ground and passed through a 200 mesh (0.075 mm) sieve, dried in an oven at 40 ℃, and then analyzed for hydration products using a Rigaku Smart-Lab X-ray diffractometer for the 28-d cured cement paste. The test conditions were as follows: scanning range of 5º–80º, scanning rate of 10 (º)/min, step size of 0.04º, tube voltage of 40 kV, and tube current of 150 mA.

The samples were cut into strips of size 5 mm × 5 mm × 10 mm, and the hydration reaction was terminated with anhydrous ethanol. They were then dried in an oven at 105°C for 24 hours. The samples were sprayed with gold, and the microscopic morphology of the hydration products was observed using a Merlin Compact scanning electron microscope manufactured by Carl Zeiss NTS GmbH, Germany. Additionally, samples were prepared as cylinders with a diameter of 15 mm and a height of 15 mm, cured for 28 days, and then removed. Hydration was terminated with anhydrous ethanol, and the samples were dried in a vacuum oven for 24 hours. The porosity of the concrete was analyzed using an AutoPore IV Mercury Piezometer manufactured by McMurray Tick Instruments Ltd.

3. Results and discussion

3.1. Determination of CCCW formulations

To improve the waterproofing performance of CCCW, this study evaluated its effectiveness using the 28-day impermeable pressure as an index. A two-step test method was employed to determine the composition of CCCW. First, the core components of CCCW were identified by constructing a seven-factor, two-level orthogonal table, and the results were analyzed using extreme difference analysis to screen the important factors

from the various influencing factors. Then, the simplex lattice method was used to optimize the ratios of these core components. Finally, the properties of the optimal CCCW ratio were tested according to GB 18445 to verify its waterproofing effectiveness.

3.1.1. Determination of core components

Table 2 shows the polar analysis of the orthogonal test using seepage resistance pressure as the index. The factors affecting the waterproofing effect, in order of influence from largest to smallest, are sodium carbonate, sodium silicate, complexing agent, calcium ion compensator, thickening agent, sodium sulfate, and retarder. Among these, sodium carbonate, sodium silicate, complexing agent, and calcium ion compensator have the most significant impact on impermeability performance. This significant effect is because sodium carbonate and sodium silicate are crystalline precipitants that provide carbonate ions to the mortar matrix. Sodium silicate reacts with the hydration product calcium hydroxide (CH) to form insoluble calcium carbonate crystals and hydrated calcium silicate (C-S-H gel). CH is a weak link in the hydration products of cement; its presence adversely affects the strength and durability of the mortar. The reaction produces a dense calcium carbonate precipitate and C-S-H gels, which fill pores and tiny cracks by consuming CH [20]. The complexing agent catalyzes the formation of induced crystals, generating calcium complexes within the matrix [21]. Due to its chemical instability, it forms insoluble crystals when encountering more stable carbonate and aluminate, with the active substances turning into free radicals. This continuous complexation-decomposition-complexation process provides conditions for long-lasting waterproofing effects [22]. The calcium ion compensator ensures a sufficient concentration of Ca^{2+} in the CCCW during the early stage of hydration, promoting rapid complexation reactions and enhancing the waterproofing performance.

Table 2. Visual analysis table of orthogonal tests

	Sodium carbonate	Sodium sulfate	Sodium silicate	EDTA-4Na	CaO	Retarder	Retarder Thickener
K1	358	417	383	383	383	408	425
K2	450	392	425	425	425	400	383
R	92	25	42	42	42	8	41

3.1.2. Formulation optimization for CCCW

The test results of mortar impermeability (as seen in Table 3) were subjected to multiple regression analysis. In the regression equation, the one-time term represented the change in a single factor, while the secondary term represented the interaction between two factors. The magnitude of the coefficient reflected the influence of each component on impermeability pressure. Notably, the one-time coefficient of the complexing agent was the largest, indicating its significant influence on impermeability performance among the four components. Conversely, the calcium ion compensator had the smallest coefficient, suggesting its relatively minor effect on impermeability performance.

