# **Recycling waste fresh paste through a Dormant & Awaking re-utilization approach**

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Received March 4, 2024 Accepted September 17, 2024 Available on line March 28, 2025

**ABSTRACT:** This paper proposed a Dormant & Awaking approach for the management and recycling of waste fresh paste (WFP), a significant byproduct of concrete mixing plants. The approach involves inducing dormancy in WFP using citric acid to slow down cement hydration; whereas calcium aluminate cement (CAC) was utilized to awaken the dormant WFP. Experimental results revealed that treating WFP with citric acid effectively impeded cement hydration, allowing for extended collection and storage time. The addition of CAC facilitates rapid hydration by absorbing and utilizing free water in the pore solutions of cement. Consequently, dormant WFP can be awakened through the rapid precipitation and assembly of hydration products. By adding 17.4% or 26.1% CAC, the compressive strength of WFP at 28 days of curing can be mostly recovered to levels comparable to the blank mixture. A flexible and efficient solution for WFP recycling is thus provided.

KEY WORDS: Waste fresh paste; Dormancy; Awakening; Hydration; Compressive strength.

**Citation/Citar como:** Zong Z, Chen P, Qian X, Li J, Fang H, Wang L. 2025. Recycling waste fresh paste through a Dormant & Awaking re-utilization approach. Mater. Construct. 75(357):e363. https://doi.org/10.3989/mc.2025.379024.

**RESUMEN:** *Reciclaje de residuos de hormigón fresco mediante enfoque de reutilización "Inactivar y Despertar.* Este artículo propone un enfoque innovador para gestionar y reciclar los residuos de hormigón fresco (WFP) de las plantas mezcladoras de hormigón mediante un método de Inactivar y Despertar. Este método utiliza ácido cítrico para inducir la inactivación en la WFP, ralentizando la hidratación del cemento, y luego emplea cemento de aluminato de calcio (CAC) para reactivar la hidratación. Los experimentos revelan que el ácido cítrico detiene efectivamente la hidratación del cemento, permitiendo un mayor tiempo de recolección y almacenamiento. La adición de CAC facilita la rápida rehidratación al absorber y utilizar el agua libre en los poros del cemento. Con una adición del 17.4% o 26.1% de CAC, la resistencia a la compresión de la WFP se recupera a niveles comparables con la mezcla original después de 28 días de curado. Este método ofrece una solución eficiente y flexible para el manejo de la WFP.

PALABRAS CLAVE: Residuos de hormigón fresco; Inactivación; Despertar; Hidratación; Resistencia a la compresión.

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## **1. INTRODUCTION**

Waste fresh concrete (WFC) represents a major waste in ready-mixed concrete plants, produced through delayed transportation, excess ordered concrete, and the clean of mixing equipment (1). It is known that about 0.3-0.4% WFC can be produced per day from a typical ready-mix concrete plant with a daily production capacity of 1000 m<sup>3</sup> (2300-2800 tons) of concrete (2). As a result, about 3000 tons of WFC would be produced from one batching plant each year. Since WFC contains partially hydrated cement under high pH conditions, it will cause serious pollution of water and soil without effective utilization, which has caused widespread concern.

Nowadays, effective methods for the treatment of WFC are still being explored, whereas few methods are available in practice. For example, researchers have proposed the use of WFC as the feedstock of degraded aggregates by crushing for new concrete production (3-5). Kou et al. (2, 6) showed that the incorporation of crushed WFC should not exceed 30% due to its unsuitability as a coarse aggregate, characterized by increased water absorption and decreased density resulting from its high porosity. Sérifou et al. (7) revealed that the application of crushed WFC as a fine aggregate seems to be more promising for fresh concrete. Although the compressive strength at 28 days of curing was reduced by only 32% when WFC replaced 100% of the total aggregate, it was limited to applications such as pavements and concrete blocks.

In other cases, the aggregates in WFC were separated for new concrete production, leaving waste fresh paste (WFP) away as a waste. Given that the cement in WFP is hydrating and far away from complete hydration, the above recycling method would fail to explore the potential of the binding capacity of WFP, which is crucial for WFC utilization through a value-elevated manner. As WFP has undergone hydration for a certain period, it encounters challenges such as limited working time due to the initiation of setting and hardening. Additionally, the generation of WFP in practice is an intermittent continuous process, with each WFC generation varying considerably in both quantity and proportion, which makes the collection and reuse of WFP difficult (6).

