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Influence of Exposure Conditions on the Carbonation and Durability of ISSA-Modified Cementitious Materials

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***Abstract* – This study explores how exposure conditions affect the carbonation process of cementitious materials that include incinerated sewage sludge ash (ISSA), to evaluate ISSA's potential as a sustainable additive in cement-based construction materials and its role in CO_2 sequestration. Hardened cement pastes with different amounts of ISSA (0 %, 20 %, 25 %, 30 %, and 35 %) were subjected to two carbonation experiments: indoor and outdoor in a natural environment. The carbonation process was tracked using phenolphthalein tests, scanning electron microscopy (SEM-EDS), X-ray diffraction (XRD), and calcium carbonate content measurement. Results show that ISSA-modified cement pastes have increased carbonation potential, especially at 20–25 % ISSA, with a rise in $CaCO_3$ formation and diverse calcite morphologies. However, higher ISSA levels (>30 %) led to decreased carbonation efficiency. The outdoor experiment revealed the significant influence of temperature and humidity changes on carbonation rates, resulting in slower CO_2 absorption compared to laboratory conditions.**

***Keywords* – CO_2 emission; hardened cement paste; mineral carbonation; sewage sludge incineration ash.**

1. INTRODUCTION

Concrete, a fundamental material in modern construction, is essential for applications ranging from residential buildings to road bases and filling materials. The main component in concrete is cement, with Portland cement being the most commonly used type [1]. In 2019, global cement production reached 4.1 gigatonnes (Gt) [2], and this figure is expected to increase significantly due to population growth and rising infrastructure demands [3]. The construction industry faces major challenges in reducing carbon dioxide (CO_2) emissions and finding sustainable solutions. Traditional cement production is a significant source of CO_2 with approximately 5–8 % of global anthropogenic emissions [4]. According to [4] and [5] around 20% of this CO_2 can be reabsorbed through carbonation during the usage phase of cement-based products. Interestingly, the guidelines of the Intergovernmental Panel on

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Climate Change (IPCC) for national greenhouse gas inventories outline methods to measure CO_2 emissions from cement production but do not consider the carbon absorption from cement materials through carbonation. Furthermore, the rate at which carbonating cement sequesters carbon is increasing rapidly, with an average annual growth rate of 5.8 % observed between 1990 and 2013, driven by the expansion of cement used in buildings and infrastructure [6].

However, the process of mineral carbonation in cementitious systems is highly complex. While calcite ($CaCO_3$) is the dominant phase formed during carbonation, other polymorphs like aragonite and vaterite, as well as amorphous carbonate phases, may also develop under certain conditions [7]. Additionally, carbonation interacts with hydration products such as portlandite and calcium silicate hydrates (C-S-H), leading to changes in pore structure, phase assemblage, and mechanical properties [8], [9]. These processes are influenced by the internal Ca/Si ratio of cementitious materials, air humidity, and temperature [10], [11], as well as the porosity and permeability of the hardened paste, which collectively control CO_2 uptake and mineral transformation.

Accelerated carbonation tests are often employed to study these mechanisms under controlled conditions, as they allow researchers to observe the kinetics and dynamics of the process faster and more directly [12]–[14]. However, such tests may not fully replicate the behaviour of carbonation under ambient conditions. Natural carbonation rates are largely controlled by external exposure conditions, including air temperature and relative humidity, the volume and duration of water contact with the concrete surface, and the partial pressure of CO_2 [15], [16]. Xi *et al*. [6] estimated that global cement stocks sequester around 1 billion tons of atmospheric CO_2 yearly.

The total amount of CO_2 that can be bound by cement, also known as the binding or buffering capacity, depends directly on the amount of calcium oxide (CaO) available to form calcium carbonate ($CaCO_3$), partially dependent on the Ca/Si ratio. A low Ca/Si ratio leads to decreased concrete strength due to the reduced concentration of Ca^{2+} in the matrix, while an excessively high ratio may adversely affect the mechanical properties and durability of the concrete [17]. The carbonation mechanism in cementitious systems containing additives differs significantly from that in cement without any additives or supplementary materials due to variations in phase assemblage evolution, pore structure, and pore solution chemistry [15]. Additives such as fly ash or metallurgical slag introduce diverse mineralogical and chemical phases that influence both carbonation kinetics and the nature of carbonation products. For example, calcium silicate hydrates (C-S-H) in Portland cement may undergo decalcification during carbonation, forming amorphous silica and crystalline $CaCO_3$ phases like calcite, vaterite, and aragonite [7], [8]. In systems with mineral additives, these reactions can be modified by the incorporation of aluminosilicates, sulfates, or trace metals, which alter the solution chemistry and favour the formation of secondary phases such as ettringite and hydrotalcite-like structures [18].

The porosity and pore size distribution of cementitious systems are also strongly affected by mineral additives, with some additives promoting finer pore structures that can inhibit CO_2 ingress and slow carbonation rates [19], [20].

Mineral carbonation has also been tested on various alkaline industrial wastes [21], [22], such as steel-making slags [23], [24], municipal solid waste fly ashes [25], municipal solid waste bottom ashes [26], and air pollution control residues (APC) [27]. Carbonated industrial wastes can partially replace clinker or concrete aggregate, indirectly reducing the use of Portland cement and thereby lowering CO_2 emissions from the cement industry [28]. While the concept of mineral carbonation in cementitious materials has gained traction, the addition

of waste incineration ashes to the cement-water mix for mineral carbonation remains a less-explored area, necessitating detailed fundamental studies.

