

Article

Effect of Calcium Aluminate and Carbide Slag on Mechanical Property and Hydration Mechanism of Supersulfated Cement

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Abstract: Supersulfated cement (SSC), a low-carbon, energy-efficient, eco-friendly cementitious material, is mainly made from industrial byproducts. However, SSC's slow early strength development leads to inadequate initial hardening and reduced durability, which restricts its practical application. This study investigated the potential enhancement of SSC by incorporating calcium aluminate (CA) and carbide slag (CS) alongside anhydrite as activators to address its slow early strength development. The effects of varying CA and CS proportions on the mechanical property and hydration mechanism of CA-CS-SSC were examined. Results indicate that employing 1% CA and 4% CS as alkaline activators effectively activates slag hydration in the 1CA-4CS-SSC, achieving a compressive strength of 9.7 MPa at 1 day. Despite the limited improvement in early compressive strength of other mixtures with higher CA and lower CS proportions in the CA-CS-SSC system, all mixtures exhibited enhanced compressive strength during long-term hydration. After 90 days, ettringite formation in the CA-CS-SSC system decelerated, whereas anhydrite remained. Concurrently, the formation of C-S-H continued to increase, promoting late compressive strength. The mechanism for enhancing the early compressive strength of the CA-CS-SSC system is attributed to the swift hydration of CA with anhydrite, dissolution of fine slag particles, and reaction with anhydrite under conditions with suitable alkali content to augment the ettringite production. This process also generates a C-S-H and OH-hydratolite to fill the void in the skeleton structure formed by ettringite, resulting in a dense microstructure that improves early compressive strength.

Keywords: supersulfated cement; calcium aluminate; carbide slag; ettringite; C-S-H



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1. Introduction

Ordinary Portland cement (OPC) is a widely used binding material in global infrastructure construction, primarily owing to its good bonding performance, reasonable strength development, and ease of use from local materials. In 2022, the global production of OPC reached 4.10 billion tonnes [1]. However, the production of OPC is associated with substantial carbon dioxide emissions. Statistics indicate that approximately 800–900 kg of carbon dioxide is released per tonne of OPC clinker [2,3]. Therefore, reducing carbon dioxide emissions during OPC production has become an urgent research topic [4,5]. One alternative method to expedite the low-carbon development of the cement industry involves the development of low-carbon cementitious materials.

Supersulfated cement (SSC) has emerged as a promising low-carbon cementitious material, comprising 70–80% granulated blast-furnace slag, 5–20% gypsum or anhydrite as the sulfate activator, and less than 5% alkali activator [6–10]. The slag, composed of CaO, SiO₂, Al₂O₃, and MgO, displays potential hydration activity and can be activated by alkaline substances and calcium sulfate. Compared with OPC, SSC exhibits superior performance attributes, encompassing low heat of hydration, elevated late strength, diminished permeability, and resistance to sulfate attack [11–15]. Furthermore, SSC has

the advantages of simple production, cost-effectiveness, and low energy consumption, significantly mitigating the environmental burden associated with resource and energy expenditure [16,17]. Consequently, SSC holds promise as a viable alternative to OPC in the construction sector from both the environmental and economic standpoints.

The low early strength of SSC is linked to the low dissolution rate of the slag, resulting in diminished production of hydration products [9,18]. While increasing the amount of OPC may increase the early strength of SSC, it negatively affects its subsequent compressive strength [19,20], possibly owing to the preferential formation of ettringite [20] or OH-hydrotalcite [21] on the slag surface in SSC with a high OPC content, obstructing further slag dissolution. An alkaline compound, such as KOH, was incorporated into SSC to enhance its early strength [20,22]. SSCs with a low-content KOH activator exhibited lower hydration kinetics, and their early strength development was also relatively slow [23]. To augment the early compressive strength of SSC, researchers have investigated the utilization of sodium lactate, suggesting that its chelation on the slag surface could accelerate its dissolution rate [10,24,25]. Integrating a high-belite calcium sulfoaluminate cement (HB-CSA) clinker into SSC has been shown to improve its early strength by accelerating the formation of early ettringite [26]. However, attaining an optimal pH range during the hydration of SSC is crucial for promoting the production of both the ettringite and C-S-H [27,28]. Researchers have proposed enhancing the early strength development of SSC by increasing the hardening temperature [29]; nonetheless, this method has shortcomings in terms of economic viability. Moreover, the hydration products within the SSC matrix, encompassing ettringite and C-S-H, become unstable with increasing temperature. Therefore, investigating more sustainable and cost-effective strategies to enhance the early performance of SSC-based materials is paramount.

Carbide slag (CS), a byproduct of the chlor-alkali industry, comprises over 80% $\text{Ca}(\text{OH})_2$ as a dry residue. In China, the annual discharge of CS surpasses 50 million tons, amounting to over 100 million tons of alkaline solid waste [30]. Owing to a lack of viable industrial applications, CS is frequently relegated to landfills, causing serious environmental pollution [31]. To broaden the applications of CS, it can be incorporated into SSC to replace Portland cement, thereby catalyzing slag dissolution [32]. This strategy reveals an alternative pathway to increase the application prospects of CS and attenuate its adverse environmental repercussions.

The deficiency in the early strength development of SSC has led to negative impacts on matrix hardening and durability, significantly limiting its widespread application in the construction industry. Addressing this issue, this study is dedicated to an in-depth analysis of the performance of SSC. It aims to overcome the challenge of the low early strength of SSC by optimizing the mix proportions of new alkaline activators to promote the rapid development of early strength, which is crucial for its replacement of OPC and promoting the development of construction materials towards a green and sustainable direction. A novel synergistic activation method is devised to rapidly promote the hydration of slag for SSC to enhance its early strength while effectively harnessing industrial CS to curtail production costs and carbon emissions and achieve energy conservation and emission reduction. In this study, calcium aluminate (CA) powder was introduced as an alkaline activator into SSC for the first time, exploiting its rapid hydration to generate ettringite in the presence of calcium sulfate. Additionally, synergistic excitation technology was employed to scrutinize the effects of CA and CS as alkali activators (the masses of both constituents were 5% of the total mass) on the mechanical property and hydration mechanism of the CA-CS-SSC system, with 15% native anhydrite utilized as a sulfate activator in the preparation of the CA-CS-SSC system. The resultant CA-CS-SSC system could attain high early and late compressive strength. The hydration kinetics of the CA-CS-SSC pastes were ascertained using isothermal calorimetry, while the evolution of the reactant phases and hydration products was determined via X-ray diffraction (XRD) and thermogravimetric analysis (TGA). The pore structure of the CA-CS-SSC pastes was characterized via mercury intrusion porosimetry (MIP). Thermodynamic modeling was

developed to simulate the hydration mechanism, gaining deeper insights into the evolution of the individual hydration phases in the CA-CS-SSC system over time [26,33].

2. Materials and Methods

2.1. Materials

The CA-CS-SSC encompasses four principal constituents: slag, anhydrite, CA, and CS, wherein CA and CS function as alkali activators, and native anhydrite acts as the sulfate activator. Exhaustive information concerning the chemical compositions and mixing ratios of each component is presented in Tables 1 and 2. The XRD patterns of the CA, CS, anhydrite, and slag are illustrated in Figure 1. The slags employed in this study predominantly comprise vitreous materials, with Figure 1d displaying the amorphous hump characteristic of slag. The specific surface area of the slag is a pivotal factor that determines its reactivity. In this investigation, the slag possessed a specific surface area of 425 m²/kg. The chemical composition of OPC, serving as the reference group for the CA-CS-SSC system, is cataloged in Table 1. The mineral constituents of CA, CS, and anhydrite are enumerated in Table 3.

Table 1. Chemical composition of slag, anhydrite, CS, and CA, (wt.%).

Material	CaO	SiO ₂	Al ₂ O ₃	SO ₃	Fe ₂ O ₃	MgO	TiO ₂	Others	SSA (m ² /kg)
Slag	40.30	30.75	14.98	3.15	0.26	10.22	0.75	0.59	425
Anhydrite	40.69	2.58	0.40	50.65	0.25	2.70	–	2.38	398
CA	46.24	6.61	25.92	10.9	6.52	0.73	1.85	1.17	430
CS	92.61	2.82	0.79	1.29	0.17	1.36	–	0.95	367
OPC	65.13	20.59	4.25	2.75	3.36	1.59	0.52	1.79	349

Table 2. Mix proportions of slag, anhydrite, CS, and CA.

	Mixture Proportions (% by Mass)			
	Slag	Anhydrite	CA	CS
1CA-4CS-SSC	80	15	1	4
2CA-3CS-SSC	80	15	2	3
3CA-2CS-SSC	80	15	3	2
4CA-1CS-SSC	80	15	4	1

Table 3. Mineralogical composition of CA, CS, and anhydrite.

