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Influence of Polyaluminum Chloride Residue on the Strength and Microstructure of Cement-Based Materials

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ABSTRACT

In this paper, cement and dechlorinated Polyaluminum Chloride Residue (PACR) have been used to prepare a net slurry and mortar specimens. Two hydration activity indicators have been used to quantitatively analyze the dechlorinated PACR hydration activity. In particular, the effect of dechlorinated PACR content on the compressive strength of mortar has been assessed by means of compressive strength tests. Moreover, X-ray diffraction (XRD) and scanning electron microscopy (SEM) have been employed to observe the microstructure of the considered hydration products. The following results have been obtained. The 28th day activity index of the dechlorinated PACR is 75%, and therefore it meets the criterion for the use of active admixture. The increase in the content of the dechlorinated PACR tends to reduce the compressive strength of mortar specimens, however, it is beneficial to its later strength growth. When the content is not greater than 10%, the strength remains unchanged, otherwise, it decreases. The PACR does not form a new crystalline phase in the cement slurry, and the dechlorinated PACR remains active until the age of the 28th day. The inclusion of the PACR mainly deteriorates the early strength of the cement slurry, but it promotes the production of hydration products in the cement slurry after the 7th day.

KEYWORDS

Dechlorinated polyaluminum chloride residue; activity index; strength; microstructure; hydration product

1 Introduction

The incorporation of mineral admixtures and industrial by-product minerals into concretes offers several advantages, including the efficient utilization of industrial solid waste and a reduction in cement consumption. This leads to a decrease in the heat generated during cement hydration and an overall improvement in concrete quality [1]. Mineral admixtures such as fly ash, silica fume, slag, and limestone powder have been widely used in concrete products, effectively enhancing concrete performance and yielding positive social and economic outcomes [2–4]. However, the distribution and properties of mineral admixtures in concrete vary due to geographical and industrial factors, making the development



and utilization of new industrial solid waste concrete admixtures and the performance of existing admixtures crucial focal points within the industry.

Polyaluminum chloride (PAC) is a widely recognized high-quality water purifying agent both domestically and internationally [5,6]. The manufacturing process involves the reaction of calcined bauxite, calcium aluminate powder, and hydrochloric acid to produce liquid polyaluminum chloride, with the resulting precipitated solid forming a waste residue primarily composed of SiO₂, Al₂O₃, CaO, and so on. Due to the increasing demand for water purifiers in urban wastewater treatment, the stock of PACR has been increasing, leading to environmental pollution when the residue is landfilled, particularly due to its acidic nature. However, through water washing and low-temperature calcination for dechlorination, the dechlorinated PACR transforms into metakaolin [7,8], a material similar to high iron and high titanium, holding potential as active admixture in cement-based materials. Modified waste residue can also enhance sludge dewatering performance [9] and be utilized in the production of baking-free bricks [10]. Although the existing research has tested and analyzed the chemical composition of PACR [11] and demonstrated its potential activity, in-depth research on the hydration activity of polyaluminum chloride and its influence on cement-based products remains limited. The hydration activity of PACR is a critical factor that determines the strength of cement-based products [12-14], necessitating a systematic investigation into PACR's hydration activity and strength, as well as its impact on the microstructure of cement-based materials.

In order to utilize industrial waste effectively and cleanly, this paper explores the possibility of PACR as an auxiliary cementitious material. Firstly, PACR undergoes pretreatment, and its chemical composition is analyzed. Subsequently, composite cementitious materials composed of ordinary silicate cement and dechlorinated PACR are used to prepare net slurry specimens and mortar specimens. The effects of dechlorinated PACR on the hydration characteristics of composite cementitious materials are analyzed through compressive strength tests, scanning electron microscopy (SEM), and X-ray diffraction (XRD). Additionally, an experimental study on the activity index of PACR is carried out by comparing mortar specimens with an equal amount of fly ash replacing cement as the control group. This helps elucidate the mechanism of dechlorinated PACR's action in the composite cementitious admixture. The results suggest that PACR can be applied to cement-containing building materials such as mortar, bricks, and concrete. Finally, by determining the optimal amount of PACR as an admixture, this research lays the foundation for future large-scale utilization of PACR.