The goal of this study was to assess the effectiveness of CO_2 sequestration through passive carbonation and evaluate the potential for incinerated sewage sludge ash (ISSA) to serve as a sustainable additive in cement production supporting the circular economy by waste utilization. The presence of ISSA introduces additional complexity to the carbonation process, influencing not only the formation of carbonate phases but also the overall characteristic. ISSA, rich in calcium and other alkaline components, may enhance the binding capacity for CO_2 while simultaneously altering hydration reactions and pore structure. Conversely, ISSA, rich in free lime and reactive aluminosilicates, may accelerate carbonation through increased CO_2-binding capacity, while also contributing to the formation of less stable phases like vaterite under certain conditions. Recent studies by Du *et al*. [29] highlight the growing practice of evaluating alternative (waste-derived) binder/mortar systems by combining performance testing with environmental assessment (e.g., life-cycle perspective), underlining the importance of linking technical behaviour with sustainability benefits.

Incorporating ISSA into cement production aligns with the principles of the circular economy by repurposing waste materials, reducing landfill dependency, and enhancing resource efficiency. This direction is consistent with the trend to develop building materials through the utilisation of manufacturing waste streams, supporting circular pathways in the construction sector [30]. By studying these processes, this research contributes to developing more sustainable construction practices while addressing global CO_2 emissions.

2. MATERIALS AND METHODS

2.1. The Incinerated Sewage Sludge Ash (ISSA)

The incinerated sewage sludge ash (ISSA) is classified as non-hazardous waste in accordance with the European List of Waste (Commission Decision 2000/532/EC and Annex III to Directive 2008/98/EC). It has a waste code 19 01 14 meaning it does not contain hazardous substances. The ash was collected from an incineration plant operating within a wastewater treatment facility serving a city of 800 000 inhabitants. The incineration process involved sludge dried to 36 % dry mass in a fluidized bed boiler (Pyrofluid™) at temperatures ranging from 850 to 900 °C. The ISSA is Si–Fe–Ca–P–Al rich material with small concentrations of Na, K, Ti, Mg Mn, C_{tot}, and S_{tot} with elevated concentrations of As, Co, Cr, Cu, Mn, Mo, Ni, Pb and Zn [31].

2.2. Preparation of Hardened Cement Paste with ISSA

To evaluate the effectiveness of CO_2 uptake, hardened cement blocks containing different ratios of ISSA were prepared (Fig. 1). A water-to-cement (w/c) ratio of 0.8 was selected to ensure sufficient workability and homogeneity, particularly at higher ISSA contents. ISSA is a lightweight, porous material with irregular particle morphology, which tends to increase water demand and reduce mix cohesion [32]. The elevated w/c ratio allowed for uniform dispersion of ISSA within the paste, minimizing clumping or segregation and facilitating reproducible casting. While this ratio is not representative of structural concrete, it was appropriate for the study's focus on carbonation behaviour, leaching, and mineral transformation rather than mechanical performance. The paste was mechanically mixed for 5 minutes, and ISSA was added in proportions ranging from 20 % to 35 % of the dry cement weight. The chosen range was consistent with ISSA usage in durability and reactivity studies, especially for exploratory or environmental applications. Several studies explore 10–40 %

substitution to evaluate performance thresholds [33]. A visible layering or tonal variation can be seen in the specimens (Fig. 1), as darker areas near the surface. These features are likely due to natural density and textural differences between the cement and the ISSA particles. ISSA, being relatively light and porous, may exhibit mild visual segregation during casting, especially in the surface layer. This behaviour was also observed by [32], who noted that ISSA's low density and irregular morphology can occasionally lead to visual artifacts on the surface of cementitious mixes without affecting overall paste uniformity. For comparison purposes, a control sample containing 0 % ISSA (pure cement-water block) was prepared to assess the effectiveness of the carbonation process. After mechanical blending of the mixtures to achieve homogeneity they were poured into cylindrical plastic moulds with a diameter of 10 cm. This geometry was chosen based on common laboratory practice for paste and mortar studies, especially when targeting durability analysis, leaching behaviour, and microstructural characterization (e.g., SEM, XRD) rather than mechanical performance. Although not standardized by ASTM or EN for carbonation testing specifically, similar mold sizes are used in durability-related investigations due to their manageable size, consistent exposure area, and compatibility with analytical equipment [34]. After casting, the samples were gently vibrated to minimize air voids and covered with plastic sheets to prevent evaporation. All samples were then cured under sealed, moist conditions at ambient temperature (~22 °C) for 28 days, allowing for sufficient hydration and strength development.

Fig. 1. Hardened cement blocks with varying proportions of incineration sewage sludge ash (ISSA).

2.3. Experimental Design

Following curing, the specimens were demoulded and cut into blocks of 1.5 cm × 5 cm for the carbonation experiments with atmospheric CO_2. Two experiments were performed to study the carbonation process under different exposure conditions: one in controlled laboratory conditions and another in an outdoor natural environment.

The first experiment ("laboratory experiment") was conducted under controlled laboratory conditions, with a constant temperature of 22 ºC and a relative humidity (RH) of approximately 45 %. A "wetting and drying" method was employed to simulate real exposure conditions, facilitating cycles of CO_2 dissolution and diffusion, but the CO_2 concentration remained at natural indoor levels (~400–420 ppm). While no standardized duration is prescribed for natural carbonation studies, the entire experimental cycle lasted 50 days (from November 23, 2023, to January 11, 2024). This duration was selected to reflect medium-term exposure under ambient atmospheric conditions, where the CO_2 concentration was not artificially increased. Under such conditions atmospheric carbonation proceeds slowly and requires extended exposure times to achieve measurable changes in the material [32]. Previous research has employed similar or longer exposure durations in natural or semi-natural conditions to evaluate carbonation progression [13]. The 50-day period allowed sufficient time to observe $CaCO_3$ formation, mass evolution, and microstructural changes in the ISSA-modified cement pastes. As the experiment was conducted under ambient laboratory

conditions, drying occurred naturally at room temperature (~22 °C, ~45 % RH) after each saturation. The cement blocks with different ISSA proportions were repeatedly saturated with 500 ml of deionized water in a porcelain evaporating dish and then allowed to dry completely at room temperature. Seven complete wetting and drying cycles occurred over the 50-day lab period. After each drying phase, the samples were weighed and tested using phenolphthalein to assess the progress of carbonate formation before repeating the cycle.