The significance analysis of the interaction between the complexing agent and calcium ion compensator yielded a P-value < 0.05, indicating their significant interaction. Moreover, the secondary term coefficient was the largest, suggesting that Ca^{2+} complexes with the complexing agent to facilitate permanent waterproofing. However, the interaction between sodium carbonate/sodium silicate and the complexing agent/calcium ion compensator was not significant ($P > 0.05$), leading to rounded-off quadratic term coefficients.

Multiple regression analysis revealed a quadratic relationship between impermeability pressure and sodium silicate, complexing agent, sodium carbonate, and calcium ion compensator. A linear regression was

performed on these factors. The model yielded an F value of 48.43 ($P < 0.05$), indicating its significance. The adjusted R2 value was 0.96, and the predicted R2 value was 0.9, with a relatively small difference between them. This analysis suggests that the linear regression model effectively characterized the relationship between the four components and impermeability pressure, making it suitable for predicting their relationship. It can be expressed as follows:

$$Y = 0.8M_{\text{sodium carbonate}} + 0.99M_{\text{sodium silicate}} + 1.05M_{\text{complexing agen}} + 0.7M_{\text{calcium ion compensator}} + 2.13M_{\text{complexing agent}}M_{\text{calcium hydroxide}}$$

Y indicates the value of impermeable pressure, while $M_{\text{sodium carbonate}}$, $M_{\text{sodium silicate}}$, $M_{\text{complexing agent}}$, $M_{\text{calcium ion compensator}}$ represent the mass fractions of sodium carbonate, sodium silicate, complexing agent and calcium ion compensator, respectively.

In summary, the optimum mixing ratio of active substances was 0.77% sodium carbonate, 0.16% sodium silicate, 0.18% complexing agent, and 0.13% calcium ion compensator, and the maximum value of impermeable pressure was 1.1 MPa.

Table 3. Test results of impermeability of mortar with different mixing ratios

Sample No.	Sodium carbonate	Sodium silicate	EDTA-4Na	CaO	Impermeability pressure ratio/(%)
1*	0.73	0.12	0.1	0.05	300
2	0.7	0.13	0.11	0.06	333
3	0.71	0.1	0.12	0.07	400
4*	0.77	0.08	0.1	0.05	267
5	0.7	0.09	0.11	0.1	367
6*	0.69	0.16	0.1	0.05	333
7*	0.69	0.08	0.1	0.13	233
8	0.69	0.12	0.1	0.09	333
9	0.7	0.09	0.15	0.06	400
10	0.69	0.08	0.14	0.09	467
11	0.69	0.08	0.18	0.05	367
12*	0.69	0.12	0.14	0.05	367
13	0.73	0.08	0.1	0.09	300
14	0.74	0.09	0.11	0.06	333
15	0.73	0.08	0.14	0.05	367

Note: Tests 1, 4, 6, 7, and 12 were repeated to increase the accuracy of the results.

3.2. Performance of CCCW

3.2.1. Effect of CCCW coatings on the impermeability of mortars

Table 4 presented the changes in the impermeability performance of both blank mortar specimens and those coated with CCCW over a 28-day period. The findings indicate that mortar coated with CCCW demonstrates significantly higher impermeability pressure ratios compared to the blank specimens. This performance meets the requirements outlined in the national standard GB-18445-2012. Specifically, the mortar with CCCW coating exhibits a 28-day impermeability pressure ratio as high as 367%, signifying a notable improvement in impermeability performance. These results underscore the effectiveness of CCCW in enhancing impermeability and waterproofing effects.

