

CO₂ storage in self-compacting concrete incorporating recycled aggregates and scallop shell powder under carbonation curing

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ABSTRACT

Carbonation curing is a promising approach for reducing the carbon footprint of construction materials. This study evaluates the carbon storage capacity of four self-compacting concrete (SCC) mixes incorporating recycled coarse aggregates (RCA), limestone powder (LSP), and scallop shell powder (SSP) under controlled conditions. CO₂ uptake is quantified using thermogravimetric analysis (TGA), mass loss, and X-ray diffraction (XRD) with Rietveld refinement, while pH evolution and microstructure are assessed through ex-situ leaching and scanning electron microscopy (SEM). The results demonstrate that RCA- and SSP-based mixes achieved the highest CO₂ uptake, reaching up to 114 kg/m³ of SCC after 90 d. Carbonation depth is not necessarily representative of the actual CO₂ uptake in SCC mixes. The different water retention capacities of SCC precursors strongly influence the moisture state after pre-curing, thereby controlling the dominant reaction mechanisms during CO₂ exposure. A reduction in crystal size and partial destabilization of the calcite phase are observed during carbonation curing. Discrepancies between CO₂ quantification methods are also identified. These findings highlight the role of the surface characteristics of SSP and RCA in modifying transport pathways and enhancing CO₂ storage efficiency, while emphasizing the need for multi-technique approaches for accurate assessment.

1. Introduction

The construction sector has a significant environmental impact, largely due to CO₂ emissions associated with the cement and concrete industry [1–5], which are projected to exceed 52% of global emissions by 2050 [6]. The implementation of carbon capture, storage, and utilization technologies (CCSU) in the construction sector represents an effective means of reducing these emissions [7–10]. These technologies represent a key strategy adopted by the European Union to achieve zero net emissions by 2050. Among them, accelerated carbonation curing enables direct CO₂ sequestration within the cement matrix through the formation of calcium carbonate (CaCO₃), while potentially enhancing early-age performance [11]. The construction industry is also known for its extensive consumption of natural resources, particularly natural coarse aggregates (NCA) and limestone powder (LSP), and for generating large quantities of construction and demolition waste [12–14]. To address this, the use of recycled coarse aggregates (RCA) and scallop shell powder (SSP) as alternatives to NCA and LSP, respectively, has

emerged as a promising strategy. These abundant waste-derived materials exhibit properties similar to those of their conventional counterparts [15–20]. When combined with carbonation curing, these approaches contribute to more sustainable practices in the construction industry by preserving natural resources and promoting CO₂ sequestration within cement-based systems.

In laboratory settings, carbonation curing is typically implemented either by injecting a controlled CO₂ gas flow into a sealed chamber or by curing specimens in carbonated water (wet carbonation) [21,22]. Although both setups promote CaCO₃ precipitation, gaseous CO₂ exposure is the most frequently adopted technique, particularly for precast concrete applications, due to its higher carbonation efficiency and superior process controllability [23–25]. For these reasons, the present study investigates the use of CO₂ gas curing for self-compacting concrete (SCC). This process generally proceeds through three sequential stages: pre-curing, CO₂ exposure, and post-conditioning [21,26]. Pre-curing, which aims to remove free water, is essential for high-slump concretes, such as SCC, playing a decisive role in regulating internal

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Table 1

Quantified phase assemblage and associated crystallographic parameters of samples after 1 d of carbonation curing. Numbers in parentheses indicate the standard deviations as 1σ on the last digits from the Rietveld refinement. Absence of standard deviations means that the values were adjusted at best manually and could not be released during the fits without instability of the refinement. Initial mineral structures were taken from the Crystallography Open Database COD [69].

Mineral name	Initial Unit-cell parameters (Å and °)	LSP $R_{wp}=7.95\%$	SSP $R_{wp}=7.47\%$	RCA $R_{wp}=10.3\%$
		wt (%)	wt (%)	wt (%)
		Unit-cell parameters (Å and °)	Unit-cell parameters (Å and °)	Unit-cell parameters (Å and °)
Calcite	-	100	99(14)	22(6)
	a = 4.98614	a = 4.98680 (6)	a = 4.98849 (6)	a = 4.9875 (1)
	c = 17.0479	c = 17.0495 (3)	c = 17.0801 (1)	c = 17.0634 (7)
Aragonite	-	-	1.1(2)	0.5(6)
	a = 4.96183	-	a = 4.961830 (13)	a = 4.9618 (13)
	b = 7.96914	-	b = 7.9776 (25)	b = 7.9691 (7)
	c = 5.742852	-	c = 5.7471 (13)	c = 5.7428 (7)
Vaterite	-	-	-	2.4(6)
Quartz	-	-	-	69(18)
Muscovite-2M1	-	-	-	1.3(4)
Orthoclase	-	-	-	0.8(3)
Albite low	-	-	-	2.3(6)
Chlorite	-	-	-	1.0(3)

moisture content, which in turn governs both CO_2 diffusivity and the kinetics of hydration and carbonation reactions [27]. Partial water removal enhances CO_2 diffusivity through the pore network, whereas excessive water removal may hinder ongoing reactions [28]. SCC presents additional complexity in this regard. Due to its high paste volume and relatively elevated water-to-cement ratio (w/c), SCC can develop a heterogeneous internal moisture distribution during the pre-curing step [29]. As a result, the optimal degree of water removal for SCC has not yet been clearly established, as it strongly depends on the water retention capacity and drying behavior of the mix constituents, which remain insufficiently characterized. Moreover, numerous studies highlighted the sensitivity of carbonation efficiency to water removal [23,30], temperature [31], relative humidity (RH) [32–36], and exposure conditions [37–39]. Consequently, differences in carbonation protocols can significantly affect CO_2 uptake, making it difficult to isolate the intrinsic influence of SCC constituents, highlighting the need for strictly controlled conditions to obtain reliable comparisons of carbonation behavior.

The CaCO_3 -bearing precursors (LSP and SSP) and coarse aggregates (NCA and RCA) exert distinct influences on transport properties and carbonation behavior. LSP and SSP consist predominantly of calcite, although SSP may also contain aragonite and residual organic content [40–42]. These compositional variations, in addition to the morphological differences of these powders [43–46], have been reported to affect the availability of nucleation sites for hydration and carbonation products [27,47]. It is widely documented that LSP accelerates clinker's hydration by providing nucleation sites on which early C–S–H can nucleate and grow. LSP particles also exhibit a strong chemical affinity with the carbonation products and provide nucleation sites for CaCO_3 precipitation [48–50]. SSP can play the same role; however, its efficiency may differ due to residual organic matter, which can partially block active nucleation sites [51–53]. Conversely, the presence of aragonite in SSP, which is more soluble than calcite, may enhance dissolution and increase Ca^{2+} availability in the pore solution,

potentially promoting carbonation and hydration reactions [54]. In the case of RCA, the old mortar surrounding their surfaces also introduces residual hydrated phases and increased porosity, which modify CO_2 diffusion pathways and local carbonation reactions [55,56]. Based on existing studies, it remains unclear whether LSP or SSP provides more effective nucleation for hydration or carbonation under coupled hydration–carbonation conditions. Moreover, previous studies have generally investigated these materials independently, and a systematic comparison of the combined effects of precursor type (LSP versus SSP) and aggregate nature (NCA versus RCA) under identical SCC carbonation-curing conditions remains lacking.

Furthermore, the carbonation efficiency of cement-based materials is commonly assessed by the CO_2 uptake, which is, in most cases, quantified using thermogravimetric analysis (TGA) [57,58], bulk mass change methods [59], X-ray diffraction (XRD) [60], or qualitatively through carbonation depth using a phenolphthalein indicator [61,62]. However, discrepancies may arise among these techniques, as each probes a different aspect of carbonation reactions. TGA, although subject to the common limitations of powder-based techniques, such as sample representativeness and phase interference, is mainly limited by the overlap of dehydration and decarbonation reactions, particularly those associated with C–S–H carbonation products. In addition, the CaCO_3 polymorphs cannot be easily differentiated based solely on their thermal decomposition behavior, particularly in the presence of CaCO_3 -bearing precursors, which further complicates the quantification of CaCO_3 [63]. XRD quantifies CO_2 uptake through the identification of crystalline carbonate phases, but it does not detect CO_2 incorporated in amorphous phases. In favorable cases, its precision is ~ 1 wt%, but it can reach ~ 5 wt% in complex matrices such as Portland cement [64]. The mass loss method estimates CO_2 uptake from mass variation within the temperature interval of CaCO_3 decomposition; however, the presence of amorphous CaCO_3 complicates quantification, as it may contain up to 20 wt% water [64,65]. Carbonation depth measurement with phenolphthalein primarily reflects changes in pH rather than the total amount of CO_2 chemically stored within the matrix. In other words, the color change is associated with acidification of the pore solution due to CO_2 dissolution and the resulting shift in the carbonate system equilibria. The indicator transition occurs around $\text{pH} \approx 9$, which lies between the carbonate system equilibria ($\text{pK}_{a1} \approx 6.3$ for the $\text{CO}_2/\text{HCO}_3^-$ system and $\text{pK}_{a2} \approx 10.3$ for the $\text{HCO}_3^-/\text{CO}_3^{2-}$ system). It therefore does not correspond to a specific carbonation step or a complete carbonate formation. The decrease in pH in cementitious materials is also linked to the consumption of $\text{Ca}(\text{OH})_2$, a major buffering phase. In contrast, CO_3^{2-} already bound in CaCO_3 crystals does not produce a color change, since the indicator responds to pH in the pore solution rather than to solid carbonates. Consequently, relying on a single technique can be misleading, and comprehensive evaluation of CO_2 diffusion, uptake, and CaCO_3 phases in SCC remains limited.

Accordingly, this study provides a direct comparison of the coupled effects of cement precursor type (LSP versus SSP) and aggregate nature (NCA versus RCA) on carbonation, while systematically investigating the CO_2 sequestration capacity of SCC through a multi-scale assessment including TGA, mass loss method, XRD combined with Rietveld refinement, scanning electron microscopy (SEM), phenolphthalein spray testing, and MATLAB-based image analysis. By combining multiple complementary characterization techniques under strictly controlled carbonation-curing conditions, the study systematically examines the relationships among CO_2 absorption, carbonation depth, pH value variations, and microstructural development, thereby providing a more comprehensive understanding of the carbonation mechanism in SCC.

2. Materials and methods

2.1. Materials

The materials used to produce the SCC mixes included Portland

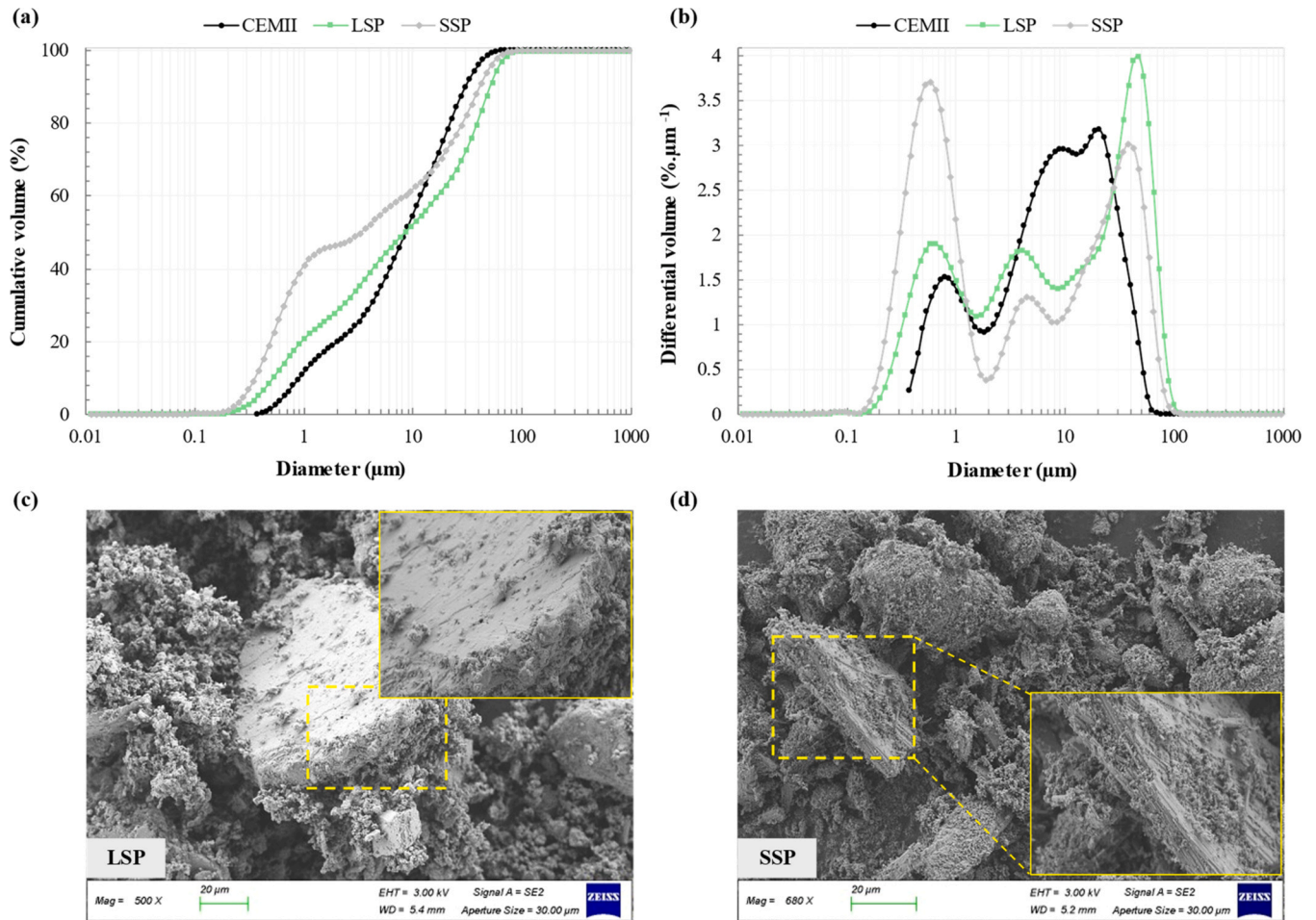


Fig. 1. Physical properties of the binder components used for SCC preparation. (a, b) Particle size distributions. (c, d) Morphologies of LSP and SSP particles.

limestone cement (CEM II/A-LL 52.5 N CE PM-CP2 NF, Ciments Calcia, Ranville, France), commercial limestone powder (Betocarb® HP-EN, Omya SAS, Entrains-sur-Nohain, France), and scallop shell powder (SSP), received in shell form from local seafood processing activities in Normandy, France, and subsequently processed in the laboratory. The aggregates comprised washed siliceous alluvial sand (0–4 mm) supplied by Oscar Savreux (Saint-Firmin-les-Crotoy, France), semi-crushed silico-calcareous natural coarse aggregates (NCA, 4–12.5 mm) supplied by Carrières STREF (Jumièges, France), and recycled coarse aggregates (RCA, 4–14 mm) supplied by Carrières de la Roche Blain – Recyclage Blainville-sur-Orne (Blainville-sur-Orne, France). Raw materials' detailed information on mineralogical compositions, quantified using powder XRD Rietveld refinement; particle size distributions; and morphologies, mainly for the binder components and RCA (Table 1 and Fig. 1), is available from [40]. LSP is composed of calcite, whereas SSP consists mainly of calcite with a minor fraction of aragonite ($\approx 1.1\%$). The slight enlargement in unit cell parameters of calcite observed for both LSP and SSP is consistent with lattice distortions commonly reported in biogenic aragonites of many shells, due to interactions between organic molecules and the mineral matrix [66–68] (Table 1). Regarding the particle size distributions, SSP is characterized by a higher fraction of fine particles than LSP and cement, with approximately 40% of its particle volume below 1 μm. LSP and cement contain about 20% and 10%, respectively (Fig. 1a). Both LSP and SSP also include coarser particles than cement, with mean sizes on the order of 40 and 50 μm (Fig. 1b). Cement particles are mainly distributed within an intermediate size range (2–40 μm), while LSP and SSP exhibit a bimodal particle size distribution, combining fine (<2 μm) and coarse (>40 μm)

fractions. SEM observations show that LSP particles are predominantly plate-like with smooth surfaces and well-defined edges (Fig. 1c). In contrast, SSP particles display more irregular, granular morphologies with rougher surfaces and higher apparent porosity (Fig. 1d), along with a greater tendency for fine particle aggregation.

2.2. SCC mix designs and curing conditions

2.2.1. SCC mix designs

In this study, two variables were investigated: (i) type of aggregates (NCA or RCA) and (ii) the CaCO_3 precursor (LSP or SSP). Based on these variables, four SCC mixtures were designed with a water-to-binder ratio (w/b) of 0.49 and a gravel-to-sand ratio (G/S) of 0.9. The binder content was maintained at 320 kg/m^3 , with 30% SSP or LSP incorporated, and NCA were fully substituted by RCA by volume, resulting in the following mixtures: LSP-NCA (30% LSP, 100% NCA), LSP-RCA (30% LSP, 100% RCA), SSP-NCA (30% SSP, 100% NCA), and SSP-RCA (30% SSP, 100% RCA).

2.2.2. Curing conditions

To evaluate CO_2 uptake in the four SCC mixes, the specimens were cured under two regimes: carbonation curing and sealed curing. The characteristics of these curing regimes are described below.

a) Carbonation curing: In this regime, the specimens first underwent an initial in-mold curing step at 20°C and 50% RH for 7 h. After this phase, the specimens were removed from their molds and pre-cured at 60°C for 17 h to eliminate free water. Following this 24 h pre-

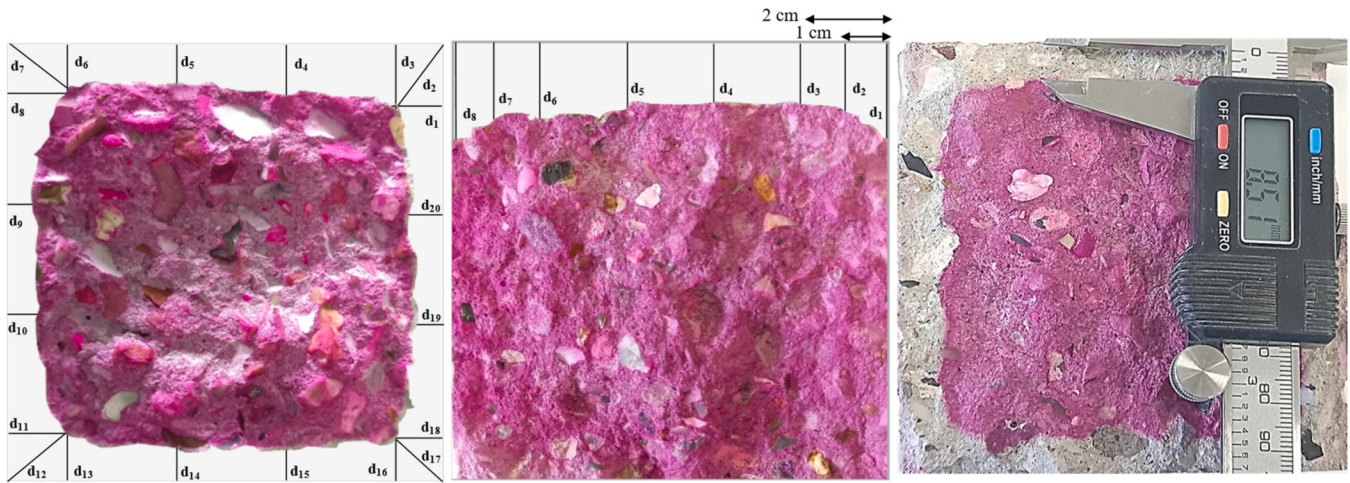


Fig. 2. Schematic for determining carbonation depth.

Table 2

CO₂ uptake calculations steps.

Step 1: Hydrate content participation in carbonations	
$m_{C-S-H \text{ ini}} = m_{C-S-H \text{ sealed}} - m_{C-S-H \text{ carbonated}}$	(Eq. 2)
Reaction mechanism of C-S-H	
$1.7\text{CaO} \cdot \text{SiO}_2 \cdot 4\text{H}_2\text{O} \rightarrow 1.7\text{CaO} \cdot \text{SiO}_2 \cdot 1.3\text{H}_2\text{O} + 2.7\text{H}_2\text{O}$	(Eq. 3)
$m_{C-S-H} = \frac{\Delta m_{150^\circ\text{C}-425^\circ\text{C}} \times M_{C-S-H}}{M_{\text{H}_2\text{O}}}$	(Eq. 4)
$m_{\text{Ca(OH)}_2 \text{ ini}} = m_{\text{Ca(OH)}_2 \text{ sealed}} - m_{\text{Ca(OH)}_2 \text{ carbonated}}$	(Eq. 5)
Reaction mechanism of Ca(OH) ₂	
$\text{Ca(OH)}_2 \rightarrow \text{CaO} + \text{H}_2\text{O}$	(Eq. 6)
$m_{\text{Ca(OH)}_2} = \frac{\Delta m_{425^\circ\text{C}-530^\circ\text{C}} \times M_{\text{Ca(OH)}_2}}{M_{\text{H}_2\text{O}}}$	(Eq. 7)
Step 2: CaCO ₃ content from C-S-H and Ca(OH) ₂ carbonation	
Reaction mechanism of C-S-H	
$1.7\text{CaO} \cdot \text{SiO}_2 \cdot 4\text{H}_2\text{O} + 1.7\text{CO}_2 \rightarrow 1.7\text{CaCO}_3 + \text{SiO}_2 \cdot 3\text{H}_2\text{O} + 0.7\text{H}_2\text{O}$	(Eq. 8)
$m_{\text{CC}_{C-S-H}} = n_{C-S-H} \times 1.7$	(Eq. 9)
$n_{C-S-H} = m_{C-S-H} / M_{C-S-H}$	(Eq. 10)
Reaction mechanism of Ca(OH) ₂	
$\text{Ca(OH)}_2 + \text{CO}_2 \rightarrow \text{CaCO}_3 + \text{H}_2\text{O}$	(Eq. 11)
$m_{\text{CC}_{\text{Ca(OH)}_2}} = n_{\text{Ca(OH)}_2} \times M_{\text{CaCO}_3}$	(Eq. 12)
$n_{\text{Ca(OH)}_2} = m_{\text{Ca(OH)}_2 \text{ ini}} / M_{\text{Ca(OH)}_2}$	(Eq. 13)
Step 3: Total carbonation (total CaCO ₃)	
$m_{\text{CaCO}_3 \text{ tot}} = m_{\text{CaCO}_3 \text{ C-S-H}} + m_{\text{CaCO}_3 \text{ Ca(OH)}_2}$	(Eq. 14)
$m_{\text{CO}_2} = m_{\text{CaCO}_3 \text{ tot}} \times M_{\text{CO}_2} / M_{\text{CaCO}_3}$	(Eq. 15)

curing, the specimens were transferred to a carbonation chamber for curing under controlled conditions. Preliminary trials were conducted to evaluate the effectiveness of the pre-curing. Initially, the specimens were air-dried to remove free water, without the 60°C drying step. After 24 h of carbonation curing, no detectable carbonation front was observed, indicating that air-drying alone was insufficient to allow CO₂ diffusion and carbonation. Consequently, pre-curing at 60°C was introduced to optimize moisture removal and enhance CO₂ uptake. This temperature was fixed as an effective and efficient drying condition based on the literature [70–72]. Internal RH was monitored at a depth of 1.5 cm using humidity sensors. After 17 h in the drying oven, the RH stabilized at 65%, and carbonation curing was initiated to avoid water starvation. The objective of this step was to reach the optimal moisture removal condition for SCC recommended in previous studies [11,23]. During carbonation curing, the CO₂-sealed chamber was regulated at 25 °C, 20% CO₂, and 70% RH. This set of parameters was defined based on ranges reported in the literature and optimized for SCC to maximize CO₂ uptake [31,39]. The specimens underwent carbonation curing for 1, 7, 28, and 90 d to evaluate CO₂ uptake and microstructural evolution from early to long-term [23,73–75].

b) **Sealed curing:** In sealed curing, the specimens underwent the same pre-curing procedure as in carbonation curing. After 24 h, they were wrapped with aluminum adhesive tape and stored in a climatic chamber at 25 °C and 70% RH.