The second experiment ("outdoor experiment") was conducted under ambient environmental conditions. The samples were placed in holders and exposed to weather variations for 62 days (late winter – early-to-mid spring; from March 6 to May 6, 2024). Their weight was measured at intervals every two to three days, with occasional longer gaps of up to six days. Temperature and humidity sensors, positioned at the height of the samples, recorded measurements every 10 minutes, enabling continuous monitoring of these parameters. Mean daily temperatures varied significantly ranging from 1.9 ºC to 20.6 ºC, while observed values peaked at 29.9 ºC and dropped as low as –4.3 ºC. Mean daily relative humidity also fluctuated considerably, between 46.3 % and 94.3 %, with momentary values peaking near saturation at 96.6 % and dropping to 25.6 %.

2.4. Analytical Methods

The elemental composition of the ISSA was analysed using inductively coupled plasma mass spectrometry (ICP-MS) and inductively coupled plasma atomic emission spectroscopy (ICP-AES) at Bureau Veritas Minerals in Vancouver, Canada. These analyses employed the LF2000 and AQ200 analytical sets, enabling precise determination of major and trace element concentrations in the raw ash samples. The ICP process was based on fusion techniques with a lithium borate/tetraborate mixture followed by nitric acid digestion to ensure the complete decomposition of the refractory matrices, allowing for the determination of total element concentrations. A 20 g portion of each sample was ground into a powder using an agate mill prior to analyses. C_{tot} and S_{tot} were measured by LECO combustion analysis, while LOI was determined by thermal methods at Bureau Veritas Minerals in Vancouver, Canada.

X-ray diffraction (XRD) analyses were conducted to analyse the phase composition of hardened cement paste with incorporated ISSA before and after the mineral carbonation experiment. The XRD analysis was performed on bulk powdered samples with a Rigaku Miniflex powder diffractometer using CuKα radiation, operating at 40 kV and 15 mA. Samples were analysed in the 2θ range of 2° to 62°, with a step width of 0.02°. Before analysis, samples were ground to a fine powder. The diffractograms were interpreted using the BGMN/Profex Rietveld program.

Hitachi S-4700 scanning electron microscope (SEM) with field emission, equipped with a NORAN energy dispersion spectrometer (EDS) was used to validate carbonate precipitation on the surface of the cement-ash blocks and the advancement of this process. Analyses were conducted at the accelerating voltage of 20 kV and a counting time of 100 s. Samples were carbon-coated to enhance conductivity using a carbon sputtering device with a turbomolecular pump before analysis.

The efficiency of the mineral carbonation process was assessed by determining the calcium carbonate content using the Scheibler volumetric method. This method quantifies the $CaCO_3$ formed during carbonation by treating samples with 4 mol/L hydrochloric acid and measuring the volume of CO_2 gas evolved in a closed system. Measurements were performed using the Eijkelkamp analytical tool, compliant with ISO 10693 standards.

To assess the relationship between atmospheric conditions and the carbonation process in the outdoor experiment, air temperature and relative humidity were measured using a HOBO U23 Pro v2 External Temperature/Relative Humidity Data Logger. The sensor was placed

40 cm above the ground, and measurements were recorded every 10 minutes throughout the entire experiment. Additionally, daily rainfall data were used to examine the natural wetting and drying cycles of the samples. Since the outdoor experiment was conducted in the university's meteorological garden, it was possible to use rainfall data directly from the experiment site.

3. RESULTS AND DISCUSSION

3.1. ISSA Chemical and Mineralogical Characterization

The elemental composition of ISSA determined by ICP-MS and ICP-AES reveals significant concentrations of key elements that influence mineral carbonation and cementitious properties. As shown in Fig. 2 (Panel A), ISSA consists predominantly of Si (17.58 %), Fe (9.91 %), Al (4.37 %), Ca (8.36 %), and Mg (2.15 %). These elements are recognized for their potential to influence pozzolanic activity and mechanical performance in cementitious materials [32]. For clarity, Fig. 2 is presented in three panels that show major elements content, potentially toxic metals content, and carbonation-relevant parameters (S_{tot}, C_{tot} and LOI).

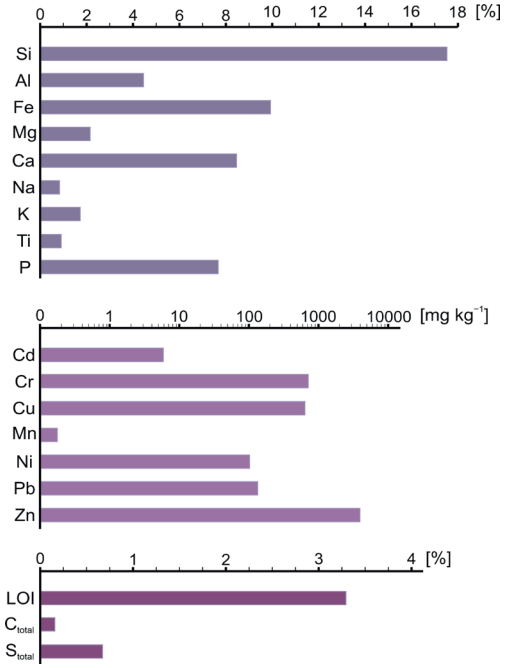

Fig. 2. Elemental composition of ISSA (major elements, potentially toxic elements and LOI, S_{tot} and C_{tot}).

These components may affect the reactivity of the binder system, including potential pozzolanic contributions and the availability of Ca-bearing phases for subsequent carbonate formation. Si, Al, and Fe play critical roles in enhancing the formation of calcium silicate hydrate (C-S-H) gel, a process known to improve the mechanical properties of concrete [35]. The presence of Ca and Mg in ISSA, essential for mineral carbonation reactions, further supports its potential utility in enhancing cementitious durability.

