

Review

# A Review of Waterborne Polymer–Cementitious Composite Repair Materials for Application in Saline Soil Environments: Properties and Progress

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**Abstract:** The properties of a large number of concrete infrastructures in China are deteriorating year by year, raising the need for repairing and strengthening these infrastructures. By introducing waterborne polymers into a cement concrete system, brittle cracks and easy bonding performance defects of concrete can be compensated for to form a long-life, semi rigid, waterborne polymer-modified cementitious repair material with a promising development prospect. This paper investigates the modification effect of polymer emulsions on ordinary cement mortar. Our research mainly focused on the physical and mechanical properties, durability, microstructure and application status of waterborne polymer-modified cementitious composites. Literature studies show that with the increase in waterborne polymer content (0 wt%–20 wt%), the performance of cement mortar significantly improves, which in turn expands its application range. Compared with ordinary cement mortar, the introduction of waterborne polymers blocks some of the pores in the cement to a certain extent, thus improving its permeability, freeze–thaw resistance and durability. Finally, this paper describes the application of waterborne polymer–cementitious composites in western saline soil environments, as well as discusses the prospects of their development.

**Keywords:** waterborne polymers; cementitious; repair materials; bonding; durability



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## 1. Introduction

During the construction of a national economy, concrete is one of the constructional materials with the widest application and the most abundant use in project works. A new and enormous market demand for concrete as well as cement products has emerged in recent decades, leading to a significant increase in the quantity of concrete produced. Nevertheless, because concrete is a nonhomogenous, multiporous and highly permeable engineering material, the combination of loading and a saline soil environment will accelerate the corrosion of concrete structures, which will have a significant impact on the service life of buildings [1,2]. There are currently about 617,000 bridges in the United States, and approximately 7.5% of them are structurally defective [3]. According to relevant data, the cumulative demand for bridge repairment in the U.S. is USD 125 billion [3]. This condition would need to be improved by increasing the bridge rehabilitation expenditures from USD 14.4 billion per year to USD 22.7 billion per year, which represents a 58% increase [4]. The current rate of investment would require waiting until 2071 to finish all the necessary restoration [3]. Over the next 50 years, the tendency to concrete deterioration seems irreversible. For a considerable amount of time to come, it is expected that the worldwide construction industry will switch from establishing new infrastructures to renovating and repairing existing infrastructures. This provides the opportunity to grow a foreign market for the development and use of repair materials [5,6].

The advantages of both inorganic and organic materials were merged in waterborne polymer–cementitious composites, which show promise for the field of concrete rehabilitation. In this type of composites, a portion of the cement is replaced with waterborne polymers, and the three-dimensional spatial network structure [7] that forms inside the cement mortar fills in some of the pores [8] to create a dense structure. This significantly improves concrete’s adhesion [9–11], flexibility [12], resistance to acid and alkali corrosion [13–15] and impermeability [16,17].

This paper focuses on the modification of common cement mortar with a waterborne polymer emulsion. Our research mainly involves the characterization of the physical and mechanical properties, durability, microstructure and application status of waterborne polymer–modified cementitious composites.

## **2. Progress of Domestic and International Research on Water-Based Polymer–Cementitious Composite Repair Materials**

### *2.1. Progress of Research on Water-Based Polymer–Cementitious Composites*

Water-based polymer–cementitious composites made their first appearance in 1923, when the British scholar Cresson [18] initially incorporated natural rubber emulsions in the form of fillers into road materials, which led to the proposal of polymer-hardened cement systems. In 1924, Lefebure et al. [19] first proposed the concept of natural rubber emulsion-modified cement mortar (concrete). In the 1930s and 1940s, natural rubber was transformed from its initial single form into various types of synthetic latexes (e.g., neoprene, polyacrylate, etc.) and resins, which are widely used in bridge deck covering, paving, flooring, and so on [20–22]. Polymer impregnation, a recently invented method to produce polymer-modified mortar, was actively implemented towards the end of the 1960s. This technology was formally introduced in China in 1973 and showed positive results in projects involving chemical flooring, gas pipelines, the improvement of salt and brine corrosion resistance, explosion-proof components, etc. However, it has limitations related to the complex processes it involves, the reuse of residual monomers, cost-effectiveness, and its few practical applications [15]. From 1970 to 1980, concrete–polymer composites gradually became the main alternative to traditional concrete in specific applications (e.g., wastewater treatment systems, surface treatment and repair works) in Japan, Europe and the United States [23]. In the late 1980s, it was found that polymer-based and dry materials can easily bind; therefore, polymer-based admixtures, hardeners, and cross-linking agents have progressively come to the attention of the general public [24].

### *2.2. Waterborne Polymer–Cement Mortar Composite Material*

#### *2.2.1. Workability*

In principle, it seems reasonable that mortar modified with waterborne polymers would flow with greater ease than ordinary cement mortar. This is mainly due to the fact that the hydrophobic groups contained in the polymer chains can help to improve the air-entraining properties of mortar and also reduce the shear stress generated during mixing [18]. Furthermore, polymer particles are encircled by other polymer particles, which creates the ‘micro-bead effect’, lowering the friction inside the cement matrix by creating ‘ball bearings’ [25]. Simultaneously, the surfactant incorporated into the latex to mitigate suspension segregation functions as a water reducer [26], considerably diminishing the amount of water required. This is reflected in the improved fluidity and decreased viscosity of the altered cement mortar. For example, Bi et al. [27] found that the larger the amount of styrene–butadiene rubber (SBR) used, the more obvious the increase in fluidity. When the polymer/cement ratio (P/C) reached 15%, the flowability reached a maximum of 270 mm, which was a significant increase of 145.5% compared to flowability of the blank control group. The same results were also obtained by Ukrainczyk et al. [28]. They found that the modified cement mortar’s consistency reached its maximum value of 195 mm with 9% of styrene–butadiene latex, which was 30% higher than the original modified cement mortar’s consistency (150 mm). Wang et al. [29] found that the styrene–acrylic emulsion

(SAE) powder has a certain water reduction effect. When the amount of doped SAE was below 3%, the water reduction rate increased linearly and reached the highest value in the presence of 3% SAE, which represented a 24% increase. There was no discernible rise in the water reduction rate with a further increase in the SAE dosage. In addition, the SAE powder showed a certain water retention ability as well. This was manifested by the fact that as the dosage increased, the water retention rate continued to improve and reached a maximum with 20% SAE, showing an increase of 16.5% compared with the water retention rate of ordinary mortar. Wong et al. [30] analyzed the ease of use of polymer–cementitious composites (PMCM) and found that the styrene–acrylic emulsion (SAE) can improve the flowability and toughness and reduce the water absorption properties of common silicate cement (OPC) or calcium sulphoaluminate (CSA)-based materials. Kim et al. [31] concluded that the incorporation of less than 2% of polyvinyl alcohol (PVA) in plain cement mortar can reduce the viscosity of the composite. Allahverdi et al. [32] investigated the effect of different water/cement ratios and polymer/cement ratios on the usability of modified cement mortar. They found that a small amount of PVA can improve the flowability of cement mortar.

Lu et al. [33] compared the effects of three polymer emulsions derived from monomers, methacrylic acid (C2), sodium persulfate (S2) and 2-acrylamido-2-methylpropanesulfonic acid (T2). The authors found that the flowability of the cement paste decreased (25–50%) and the plastic viscosity increased (25–250%) with the increasing polymer dosage.

This contrasting rheological properties can be attributed to differences in the apparent viscosity of the polymer used, the solids content of the suspension, the surface charge of the polymer, the polymer particle size and the glass transition temperature [34]. Some water-soluble latexes agglomerate in the gelling matrix, which tends to increase the viscosity of the interstitial phase [35]. In addition, polymers with higher viscosity and solids content tend to reduce the fluidity of the cement matrix compared to polymers with lower viscosity and solids content.

For polymer-modified cement mortars with increased plastic viscosity and poor ease of use, Kim et al. [36] suggested that pre-wetting methods or nano-SiO<sub>2</sub> can be used to improve the ease of use.

In summary, there is an ongoing debate on the impact of polymers on the rheological properties of cement mortars. Furthermore, cement will eventually solidify and harden, turning from a plastic slurry to a stone-like body with a certain strength after being mixed with water. At the same time, this process is accompanied by phenomena such as exothermic hydration, volume change and strength growth, which indicates that a series of complex physical, chemical and physicochemical changes are produced. In addition, the reaction of polymers in cement mortars is not completely controllable. Further research should address the effect of polymers on the flow properties of cement mortar materials to ensure the control of the polymer effects.

### 2.2.2. Mechanical Properties

To achieve the long-term stability of a construction, appropriate mechanical qualities must be available. While it is cheap and compressive, ordinary cement mortar is not very flexible. Materials made of organic polymers are more flexible and have stronger bonds and better water retention. Thus, the performance of cementitious materials can be effectively improved with the addition of polymers, which has been a topical research issue in recent years [37].

Pang et al. [38] investigated the effect of self-emulsifying epoxy resin (EEP) and externally emulsifying epoxy resin (NEP) dosing from 5% to 80% on the mechanical properties of cement mortar. The findings demonstrated that while the extensibility increased, the compressive strength of cement mortar tended to decrease with the increase in the resin content. Apparently, a stronger toughening effect of NEP compared to EEP was observed; moreover, the flexural strength of NEP-modified cement mortar reached a minimum value,

which was still higher than the flexural strength of the whole group of EEP-modified cement mortars after 28 days, with an epoxy resin amount of 40%.

Wang et al. [39] measured the compressive and flexural strengths of cement mortar modified with styrene–butadiene latex (SBR) under various curing conditions (wet curing and mixed curing). The results showed that, under wet and mixed curing circumstances, the compressive and flexural strengths of the composites increased initially, then dropped as the polymer dosage increased, and finally remained almost constant. However, in general, the flexural strengths increased. Furthermore, the longer the curing time, especially in mixed settings, the greater the strength at the same polymer/cement ratio. For example, adding 8% of styrene–butadiene latex (SBR) increased the flexural strength at 28 days of age under mixed curing conditions by approximately 25.4% compared to the value obtained in wet curing conditions.

Fan et al. [40] created a modified cement mortar by including carbon nanotubes (CNTs) and the styrene–acrylic emulsion (SAE) and then conducted trials to measure the mortar's compressive and flexural strengths. It was discovered that the addition of the styrene–acrylic emulsion (SAE) significantly increased the mortar's flexural strength while having the reverse impact on its compressive strength. When it came to modified cement mortar with ages of 3, 7 and 28 days, a 15% SAE content produced an approximately 15%, 11 and 12% higher flexural strength compared to that of regular cement mortar, respectively. At this polymer/ash ratio, the modified cement mortar's compressive strength value was minimized concurrently.