Table 4. 28-d impermeability test results of mortar blank and coated specimens

Sample no.	Impermeability pressure (MPa)	Impermeability pressure ratio (%)	National standard (%)
blank sample	0.3	--	≥ 250
Blocks with CCCW	1.1	367	

3.2.2. Effect of CCCW coating on mechanical properties of mortar

Figure 1 illustrates the variations in mechanical properties observed in both blank specimens and those coated with CCCW under different curing times. The results indicate that the compressive strengths at 3, 7, and 28 days of CCCW-coated specimens are higher compared to the baseline specimens. Specifically, at the 28-day mark, the compressive strength of the coated specimens was 8% higher than that of the blank specimens. This suggests that coating the substrate surface with CCCW significantly enhances the compressive strength of the mortar. This improvement can be attributed to several factors. Firstly, calcium carbonate in CCCW reacts with calcium ions to produce water-insoluble calcium carbonate crystals. Secondly, sodium silicate reacts with calcium ions to generate more C-S-H gels, which fill internal pores and cracks. Lastly, the complexing agent promotes cement hydration, resulting in a denser internal structure and improved specimen strength, thereby indicating CCCW's beneficial compressive effect.

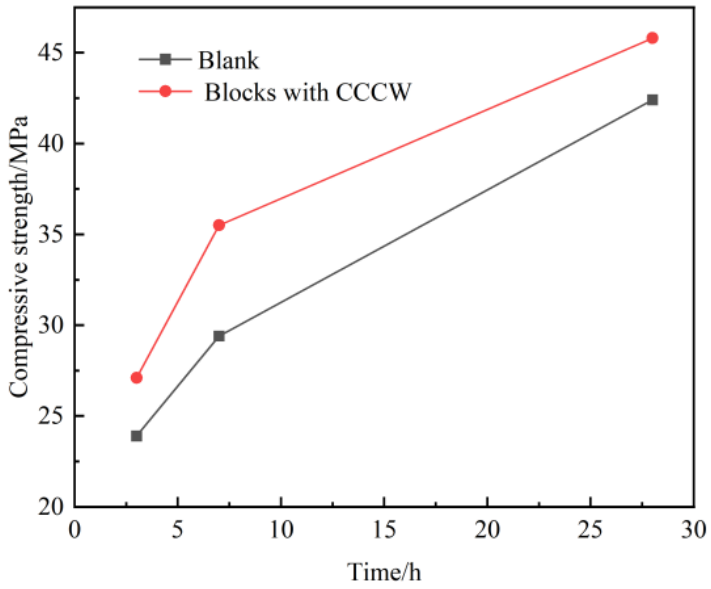

Figure 1. Mechanical properties of blank and CCCW-coated specimens under different curing times

3.2.3. Effect of CCCW coating on water absorption of mortar

Figure 2 illustrates the changes in water absorption observed in both blank specimens and those coated with CCCW under various immersion times. It is evident from the figure that as the immersion time increases, the water absorption rate of the CCCW-coated specimens gradually levels off. Within 48 hours, the coated specimens reach a state of water absorption saturation, while the blank specimens continue to absorb water. This suggests that the active substances in CCCW contribute to waterproofing the specimens. Additionally, regardless of the immersion time, the water absorption rate of the CCCW-coated specimens remains lower than that of the blank specimens. This indicates that the CCCW coating can penetrate the substrate, generate insoluble crystals, and block cracks and capillaries, thereby reducing the porosity of the substrate specimens and effectively waterproofing them.