However, the Ca/Si ratio of 0.475 in ISSA may indicate insufficient calcium availability, potentially resulting in slower or incomplete hydration and weaker concrete as well as limited calcium availability for carbonation. This could affect both the carbon sequestration potential and the long-term durability of the studied material.

Additionally, [36] identified crystalline phases in ISSA, including quartz, feldspar, muscovite, hematite, Ca-Fe-Mg rich whitlockite, and Fe-PO_4. These phases may contribute to ISSA's overall reactivity and performance in cementitious applications, influencing both pozzolanic activity and potential mineral carbonation reactions.

3.2. Carbonation Indicators (Phenolphthalein Test and Macroscopic Observations)

Carbonation progression in hardened cement pastes was evaluated using phenolphthalein staining. The indicator response showed clear differences in carbonation depth between mixes and exposure conditions as seen in Fig. 3. In general, carbonation effects were most pronounced in the near-surface zone, consistent with the diffusion-controlled nature of CO_2 ingress into cementitious materials. Macroscopic observations after exposure confirmed visible changes associated with carbonation and weathering, and the comparison between laboratory exposure and outdoor exposure indicated that environmental variability can influence the spatial extent and uniformity of carbonation (Fig. 4).

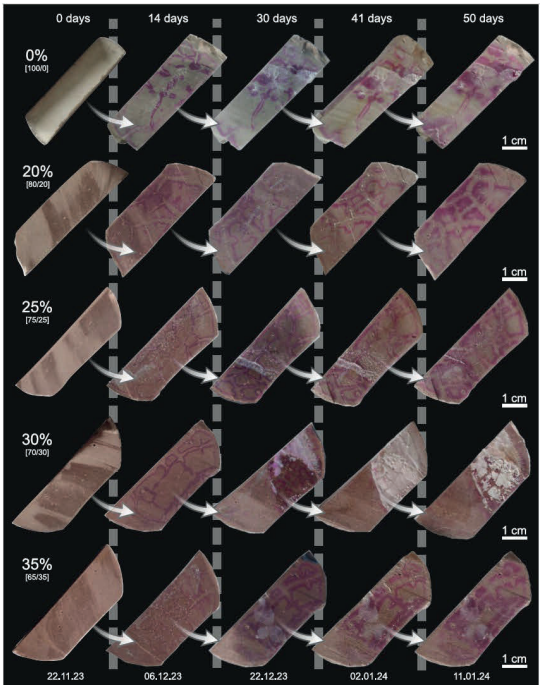

Fig. 3. Hardened cement pastes with different ISSA proportions (0 %, 20 %, 25 %, 30 % and 35 %) before and during the laboratory experiment. The advancement of mineral carbonation was tested with phenolphthalein, which turns purple in carbonated areas.

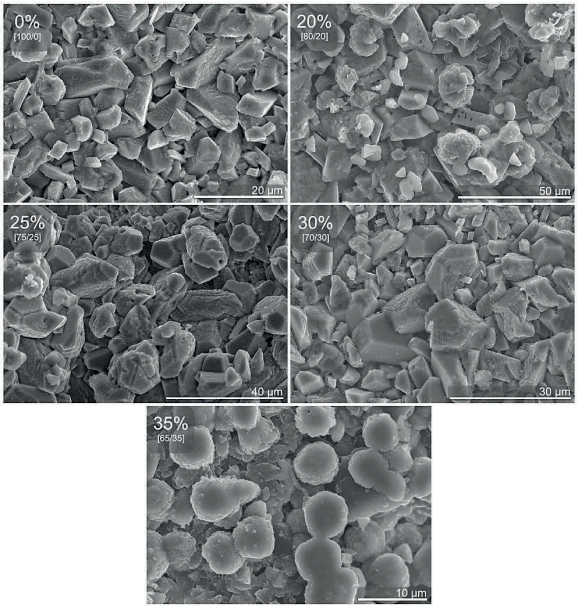

Fig. 4. SEM micrographs showing the morphology of calcium carbonate crystals formed on the surface of hardened cement pastes containing different proportions of ISSA (0 %, 20 %, 25 %, 30 %, and 35 %) after 50 days of natural carbonation under laboratory conditions. The images highlight variations in crystal shapes (e.g., rhombohedral, spherical, and irregular forms) associated with ISSA-induced changes in pore chemistry and nucleation behaviour. Morphological diversity reflects the influence of ISSA composition on carbonate precipitation, not structural differences between samples.

These preliminary results suggest that the chosen experimental parameters were sufficient to enable the dissolution of CO_2 without excessively filling the pores, allowing for effective diffusion of CO_2 gas into the cement matrix. The conditions facilitated a balanced environment where moisture levels supported the formation of carbonic acid and subsequent carbonation reactions, while the pores remained adequately open for CO_2 diffusion.

The crystalline morphology of calcite exhibited diverse forms including cubes, prisms, spherical aggregates, and irregular shapes. As suggested by Lu *et al*. [37], variations in $CaCO_3$ crystal structure are associated with crystallization processes, which are highly dependent on carbonation environments. Since all samples in our study were exposed to the same experimental conditions, differences in calcium carbonate morphology resulted from the varying ISSA content. The incorporation of such additives can alter the nucleation and growth of calcite crystals, leading to differences in size, shape, and overall structure.

It is important to emphasize that the SEM images are presented for the purpose of morphological illustration only. They were taken from representative surface regions at the end of the carbonation experiment and were not intended for quantitative or spatially resolved comparison. Rather, the goal was to highlight how varying ISSA content influences carbonate crystal formation under identical environmental conditions. This approach is consistent with similar morphological studies in cement-based carbonation research [37], [38].