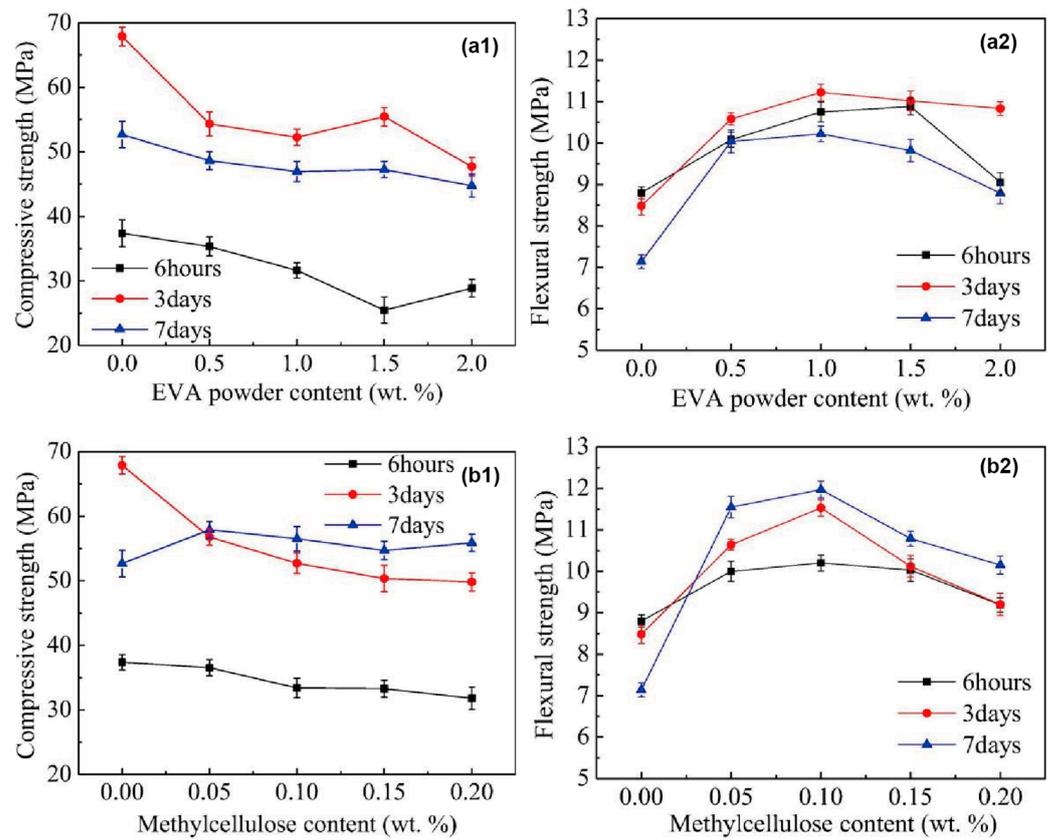
Chen et al. [41] examined how ethylene vinyl acetate (EVA) affected cement mortar's mechanical strength. Figure 1 demonstrates how the composites' compressive strength steadily decreased when the EVA dose was increased at the ages of 6 h, 3 days and 7 days. The decrease in the composites' compressive strength after 3 days was more pronounced when the poly/ash ratio remained constant at 0% wt~2% wt. When the EVA content was 1.0%, the modified mortar's flexural strength peaked, showing a tendency to increase before decreasing. At this point, the flexural strength at the 6 h, 3 d and 7 d ages increased by 25%, 37.5% and 42.9%, respectively, in comparison to that of regular cement mortar.

Bi [42] investigated the impact of waterborne polyurethane on the mechanical characteristics of cement mortar at various cement-to-polymer ratios. According to the results, which are displayed in Figure 2, ordinary mortar has a compressive strength between 67 MPa and 72 MPa, whereas waterborne polyurethane–cement mortar has a compressive strength between 48 MPa and 56 MPa. Additionally, when waterborne polyurethane was added, the compressive strength of the mortar after 28 days was about 30% lower than that of ordinary mortar. In contrast, the materials' flexibility and stickiness were enhanced by the addition of waterborne polyurethane; after 28 days, the flexural strength and tensile strength reached 12 MPa~14 MPa and 6 MPa~8 MPa, respectively.

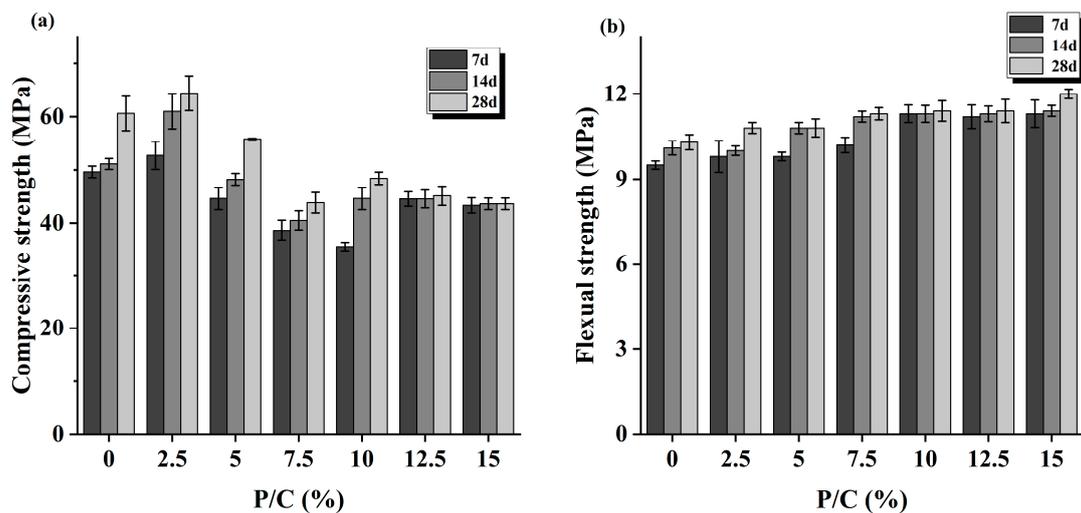
### 2.2.3. Bonding Strength

For concrete surfaces, bonding strength can generally be defined as the tensile strength perpendicular to the interface between the two layers in concrete [43]. The bonding strength between cementitious repair materials and the substrate can be evaluated by various test methods, and the available test methods can be categorized as follows (Figure 3):

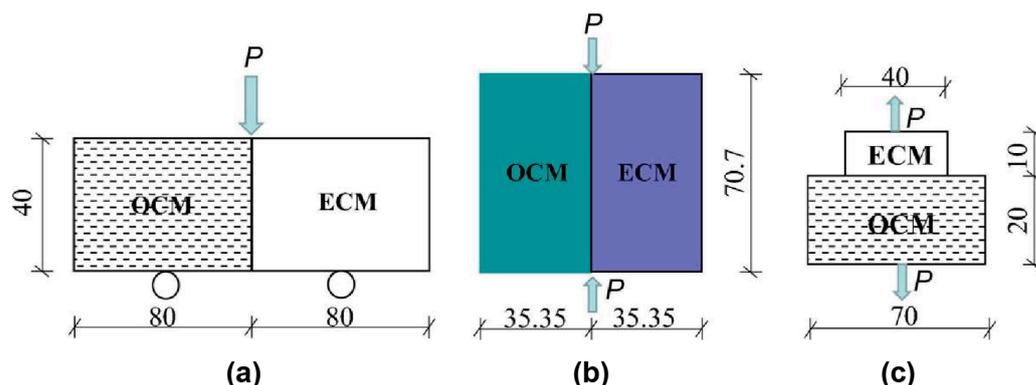
- (1) Tensile bonding. It mainly includes positive tensile bonding [44] and split bonding [45].
- (2) Direct shear bonding. It mainly includes L-type, single-sided shear [46], and double-sided shear.
- (3) Bending and pulling bonding.
- (4) Compression diagonal shear bonding.



**Figure 1.** (a1) Influence of EVA on the compressive strength of CSA cement mortar; (a2) Influence of EVA on the flexural strength of CSA cement mortar; (b1) Influence of Methylcellulose on the compressive strength of CSA cement mortar; (b2) Influence of Methylcellulose on the flexural strength of CSA cement mortar [41].



**Figure 2.** (a) Compressive strength of mortar at different P/C ratios; (b) flexural strength of mortar at different P/C ratios.



**Figure 3.** Geometry and dimensions of the specimens for different bonding strength tests (unit, mm), (a) Flexural bonding strength test; (b) direct interface shear strength test; (c) interface tensile strength test. OCM—ordinary cement mortar (substrate); ECM—epoxy resin-modified OPC mortar [9].

With the exception of the sample preparation technique, the flexural bond strength test and the flexural strength test are comparable. They not only establish the flexural bond strength, but also show the overall damage pattern at the repair interface through different fracture locations and section conditions [47]. Testing the shear bond strength of a fixed specimen entails applying a weight perpendicular to the interface. It can ascertain the repair material's shear resistance [48]. The tensile strength can be directly determined using the pull-out bond strength test. In this test, a square steel block is effectively bonded to the sample surface using an epoxy adhesive, and then a vertical tensile force is applied until the interface breaks. The bending resistance and monolithic damage pattern of the repaired interface are determined by the bending bond strength test; the shear bond strength test, tilt shear test, and push-out test are used to determine the shear resistance, while the pull-out bond strength test, split-pull bond strength test, and core-pull-release bond strength test are designed to determine the tensile strength.

Given variations in the modulus and coefficient of thermal expansion, the aggregate and mortar may expand and contract to various degrees inside the high-porosity [49–51] interfacial transition zone (ITZ) that forms between the old concrete and the repair material. Under the effect of water infiltration, microcracks within the ITZ expand [52], and the interfacial stress concentration leads to interfacial delamination. In order to ensure the integrity and operation of the restored structure, effective bonding is therefore necessary to withstand the aforementioned forces [53].

He et al. [54] discovered a linear relationship between the mechanical strength and interfacial roughness of both new and old concrete. This implies that increasing the interfacial roughness can significantly improve the mechanical interlocking effect between the two types of concrete, with interfacial roughness being the primary mechanism for bonding the two types of concrete. Nevertheless, there are still pores in the concrete between the old and the new layers, and microcracks will continue to form. The organic–inorganic composite matrix [55] and the polymer particles formed by cross-linking will force the denser hydration products or bonding paste to penetrate the small pores or interstices of the old concrete, causing the pores in the area to be filled and reducing the porosity [56]. Polymer particles can be adsorbed on top of cement particles and gravel particles at the same time and as cement hydration proceeds. A denser transition zone between the old and the new concrete is anticipated to be created by the hydrophilic polymers' ability to adsorb moisture or water and adhere to the aggregate's surface. Simultaneously, the dehydration-induced polymer film can act as a bridge between the aggregate and the rigid mortar, efficiently dissipating the fracture energy during the bending and tensile deformation processes. This allows the energy to be transferred and stresses within the concrete matrix to be evenly distributed, which rapidly alleviates the pore structure expansion and contraction caused by the external force [57]; otherwise, the cross-linking effect based on the interfacial adherence to the interpenetrating mesh structure within the matrix can be achieved through

crack deflection and the bridging effect to delay the emergence and expansion of cracks [58,59], also thanks to the strong adhesion to the surface of the old concrete substrate.

Pang [60] analyzed the bond strength of two types of waterborne epoxy (self-emulsifying emulsion NEP, external emulsion EP)–cementitious composites (NEPC and EPC) at the age of 7 days and 28 days by testing the positive tensile bond strength and the compressive slant shear strength. The findings demonstrated that there was no discernible improvement in the compressive diagonal shear strength, but the positive tensile bond strength of both kinds of modified mortars tended to rise with an increase in the P/C ratio and a prolongation of the curing time. When compared to the unmodified mortar, the 28-day bond strength in the case of NEPC was the highest at the 80% epoxy emulsion dosage, reaching a tensile bond strength of 2.6 MPa and a compressive shear strength of 8.1 MPa, corresponding to increases of 36.8% and 5.2%.

Kim et al. [61] examined the effects on the adhesive properties of cement mortar, which were defined by measuring the positive tensile bond strength, of adding four different polymers to the mortar at the same water/cement and polymer/cement ratios. The polymers used were an acrylic emulsion (AC), polyvinyl alcohol (PVA), styrene–butadiene rubber latex (SBR), and a redispersible latex powder (EVA). It was demonstrated that adding polymers to mortar enhances its adhesive qualities and bond strength as the curing period is extended. For instance, at the age of 28 days, the bond strengths of the four modified mortars, in the order of AC-modified mortar, PVA-modified mortar, SBR-modified mortar and EVA-modified mortar, were 3.41 MPa, 3.68 MPa, 3.71 MPa and 3.42 MPa, respectively, showing a noticeable increase (18.5%~27.9%) compared with the bond strength of ordinary cement mortar (2.88 MPa). It is clear that SBR induced the highest increase.