Therefore, a method named Dormant & Awaking treatment is proposed in this study for achieving an optimal re-utilization of WFP. Dormancy involves slowing down the cement hydration process to a nearly stagnant state while still maintaining a flowable state for extended period, giving sufficient time for collecting WFP. Awakening, on the other hand, entails accelerating the cement hydration process, causing the dormant concrete to set and harden again with satisfactory performance. Through such a way, the hydrating cement can still be used to bind aggregates without generating additional waste, towards to a cleaner way. This treatment can be carried out simply by employing a variety of commercial retarders, i.e., hydroxy carboxylic acids, polyols, and saccharide compounds (8-10). However, the main challenge of the dormancy-awakening method lies in breaking the dormant state of the WFP to enable continuous hydration and setting as required.

To this regard, calcium aluminate cement (CAC) was applied in this work as the awakener for dormant WFP to achieve rapid setting and hardening (11). CAC, primarily composed of monocalcium aluminate, initiates reactions distinct from those in ordinary Portland cement (OPC) hydration. Specially, CAC is widely used in rapid military repair and construction because it can achieve strength equivalent to OPC at 28 days of curing within just 5-6 hours (12). The hydration of CAC leads to the rapid formation of calcium aluminate hydrates such as CAH<sub>10</sub> and C<sub>2</sub>AH<sub>8</sub>. In contrast, OPC hydration involves the formation of calcium silicate hydrates and portlandite, which typically result in slower strength development (13).

When CAC is combined with OPC, the interactions between the different hydrates can create complex synergistic effects. Previous studies have revealed that the aluminate phases from CAC reacts rapidly with the hydration by-product calcium hydroxide in OPC to form additional calcium aluminate hydrates, contributing to a fast set in binary mixes of OPC/CAC (14-16). Additionally, Le Saout et al. (17) describes that as well as calcium aluminate hydrates, the precipitation of ettringite from the dissolution of monocalcium aluminates contained in CAC and the available sulfate may also dominate the early strength. Gu et al. (18) further proclaimed that the exothermic nature of CAC hydration increases the internal temperature of the cement paste and promotes the hydration reaction, addressing the limitations of OPC related to slow mechanical strength development and prolonged setting duration.

In summary, a dormancy-awakening treating method is implemented for WFP recycle in this paper using citric acid as the dormancy agent and CAC as the awakener. In order to evaluate the recyclability of fresh WFP, a reasonable concentration of CAC was searched to utilize for waste reduction. A comprehensive study is carried out to analyze the properties of products of dormant WFP due to different concentration of CAC; and the setting time, hydration products, compressive strength, pore structure, and microstructure of awakened WFP were studied.

# 2. MATERIALS AND METHODS

# 2.1. Materials

Ordinary Portland cement (P. O. 42.5) was used, sourced from Anhui Conch Cement Company. Calcium aluminate cement (CAC) of CA50 grade was procured from Tianjin Zhiyuan Chemical Reagent Co., Ltd., in China. The chemical composition, density, BET area, and the particle size distributions of the two materials were presented in Table 1 and Figure 1. City tap water was used for the mixtures. Additionally, the used citric acid was supplied by Tianjin Zhiyuan Chemical Reagent Co., Ltd.

# 2.2. Mixing proportion and preparation process

A cement paste with w/c of 0.45 was prepared in a laboratory to simulate WFP in this paper. The dormancy treatment of this WFP was achieved using commercial citric acid as the retarder, 0.5wt% of OPC, as presented in Table 2. A total of six mixtures were prepared in which L0 was treated with citric acid only in a dormant state. Specially, the dormant WFPs were placed in a closed apparatus in a laboratory environment for 24 hours, considering a proper flowability. Then, paste samples were prepared for analysis when CAC with mass fractions of 8.7%, 17.4%, 26.1%, and 34.8% was added to the dormant WFPs, and named as L3, L6, L9, and L12, respectively.