2.3. Methods

2.3.1. Pre-curing phase monitoring

Six cubic, 10 cm-edged pre-cured specimens were evaluated based on their water loss. During drying at 60 °C, the specimens' mass was recorded hourly, and the corresponding water content (*w* %) was calculated using Eq. 1.

$$w \text{ (\%)} = \frac{M_i - M_f}{M_i} \times 100 \quad (1)$$

Where:

- *w* (%): water content.
- *M_i*: mass of the fresh SCC specimens.
- *M_f*: mass of the specimens measured after drying at 105 °C.

2.3.2. Carbonation depth

Carbonation depth was evaluated under two conditions: unidirectional exposure (one surface exposed) and generalized exposure (all surfaces exposed to CO₂). After the exposure period, three cubic specimens of each SCC mix design were split into two pieces. The cross-section was sprayed with phenolphthalein to identify the carbonated zone. The carbonation depth was measured using digital calipers as the average of 8 readings for unidirectional exposure and 20 readings for generalized exposure (Fig. 2). In addition, MATLAB was used to estimate the carbonated area.

2.3.3. Estimation of CO₂ uptake

a) Thermogravimetric analysis (TGA/DTG)

The analysis was conducted using a Thermo Scientific Jupiter STA 449 F5 thermo-gravimetric analyzer (Thermo Fisher Scientific, Waltham, MA, USA), which continuously records mass changes as the temperature increases from 25 °C to 1200 °C at a heating rate of 10 °C/min under a nitrogen atmosphere. The quantitative analysis is based on the following assumptions: (i) during the C-S-H carbonation, the reaction leads to its complete decalcification; (ii) the mass loss occurring between 740 and 920 °C corresponds only to CO₂ release [76]. Based on these assumptions, the CO₂ uptake of SCC mixes prepared for this study is estimated through three steps: (i) calculating the initial mass of C-S-H and Ca(OH)₂, the mass of the hydrate participating in the carbonation

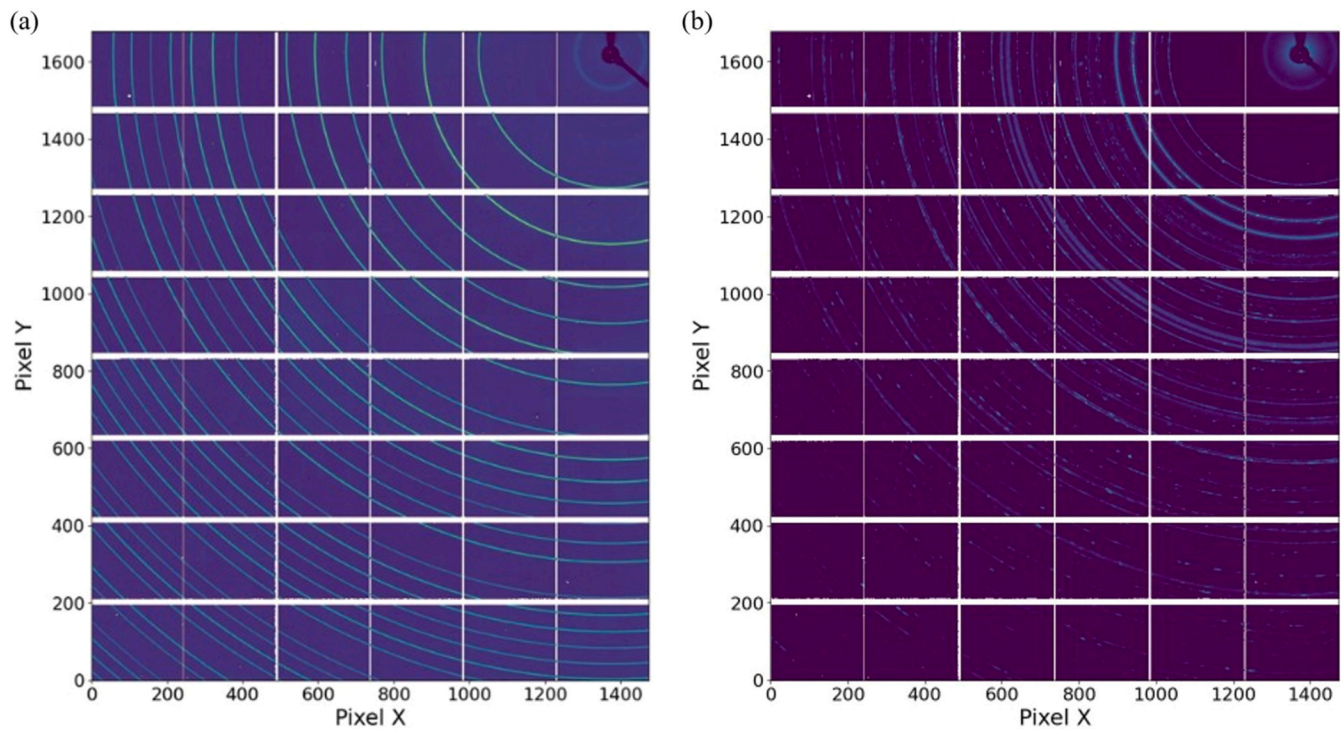


Fig. 3. Illustration of a Debye-Scherrer experiment conducted at ID31, showing (a) the LaB₆ 660b NIST reference powder employed to determine the instrument's contribution, and (b) the LSP-RCA sample after 1 d of carbonation.

reaction, by subtracting their amount in sealed cured samples from their carbonated-cured samples (Eq. 2 to Eq. 7). (ii) calculating the mass of CaCO₃ resulting from their carbonation (Eq. 8 to Eq. 13). (iii) calculating the total CaCO₃ resulting from each carbonation reaction (Eq. 14). The total obtained is used to calculate the CO₂ uptake of the four mixes (Eq. 15). Table 2 summarizes the steps to estimate the CO₂ uptake.

b) Mass loss method

Three cylindrical specimens (Ø 4 cm × H 10.4 cm) from each mix were initially dried in an oven at 105 °C for 24 h to remove moisture. Following this drying stage, the specimens were subjected to a controlled thermal treatment in a muffle furnace. The furnace temperature increased from 25 °C to 525 °C at a controlled heating rate, reaching the target temperature within 49 min, after which it was held at 525 °C for 2 h. Subsequently, the temperature was further increased from 525 °C to 1000 °C, and was then maintained at 1000 °C for an additional 2 h. Afterward, their CO₂ uptake was determined using Eq. 16 [59].

$$CO_{2\text{uptake}}(\%) = \frac{\Delta m_{525^\circ\text{C}-1000^\circ\text{C}}^{\text{Carb}} - \Delta m_{525^\circ\text{C}-1000^\circ\text{C}}^{\text{Cseald}}}{m_{1000^\circ\text{C}}} \times 100 \quad (16)$$

Where:

- $\Delta m_{525^\circ\text{C}-1000^\circ\text{C}}^{\text{Carb}}$: mass loss between 525 °C and 1000 °C of carbonated and sealed specimens.
- $m_{1000^\circ\text{C}}$: sample mass after 1000 °C.

c) X-Ray Diffraction (XRD)

Synchrotron radiation experiments were conducted at the ESRF facility under the Momentum Transfer program of the 4-year EU project OSCARS [77]. A total of 15 powder samples were measured at the ID31 beamline in transmission mode using high-energy X-rays (~75 keV). The detector used on this instrument consists of 48 detecting elements in a Pilatus CdTe 2 M flat panel positioned 1.5 m from the sample. After acquisition, the detector images (Fig. 3) were integrated over the diffraction rings to reconstruct 2θ diffraction patterns for Rietveld

analysis. Full-profile refinements were performed using the Rietveld method implemented in the software MAUD (Materials Analysis Using Diffraction) [78]. MAUD uses an extended Rietveld code that can model and fit all characteristics (structure, texture, phase fractions, stresses, etc.) simultaneously [79]. When required, standard function models were used to account for preferred-orientation effects in certain phases. For all samples, the crystallite size and microstrain of the main crystalline phases (quartz and calcite) were modeled, and in some cases, this approach was also applied to minor phases. Isotropic or anisotropic size and microstrain models were used following the Popa approach [80]. Despite the use of Rietveld refinement, assumptions are required to describe the C-S-H gels, for which appropriate initial structural models must be selected for the starting crystalline phases (e.g., C₃S, C₂S, C-S-H, C₄AH). In this work, the Le Bail method [81] was used to represent these gels in the diffraction patterns. Instrumental characteristics were taken into account during Rietveld refinement by measuring the LaB₆ srm 660c from NIST under the same experimental conditions.

2.3.4. pH

The pH was determined using the ex-situ leaching method [82], which involves mixing powdered SCC with a solvent. The samples were extracted from both the carbonated and uncarbonated zones, manually ground, and sieved through a 60 μm sieve. The passing powder was blended with distilled water with a liquid-to-solid (L/S) ratio of 1. The resulting suspension was agitated for 3 min, after which the liquid was filtered, and the pH was immediately measured using a digital pH meter.

2.3.5. Scanning Electron Microscopy (SEM)

Scanning Electron Microscopy (SEM) was performed to investigate the morphology of CaCO₃ crystals and their evolution during carbonation curing, provided that the phases could be identified based on crystal or grain shape, as SEM does not probe crystal planes. The microstructure was examined using a Carl ZEISS SUPRA 55 microscope (Carl Zeiss AG, Oberkochen, Germany) in secondary electron mode, with an applied voltage of 3 kV, a 60 μm aperture, and a working distance of 3–5 mm to minimize charging. Observations were carried out on non-

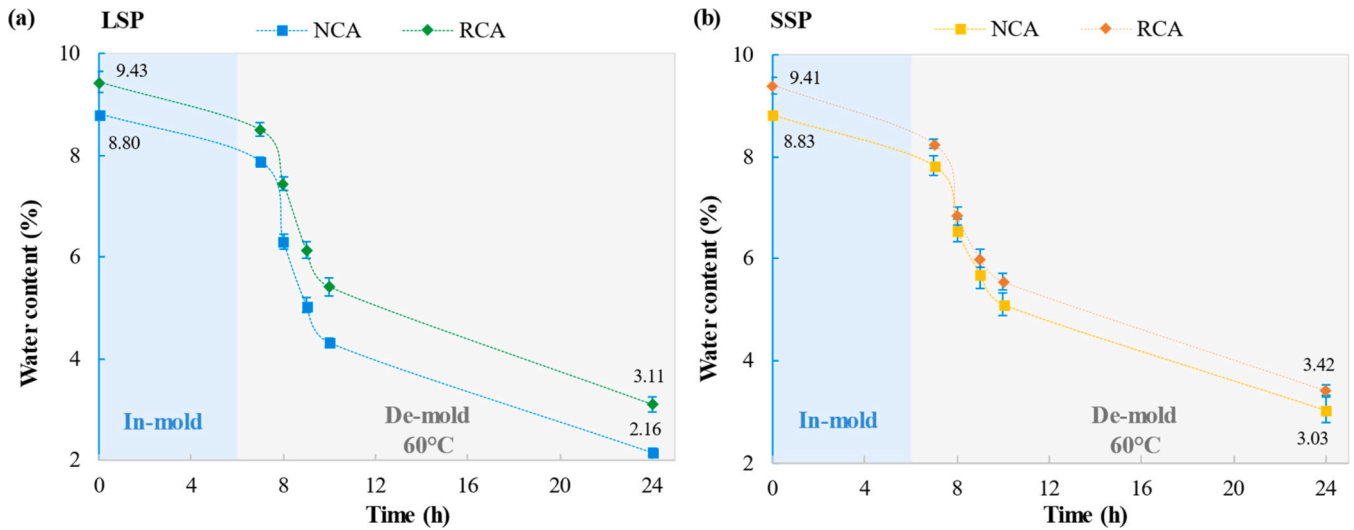


Fig. 4. Water content variation during the pre-curing step: (a) LSP-based and (b) SSP-based specimens.

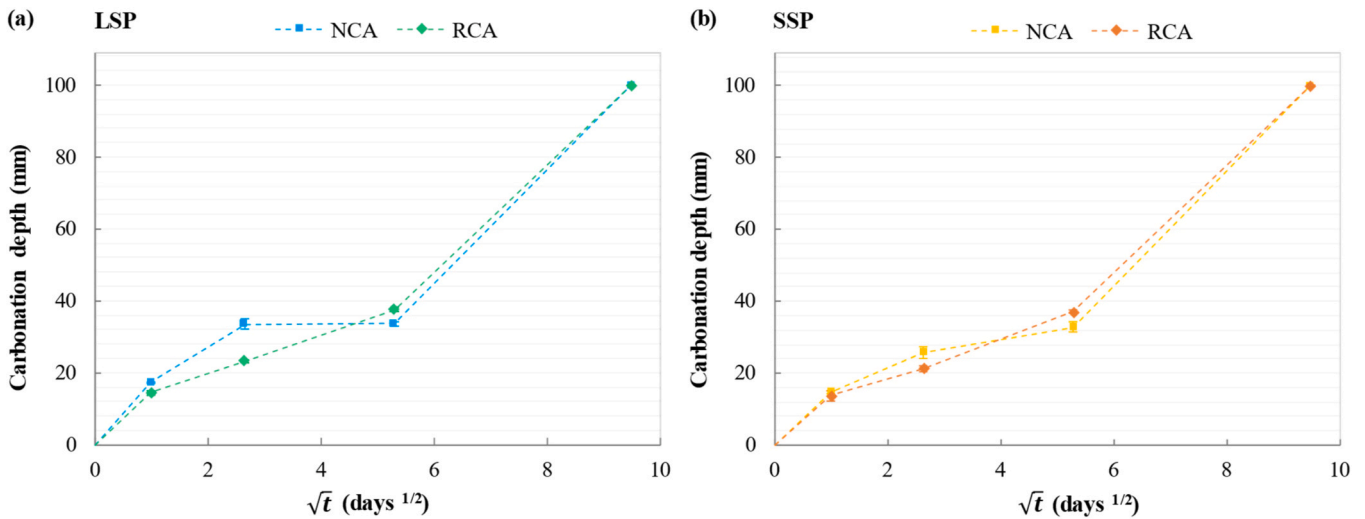


Fig. 5. One-dimensional carbonation depth of (a) LSP-based and (b) SSP-based specimens over time.

coated bulk samples, mounted on conductive carbon tape and surrounded with silver lacquer.

3. Experimental results and analysis

3.1. Pre-curing phase monitoring

The water content of SCC specimens, from in-mold curing through the pre-curing step, is illustrated in Fig. 4, revealing that SCC specimens initially fall into two groups based on aggregate type: NCA-based specimens exhibit lower initial water contents, while RCA-based specimens show higher values. For NCA specimens, LSP-NCA (Fig. 4a) and SSP-NCA (Fig. 4b) initially exhibit similar in-mold water contents of $8.80 \pm 0.01\%$ and $8.83 \pm 0.01\%$, respectively. During the pre-curing step (60°C), water content decreases rapidly between 7 and 10 h, then gradually slows. After 24 h, LSP-NCA specimens reach $2.2 \pm 0.1\%$, lower than SSP-NCA ($3.0 \pm 0.2\%$) and the lowest among all four mixes. This behavior is consistent with the anomalous or non-Fickian drying kinetics reported in cementitious materials [83,84]. Qi *et al.* [72] highlighted that water removal from cement-based porous materials can be governed by moisture diffusion, capillary flow, and sorption-based mechanisms, thereby generating various drying models. In SCC, these

observed differences may be attributed to the finer particle-size distribution of SSP, which increases the contact surface area with water, thereby increasing water retention. Moreover, the hydrophilic character of the intracrystalline organic matrix, which is rich in amino acids with carboxyl functional groups, may promote hydrogen bonding with water molecules [85–87]. Consequently, the retention of physically and chemically bound water reduces the amount of free water available for evaporation [86,88]. In contrast, the water content in LSP-NCA is influenced by LSP's distinct properties compared to SSP, as well as the higher bleeding of the fresh mix and delayed hydration kinetics previously reported [40], which increases the free water available for evaporation and leads to the lowest residual water content among the specimens. Following the same trend, RCA-based specimens start with similar in-mold water contents and reach final values of $3.11 \pm 0.02\%$ and $3.42 \pm 0.01\%$ for LSP-RCA and LSP-NCA, respectively, at the end of pre-curing. These values remain slightly higher than in NCA-based specimens due to the structure of the mortar adhering to the RCA surfaces. This mortar is characterized by heterogeneous capillary pore networks and high tortuosity, which increases the energy required and prolongs the time needed for moisture removal during drying [72,89,90].

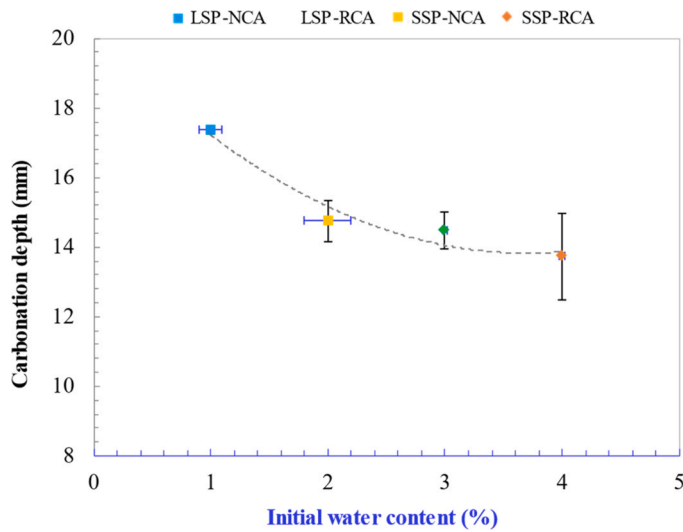


Fig. 6. Relationship between the specimens' initial water content before carbonation curing and the 1-d carbonation depths.

3.2. Carbonation depth

a) One-dimensional diffusion

The carbonation depth progression within SCC specimens as a function of the square root of time up to 90 d of curing (Fig. 5) shows a non-linear trend, deviating from the classical $\sqrt{t}$ law (as would result from Fick-like solid-state diffusion) [91]. This deviation has typically been attributed to variation in carbonation-induced microstructure, environmental relative humidity, and temperature [92]. LSP-NCA specimens (Fig. 5a) show slightly higher carbonation depths of 17.36 ± 0.09 mm compared to 14.75 ± 0.60 mm for SSP-NCA specimens (Fig. 5b). A greater carbonation depth is observed in NCA-based specimens relative to their RCA-based counterparts, when LSP-RCA and SSP-RCA specimens exhibit comparable depths of 14.75 ± 0.52 mm and 13.75 ± 1.24 mm, respectively. After 7 d, the difference between LSP-NCA and SSP-NCA specimens becomes more pronounced, with LSP-NCA reaching 33.50 ± 1.48 mm compared to 25.97 ± 1.65 mm for SSP-NCA. Meanwhile, the carbonation depths of LSP-RCA and SSP-RCA (Fig. 5a and b) specimens remain lower compared to those of their NCA counterparts, measuring 23.24 ± 0.32 mm and 21.29 ± 0.94 mm, respectively. It is well established in the literature that, at early ages

(particularly at 1 d), carbonation is a solid-state diffusion-controlled process in which CO_2 transport is primarily governed by its movement through the specimen's pore network [93]. This transport within the specimens appears to be strongly influenced by internal water content after pre-curing, which reduces effective gas diffusion by partially blocking capillary pores, thereby limiting CO_2 transport pathways and delaying carbonation progression [34,92,94–96]. This effect appears to be more influential on early-age carbonation than the intrinsic porosity of the cement precursor (SSP and LSP, Fig. 1). In addition, the intrinsic characteristics of RCA are not favorable to CO_2 diffusion, particularly during the early stages of carbonation curing. According to Mi *et al.* [97], CO_2 diffusion in RCA concrete occurs through five main pathways, including natural mortar (NM), old mortar (OM), the original aggregate (OA)-NM-interfacial transition zone (ITZ), the OA-OM-ITZ, and the NM-OM ITZ. The structural complexity and heterogeneity of these pathways may impede the early progression of carbonation into the deeper layers of the concrete [56]. These results indicate that pore connectivity and saturation state are more critical to carbonation than total porosity alone. The strong correlation between initial water content and early carbonation supports this assumption, with specimens exhibiting lower internal water showing higher carbonation depths (Fig. 6), in line with previous findings [34,94–96].

As curing time progresses, all specimens show an increase in carbonation depth, with LSP-NCA (Fig. 5a) and SSP-NCA (Fig. 5b) exhibiting similar values. However, the evolution of carbonation depth between 7 and 28 d shows different patterns. In LSP-NCA specimens, the carbonation front at 7 d is clearly defined at 33.50 ± 1.48 mm, with a 1 mm intermediate zone containing uncarbonated spots across the cross-section. This indicates that carbonation progresses rapidly during the initial stage, but is not fully completed within this intermediate zone [61,98]. Consequently, carbonation remains concentrated within this zone rather than advancing deeper (Figs. 5, 7d), reflecting saturation until 28 d. The ongoing carbonation within this intermediate zone can be attributed to the initially low internal water content of these specimens after pre-curing. Under the 70% RH CO_2 exposure conditions, moisture is gradually re-adsorbed and redistributed within the pore structure, creating more favorable conditions for CO_2 dissolution and continued carbonation within the partially carbonated zone. In contrast, SSP-NCA specimens exhibit a steady increase in depth, indicating progressive carbonation during the curing period. By 90 d, the carbonation front reaches the core of both specimens. Regarding RCA-based specimens, the carbonation depth in LSP-RCA and SSP-RCA reaches comparable levels, slightly higher than those observed in their NCA counterparts at 28 d, likely reflecting the higher porosity of RCA. By 90

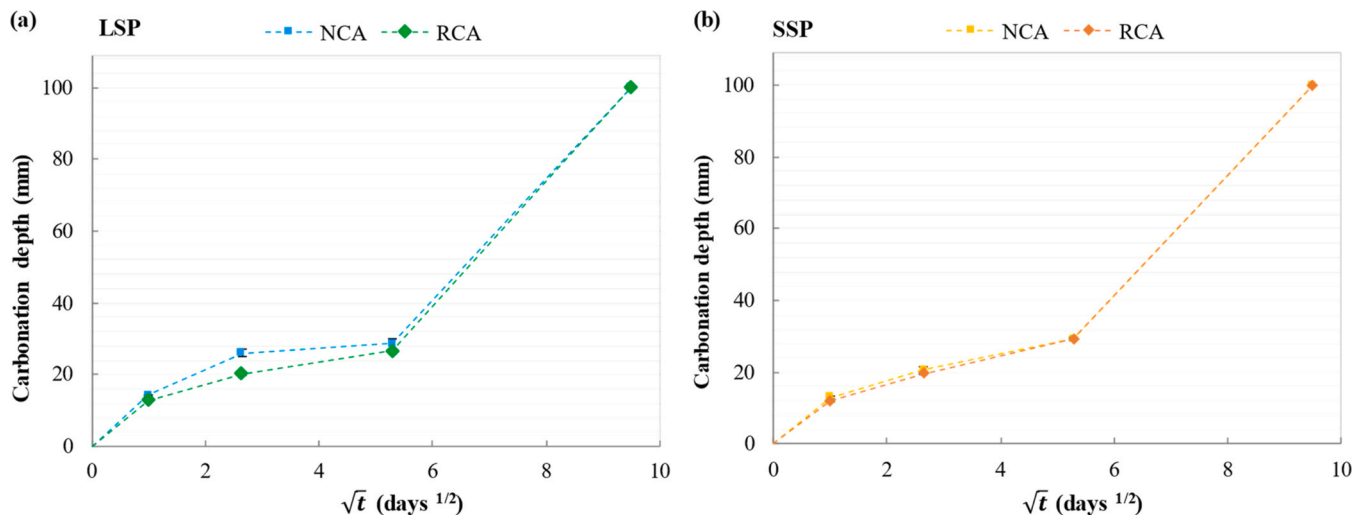


Fig. 7. Generalized carbonation depth of (a) LSP-based and (b) SSP-based specimens over time.