Mineralogical Composition (wt.%)			
ID	CA	CS	Anhydrite
CA ₁	27.54	–	–
CA ₂	36.72	–	–
C ₂ AS	34.56	–	–
CaSO ₄	–	–	89.64
CaSO ₄ ·H ₂ O	–	–	5.74
Ca(OH) ₂	–	83.34	–
CaCO ₃	–	14.61	3.52
SiO ₂	–	2.05	–
Other	1.27	–	1.44

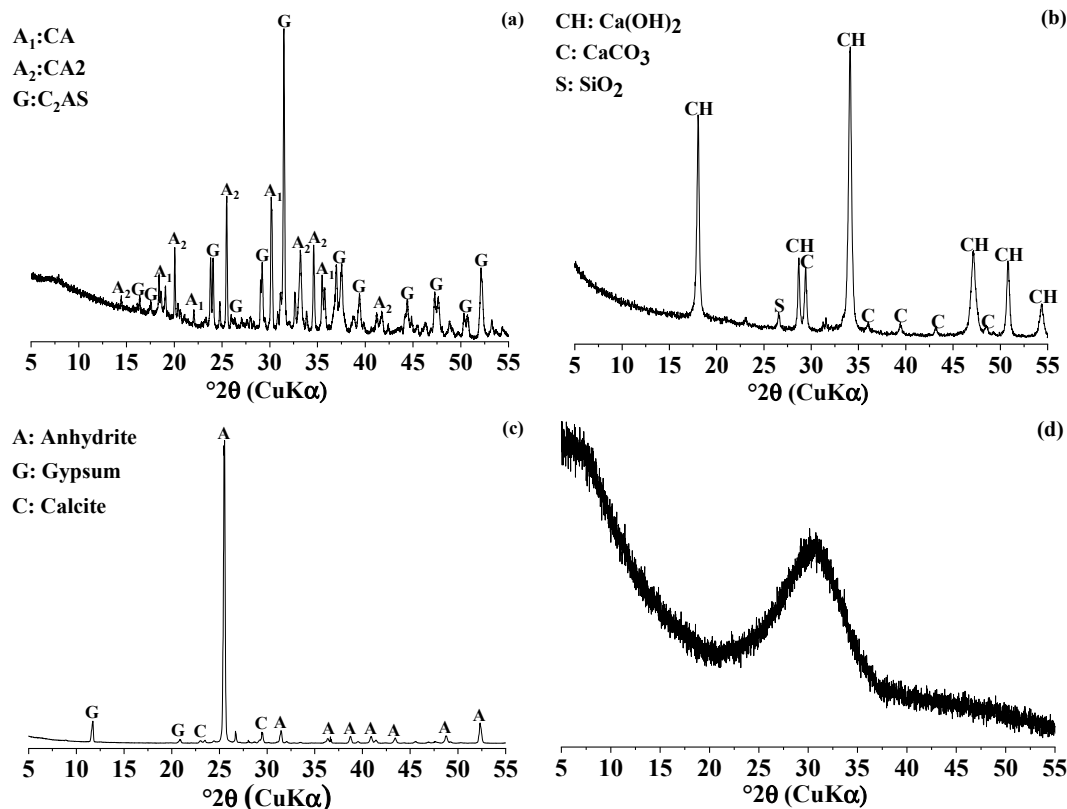


Figure 1. X-ray diffraction pattern of (a) CA, (b) CS, (c) anhydrite, and (d) slag.

2.2. Sample Preparation

Cement pastes were prepared with a water-to-cement ratio of 0.4 and cast into molds measuring 40 mm × 40 mm × 160 mm. After casting, the molds were cured in a chamber maintained at 20 ± 2 °C and 95% relative humidity for 24 h before being demolded. Following demolding, they were cured in water at 20 ± 1 °C until they attained the desired age. To halt hydration, the cured cement specimens were fragmented into roughly 1 g pieces using a hammer and immersed in isopropanol, maintaining a cement-to-isopropanol volume ratio of 1:5.

The termination of the hydration process was executed in two phases. Initially, the crushed samples were submerged in isopropanol for 12 h, followed by transfer into another isopropanol for an additional 48 h immersion. Following this, the samples were dried at 40 °C for 3 days, and a segment of the crushed samples was pulverized into a fine powder sieved through a 63 μm square mesh. The sieved powder samples were promptly sealed and reserved for subsequent TGA and XRD analyses. Block samples were utilized for the porosity examination.

2.3. Analytical Methods

2.3.1. Compressive Strength

The compressive strength of the mortar was ascertained at a constant loading rate of 0.02 mm/s in accordance with ASTM C109 [34]. Mortar specimens were fabricated with a sand-to-binder ratio of 1:3 and a water-to-binder ratio of 0.5, each having dimensions of 40 mm × 40 mm × 40 mm. The compressive strength of the mortar was evaluated at 1, 3, 7, 14, 28, 90, and 180 days to assess the early and late strength development of the CA-CS-SSC system.

2.3.2. Pore Structure Measurement

A mercury porosimeter, specifically the AutoPore IV9500 V1.09 (Micromeritics, Norcross, GA, USA), was employed to analyze the pore structure. This instrument is capable of exerting a maximum pressure of approximately 60,000 psia, and it can measure pore sizes spanning from 3 to 400,000 nm. The sample cell encompassed a volume of 5 cc, and sample sizes were selected between 5–9 mm to determine the pore size and porosity of the hardened cement paste. The contact angle, presumed to be 130°, was established between the mercury and sample surface.

2.3.3. Hydration Heat

A Thermometric TAM Air calorimeter was employed to gauge the hydration heat rate and cumulative heat release of cement. The assays were executed at an ambient temperature of 20 °C, with a cement mass of 4.0 ± 0.05 g and a water-to-cement ratio of 0.4. Within the experiment, 0.8 mL of deionized water was drawn into the needle of a needle-type mixer. Following the drying of the mixer surface, it was affixed to an ampoule bottle and positioned within a calorimeter. Deionized water was injected into the ampoule bottle and subjected to internal mixing for 1 min. The hydration heat tests spanned 168 h.

2.3.4. XRD

Hydration phase analysis of the cement specimens at varying hydration ages was performed using a German Bruker D8 ADVANCE X-ray diffractometer at room temperature. The diffractometer deployed CuK α radiation ($\lambda = 1.5405$ Å) with a tube current and voltage of 40 mA and 40 kV, respectively. The scanning range was 5–55°, with a step size of 0.0102 and a counting time of 0.2 s. Each scan lasted approximately 20 min. The crystal structures of the identified crystalline phases were retrieved from the inorganic crystal structure database (ICSD). An internal standard method was employed to compute the amorphous content of the paste sample, which was blended with a 20% ZnO internal standard.

The quantities of the principal crystalline and slag phases within the samples were ascertained via Rietveld analysis using the Highscore Plus 4.9 software. For a quantitative analysis of the amorphous materials in slag, PONKCS phase fitting was applied to the amorphous segment, where the pseudo-phase was determined using partial structural information with the “hkl phase”. The amorphous morphology of the slag was explored by hump fitting the (hkl) phase in the precursor, which was further refined employing free lattice parameters, scale factors, profile parameters, and phase positions. A homogeneous mixture of ZnO and slag was generated during agate grinding to accurately model the X-ray scattering contribution of the slag. ZnO was utilized to substitute the slag in proportions of 20%, 30%, and 50%. Reliable fitting results could be attained by setting the background parameter of the PONKCS phase to a first-order polynomial background with a 1/2 θ parameter. The raw materials and hydrated samples were analyzed using the method described in [35].

2.3.5. TGA

The powder specimens were examined via TGA using a Mettler-Toledo TGA/DSC1 simultaneous thermal analyzer. Before the test, 20 ± 0.3 mg of the sample were weighed, placed in a 30- μ L aluminum oxide crucible, covered, and stationed on the balance support of the instrument. Nitrogen gas served as a protective gas throughout the TGA test, with a heating rate of 10 °C/min and a heating temperature range spanning from 30 to 800 °C.

2.3.6. Thermodynamic Modeling

The GEM-Selector program, widely utilized for geochemical modeling, facilitates calculating the combination and morphology of equilibrium phases based on the elemental composition of complex chemical systems [36,37]. Amid the hydration process of the CA-CS-SSC system, the solid-phase components were explored using Gibbs Energy Minimization software (GEMS-PSI 3.5) [38,39] combined with the cement database CEMDATA

18.01 [40]. Based on the input initial raw-material composition and thermodynamic data of the simulated phase, thermodynamic modeling can predict the solid-phase assemblage of the system at equilibrium [33]. Within this study, the CSH3T model was deployed to elucidate the C-S-H gel formation. The extant literature signifies that amorphous aluminum hydroxide ($\text{Al}(\text{OH})_3(\text{am})$) can be discerned in SSC stimulated by high-belite calcium sulfoaluminate cement (HB-CSA) clinker [26]. Furthermore, data concerning $\text{Al}(\text{OH})_3(\text{am})$ were included in the study.

Based on the thermodynamic modeling of CA-CS-SSC system hydration, the following assumptions were posited:

1. A thermodynamic equilibrium exists between the precipitated hydration products and the pore water solution;
2. According to the 5% (CA + CS) alkaline activators and 15% natural anhydrite, the active mineral components specified in Table 3 were designated as the input, while the quantitative results of the slag reaction percentage based on XRD-Rietveld-PONKCS refinement were used as input;
3. The mass alteration of the hydration products was computed as a function of the hydration reaction time.

3. Results and Discussion

3.1. Compressive Strength

The compressive strength of the CA-CS-SSC system mortars at intervals of 1, 3, 7, 14, 28, 90, and 180 days are depicted in Figure 2. In the study of the compressive strength of CA-CS-SSC system mortars, it was observed that their compressive strength gradually increased throughout the hydration process. Notably, after 3 days of hydration, the compressive strength of the CA-CS-SSC system mortars exhibited a significant enhancement, surpassing that of the OPC mortar. The rapid improvement in the compressive strength of the CA-CS-SSC system mortars after 3 days of hydration was primarily attributed to the alkaline environment provided by CS, which promoted the rapid hydration of slag and generated a large amount of hydration products, making the matrix denser.