2 Test Materials and Test Design

2.1 Test Materials and Components

2.1.1 Test Material

The specific materials used in this test encompass P.O 42.5 ordinary silicate cement, ISO standard sand, and river sand. The ISO standard sand serves as a reference for the strength comparison activity index test. The river sand, on the other hand, is utilized for the mortar strength test, after being sieved with only particles below 1.18 mm retained. The density of the river sand is 2.58 g/cm³, with a fineness modulus of 2.4. Furthermore, the test incorporates dechlorinated PACR, which is derived from industrial processes involving water washing, dichlorination treatment, and low-temperature calcination of waste materials. The resulting polymeric aluminum chloride water purification agent is subsequently crushed and ground to form a brownish-yellow powder. It possesses a neutral pH and a density of 2.23 g/cm³.

2.1.2 Dechlorinated Polyaluminum Chloride Residue

The PACR test involves a series of steps. First, PACR is mixed with quicklime at a mass ratio of 20:1 and stirred to ensure even distribution. Next, water is added to create a slurry, which is stirred for 30 min. The

slurry is then left to settle for 15 min, during which time natural precipitation occurs, resulting in supernatant and waste residue. Once the precipitation is complete, the supernatant is carefully removed, and the waste residue undergoes low-temperature calcination and drying at $150^{\circ}C-200^{\circ}C$. This process aims to eliminate chloride ions and acids, as the supernatant contains high levels of CaCl₂ and Ca(OH)₂. Finally, after undergoing procedures such as crushing, screening, grinding, and other procedures by ball mills, the lumpy waste residue is reduced to particles ranging from 0–0.075 mm, resulting in dechlorinated PACR. This dechlorinated PACR is illustrated in Fig. 1.



Figure 1: 0–0.075 mm dechlorinated PACR

Table 1 presents the main chemical components of dechlorinated PACR and cement. It is evident that the dechlorinated PACR has a significant composition of SiO₂ and Al₂O₃, and after the dichlorination process, the chloride ion content is minimal, making it suitable for use as raw materials in cement-based building materials. Fig. 2 illustrates the analysis of crystal composition, revealing the presence of crystalline phases such as SiO₂, Al₂O₃, CaCO₃, and CaTiO₃ in the dechlorinated PACR. The diffraction peak of SiO₂ is the most obvious. The reactivity of silicon depends on its phase form, whether crystalline or non-crystalline. Generally, higher silicon content leads to increased reactivity of volcanic ash after calcination [15–17]. The dechlorinated PACR prepared in this study underwent a chemical composition test, and the results are displayed in Fig. 2. The treatment process involving low-temperature calcination and subsequent procedures suggest the potential reactivity of dechlorinated PACR. Furthermore, the alkaline environment generated during cement hydration may trigger a reaction with the PACR due to its volcanic ash properties.

 Table 1: Main chemical composition of dechlorinated PACR and cement (%)

Kind	SiO ₂	Al_2O_3	Fe ₂ O ₃	CaO	MgO	TiO ₂	Cl-	Other
OPC	20.15	4.76	3.18	64.87	2.87	_	_	1.74
PACR	45.72	27.80	3.42	5.68	1.34	3.86	0.25	10.39

2.2 Test Methods

2.2.1 Polyaluminum Chloride Residue Total Activity Test

The test was conducted following the standard industrial waste activity test method, maintaining a fixed water-binder ratio of 0.5. Initially, the dechlorinated PACR in the 0-0.075 mm size range was mixed with cement at a mass ratio of 3:7. This combination was then used as the cementing material to produce mortar specimens (H1). To compare the activity level of PACR with that of an active admixture fly ash, first-grade fly ash and cement were mixed at a mass ratio of 3:7 to create cementitious material for mortar

specimens (FA). The mortar ratios for each group can be found in Table 2, and the prepared mortar underwent standard maintenance. Compressive strength measurements were taken at different ages, and the resulting values were utilized in the admixture activity index equation to calculate the PACR's activity level.