#### 2.2.4. Durability

Aggarwal et al. [62] created a polymer system based on an epoxy emulsion and compared the durability characteristics of acrylic-modified mortar with those of the new cement mortar modified by the epoxy emulsion. The epoxy emulsion-based cement demonstrated superior resistance to carbonation. For instance, the carbonation depth of the epoxy emulsion-based mortar was 45% lower than that of the ordinary cement mortar at a poly/cement ratio of 10%, while that of the acrylic-based mortar was reduced by 28%. When the epoxy emulsion was incorporated at 20%, the carbonation depth was greatly reduced (by about 75%). Similarly, the resistance to chloride penetration improved with an increasing polymer dosage. At 10% epoxy or 20% acrylic doping, the depth of chloride penetration resistance of the composite was reduced by approximately 40%.

Ying et al. [63] compared the effects of ordinary epoxy resin emulsion and graphene-modified epoxy resin emulsion on the durability of cement mortar. The results showed that after 175 freeze–thaw cycles, the mass loss of ordinary mortar was as high as 5.13%, which was much larger than those of EP–mortar (0.71%) and modified EP–mortar (0.64%). The mass losses of modified EP–mortars and EP–mortars after 300 freeze–thaw cycles were 1.38% and 1.30%, respectively, and were still significantly lower than the 5.13% mass loss of regular mortar. The relative dynamic modulus of regular mortar underwent a substantial fall after 175 freeze–thaw cycles, while the two ordinary mortars modified with epoxy showed a minimal decrease, leading to 99.4% (EP–mortar) and 99.6% (modified EP–mortar) of the relative dynamic modulus after the same number of cycles. When comparing the three mortars' respective chloride concentrations on the exposed surface at different depths, regular mortar showed higher concentrations than the EP–mortar and the modified EP–mortar throughout nearly the whole depth range from 0 mm to 30 mm. The results suggest that the epoxy resin or graphene-reinforced epoxy resin is useful in preventing environmental chloride from penetrating mortar.

#### 2.2.5. Modification Mechanism of Waterborne Polymers

Above all, the addition of water-based polymers has significantly enhanced the bond strength, flexural strength, and durability (especially carbonation, chloride penetration,

and frost resistance) of cementitious materials, while negatively affecting their compressive strength. Some scholars explained the above phenomenon as reported below.

### Cement Hydration Delay

Based on research by Taylor [64], the typical calorimetric curve of ordinary silicate cement is shown in Figure 4. It is usually divided into five phases, i.e., the initial phase, the induction phase, the acceleration phase, the deceleration phase and the prolonged slow reaction phase. When polymers are added to a cement mortar system, they have the potential to partially cover the surface of some unhydrated cement particles and slow down the hydration of the cement in two different ways: one way is by delaying the appearance of the main hydration peak, and the other way is by delaying the start of the accelerated (or induced) phase [65]. In addition, polymer latexes are dispersions in water of polymer particles with sizes of 0.05–5.00  $\mu\text{m}$ . During the ion solubilization and dynamic equilibrium phases, the polymer particles with reactive groups (e.g., hydroxyl groups, carboxylates) and negative charges can induce the adsorption of calcium and magnesium ions present in the aqueous solution and the encapsulation of cement particles [66]. This adsorption prolongs the time for the ionic concentration around the cement particles to reach supersaturation, leading to a decrease in the pH of the pore solution and a prolongation of the induction period [67–70], a process that facilitates the exothermic dissolution of the cement. Encapsulation inhibits the cement's contact with water and lengthens its hydration time, while also increasing the spatial site resistance between the cement particles, which prevents the formation of hydration products and the cross-linking of the cement. Kong et al. [65] noted that there may be a considerable correlation between the system's carboxyl group concentration and the slowing effect on cement hydration. They discovered that the main hydration peak in the accelerated period continuously moved to the right, that the height of the hydration peak decreased as the polymer dosage increased (Figure 5) and that the polymer latex (L1) made from a copolymer of styrene and acrylic acid (MAA) promoted a longer induction period. L1 contains more R-COO<sup>-</sup> groups, as determined by potentiometric titration. In other words, the electrostatic interaction between anions with a high charge density (R-COO<sup>-</sup> groups) and metal cations on the mineral surface is stronger, and the delaying effect is more pronounced (Figure 6). It was suggested that the polymer latex would also achieve the retardation effect by reducing the hydration rate in the accelerated period and the total heat generation at a certain age [71,72], besides prolonging the induction period. This is consistent with the findings of Kong et al. In contrast, in most cases, delays in setting and hydration lead to a reduction in the early strength of the cementitious matrix [73,74].

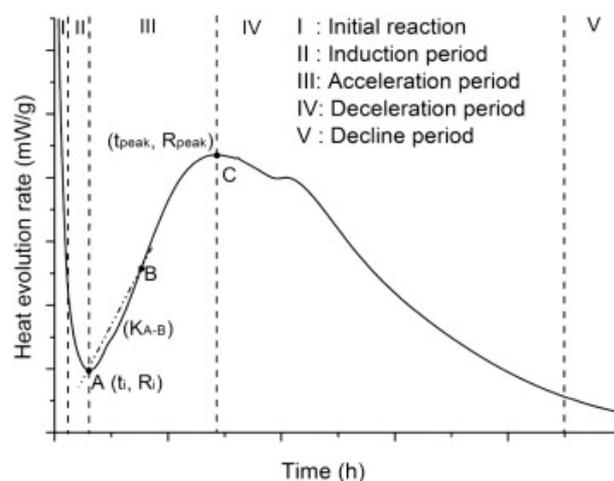
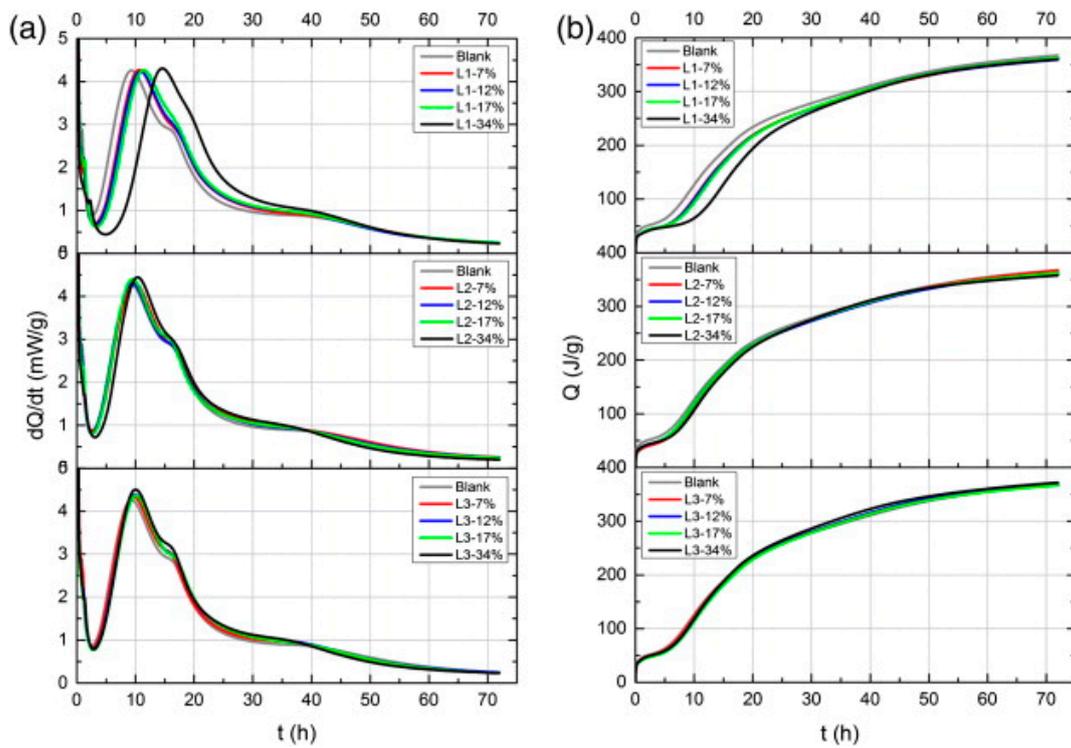
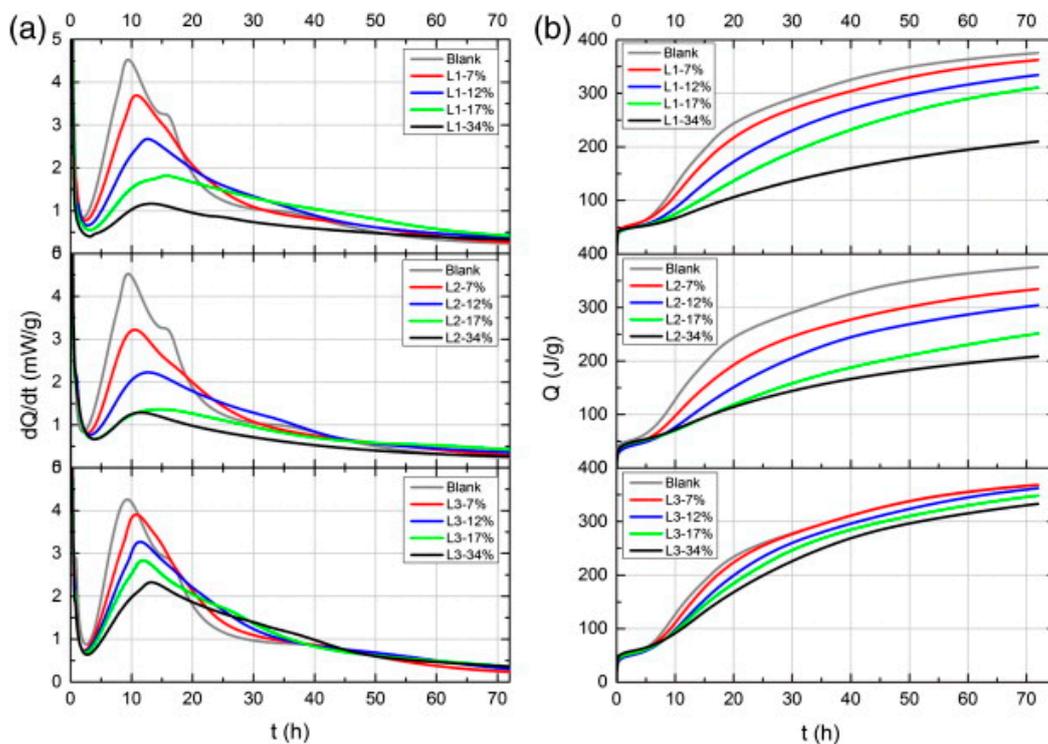


Figure 4. A typical heat evolution curve of cement hydration [65].



**Figure 5.** Calorimetry curves of cement pastes in the presence of various latexes. (a) Differential heat flow; (b) cumulative heat flow [65].



**Figure 6.** Calorimetry curves of cement pastes in the presence of the serums from various latexes. (a) Differential heat flow; (b) cumulative heat flow [65].

In summary, the polymer inhibits the contact between cement and moisture mainly through the complementation of polymer particles with  $\text{Ca}^{2+}$ , hindering the formation and cross-linking of hydration products (especially, hydrated calcium silicate), prolonging the

hydration time and lowering the early strength of modified mortar. This phenomenon is more obvious in polymer–cement mortar composites containing reactive groups.