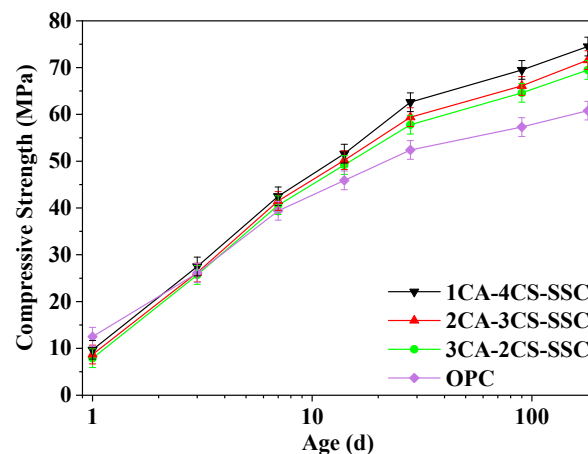


Figure 2. Compressive strength development of the CA-CS-SSC system mortars at 1, 3, 7, 14, 28, 90, and 180 days.

The experimental results demonstrated that the 1CA-4CS-SSC mortar, prepared using 1% CA and 4% CS as alkaline activators, exhibited superior compressive strength at all the ages tested. Specifically, the compressive strength of the 1CA-4CS-SSC mortar reached 9.7 MPa after 1 day of hydration, slightly lower than that of OPC mortar. The compressive strength of 1CA-4CS-SSC mortar achieved an increase of 84.6% from 3 to 7 days of hydration, reaching 42.5 MPa at 7 days. By 14 days of hydration, the compressive strength of the 1CA-4CS-SSC mortar further increased to 51.6 MPa, surpassing that of the OPC mortar by 12.6%. Moreover, from 14 to 28 days of hydration, the compressive strength of the

1CA-4CS-SSC mortar increased by 21.3%, achieving 62.6 MPa, which was 19.5% higher than that of the OPC mortar. These results demonstrated the effectiveness and optimal synergistic action of 1% CA and 4% CS as alkaline activators.

In the CA-CS-SSC system mortars, the enhancement of early compressive strength is primarily achieved by adjusting the proportions of CA to CS used as alkaline activators, promoting synergistic control over the formation of hydration products and pore structure in the hardened matrix. The synergistic action between CA and CS is primarily realized through the rapid hydration of CA with anhydrite to form ettringite, while CS provides an alkaline environment for the dissolution of fine slag particles, accelerating the reaction between fine slag particles and anhydrite to form ettringite, while concurrently generating C-S-H and OH-hydratocalcite. These hydration products fill the pore within the skeletal structure formed by ettringite, thereby resulting in a denser microstructure. This dense microstructure is critical to developing early compressive strength in the CA-CS-SSC system mortars.

The compressive strength of the CA-CS-SSC system mortars also surpassed that of the OPC mortar in the late hydration stage. In the late stage, the growth rate of the compressive strength of the CA-CS-SSC system mortars gradually decelerated from 28 to 180 days, influenced by the hydration process and the diffusion coefficient. Notably, the growth rate of the compressive strength of the 1CA-4CS-SSC mortar from 28 to 180 days of hydration decreased to 15.9%. Throughout the hydration period, spanning from 28 to 90 days, the compressive strength of the 1CA-4CS-SSC, 2CA-3CS-SSC, 3CA-2CS-SSC, and 4CA-1CS-SSC mortars underwent increments of 11.1%, 11.3%, 11.8%, and 13.1%, respectively. Conversely, the compressive strength of the OPC mortar increased by 9.4% over the identical test interval (28–90 days), culminating at 57.1 MPa. From 90 to 180 days of hydration, the compressive strength of the 1CA-4CS-SSC mortar increased by 7.2%, achieving 74.5 MPa. The late compressive strength growth of the CA-CS-SSC system mortars is predominantly influenced by the amount of C-S-H generated, which is consistent with previous research findings [18,26]. Studies have elucidated that, in lower alkaline environments, the late strength growth of SSC dwindles primarily owing to the sluggish diffusion of silicate ions [41]. A layer enriched in silicon and aluminum forms around the dissolved slag particles, causing C-S-H to precipitate from around the slag, which in turn retards the diffusion rate of free water to the slag surface [41], culminating in a reduction in slag dissolution. Nevertheless, the compressive strength of the CA-CS-SSC system mortars did not increase with an increase in the amount of CA. Hence, the proportions and dosages of CA and CS within the CA-CS-SSC system matrix need to be matched to synergistically stimulate paste hydration and concurrently enhance the mechanical attributes of the mortars.

3.2. Microstructure Analysis

The porosity and pore size distributions of the CA-CS-SSC system pastes post-hydration for 1, 28, and 180 days are illustrated in Figure 3, while the pore volume fractions of the CA-CS-SSC system pastes are depicted in Figure 4. A bimodal pore size distribution can be observed for the CA-CS-SSC system pastes. In the CA-CS-SSC system pastes, at 1 day of hydration, 1CA-4CS-SSC paste demonstrated the lowest porosity (37.02%), and its compressive strength was the highest. Further analysis revealed that the volume fraction of a pore size smaller than 20 nm increased as the CA content increased during the early stage of hydration (1 day) in the CA-CS-SSC system pastes. Increasing the CA content in the CA-CS-SSC system pastes enhanced the amount of ettringite generated by its reaction with anhydrite, thereby increasing the volume fraction of small pores. Nevertheless, the content of CS in the alkaline activators decreased concomitantly, diminishing the alkaline environment provided for slag hydration. This reduction weakened the hydration of the slag, consequently affecting the decrease in the total amount of ettringite formed at the early stage (within 1 day) (as shown in Figure 7). Moreover, the porosity and compressive strength of the CA-CS-SSC system did not show a corresponding increasing trend with the addition of CA content.

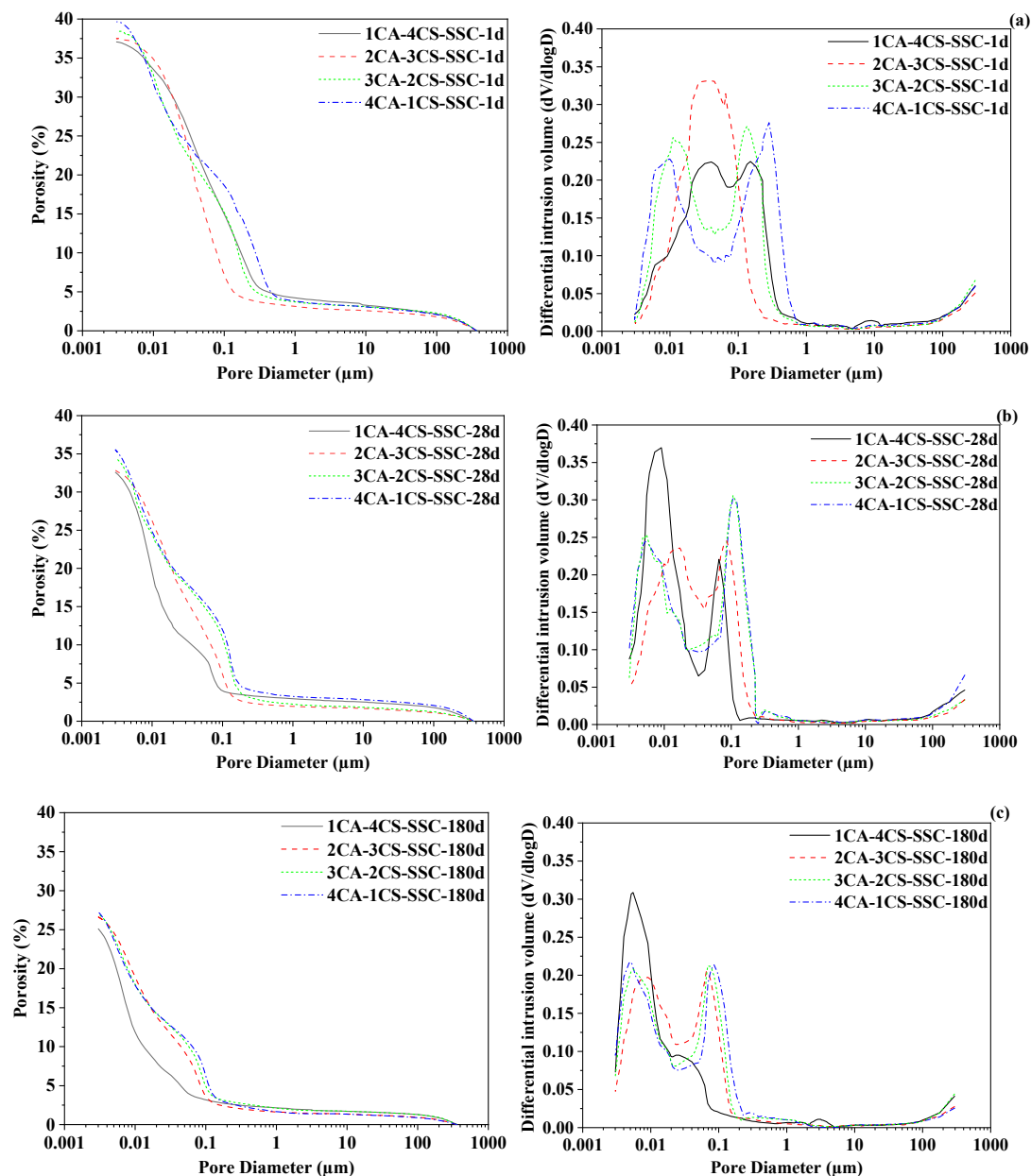


Figure 3. Porosity and differential intrusion curve of the CA-CS-SSC system pastes at (a) 1 day, (b) 28 days, and (c) 180 days.