In a typical mixing procedure, a citric acid aqueous solution was first prepared. This involved adding citric acid to water and thoroughly mixing to ensure complete dissolution. Subsequently, the solution was cooled to room temperature and mixed with cement in a mixer for 3 mins to be used as a laboratory simulation of dormant WFP. After 24 hours, CAC was added to the dormant paste, followed by an additional 3 mins of mixing to obtain a fresh paste. The whole experiment was carried out in the laboratory at a temperature of  $23 \pm 1$  °C.

## 2.3. Experimental methods

## 2.3.1. Setting time

Vicat needle test was used to test the initial and final setting times of pastes according to ASTM C191 (19) in a standard curing room set at  $23 \pm 1$  °C and  $\geq 95\%$  relative humidity.

	Chemical composition / %						Density /	BET area /	
	CaO	SiO <sub>2</sub>	Fe <sub>2</sub> O <sub>3</sub>	Al <sub>2</sub> O <sub>3</sub>	SO <sub>3</sub>	MgO	Others	g/cm <sup>3</sup>	$m^2/g$
OPC	49.70	29.60	2.40	7.77	4.70	3.60	2.23	3.1	1.7
CAC	31.40	7.57	1.58	54.86	0.60	0.48	3.51	3.0	4.1

TABLE 1. The chemical composition of OPC and CAC.



TABLE 2. Mixing	proportions (	kg/m <sup>3</sup>	).
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Group	OPC	Water	Citric acid	CAC
NM	1839	826	0	0
L0	1839	826	9	0
L3	1839	826	9	160
L6	1839	826	9	320
L9	1839	826	9	480
L12	1839	826	9	640

FIGURE 1. Particle size distributions of OPC and CAC.

#### 2.3.2. Compressive strength

Following the guidelines outlined in ASTM C192 (20), the procedure involves pouring the fresh mortar into cubic molds that have been prepared in advance, with each side measuring 50 mm. The next step was to vibrate the molds to remove any trapped air and ensure uniform filling. Subsequently, the samples should be covered with plastic film and left to cure for 24h. The samples were then demolded and still cured in the room till the ages of 3, 7, and 28 days prior to testing by a DYE-300A presser. The average value and standard error were estimated based on triplicate samples.

# 2.3.3. Hydration products analysis

The specimens were first immersed in isopropanol for 7 days to halt cement hydration, following by 24 hours of vacuum drying and pulverized to a particle size range of less than 75µm. The mineralogical compositions of pastes were analyzed using a TTR-III h/h rotating anode X-ray diffractometer (XRD) with Cu K $\alpha$  radiation (40 kV, 200 mA). XRD scans were performed at a rate of 5 °/min over a 2 $\theta$  range of 5°-60°. Subsequently, a TGA/DSC 1/1600 thermogravimetric analyzer (TGA) was employed. Samples were heated in an alumina crucible under N<sub>2</sub> atmosphere from room temperature to 900 °C at a rate of 20 °C/min. The sample preparation method for TGA analysis was identical to that of XRD analysis.

#### 2.3.4. Mercury intrusion porosimetry (MIP)

The pore structures of pastes at 28 days of curing involved the utilization of a MicroActive AutoPore V 9620 MIP. Cube samples (20 mm  $\times$  20 mm  $\times$  20 mm) were immersed in isopropanol during 7 days and vacuum dried for 24 hours before analysis.

# 2.3.5 Microstructure analysis

A Flex1000 scanning electron microscope (SEM) was utilized to examine the microstructure of 28d pastes. The specimens were cut into cubic samples of  $20 \text{mm} \times 20 \text{mm} \times 20 \text{mm}$  size, soaked in isopropyl alcohol for 7 days to terminate hydration and dried in vacuum before internal samples were taken. Before analysis, a layer of gold particles was sputtered for 120s to gain a good conductivity.

# **3. RESULTS AND DISCUSSION**

## 3.1. Setting time

Table 3 presents the setting time of pastes. It clearly indicates that the addition of 0.5% citric acid to L0 significantly prolonged both the initial and final setting times of the sample by 13 and 26 times, respectively, relative to NM. This extension in setting times allows for a longer working time in practical engineering, particularly for the collection and accumulation of waste concrete. As 8.7% CAC (L3) was added, the setting time of awakened pastes notably shorter than that of L0, suggesting the efficient effect of CAC as an awakener. This is mainly because that CAC hydrates rapidly, and its hydration products contribute to construct the initial solid skeleton of the fresh paste (12). Furthermore, the hydration of CAC with water led to a reduction in the water content in the pore solutions of cement, which in turn enhanced the concentration of ions in the pore solutions, thereby accelerating the cement hydration process (15, 16).