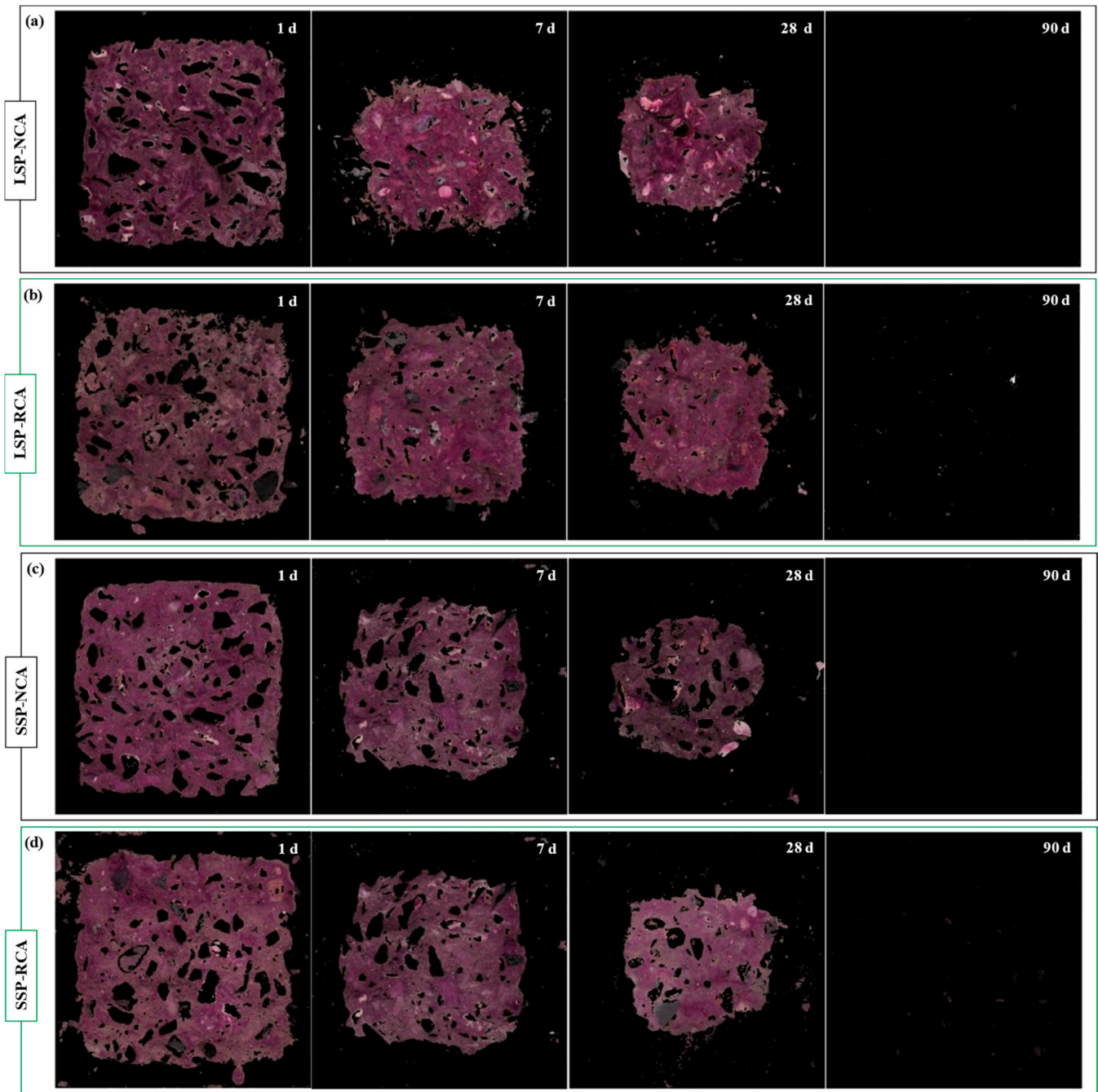


Fig. 8. Temporal evolution of the carbonated area determined by MATLAB image analysis of: (a-b) LSP-based and (c-d) SSP-based specimens.

d, these specimens are fully carbonated. These findings indicate that CO_2 diffusion is initially controlled by mix-design porosity and internal water content, and at later ages, increasingly influenced by ongoing carbonation, hydration reactions, and microstructural changes [99,100].

b) Generalized exposure

When the entire specimen's surfaces are exposed to generalized carbonation, additional features emerge (Fig. 7). In SSP-based and LSP-RCA specimens, carbonation progresses stably and uniformly over time across the exposed surfaces (Fig. 7a and d) [101]. LSP-NCA specimens (Fig. 7a) represent a specific case, exhibiting higher carbonation depths at 7 d, consistent with one-dimensional exposure due to the presence of the previously identified intermediate, partially carbonated zone. These observations are based on carbonation depths measured under generalized exposure, which are lower than those obtained under one-dimensional carbonation. This difference reflects non-uniform

development across surfaces, resulting from variations in surface porosity, local moisture content, aggregate distribution, and demolding-induced microcracks [102]. Consequently, CO_2 distribution is heterogeneous, with the greatest variability observed in LSP-NCA specimens at early ages and in LSP-RCA at 28 d.

c) Image analysis

The total carbonated area was calculated using MATLAB-based image analysis, enabling automated segmentation and area computation. Time-series images of the carbonation front (Fig. 8) show that carbonation initially progresses uniformly, broadly reflecting the original specimen geometry. This progression is governed by CO_2 diffusion through the tortuous pore network at exposed surfaces. As carbonation advances, the front becomes increasingly heterogeneous, with preferential penetration at edges and corners. It then evolves in a quasi-radial manner as the uncarbonated core shrinks until it is completely

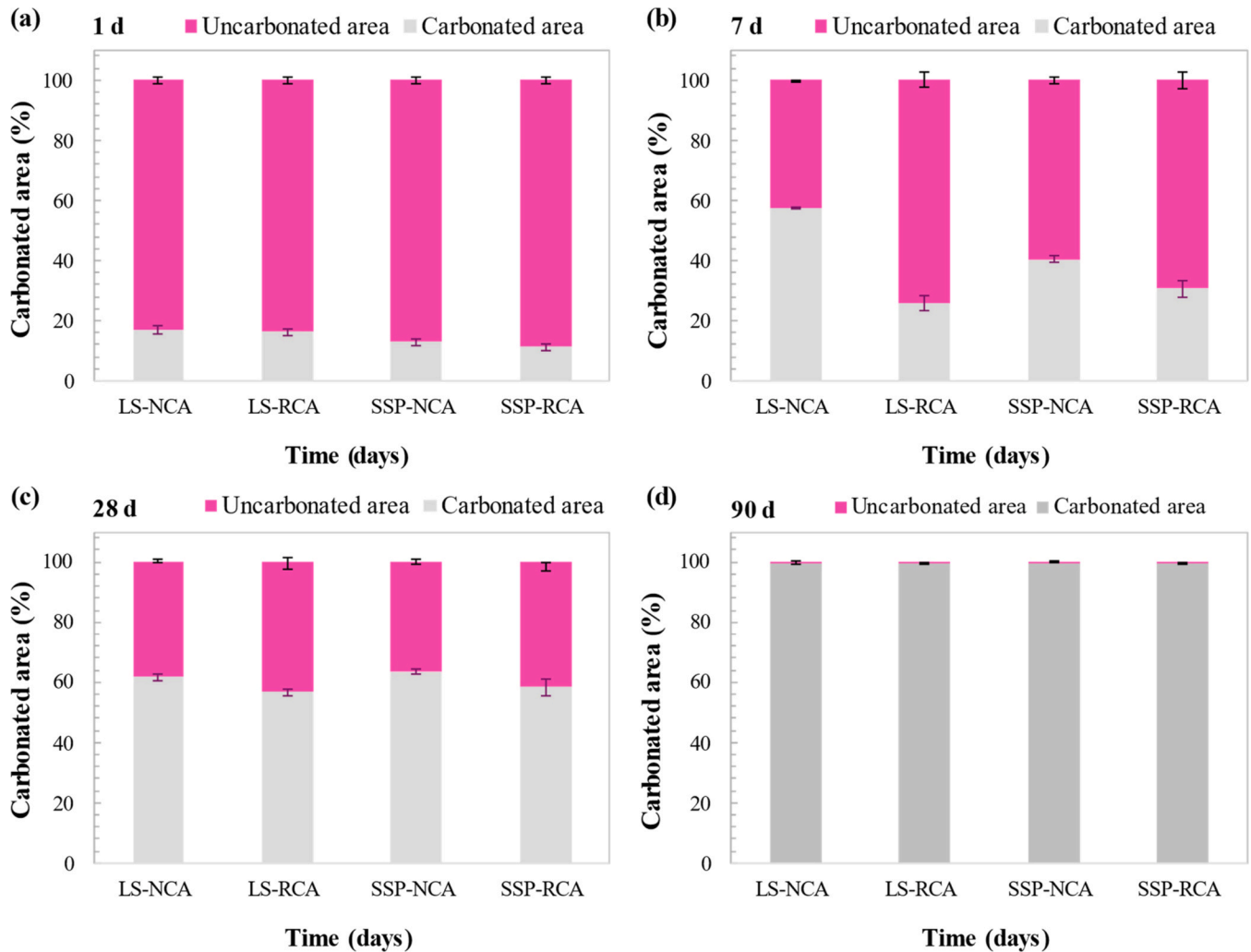


Fig. 9. Carbonated area of specimens after (a) 1 d, (b) 7 d, (c) 28 d, and (d) 90 d of carbonation curing, as determined by MATLAB image analysis.

consumed, at which point the specimens are fully carbonated. This behavior is consistent with previous studies [70,103–105].

Quantitative analysis based on surface integration of Fig. 8 shows that SSP-based specimens exhibit slightly lower carbonated areas than LSP-based specimens after 1 d of carbonation curing (Fig. 9a). At this early age, no notable differences are observed between NCA- and RCA-based specimens. Differences become more pronounced after 7 d (Fig. 9b), with LSP-NCA specimens displaying the largest carbonated area. By 28 d (Fig. 9c), all specimens reach comparable carbonated areas, with slightly higher values in NCA-based specimens. At 90 d (Fig. 9d), carbonation approaches ~99% in all specimens, broadly consistent with one-dimensional measurements obtained using a digital caliper, despite minor discrepancies. Measurement challenges were observed in both methods: caliper readings were accurate when the carbonation front retained the specimen's original shape, but became less reliable as the front became circular. At the same time, coarse aggregates occasionally interfered with MATLAB-based surface calculations. Despite these uncertainties, the results consistently demonstrate nonlinear CO_2 diffusion across all specimens. LSP-NCA exhibits faster early diffusion (up to 28 d) and greater heterogeneity. An intermediate regime, typically 4–30 d, during which carbonation front development is modulated by the various constituent materials in the specimens; these are smoothed out for longer reaction times, up to complete carbonation.

3.3. Estimation of CO_2 uptake

3.3.1. Thermogravimetric analysis (TGA/DTG)

a) Analysis of TGA/DTG curves

The ATG/DTG measurements of carbonated- and sealed-cured specimens were performed on powdered samples collected from regions corresponding to the carbonation depth at each curing age, as determined by the phenolphthalein spray test. For consistency, samples from the sealed-cured specimens were extracted at the corresponding depths (Fig. 10). DTG analysis shows that the peak observed near 100 °C in sealed-cured specimens exhibits the lowest intensity in LSP-based samples, particularly after 1 d of curing (Fig. 10a and b), compared to SSP-based samples (Fig. 10c and d), suggesting a partial inhibition of hydration reactions in these systems. These differences in hydration behavior can be associated with the residual water content in the SCC matrix after the pre-curing stage, as the specimens exhibited varying moisture levels before carbonation [106], as previously described in (§ 3.1). Moreover, the DTG peaks around 170 °C, corresponding mainly to the thermal decomposition of the carboaluminate phases (Hc/Mc), resulting from interactions between SSP- or LSP-derived CaCO_3 and C_3A [107], are slightly more intense in SSP-based samples, particularly from 7 d onward (Fig. 10c and d), compared to LSP-based samples (Fig. 10a and b). This may indicate a slightly higher reactivity of SSP, although confirmation of this trend would require complementary XRD analysis. In addition, the increase in mass loss observed up to 180 °C in carbonation-cured specimens compared to their sealed-cured

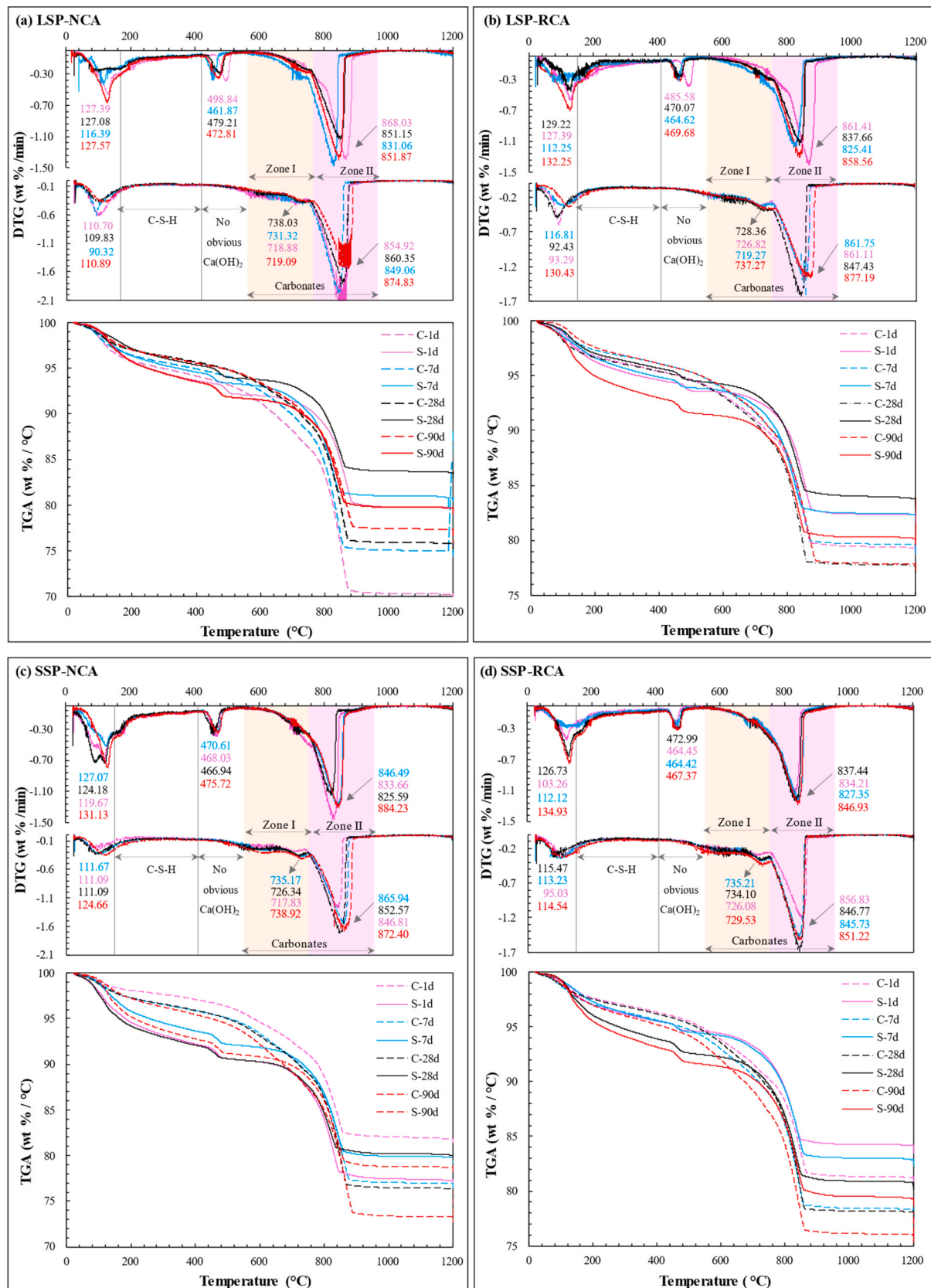


Fig. 10. TGA/DTG curves of sealed- and carbonation-cured specimens over time: (a, b) LSP-based mixes; (c, d) SSP-based specimens.

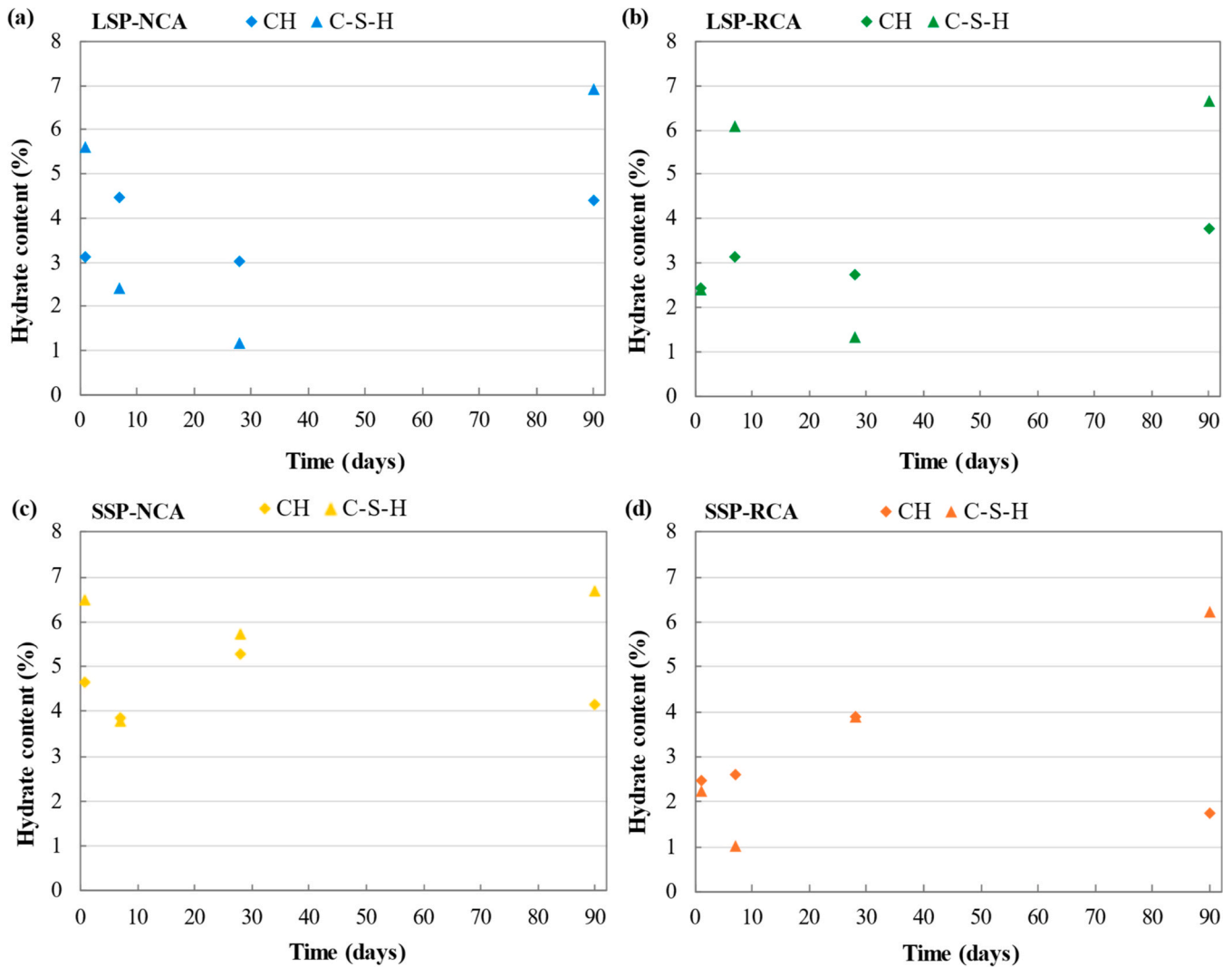


Fig. 11. Ca(OH)_2 (CH) and C-S-H content participating in carbonation over time: (a-b) LSP-based mixtures; (c-d) SSP-based specimens.

counterparts indicates that the carbonation reaction releases physically bound water, thereby promoting hydration. However, it remains challenging to determine whether the additional physical water originates from Ca(OH)_2 or C-S-H carbonation using TGA.

The TGA curves further show that the mass loss from 150 °C up to 425 °C, associated with C-S-H dehydration [58,108], varies across the carbonation-cured specimens, indicating differences in the carbonation behavior of C-S-H within each specimen (Fig. 10). Liu *et al.* [109] identified three successive stages in the carbonation of C-S-H: a dissolution period characterized by a rapid release of Ca^{2+} ions from the interlayer of C-S-H; an initial dissolution stage involving rapid release of Ca^{2+} from the C-S-H interlayer, a diffusion-controlled stage governed by Ca^{2+} availability, and a slower reaction stage resulting from the thickening of carbonation products on C-S-H surface. The transition between these stages may be affected by the internal moisture content of the carbonation-cured specimens [110]. Moreover, according to Liu *et al.* [111], in agreement with Borges *et al.* [106], reported that carbonation curing can concurrently promote hydrate formation and carbonation products, thereby reducing differences in C-S-H-related mass loss between sealed and carbonation-cured specimens. No detectable mass loss was observed between 425 °C and 530 °C in any carbonation-cured specimen, regardless of curing age. The absence of dehydration within this temperature range suggests rapid conversion of Ca(OH)_2 into CaCO_3 [112] (Fig. 10, DTG curves). According to Lu *et al.* [113], this behavior is evidence of a fast carbonation process, in which

CaCO_3 forms before any accumulation of Ca(OH)_2 . Therefore, the absence of Ca(OH)_2 crystals before CaCO_3 precipitation, along with the subsequent pH reduction (to be discussed later), confirms that the carbonation of Ca(OH)_2 proceeds via a dissolution-precipitation mechanism rather than a solid-state transformation [114–116]. This provides evidence that helps resolve the ongoing debate regarding the mechanism of Ca(OH)_2 carbonation.