With the progression of hydration, the pore size distribution in the CA-CS-SSC paste system gradually transitioned towards smaller pores. Specifically, at 28 days of hydration, the volume fraction of pore size smaller than 20 nm in the 1CA-4CS-SSC paste reached 63.5%, indicating that pore size ranging from 20 to 5000 nm progressively transformed into pore size smaller than 20 nm. Moreover, from 1 to 28 days of hydration, the porosity of 1CA-4CS-SSC, 2CA-3CS-SSC, 3CA-2CS-SSC, and 4CA-1CS-SSC pastes decreased by 13.83%, 14.25%, 11.45%, and 11.67%, respectively. From 28 to 180 days of hydration, the porosity of 1CA-4CS-SSC, 2CA-3CS-SSC, 3CA-2CS-SSC, and 4CA-1CS-SSC pastes further decreased by 29.73%, 23.25%, 27.50%, and 30.27% respectively, with the 1CA-4CS-SSC paste achieving the lowest porosity at 25.10%. This change reflects a synergistic interaction between CA and CS, whereby the rapid reaction of CA with anhydrite to form ettringite, along with the reaction of small slag particles dissolved from CS with anhydrite forms ettringite. Concurrently, the formation of C-S-H and OH-hydrotalcite served to fill the

pores within the skeletal structure created by ettringite formation, thereby optimizing the microstructure of the paste and enhancing its early-stage density.

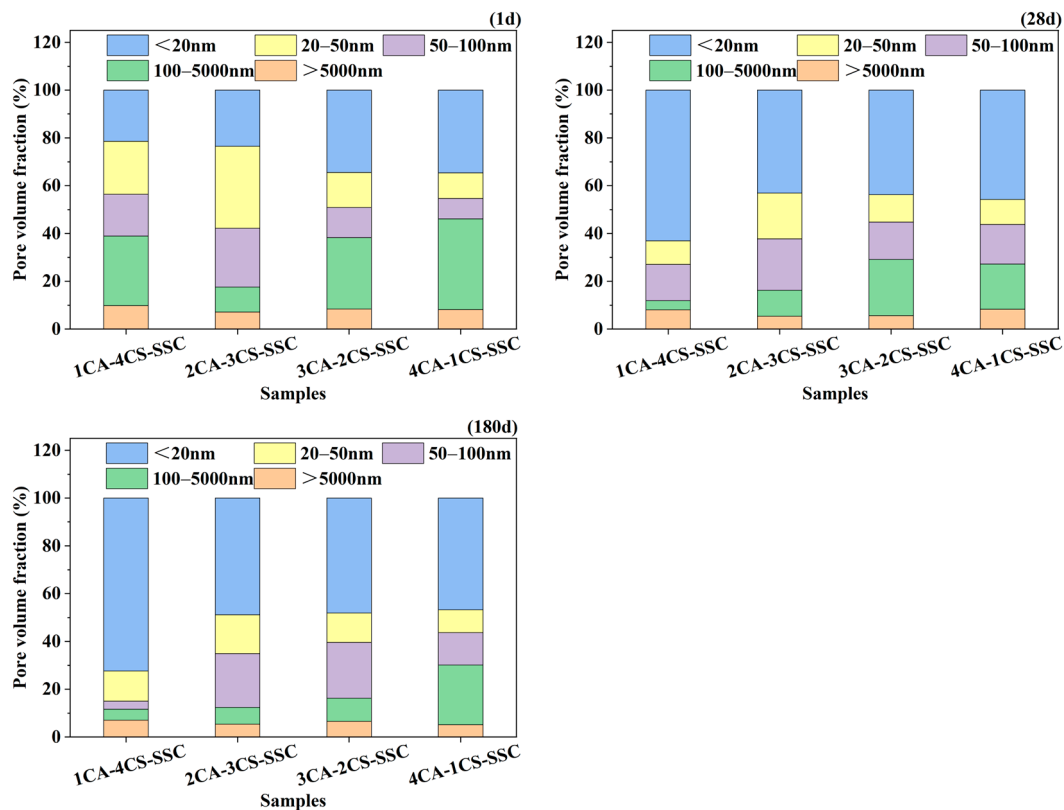


Figure 4. Pore volume fraction of the CA-CS-SSC system pastes at 1 day, 28 days, and 180 days.

At 180 days of hydration, pore size smaller than 20 nm in the 1CA-4CS-SSC paste accounted for 73.3% of the total pore volume. Nonetheless, as age increased, the volume of macropores larger than 5000 nm in the 1CA-4CS-SSC paste remained unchanged. This result was attributed to the rapid hydration of CA with anhydrite to form ettringite while enhancing the early compressive strength of the CA-CS-SSC system, causing expansion of the matrix due to ettringite formation, leading to an increase in some of the larger pores. The hydration products formed from the late hydration of slag were insufficient to fill these macropores. As the age increases, the volume fraction of pore size smaller than 20 nm in the 2CA-3CS-SSC, 3CA-2CS-SSC, and 4CA-1CS-SSC pastes increased at a modest pace, and their respective compressive strengths evolved relatively slowly. CA and CS, serving as alkaline activators, primarily functioned to regulate the quantity and proportion of hydration products formed, thereby optimizing the microstructure of the matrix—including enhancing density and reducing porosity. Compared to merely increasing the dosage of alkaline compounds, this synergistic interaction showed a more significant effect in reducing porosity, enhancing the formation of hydration products, and improving the early strength of the mortar.

3.3. Heat Hydration

The effects of CA and CS as alkaline activators on the hydration kinetics of the CA-CS-SSC system pastes were investigated by evaluating the hydration heat release rate and accumulated heat, as delineated in Figure 5. Throughout this 168.0 h hydration process, the CA-CS-SSC system pastes manifested three hydration exothermic peaks. The first peak may be attributed to the dissolution and wetting of the reactant phase in the pastes, while the second and third peaks emerge between 3.5–18.0 h and 18.0–48.0 h, respectively. Notably, despite adjustments in the mix proportions of alkaline activators, the induction-

period duration remained constant across all CA-CS-SSC system pastes, with an induction duration of 2.0 h (1.5–3.5 h). The CA-CS-SSC system pastes display a truncated acceleration period of the second main peak, roughly 3.0–5.0 h (3.3–10.0 h). This is predominantly ascribed to the CA_1 and CA_2 in CA, which interacted with anhydrite, enabling swift early hydration and brief exothermic concentrations and durations. The second exothermic peak of hydration in the 1CA-4CS-SSC paste exhibited a faster hydration rate compared to 2CA-3CS-SSC, 3CA-2CS-SSC, and 4CA-1CS-SSC, with the acceleration period lasting for 3.0 h (3.3–6.3 h), which is consistent with its early compressive strength development. All CA-CS-SSC system pastes entered a hydration deceleration phase after 6.0–10.0 h of hydration, and with the increase in CA addition and decrease in CS addition, the second main peak of the pastes gradually delayed and broadened, and the slope of the hydration curve deceleration phase gradually decreased. This implies a gradual reduction in the dissolution rate of slag and a decrease in the rate at which aluminates from slag and anhydrite form ettringite. Post 20.0–48.0 h of hydration, all pastes within the CA-CS-SSC system pastes exhibited a third hydration exothermic peak (second aluminate peak), likely originating from the reaction of aluminum in the slag with sulfate, culminating in an ettringite precipitate [42]. The third peak of the 1CA-4CS-SSC paste is more exothermic, owing to its elevated hydration level. Quantitative XRD analysis revealed that the CA-CS-SSC system pastes generated a large amount of ettringite and C-S-H at 1 day of hydration. Notably, the 1CA-4CS-SSC paste with a mix of 1% CA and 4% CS as alkaline activators exhibited the most production of the hydration products.

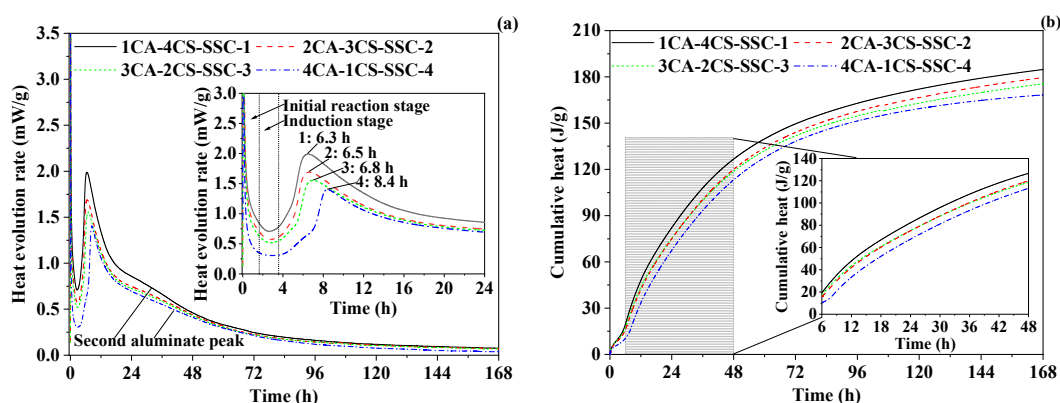


Figure 5. Measured hydration heat: (a) heat evolution rate; (b) cumulative heat.