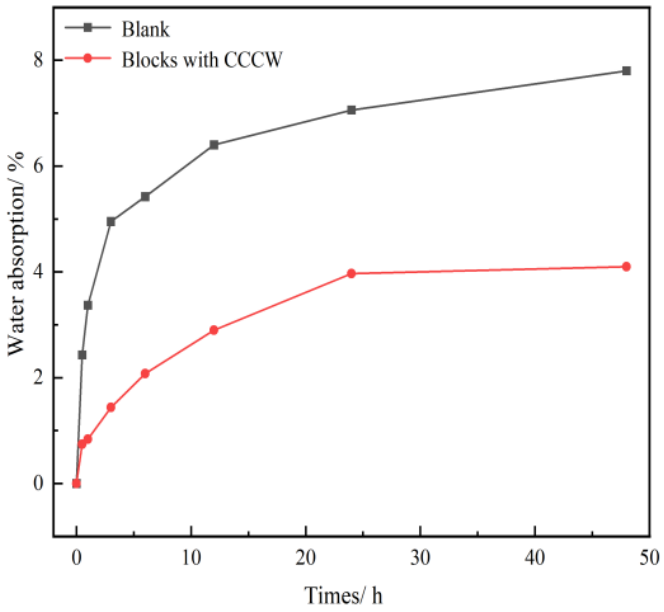

Figure 2. Water absorption test results for blank and CCCW-coated specimens

3.2.4. Effect of CCCW coating on the pore structure of mortar

Figure 3 depicts the piezometric analysis of both blank and coated specimens after 28 days of maintenance. The test results indicated that the porosity of the blank specimen and the waterproof coating with CCCW was 29% and 20%, respectively. This suggests that compared with the blank specimen, the CCCW coating effectively reduced porosity and significantly decreased the number of harmful holes. These findings underscored the significant improvement CCCW brought to the densification of cement mortar.

Observations from **Figure 3** revealed that after CCCW was applied to the matrix surface, the coated specimens not only reduced the number of gel pores and capillaries but also decreased the number of macropores. This further indicated that the active substances in CCCW could penetrate the mortar matrix, with the resulting crystals blocking the pores and cracks, thus enhancing the densification and impermeability of the mortar specimens.

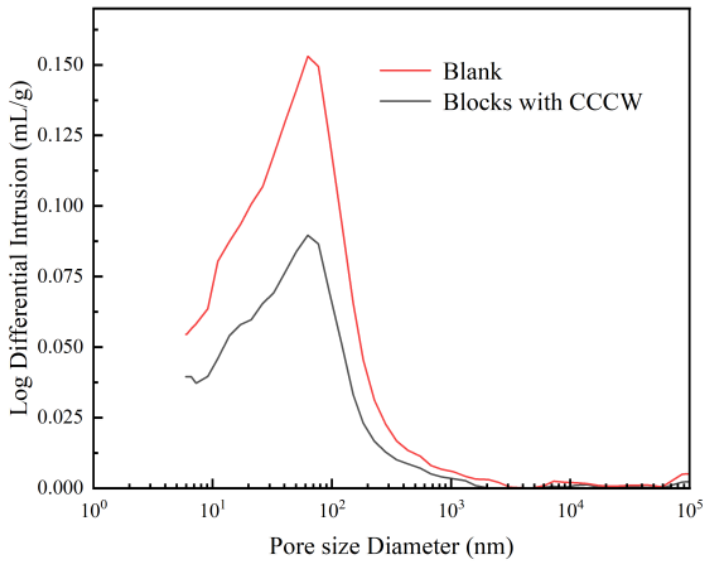

Figure 3. Pore size distribution of blank and coated specimens

3.2.5. Effect of CCCW coatings on mortar hydration products

Figure 4 displays the XRD patterns of hydration products from both blank and coated specimens after 28 days. The results revealed that the coated specimen exhibited stronger diffraction peaks for C-S-H and calcium carbonate compared to the blank specimen, while the diffraction peaks of CH showed a decreasing trend. This indicated that during the hydration process, the infiltration of active chemicals from the coated specimen consumed CH, resulting in the generation of more C-S-H gel and calcium carbonate. The generated insoluble crystals played a role in filling pores and cracks, thereby densifying the internal structure of the mortar and enhancing its impermeability.

Furthermore, the hydration products of both the blank and coated specimens were identical, suggesting that during the hydration process of CCCW, the generated compounds were consistent with the hydration products of cement. This indicated no compatibility issues between CCCW and the cement mortar matrix.