Furthermore, the thermal decomposition of CaCO_3 in carbonation-cured samples occurs over the range of 550–1000 °C and can be divided into two distinct regions, as evidenced by DTG peaks [117]. The peaks appearing between 550 °C and 680 °C, which are slightly more intense in SSP-based samples (Fig. 10c and d), are associated with the newly formed amorphous CaCO_3 . Hay *et al.* [118] reported that SSP and LSP's nucleation effect promotes the formation of amorphous CaCO_3 phases. However, this interpretation appears to be partly inconsistent with the known tendency of aragonite in SSP to exhibit limited nucleation sites for amorphous CaCO_3 , owing to its elongated morphology, which promotes bridge-like particle networks, its relatively high thermodynamic stability, and its surface structure lacking an oxygen-atom layer [54]. In contrast, the second region peaks at around 719 °C and 738 °C, which are more intense in SSP-based samples and not observed in sealed-cured specimens at later ages, indicating the presence of vaterite and/or aragonite. According to Shao *et al.* [98], these polymorphs are reported as carbonation products of C-S-H present in higher content, consistent with the enhanced hydration observed in SSP-based

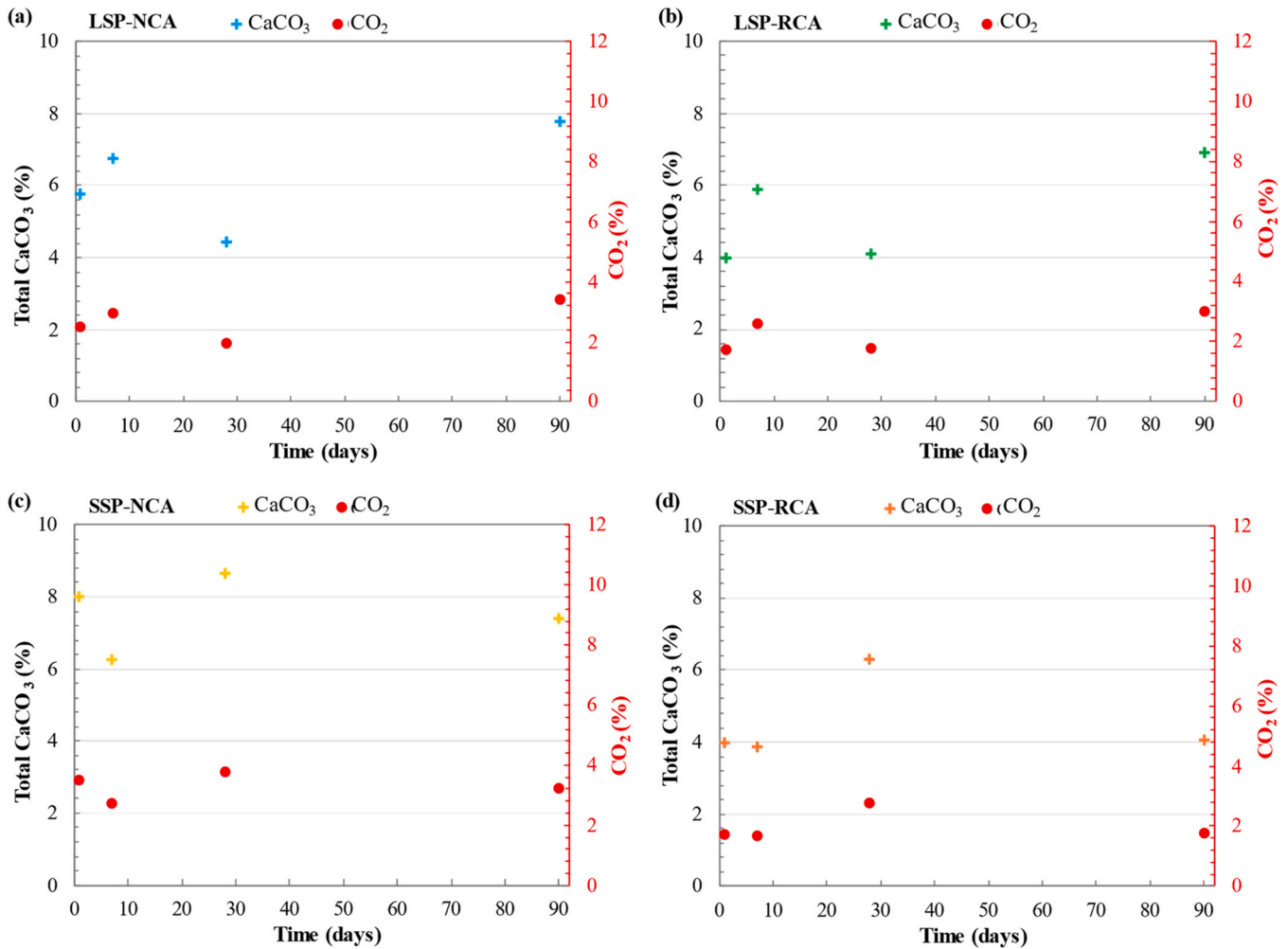


Fig. 12. Evolution of total CaCO_3 content and CO_2 uptake over time: (a-b) LSP-based mixtures; (c-d) SSP-based specimens.

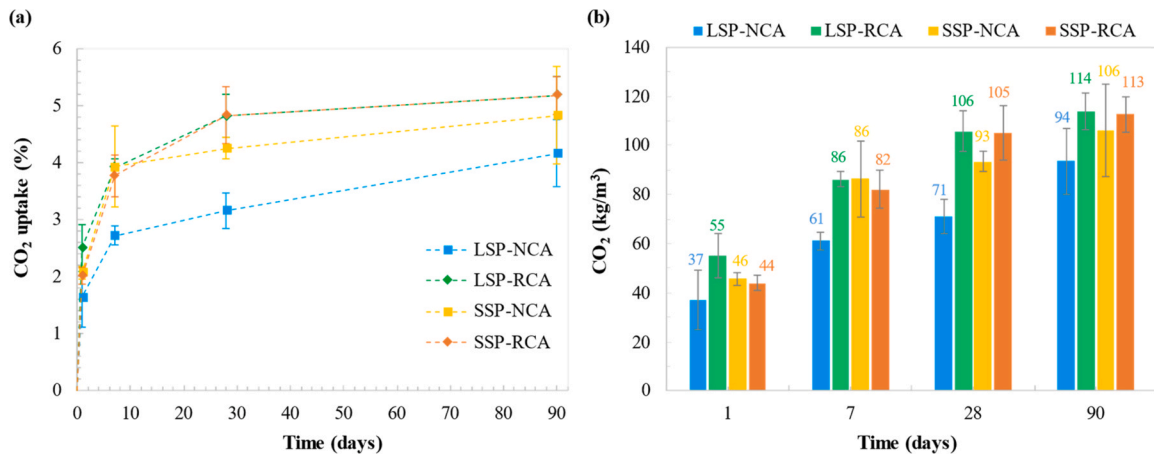


Fig. 13. (a) Mass loss CO_2 uptake measurements and (b) corresponding CO_2 storage (kg/m^3).

samples [119]. It should be noted that part of the aragonite detected in SSP-based samples originates from the SSP itself rather than being newly formed during carbonation. However, its low content (a few percent) is unlikely to significantly affect the DTG results. The decomposition of calcite typically occurs between 740 °C and 990 °C [120], although carbonation curing may shift this decomposition range. This shift suggests that CaCO_3 crystallinity is more strongly influenced by local CO_2

availability than by carbonation duration [121].

b) $\text{Ca}(\text{OH})_2$ and C-S-H carbonation

The extent of carbonation was evaluated based on the contents of $\text{Ca}(\text{OH})_2$ and C-S-H, the main phases involved in the reaction over time (Fig. 10). After 1 d of carbonation curing, the LSP-RCA and SSP-RCA samples (Fig. 10b and d) exhibit lower contents of carbonated $\text{Ca}(\text{OH})_2$ and C-S-H compared to their counterparts with NCA (Fig. 10a

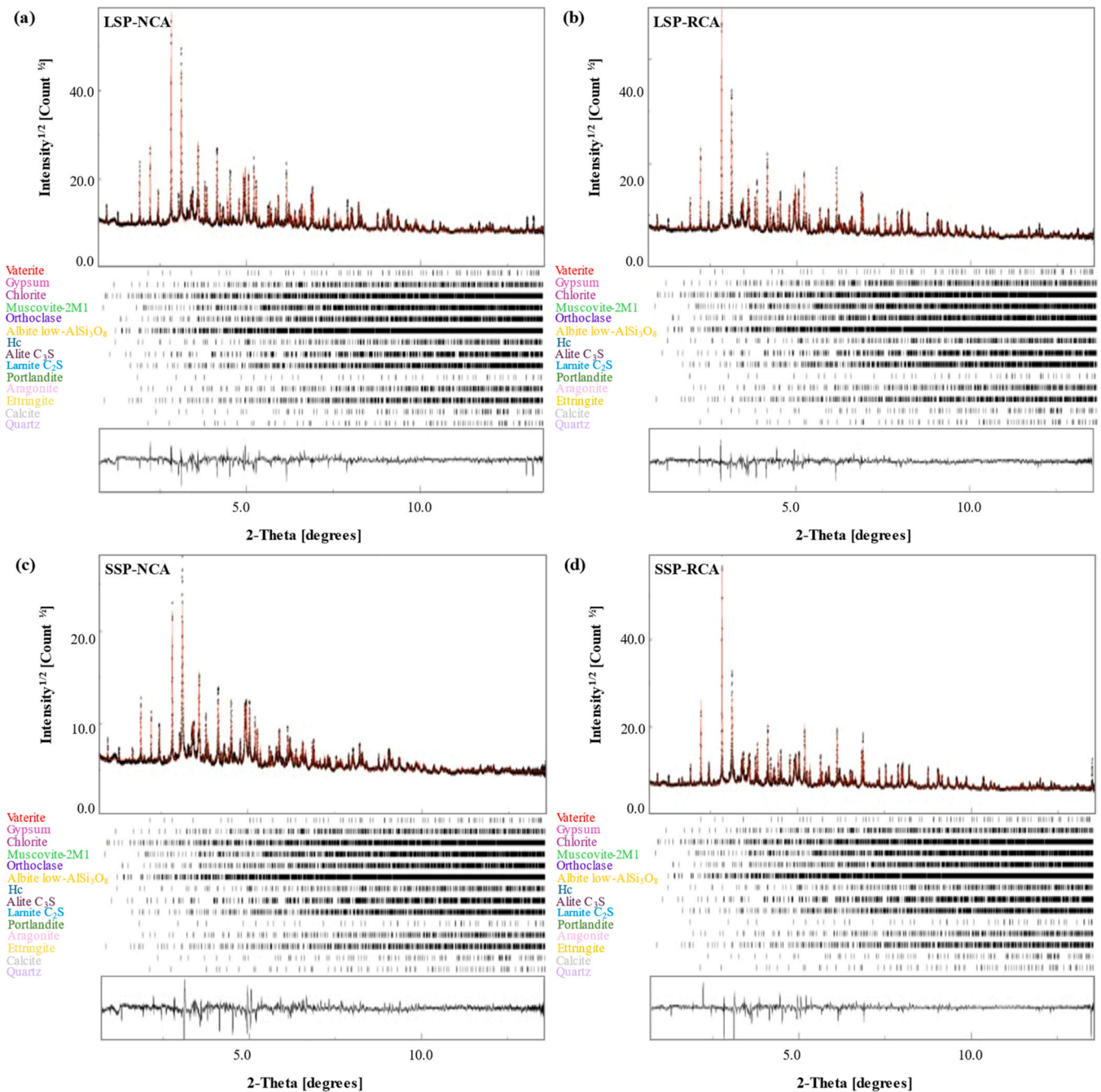


Fig. 14. XRD patterns of 1 d sealed-cured SCC samples.

and c), consistent with slower CO_2 diffusion as discussed in § 3.2. Among the NCA-based samples, the SSP-NCA exhibited the highest carbonated $\text{Ca}(\text{OH})_2$ content, resulting in a greater C–S–H carbonation extent. According to Borges *et al.* [106], in agreement with Lu *et al.* [122], when $\text{Ca}(\text{OH})_2$ carbonation increases, the buffering capacity of the pore solution is impaired (pH drop), causing rapid C–S–H decalcification. At this stage, the carbonation process is primarily controlled by the availability of early-formed hydrates in the outer layer. This availability is likely higher in SSP-NCA than in LSP-NCA, as previously indicated by calorimetry results showing delayed early hydration and lower heat release in LSP-NCA [40]. As carbonation progresses, the $\text{Ca}(\text{OH})_2$ and C–S–H contents undergoing carbonation exhibit a non-monotonic trend across all specimens. These non-monotonic trends may be attributed not only to variations in CO_2 diffusion rate, which are strongly influenced by changes in open porosity within the specimens, but also to the shifting

balance between hydration-controlled phase development and carbonation-induced passivation effects [98] at different depths within each specimen. At certain ages, ongoing hydration can increase $\text{Ca}(\text{OH})_2$ and C–S–H contents, thereby making these phases available for carbonation, explaining the observed increases in hydration products during carbonation. Meanwhile, the subsequent decrease in carbonation may be due to the formation of CaCO_3 layers on hydrate surfaces, which limit further reaction [106,123]. However, these trends cannot be fully elucidated by TGA alone, particularly given that the C–S–H carbonation does not always directly correlate with the simultaneous consumption of $\text{Ca}(\text{OH})_2$. For example, at 90 d, all specimens exhibit a greater content of C–S–H undergoing carbonation than $\text{Ca}(\text{OH})_2$, due to the cumulative effects of hydration, passivation, and diffusion-related factors mentioned earlier. A more comprehensive interpretation can be obtained by incorporating complementary evidence from the XRD results

Table 3

Quantitative phase composition and corresponding unit-cell parameters of samples after 1 d of sealed curing. Numbers in parentheses indicate the standard deviations as 1σ on the last digits from the Rietveld refinement. Absence of standard deviations means that the values were adjusted at best manually and could not be released during the fits without instability of the refinement.

Mineral name	COD	Initial Unit-cell parameters (Å and °)	LSP-NCA	LSP-RCA	SSP-NCA	SSP-RCA
			wt (%)	wt (%)	wt (%)	wt (%)
			Unit-cell parameters (Å and °)	Unit-cell parameters (Å and °)	Unit-cell parameters (Å and °)	Unit-cell parameters (Å and °)
Quartz	1011097	-	27.08(1.2)	34.2(0.2)	14.26(0.1)	38.16(0.3)
Calcite	1547348	-	39(2)	36.9(0.2)	51.5(3)	34.84(0.3)
		a = 4.98614(1)	a = 4.85660(8)	a = 4.9856(7)	a = 4.98566(8)	a = 4.98566(8)
		c = 17.0479(3)	c = 17.0477(3)	c = 17.0477(3)	c = 17.0477(3)	c = 17.0477(3)
Portlandite	1001768	-	7.8(0.3)	3.4(0.05)	6.8(0.1)	2.60(0.06)
Ettringite	9011576	-	2.3(0.1)	2.02(0.1)	2.31(0.2)	1.41(0.1)
Alite (C ₃ S)	9016125	-	2.8(0.2)	4.9(0.1)	4(0.1)	6.30(0.1)
		a = 11.6389	a = 12.223(2)	a = 12.234(1)	a = 12.248(1)	a = 12.2334(10)
		b = 14.1716	b = 7.069(1)	b = 7.0701(7)	b = 7.0620(8)	b = 7.0706(5)
		c = 13.6434	c = 9.295(1)	c = 9.2919(8)	c = 9.300(1)	c = 9.2919(7)
		β = 94.622	β = 116.13°(3)	β = 116.19(9)	β = 116.2(1)	β = 116.17(1)
Larnite-(C ₂ S)	9017424	-	1.9(1)	16.11(0.3)	15.5(0.4)	14.06(0.4)
		a = 5.5051	a = 5.514(4)	a = 5.515(1)	a = 5.514(2)	a = 5.511(2)
		b = 6.7551	b = 6.767(2)	b = 6.7632(2)	b = 6.766(2)	b = 6.770(2)
		c = 9.3108	c = 9.336(2)	c = 9.373(2)	c = 9.370(3)	c = 9.369(3)
		β = 90.59	β = 93.60°(2)	β = 93.6°(2)	β = 93.7°(3)	β = 93.6 (3)
Hc	2105251	-	0.28(0.04)	0.26(0.4)	0.24(0.06)	0.2(0.05)
Albite low	2107372	-	0.20(0.05)	0.2(0.04)	0.21(0.06)	0.14(0.04)
Muscovite	1011049	-	0.013(0.03)	0.02(0.1)	0.05(0.03)	0.08(0.08)
Aragonite	2100187	-	0.7(0.1)	0.7(0.1)	1.31(0.1)	1.09(0.014)
Chlorite	9000158	-	0.1(0.2)	0.02(0.05)	0.23(0.2)	0.26(0.2)
Orthoclase	9000304	-	0.4(0.1)	0.5(0.1)	0.21(0.2)	0.52(0.2)
Vaterite	9007475	-	0.13(0.9)	0.25(0.1)	0.02(0.2)	0.27(0.1)
Gypsum	1010981	-	0.1(0.1)	0.1(0.1)	0.22(0.12)	0.03(0.09)

Table 4

Quantitative phase composition and corresponding unit-cell parameters of samples after 1 d of carbonation curing. Numbers in parentheses indicate the standard deviations as 1σ on the last digits from the Rietveld refinement. Absence of standard deviations means that the values were adjusted at best manually and could not be released during the fits without instability of the refinement.

Mineral name	Initial Unit-cell parameters (Å and °)	LSP-NCA	LSP-RCA	SSP-NCA	SSP-RCA
		wt (%)	wt (%)	wt (%)	wt (%)
		Unit-cell parameters (Å and °)	Unit-cell parameters (Å and °)	Unit-cell parameters (Å and °)	Unit-cell parameters (Å and °)
Calcite	-	41(1)	52(0.4)	55(1)	54(1)
	a = 4.98614	a = 4.9843(3)	a = 4.98376(9)	a = 4.9850(2)	a = 4.9847(1)
	c = 17.0479	c = 17.083(2)	c = 17.0822(6)	c = 17.105(1)	c = 17.10352(5)
Aragonite	-	2.06	2.8(0.1)	1.33(0.2)	1.75(0.1)
Vaterite	-	0.96	1.3(0.1)	0.6(0.2)	0.78(0.07)
Portlandite	-	-	0.2(0.03)	-	0.15(0.03)
Ettringite	-	0.2(0.6)	0.2(0.1)	0.2(0.9)	0.2(0.1)
Alite-C ₃ S	-	1.9(0.3)	1.5(0.1)	2.4(0.2)	2.5(0.1)
Larnite-C ₂ S	-	-	2.7(0.1)	1.8(0.3)	2.58(0.1)
Gypsum	-	0.81	0.5(0.1)	0.1(0.2)	0.36(0.7)
Quartz	-	50(1)	37(0.3)	38(0.5)	34.7(0.2)
Hc	-	-	0.002(0.035)	0.7(0.1)	0.007(0.04)

section.

c) CO₂ uptake

The non-monotonic trends observed in the Ca(OH)₂ and C–S–H results indicate spatially heterogeneous CaCO₃ precipitation, independent of sampling depth (Fig. 12). After 1 d of carbonation curing, SSP-NCA samples show a higher extent of CaCO₃ precipitation and, consequently, greater CO₂ uptake than their LSP-NCA counterparts, reflecting more pronounced early carbonation (Fig. 12a and c). Meanwhile, the RCA-based samples exhibit lower CaCO₃ precipitation and lower CO₂ uptake than their NCA counterparts (Fig. 12b and d). After 7 d, CaCO₃ precipitation increases in LSP-based samples but decreases in SSP-based samples. By day 28, CaCO₃ precipitation decreases in LSP-based samples, while it recovers in SSP-based specimens. By 90 d, CaCO₃ precipitation becomes comparable across most samples, except for the SSP-RCA samples (Fig. 12d). These findings suggest that the finer SSP

particles may promote more rapid initial CO₂ uptake in NCA-based systems, followed by a subsequent decline. It has been reported that finer particles increase the surface area, enabling more surfaces to host water films, which in turn provide more active sites for CO₂ and Ca²⁺ dissolution and shorten the Ca²⁺ diffusion path, thereby accelerating CaCO₃ precipitation [98]. In contrast, LSP particles appear to support a slower but more sustained CO₂ uptake over time. Once again, however, the mechanisms governing these behaviors remain unclear at this stage. The inclusion of RCA generally leads to slower and lower CO₂ uptake throughout the curing period. Saikia *et al.* [124] reported similar behavior, attributing the uneven CO₂ uptake across the specimen depth to an initial dominance of heterogeneous surface-reaction kinetics, followed by a transition to liquid-phase diffusion control. A similar tendency is observed in the present carbonation depth results § 3.2.

Overall, the TGA/DTG results suggest that SSP-based specimens may

Table 5

Quantitative phase composition and corresponding unit-cell parameters of samples after 7 d of carbonation curing. Numbers in parentheses indicate the standard deviations as 1σ on the last digits from the Rietveld refinement. Absence of standard deviations means that the values were adjusted at best manually and could not be released during the fits without instability of the refinement.

Mineral name	LSP-NCA	LSP-RCA	SSP-NCA	SSP-RCA
	wt (%)	wt (%)	wt (%)	wt (%)
Calcite	29.2(0.1)	65(0.4)	57.5(0.3)	65.6(0.2)
Aragonite	1.25(0.1)	3.1(0.1)	0.1(0.1)	1.9(0.2)
Vaterite	0.4(0.1)	2.5(0.1)	0.01(0.06)	1.1(0.1)
Portlandite	9.6(0.1)	0.17(0.03)	0.004(0.03)	0.01(0.07)
Ettringite	4.1(0.1)	0.04(0.08)	0.003(0.064)	0.001(0.041)
Alite-C ₃ S	1.6(0.1)	0.6(0.1)	0.02(0.08)	0.01(0.08)
Larnite-C ₂ S	7.2(0.1)	1.4(0.1)	0.01(0.3)	0.6(0.2)
Gypsum	4.5(0.5)	0.1(0.1)	5.1(0.6)	0.9(0.2)
Quartz	39(0.2)	23.9(0.1)	35.7(0.2)	26.5(0.3)
Hc	-	0.001(0.055)	0.4(0.1)	0.002(0.019)

exhibit higher CO₂ uptake compared to their corresponding LSP-based specimens. The higher porosity of SSP also contributes to the rapid initial CO₂ uptake (Fig. 1), while the uptake after 90 d is relatively identical. In contrast, the porosity of RCA does not appear to significantly accelerate CO₂ uptake, as RCA-based specimens exhibit lower uptake values than NCA-based specimens, according to TGA analysis. The observed fluctuations in the data highlight the inherent variability of the carbonation process, which may be influenced by sampling depth.