A rapid increase in the amount of accumulated heat was observed throughout the hydration trajectory of the CA-CS-SSC system pastes during the initial 48 h, after which the growth rate gradually decreased. The heat release rate and accumulated heat of the CA-CS-SSC system pastes progressively diminished with an increase in the added CA content and a decrease in the added CS content. The heat release rate and cumulative heat of the 1CA-4CS-SSC pastes surpassed those of the other mixtures, and the mechanical properties exhibited a more rapid improvement. The study demonstrated that the 1CA-4CS-SSC paste, containing 1% CA and 4% CS as alkaline activators, exhibited the highest cumulative heat release during the hydration process. The cumulative heat released during early hydration, to some extent, reflected the degree of slag hydration. Specifically, at a hydration duration of 72 h, the accumulated heat for the 1CA-4CS-SSC, 2CA-3CS-SSC, 3CA-2CS-SSC, and 4CA-1CS-SSC pastes were recorded as 149, 143, 141, and 138 J/g, respectively. It was estimated that, after 168 h of hydration, the accumulated heat for the 1CA-4CS-SSC, 2CA-3CS-SSC, 3CA-2CS-SSC, and 4CA-1CS-SSC pastes was 184, 179, 175, and 168 J/g, respectively. An additional advantage of the CA-CS-SSC system pastes is that, at both 72 and 168 h of hydration, the accumulated heat is lower than the stipulated requisites for No. 42.5 low-heat Portland cement as delineated in the Chinese standard GB/T 200-2017 [43]. This standard articulates that the cumulative heat of cement hydration at the 72 h and 168 h hydration intervals should not surpass 230 and 260 J/g, respectively.

3.4. XRD

Per the XRD patterns of the CA-CS-SSC system pastes at hydration ages of 1, 3, 28, 90, and 180 days, as illustrated in Figure 6 and in conjunction with the XRD quantitative outcomes in Figure 7, it can be detected that the main crystalline hydration product is ettringite, whose formation predominantly depended on the reaction between CA and anhydrite and the dissolution of slag, as well as the hydration between slag and anhydrite, consistent with previous research findings [8,9]. In the XRD patterns of the CA-CS-SSC system pastes, an amorphous is discernible as a hump at approximately 30° in 2θ , which mainly consists of unhydrated slag, C-S-H, OH-hydrotalcite, and $\text{Al}(\text{OH})_3(\text{am})$. During the early stage of hydration in the CA-CS-SSC system, an augmentation in the CA content contributed to a marginal increase in ettringite production, originating from the reaction between CA_1 and CA_2 in CA with anhydrite. The thermodynamic modeling computations suggest that the quantities of ettringite produced by the reaction of CA_1 and CA_2 in CA with anhydrite in the 1CA-4CS-SSC, 2CA-3CS-SSC, 3CA-2CS-SSC, and 4CA-1CS-SSC pastes are 1.31, 2.62, 3.93, and 5.25 g, respectively. By merging insights from the XRD quantitative data and thermodynamic modeling simulation results, it can be concluded that the formation of ettringite in the 1CA-4CS-SSC paste primarily originates from the swift hydration of the fine-grained slag and its reaction with anhydrite. Moreover, the quantitative XRD results affirmed that, at 1 day of hydration, the 1CA-4CS-SSC paste exhibited superior slag hydration and amorphous formation compared to the other mixtures. After 1 day of hydration, the characteristic peaks of CA_1 and CA_2 vanished in the CA-CS-SSC system pastes, indicating their complete hydration, with concurrent ettringite formation.

Quantitative XRD results confirmed that at 1 day of hydration, the 1CA-4CS-SSC paste exhibited a greater amount of ettringite formation compared to the 2CA-3CS-SSC, 3CA-2CS-SSC, and 4CA-1CS-SSC pastes, reaching 18.99%. From 1 to 3 days of hydration, the amount of ettringite formed in the 1CA-4CS-SSC, 2CA-3CS-SSC, 3CA-2CS-SSC, and 4CA-1CS-SSC pastes increased by 23.59%, 22.36%, 20.71%, and 20.20%, respectively, indicating a rapid growth trend in ettringite formation. This growth trend reflected the formation of ettringite facilitated by the hydration of slag with anhydrite. As the content of anhydrite gradually decreased, the growth rate of ettringite formation decreased. By 28 days of hydration, the amount of ettringite formed in the 1CA-4CS-SSC paste showed the highest value compared to the 2CA-3CS-SSC, 3CA-2CS-SSC, and 4CA-1CS-SSC pastes, at 26.38%. However, from 28 to 90 days of hydration, the main hydration products in the 1CA-4CS-SSC, 2CA-3CS-SSC, 3CA-2CS-SSC, and 4CA-1CS-SSC pastes gradually became amorphous hydration products (mainly C-S-H, OH-hydrotalcite, and $\text{Al}(\text{OH})_3(\text{am})$), and their formation quantities progressively increased, with the amorphous hydration products increasing by 12.15%, 16.27%, 13.40%, and 18.23%, respectively. From 90 to 180 days of hydration, as the anhydrite was consumed, the growth rate of ettringite formation in all pastes decreased, with the amount of ettringite formation in the 1CA-4CS-SSC, 2CA-3CS-SSC, 3CA-2CS-SSC, and 4CA-1CS-SSC pastes only increasing by 1.20%, 1.96%, 3.17%, and 6.10%, respectively, but the amount of amorphous hydration products increased by 11.97%, 7.02%, 8.84%, and 10.37%, respectively. TGA results also confirmed that, after 28 days of hydration, the main hydration products in the CA-CS-SSC system pastes gradually transitioned from ettringite to amorphous hydration products, significantly increasing the amount of C-S-H. No characteristic peak of $\text{Ca}(\text{OH})_2$ was discerned in the XRD pattern of the CA-CS-SSC system pastes after 90 days of hydration, suggesting a complete reaction of $\text{Ca}(\text{OH})_2$. Given that amorphous phases like the C-S-H, OH-hydrotalcite, and $\text{Al}(\text{OH})_3(\text{am})$ could not be analyzed via XRD, they were characterized via TGA.

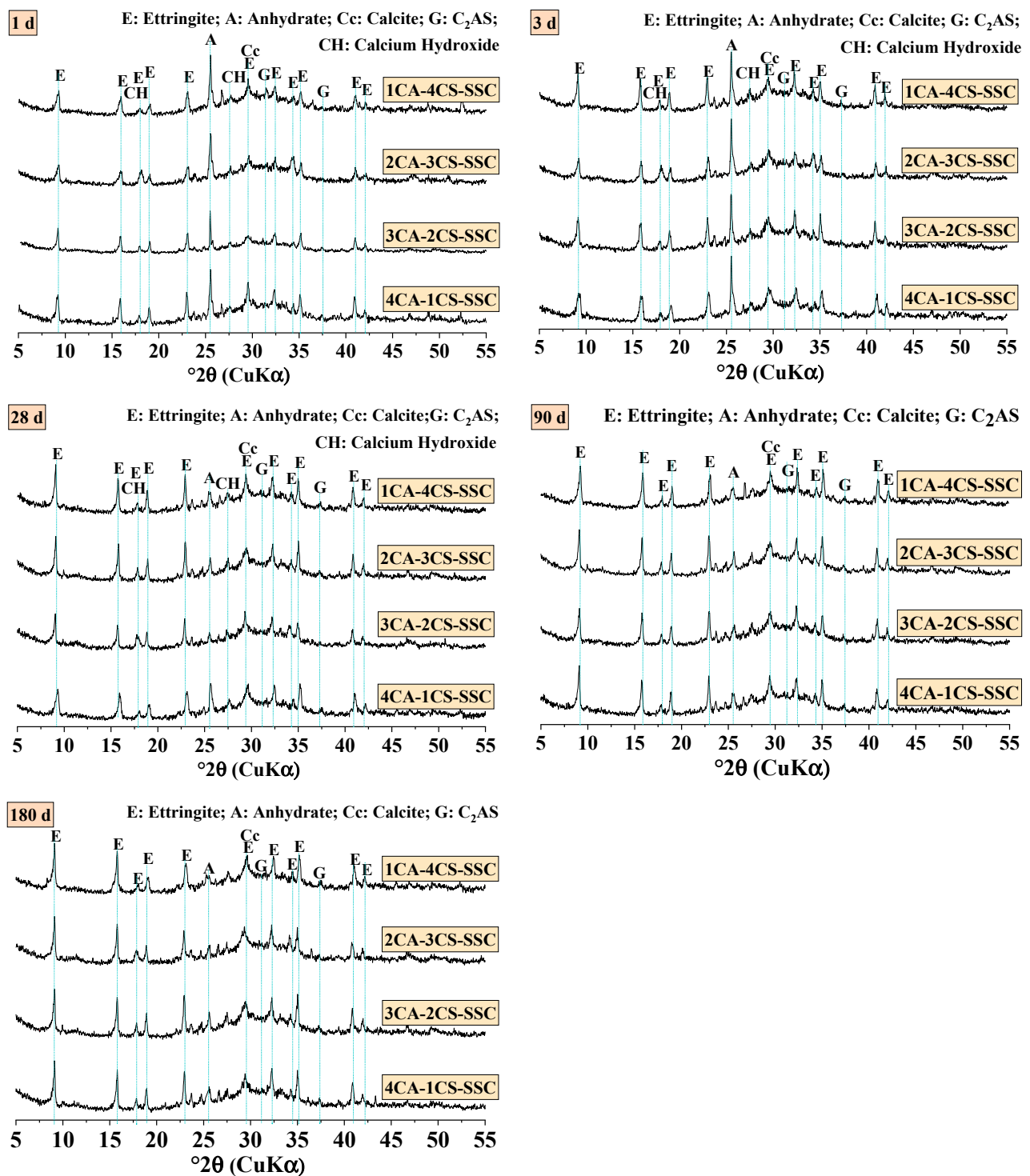


Figure 6. XRD patterns of the CA-CS-SSC system pastes at 1 day, 3 days, 28 days, 90 days, and 180 days.