#### Air Entrainment

It is typical that the polymer emulsions used in the preparation of mortars usually entrain a considerable amount of air [75–77]. Air is trapped inside the cement paste as whirlpools that form while mixing [78]. For these gasses, the hydrophobic groups carried by the polymer will help to generate and stabilize bubbles. The reciprocal repulsion of the groups causes these bubbles to gradually disperse throughout the cement mix. Air replaces a portion of the slurry's particles, creating closed, spherical pores with pore sizes between 10 and 300  $\mu\text{m}$ . These pores can make the modified mortar less dense and more porous, forming a hollow and collapsed structure inside the mortar that is unable to withstand larger loads and manifests itself as a reduction in compressive strength. These pores can reduce the sliding resistance of the cement particles as well as the transport of aggressive ions and gases and significantly improve the fluidity and frost resistance of the mortar [79,80]. As concluded by other researchers [81,82], the compression-related weaknesses of polymer-modified cements can be compensated to a large extent by their improved flowability.

In conclusion, on the one hand, the air-entraining effect of polymers significantly improves the compactness and fluidity of mortar due to the formation of closed pores, and on the other hand, the porosity increases, and the mechanical properties decrease.

#### Curing Condition

Shi et al. [83] investigated the effects of the styrene–acrylic emulsion (SAE) and styrene–butadiene latex (SBR) on the mechanical properties of ternary binders composed of silicate cement, alumina cement and hemihydrate gypsum under various humidity curing conditions (water curing condition: 100% humidity,  $22 \pm 2$  °C; natural curing condition:  $50 \pm 10\%$  humidity,  $22 \pm 2$  °C; and standard curing condition:  $90 \pm 5\%$  humidity,  $22 \pm 2$  °C). The results showed that the flexural and compressive strengths of the two polymer latex-modified mortars gradually increased with the extension of the curing time, regardless of the humidity conditions, but both were lower than those of the unmodified cement mortar. It is noteworthy that both types of composites' flexural strength increased significantly with the increasing polymer dosage at 28 days and under water-curing conditions (100% humidity and  $22 \pm 2$  °C). The maximum flexural strength of the was reached at P/C = 16% (SBR) and 12% (SAE), which represented increases of 11% and 19%, respectively, over the unmodified mortar. Chen et al. [84] examined the effects of SBR and SAE on silicate cement–aluminate cement–gypsum (OPC–CA–gypsum) at different curing temperatures (40 °C, 60 °C). The findings indicated that while both SBR and SAE were useful for raising the flexural strength and toughness of OPC–CA–gypsum mortar, SAE performed the best. So, it is implied that the curing temperature affects the morphology of the polymer film and hydration products and the extent of their formation, which consequently affects the performance of polymer-modified OPC–CA gypsum. This was verified in the study of Li et al. [85]. The conclusion was reached that high temperatures promote the creation of compounds containing carboxylates as well as pore refinement, a decrease in the thickness of the polymer film, and an increase in the number of pores in the polymer film, all of which alter the mortar mechanical properties. In the above study [68], Wang et al. found that the addition of polymer can greatly improve the flexural properties of mortar under wet–solid mixed curing conditions. Some scholars proposed the following two ways of wet–solid mixed maintenance. One technique involves initially setting the mortar molding at  $20 \pm 3$  °C with a relative humidity of 90% or higher in the fog room for 7 d (wet maintenance) and then setting it at ambient temperature for 21 d (dry maintenance); the other involves placing the mortar molding in water at  $20 \pm 3$  °C for 5 d and then at  $20 \pm 3$  °C with relative humidity of 60% with respect to that of the maintenance conditions for 21 d. The first setting involves operating in  $20 \pm 3$  °C water with 80% or higher hu-

midity with respect to that of wet maintenance for 2 d. Combined with the results of a previous study [37] and others in the literature [86], it can be seen that water curing is more conducive to the hydration of ordinary cement mortar compared with wet curing, and the strength of the cement grows; however, this is not observed for polymer-modified mortar. Dry curing at a later stage is conducive to polymer (latex, powder) agglomeration on the hydrated cement [87–89], resulting in the creation of a polymer film or a network structure. This process produces higher strengths more quickly than those observed in regular cement mortars.

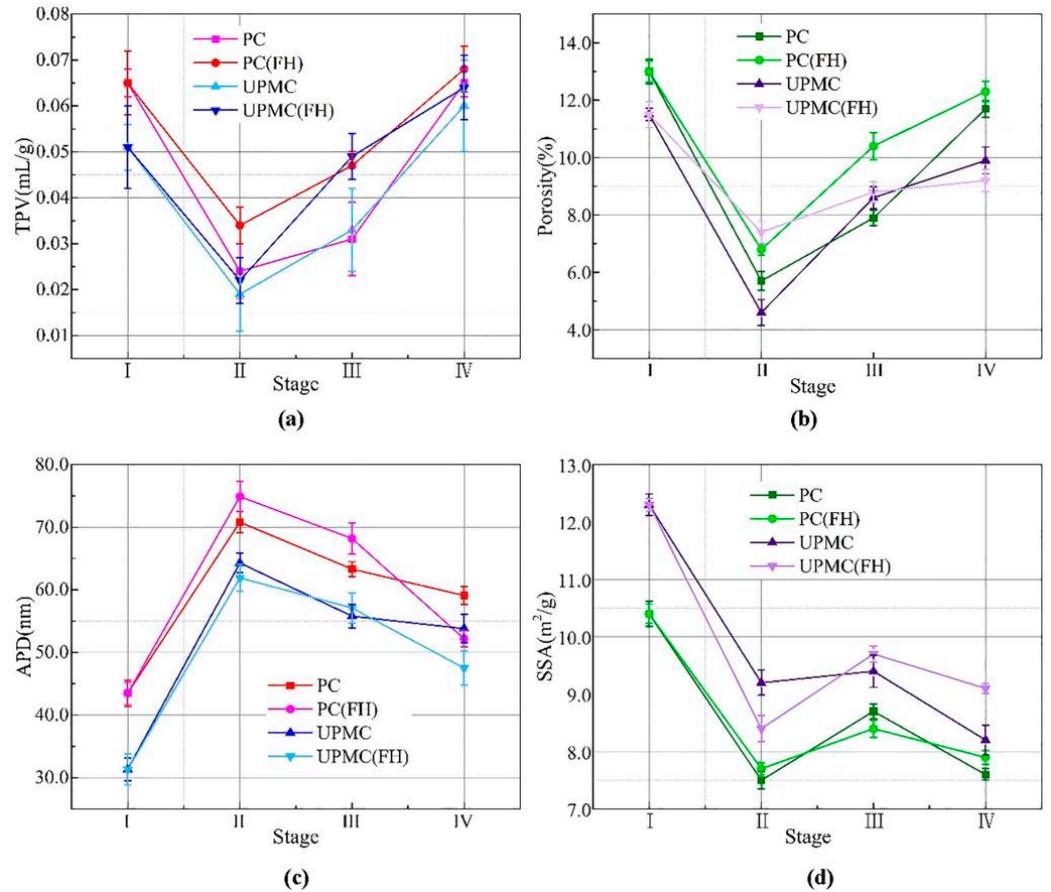
Furthermore, Beeldens et al. [90] and Li Bei et al. [91] concluded that the curing temperature should be higher than the minimum polymer film-forming temperature, or the tightly packed polymer particles could not form a continuous film or reticular structure [92].

In conclusion, conditioning temperature and conditioning humidity have the biggest effects on polymer modification under conditioning circumstances. The polymer's ability to form a film and fully utilize its high flexibility and elasticity is determined by the maintenance temperature. The rate at which mortar loses water is influenced by the humidity; the faster the mortar dries out, the more favorable conditions exist for the polymer to form a film and a three-dimensional mesh structure.

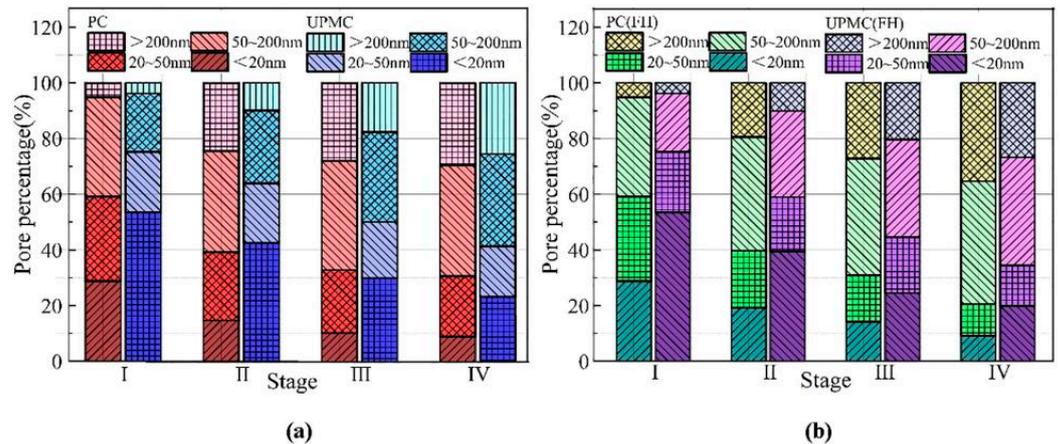
### Pore Structure

When concrete is solidified, the 'wall effect' and 'microleakage effect' within the matrix induce a water film to form around the aggregate; at this time, the water/cement ratio near the aggregate is higher than in the mortar, and the pore space in the interface area increases [93]. When a polymer emulsion (powder) is mixed as an additive (such as a plasticizer) [94], the concrete's internal stress tends to balance (similar to drying shrinkage), and the process creates a significant number of bubbles and pores. The polymer intermolecular tensions will rise once again. A 10% increase in pore volume will result in a 50% decrease in compressive strength. On the one hand, the introduced gas will form pores in the mortar after it hardens, affecting the internal compactness. On the other hand, the increase in pores will cause a decrease in the compressive strength of the concrete, especially under fatigue loading [95].

Zhang et al. [96] studied and compared the changes in the pore structure parameters (characterized by TPV, porosity, SSA, and APD) of plain concrete (PC) and unsaturated polyester resin-modified concrete (UPMC) under single loading as well as under the interaction of hygrothermal effect and fatigue loading (FH) and classified the effect of the force into four stages (i.e., I, II, III and IV), as shown in Figures 7 and 8. In the initial stage, the compound effect of early compression and splitting of the pores causes an increase in the density of concrete. Under the influence of loading and of the interaction of FH with the hygrothermal effect, the pores are stimulated to develop, small pores compress, and large pores expand, and the proportion of harmful pores increases [97]. For example, when a single loading is applied to ordinary concrete, the volume fraction of the more hazardous pores increases by 371.48% (stage II), 439.31% (stage III), and 469.17% (stage IV). The ultimate expansion of the large pores affects the small pores with the continuous addition of unsaturated polyester resin and the extension of the phase of action of the applied force, as shown by an increase in the APD values of UPMC materials, under the interaction of FH with the hygrothermal effect, by 92.12% (stage II), 80.23% (stage III), and 51.75% (stage IV) relative to the initial values. Additionally, at every stage of external activity, the SSA value of UPMC is lower than that of regular concrete, and the pore structure is continuously homogenized and polished. Concurrently, the film resulting from the desiccation of the unsaturated polyester resin is affixed to the inflexible cement mortar. This mitigates the extent of pore structure expansion [57] and contraction caused by external forces by evenly distributing the stress within the concrete matrix. Furthermore, in comparison to that of regular concrete, the porosity reduction rate of UPMC is 11.54% (stage I), 19.30% (stage II), 6.21% (stage III), and 15.38% (stage IV).



**Figure 7.** Variation in the pore parameters of different concretes subjected to different action types at various stages. (a) TPV; (b) porosity; (c) APD; (d) SSA, vs. action stages [96].



**Figure 8.** Variation in the pore diameter distribution of different concretes subjected to different action types at various stages, (a) Single-load action; (b) interaction of hygrothermal effect and fatigue loading [96].