3.3.2. Mass loss method

CO₂ uptake was estimated by calculating the difference in mass loss between carbonation-cured and sealed-cured specimens over the temperature range 525 °C to 1000 °C for both carbonation-cured and sealed-

cured specimens at each curing age. Analyzing the CO₂ uptake trend (Fig. 13.a), the estimated CO₂ uptake increases and stabilizes over time across all specimens, with the highest values of 55 ± 9 and 46 ± 3 kg/m³ observed in LSP-RCA and SSP-NCA specimens after 1 d, rising to maximum values of 114 ± 7 and 113 ± 7 kg/m³ in RCA-based specimens at 90 d (Fig. 13.b). This trend is consistent with previous studies [23,58,125]. The magnitude and evolution of the changes vary among the specimens. Interestingly, LSP-NCA specimens, which showed the greatest carbonation depth (§ 3.2), exhibit the lowest CO₂ uptake among SSP-NCA and RCA-based counterparts from 7 d onward. Yuan *et al.* [126] predicted that higher porosity facilitates deeper carbonation but may reduce overall CO₂ uptake, which is consistent with the behavior observed in LSP-NCA specimens. Moreover, although LSP is less porous (Fig. 1), it can contribute to increased intergranular porosity in the SCC

Table 6

Quantitative phase composition and corresponding unit-cell parameters of samples after 28 d of carbonation curing. Numbers in parentheses indicate the standard deviations as 1σ on the last digits from the Rietveld refinement. Absence of standard deviations means that the values were adjusted at best manually and could not be released during the fits without instability of the refinement.

Mineral name	LSP-NCA	LSP-RCA	SSP-NCA	SSP-RCA
	wt (%)	wt (%)	wt (%)	wt (%)
Calcite	57(1)	62.7(0.6)	69 (1.6)	59.3(0.4)
Aragonite	-	0.005(0.194)	-	0.6(0.2)
Vaterite	-	0.3(0.2)	-	0.3(0.2)
Portlandite	0.1(0.1)	0.002(0.06)	-	0.27(0.06)
Gypsum	-	0.007(0.220)	0.005(0.542)	0.2(0.2)
Quartz	42(1)	37(1)	31(2)	39(0.5)
Hc	0.7(22)	-	-	-

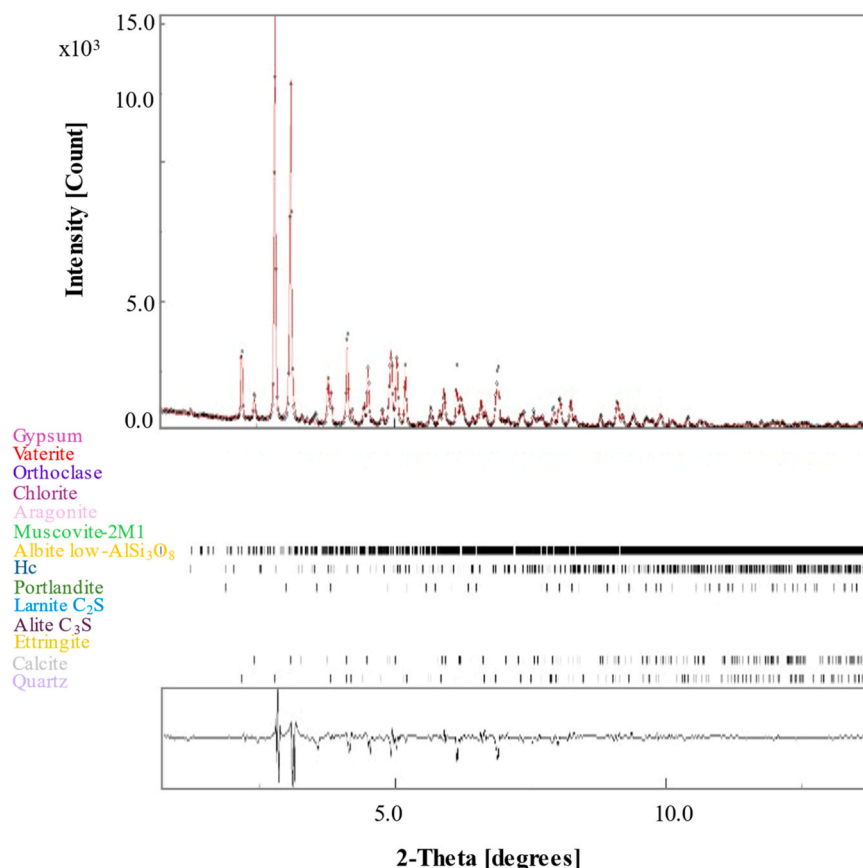


Fig. 15. XRD pattern of the 28 d carbonation-cured LSP-NCA sample showing calcite peak broadening.

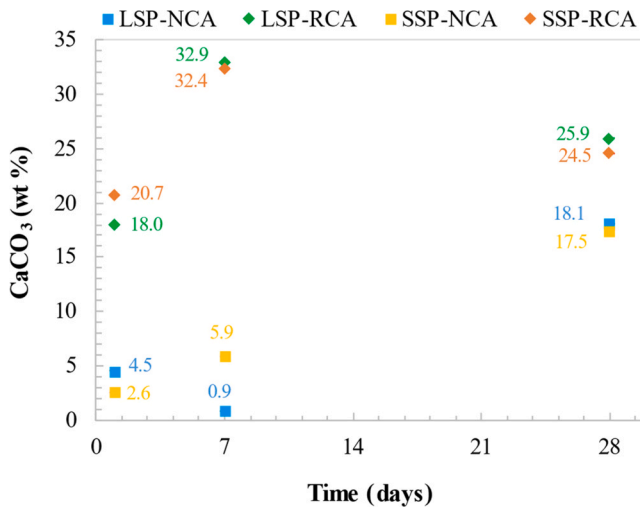


Fig. 16. Estimation of CO₂ uptake via Rietveld-based CaCO₃ content up to 28 d.

matrix. In contrast, SSP and RCA introduce additional intragranular porosity, which may enhance CO₂ uptake despite limiting penetration depth. As a result, specimens with LSP may exhibit greater carbonation depth but lower overall CO₂ uptake.

3.3.3. X-Ray Diffraction (XRD)

XRD was further employed to quantify the relative increase in CaCO₃ content compared with sealed-cured reference specimens. The 1 d sealed-cured samples were used as the baseline, as they represent the initial hydration state before carbonation. The additional CaCO₃ content is defined as the amount exceeding this initial content. The XRD patterns of sealed-cured samples are shown in Fig. 14. Ca(OH)₂ is identified as the dominant crystalline hydration product, with the highest peak intensities observed in LSP-NCA and SSP-NCA samples (Fig. 14a and c) compared to their RCA counterparts (Fig. 14b and d). Ettringite is detected in all samples through low-intensity reflections. The Hc phase is identified by weak peaks in LSP-NCA and only marginally in the other samples, while no reflections associated with Mc are observed. The primary clinker phases (C₃S and C₂S) are present in all samples, with C₃S showing higher peak intensities. Quartz exhibits the strongest reflections, followed by calcite, except in SSP-NCA, where both phases display comparable intensities. Minor phases, including albite, muscovite, chlorite, orthoclase, gypsum, aragonite, and vaterite, do not generate prominent diffraction peaks but are included in the Rietveld

refinement.

Building on the qualitative observations from the diffraction patterns, phase assemblages differ among the samples (Table 3). Calcite and quartz contents vary from approximately 34.80–51.50 wt% and 14.26–38.16 wt%, respectively. These variations are mainly attributed to sample heterogeneity rather than differences in chemical reactions. However, in LSP-NCA, the calcite a-unit-cell parameter ($a = 4.856 \text{ \AA}$) contracted relative to pure calcite ($a = 4.986 \text{ \AA}$), suggesting lattice distortion potentially due to composition variations toward magnesio-calcite. Ca(OH)₂ content is higher in NCA-based samples ($\approx 7.80 \text{ wt\%}$ in LSP-NCA and $\approx 6.85 \text{ wt\%}$ in SSP-NCA) than in RCA-based ones ($\approx 3.40 \text{ wt\%}$ in LSP-RCA and $\approx 2.60 \text{ wt\%}$ in SSP-RCA), indicating enhanced early hydration in NCA-based samples. The relative variations in ettringite content ($\approx 3.32 \text{ wt\%}$ in LSP-NCA and SSP-NCA; $\approx 2.02 \text{ wt\%}$ and $\approx 1.41 \text{ wt\%}$ in LSP-RCA and SSP-RCA, respectively) follow a similar trend to that observed for Hc. This suggests that higher Hc contents may contribute to ettringite stabilization [50]. In addition, the comparable Hc contents across all mixtures indicate that SSP and LSP exert similar effects on early-age hydration after 1 d of sealed curing. Among the clinker phases, LSP-NCA exhibits the lowest C₃S content ($\approx 2.85 \text{ wt\%}$) and the highest C₂S content ($\approx 19.10 \text{ wt\%}$) relative to the other samples. This compositional distribution is consistent with rapid early hydration and potential for further hydration. The refined unit-cell parameter (c) of the two phases shows an enlargement across all samples. Other minor crystalline phases are detected only in trace amounts.

The tendency of these phases to carbonate or undergo further hydration will be examined using XRD analysis of the carbonated samples. The three CaCO₃ polymorphs (calcite, aragonite, and vaterite) are considered collectively for quantification, while phases present in trace amounts are excluded. After 1 d of carbonation curing, XRD patterns show stronger calcite reflections across all carbonation-cured specimens than in their 1d sealed-cured counterparts. The contributions of vaterite and aragonite to the diffraction peak intensities show a slight increase. In contrast, the peaks associated with C₂S and C₃S decrease in intensity, with C₂S becoming undetectable in LSP-NCA samples. No distinct Bragg reflections corresponding to Ca(OH)₂ are identified in the NCA-based specimens.

Rietveld quantification confirms an overall increase in CaCO₃ content, with calcite as the primary polymorph (Table 4). The relative proportion of the metastable polymorphs (aragonite and vaterite) also slightly increases [127]. These phases typically form during early carbonation, before or alongside calcite precipitation, and eventually revert to calcite at later ages [128–131]. Interestingly, the carbonated LSP-based samples show higher contents of vaterite and, more distinctly, aragonite than their SSP-based counterparts (Table 4). As reported

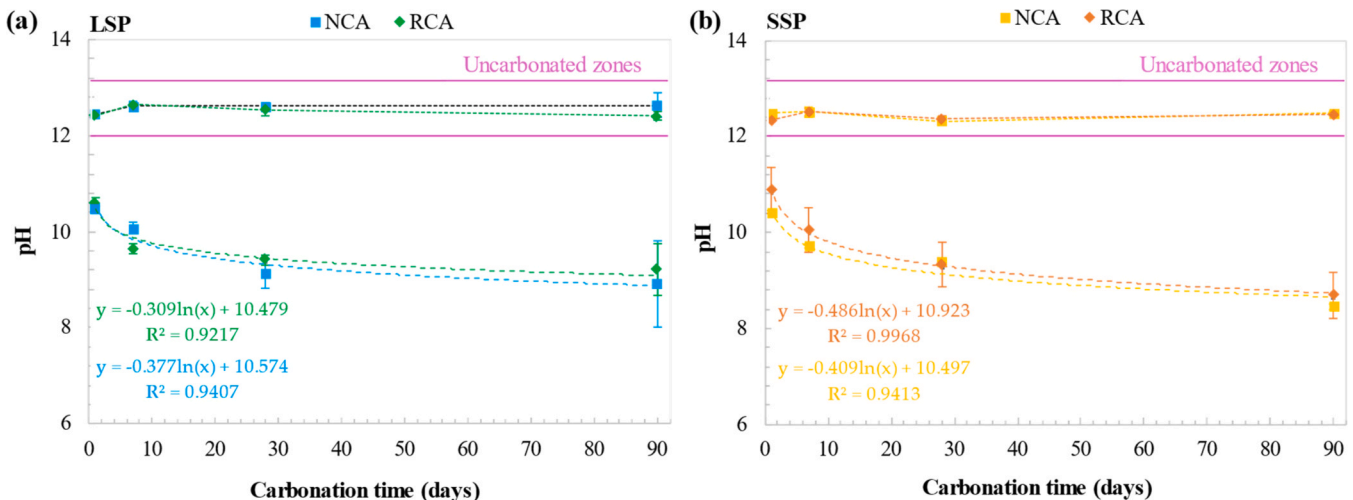


Fig. 17. Evolution of pH of SCC specimens at different carbonation curing ages.

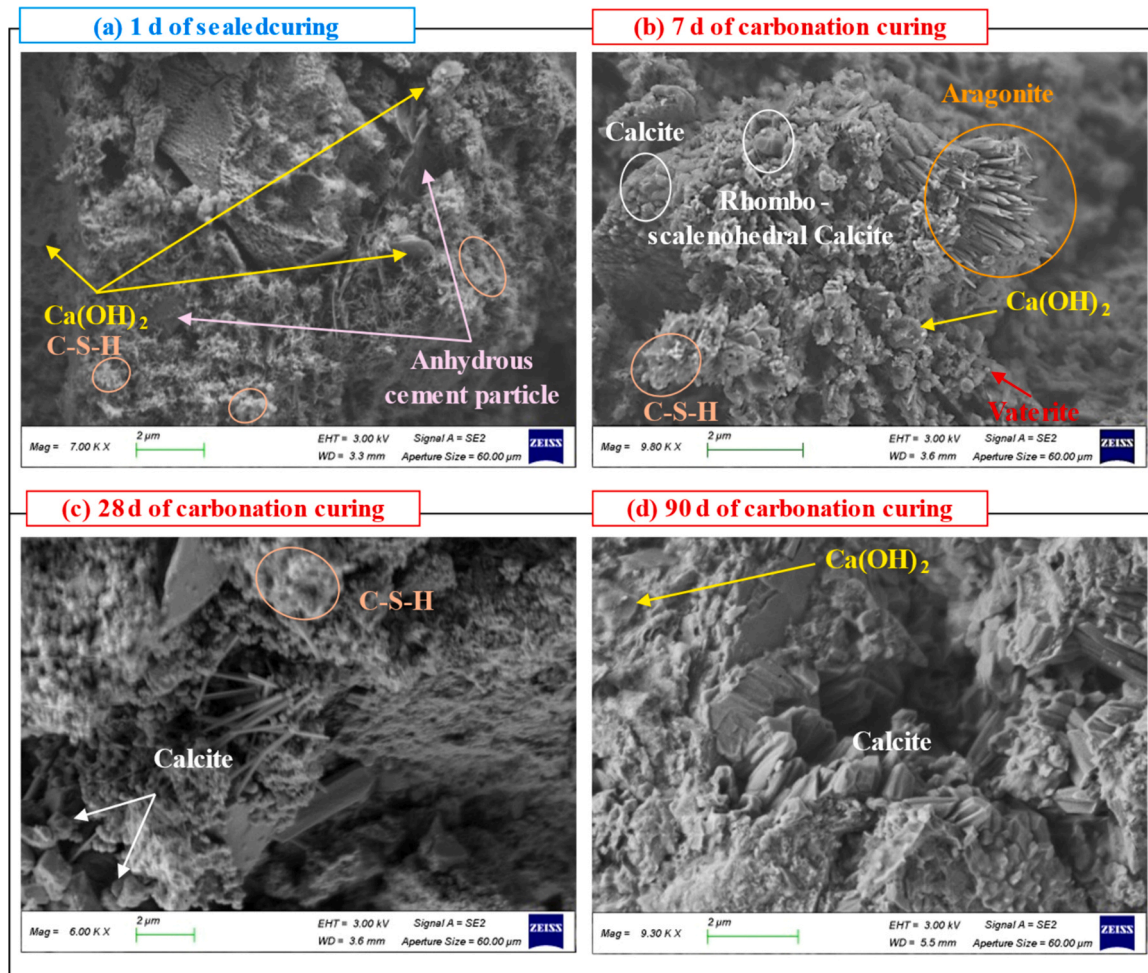


Fig. 18. SEM images showing the microstructural evolution from (a) sealed-cured specimens after 1 d to (b-d) carbonation-cured specimens at different ages for various mix designs.

[132], CaCO_3 polymorphism in comparable mineral systems is strongly influenced by physico-chemical conditions, including temperature, pH, moisture content, and pore solution chemistry. The variations observed among these samples may be attributed to the pre-curing stage, which resulted in lower residual water content in the LSP-based specimens, particularly in LSP-NCA, thereby restricting the availability of water for polymorphic transformation [133]. Upon exposure to CO_2 , all hydrated phases undergo carbonation simultaneously [134,135], leaving only trace amounts of ettringite and Hc in all specimens. The C_2S and C_3S depletion, together with Ca^{2+} release from the aforementioned Ca-bearing phases and its subsequent reaction with CO_3^{2-} ions to form CaCO_3 , confirms the simultaneous progression of hydration and carbonation mechanisms. Moreover, the crystalline phases exhibit variations in their unit-cell parameters (Table 4). In particular, calcite shows a slight enlargement in the c-parameter, consistent with its elastic anisotropy, where the structure is more compliant along the c-axis than the a-axis [136]. During early carbonation, calcite nucleates in a Ca^{2+} - and OH^- -rich supersaturated solution derived from Ca(OH)_2 dissolution. Under these conditions, OH^- ions may be incorporated into the growing calcite structure before full structural relaxation, leading to the observed lattice distortion [137,138]. Impurities derived from minor phases in the reaction medium may also contribute to calcite lattice distortion.

After 7 d, calcite reflections intensify in LSP-RCA, SSP-NCA, and SSP-RCA, while LSP-NCA shows more pronounced diffraction peaks of Ca(OH)_2 and ettringite. Vaterite and aragonite remain detectable in LSP-NCA, LSP-RCA, and SSP-RCA, but exhibit very low intensities in SSP-NCA. Rietveld refinement results confirm continued calcite formation

in LSP-RCA, SSP-NCA, and SSP-RCA, as reflected by increased calcite contents (Table 5). In contrast, LSP-NCA shows a marked reduction in calcite content (from ≈ 49.93 wt% to ≈ 29.22 wt%), which is also lower than that of the sealed-cured reference ($\approx 36.8\%$). This reduction is attributed to partial dissolution of pre-existing limestone-derived calcite, which is strongly dependent on pH and the pore solution's chemistry during carbonation. Under typical conditions, such dissolution is expected to be followed by recrystallization once pH increases and sufficient Ca^{2+} and CO_3^{2-} ions become available [139]. However, in LSP-NCA specimens, reprecipitation is limited because the released Ca^{2+} is rapidly consumed by hydration reactions, as evidenced by the increased Ca(OH)_2 and ettringite contents, and eventually C-S-H formation [73]. On the other hand, vaterite and aragonite contents are approximately 1.25 wt% and 0.42 wt% in LSP-NCA, 3.09 wt% and 2.47 wt% in LSP-RCA, and 1.91 wt% and 1.14 wt% in SSP-RCA, while remaining negligible in SSP-NCA, indicating nearly complete transformation toward calcite in this sample.

After 28 d, aragonite and vaterite peaks are no longer observed in LSP-NCA samples and SSP-NCA samples, indicating the transformation of these metastable phases into the more stable calcite (Ostwald's step rule). Reflections of C_2S and C_3S are no longer detected. However, it remains unclear whether these phases first hydrate or directly destabilize under carbonation conditions. Ibáñez *et al.* [140] reported deeper carbonation penetration in C_2S than in C_3S , consistent with the faster hydration kinetics of C_3S , which have been reported to be further accelerated under carbonation curing [141]. Rapid C_3S hydration produces Ca(OH)_2 near the surface, which quickly carbonates to a dense

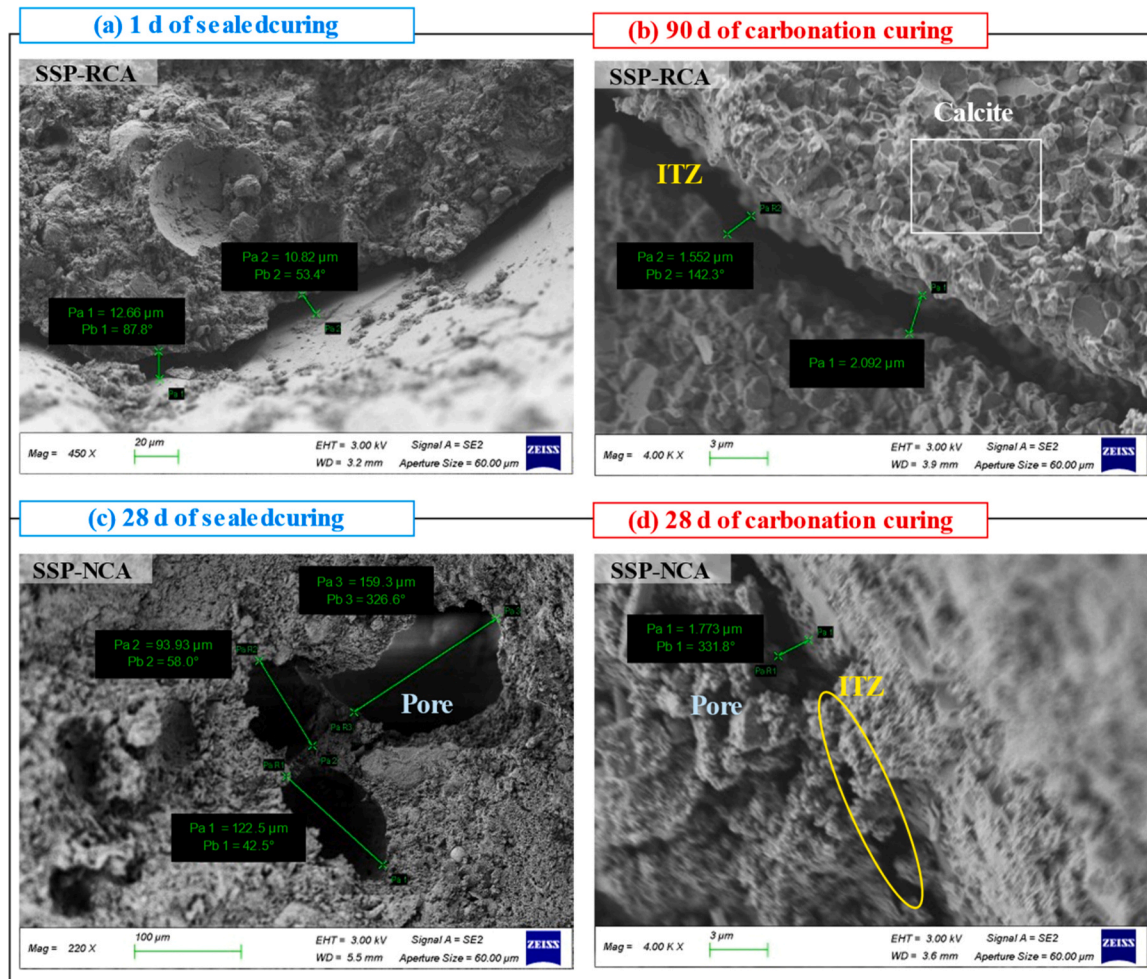


Fig. 19. SEM images of the ITZ and the adjacent matrix before and after carbonation curing.

CaCO_3 layer that may hinder further CO_2 ingress. In contrast, the slower hydration of C_2S allows deeper carbonation, leading to greater destabilization of this phase during prolonged carbonation curing. A minor contribution of Hc remains in the LSP-NCA samples (Fig. 15). During carbonation, Hc is expected to destabilize into Mc and subsequently into strätlingite. However, none of these secondary phases are detected in the XRD patterns throughout the carbonation curing period. This is likely due to the rapid destabilization of Hc, which prevented its conversion into these phases, and/or to low initial Hc content in the system [142]. Moreover, Rietveld refinement reveals that calcite peaks broaden with carbonation time across all samples, reflecting a decrease in crystallite size. Takasaki *et al.* [143] reported that calcite nanocrystals evolve during carbonation under decreasing pH via ion-by-ion assembly from amorphous CaCO_3 , initially forming ~ 50 nm bundles that later fragment into monodispersed nanograins. The reduced crystallite size from Rietveld refinement reflects this fragmentation process, indicating a shift toward nucleation-dominated formation and finer calcite domains. A concurrent decrease in amorphous phases in carbonation-cured samples further indicates C–S–H carbonation [135].