Figure 2: XRD diffraction pattern of the dechlorinated PACR

Specimen number	Water-binder ratio	Material consumption per unit volume $(kg \cdot m^{-3})$			
		Cement	PACR	FA	Standard sand
H0	0.5	450	0	0	1350
H1	0.5	315	135	0	1350
FA	0.5	315	0	135	1350

 Table 2: Mix ratio of mortar specimens

In this study, the hydration activity of dechlorinated PACR was assessed using two indices: the compressive strength activity index A and the hydration activity contribution rate P_p . Previous research conducted by scholars on the hydration activity of mineral admixtures in cement concrete has yielded valuable findings [18–21]. Following the guidelines specified in GB/T1596-2017 "Fly Ash Used in Cement and Concrete," the compressive strength of the mortar specimen FA, which contained 30% fly ash as a replacement for cement in equal quantity, was measured and compared to the test mortar. The strength activity index A of the specimen was determined by calculating the ratio of the compressive strengths of the two specimens at 28 days, As shown in Eq. (1).

$$A = \frac{H_{\rm t}}{H_0} \times 100\% \tag{1}$$

The compressive strength activity index A for different specimens can be calculated by substituting by substituting the compressive strength test results into Eq. (1). Here, H_t represents the 28th day compressive strength of PACR cement mortar, and H_0 denotes the 28th day compressive strength of cement mortar base group.

To evaluate the effect of the dechlorinated PACR on the compressive strength of the specimens, the mineral admixture activity index evaluation method proposed by Pu [22] was employed. The hydration

activity contribution of the admixture P_p was then calculated using Eqs. (2)–(5), based on the compressive strength test results of the specimens at different ages.

$$P_{\rm p} = \frac{R_{\rm p}}{R_{\rm t}} \tag{2}$$

$$R_{\rm p} = R_{\rm t} - R_0 \tag{3}$$

$$R_{\rm t} = \frac{f}{q} \tag{4}$$

$$R_0 = \frac{R_c}{100} \tag{5}$$

Here, R_t represents the strength provided by the unit cement content to the cement-based material in the admixture cement mortar, while R_0 denotes the strength provided by unit cement content to the cement-based material in the cement mortar. The strength of the admixture cement-based material is denoted as f, and the mass percentage of cement in the admixture cement-based material is represented by q. Furthermore, the strength value of the cement-based material in the control group is labelled as R_c .

2.2.2 Effect of Polyaluminum Chloride Residue Content on Compressive Strength of Cement Mortar

The cement mortar specimens were prepared using the 0–0.075 mm dechlorinated PACR and a fixed water-cement ratio of 1.0, as outlined in Table 3. Five groups of specimens were configured, each with a different residue content. The dechlorinated PACR replaced varying percentages of the cement content in the mortar specimens, namely 0%, 5%, 10%, 15%, and 20% by mass. Each group consisted of three specimens, labelled as PM1, PM2, PM3, PM4, and PM5. The specimen size was 70.7 mm × 70.7 mm × 70.7 mm. After pouring, the specimens were placed in a standard curing box with a temperature of 20°C \pm 2°C and a relative humidity exceeding 90%. Following 24-h curing period, the specimens were demolded and numbered accordingly. Subsequently, the specimens were further cured under the standard environmental conditions, and the compressive strength test corresponding to each age was conducted.

Number	Cement	PACR	River sand	Water	Water-binder ratio
PM1	342.0	0	1411.0	342.0	1.0
PM2	324.9	17.1	1411.0	342.0	1.0
PM3	307.8	34.2	1411.0	342.0	1.0
PM4	290.7	51.3	1411.0	342.0	1.0
PM5	273.6	68.4	1411.0	342.0	1.0

Table 3: Mix ratio of cement mortar specimens (kg·m⁻³)

Note: PM1 (0% dechlorinated PACR cement mortar), PM2 (5% dechlorinated PACR cement mortar), PM3 (10% dechlorinated PACR cement mortar), PM4 (15% dechlorinated PACR cement mortar), PM5 (20% dechlorinated PACR cement mortar).