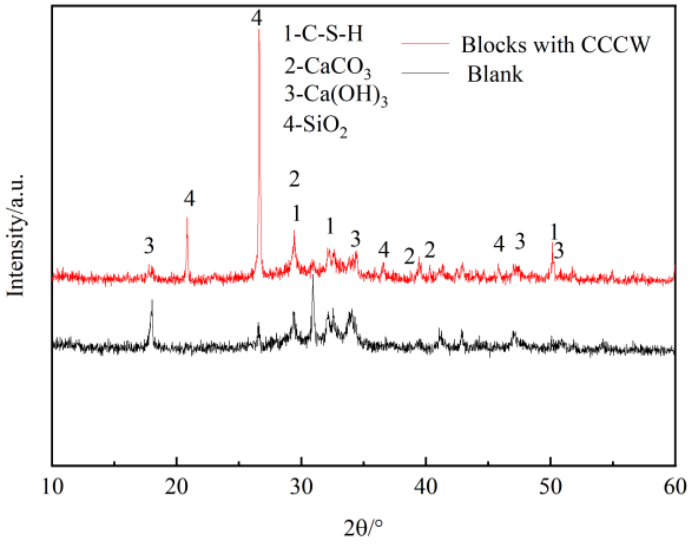

Figure 4. Results of 28 d XRD tests on blank and coated specimens

3.2.6. Effect of CCCW coating on the microstructure of mortar

Figure 5 compares the micro-morphology of both blank and coated specimens at various depths from the surface layer. Figures 5(a), (b), and (c) display SEM images of the blank specimen at 20 mm, 10 mm, and 5 mm from the surface layer, respectively. These images reveal the presence of unhydrated cement particles and non-uniformly distributed cracks, predominantly composed of calcium hydroxide crystals and hydrated calcium silicate gels, with no significant morphological features. In contrast, the overall structure of the coated specimens appeared denser (Figures (d), (e), and (f)), with dendritic crystals becoming more prevalent closer to the coating. This densification was attributed to the higher concentration of active substances in the surface layer, resulting in the generation of more dendritic crystals that clogged the pores and microcracks. Dendritic crystals were still present at 20 mm from the CCCW coating, indicating the superior waterproofing performance of CCCW to the mortar matrix.

This enhancement stemmed from the reaction between the active substances in CCCW and the hydration products of cement, which generated more insoluble crystals, filling and blocking pores and cracks, thereby densifying the internal structure of the mortar and further improving its impermeability performance. Meanwhile, the complexing agent acted catalytically and did not participate in the hydration reaction. However, when the complexing agent penetrated the matrix interior, it combined with calcium ions to form a complex that was easily soluble in water. This cyclic process provided conditions for permanent waterproofing.