Regarding phase contents (Table 6), calcite shows a marked increase in SSP-NCA samples compared with the other mixtures. Trace amounts of aragonite and vaterite persist in RCA-based samples. Ettringite appears to be fully destabilized under carbonation, whereas minor quantities of $\text{Ca}(\text{OH})_2$ remain in LSP-NCA, LSP-RCA, and SSP-RCA. The absence of C_2S and C_3S indicates the extra reaction of the anhydrous cement during the carbonation, as well as near-complete carbonation of the specimens' inner layer [135,144].

Estimation of the CO_2 uptake. CO_2 uptake is expressed as CaCO_3 content in carbonation-cured samples. The CaCO_3 precipitated during 28 d of carbonation curing (Fig. 16) shows that RCA-based samples exhibit higher CO_2 uptake than NCA-based counterparts. Both LSP-RCA and SSP-RCA display rapid early carbonation within the first 7 d, reaching ~ 32 – 33 wt% CaCO_3 , followed by a slight decrease to 25–26 wt % at 28 d. NCA-based samples exhibit similar CO_2 uptake during early carbonation and maintain comparable levels at the final curing age. Differences arise at intermediate carbonation ages, with SSP-NCA samples retaining more CO_2 , whereas LSP-NCA samples favor continued hydration over carbonation. These results align with mass loss measurements (§ 3.3.2).

3.4. pH

Carbonation significantly reduces the pH of cement-based materials, as observed in the carbonation-cured SCC specimens (Fig. 17). After 1 d, the pH decreases from an average of 12.43–10.61 across all samples. This drop is primarily associated with the consumption of $\text{Ca}(\text{OH})_2$, which governs pore solution alkalinity. As carbonation progresses, the depletion of OH^- ions reduces the buffering capacity of the system, leading to further acidification. At this stage, other cementitious phases are also destabilized, as confirmed by XRD results. After 7 d, the pH further decreases but remains above 10 units in LSP-NCA and SSP-RCA, while reaching 9.65 and 9.72 in LSP-RCA and SSP-NCA samples (Fig. 17a and b). This behavior may indicate either incomplete $\text{Ca}(\text{OH})_2$ consumption or the subsequent formation of hydrated products that

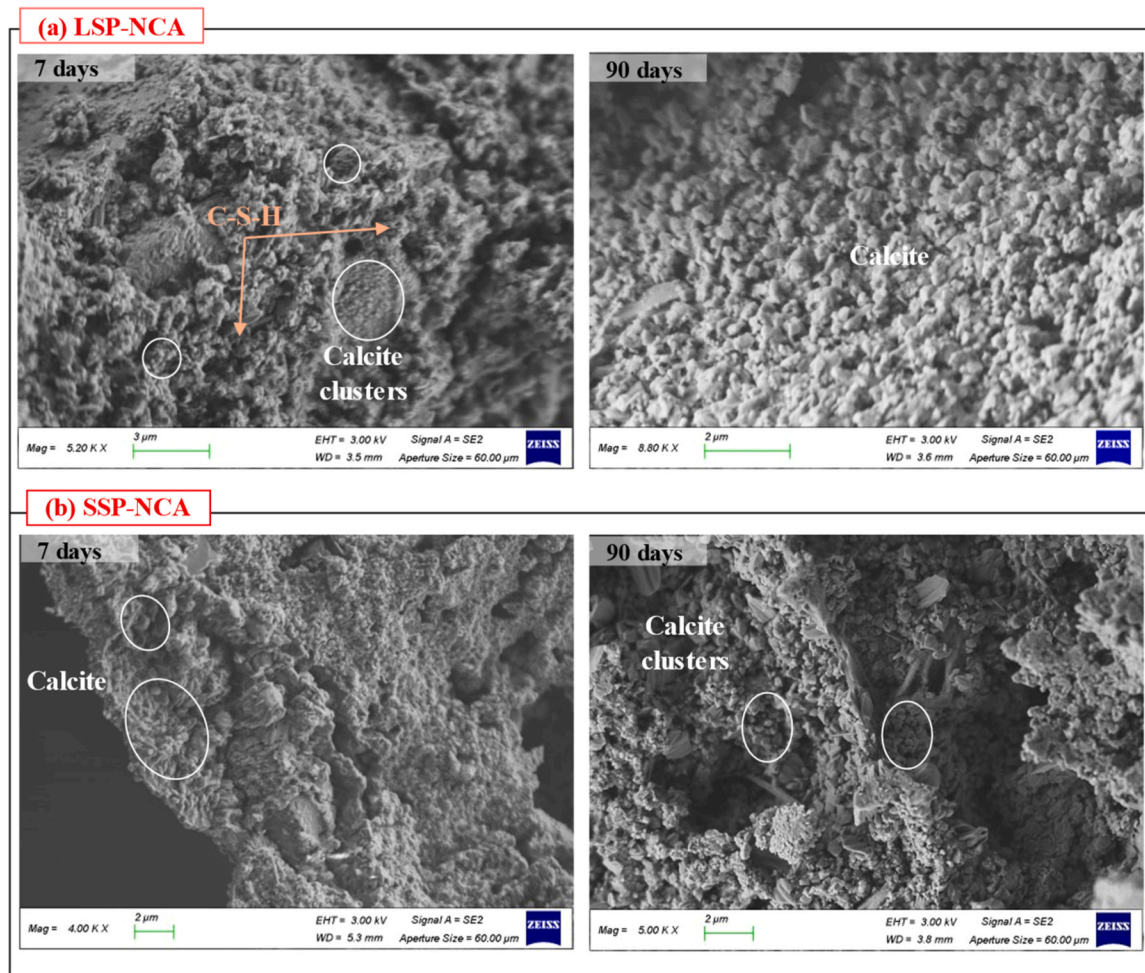


Fig. 20. SEM images showing the spatial distribution of CaCO_3 crystals within (a) LSP-NCA and (b) SSP-NCA specimens after different carbonation curing ages.

buffer the pore solution, delaying further pH decrease [124]. The delayed pH decrease in LSP-NCA is consistent with its higher $\text{Ca}(\text{OH})_2$ content, as confirmed by XRD. At later ages, the pH continues to decrease, reaching final values at 90 d of 8.92, 8.45, 9.22, and 8.69 for LSP-NCA, SSP-NCA, LSP-RCA, and SSP-RCA, respectively. At 28 d, Rietveld quantification indicates that no residual $\text{Ca}(\text{OH})_2$ remains in any of the samples.

The progressive decrease in pore solution pH observed across all specimens can be described as a two-stage process. In the initial stage, the pH begins to drop as CO_2 dissolves and CO_3^{2-} ions form by consuming OH^- from $\text{Ca}(\text{OH})_2$, resulting in an initial pH decrease from approximately 12.4 to around 10.6 across all specimens [145,146]. Once $\text{Ca}(\text{OH})_2$ is depleted at 28 d, a second stage begins in which continued CaCO_3 precipitation shifts the $\text{CO}_3^{2-}/\text{HCO}_3^-$ equilibrium, releasing H^+ and further lowering the pH. When the pH approaches approximately 8, CO_3^{2-} ions are no longer stable in solution, and the carbonation process effectively stops, as observed after 90 d in all specimens [23,145,147]. It is worth noting that such a decrease in pH, particularly in reinforced concrete, may compromise the stability of the passive film protecting steel reinforcement, thereby promoting the initiation of corrosion [148]. However, reinforced concrete applications are beyond the scope of the present study.

3.5. Scanning Electron Microscopy (SEM)

The microstructural observations from SEM images of carbonation-cured specimens, compared to their sealed-cured counterparts, are interpreted based on XRD results and morphological features reported in

the literature [50,149–151] (Fig. 18). All carbonation-cured specimens exhibit a denser microstructure, which evolves with the duration of carbonation curing, characterized by a substantial amount of CaCO_3 crystals, which are less prominent in the sealed-cured specimens (Fig. 18a) [113,135,152]. The three polymorphs detected in XRD analysis are visible in SEM images: spheroidal vaterite, rod-like aragonite crystallites, and calcite rhombs [113] (Fig. 18b, c, and d). Moreover, the ITZ undergoes significant refinement during carbonation curing across all specimens. For example, the ITZ thickness in SSP-RCA decreases from approximately 13 μm under sealed curing to about 2 μm after 90 d of carbonation curing (Fig. 19a and b). A similar trend is observed in SSP-NCA, where the ITZ thickness is only about 1.7 μm after 28 d of carbonation curing (Fig. 19d). In parallel, the adjacent mortar exhibits a noticeable reduction in pore volume. Pores appear as dark-contrast regions in the SEM images with low electron intensity, corresponding to voids or microcracks within the matrix. These features were identified qualitatively based on grayscale contrast differences and are shown only in selected images as representative examples to highlight the microstructural characteristics (Fig. 19c and d). These changes indicate an improved bond between the aggregate and the mortar matrix, resulting from the precipitation of CaCO_3 (calcite) within pores and microcracks of the ITZ, which progressively reduces its thickness and densifies the matrix [153].

Further insight is obtained by comparing the arrangement of CaCO_3 in SCC matrices. In comparison with the carbonation-cured SSP-NCA specimens, the spatial distribution of CaCO_3 crystals, particularly calcite, within the LSP-NCA matrix shows notable differences across curing ages (Fig. 20). From 7 d onward, the LSP-NCA specimens exhibit

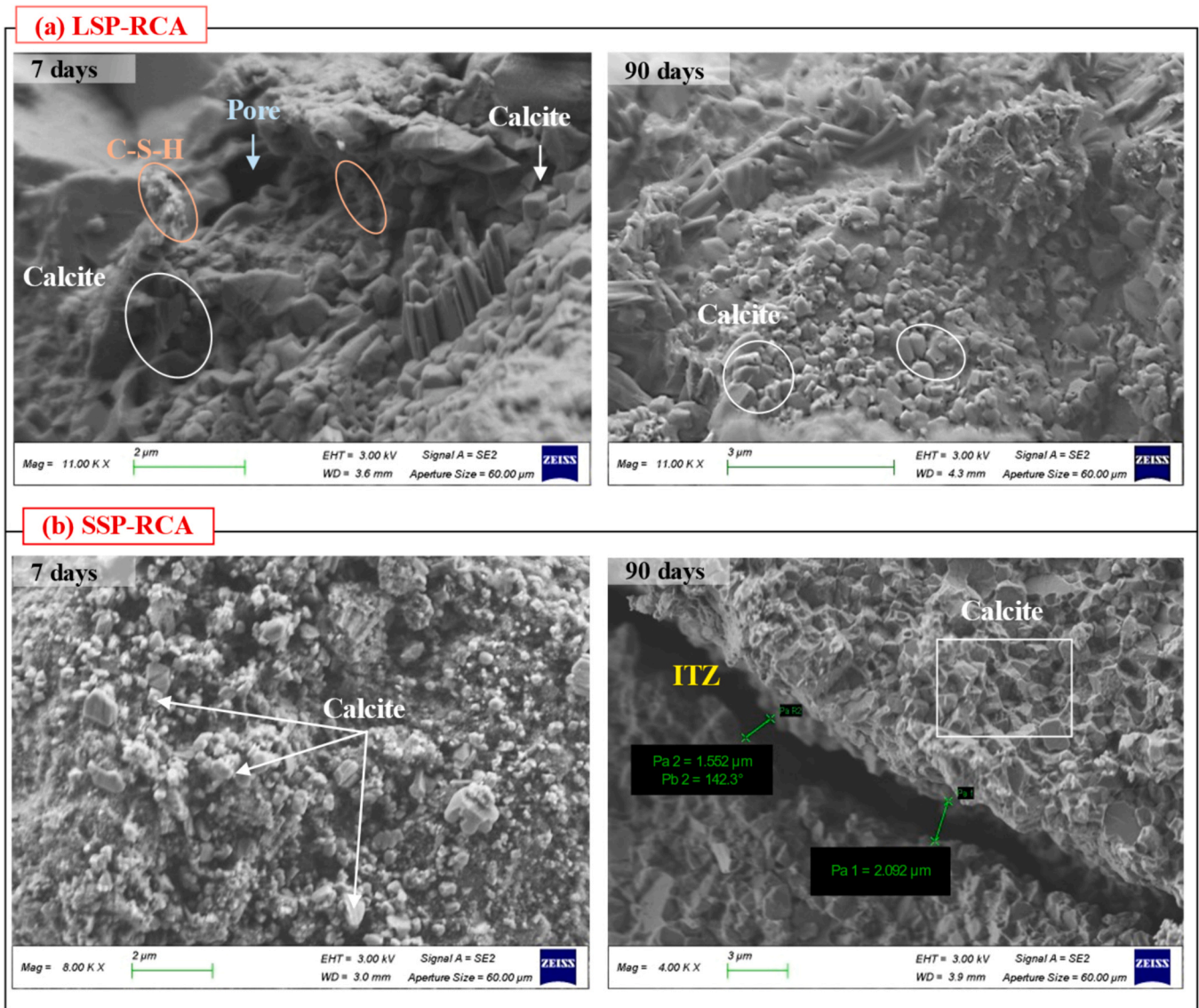


Fig. 21. SEM images showing the spatial distribution of CaCO_3 crystals within (a) LSP-RCA and (b) SSP-RCA specimens after different carbonation curing ages.

denser accumulations of finely dispersed calcite, often forming compact clusters predominantly located near the surfaces of the C–S–H gel (Fig. 20a). Individual calcite crystals are also observed either embedded in or on the C–S–H gel surface [153]. This distribution results in a strong bond between calcite crystals and the C–S–H gel [154]. Although calcite clusters are also present in SSP-NCA specimens, they tend to be less densely packed. The C–S–H gel is less discernible in these specimens, likely due to its greater consumption during the carbonation process (Fig. 20b). For RCA-based specimens, the morphology and size of the calcite crystals are more heterogeneous, with a more clustered distribution within the ITZ and a sparser, irregular dispersion in the bulk matrix (Fig. 21). Microcracks are also more prevalent and often partially unfilled, in contrast to NCA-based counterparts (Fig. 20).

The shape of calcite crystals changes over time as the chemical environment and pH evolve. On the first d, nanometer-sized rhombohedral crystals form (Fig. 22a) when rapid nucleation dominates. By around 7 d, the crystals grow into rhombo-scalenohedral (Fig. 22b) or scaleno-rhombohedral (Fig. 22c) as the pH drops to ~ 10 , allowing more directional growth. At later stages, scalenohedral crystals (Fig. 22d) appear under lower pH, where slower growth and reduced supersaturation favor the development of larger, elongated crystals. These observations align with previous studies [130,155,156] and are further

supported by the XRD analyses, which indicated calcite as the predominant carbonate phase in the carbonated samples.

4. Conclusions

The carbon storage capacity and carbonation behavior of self-compacting concrete (SCC) incorporating different precursors (LSP, SSP, NCA, and RCA) were investigated under controlled carbonation curing with regulated internal moisture. The main findings are summarized below.

- **CO_2 uptake:** Mass loss and Rietveld refinement indicated higher CO_2 uptake in RCA- and SSP-based specimens, with values increasing from 55 ± 9 and $46 \pm 3 \text{ kg/m}^3$ at 1 d in LSP-RCA and SSP-NCA, respectively, to 114 and 113 kg/m^3 of SCC at 90 d in LSP-RCA and SSP-RCA, based on the mass loss method. Rietveld refinement showed an average CaCO_3 content of 19 wt% at 1 d, rising to 25 wt% in RCA-based specimens at 90 d.
- **Carbonation depth:** Carbonation depth was not directly correlated with CO_2 uptake. For instance, in LSP-NCA specimens at 1 d of carbonation, a carbonation depth of 14 mm corresponded to 2.5% CO_2 uptake, whereas 17 mm corresponded to only 1.6%, based on

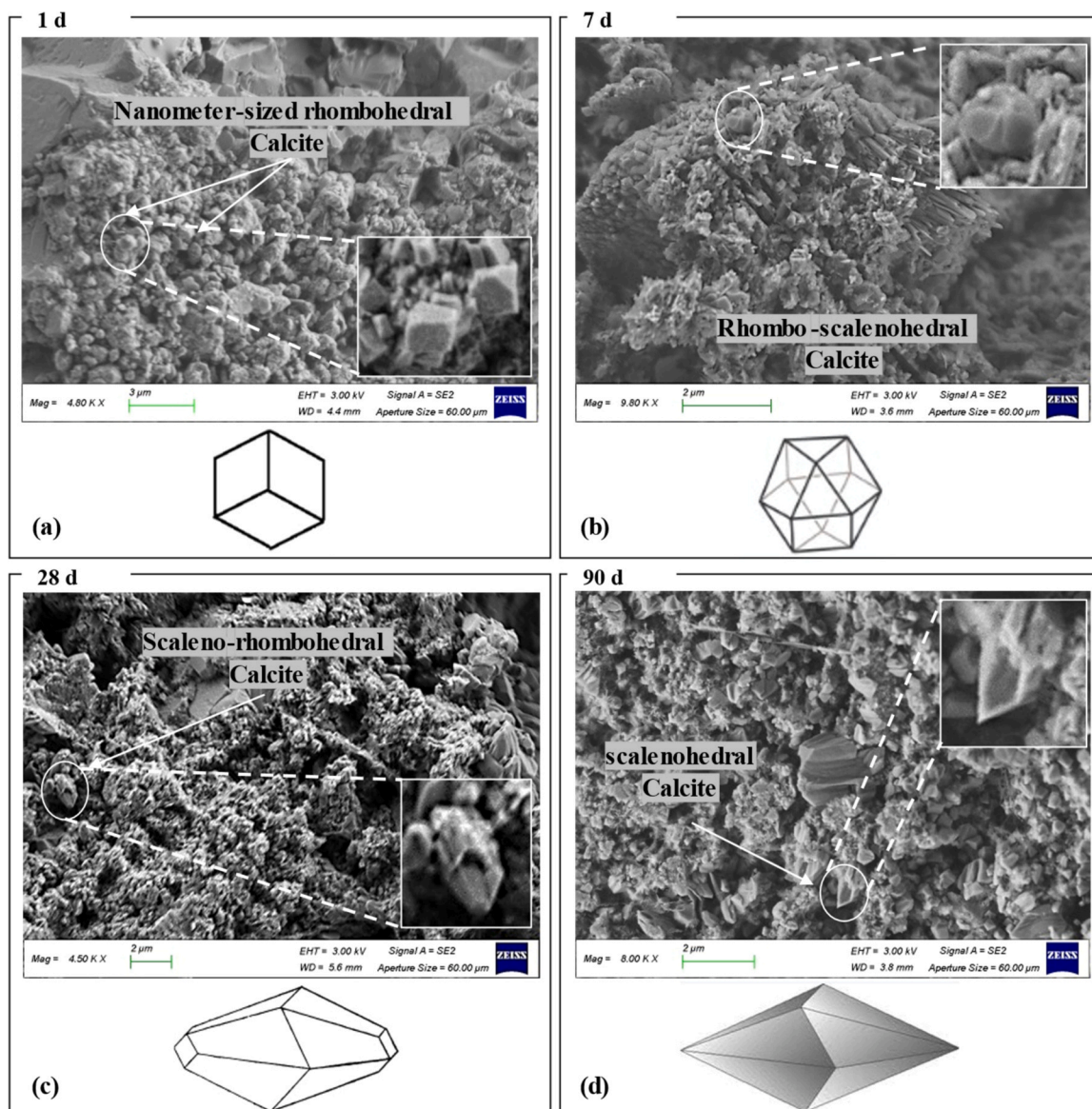


Fig. 22. SEM images showing the morphological evolution of CaCO_3 crystals during carbonation-induced pH drop.

one-dimensional diffusion and the mass loss method, indicating that deeper CO_2 penetration does not necessarily result in higher CaCO_3 formation or CO_2 sequestration.

- Microstructural evolution and phase development:** Carbonation curing progressively densified the SCC microstructure, promoting the formation and evolution of CaCO_3 polymorphs, amorphous- CaCO_3 , vaterite, aragonite, and calcite at early ages, which transformed mainly into calcite at later ages within the matrix and ITZ. RCA-based specimens exhibited more heterogeneous distributions and more microcracks than NCA-based counterparts. After 90 d of carbonation curing, carbonation was considered complete in all specimens, as CO_3^{2-} ions became unstable at a pH of approximately 8, limiting further carbonation.
- Chemical reactivity and physical effects of SSP and LSP:** Both SSP and LSP showed limited reactivity toward aluminate phases, as indicated by the low Hc content, resulting in a negligible chemical contribution to carbonation. However, the finer particle size and the rough, irregular morphology of SSP enhanced carbonation by increasing reactive surface area and providing additional nucleation sites for CaCO_3 precipitation. Carbonation kinetics remained strongly dependent on internal moisture availability.

Despite identical carbonation curing conditions, discrepancies among CO_2 quantification methods highlight the need for a multi-method approach or a refined calculation protocol to achieve a more reliable assessment of CO_2 uptake.

CRediT authorship contribution statement

Stéphanie Gascoin: Visualization, Validation. **Daniel Chateigner:** Writing – review & editing, Visualization, Validation, Supervision, Methodology, Investigation. **Fouzia Khadraoui:** Visualization, Validation, Supervision. **El Moustapha Bouha:** Writing – review & editing, Validation, Supervision, Methodology. **GUESSOUM Meriem:** Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis, Data curation, Conceptualization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

Data will be made available on request.