2.2.3 Microscopic Test

To investigate the correlation between the activity, hydration products, micromorphological characteristics, and strength of the dechlorinated PACR cement-based composite slurry, various experimental procedures were conducted using pure slurry specimens with different PACR content. The specific ratios used in these specimens are provided in Table 4. Firstly, X-ray diffraction (XRD) analysis was used to observe the hydration products of the pure slurry specimens, both from the reference group and those containing different PACR content, on the 7th and 28th days. This allowed for the examination

of the relationship between activity, hydration products, and PACR content. Secondly, scanning electron microscopy (SEM) was employed to study the microstructure of the cement mortar specimens. This involved comparing the reference group specimens with those containing the 0–0.075 mm dechlorinated PACR. Furthermore, SEM was employed to investigate the effect of dechlorinated PACR content on hydration products and the microstructure of cement-based materials. In particular, the pure slurry specimens with 5% and 20% PACR were examined using SEM. However, before conducting the microscopic tests, it was necessary to wait until the specimens were cured to the age of 7th day and 28th day. Specimens with diameters of 5–10 mm were taken from the center of the mortar specimens, stored in glass jars with anhydrous ethanol, and sealed to prevent the continuation of the hydration process. The mortar specimens were then taken out and air-dried before the SEM test to facilitate the examination.

	-	-	
Specimen number	Water-binder ratio	Quality percentage (%)	
		Cement	Chlorinated PACR
P1	1	100	0
P2	1	95	5
P3	1	90	10
P4	1	85	15
P5	1	80	20

Table 4: Proportion of dechlorinated PACR pure slurry specimens

3 Test Results and Analysis

3.1 Hydration Activity of the Dechlorinated Polyaluminum Chloride Residue

According to the results of the compressive strength test, substituting into Eq. (1), the compressive strength activity index A of different specimens can be calculated, and the findings are tabulated in Table 5.

Number	7th day	14th day	28th day
H0	100	100	100
H1	68.1	72.5	75
FA	72.3	78.4	83.9

Table 5: Compressive strength activity index *A* of each specimen (%)

The compressive strength activity index of the dechlorinated PACR at the same age is lower than that of fly ash, indicating that the dechlorinated PACR is a weak active admixture. However, the 28th day activity index of the dechlorinated PACR is 75% according to Table 5, exceeding the critical value of 65% specified in national standards [23] for a volcanic ash activity index. This suggests that the dechlorinated PACR can still be utilized as an active admixture in cement. The activity index of the dechlorinated PACR and fly ash increases with the growth of hydration age.

The contribution of the hydration activity of the dechlorinated PACR with respect to its age was determined by calculating the values of Eqs. (2)–(5) using the compressive strength test results obtained from each specimen at various ages. These calculations were then utilized to create graphs showcasing

the relationship between the hydration activity contribution rate and the age of the dechlorinated PACR, as depicted in Fig. 3.



Figure 3: Hydration activity contribution rate of the dechlorinated PACR

Fig. 3 demonstrates the relationship between the contribution rate of FA hydration activity and age. It shows a gradual increase in the contribution rate with age. The contribution of dechlorinated PACR to hydration activity, however, initially had a negative impact until the 7th day. This suggests that under these conditions, the presence of dechlorinated PACR had some adverse effect on slurry properties. With the exception of the 7th day, the contribution of dechlorinated PAC sludge to hydration activity gradually increased with age, although the rate of increase slowed down as the age progressed. This behavior can be attributed to the decrease in cement and calcium ion content in the slurry due to the replacement of cement with dechlorinated PACR. Additionally, before the 7th day, the low hydration degree of cement led to low alkalinity in the slurry, which did not stimulate the activity of dechlorinated PACR. Furthermore, as an inert admixture, dechlorinated PACR does not undergo a volcanic ash reaction. As the age increased, the hydration degree deepened, resulting in higher alkalinity and Ca(OH)₂ content in the slurry. This stimulated the activity of dechlorinated PACR, resulting in an increased contribution to hydration activity.

3.2 Influence of Polyaluminum Chloride Residue Content on Compressive Strength

The test results are illustrated in Fig. 4 and Table 6, which involved the design of five groups of cement mortars. The 0–0.0.75 mm dechlorinated PACR was used to replace cement in equal amounts at mass percentages of 0%, 5%, 10%, 15%, and 20%.