The chemical complexity of raw materials significantly affects the crystal morphology and microstructure of carbonated materials. For instance, Mg, K, and SO_3 present in cement and fillers, can influence the crystallization process of calcite during carbonation. Acting as impurities or additives, these elements alter the nucleation and growth dynamics of calcite crystals [38]. Consequently, variations in crystal shapes and microstructural characteristics

arise due to the intricate interactions between these elements and the carbonation environment.

XRD analysis confirmed the presence of key cement minerals including portlandite, ettringite, gypsum, and calcite, as well as primary ISSA-derived minerals such as quartz, hematite, and whitlockite. After the mineral carbonation, a relative increase in calcite content was observed, along with the appearance of small peaks indicative of magnesite. The persistence of portlandite indicates that carbonation did not fully consume $Ca(OH)_2$ within the experiment duration, which is consistent with diffusion-controlled atmospheric carbonation. During atmospheric ageing, carbonation converts portlandite into calcite according to $Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O$, which is consistent with the phase trends observed in cementitious matrices during long-term exposure [39].

Both the laboratory and the outdoor experiments demonstrated that mineral carbonation was most efficient in samples containing 20 % and 25 % of ISSA (Table 1). This suggests that ISSA contents in the 20–25 % range may be a practical target for non-structural cementitious products where CO_2 uptake and durability-related microstructural densification are desired. At higher concentrations (30 % and 35 %) ISSA exceeded the optimal dosage required for effective catalysis or nucleation in the carbonation process. This excess additive likely leads to overcrowding of nucleation sites or inhibits crystal growth, thereby reducing or slowing down the efficiency of carbonate mineral formation. Additionally, the presence of other elements in ISSA, such as P, K and S, at higher concentrations could potentially interfere with the nucleation and growth of carbonate minerals. These elements may form complexes or precipitates that compete with or inhibit the formation of calcite crystals during carbonation. K, often in soluble forms, can enhance pore solution alkalinity, which indirectly supports carbonation by stabilizing $Ca(OH)_2$ necessary for CO_2 uptake [40], [41]. On the other hand, P forms less soluble compounds with Ca, potentially reducing the availability of free Ca^{2+} ions for carbonation reactions. The presence of these elements might also modify the microstructure, affecting porosity and diffusion pathways for CO_2 [42]. Also, as suggested by Nielsen *et al*. [43] Mg is known to significantly influence the growth and dissolution of calcite in the mineral carbonation process. The presence of Mg in raw materials or additives like ISSA can thus play a significant role in shaping the crystallization kinetics, morphology, and overall efficiency of the carbonation process. Similar observations were made by Zhang and Dawe [44], who discovered that the presence of Mg_{2+} ions in a $CaCO_3$ supersaturated solution inhibits the growth of calcite primarily by interfering with crystal lattice formation, adsorbing onto crystal surfaces, and altering the surface energetics. These mechanisms collectively reduce the growth rate of calcite crystals, highlighting the significant role of Mg_{2+} ions in controlling mineral precipitation kinetics and crystal morphology during mineral carbonation.

Samples containing higher percentages of ISSA generally show lower $CaCO_3$ content post-carbonation compared to those with lower ISSA percentages, however, in the outdoor experiment, the relative increase in $CaCO_3$ was smaller than in the control sample (Table 1). The observed differences in CO_2 sequestration between laboratory and outdoor conditions can be attributed to weather fluctuations during the experiment. Additionally, the presence of water in the vessels where samples were placed may have affected carbonation efficiency by limiting CO_2 diffusion, prolonging pore saturation, and potentially dissolving metastable carbonate phases as also suggested by Lin *et al*. [45].

3.3. Impact of ISSA Addition on CO_2 Sequestration Efficiency under Controlled and Natural Conditions

The Scheibler method of measuring $CaCO_3$ reveals consistent variations in exposure conditions and ISSA concentrations (Table 1). The highest carbonation efficiency is observed

for mixes with 20–25 % ISSA, while higher ISSA contents (>30 %) exhibit reduced $CaCO_3$ formation. As shown in Table 1, the laboratory exposure yields higher carbonation efficiency than outdoor exposure. According to our results, increasing the ISSA content to 25 % in cement paste had a significant effect on CO_2 sequestration, capturing approximately 69 kg of CO_2 per ton under controlled laboratory conditions and 34 kg of CO_2 per ton in natural conditions. These results suggest that under controlled conditions, where environmental factors such as temperature and humidity can be precisely regulated, the efficiency of CO_2 capture is 35 % higher compared to samples without ISSA. However, when compared to pure cement paste without any ISSA addition, the CO_2 uptake is 35 % lower under natural conditions (Table 1).

TABLE 1. THE $CaCO_3$ % FORMATION MEASUREMENTS FOR RAW SAMPLES AND AFTER BOTH THE LABORATORY AND OUTDOOR EXPERIMENTS BY THE SCHEIBLER VOLUMETRIC METHOD

	$CaCO_3$, %, in raw samples	**$CaCO_3$, %, after laboratory experiment**	**increase in $CaCO_3$, %**	**$CaCO_3$, %, after outdoor experiment**	**increase in $CaCO_3$, %**
0 %	29.69	40.1	10.41	40.21	10.52
20 %	17.73	30.68	12.95	25	7.27
25 %	**14.32**	**30.08**	**15.76**	**22.02**	**7.7**
30 %	23.23	24.04	0.81	25.83	2.6
35 %	26.82	26.09	–0.73	20.41	–6.41

Liu *et al*. [26] found that the maximum carbon uptake reached up to 26.67 kg per ton at a 5 % ISSA incorporation rate under natural environmental conditions. When compared with our results, this indicates that 5 % ISSA may be too low to significantly modify the binder system and provide conditions such as availability of Ca-bearing phases and a favourable microstructural and transport environment for CO_2 ingress that maximize $CaCO_3$ formation.