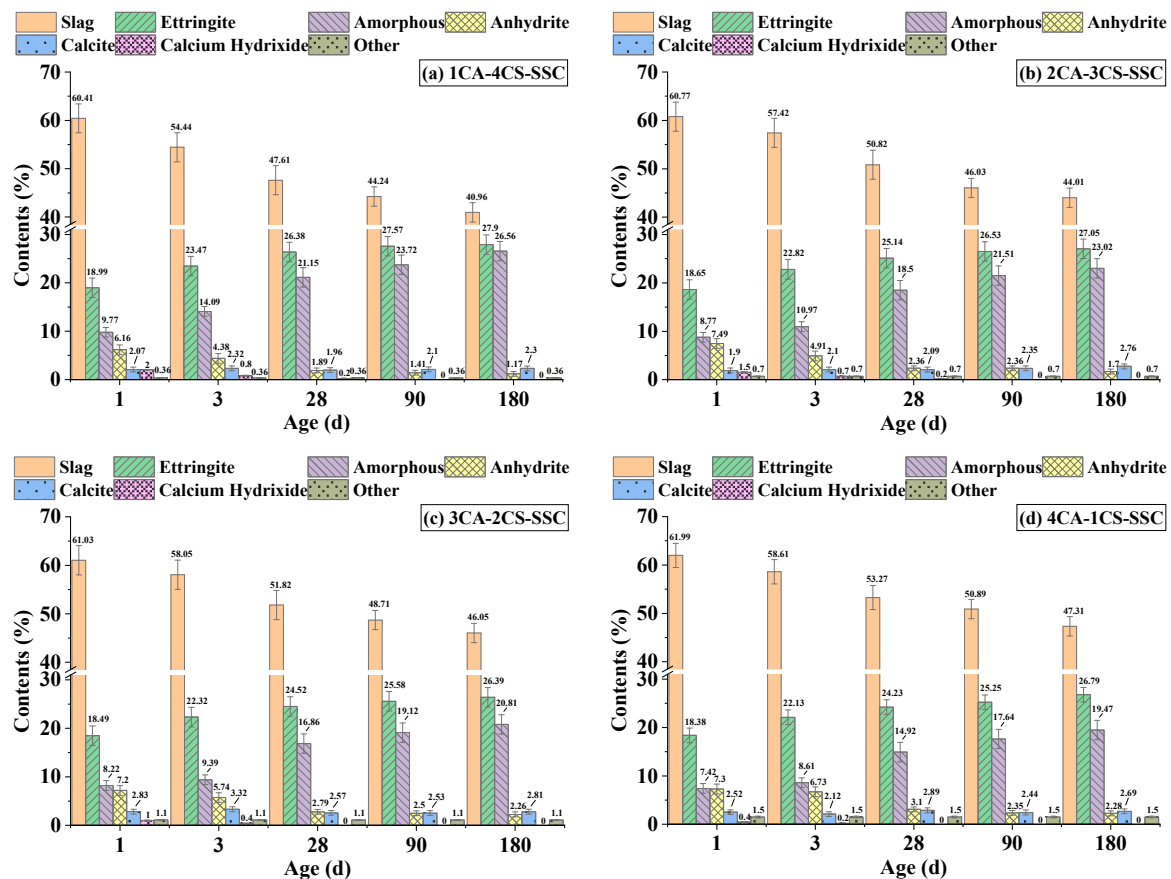


Figure 7. Phase evolution of (a) 1CA-4CS-SSC paste, (b) 2CA-3CS-SSC paste, (c) 3CA-2CS-SSC paste, and (d) 4CA-1CS-SSC paste for up to 180 days.

3.5. TGA

TGA and differential thermogravimetry (DTG) were conducted on the CA-CS-SSC system pastes following 1, 3, 28, 90, and 180 days of hydration, as depicted in Figure 8. Additionally, Figure 9 displays the non-evaporated water content within the 30–600 °C temperature range at 1, 3, 28, 90, and 180 days of hydration. An appreciable endothermic peak is evident around 120 °C, attributed to non-evaporative water loss by ettringite and C-S-H. The experimental results indicate that the main hydration products of the CA-CS-SSC system pastes were ettringite (70–450 °C) and C-S-H (50–600 °C). The dehydration curves for ettringite and C-S-H overlap with those of OH-hydratolite within the temperature range of 300–400 °C. The CA-CS-SSC system pastes generated a substantial quantity of ettringite following 1 day of hydration, with no detection of gypsum. Between temperatures of 30 and 300 °C, the mass loss of the CA-CS-SSC system pastes at 1 day of hydration ranged from 7.381 to 10.339%, while at 28 days of hydration, the mass loss increased to 11.031–12.480%. After 1 day of hydration, the XRD quantitative outcomes revealed that the amount of ettringite produced by all pastes was comparable, though the extent of amorphous formation varied significantly. Concurrently, TG analysis also revealed that the 1CA-4CS-SSC paste underwent the highest mass loss among the other mixtures within the temperature range of 30 and 500 °C. At 1 day of hydration, based on the XRD quantitative results, the TG analysis indicated that, between 30 and 500 °C, the disparity in mass loss among mixtures in the CA-CS-SSC system pastes was primarily reflected in the amount of amorphous formation. This result aligns with the quantitative XRD result and reveals a close relationship between the improved early compressive strength of the CA-CS-SSC system mortars and the amount of amorphous hydration products formed. With increased CA content in the CA-CS-SSC system pastes, the amount of amorphous hydration products gradually decreased, suggesting that the hydration of slag might not have been sufficiently

activated in the CA-CS-SSC system pastes with a high CA content. Furthermore, from 1 to 28 days of hydration in the CA-CS-SSC system pastes, the minor weight loss peak of $\text{Ca}(\text{OH})_2$ was observed around 450 °C, indicating that the slag has been dissolved and a hydration reaction has occurred to generate hydration products. The elevated mass loss of the 1CA-4CS-SSC paste affirmed its high degree of hydration and demonstrated that 1% CA and 4% CS, acting as alkaline activators, were instrumental in promoting the formation of hydration products.

According to the DTG pattern of the CA-CS-SSC system pastes, as the curing age advanced, the dehydration weight loss peak of ettringite incrementally increased, suggesting an increased production. Within the initial 28 days of hydration in the CA-CS-SSC system pastes, the weight loss peak of ettringite overlapped with that of C-S-H, and due to the substantial formation of ettringite during this phase, the weight loss peak of ettringite rapidly increased, while that of C-S-H rose more slowly. Moreover, given that the bound water content of C-S-H is relatively lower than that of ettringite, the weight loss peak of C-S-H was insignificant during the initial 28 days of hydration. However, at 90 days of hydration, the weight loss peak of C-S-H significantly increased in the DTG patterns of the CA-CS-SSC system pastes, indicating a substantial generation of C-S-H in the late stage of hydration. In contrast, the weight loss peak of ettringite at 90 days of hydration slightly increased compared to that at 28 days of hydration, further confirming that the production of ettringite was still slowly increasing in the late stage of hydration. These results suggest that, in the CA-CS-SSC system pastes, with the extension of hydration time, the formation of ettringite and C-S-H exhibits distinct dynamic changes, particularly with the rapid increase of ettringite formation dominating at the early stage of hydration, while the substantial generation of C-S-H became a significant characteristic at the late stage of hydration.

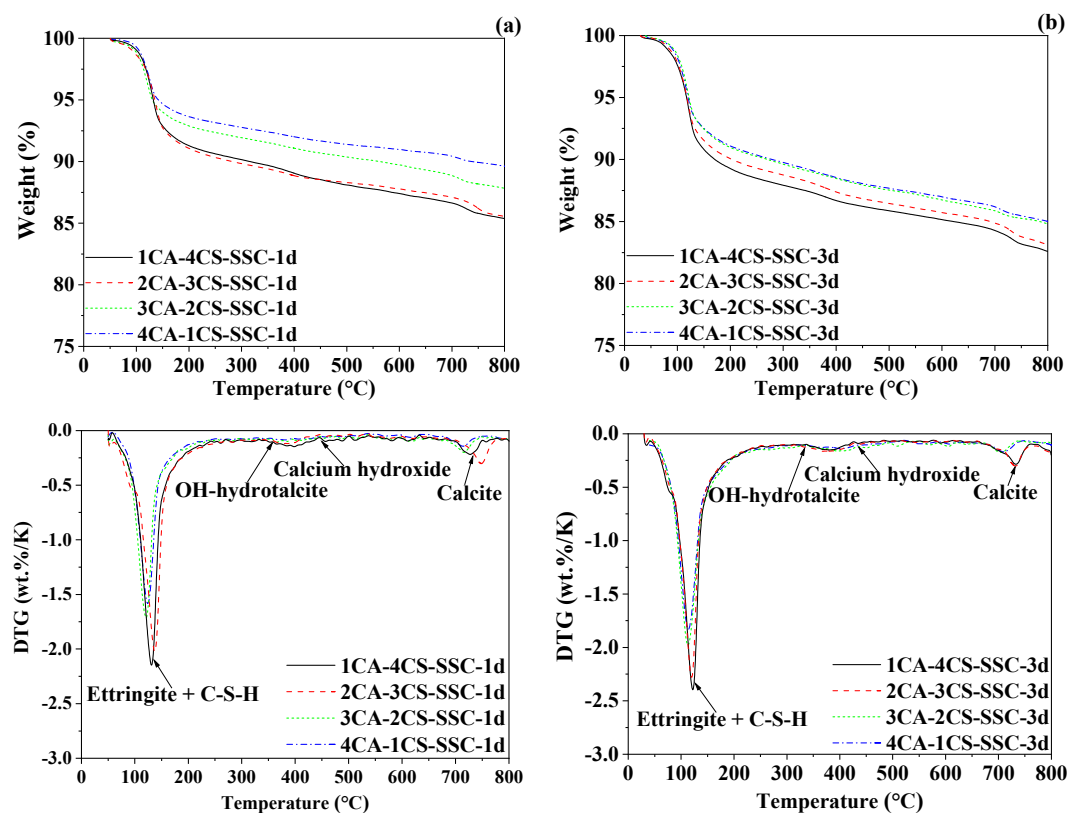


Figure 8. Cont.