On the difference between these pastes with CAC, it was observed that when the CAC content exceeded 17.5% (L6), there was an acceleration in the setting process of the pastes. However, further increases in CAC showed diminishing returns in awakening dormant pastes, as indicated by the similar setting times of pastes L6, L9, and L12. In other words, the awakening effect of CAC is more pronounced at lower concentrations, and excessive CAC appears to be unnecessary. Consequently, the results suggest the importance of optimizing the dosage of set retarder and awakener in a synergistic manner to establish an efficient dormancy-awakening system (11).

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	NM	L0	L3	L6	L9	L12
Initial setting	8.8	116.4	7.5	3.6	3.1	2.2
Final setting	9.2	242.8	8.9	4.1	3.4	2.9

TABLE 3. The setting times of pastes (h).

Materiales de Construcción 75 (357), January-March 2025, e363. ISSN-L: 0465-2746. https://doi.org/10.3989/mc.2025.379024

# 3.2. Hydration products of pastes

#### 3.2.1. XRD analysis

Figure 2 illustrates XRD patterns of pastes at 3 and 28 days of curing, revealing the identification of five substances, including Ca(OH)<sub>2</sub> (CH)), alumina ferric trisulfate (AFt), tricalcium silicate (C<sub>3</sub>S), dicalcium silicate ( $C_2S$ ), and calcium aluminate hydrate ( $C_3AH_6$ ) based on references and JCPDS files (11, 16). The impact of citric acid on the early-age hydration of cement is evident in Figure 2a, where the peaks of C<sub>2</sub>S and C<sub>3</sub>S of L0 show an increase compared to NM, indicating a significant retardation. Upon the addition of CAC, a new mineral, C<sub>3</sub>AH<sub>6</sub>, is observed at 20=11.0°, 32.2°, and 37.1°, attributed to the hydration of monocalcium aluminate in CAC. Previous studies have indicated that CAC can rapidly hydrate to produce metastable hydrates  $CAH_{10}$  and  $C_2AH_8$  phases, which subsequently transform into the more stable cubic hydrogarnet phase  $(C_3AH_6)$  with a reduction in the volume of solid phase (11, 21). Ding et al. (22) showed that the conversion of CAH10 to C3AH6 resulted in a decrease of about 50% while the conversion of C<sub>2</sub>AH<sub>8</sub> to the cubic phase resulted in a decrease of about 65% of the original volume of the reactants. Therefore, the conversion process would cause an increase in porosity and a decrease in mechanical strength of the paste. Although Ca-bearing hydrates such as CAH<sub>10</sub> and C<sub>2</sub>AH<sub>8</sub>, were not detected by XRD analysis, several factors could account for their absence. These hydrates may be present in quantities below the detection limit of XRD, and their low crystallinity makes them difficult to detect. On the other hand, these phases rapidly transform into the more stable C<sub>3</sub>AH<sub>6</sub> as a result of temperature increase under hydration conditions (23). Furthermore, the addition of CAC impacts the formation of AFt, as evidenced by the increase in intensity of the peak at  $2\theta=11.0^{\circ}$  with the inclusion of CAC. Given that the generation of AFt is closely linked to the presence of aluminum phases, the dissolution of CAC is likely the primary factor contributing to the formation of AFt, as it can supply abundant Al phases (15).



FIGURE 2. XRD patterns of pastes cured at (a) 3 and (b) 28 days.

In addition, the peak intensities of  $C_2S$  and  $C_3S$  in the presence of CAC exhibit significantly higher values compared to L0. And a substantial addition of CAC leads to a weakening trend in CH peaks, suggesting a retarding effect on the hydration process of OPC. Essentially, the presence or hydration of CAC inhibits the hydration of OPC. This phenomenon can be attributed to the rapid consumption of mixing water by CAC during its hydration process, consequently reducing the availability of free water for the hydration of OPC. Moreover, the by-products formed during the hydration of CAC may precipitate on the grains or initial hydration products of OPC, thereby hindering the diffusion of ions and delaying the hydration of OPC (24).