It should be noted that while SEM observations in this study revealed that calcium carbonate precipitation was mainly concentrated at the surface, this is typical of natural or early-stage carbonation processes, where CO_2 penetration is limited by diffusion and reaction kinetics [32]. Surface carbonation should not be misinterpreted as superficial or ineffective, especially under atmospheric conditions. The Scheibler method, applied in this study, quantitatively confirmed $CaCO_3$ formation throughout the specimen volume, validating that the observed surface crystallization corresponded with significant bulk CO_2 sequestration. In practice, this near-surface precipitation is relevant because it is the exposed zone that largely controls permeability and durability in non-structural elements

Although ISSA-modified cement pastes showed slightly lower carbonation efficiency in natural conditions compared to pure cement, they still contributed significantly to CO_2 sequestration and material circularity, making them a viable option for sustainable construction.

3.4. Environmental Controls: Temperature, RH and Moisture Effects

The weather conditions during the outdoor experiment were far less favourable for optimal carbonation. Air temperature exceeded 20.0 °C for only 13 % of the experiment's duration and remained below 15.0 °C for three-quarters of the time (Fig. 5). The reduced carbonation efficiency under such conditions is a known limitation, as studies have shown that lower temperatures significantly hinder carbonation by slowing down reaction rates and reducing

the efficiency of $CaCO_3$ formation. The laboratory environment, on the other hand, which is carefully controlled, offers optimal conditions for carbonation, where temperatures and humidity levels can be maintained to encourage the precipitation of calcium carbonate. This process is highly dependent on temperature, as higher temperatures accelerate the carbonation reaction, leading to greater CO_2 uptake and the formation of stable $CaCO_3$ crystals. According to Verbeck [46], the carbonation rate in cementitious materials reaches its maximum at 20 °C.

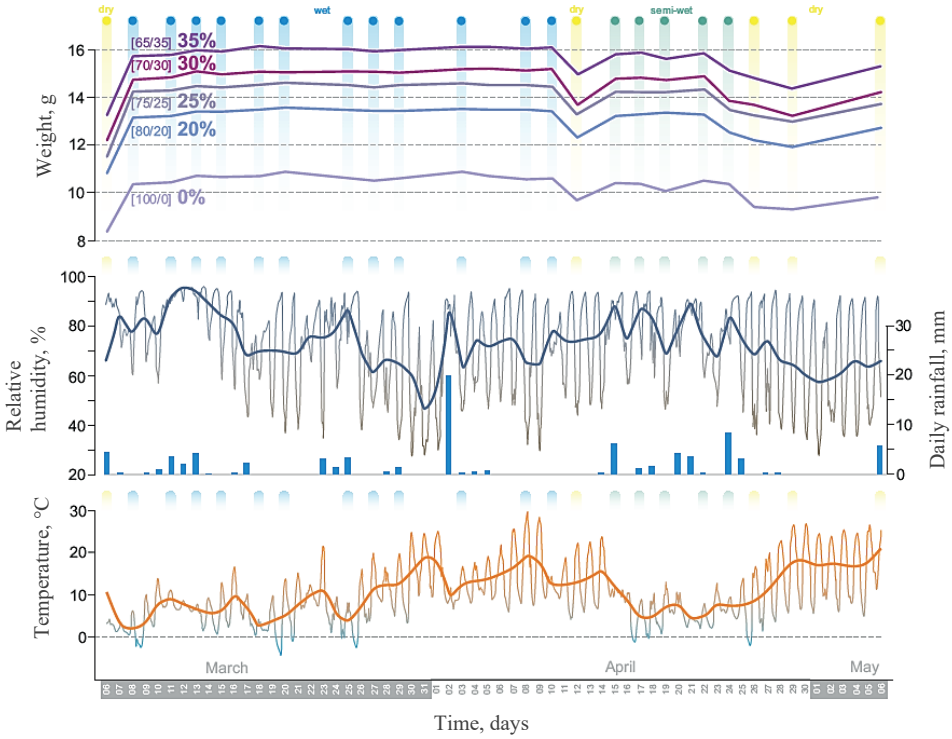

Fig. 5. The variations in relative humidity (with daily average record), temperature (with daily average record), daily rainfall and mass of the samples during the outdoor experiment. The condition of samples during weighing was marked above graphs as follows: yellow – dry, green – semi-wet, blue – wet.

Additionally, fluctuations in relative humidity likely influenced the carbonation process. While laboratory conditions allow for maintaining a constant and optimal RH, in natural settings, RH can vary widely, affecting CO_2 diffusion and overall carbonation efficiency. Previous research has shown that carbonation in plain cementitious materials is most effective when RH is approximately 50–55 % [47], while Verbeck [46] estimated a broader optimal range of 40–80 %. Deviations from this range can either lead to excessive moisture, which impedes CO_2 diffusion, or insufficient moisture, which prevents carbonation from progressing effectively. During the outdoor experiment, RH remained within the 40–80 % range for more than half of the time (Fig. 5). However, when considering air temperature and humidity together, optimal conditions for carbonation occurred for only 10 % of the experiment's duration. Moreover, large daily variations in these parameters likely further inhibited the full development of the carbonation process. Finally, the carbonation process under natural conditions was impacted by the high moisture content of the samples, resulting from rainfall, relatively low temperatures and high humidity. Despite these issues, the

addition of 25 % ISSA to cement paste consistently yields the best results in both controlled and natural conditions, compared to other sample compositions (Table 1). The ISSA acts as a supplementary material that enhances the carbonation process and contributes to the sequestration of CO_2.