### Polymers

Most of the polymer emulsions used in the field of modified cements can be classified into two groups due to their differences in the polymer chains. One type of polymer has no reactive groups in their polymer chains, while the other type contains them. For the purpose of modifying cement mortars, Wang et al. [98] examined and compared the modification methods of two typical polymers (XSBRI, a carboxy styrene-butadiene latex, and SBR, a styrene-butadiene latex). As shown in Figure 9, there is a noticeable difference between

the infrared spectrum plots of the two polymers. It is clear that the tensile vibration peaks of XSBRI at  $1700\text{ cm}^{-1}$  and  $1150\text{ cm}^{-1}$  corresponding to C=O and C-O in the carboxylate group (-COOH) [99], respectively, are not obvious in the SBR plots. The strength of the SBR-modified mortar was consistently higher than that of XSBRI, despite the fact that the compressive strength of the composites decreased in the presence of the polymer. Furthermore, SBR did not significantly increase the flexural strength as much as XSBRI. There might be a solution to this from a micro-perspective. The authors found that the  $\Delta H$  of the SBR-modified cement mortar did not change significantly with the increase in polymer amount, as shown in the DSC plots (Figure 10), which implied that the amount of CH crystals in the composite was almost unchanged, as evidenced by the infrared spectroscopy (FITR) plots (Figure 11a) in the presence of 12.5% SBR doping after 28 days. The characteristic peaks of the SBR latex polymer chains are still present, despite a change in shape, and this result also tentatively indicates that no chemical reaction occurred in the SBR-modified mortar, but there was a physical reaction. The  $\Delta H$  values of the XSBRI-modified cement mortar decreased from the initial  $116.57\text{ J/g}^{-1}$  to  $75.34\text{ J/g}^{-1}$ , showing a decrease in CH crystals. The vibrational peak of C=O at  $1700\text{ cm}^{-1}$  nearly vanished in the related FITR plot (Figure 11b), and the distinctive carboxylate (-COO-) peaks emerged at  $1580\text{ cm}^{-1}$  and  $1410\text{ cm}^{-1}$  with 12.5% polymer doping [100–102]. It can be guessed that this was due to the fact that the  $\text{H}^+$  ion on the carboxylate group in XSBRI was replaced by metal ions in the pore solution to form -COO- [103–105], and the CH crystals in the system were constantly consumed. In other words, the carboxyl group (-COOH) in the XSBRI-modified mortar was involved in the chemical reaction. Furthermore, SEM analysis revealed that the hydrated crystals were covered by both SBR (Figure 12) and XSBRI (Figure 13) latex films, which simultaneously filled the pores and fissures of the modified cement system. It is interesting to observe that the formation of fibrous membranes and interconnecting hydrated crystals in the presence of XSBRI, as shown in Figures 12 and 13—that is, the formation of a network structure in the XSBRI-modified mortar—did not occur in the presence of SBR.

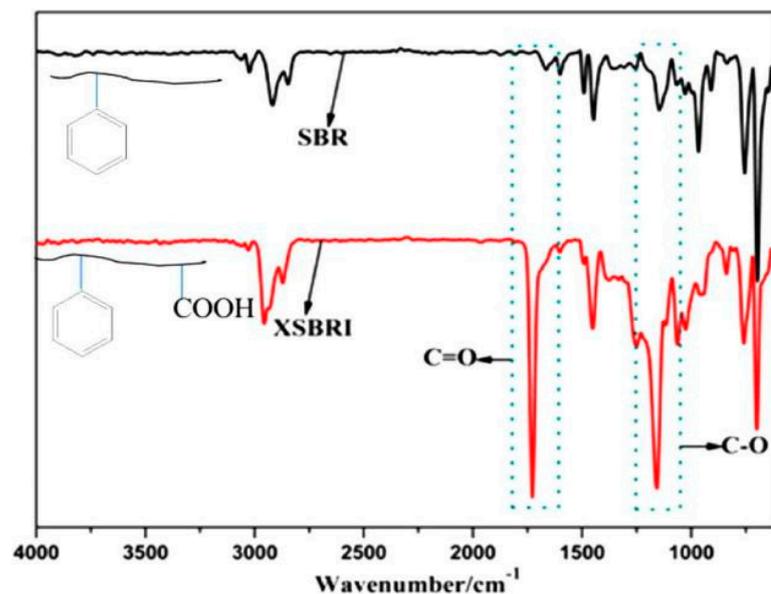
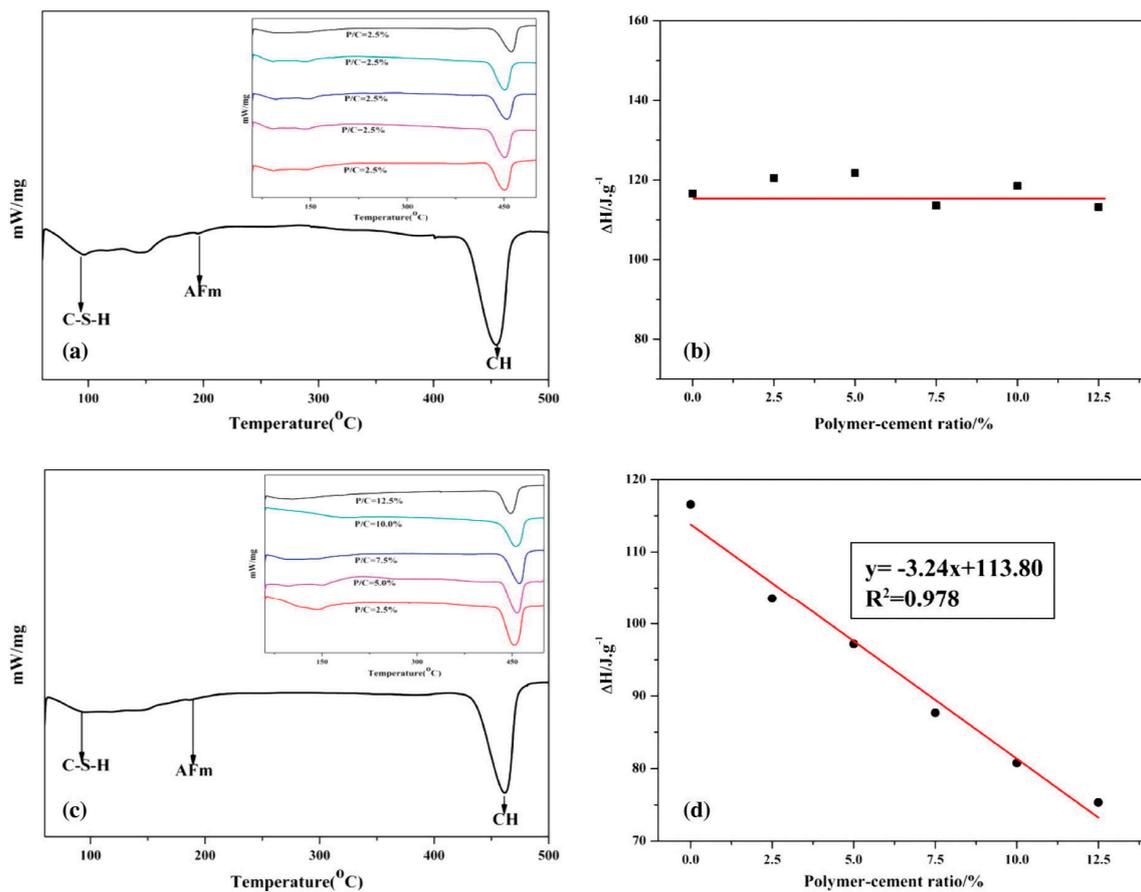
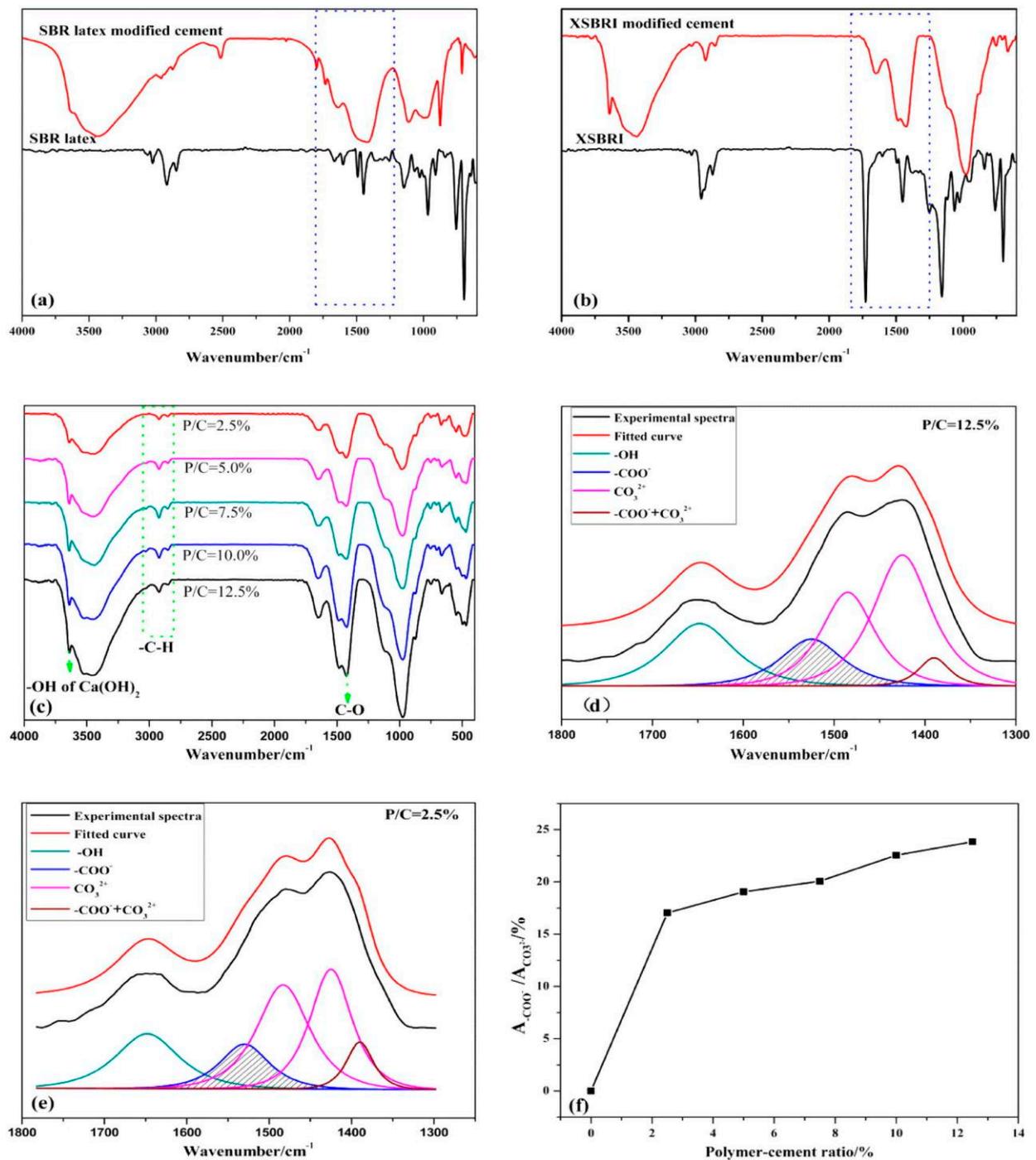


Figure 9. The ATR-IR spectra of SBR and XSBRI latexes [98].