At 28 days, XRD pattern showed a decrease in the intensities of diffraction peaks corresponding to  $C_2S$  and  $C_3S$  in comparison to the results at 3 days. This decline indicates that a significant portion of the cement clinker had undergone reactions by this stage, resulting in a marked increase in the generation of CH. The peaks associated with the unhydrated cement particles of L3 and L6 have been diminished to levels similar to that of NM, with comparable CH peak intensities. Consequently, for practical engineering applications and to enhance the overall progress of cement hydration, optimizing the CAC/cement ratio is crucial.

Materiales de Construcción 75 (357), January-March 2025, e363. ISSN-L: 0465-2746. https://doi.org/10.3989/mc.2025.379024

#### 3.2.2. TGA-DTG

Figure 3 presents the TGA-DTG thermograms of pastes at 3 and 28 days of curing. Five endothermic peaks are identified in DTG curves, reflecting the decompositions of AFt and C-S-H (80-130°C), AFm (180-200°C),  $C_3AH_6$  (280-350°C),  $Ca(OH)_2$  (400-500°C) and  $CaCO_3$  (600-800°C) (10, 24-26). The presence of CaCO<sub>3</sub> may result from the carbonation of hydrated Ca(OH)<sub>2</sub> during the curing and processing processes of samples.



FIGURE 3. TGA-DTG thermograms of pastes cured at (a) 3 and (b) 28 days.

It can be seen that the total mass losses of pastes cured at 3 days containing citric acid are less than that of NM, revealing the generation of less hydration products due to the retarding effect of citric acid. With the addition of CAC, the dormant pastes have experienced more mass losses during the heating process compared with L0, where a higher dosage of CAC has resulted in a larger mass loss. It is thus suggested that the elevated dosage of CAC accelerated the early-age hydration within the composite system. Although the Ca(OH)<sub>2</sub> peaks in the D-TG curves of CAC are more intense than that of L0, which still less pronounced relative to NM, indicating that the hydration promotion effect of CAC was dominant in the early-age hydration of composite cement. Clearly, the presence of larger amounts of CAC in the pore solution retards the hydration of OPC at early-ages as previously reported. Analysis of the curves shows that L3 exhibited the highest mass loss of Aft, with higher CAC dosages associated with lower mass losses. Simultaneously, the mass loss of AFm tended to increase, particularly for L9 and L12, where the mass loss of AFm exceeded that of NM. It clearly confirms that the addition of CAC promotes the presence of aluminum phases in pastes and accelerates the transition from acicular AFt to plate AFm according to Redondo-Soto et al (24). For a cement hydration system, the formation of AFt results in a solid phase volume increase of 164%, whereas the formation of AFm only increases the solid phase volume by 79.2% (27). Therefore, the transformation of a large amount of AFt into AFm under high dosage of CAC in the composite system inevitably leads to the decrease of solid phase volume and the increase of porosity (28). After 28 days of hydration, it was observed that the mass loss of L0 closely resembled that of NM, in contrast to the noticeable difference observed at 3 days, and the total mass losses of hydration products of CAC-added pastes all exceeded L0 and NM, except for L3. This implies that the retarding effect of citric acid gradually diminished, allowing the long-term hydration to be predominantly influenced by the hydration promotion effect of CAC.

Quantitative calculations were performed for hydration products to assess the effect of CAC on the hydration products of pastes. The mass loss of AFm  $(180\sim200^{\circ}C)$ , Ca(OH)<sub>2</sub>  $(400\sim500^{\circ}C)$  and CaCO<sub>3</sub>  $(600\sim800^{\circ}C)$  are given in Figure 4. The mass losses of Ca(OH)<sub>2</sub> in all samples are consistently below L0 at 28 days and gradually decrease, which suggests that CAC suppresses the long-term hydration of OPC. This may be also attributed to the reaction between CAC and CH, leading to the formation of calcium aluminate hydrate (12). Moreover, the increased mass loss of CaCO<sub>3</sub> in L0 could be attributed to the slow condensation of the paste after extended carbonation. In contrast, composite pastes containing CAC exhibit lower mass losses of CaCO<sub>3</sub> compared to NM, representing a significant improvement from L0. This highlights that the incorporation of CAC could enhances carbonation resistance.