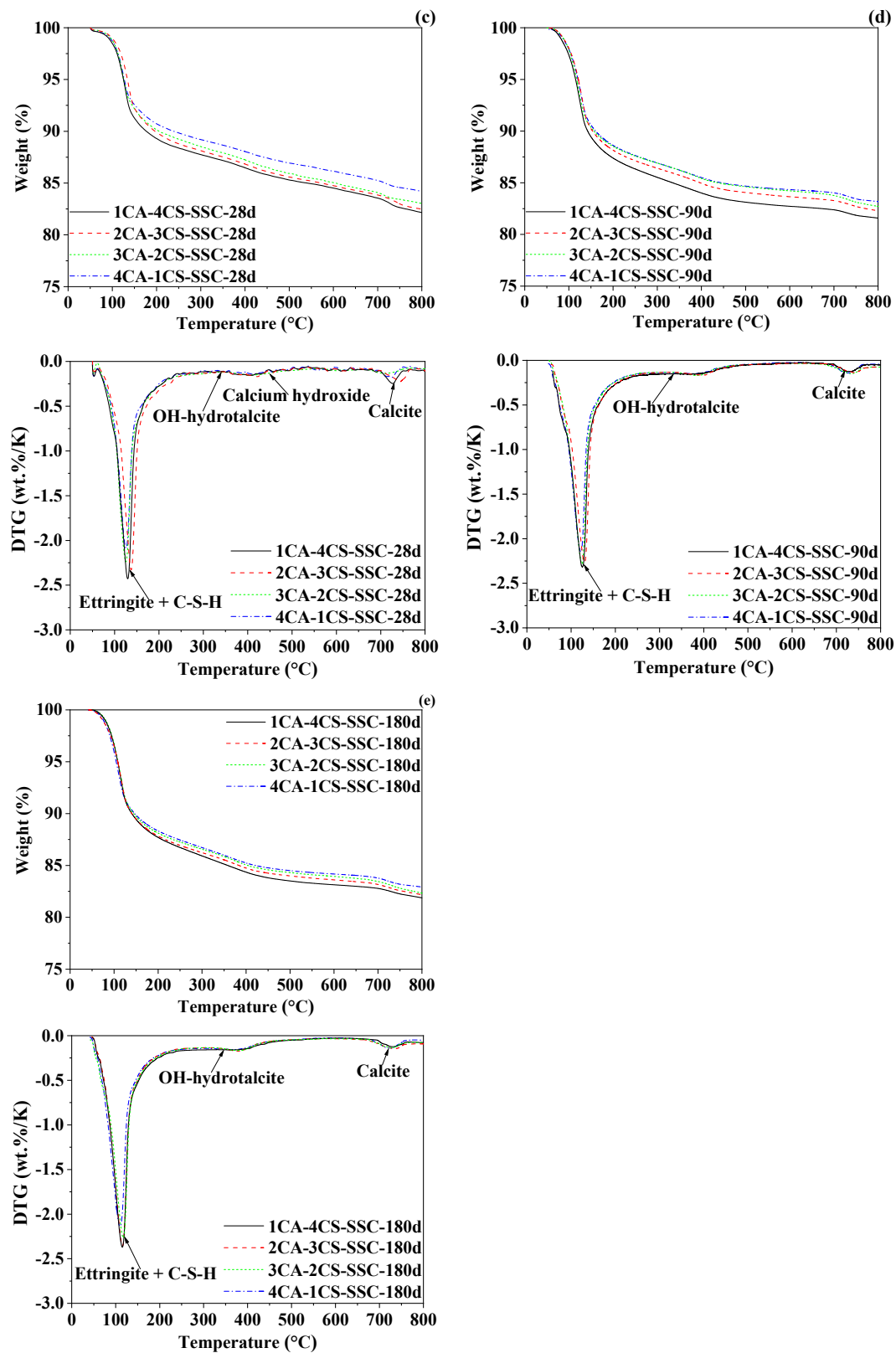


Figure 8. TGA and DTG curves of the CA-CS-SSC system pastes at (a) 1 day, (b) 3 days, (c) 28 days, (d) 90 days, and (e) 180 days.

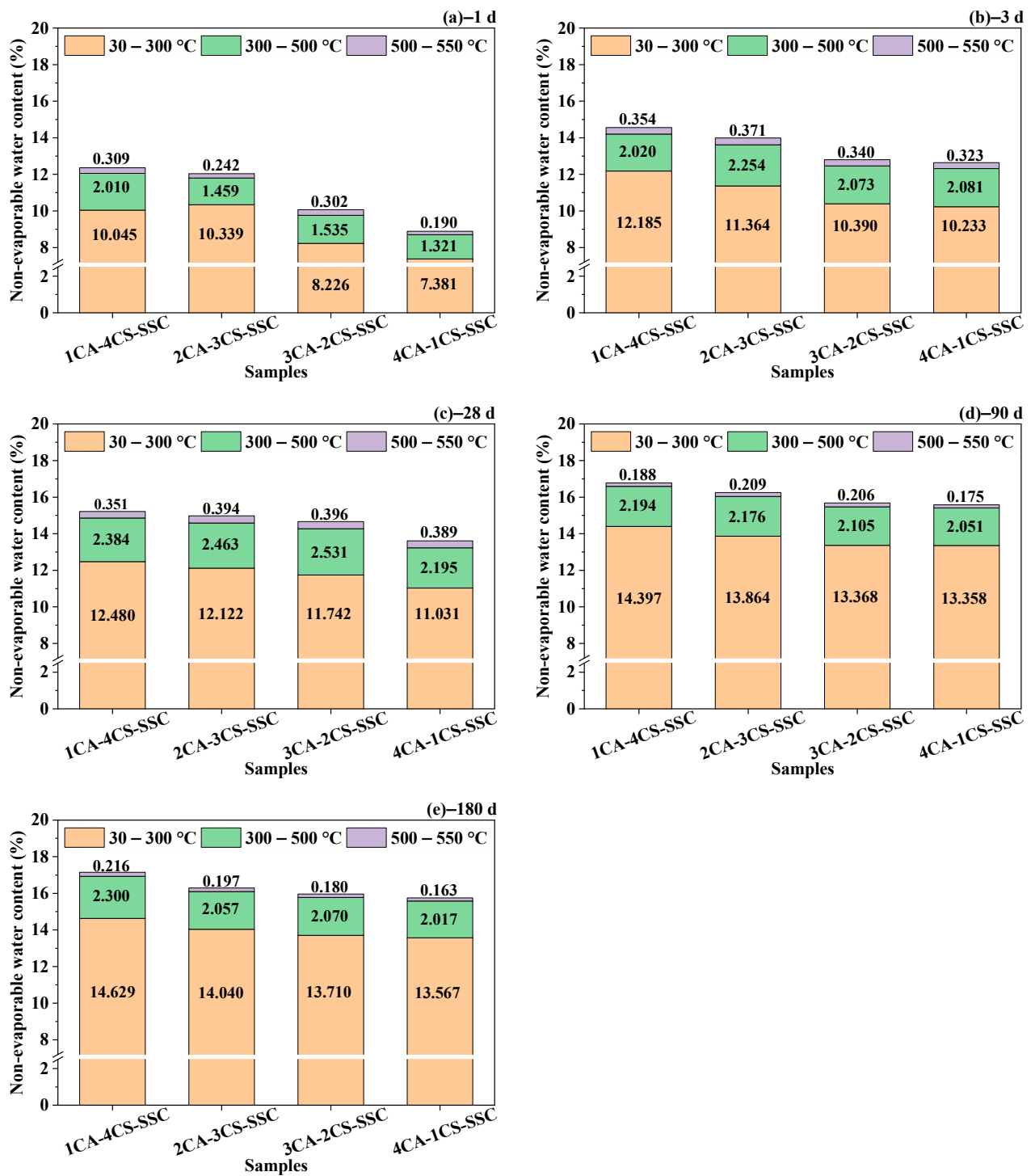


Figure 9. Non-evaporative water content (30–600 °C) of the CA-CS-SSC system pastes at (a) 1 day, (b) 3 days, (c) 28 days, (d) 90 days, and (e) 180 days.

Notably, the compressive strength of 1CA-4CS-SSC mortar was significantly superior to that of other mixtures. Moreover, OH-hydrotalcite was produced within the CA-CS-SSC system pastes, with its quantity increasing as hydration proceeded. Because $\text{Ca}(\text{OH})_2$ (400–500 °C) has a low presence in the CA-CS-SSC system pastes, and its dehydration curve overlaps with those of ettringite, C-S-H, and OH-hydrotalcite, the precise determination of its content is difficult.