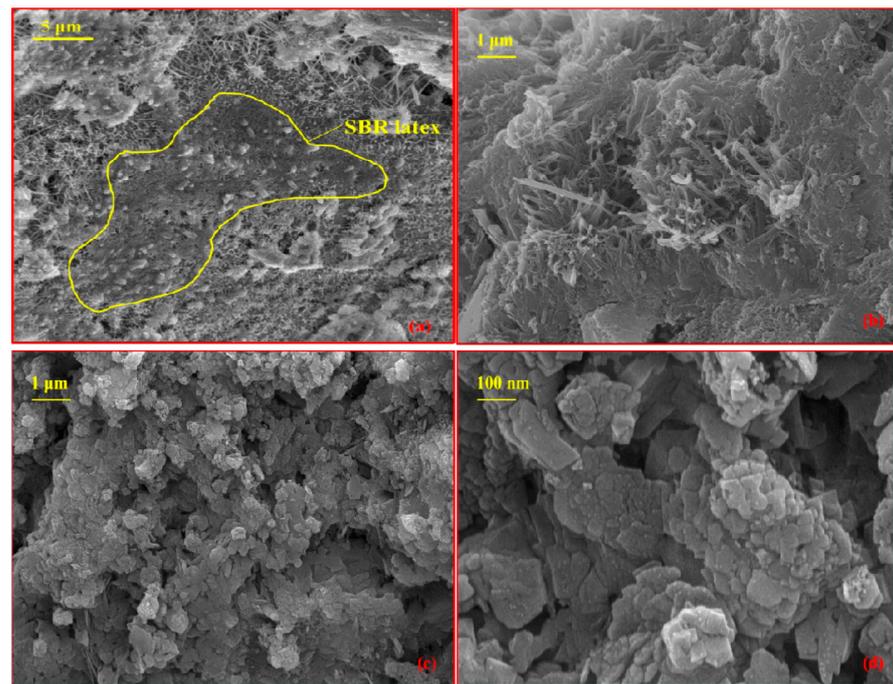


**Figure 10.** (a) DSC plot of the SBR latex-modified cement at 28 days; (b) estimated variation of the enthalpy change due to  $\text{Ca}(\text{OH})_2$  decomposition in the presence of the SBR latex; (c) DSC plot of the XSBRI-modified cement at 28 days; (d) estimated variation of the enthalpy change due to  $\text{Ca}(\text{OH})_2$  decomposition in the presence of XSBRI [98].

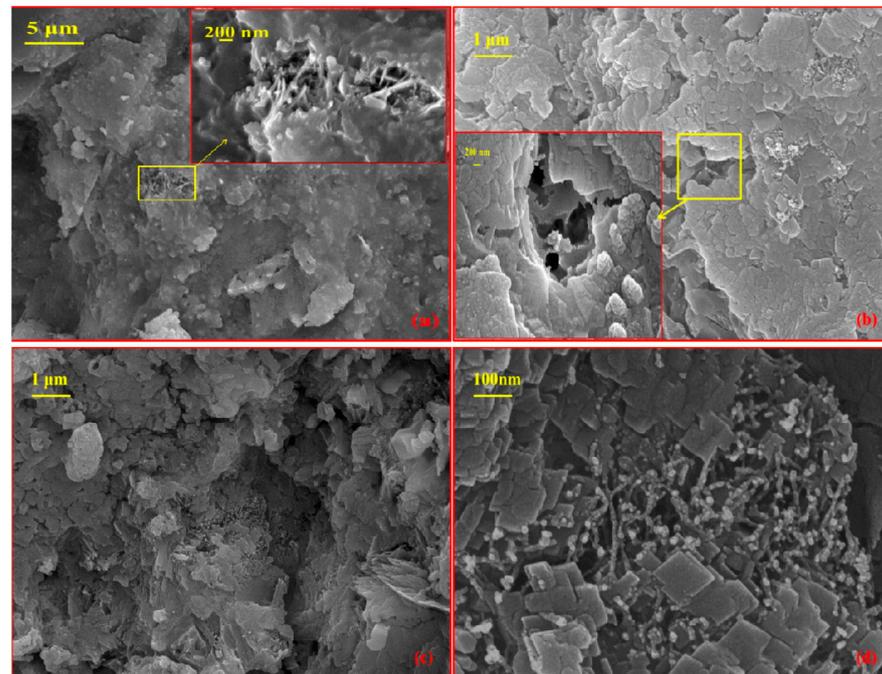
Given the above, for polymer latexes (powders) that do not contain reactive groups, the reaction process with cement mortar consists of physical reactions only, which means that the polymers are connected from flocs to a film in the cement slurry along with the process of hydration of the cement. The coherent polymer film will create an internal barrier to free water movement, filling the cracks and pores of the cement (filling effect). This will enhance internal curing (i.e., reduce porosity) and reduce the expansion of microcracks within the matrix [72], as demonstrated by the studies mentioned above, which also showed decreased water absorption and increased toughness, impermeability, and frost resistance. This mechanism is called physical modification mechanism (Figure 14). Additionally, the reactive groups will react with the calcium ions ( $\text{Ca}^{2+}$ ) released during cement hydration to form new compounds. This process will increase the cement material's degree of hydration, inhibit or shrink the nucleation of crystallization, as well as decrease the crystal orientation of calcium hydroxide (C-H) within the ITZ and promote the formation of polymer-cement hydration products and the composite structure of dense calcium-silicate-hydrate (C-S-H) gels [106,107]. This phenomenon increases the amount of hydration products and makes up for the strength loss brought on by the excessive porosity caused by air entrainment. The flexural strength of mortar treated with XSBRI latex was demonstrated to be greater than that of mortar treated with SBR latex in a study conducted by Wang et al. [98]. Chemical modification mechanism is the name given to this mechanism (Figure 15).



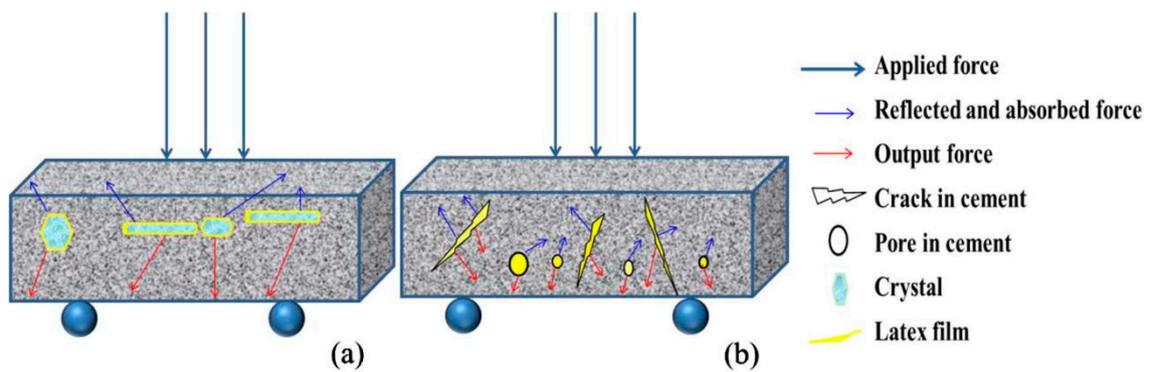
**Figure 11.** (a) Comparison of the FTIR spectra of SBR latex and SBR latex-modified cement; (b) comparison of the FTIR spectra of XSBRI and XSBRI-modified cement; (c) FTIR spectra of XSBRI-modified cement at 28 days; (d) FTIR spectrum fitted result of XSBRI-modified cement at 1800–1250 cm<sup>-1</sup> with a polymer/cement ratio of 12.5%; (e) FTIR spectrum fitted result of XSBRI-modified cement at 1800–1250 cm<sup>-1</sup> with a polymer/cement ratio of 2.5%; (f) variation of  $A_{\text{-COO}^-} / A_{\text{CO}_3^{2-}}$  with the polymer/cement ratio [98].



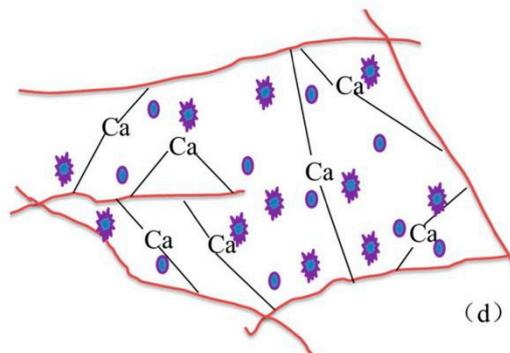
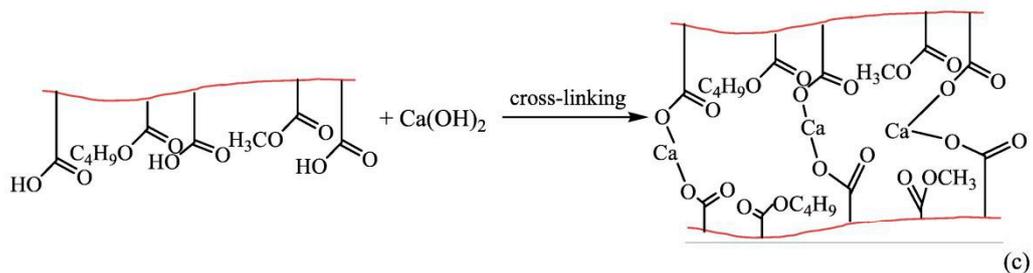
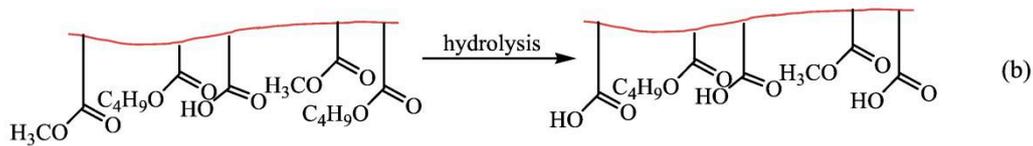
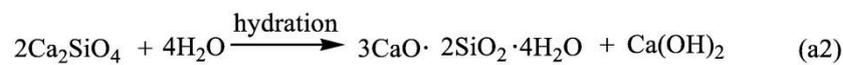
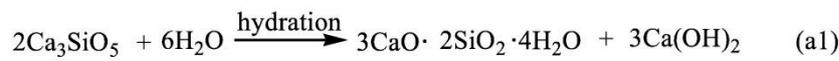
**Figure 12.** The microstructure of SBR latex modified cement at 28 days, (a) the SBR latex film covered the surface of needle-like, rod-like and bar-like crystals; (b) SBR latex film filled into pores and cracks of cement; (c,d) the polymer film and particles covered the surface of bar-like crystals and filled in the pores and cracks in SBR latex modified cement system [98].



**Figure 13.** The microstructure of XSBRI-modified cement at 28 days, (a) the XSBRI covered the surface of hydration crystals; (b) XSBRI latex film filled into pores and cracks of cement; (c,d) the XSBRI fiber-like film and hydration crystals connected with each other and latex particles filled in the pores of cement [98].



**Figure 14.** The physical model of polymer latex-modified cement. (a) The polymer latex covers the hydration crystals; (b) the polymer particles and film fill in the cracks and pores [98].



— The backbone of PA latex    \* The product of cement hydration    ● The unhydrated cement particles

**Figure 15.** (a1,a2) The hydration reactions of cement. (b) The hydrolysis of polyacrylate latex in the modified system; (c) the crosslinking reaction between polyacrylate latex and cement; (d) the cross-linked network structure which was obtained by chemical reactions in the polyacrylate latex-modified cement system [67].