FIGURE 4. Mass loss rate of different hydration products obtained from the TGA results of pastes cured at (a) 3 and (b) 28 days.

## 3.3. Compressive strength

Figure 5 shows the influence of different concentration of CAC on the compressive strength of pastes. The addition of citric acid has strongly retarded the hydration of cement, with L0 failing to achieve hardening within the initial 7 days of curing. Therefore, the dormant paste allowed for extended operational time for their subsequent for reutilization, a crucial aspect in practical engineering applications involving the collection and accumulation of waste fresh concrete before awakening CAC. As 8.7% CAC was added, the dormant state of the paste was converted into an awakened state, as L3 exhibited early-age hardening with a 3-day compressive strength of 3.6 MPa, suggesting the insufficient presence of AC to trigger a high-level hydration of cement. However, the 28-day compressive strength of L0 developed to a close value to that of NM with 12.3% reduction. It has been well clarified that the addition of set retarder in cement could homogenize the hydration of cement for the generation and precipitation of hydration products in slower ways (29, 30). The overall hydration degree of cement can be promoted at a sufficient long period of curing, e.g., 180 days (31).

Nevertheless, extending the curing period to 28 days results in L3 exhibiting compressive strength surpassing that of NM by 17.2%. This implies a synergetic hydration effect on cement has been contributed with the addition of CAC (32). As discussed earlier, in the pore solution of cement, the presence of CAC with high reactive minerals could absorb and consume some of free water for its rapid hydration. The hydration of cement in the state of dormancy retarded by citric acid can be thus rebooted as the ions concentration of pore solutions increased (33). Therefore, as more stress-bearing products were produced, the paste can gradually harden to achieve a compressive strength at early ages despite very low values. This means that the retarding effect of citric acid may still dominant the overall hydration of the blended cement due to insufficient CAC. Then, the hydration homogenizing effect of set retarder further enhanced the compressive strength of pastes based on the synergetic hydration effect owing to CAC.

With the increase of the CAC addition, one can find that the 3-day and 7-day compressive strengths of pastes increase gradually, implying the enhancement in the synergetic hydration effect of CAC addition, while the 28-day compressive strength of pastes tends to decrease compared with L3. This indicates that by adding more CAC, the hydration homogenizing effect of CAC has been weakened to exert a robust influence on the precipitation and assembly of hydration products. By the way, in consideration of the balance between early strength gains, long-term strength development and material costs, 17.4% (L6) or 26.1% (L9) can be optimal ones to achieve a dormancy-wakening technology application that requires both performance and economic efficiency. This is particularly important in large infrastructure projects, precast concrete production, and situations requiring staged construction processes.

#### 3.4. Pore structure

Pore structures of the pastes cured at 28 days were obtained by MIP analysis and shown in Figure 6. Two major peaks appear in the pore size distribution curve of NM at 5 nm and 100 nm, representing gel pores and capillary pores (34). When added with citric acid for dormant treatment, significant changes in the pore volume of L0 were observed, with an increase in the volume of pores larger than 300 nm. This increase suggests higher porosity due to gas residue formation induced by retardation. In contrast to gel and capillary pores, which typically result from the accumulation of hydration products, the presence of large pores exceeding 100 nm is often attributed to the entrapment of air voids during sample preparation and

curing. The evolution of capillaries in the range of 10-100 nm led to a shift in the peak of capillary pore size from 100 nm in NM to 77 nm in L0. The significant changes in the volume of these pores underscore the pore-refining effect of citric acid as a retarder. This effect is linked to the homogenization of hydration facilitated by the delayed setting of cement and the slower generation and precipitation of hydration products. Additionally, a slight tendency towards increased capillary pore volume was also observed.

The introduction of CAC during the dormant phase of the paste significantly alters the pore structure of the samples. There is a marked decrease in the pore volume below 100 nm, along with a noticeable leftward shift tendency in the pore volume of the samples with the added CAC. This shift suggests a more pronounced refinement in pore size, highlighting the effect of the synergistic interaction between CAC and cement on the precipitation and arrangement of hydration products (35). However, it is worth noting that the samples mixed with CAC were overall lower than NM in the gel pore range, revealing that accelerated hydration could have a detrimental effect on the formation of gel products.



distributions of pastes cured at 28 days.