The mass change trends were observed in the cement blocks in the case of both experiments. The laboratory experiment revealed two distinct phases (Fig. 5). During the initial phase, a decrease in mass was observed across all samples, particularly noticeable in the first 30 days of the experiment duration. This can be attributed to the drying stage of the wetting and drying cycles, where water retained in the pores evaporated, leading to mass loss. In the subsequent phase, a gradual increase in mass was recorded, which correlates with the onset of carbonation. This process involves the reaction of calcium hydroxide ($Ca(OH)_2$) in the cement matrix with atmospheric CO_2 to form calcium carbonate ($CaCO_3$), resulting in a net mass gain. The dynamics of mass change varied slightly depending on the ISSA content, with higher ISSA proportions potentially influencing porosity and reactivity showing the interplay between drying, carbonation, and ISSA content in determining the overall mass evolution of the blocks. The following data presents mass measurements over multiple dates for various cement-to-ISSA ratios, highlighting the dynamic effects of ISSA content on the hydration or carbonation processes. The control samples show a consistent increase in mass over time, indicating "normal" carbonation processes without the presence of ISSA. The mass increases slightly over time for the samples containing 20 %, 25 %, and 30 % of ISSA, indicating some degree of continuing process over the measurement period, however in the case of the sample containing 35 % of ISSA there is variability in mass measurements, with fluctuations across different periods. This trend can be attributed to competing processes of hydration and carbonation, where higher ISSA ratios may enhance hydration due to the presence of reactive phases in sewage sludge ash. Since calcite formation analysis (via multiple methods) indicates the largest CO_2 uptake for samples containing 20 % and 25 % ISSA, we may assume that the increase in mass for samples with higher ISSA content may reflect combined hydration and carbonation. In cement-ash mixtures, $Ca(OH)_2$ is primarily produced through the hydration of tricalcium silicate (C_3S) and dicalcium silicate (C_2S). The presence of sewage sludge ash can influence the availability and distribution of $Ca(OH)_2$ through pozzolanic reactions, where reactive components in the ash react with $Ca(OH)_2$ to form additional cementitious phases [48]. While these pozzolanic reactions may reduce the availability of $Ca(OH)_2$ for carbonation, they do not completely inhibit the process. Residual $Ca(OH)_2$ can still react with CO_2 from the atmosphere to form calcium carbonate ($CaCO_3$), particularly at the material's surface, as observed in SEM images. Hydration is indeed a prerequisite for carbonation in cement-based materials; it provides the necessary $Ca(OH)_2$, which is essential for the carbonation process. Understanding this relationship helps in managing and predicting the long-term durability and performance of concrete structures, especially in environments with significant CO_2 exposure [49].

The addition of ISSA introduces aluminosilicates that can undergo pozzolanic reactions, consuming $Ca(OH)_2$ and influencing the availability of phases for carbonation.

Even in the presence of Al and Si-rich ash, which promotes pozzolanic reactions consuming some $Ca(OH)_2$ [50], there still should be $Ca(OH)_2$ available for carbonation. The pozzolanic reactions may inhibit some degree of carbonation, but not completely prevent it, allowing for the formation of $CaCO_3$ especially at the surface of the material as we could observe in the SEM images. The observed mass changes reflect this balance, with an initial mass loss caused by drying during the early cycles, followed by a gradual increase as carbonation and hydration advanced.

3.5. Mass Evolution during Exposure

The mass of the samples in ambient conditions also changed during the experiment suggesting varying degrees of moisture absorption and evaporation. The mass of the samples gradually increased over time, particularly for the samples with higher ISSA content. The 0 % ISSA sample showed a relatively lower and more stable mass, indicating lower moisture retention compared to ISSA-modified samples. The RH was highly variable, often reaching near saturation (~100 %) and then dropping. This suggests frequent wetting and drying cycles, which impacted mass changes. During drying periods, the reduction in moisture content can enhance CO_2 diffusion but may also lead to the partial decomposition of metastable carbonate phases, such as vaterite, back into more soluble forms [51]. Excessively high humidity could limit CO_2 diffusion, affecting the overall sequestration process, as the pores were saturated with water, and therefore limit the amount of CO_2 that could penetrate the material and thus slowing down the mineral carbonation process [52].

3.6. The Potential Application of Carbonated Hardened Cement Pastes

Incorporating ISSA into cementitious materials can effectively support the waste utilization, reducing the need for raw materials and mitigating CO_2 emissions. From a practical perspective, the key outcome of this study is that ISSA-modified cement pastes enable measurable CO_2 uptake through passive carbonation, with the highest carbonation efficiency observed at low-to-moderate ISSA contents (20–25 %) (Table 1). For non-reinforced elements, carbonation does not pose a corrosion risk and may be beneficial by promoting microstructural changes associated with $CaCO_3$ precipitation, potentially contributing to near-surface densification and reduced permeability [53]. Therefore, the investigated material concept is most relevant for non-structural, non-reinforced cement-based products**,** where service performance is governed mainly by durability and permeability rather than protection of steel reinforcement [33], [54]. In practice, this includes small, prefabricated components and minor public-space infrastructure without reinforcement (e.g., paving units, kerbs, blocks, protective layers, and urban small architecture), which also provides an appropriate setting for pilot-scale implementation under real exposure conditions. The comparison between laboratory and outdoor exposure further indicates that carbonation performance is strongly dependent on environmental variability, which should be considered when selecting curing and exposure regimes for practical deployment [11], [46], [47].

4. CONCLUSION

This study demonstrated that incorporating incinerated sewage sludge ash (ISSA) into hardened cement pastes can enhance mineral carbonation and CO_2 uptake, particularly at 20–25 % ISSA content, as evidenced by quantitative $CaCO_3$ formation measured via the Scheibler method. Under controlled laboratory conditions (22 °C, ~45 % RH), carbonation was more efficient, while natural environmental conditions with highly fluctuating temperature and humidity slowed down carbonation rates due to inconsistent moisture levels affecting CO_2 diffusion. Higher ISSA contents (>30 %) resulted in reduced carbonation efficiency, likely due to pore structure modifications and inhibitory effects of elements such as phosphorus, potassium, and magnesium. These findings highlight the influence of environmental exposure on carbonation kinetics and the importance of optimizing ISSA incorporation in cement-based applications. Utilizing ISSA as an additive in cement-based materials supports circular economy principles by repurposing waste, reducing landfill dependency, lowering clinker demand, and contributing to CO_2 sequestration in construction materials. This research provides insights into the potential of ISSA-modified cementitious

materials for sustainable building practices, but further studies are needed to refine exposure conditions and long-term durability assessments under real-world scenarios. Carbonation trends were consistently supported by Scheibler $CaCO_3$ quantification, XRD phase evidence, and SEM-EDS morphological observations.