Furthermore, the early stage of polymer inclusion shows a tiny rise in compressive strength, but a decrease in later stages. It may be due to the filling effect of the polymer that slightly increases the compressive strength. However, when doping increases, the polymer

film—which previously exhibited great elasticity and flexibility—becomes thicker [108], worsening the mechanical properties [109–111], increasing rigidity, and lessening the sensitivity to external stress; conversely, the composite’s water transport is impeded by the continuous polymer sheet. As a result, less water is needed for the cement to hydrate, which lowers the rate of hydration and leads to fewer hydration products. The compressive strength consequently drops. This explains why flexural strength decreases with the increasing polymer dosage.

### 3. Application of Waterborne Polymer–Cement-Based Composite Repair Materials in the Western Saline Environment

According to the global map of saline soils released by the Food and Agriculture Organization of the United Nations on 20 October 2021, there are currently more than 833 million hectares of saline soils globally, accounting for about 8.7% of the Earth’s total area [112]. In China, the area of saline soil is about  $3.69 \times 10^7$  hm<sup>2</sup>, and the area of saline soil in northwest China is about 69.03% of the national area [113,114]. A broad word encompassing saline, alkaline and different types of salinized and alkalinized soils that contain high levels of calcium carbonate, sodium sulfate and calcium chloride is ‘saline soil’. Long-term exposure to a saline environment causes a large number of corrosive ions, such as  $\text{SO}_4^{2-}$ ,  $\text{Cl}^-$ ,  $\text{Mg}^{2+}$  and  $\text{CO}_3^{2-}$ , to diffuse, crystallize, dissolve, recrystallize, expand and deform through the initial cracks and pores in concrete structures. This eventually causes cross-cracks to form on the inside and outside of a structure, which ultimately lead to structure instability or even collapse [115]. In addition, compared with mild-climate areas, Northwest China has a large temperature difference between day and night, lower humidity, dryness [116] and other environmental characteristics that cause the internal evaporation and migration of water in concrete [117]. Thus, concrete’s mechanical qualities [118] will be diminished by increased capillary stress and uneven temperature stress, which will make concrete more prone to cracking.

As mentioned previously, polymers can effectively improve the compactness of mortar, increasing its resistance to erosion by external corrosive ions. Li et al. [74] prepared SBR-, SAE- and polyacrylic acid emulsion (PAE)-modified CSA cement mortars and evaluated the changes in the mechanical properties of all three materials under sulfate attack. As shown in Figure 16, when subjected to sulfate attack, the flexural strength of the cement mortar was not significantly affected, but sulfate attack had a large effect on the compressive strength. With the increase in sulfate solution concentration and polymer dosage, the compressive strength of SAE- and PAE-modified cement mortar showed a decreasing trend. In addition, the flexural strength coefficients of SAE- and PAE-modified mortars increased with the addition of 20% polymer in 5% sodium sulfate solution (Figure 17), which showed that the addition of polymer latex helped to improve the resistance to sulfate attack.

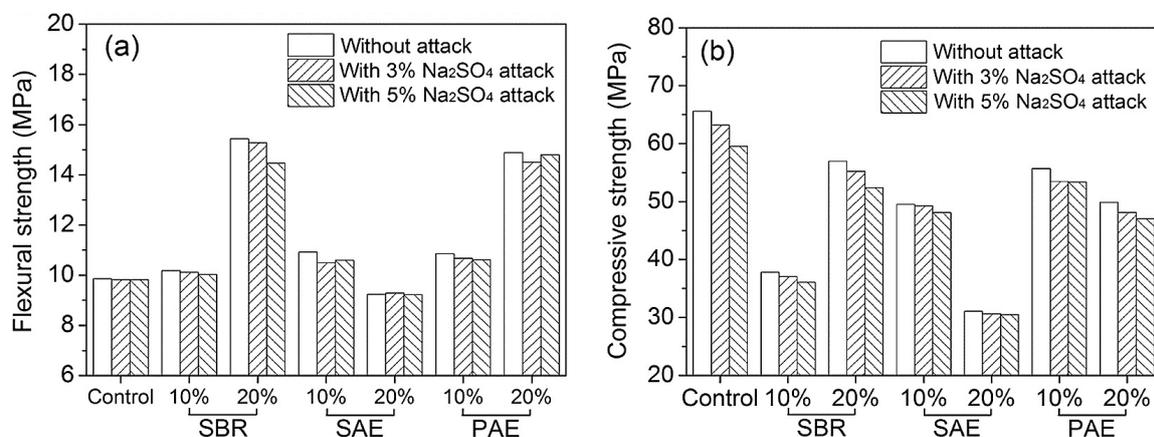


Figure 16. Strength before and after sulfate attack. (a) Flexural strength; (b) compressive strength [74].

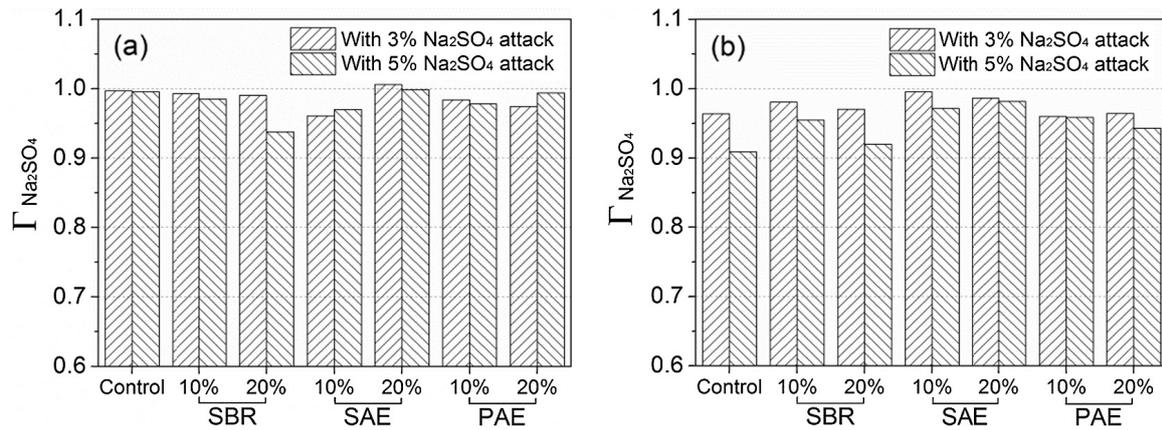


Figure 17. Coefficient of sulfate attack resistance. (a) Flexural strength; (b) compressive strength [74].

Lukowski et al. [119] created a polyacrylate–cementitious composite and compared the elongation, mass, and compressive strength of composite samples submerged in water and in a 5% mass fraction solution of magnesium sulfate. While the specimens submerged in water showed almost no significant changes, it was indicated that the elongation, mass, and compressive strength of the composites decreased continuously with an increase in the polymer dosage under sulfate attack. Additionally, the elongation showed an increase with age at the same poly/cement ratio. At 42 months of age, the elongation of the modified mortar with P/C = 20% was 44% less than that of the unmodified cement mortar, as illustrated in Figure 18. In terms of mass (Figure 19) and compressive strength (Figure 20), the composites showed an increasing and then a decreasing trend with age, and the higher the content of the polyacrylic acid emulsion, the more significant the loss of mass, so that the mortar mass reached its lowest value corresponding to 20% of the original mass, which was 38.6% lower than that of the control group (Figure 19).

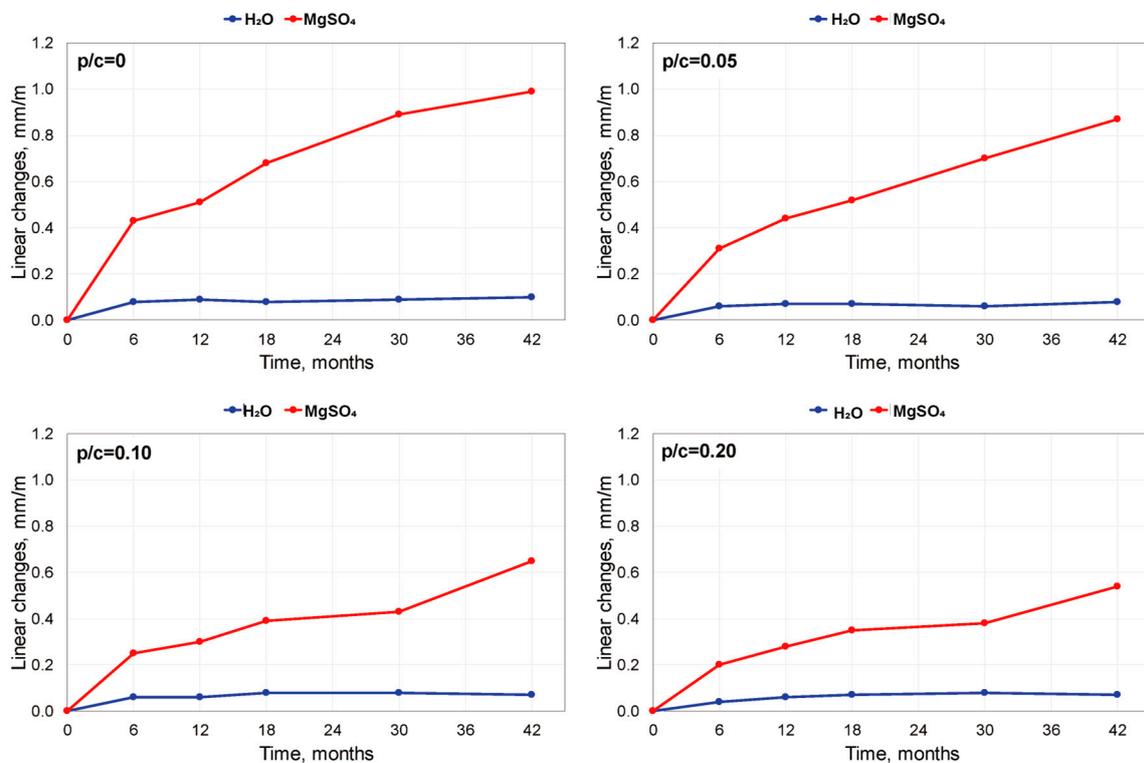
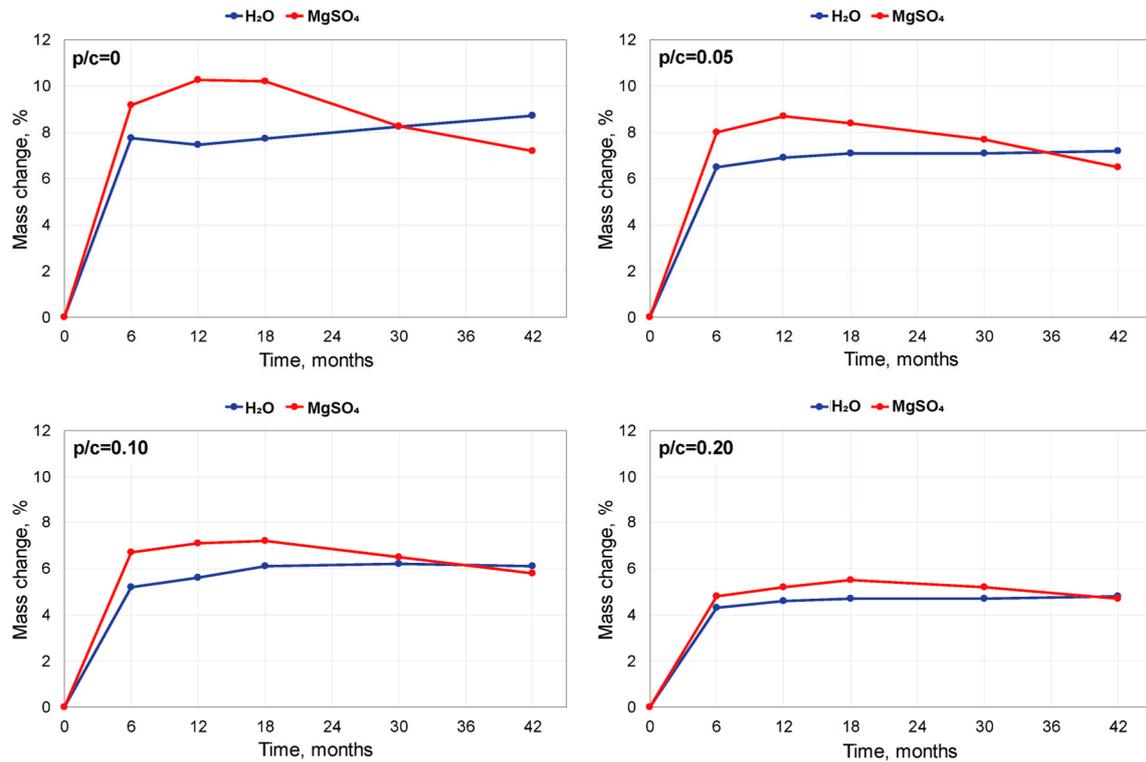
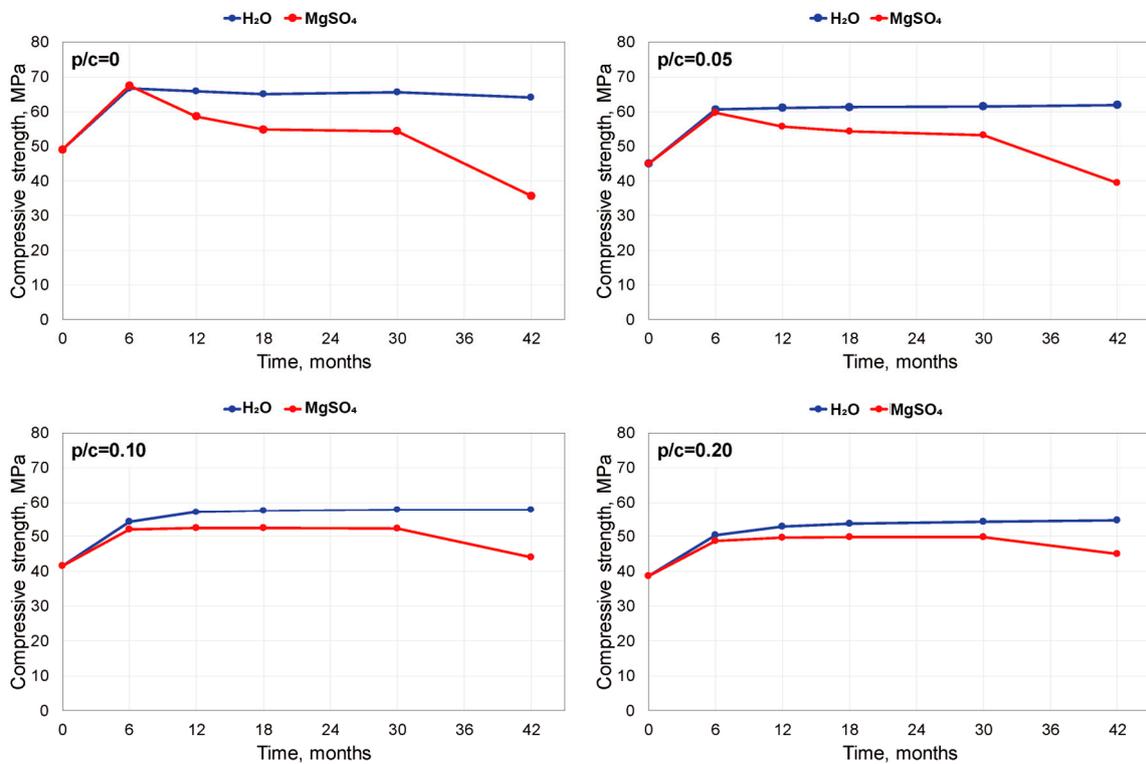