References

- [1] P. Guiraud, Fabrication du ciment et empreinte CO₂, Infociments. (2020). (<https://www.infociments.fr/enjeux-societe/la-fabrication-du-ciment-source-maitrisee-de-co2>) (accessed September 11, 2023).
- [2] Y. Li, D. Han, H. Wang, H. Lyu, D. Zou, Carbonation curing of mortars produced with reactivated cementitious materials for CO₂ sequestration, *J. Clean. Prod.* 383 (2023) 135501, <https://doi.org/10.1016/j.jclepro.2022.135501>.
- [3] B. El Moustapha, S. Bonnet, A. Khelidj, N. Leklou, N. Maranzana, I. Ahmedou Babah, Influence of microencapsulated phase change materials on drying shrinkage, pore structure, and gas permeability of blast-furnace slag and metakaolin-based geopolymers, *J. Sustain. Cem. Based Mater.* 14 (2025) 2424–2440, <https://doi.org/10.1080/21650373.2025.2532184>.
- [4] B. El Moustapha, F. Boukhelf, S. Gascoin, X. Larose, F. Khadraoui, D. Chateigner, Enhancing recycled aggregate-based geopolymer concrete: The effect of metakaolin on performance and microstructure, *Open. Ceram.* 25 (2026) 100931, <https://doi.org/10.1016/J.OCERAM.2026.100931>.
- [5] Chloride Diffusion in Geopolymers Containing Phase Change Materials, in: SP-362: ICCM2024, American Concrete Institute, 2024. <https://doi.org/10.14359/51742007>
- [6] L. Zhang, B. Liu, J. Du, C. Liu, H. Li, S. Wang, Internationalization trends of carbon emission linkages: A case study on the construction sector, *J. Clean. Prod.* 270 (2020) 122433, <https://doi.org/10.1016/j.jclepro.2020.122433>.
- [7] N. Thonemann, L. Zacharopoulos, F. Fromme, J. Nühlen, Environmental impacts of carbon capture and utilization by mineral carbonation: a systematic literature review and meta life cycle assessment, *J. Clean. Prod.* 332 (2022) 130067, <https://doi.org/10.1016/j.jclepro.2021.130067>.
- [8] T. Chen, M. Bai, X. Gao, Carbonation curing of cement mortars incorporating carbonated fly ash for performance improvement and CO₂ sequestration, *J. CO₂ Util.* 51 (2021) 101633, <https://doi.org/10.1016/j.jcou.2021.101633>.
- [9] J.-M. Torrenti, Résultats du projet national FastCarb: stockage de CO₂ par carbonatation accélérée du béton recyclé, in: NOMAD 2022-4e Conférence Internationale Francophone Nouveaux Matériaux et Durabilité, 2022.
- [10] M. Zajac, J. Skocek, M. Ben Haha, J. Deja, CO₂ mineralization methods in cement and concrete industry, *Energ. (Basel)* 15 (2022) 3597, <https://doi.org/10.3390/en15103597>.
- [11] C. Liang, B. Li, M.-Z. Guo, S. Hou, S. Wang, Y. Gao, X. Wang, Effects of early-age carbonation curing on the properties of cement-based materials: a review, *J. Build. Eng.* (2024) 108495, <https://doi.org/10.1016/j.jobe.2024.108495>.
- [12] A. Diemer, C.E. Nedelciu, M.E. Morales, C. Batisse, C. Cantuarias-Villesuzanne, Waste management and circular economy in the french building and construction sector, *Front. Sustain.* 3 (2022), <https://doi.org/10.3389/frsus.2022.840091>.
- [13] M.A. Mahi, F. Becquart, A. Sadok, T. Gaudron, R. Zentar, Self-compacting earth-based concretes with complete replacement of natural aggregates by excavated soils and recycled concrete aggregates, stabilised with low-carbon binders: a performance-based case study from the Grand Paris Express, *Case Stud. Constr. Mater.* 24 (2026) e06047, <https://doi.org/10.1016/J.CSCM.2026.E06047>.
- [14] MineralInfo, Careers in France, (<https://www.MineralInfo.Fr/Fr/Ressources-Minerales-France-Gestion/Carrieres-France>) (2020).
- [15] R. Fernández-Penas, C. Verdugo-Escamilla, C. Triunfo, S. Gärtner, A. D'Urso, F. Oltolina, A. Follenzi, G. Maoloni, H. Cölfen, G. Falini, J. Gómez-Morales, A sustainable one-pot method to transform seashell waste calcium carbonate to osteoinductive hydroxyapatite micro-nanoparticles, *J. Mater. Chem. B* 11 (2023) 7766–7777, <https://doi.org/10.1039/D3TB00856H>.
- [16] B. De Pascale, G. Tarsi, P. Tataranni, C. Sangiorgi, Potential application of waste bivalve shells as recycled filler in porous asphalt concrete through rheo-mechanical analysis, *Resour. Conserv. Recycl.* 209 (2024) 107830, <https://doi.org/10.1016/j.resconrec.2024.107830>.
- [17] M. Zelloufi, V. Ramos, B. El Haddaji, A. Bourguiba, G.K. Rollinson, J. Andersen, N. Sebaibi, M. Boutouil, Bio-colonisation, durability, and microstructural analysis of concrete incorporating magnetite and oyster shell waste aggregates in marine environment, *Constr. Build. Mater.* 456 (2024) 139269, <https://doi.org/10.1016/J.CONBUILDMAT.2024.139269>.
- [18] M. Zelloufi, V. Ramos, B. El Haddaji, A. Bourguiba, S. Broom-Fendley, G. K. Rollinson, A. Guedes, J. Andersen, N. Sebaibi, M. Boutouil, Assessment of bio-colonisation, durability and microstructure of concrete exposed to intertidal and subtidal marine environments, *Case Stud. Constr. Mater.* 23 (2025) e05119, <https://doi.org/10.1016/J.CSCM.2025.E05119>.
- [19] G. Hondroyiannis, E. Sardanou, V. Nikou, K. Evangelinos, I. Nikolaou, Recycling rate performance and socioeconomic determinants: evidence from aggregate and regional data across European Union countries, *J. Clean. Prod.* 434 (2024) 139877, <https://doi.org/10.1016/j.jclepro.2023.139877>.
- [20] M. Zelloufi, N. Sebaibi, J. Waligora, A. Elbarhoum, S. Pouget, Potential of scallop powder in producing low-carbon cement: experimental approach and environmental impact, *Constr. Build. Mater.* 498 (2025) 144003, <https://doi.org/10.1016/j.conbuildmat.2025.144003>.
- [21] N. Li, C. Unluier, Enhancement of the wet carbonation of artificial recycled concrete aggregates in seawater, *Cem. Concr. Res.* 175 (2024) 107387, <https://doi.org/10.1016/j.cemconres.2023.107387>.
- [22] C. Zhang, S. Liu, P. Tang, X. Guan, C. Shi, Enhancing the hardening properties and microstructure of magnesium slag blocks by carbonation-hydration sequential curing, *J. Build. Eng.* 76 (2023) 107414, <https://doi.org/10.1016/J.JOBE.2023.107414>.
- [23] D. Zhang, Y. Shao, Early age carbonation curing for precast reinforced concretes, *Constr. Build. Mater.* 113 (2016) 134–143, <https://doi.org/10.1016/j.conbuildmat.2016.03.048>.
- [24] D. Suescum-Morales, J.M. Fernández-Rodríguez, J.R. Jiménez, Use of carbonated water to improve the mechanical properties and reduce the carbon footprint of cement-based materials with recycled aggregates, *J. CO₂ Util.* 57 (2022) 101886, <https://doi.org/10.1016/j.jcou.2022.101886>.
- [25] D. Zhang, Y. Shao, Early age carbonation curing for precast reinforced concretes, *Constr. Build. Mater.* 113 (2016) 134–143, <https://doi.org/10.1016/j.conbuildmat.2016.03.048>.
- [26] Z. Liu, W. Meng, Fundamental understanding of carbonation curing and durability of carbonation-cured cement-based composites: a review, *J. CO₂ Util.* 44 (2021) 101428, <https://doi.org/10.1016/J.JCOU.2020.101428>.
- [27] Q.T. Phung, N. Maes, D. Jacques, E. Bruneel, I. Van Driessche, G. Ye, G. De Schutter, Effect of limestone fillers on microstructure and permeability due to carbonation of cement pastes under controlled CO₂ pressure conditions, *Constr. Build. Mater.* 82 (2015) 376–390, <https://doi.org/10.1016/j.conbuildmat.2015.02.093>.
- [28] D. Zhang, Z. Ghoulah, Y. Shao, Review on carbonation curing of cement-based materials, *J. CO₂ Util.* 21 (2017) 119–131, <https://doi.org/10.1016/j.jcou.2017.07.003>.
- [29] P.L. Domone, A review of the hardened mechanical properties of self-compacting concrete, *Cem. Concr. Compos.* 29 (2007) 1–12, <https://doi.org/10.1016/j.cemconcomp.2006.07.010>.
- [30] C. Shi, Y. Wu, Studies on some factors affecting CO₂ curing of lightweight concrete products, *Resour. Conserv. Recycl.* 52 (2008) 1087–1092, <https://doi.org/10.1016/j.resconrec.2008.05.002>.
- [31] M.F. Bertos, S.J.R. Simons, C.D. Hills, P.J. Carey, A review of accelerated carbonation technology in the treatment of cement-based materials and sequestration of CO₂, *J. Hazard. Mater.* 112 (2004) 193–205, <https://doi.org/10.1016/j.jhazmat.2004.04.019>.
- [32] P. He, C. Shi, Z. Tu, C.S. Poon, J. Zhang, Effect of further water curing on compressive strength and microstructure of CO₂-cured concrete, *Cem. Concr. Compos.* 72 (2016) 80–88, <https://doi.org/10.1016/j.cemconcomp.2016.05.026>.
- [33] L. Mo, F. Zhang, M. Deng, Mechanical performance and microstructure of the calcium carbonate binders produced by carbonating steel slag paste under CO₂ curing, *Cem. Concr. Res.* 88 (2016) 217–226, <https://doi.org/10.1016/j.cemconres.2016.05.013>.
- [34] D. Zhang, X. Cai, Y. Shao, Carbonation curing of precast fly ash concrete, *J. Mater. Civil. Eng.* 28 (2016), [https://doi.org/10.1061/\(ASCE\)MT.1943-5533.0001649](https://doi.org/10.1061/(ASCE)MT.1943-5533.0001649).
- [35] A. Neves Junior, R.D. Toledo Filho, E. de Moraes Rego Fairbairn, J. Dweck, The effects of the early carbonation curing on the mechanical and porosity properties of high initial strength Portland cement pastes, *Constr. Build. Mater.* 77 (2015) 448–454, <https://doi.org/10.1016/j.conbuildmat.2014.12.072>.
- [36] Z. Tu, M. Guo, C.S. Poon, C. Shi, Effects of limestone powder on CaCO₃ precipitation in CO₂ cured cement pastes, *Cem. Concr. Compos.* 72 (2016) 9–16, <https://doi.org/10.1016/j.cemconcomp.2016.05.019>.
- [37] M.F. Bertos, S.J.R. Simons, C.D. Hills, P.J. Carey, A review of accelerated carbonation technology in the treatment of cement-based materials and sequestration of CO₂, *J. Hazard. Mater.* 112 (2004) 193–205, <https://doi.org/10.1016/j.jhazmat.2004.04.019>.
- [38] D. Wang, T. Noguchi, T. Nozaki, Increasing efficiency of carbon dioxide sequestration through high temperature carbonation of cement-based materials, *J. Clean. Prod.* 238 (2019) 117980, <https://doi.org/10.1016/j.jclepro.2019.117980>.
- [39] I.-S. Yoon, O. Çopuroğlu, K.-B. Park, Effect of global climatic change on carbonation progress of concrete, *Atmos. Environ.* 41 (2007) 7274–7285, <https://doi.org/10.1016/j.atmosenv.2007.05.028>.
- [40] M. Guessoum, B. El Moustapha, F. Khadraoui, D. Chateigner, S. Gascoin, W. Maherzi, Effect of scallop shell powder addition on fresh and hardened properties of self-compacting concrete (SCC) based on recycled aggregates, *Constr. Build. Mater.* 495 (2025) 143660, <https://doi.org/10.1016/J.CONBUILDMAT.2025.143660>.

- [41] M.L. Basile, C. Triunfo, S. Gärtner, S. Fermani, D. Laurenzi, G. Maoloni, M. Mazzon, C. Marzadori, A. Adamiano, M. Iafisco, D. Montroni, J. Gómez Morales, H. Cölfen, G. Falini, Stearate-coated biogenic calcium carbonate from waste seashells: a sustainable plastic filler, *ACS Omega* 9 (2024) 11232–11242, <https://doi.org/10.1021/acsomega.3c06186>.
- [42] S. Yalcin, D. Chateigner, L. Le Pluart, S. Gascoin, S. Eve, Investigation of structural and mechanical properties of BioCaCO₃-LDPE composites, *Mater. Int.* 1 (2019) 29–43, <https://doi.org/10.33263/Materials12.029043>.
- [43] B. Felekoglu, Utilisation of high volumes of limestone quarry wastes in concrete industry (self-compacting concrete case), *Resour. Conserv. Recycl.* 51 (2007) 770–791, <https://doi.org/10.1016/j.resconrec.2006.12.004>.
- [44] M.K. Mohammed, A.R. Dawson, N.H. Thom, Production, microstructure and hydration of sustainable self-compacting concrete with different types of filler, *Constr. Build. Mater.* 49 (2013) 84–92, <https://doi.org/10.1016/j.conbuildmat.2013.07.107>.
- [45] H.T. Cam, N. Neithalath, Moisture and ionic transport in concretes containing coarse limestone powder, *Cem. Concr. Compos.* 32 (2010) 486–496, <https://doi.org/10.1016/j.cemconcomp.2010.04.002>.
- [46] P. Thongsanitgarn, W. Wongkeo, A. Chaipanich, C.S. Poon, Heat of hydration of Portland high-calcium fly ash cement incorporating limestone powder: effect of limestone particle size, *Constr. Build. Mater.* 66 (2014) 410–417, <https://doi.org/10.1016/j.conbuildmat.2014.05.060>.
- [47] R.-S. Lin, X.-Y. Wang, Effects of cement types and addition of quartz and limestone on the normal and carbonation curing of cement paste, *Constr. Build. Mater.* 305 (2021) 124799, <https://doi.org/10.1016/j.conbuildmat.2021.124799>.
- [48] D. Zhao, J.M. Williams, Z. Li, A.-H.A. Park, A. Radlinska, P. Hou, S. Kawashima, Hydration of cement pastes with calcium carbonate polymorphs, *Cem. Concr. Res.* 173 (2023) 107270, <https://doi.org/10.1016/j.cemconres.2023.107270>.
- [49] Y. Briki, M. Zajac, M.Ben Haha, K. Scrivener, Impact of limestone fineness on cement hydration at early age, *Cem. Concr. Res.* 147 (2021) 106515, <https://doi.org/10.1016/j.cemconres.2021.106515>.
- [50] X. Gu, B. Yang, Z. Li, B. Liu, J. Liu, Q. Wang, M.L. Nehdi, Elucidating the reaction of seashell powder within fly ash cement: A focus on hydration products, *Constr. Build. Mater.* 428 (2024) 136331, <https://doi.org/10.1016/j.conbuildmat.2024.136331>.
- [51] T. Danner, H. Justnes, M. Geiker, R.A. Lauten, Early hydration of C3A-gypsum pastes with Ca- and Na-lignosulfonate, *Cem. Concr. Res.* 79 (2016) 333–343, <https://doi.org/10.1016/j.cemconres.2015.10.008>.
- [52] C. Ma, B. Chen, L. Chen, Effect of organic matter on strength development of self-compacting earth-based construction stabilized with cement-based composites, *Constr. Build. Mater.* 123 (2016) 414–423, <https://doi.org/10.1016/j.conbuildmat.2016.07.018>.
- [53] S. Gowthaman, M. Chen, K. Nakashima, S. Kawasaki, Effect of scallop powder addition on MICP treatment of amorphous peat, *Front. Environ. Sci.* 9 (2021), <https://doi.org/10.3389/fenvs.2021.690376>.
- [54] M. Zhou, S. Chen, H. Chao, Y. Ma, Z. Li, X. Ouyang, Impact of the aragonite-to-calcite transition in bio-fillers on cement hydration, *J. Build. Eng.* 117 (2026) 114885, <https://doi.org/10.1016/j.jobe.2025.114885>.
- [55] W. Zuo, X. Zhao, J. Luo, Z. He, J. Zhao, Investigating the impact of carbonation curing concentrations on the mechanical properties and microstructure of recycled concrete, *J. Build. Eng.* 96 (2024) 110465, <https://doi.org/10.1016/J.JOBE.2024.110465>.
- [56] C. Liang, N. Lu, H. Ma, Z. Ma, Z. Duan, Carbonation behavior of recycled concrete with CO₂-curing recycled aggregate under various environments, *J. CO₂ Util.* 39 (2020) 101185, <https://doi.org/10.1016/j.jcou.2020.101185>.
- [57] T. Kim, J. Olek, Effects of sample preparation and interpretation of thermogravimetric curves on calcium hydroxide in hydrated pastes and mortars, *Transp. Res. Rec. J. Transp. Res. Board.* 2290 (2012) 10–18, <https://doi.org/10.3141/2290-02>.
- [58] M.-Y. Xuan, H.-K. Cho, X.-Y. Wang, Performance improvement of waste oyster-shell powder-cement binary system via carbonation curing, *J. Build. Eng.* 70 (2023) 106336, <https://doi.org/10.1016/j.jobe.2023.106336>.
- [59] D. Xuan, B. Zhan, C.S. Poon, A maturity approach to estimate compressive strength development of CO₂-cured concrete blocks, *Cem. Concr. Compos.* 85 (2018) 153–160, <https://doi.org/10.1016/j.cemconcomp.2017.10.005>.
- [60] N. Kim, S. Choi, I.T. Amr, B.A. Fadhel, J. Park, J. Seo, H.K. Lee, Synergistic effects of CO₂ mixing and CO₂ curing in CO₂ uptake capacity of Portland cement, *Constr. Build. Mater.* 448 (2024) 138217, <https://doi.org/10.1016/j.conbuildmat.2024.138217>.
- [61] M. Sakoparnig, I. Galan, B. Müller, F. Mittermayr, J. Juhart, C. Grengg, Optical pH imaging reveals what phenolphthalein conceals – A carbonation study on blended cements, *Cem. Concr. Res.* 186 (2024) 107681, <https://doi.org/10.1016/j.cemconres.2024.107681>.
- [62] L. Mo, D.K. Panesar, Effects of accelerated carbonation on the microstructure of Portland cement pastes containing reactive MgO, *Cem. Concr. Res.* 42 (2012) 769–777, <https://doi.org/10.1016/j.cemconres.2012.02.017>.
- [63] G. Villain, M. Thiery, G. Platret, Measurement methods of carbonation profiles in concrete: thermogravimetry, chemical analysis and gammadensimetry, *Cem. Concr. Res.* 37 (2007) 1182–1192, <https://doi.org/10.1016/j.cemconres.2007.04.015>.
- [64] T. Matschei, S. Braymand, N. Garg, B. Huet, D. Jansen, R.J. Myers, B. Scruggs, E. Bernard, J. Bertola, B. Cazacliu, L. Ellwood, C. Felten, G. Ferrara, E. Garboczi, A. Gholizadeh Vayghan, H. Hafez, T. Hanein, F. Kanavaris, M. Mahoutian, I. Maruyama, S. Roux, P. Turcry, J. Skibsted, R. Snellings, S. Srivastava, V. Vial, J. Wei, M. Zajac, Z. Zhang, P. Suraneni, Test methods for CO₂ mineralization in cement-based materials: a review by RILEM TC 309-MCP, *Mater. Struct.* 59 (2026) 175, <https://doi.org/10.1617/s11527-026-03064-x>.
- [65] A.V. Radha, T.Z. Forbes, C.E. Killian, P.U.P.A. Gilbert, A. Navrotsky, Transformation and crystallization energetics of synthetic and biogenic amorphous calcium carbonate, *Proc. Natl. Acad. Sci.* 107 (2010) 16438–16443, <https://doi.org/10.1073/pnas.1009959107>.
- [66] D. Chateigner, S. Ouhenia, C. Krauss, C. Hedegaard, O. Gil, M. Morales, L. Lutterotti, M. Rousseau, E. Lopez, Voyaging around nacre with the X-ray shuttle: from bio-mineralisation to prosthetics via mollusc phylogeny, *Mater. Sci. Eng. A* 528 (2010) 37–51, <https://doi.org/10.1016/j.msea.2010.07.032>.
- [67] B. Pokroy, J.S. Fieramosca, R.B. Von Dreelle, A.N. Fitch, E.N. Caspi, E. Zolotoyabko, Atomic Structure of Biogenic Aragonite, *Chem. Mater.* 19 (2007) 3244–3251, <https://doi.org/10.1021/cm070187u>.
- [68] S. Ouhenia, D. Chateigner, M.A. Belkhir, E. Guillemeau, Microstructure and crystallographic texture of Charonia lampas lampas shell, *J. Struct. Biol.* 163 (2008) 175–184, <https://doi.org/10.1016/J.JSB.2008.05.005>.
- [69] S. Grazulis, D. Chateigner, R.T. Downs, A.F.T. Yokochi, M. Quirós, L. Lutterotti, E. Manakova, J. Butkus, P. Moeck, A. Le Bail, Crystallography Open Database – an open-access collection of crystal structures, *J. Appl. Crystallogr.* 42 (2009) 726–729, <https://doi.org/10.1107/S0021889809016690>.
- [70] N.Ap.M. Beltrame, R.L. Dias, F.B. Witzke, R.A. Medeiros-Junior, Effect of carbonation curing on the physical, mechanical, and microstructural properties of metakaolin-based geopolymer concrete, *Constr. Build. Mater.* 406 (2023) 133403, <https://doi.org/10.1016/j.conbuildmat.2023.133403>.
- [71] W. Li, H. Wang, Z. Liu, N. Li, S. Zhao, S. Hu, Steel slag accelerated carbonation curing for high-carbonation precast concrete development, *Materials* 17 (2024) 2968, <https://doi.org/10.3390/ma17122968>.
- [72] Q. Zhang, Q. Zeng, D.C. Zheng, J. Wang, S.L. Xu, Oven drying kinetics and status of cement-based porous materials for in-lab microstructure investigation, *Adv. Cem. Res.* 30 (2018) 204–215, <https://doi.org/10.1680/jadcr.17.00075>.
- [73] V. Rostami, Y. Shao, A.J. Boyd, Z. He, Microstructure of cement paste subject to early carbonation curing, *Cem. Concr. Res.* 42 (2012) 186–193, <https://doi.org/10.1016/J.CEMCONRES.2011.09.010>.
- [74] D. Zhang, Y. Shao, Effect of early carbonation curing on chloride penetration and weathering carbonation in concrete, *Constr. Build. Mater.* 123 (2016) 516–526, <https://doi.org/10.1016/j.conbuildmat.2016.07.041>.
- [75] F. Ren, C. Zhou, Z. Zhang, C.H. Dreimol, U. Angst, Effect of accelerated carbonation on long-term water absorption behavior of cement-based materials, *Mater. Struct.* 58 (2025) 16, <https://doi.org/10.1617/s11527-024-02533-5>.
- [76] L.P. Singh, A. Goel, S.K. Bhattacharyya, S. Ahalawat, U. Sharma, G. Mishra, Effect of Morphology and Dispersibility of Silica Nanoparticles on the Mechanical Behaviour of Cement Mortar, *Int. J. Concr. Struct. Mater.* 9 (2015) 207–217, <https://doi.org/10.1007/s40069-015-0099-2>.
- [77] OSCARS | Fostering the uptake of Open Science in Europe. (<https://oscars-project.eu/>) (accessed October 21, 2025).
- [78] Program MAUD, Available from: <http://www.ing.unitn.it/maudyesqui>.
- [79] L. Lutterotti, D. Chateigner, S. Ferrari, J. Ricote, Texture, residual stress and structural analysis of thin films using a combined X-ray analysis, *Thin Solid. Films* 450 (2004) 34–41, <https://doi.org/10.1016/J.TSF.2003.10.150>.
- [80] N.C. Popa, The (hkl) dependence of diffraction-line broadening caused by strain and size for all laue groups in rietveld refinement, *J. Appl. Crystallogr.* 31 (1998) 176–180, <https://doi.org/10.1107/S0021889897007995>.
- [81] A. Le Bail, Modelling the silica glass structure by the Rietveld method, *J. Non Cryst. Solids* 183 (1995) 39–42, [https://doi.org/10.1016/0022-3093\(94\)00664-4](https://doi.org/10.1016/0022-3093(94)00664-4).
- [82] L. Li, J. Nam, W.H. Hartt, Ex situ leaching measurement of concrete alkalinity, *Cem. Concr. Res.* 35 (2005) 277–283, <https://doi.org/10.1016/J.CEMCONRES.2004.04.024>.
- [83] J.M. de Burgh, S.J. Foster, Influence of temperature on water vapour sorption isotherms and kinetics of hardened cement paste and concrete, *Cem. Concr. Res.* 92 (2017) 37–55, <https://doi.org/10.1016/j.cemconres.2016.11.006>.
- [84] C. Hall, Anomalous diffusion in unsaturated flow: fact or fiction? *Cem. Concr. Res.* 37 (2007) 378–385, <https://doi.org/10.1016/j.cemconres.2006.10.004>.
- [85] O. Oueslati, J. Duchesne, The effect of SCMs and curing time on resistance of mortars subjected to organic acids, *Cem. Concr. Res.* 42 (2012) 205–214, <https://doi.org/10.1016/J.CEMCONRES.2011.09.017>.
- [86] S. Ge, Y. Pan, L. Zheng, X. Xie, Effects of organic matter components and incubation on the cement-based stabilization/solidification characteristics of lead-contaminated soil, *Chemosphere* 260 (2020) 127646, <https://doi.org/10.1016/J.CHEMOSPHERE.2020.127646>.
- [87] E. Lipczynska-Kochany, Humic substances, their microbial interactions and effects on biological transformations of organic pollutants in water and soil: a review, *Chemosphere* 202 (2018) 420–437, <https://doi.org/10.1016/J.CHEMOSPHERE.2018.03.104>.
- [88] G. Magnabosco, D. Giuri, A.P. Di Bisceglie, F. Scarpino, S. Fermani, C. Tomasini, G. Falini, New material perspective for waste seashells by covalent functionalization, *ACS Sustain. Chem. Eng.* 9 (2021) 6203–6208, https://doi.org/10.1021/ACSUSCHEMENG.1C01306/ASSET/IMAGES/LARGE/SC1C01306_0005.JPEG.
- [89] J. Liu, K. Ma, J. Shen, J. Zhu, G. Long, Y. Xie, B. Liu, Influence of recycled concrete aggregate enhancement methods on the change of microstructure of ITZs in recycled aggregate concrete, *Constr. Build. Mater.* 371 (2023) 130772, <https://doi.org/10.1016/j.conbuildmat.2023.130772>.
- [90] T. Ikeda, S. Yamane, A. Sakamoto, Strengths of concrete containing recycled concrete aggregate, in: *Demolition Reuse Conc Mason V2*, CRC Press, 2023, pp. 585–594.