3.6. Thermodynamic Modeling

The hydrate assemblages of the CA-CS-SSC system pastes simulated by the thermodynamic modeling are illustrated in Figure 10a–d. The reaction quantities of slag, anhydrite, CS, and CA, serving as input data for the system, were based on the experimental findings acquired via XRD-Rietveld-PONKCS refinement (refer to Figure 7a–d). Predicated on the supposition of uniform slag dissolution, thermodynamic equilibrium was attained at each time juncture, facilitating the computation of the total quantity of hydration products generated. The slag, primarily composed of CaO, SiO₂, Al₂O₃, and MgO, exhibited significantly enhanced hydration reactivity upon activation in an alkaline pore solution. Upon activation, the slag's glassy structure dissolved, leading to increased concentrations of calcium, aluminum, and silicon active components in the liquid phase. In the presence of anhydrite, these active components underwent hydration reactions, resulting in the formation of fine crystalline hydration products, such as ettringite and C-S-H. These hydration products reduced nucleation barriers, facilitating further slag hydration. Introducing an appropriate amount of CS into the SSC further enhanced C-S-H formation, thereby improving the microstructure and mechanical properties of the hardened SSC paste. The thermodynamic modeling simulations revealed that ettringite and C-S-H emerged as the main hydration products of the CA-CS-SSC system pastes, as the slag underwent dissolution and the CA interacted with anhydrite. Additionally, a nominal quantity of OH-hydrotalcite and Al(OH)₃(am) was predicted to form. The C-S-H, OH-hydrotalcite, and Al(OH)₃(am) manifest as amorphous hydration products in the CA-CS-SSC system pastes; nonetheless, their amorphous structure renders them elusive to XRD observation. Hence, the simulated quantities of C-S-H, OH-hydrotalcite, and Al(OH)₃(am) may offer a more reliable estimation. The results from thermodynamic modeling calculations did not exhibit the formation of the Al(OH)₃(am) for 1CA-4CS-SSC paste with a high degree of slag reaction. The thermodynamic modeling results illustrate that the quantity of ettringite generated in 1CA-4CS-SSC paste, computed by accounting for the solid hydration products and unreacted phases based on 100% content, aligns well with the XRD-Rietveld quantitative findings. Moreover, the quantities of C-S-H and OH-hydrotalcite paralleled the total amorphous amount ascertained by XRD-Rietveld. The quantity of ettringite produced by 1CA-4CS-SSC paste at 1 day of hydration was similar to that of other mixtures, yet a significant disparity was observed in the quantities of C-S-H and OH-hydrotalcite produced. These conclusions are consistent with the XRD and TGA results.

For mixtures 2CA-3CS-SSC, 3CA-2CS-SSC, and 4CA-1CS-SSC pastes with higher CA contents, the Al(OH)₃(am) emerged during the early stage of hydration. Nonetheless, in 2CA-3CS-SSC paste, no Al(OH)₃(am) phase was produced during 3–28 days of hydration. The Al(OH)₃(am) phase formed only after 28 days of hydration. Following 90 days of hydration, the variations in the quantities of ettringite and OH-hydrotalcite generated by each mixture within the CA-CS-SSC system pastes were negligible; however, a substantial disparity was noted in the quantity of C-S-H generated.

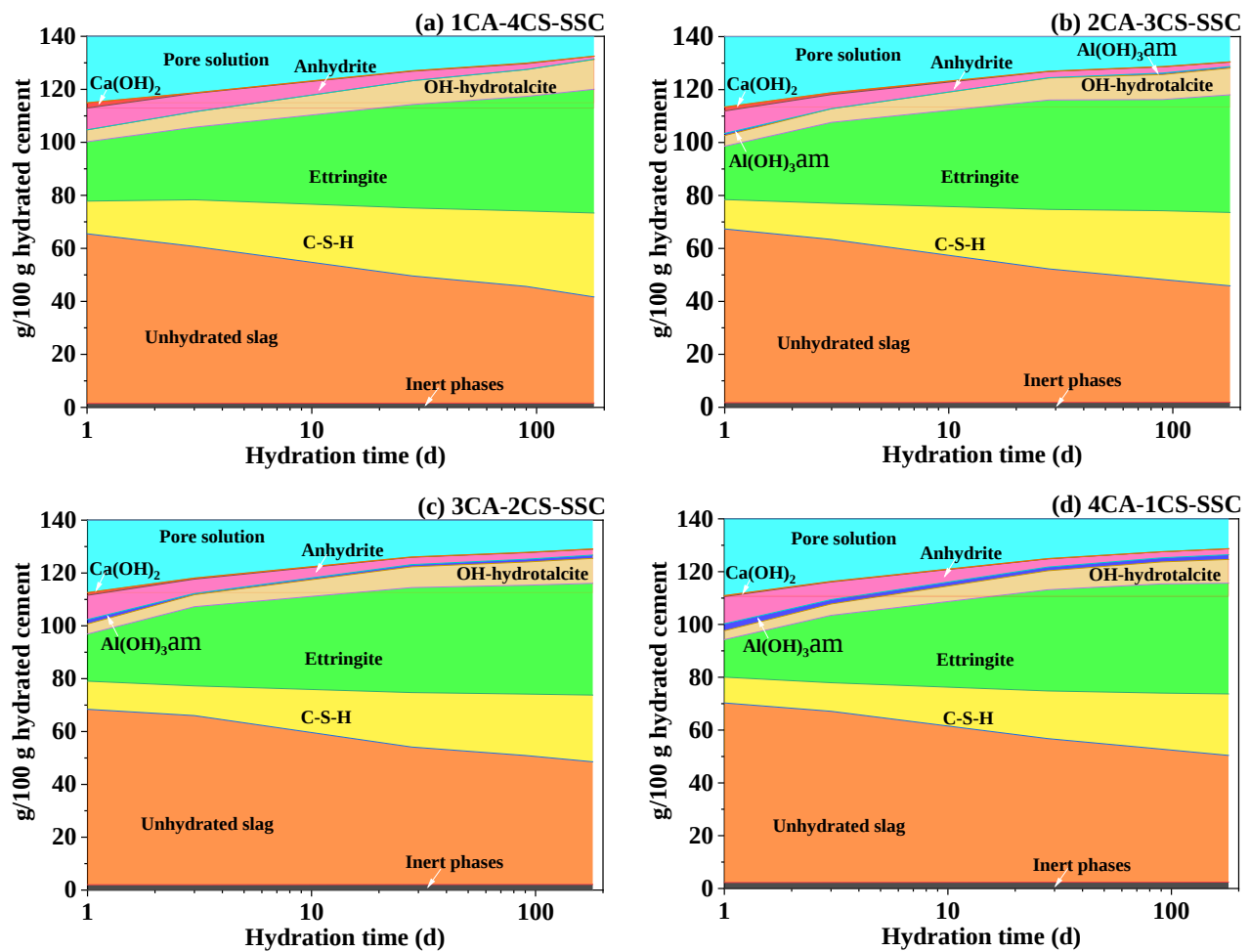


Figure 10. Modeled changes of phase masses of (a) 1CA-4CS-SSC paste, (b) 2CA-3CS-SSC paste, (c) 3CA-2CS-SSC paste, and (d) 4CA-1CS-SSC paste for up to 180 days.

4. Conclusions and Future Prospects

4.1. Conclusions

The investigative findings enrich the understanding of the viability of invigorating SSC adhesives using alkali-containing solid waste materials and reveal the potential for harnessing multiple solid waste materials for SSC preparation in forthcoming endeavors. Through meticulous experimental examination, this study scrutinized the impact of CA and CS on the hydration and hardening attributes of CA-CS-SSC, culminating in the following conclusions:

1. Amplifying the quantity of CA incorporated into the CA-CS-SSC system pastes fostered early ettringite formation within the matrix. Nevertheless, this concurrently procrastinated the slag hydration process. This delayed effect curtails the rate of amorphous gel formations, such as C-S-H and OH-hydrotalcite, thereby making filling the voids generated during ettringite formation difficult. Consequently, despite the augmentation of added CA, it did not significantly increase the early compressive strength of the CA-CS-SSC system mortars;
2. The 1CA-4CS-SSC paste prepared with 1% CA and 4% CS as alkaline activators, rapidly generated a substantial quantity of ettringite in the initial hydration phases, partially owing to slag dissolution with anhydrite. Concurrently, an increased formation of amorphous gels, such as C-S-H and OH-hydrotalcite, occurred to fill the voids generated during ettringite formation, markedly augmenting both the early and late compressive strength;

3. During the early stage of hydration in the CA-CS-SSC system pastes, CA₁ and CA₂ rapidly interacted with anhydrite, producing a significant quantity of ettringite. Simultaneously, the dissolution of calcium hydroxide and anhydrite underwent hydration reactions with small-particle slag, yielding hydration products such as ettringite, C-S-H, and OH-hydrotalcite. With the progression of hydration, the continual consumption of anhydrite led to a deceleration in ettringite formation, eventually rendering C-S-H as the primary hydration product within the CA-CS-SSC system pastes;
4. The proportionate addition of CA to CS in the CA-CS-SSC system pastes had a relatively insubstantial impact on the early ettringite production. However, it wielded a more pronounced influence on creating amorphous hydration products, such as C-S-H and OH-hydrotalcite, in the CA-CS-SSC system pastes with a comparatively higher CA content. As the quantity of added CA decreased and the quantity of added CS increased in the CA-CS-SSC system pastes, the formation of C-S-H and OH-hydrotalcite gradually amplified, concurrently diminishing the porosity.

4.2. Future Prospects

The SSC is gradually emerging as a promising alternative to OPC, primarily due to its lower carbon footprint and efficient utilization of industrial byproducts. This article underscores the significance of several research directions for the future of SSC, which are crucial for enhancing its application performance and environmental sustainability:

1. Conduct in-depth studies on the resistance to sulfate, chloride, and carbonation of SSC, which are crucial for improving the durability of the structures made from SSC. Through long-term exposure experiments under various environmental conditions, a better understanding of the chemical stability and physical properties of SSC can be achieved, thus providing a scientific basis for its application in specific environments;
2. Optimize the mix design of SSC by altering its composition ratio (including blast-furnace slag, gypsum, and alkaline activators) to find the optimal material proportion that balances high performance and cost-effectiveness. The characteristics of materials from different sources should be considered, as well as how they affect the mechanical properties of SSC;
3. Explore the use of various industrial solid waste materials as partial substitutes in SSC to increase the solid waste utilization rate, thereby further enhancing the environmental sustainability of SSC. Investigate the chemical and physical properties of these substitute materials, as well as how they influence the mechanical performance and durability of SSC.

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