Figure 18. Elongation of mortar specimens exposed to a 5% MgSO<sub>4</sub> solution and to distilled water [119].



**Figure 19.** Mass changes of mortar specimens exposed to a 5% MgSO<sub>4</sub> solution and to distilled water [119].



**Figure 20.** Compressive strength of mortar specimens exposed to a 5% MgSO<sub>4</sub> solution and to distilled water [119].

Lu et al. [120] used sodium persulfate (initiator) and an emulsifier in dosages of 4% and 5%, respectively, to produce the styrene–acrylic emulsion (SAE), which was then

incorporated into alumina sulphate cement, and evaluated the changes in the flexural strength and sulphate erosion resistance coefficients of the modified mortars when they were placed in a sodium sulphate solution with a mass fraction of 3% and in water. The findings demonstrated that, in all environments, the flexural strength of the polypropylene emulsion–cement mortar composites with an initiator of 5% was always greater than that of the modified mortar containing 4% of the initiator. This strength developed as the polymer content increased and peaked at P/C = 7.5%, with values of 10.53 MPa and 9.92 MPa after immersion in 3% sodium sulfate and in water, respectively, exceeding that of the blank control group. It can be seen that the polypropylene emulsion improved the ability of cement mortar to resist sulfate erosion.

#### 4. Summary and Prospects

Through summarizing the effects of different polymers on the workability, mechanical properties and durability properties of cement mortar, this paper found that polymers added to cement mortar have the following three main effects:

- (1) Polymer particles are adsorbed on the surface of cement, preventing the contact between cement and water and delaying the hydration of cement, which is manifested in the prolongation of the setting time and the reduction in early strength;
- (2) In addition to improving ease of use, impermeability, etc., and the reduction of viscosity, with the hydration of cement, the aggregate and the aggregated polymer particles crosslink to form a thin film layer. On the one hand, this film can improve the flexibility, toughness, adhesion and durability of cement mortar by taking advantage of high elasticity and filling effects; on the other hand, it hinders water transport and lowers the rate of hydration, particularly in the presence of high polymer dosages, and manifests as a decrease in mechanical strength;
- (3) The polymer will have a significant impact on cement mortar depending on curing conditions (such as temperature, humidity), pore structure, polymer phase, cement hydration and other factors. The detrimental effects of these conditions should be taken into account in the subsequent application processes, and positive and effective steps should be taken to reduce them.

Many thermoplastic or other thermosetting polymers are now widely used to modify cementitious systems for repair, injection, bonding [121] and waterproofing [122] applications. They are used in various forms such as liquid resins, latexes, redispersible powders and water-soluble homopolymers or copolymers. The most important characteristic of a composite material is the designability of its properties and structure, which are found in polymer–cementitious composites. Therefore, in order to achieve an optimized design and enable the adaptation of the obtained material to various occasions, it is necessary to select appropriate polymers and design reasonable structures based on the properties of the polymers and the cement, in accordance with the requirements of the material properties at the site of use. For the purpose of providing some reference value to real projects, this study summarized and compared the performance of polymer latex (powder) used in the aforementioned research. The results, shown in Table 1, indicated that SBR showed an excellent performance in terms of both mechanical properties and durability. It is made of copolymerized butadiene and styrene, and a study [84] cited above attests to the fact that it has higher heat resistance than natural rubber. As a result, SBR can be suitable in hot, dry climates (particularly for projects that cannot be water-cured after construction). Additionally, SBR has exceptional bonding qualities even in damp environments and a certain level of crack resistance. SBR is also one of the repair materials with ideal comprehensive performance and a fairly reasonable cost. Comprehensively speaking, SBR–cement mortar composite repair materials can be applied to roads, bridges, hydropower dams and municipal engineering and other repair projects. Epoxy-modified mortar shows significantly advantageous mechanical properties, which may be due to the epoxy resin emulsion being a thermosetting polymer, the high degree of inter molecular crosslinking and the formation of a body-type mesh structure, leading to strong rigidity

and very good heat resistance. While polyurethane emulsion-modified mortar has a slower curing reaction [123,124] and is more prone to chalking, yellowing and discoloration than epoxy resin emulsions, the latter are more durable and cannot be readily destabilized in a medium-alkaline environment. Therefore, a polymer-modified cement mortar based on an epoxy emulsion should be more appropriate for use in structures exposed to high humidity conditions or immersed in water, structural repair projects requiring a high degree of strength, and repair projects involving marine engineering and water conservation with high requirements for corrosion resistance. Nevertheless, epoxy resin is easy to degrade, its chains can break under ultraviolet irradiation [62,125,126], and it has poor aging resistance; so, it is not applicable in highway repair projects. Composite materials consisting of PVA, EVA, VAE, AC, PAE, SAE and cement mortar have relatively favorable adhesion, mechanical properties, seepage resistance, corrosion resistance, etc., and can be ideal for patching projects with low performance requirements. However, the EVA emulsion is a linear polymer material with high molecular polarity that can easily become brittle and hard when heated; therefore, its aging resistance and water resistance are poor [127], and it is not suitable for highway engineering.

On top of that, a single polymer can be expensive, and there are ongoing issues like short storage times and environmental contamination brought on by variables like the synthesis method, the raw ingredients used, storage settings, etc., making it impossible to guarantee a product's performance. The performance of cement mortars is significantly impacted by polymers modified with other admixtures (fly ash, silica fume, silica), polymers and admixtures (water reducers, defoamers), polymer compounds (waterborne polyurethane emulsions, waterborne epoxy resin emulsions) and modified polymers (emulsions and water-soluble polymers can be compositely modified, and emulsions can be modified with ultrafine mineral powder admixtures, water glass composites, polymer fibers and other composite materials); otherwise, those materials can effectively minimize the production costs due to the reduction in the amount of polymer admixture, environmental pollution, and so on.

**Table 1.** Comparison of different waterborne polymers–cement mortar properties.

Polymer	Performance Parameter							Performance Comparison
	Compressive Strength	Flexural Strength	Bond Strength	Corrosion Resistance	Freeze–Thaw Resistance	Anticarbonization Performance	Impermeability	
SAE	[117]	50.1 MPa	11.3 MPa		39.9% (A)	43.4% (A)	37.5% (A)	1. Comparison of compressive and antifolding performance [117] EP > WPU > SBR > PAE > SAE > VAE [53] EP > WPU > EVA > PVA > SBR > AC > VAE 2. Bond strength comparison SBR > PVA > EVA > AC > VAE > SAE > EP > WPU 3. Corrosion resistance comparison [117] SAE > SBR > PAE (A) [128] VAE > SBR (C) 4. Comparison of freeze–thaw resistance [117,129] SBR > PVA > PAE > SAE (A) 5. Comparison of anticarbonization performance [117] PAE > SBR > SAE (A) 6. Impermeability contrast [128] SBR > VAE (C)
SBR	[117]	62.3 MPa	15 MPa	2.72 MPa	34% (A)	16.1% (A)	31.9% (A)	
	[52]	44.1 MPa	9.62 MPa					
PAE	[128]			3.71 MPa	1.04 (C)			
	[117]	61 MPa	13 MPa		17.6% (A)	36.9% (A)	16.4% (A)	
EP	[27]	90 MPa	17.8 MPa	2.6 MPa	13.6% (B)			
	[53]				16% (B)			
	[56]					1.38% (B)	1.6 (C)	
PVA	[53]	49.9 MPa	9.91 MPa	3.68 MPa				
	[129]				35.28% (A)			
					1.07% (B)			
AC	[53]	32.34 MPa	7.92 MPa	3.41 MPa				
EVA	[53]	50.27 MPa	11.09 MPa	3.42 MPa				
WPU	[42]	64.4 MPa	12 MPa	2.33 MPa	18% (B)	18% (B)		
VAE	[128]			3.35 MPa				
	[129]	27.9 MPa	7.23 MPa		1.07 (C)		0.1 (C)	

Note: In this table, A shows the loss of compressive strength, B indicates the mass loss, and C indicates the corrosion resistance coefficient.

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