#### **3.5.** Microstructure analysis

Figure 7 presents the SEM images of pastes cured at 28 days. As can be seen from Figure 7a, the microstructure of NM is dense, with clearly visible Portlandite crystals. The  $Ca(OH)_2$  crystals appear as finely dispersed particles with complex and irregular shapes. When citric acid was added, as shown in Figure 7b, significant changes were observed with no formation of Portlandite crystals in L0. Previous studies using malic acid, a carboxylic acid from the same group as citric acid, also showed minimal portlandite formation (36). Rai et al. (37) suggested that during the hydration process, portlandite can reacts with citric acid evolving heat of neutralization leading to the disappearance of portlandite. Additionally, obvious cracks appear in the interfacial transition zone of the hydration products, indicating a weakening of the cement bonding properties. The hydration of cement was strongly retarded by citric acid, which prevented more free water and unreacted components from forming a strong bond.

Figure 7c further illustrates the microscopic morphology of the composite system obtained after the addition of CAC, where a certain amount of flocculent C-S-H gel, plate Ca(OH)<sub>2</sub>, and acicular AFt can be seen. Unlike NM, the Ca(OH)<sub>2</sub> crystals in L3 have grown in large concentrated masses in the form of clusters. Furthermore, the presence of larger Portlandite crystal formations which have grown on existing clusters rather than precipitating new crystals within the microstructure, supporting the idea that CAC promotes cement hydration. However, as the CAC dosage further increases (Figure 7e), the paste exhibits a large amount of plate substances without observable acicular AFt. Combined with the above TGA results, the plate substances in the paste are a mixture of calcium aluminate hydrate and AFm (38). The former is directly hydrated by CAC, while the latter results from the transformation of early-formed AFt due to insufficient sulfate sources in the paste (39). Moreover, a high CAC addition enables a looser cross-section microstructure, and the presence of a large number of large pores with diameters up to several microns is visible. This could be attributed to the transformation of an amount of AFt to AFm and the phase transformation of early-formed unstable hydrated calcium aluminate, which results in a reduction in the volume of solid phase and an increase in porosity within the cement hydration system (22).



FIGURE 7. SEM images of pastes cured at 28 days: a) NM; b) L0; c) L3; d) L6; e) L9; f) L12.

# 4. CONCLUSIONS

This study explored the use of CAC as an awakening agent for accelerating the solidification process of paste subsequent to citric acid-induced dormancy. This approach facilitated the recycling of WFP through a Dormant & Awaking approach. Results demonstrated that following a 24 hours dormancy period, CAC significantly reduced the setting time of the paste by facilitating the rapid involvement of the highly reactive minerals present in CAC. The impact of CAC on cement hydration was primarily associated with the dosage increment. TG-DTG curves illustrated the significant role of CAC in enhancing the early-stage hydration of composite cement, while the presence of a considerable amount of CAC in the pore solution led to a delay in the early hydration of OPC. In this case, the synergistic hydration of CAC with OPC strongly influenced the hydration product precipitation and assembly. Consequently, the compressive strength of L3 sample exhibited a 17.2% enhancement at 28 days of curing compared to NM sample, accompanied by a densification of pore structure at 28 days. The proposed technique is simple and eco-friendly, without complex requirements conditions or specialized equipment, making it easily applicable in practical engineering.

# Acknowledgements

This study was funded by the National Natural Science Foundation of China (52008003), Outstanding Youth Project of Natural Science Research in Universities of Anhui Province (23AH030043), National Key Research and Development Plan (2021YFB3401504), Graduate Innovation Fund of AUST (2023cx2043).

## Authorship contribution statement

Zijian Zong: Conceptualization, Data cleansing, Formal analysis, Research, Visualization, Writing -original draft.

**Pei Yuan Chen:** Conceptualization, Methodology, Supervision, Resources, Writing - review & editing. **Xinliang Qian:** Formal analysis, Methodology, Supervision.

Jin Li: Formal analysis, Supervision.

Hu Fang: Formal analysis, Validation.

Lei Wang: Formal analysis, Validation.

# **Declaration of competing interest**

The authors of this article declare that they have no financial, professional or personal conflicts of in-terest that could have inappropriately influenced this work.

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