- [91] S. von Greve-Dierfeld, B. Lothenbach, A. Vollpracht, B. Wu, B. Huet, C. Andrade, C. Medina, C. Thiel, E. Gruyaert, H. Vanoutrive, Understanding the carbonation of concrete with supplementary cementitious materials: a critical review by RILEM TC 281-CCC, *Mater. Struct.* 53 (2020) 136, <https://doi.org/10.1617/s11527-020-01558-w>.
- [92] L. Cheng, R. Kurihara, Z. Yang, T. Ohkubo, R. Kitagaki, A. Teramoto, Y. Suda, I. Maruyama, Proposal of a new pretreatment of accelerated carbonation for evaluating material potential, *Cem. Concr. Compos.* 171 (2026) 106633, <https://doi.org/10.1016/j.cemconcomp.2026.106633>.
- [93] H.-W. Song, S.-J. Kwon, K.-J. Byun, C.-K. Park, Predicting carbonation in early-aged cracked concrete, *Cem. Concr. Res.* 36 (2006) 979–989, <https://doi.org/10.1016/j.cemconres.2005.12.019>.
- [94] J.J. Thomas, J.J. Chen, A.J. Allen, H.M. Jennings, Effects of decalcification on the microstructure and surface area of cement and tricalcium silicate pastes, *Cem. Concr. Res.* 34 (2004) 2297–2307, <https://doi.org/10.1016/j.cemconres.2004.04.007>.
- [95] K. De Weert, M. Ben Haha, G. Le Saout, K.O. Kjellsen, H. Justnes, B. Lothenbach, Hydration mechanisms of ternary Portland cements containing limestone powder and fly ash, *Cem. Concr. Res.* 41 (2011) 279–291, <https://doi.org/10.1016/j.cemconres.2010.11.014>.
- [96] J.G. Jang, G.M. Kim, H.J. Kim, H.-K. Lee, Review on recent advances in CO₂ utilization and sequestration technologies in cement-based materials, *Constr. Build. Mater.* 127 (2016) 762–773, <https://doi.org/10.1016/j.conbuildmat.2016.10.017>.
- [97] R. Mi, K.M. Liew, G. Pan, New insights into diffusion and reaction of CO₂ gas in recycled aggregate concrete, *Cem. Concr. Compos.* 129 (2022) 104486, <https://doi.org/10.1016/j.cemconcomp.2022.104486>.
- [98] Z. Shao, Y. Sakai, Low-carbon construction material from waste concrete powder through compaction and carbonation: Effect of powder size, *Constr. Build. Mater.* 514 (2026) 145555, <https://doi.org/10.1016/j.conbuildmat.2026.145555>.
- [99] B. Šavija, M. Luković, Carbonation of cement paste: Understanding, challenges, and opportunities, *Constr. Build. Mater.* 117 (2016) 285–301, <https://doi.org/10.1016/j.conbuildmat.2016.04.138>.
- [100] S. Kashaf-Haghighi, S. Ghoshal, Physico-chemical processes limiting CO₂ uptake in concrete during accelerated carbonation curing, *Ind. Eng. Chem. Res.* 52 (2013) 5529–5537, <https://doi.org/10.1021/ie303275e>.
- [101] B. Li, Y. Tian, G. Zhang, Y. Liu, H. Feng, N. Jin, X. Jin, H. Wu, Y. Shao, D. Yan, Z. Zhou, S. Wang, Z. Zhang, J. Chen, X. Chen, Y. Lu, X. Li, J. Wang, Comparison of detection methods for carbonation depth of concrete, *Sci. Rep.* 13 (2023) 19980, <https://doi.org/10.1038/s41598-023-47443-8>.
- [102] J. Han, W. Sun, G. Pan, W. Caihui, Monitoring the evolution of accelerated carbonation of hardened cement pastes by X-ray computed tomography, *J. Mater. Civil. Eng.* 25 (2013) 347–354, [https://doi.org/10.1061/\(ASCE\)MT.1943-5533.0000610](https://doi.org/10.1061/(ASCE)MT.1943-5533.0000610).
- [103] Y. Wang, Y. Wang, R. Kitagaki, D. Oh, H. Takahashi, K. Shibuya, T. Ohkubo, A. Teramoto, Y. Suda, I. Maruyama, Spectral characterization and carbonation depth visualization of concrete using hyperspectral imaging, *Cem. Concr. Compos.* 168 (2026) 106498, <https://doi.org/10.1016/j.cemconcomp.2026.106498>.
- [104] F. Winnefeld, A. Leemann, A. German, B. Lothenbach, Multi-modal imaging of the cementitious carbonation front: Evidence for pore refinement, *J. CO₂ Util.* 90 (2024) 102993, <https://doi.org/10.1016/j.cogsc.2022.100672>.
- [105] H. Bao, M. Yu, Y. Liu, J. Ye, Experimental and statistical study on the irregularity of carbonation depth of cement mortar under supercritical condition, *Constr. Build. Mater.* 174 (2018) 47–59, <https://doi.org/10.1016/j.conbuildmat.2018.04.100>.
- [106] P.H.R. Borges, J.O. Costa, N.B. Milestone, C.J. Lynsdale, R.E. Streatfield, Carbonation of CH and C–S–H in composite cement pastes containing high amounts of BFS, *Cem. Concr. Res.* 40 (2010) 284–292, <https://doi.org/10.1016/j.cemconres.2009.10.020>.
- [107] Y.-S. Wang, S.-H. Tae, R.-S. Lin, X.-Y. Wang, Effects of Na₂CO₃ on engineering properties of cement–limestone powder–slag ternary blends, *J. Build. Eng.* 57 (2022) 104937, <https://doi.org/10.1016/j.jobe.2022.104937>.
- [108] B.A. Tayeh, M.W. Hasaniyah, A.M. Zeyad, M.M. Awad, A. Alaskar, A. M. Mohamed, R. Alyousef, Durability and mechanical properties of seashell partially-replaced cement, *J. Build. Eng.* 31 (2020) 101328, <https://doi.org/10.1016/j.jobe.2020.101328>.
- [109] X. Liu, P. Feng, Y. Cai, X. Yu, C. Yu, Q. Ran, Carbonation behavior of calcium silicate hydrate (CSH): Its potential for CO₂ capture, *Chem. Eng. J.* 431 (2022) 134243, <https://doi.org/10.1016/j.cej.2021.134243>.
- [110] W. Ashraf, J. Olek, S. Sahu, Phase evolution and strength development during carbonation of low-lime calcium silicate cement (CSC), *Constr. Build. Mater.* 210 (2019) 473–482, <https://doi.org/10.1016/j.conbuildmat.2019.03.038>.
- [111] J. Liu, C. Zeng, Z. Li, G. Liu, W. Zhang, G. Xie, F. Xing, Carbonation of steel slag at low CO₂ concentrations: Novel biochar cold-bonded steel slag artificial aggregates, *Sci. Total. Environ.* 902 (2023) 166065, <https://doi.org/10.1016/J.SCI.TOTENV.2023.166065>.
- [112] X. Xian, Y. Shao, Microstructure of cement paste subject to ambient pressure carbonation curing, *Constr. Build. Mater.* 296 (2021) 123652, <https://doi.org/10.1016/j.conbuildmat.2021.123652>.
- [113] B. Lu, S. Drissi, J. Liu, X. Hu, B. Song, C. Shi, Effect of temperature on CO₂ curing, compressive strength and microstructure of cement paste, *Cem. Concr. Res.* 157 (2022) 106827, <https://doi.org/10.1016/j.cemconres.2022.106827>.
- [114] I. Galan, F.P. Glasser, D. Baza, C. Andrade, Assessment of the protective effect of carbonation on portlandite crystals, *Cem. Concr. Res.* 74 (2015) 68–77, <https://doi.org/10.1016/j.cemconres.2015.04.001>.
- [115] E. Ruiz-Agudo, K. Kudlacz, C.V. Putnis, A. Putnis, C. Rodriguez-Navarro, Dissolution and carbonation of portlandite (Ca (OH)₂) single crystals, *Environ. Sci. Technol.* 47 (2013) 11342–11349, <https://doi.org/10.1021/es402061c>.
- [116] D.R. Moorehead, Cementation by the carbonation of hydrated lime, *Cem. Concr. Res.* 16 (1986) 700–708, [https://doi.org/10.1016/0008-8846\(86\)90044-X](https://doi.org/10.1016/0008-8846(86)90044-X).
- [117] M. Liu, S. Hong, Y. Wang, J. Zhang, D. Hou, B. Dong, Compositions and microstructures of hardened cement paste with carbonation curing and further water curing, *Constr. Build. Mater.* 267 (2021) 121724, <https://doi.org/10.1016/J.CONBUILDMAT.2020.121724>.
- [118] R. Hay, B. Peng, K. Celik, Filler effects of CaCO₃ polymorphs derived from limestone and seashell on hydration and carbonation of reactive magnesium oxide (MgO) cement (RMC), *Cem. Concr. Res.* 164 (2023) 107040, <https://doi.org/10.1016/j.cemconres.2022.107040>.
- [119] Z. Shao, Z. Fang, Y. Sakai, Enhancement of compacted hardened cement paste powder through carbonation: evolution of CO₂ diffusion, phase assemblage and pore structure, *Case Stud. Constr. Mater.* 23 (2025) e05293, <https://doi.org/10.1016/j.cscm.2025.e05293>.
- [120] J.H. Seo, S.M. Park, H.K. Lee, Evolution of the binder gel in carbonation-cured Portland cement in an acidic medium, *Cem. Concr. Res.* 109 (2018) 81–89, <https://doi.org/10.1016/j.cemconres.2018.03.014>.
- [121] T. Chen, X. Gao, Effect of carbonation curing regime on strength and microstructure of Portland cement paste, *J. CO₂ Util.* 34 (2019) 74–86, <https://doi.org/10.1016/j.jcou.2019.05.034>.
- [122] B. Lu, Z. Huo, Q. Xu, G. Hou, X. Wang, J. Liu, X. Hu, Characteristics of CSH under carbonation and its effects on the hydration and microstructure of cement paste, *Constr. Build. Mater.* 364 (2023) 129952, <https://doi.org/10.1016/j.conbuildmat.2022.129952>.
- [123] D. Xuan, B. Zhan, C.S. Poon, A maturity approach to estimate compressive strength development of CO₂-cured concrete blocks, *Cem. Concr. Compos.* 85 (2018) 153–160, <https://doi.org/10.1016/j.cemconcomp.2017.10.005>.
- [124] S.K. Saikia, A.S. Rajput, Effect of carbon sequestration methods on uptake potential and characteristics of ordinary portland cement-based concrete, *Constr. Build. Mater.* 418 (2024) 135330, <https://doi.org/10.1016/J.CONBUILDMAT.2024.135330>.
- [125] S. Ahmad, R.A. Assagaf, M. Maslehuddin, O.S.B. Al-Amoudi, S.K. Adekunle, S. I. Ali, Effects of carbonation pressure and duration on strength evolution of concrete subjected to accelerated carbonation curing, *Constr. Build. Mater.* 136 (2017) 565–573, <https://doi.org/10.1016/J.CONBUILDMAT.2017.01.069>.
- [126] W. bin Yuan, Y. Zheng, N. ting Yu, Y. Shen, X.K. Hao, Comparative analysis of time-dependent CO₂ uptake in cement and geopolymers concretes via diffusion-induced carbonation, *Case Stud. Constr. Mater.* 22 (2025) e04573, <https://doi.org/10.1016/J.CSCM.2025.E04573>.
- [127] S.A. Yaseen, G.A. Yiseen, Z. Li, Elucidation of calcite structure of calcium carbonate formation based on hydrated cement mixed with graphene oxide and reduced graphene oxide, *ACS Omega* 4 (2019) 10160–10170, <https://doi.org/10.1021/acsomega.9b00042>.
- [128] L. Black, C. Breen, J. Yarwood, K. Garbev, P. Stemmermann, B. Gasharova, Structural Features of C–S–H(I) and Its Carbonation in Air—A Raman Spectroscopic Study. Part II: Carbonated Phases, *J. Am. Ceram. Soc.* 90 (2007) 908–917, <https://doi.org/10.1111/j.1551-2916.2006.01429.x>.
- [129] C. Rodriguez-Navarro, K. Kudlacz, Ö. Cizer, E. Ruiz-Agudo, Formation of amorphous calcium carbonate and its transformation into mesostructured calcite, *CrystEngComm* 17 (2015) 58–72, <https://doi.org/10.1039/C4CE01562B>.
- [130] Ö. Cizer, C. Rodriguez-Navarro, E. Ruiz-Agudo, J. Elsen, D. Van Gemert, K. Van Balen, Phase and morphology evolution of calcium carbonate precipitated by carbonation of hydrated lime, *J. Mater. Sci.* 47 (2012) 6151–6165, <https://doi.org/10.1007/s10853-012-6535-7>.
- [131] H. El-Hassan, Y. Shao, Z. Ghouleh, Reaction products in carbonation-cured lightweight concrete, *J. Mater. Civil. Eng.* 25 (2013) 799–809, [https://doi.org/10.1061/\(ASCE\)MT.1943-5533.0000638](https://doi.org/10.1061/(ASCE)MT.1943-5533.0000638).
- [132] O. Shtepenko, C. Hills, A. Brough, M. Thomas, The effect of carbon dioxide on β -dicalcium silicate and Portland cement, *Chem. Eng. J.* 118 (2006) 107–118, <https://doi.org/10.1016/J.CEJ.2006.02.005>.
- [133] Z. Xiao, J. Zhang, X. Hu, A. Khakhoussi, P. He, X. You, X. Su, W. Chen, C. Shi, An overview on polymorphs of calcium carbonate formed during carbon mineralization of cementitious materials, *J. Sustain. Cem. Based Mater.* (2025) 1–23, <https://doi.org/10.1080/21650373.2025.2471820>.
- [134] M. Castellote, C. Andrade, X. Turrillas, J. Campo, G.J. Cuello, Accelerated carbonation of cement pastes in situ monitored by neutron diffraction, *Cem. Concr. Res.* 38 (2008) 1365–1373, <https://doi.org/10.1016/J.CEMCONRES.2008.07.002>.
- [135] V. Shah, K. Scrivener, B. Bhattacharjee, S. Bishnoi, Changes in microstructure characteristics of cement paste on carbonation, *Cem. Concr. Res.* 109 (2018) 184–197, <https://doi.org/10.1016/j.cemconres.2018.04.016>.
- [136] D.C. Green, J. Ihli, P.D. Thornton, M.A. Holden, B. Marzec, Y.-Y. Kim, A.N. Kulak, M.A. Levenstein, C. Tang, C. Lynch, S.E.D. Webb, C.J. Tynan, F.C. Meldrum, 3D visualization of additive occlusion and tunable full-spectrum fluorescence in calcite, *Nat. Commun.* 7 (2016) 13524, <https://doi.org/10.1038/ncomms13524>.
- [137] Y.-Q. Niu, J.-H. Liu, C. Aymonier, S. Feriani, D. Kralj, G. Falini, C.-H. Zhou, Calcium carbonate: controlled synthesis, surface functionalization, and nanostructured materials, *Chem. Soc. Rev.* 51 (2022) 7883–7943, <https://doi.org/10.1039/D1CS00519G>.
- [138] L.S. Gomez-Villalba, P. López-Arce, M. Alvarez de Buergo, R. Fort, Atomic Defects and Their Relationship to Aragonite–Calcite Transformation in Portlandite Nanocrystal Carbonation, *Cryst. Growth Des.* 12 (2012) 4844–4852, <https://doi.org/10.1021/cg300628m>.

- [139] Ö. Cizer, C. Rodríguez-Navarro, E. Ruiz-Agudo, J. Elsen, D. Van Gemert, K. Van Balen, Phase and morphology evolution of calcium carbonate precipitated by carbonation of hydrated lime, *J. Mater. Sci.* 47 (2012) 6151–6165, <https://doi.org/10.1007/s10853-012-6535-7>.
- [140] J. Ibáñez, L. Artús, R. Cuscó, Á. López, E. Menéndez, M.C. Andrade, Hydration and carbonation of monoclinic C_2S and C_3S studied by Raman spectroscopy, *J. Raman Spectrosc.* 38 (2007) 61–67, <https://doi.org/10.1002/JRS.1599>.
- [141] B.J. Zhan, D.X. Xuan, C.S. Poon, C.J. Shi, Effect of curing parameters on CO_2 curing of concrete blocks containing recycled aggregates, *Cem. Concr. Compos.* 71 (2016) 122–130, <https://doi.org/10.1016/j.cemconcomp.2016.05.002>.
- [142] K.C. Reddy, N.S. Melaku, S. Park, Thermodynamic Modeling Study of Carbonation of Portland Cement, 5060, *Materials* 2022 15 (15) (2022) 5060, <https://doi.org/10.3390/MA15145060>.
- [143] M. Takasaki, Y. Kezuka, M. Tajika, Y. Oaki, H. Imai, Evolution of Calcite Nanocrystals through Oriented Attachment and Fragmentation: Multistep Pathway Involving Bottom-Up and Break-Down Stages, *ACS Omega* 2 (2017) 8997–9001, <https://doi.org/10.1021/acsomega.7b01487>.
- [144] L. Qin, X. Gao, T. Chen, Influence of mineral admixtures on carbonation curing of cement paste, *Constr. Build. Mater.* 212 (2019) 653–662, <https://doi.org/10.1016/j.conbuildmat.2019.04.033>.
- [145] J.H.M. Visser, Influence of the carbon dioxide concentration on the resistance to carbonation of concrete, *Constr. Build. Mater.* 67 (2014) 8–13, <https://doi.org/10.1016/J.CONBUILDMAT.2013.11.005>.
- [146] T. Nakagaki, I. Tanaka, Y. Furukawa, H. Sato, Y. Yamanaka, Experimental Evaluation of Effect of Oxidative Degradation of Aqueous Monoethanolamine on Heat of CO_2 Absorption, Vapor Liquid Equilibrium and CO_2 Absorption Rate, *Energy Procedia* 63 (2014) 2384–2393, <https://doi.org/10.1016/J.EGYPRO.2014.11.260>.
- [147] K. Maekawa, T. Ishida, T. Kishi, Multi-scale Modeling of Concrete Performance Integrated Material and Structural Mechanics, *J. Adv. Concr. Technol.* 1 (2003) 91–126, <https://doi.org/10.3151/jact.1.91>.
- [148] N. Singh, S.P. Singh, Carbonation and electrical resistance of self compacting concrete made with recycled concrete aggregates and metakaolin, *Constr. Build. Mater.* 121 (2016) 400–409, <https://doi.org/10.1016/J.CONBUILDMAT.2016.06.009>.
- [149] N.O. Emekli, M. Saydan, A. Ünal, Effect of hydration products on the mechanical behavior of reinforced concrete slabs made with ternary systems, *J. Build. Eng.* 106 (2025) 112625, <https://doi.org/10.1016/j.job.2025.112625>.
- [150] X. Feng, J. Li, Q. Chen, J. Xu, G. Chen, Y. Sun, Z. Jiang, H. Zhu, Effect of flowing water on the ettringite-induced healing of concrete cracks by electrodeposition: Experiments and molecular dynamics simulations, *Constr. Build. Mater.* 443 (2024) 137718, <https://doi.org/10.1016/j.conbuildmat.2024.137718>.
- [151] Y. Chen, F. Al-Neshawy, J. Punkki, Investigation on the effect of entrained air on pore structure in hardened concrete using MIP, *Constr. Build. Mater.* 292 (2021) 123441, <https://doi.org/10.1016/j.conbuildmat.2021.123441>.
- [152] H.K. Kim, S.J. Park, J.I. Han, H.K. Lee, Microbially mediated calcium carbonate precipitation on normal and lightweight concrete, *Constr. Build. Mater.* 38 (2013) 1073–1082, <https://doi.org/10.1016/J.CONBUILDMAT.2012.07.040>.
- [153] B. Guo, G. Chu, R. Yu, Y. Wang, Q. Yu, D. Niu, Effects of sufficient carbonation on the strength and microstructure of CO_2 -cured concrete, *J. Build. Eng.* 76 (2023) 107311, <https://doi.org/10.1016/J.JOBE.2023.107311>.
- [154] B.J. Zhan, D.X. Xuan, C.S. Poon, C.J. Shi, Mechanism for rapid hardening of cement pastes under coupled CO_2 -water curing regime, *Cem. Concr. Compos.* 97 (2019) 78–88, <https://doi.org/10.1016/J.CEMCONCOMP.2018.12.021>.
- [155] J. García, Carmona, J. Gómez Morales, R. Rodríguez, Clemente, Rhombohedral–scalenohedral calcite transition produced by adjusting the solution electrical conductivity in the system $Ca(OH)_2$ – CO_2 – H_2O , *J. Colloid Interface Sci.* 261 (2003) 434–440, [https://doi.org/10.1016/S0021-9797\(03\)00149-8](https://doi.org/10.1016/S0021-9797(03)00149-8).
- [156] F. Liendo, M. Arduino, F.A. Deorsola, S. Bensaid, Factors controlling and influencing polymorphism, morphology and size of calcium carbonate synthesized through the carbonation route: a review, *Powder Technol.* 398 (2022) 117050, <https://doi.org/10.1016/J.POWTEC.2021.117050>.