Fig. 4 illustrates that the 7th day compressive strength of dechlorinated PACR cement mortar gradually decreases as the PACR content increases. However, for contents below 10% (PM2 and PM3 groups), the 28th day compressive strength of dechlorinated PACR cement mortar remains comparable to that of the PM1 group without dechlorinated PACR. Moreover, as indicated in Table 6, when the PACR content increases from 15% to 20% (PM4 and PM5 groups), the average 7th day compressive strength of dechlorinated PACR cement mortar experiences the most significant decrease, measuring 1.7 MPa. Conversely, the average 28th day compressive strength of these two groups of specimens shows a lesser decline, with a decrease of 0.9 MPa. Notably, there is even a slight increase in strength after substituting 5% dechlorinated PACR for cement. This suggests that PACR primarily influences the early strength of mortar and reacts with cement hydration products during later stages, resulting in the formation of new hydration products and improved strength of the 28th day.



Figure 4: Compressive strength of 0–0.0.75 mm dechlorinated PACR mortar specimens with different contents

Table 6: Average compressive strength of 0-0.0.75 mm dechlorinated PACR mortar specimens with different contents (MPa)

Number	7 d	28 d
PM1	13.5	17.9
PM2	12.5	19.3
PM3	11.2	17.9
PM4	10.4	16.2
PM5	8.7	15.1

3.3 Hydration Activity of Dechlorinated Polyaluminum Chloride Residue

The results of XRD analysis for cement slurry specimens are presented in Fig. 5. The specimens include pure cement net slurry as well as slurry mixed with 5%, 10%, 15%, and 20% dechlorinated PACR. The analysis was conducted at 7 days and 28 days of age.



Figure 5: XRD spectrum of cement pure slurry specimen

Fig. 5 shows that the hydration products of pure slurry specimens remain consistent regardless of the addition of dechlorinated PACR. These products mainly consist of Ca(OH)₂, calcium carbonate (CaCO₃), tricalcium silicate (Ca₃SiO₅), and a small amount of ettringite (AFt). The chromatogram of specimens mixed with dechlorinated PACR at 28 days reveals a diffraction peak of SiO₂, indicating the presence of this crystal phase introduced directly by the active substance of dechlorinated PACR after precipitation. It is evident that dechlorinated PACR does not generate new crystal phases in the slurry. Comparing the hydration products of net slurry specimens with and without PAC slag doping, there is no significant difference observed in the main peak of Ca(OH)₂ and the characteristic peak of Ca₃SiO₅ in the hydration product at 7 days. However, at 28 days, these peaks weaken. This phenomenon is attributed to the low substitution rate of slag replacing cement and the reduction of cement content in the system, resulting in minimal changes in the crystalline phase. Additionally, the insufficient release of PACR activity at 7 days and the volcanic ash reaction consuming a portion of Ca(OH)₂ with increasing age contribute to these changes. When comparing net slurry specimens with different dosages of PACR, the waste slag dosage varies (0%, 10%, 20%). By observing the characteristic peaks of CaCO₃ and Ca₃SiO₅ in the hydration products of 7 days and 28 days, it can be seen that the enhancement of the hydration products of 28 days is obvious, and at the same time, the characteristic peaks of SiO₂ are clearly enhanced. However, in the 28 days plots, it can be seen that the hydration products of the three groups of net slurry specimens with different dosages of waste slag are essentially the same. This finding indicates that the addition of PACR admixture does not substantially alter the hydration reactants of cement, but only has a marginal effect on the rate of hydration.

Electron microscopy was employed to analyze the internal microstructure of mortar specimens from two groups: the H0 group (specimens without dechlorinated PACR) and the H1 group (specimens with 0–0.075 mm dechlorinated PACR). Scans were conducted on the 7th and 28th days, and the results are displayed in Fig. 6.

The existing studies have indicated that the microscopic morphology of cement hydration products, specifically C-S-H gels, typically exhibit spiny-like, needle-like, column-like, and network-like gel particles. Ettringite (AFt) is predominantly needle-like [24,25]. From Figs. 6a and 6b, it is apparent that on the 7th day, a large number of "network-like" C-S-H gels and some "needle-like" AFt were observed in the baseline group H0. The hydration products were interlaced, and the overall structure was dense. In contrast, in group H1, which was mixed with the dechlorinated PACR, a small number of C-S-H gels were observed, showing cluster radial growth. The microstructure of the H1 group contained more pores, generated fewer hydration products compared to the H0 group during the same period, and appeared looser due to the accumulation of unreacted dechlorinated PACR particles. Analysis of Figs. 6c and 6d a reveals that on the 28th day, the H0 group exhibited a dense skeleton attributed to the encapsulation of AFt by the settled C-S-H gel. Microporosity was reduced. On the other hand, the microstructure of the H1 group contained more pores, and the presence of these pores offered less sufficient support from AFt generation. In comparison to the H0 group during the same period, the growth of hydration products in the H1 group was more pronounced, accompanied by the formation of a C-S-H gel matrix and inter-pore AFt. This indicates that as the hydration reaction progresses, the dechlorinated PACR activity is gradually released, and a volcanic ash reaction occurs, promoting the generation of hydration products in the cement-dechlorinated PACR composite cementitious system.