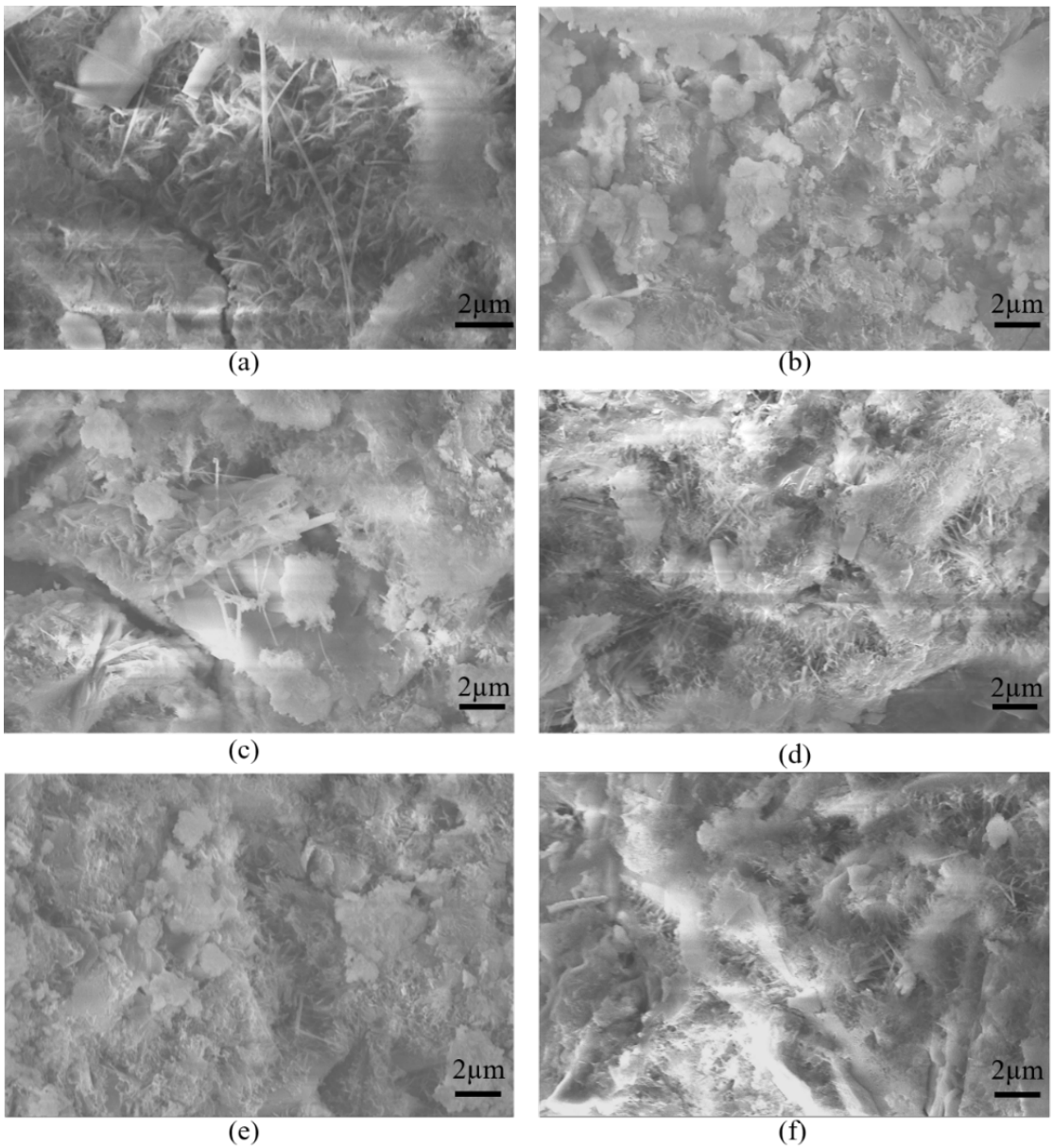

Figure 5. SEM images of blank specimen and coated specimen

4. Conclusion

(1) The orthogonal test method was initially employed to screen factors with significant influence on impermeability. Within the component design range, sodium sulfate, retarding agent, and surfactant had minimal impact on impermeability performance. Conversely, the complexing agent, sodium silicate, sodium carbonate, and calcium ion compensator exhibited more substantial effects on mortar impermeability.

(2) EDTA-4Na, sodium silicate, sodium carbonate, and calcium ion compensator were selected as influencing factors for mixing design. Multiple regression analysis was conducted on the test results. The optimal ratio for achieving maximum anti-seepage pressure was determined to be 0.77% sodium carbonate, 0.16% sodium silicate, 0.18% EDTA-4Na, and 0.13% calcium ion compensator. A quadratic function relationship was observed between impermeability pressure and sodium silicate, EDTA-4Na, sodium carbonate, and calcium ion compensator. This relationship effectively characterized the

impermeability pressure and each component. Comparing the coefficients in this function revealed that the interaction between EDTA-4Na and calcium ion compensator had the most significant effect on mortar impermeability. This indicates that the interaction of these two waterproofing mechanisms can be well understood in the CCCW reaction process.

(3) The optimized ratios complied with relevant national specifications across all performance indexes, exhibiting excellent waterproofing performance, improved mechanical properties, and effective reduction in the number of harmful pores. XRD and SEM analyses revealed that the infiltration of active substances consumed CH, resulting in the generation of more hydrated calcium silicate to fill capillary pores and microcracks within the mortar. This densified the internal structure of the mortar, achieving a better waterproofing effect.

Disclosure statement

The authors declare no conflict of interest.

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