To investigate the impact of chlorinated PACR content on hydration products and microstructure in cement-based systems, SEM was utilized to observe the micro-morphology of pure slurry specimens with 5% (P2) and 20% (P5) dechlorinated PACR. The results are presented in Fig. 7.



Figure 6: Comparison of microstructure between cement mortar specimen and dechlorinated PACR cement mortar specimen

(d) Specimen H1 group on the 28th day

(c) Specimen H0 group on the 28th day

Upon comparing (a) and (b) in Fig. 7, it is evident that on the 7th day, the P2 specimen exhibited a significantly higher amount of hydration products compared to the P5 specimen. The P2 specimen contained numerous needle rod-like AFt structures within the pores. This finding aligns with a study conducted by Liu et al. [26], which suggests that when weak active admixture is used instead of cement, it fails to stimulate activity due to insufficient alkalinity. In the P5 specimen, PACR particles accumulated, and although some hydration products were present on the surface of the waste residue particles, their integrity was poor. Furthermore, several pores and cracks were observed between the structures, indicating insufficient alkalinity in the slurry. Consequently, the hydration reaction was reduced, leading to fewer hydration products to fill the pores of slurry on the 7th day.

Examining Figs. 7c and 7d reveals that on the 28th day, the P2 specimen contained a considerable amount of columnar C-S-H gel, partially laminated sheet $Ca(OH)_2$ crystals, and acicular AFt. These findings indicate a compact structure within the slurry. In contrast, the unreacted PACR particles disappeared in the P5 specimen, which exhibited abundant hydration products with tightly bonded structures. This outcome can be attributed to the increased hydration degree and alkalinity of the slurry as

it aged. When comparing cement specimens with 5% and 20% PACR at the same age, it is evident that the latter still contained a significant number of pores within the microstructure and exhibited poor microstructural integrity. This explains why the compressive strength of PACR cement mortar with 20% PACR content at the same age is lower than that of PACR mortar with 5% PACR content.



(c) Specimen P2 group on the 28th day

(d) Specimen P5 group on the 28th day

Figure 7: Comparison of microstructure between cement mortar specimen and dechlorinated PACR cement mortar specimen

4 Conclusion

Through various test methods such as the PACR composition test, PACR activity index test, the effect of PACR content on compressive strength of cement mortar, and microscopic examination, the collected data will be organized and analyzed. Based on the obtained results, the following key findings can be made:

1. The dechlorinated PACR exhibits a low strength activity index in the early stages. However, with increasing age, the activity index gradually improves, reaching 75% on the 28th day. When compared to fly ash, dechlorinated PACR behaves as an admixture with weak activity, as its strength activity index is lower.

2. As the content of dechlorinated PACR increases, the compressive strength of the dechlorinated PACR-cement mortar decreases. The addition of PACR mainly deteriorates the early strength of the mortar. When the PACR content is small (not more than 10%), the 28th day compressive strength of the mortar remains relatively unchanged.

3. The dechlorinated PACR does not introduce new crystalline phases in the cement paste. On the 7th day, the reaction degree of the dechlorinated PACR is low, while the hydration cementation products increase significantly by the 28th day of age. This indicates that PACR contributes to the later strength growth of the cement paste.

4. With an increase in dechlorinated PACR dosage, the early hydration products of cement slurry reduce, leading to an increase in structural porosity and an impact on mortar strength. However, as the hydration age progresses, the internal structure of the slurry undergoes significant improvement. Notably, the dosage of PACR in cement mortar should not exceed 10%.

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