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REFERENCES

[1] Moro C., Francioso V., Lopez-Arias M., Velay-Lizancos M. The impact of CO_2 uptake rate on the environmental performance of cementitious composites: A new dynamic Global Warming Potential analysis. *Journal of Cleaner Production* 2022:375:134155 https://doi.org/10.1016/j.jclepro.2022.134155

[2] Nunes L. J. R. The Rising Threat of Atmospheric CO_2: A Review on the Causes, Impacts, and Mitigation Strategies. *Environments* 2023:10(4):66. https://doi.org/10.3390/environments10040066

[3] Godbold J. A., Solan M. Long-term effects of warming and ocean acidification are modified by seasonal variation in species responses and environmental conditions. *Philosophical Transactions of the Royal Society B* 2013:368(1627):20130186. https://doi.org/10.1098/rstb.2013.0186

[4] Steiner S., Lothenbach B., Proske T., Borgschulte A., Winnefeld F. Effect of relative humidity on the carbonation rate of portlandite, calcium silicate hydrates and ettringite. *Cement and Concrete Research* 2020:135:106116. https://doi.org/10.1016/j.cemconres.2020.106116

[5] Stripple H., Ljungkrantz C., Gustafsson T., Andersson R. CO_2 Uptake in Cement Containing Products: Background and Calculation Models for IPCC Implementation. *IVL Swedish Environmental Research Institute* 2018.

[6] Xi F., Davis S. J., Ciais P., Crawford-Brown D., Guan D., Pade C., Shi T., Syddall M., Lv J., Ji L., Bing L., Wang J., Wei W., Yang Z.-H., Lagerblad B., Galan I., Andrade C., Zhang Y., Liu Z. Substantial global carbon uptake by cement carbonation. *Nature Geoscience* 2016:9(12):880–883. https://doi.org/10.1038/ngeo2840

[7] Matschei T., Lothenbach B., Glasser F. P. The role of calcium carbonate in cement hydration. *Cement and Concrete Research* 2007:37(4):551–558. https://doi.org/10.1016/j.cemconres.2006.10.013

[8] Fernández-Carrasco L., Torrens-Martín D., Morales L. M., Martínez-Ramírez S. Infrared Spectroscopy in the Analysis of Building and Construction Materials. *Infrared Spectroscopy – Materials Science, Engineering and Technology,* 2012. https://doi.org/10.5772/36186

[9] Boumaaza M., Turcry P., Huet B., Aït-Mokhtar A. Influence of carbonation on the microstructure and the gas diffusivity of hardened cement pastes. *Construction and Building Materials* 2020:253:119227. https://doi.org/10.1016/j.conbuildmat.2020.119227

[10] Wang H., Liu T., Zhang Z., Zou D., Zhou A., Li Y. Durability of SFCB reinforced low-alkalinity seawater sea sand concrete beams in marine environment. *Engineering Structures* 2024:317:118616. https://doi.org/10.1016/j.engstruct.2024.118616

[11] Xu Z., Zhang Z., Huang J., Yu K., Zhong G., Chen F., Chen X., Yang W., Wang Y. Effects of temperature, humidity and CO_2 concentration on carbonation of cement-based materials: A review. *Construction and Building Materials* 2022:346:128399. https://doi.org/10.1016/j.conbuildmat.2022.128399

[12] Fattuhi N. I. Carbonation of concrete as affected by mix constituents and initial water curing period. *Materials and Structures* 1986:19:131–136. https://doi.org/10.1007/BF02481757

[13] Sanjuan M. A., Andrade C., Cheyrezy M. Concrete carbonation tests in natural and accelerated conditions. *Advances in Cement Research* 2003:15:171–180. https://doi.org/10.1680/adcr.15.4.171.36711

[14] Castellote M., Andrade C., Turrillas X., Campo J., Cuello G. J. Accelerated carbonation of cement pastes in situ monitored by neutron diffraction. *Cement and Concrete Research* 2008:31(12):1365–1373. https://doi.org/10.1016/j.cemconres.2008.07.002

[15] von Greve-Dierfeld S., Lothenbach B., Vollpracht A., Wu B., Huet B., Andrade C., Medina C., Thiel C., Gruyaert E., Vanoutrive H., del Bosque I. F. S., Ignjatovic I., Elsen J., Provis J. L., Scrivener K., Thienel K.-C., Sideris K., Zajac M., Alderete N., Cizer Ö., Van den Heede P., Hooton R. D., Kamali-Bernard S., Bernal S. A., Zhao Z. F., Shi Z. G., De Belie N. Understanding the carbonation of concrete with supplementary cementitious materials: a critical review by RILEM TC 281-CCC. *Materials and Structures* 2020:53:136. https://doi.org/10.1617/s11527-020-01558-w

[16] Neville A. M. Properties of Concrete, Fifth ed., Prentice Hall, Malaysia, 2011. [Online]. [Accessed 21.05.2024]. Available: http://refhub.elsevier.com/S0950-0618(18)31238-8/h0060
[17] Fan M. X., Chen F. X., Zhang X. Y., Wang R. K., Yu R. Effect of Ca/Si ratio on the characteristics of alkali-activated ultra-high performance concrete (A-UHPC): From hydration kinetics to microscopic structure development. *Construction and Building Materials* 2023:394:132158. https://doi.org/10.1016/j.conbuildmat.2023.132158
[18] Glasser F. P., Marchand J., Samson E. Durability of concrete — Degradation phenomena involving detrimental chemical reactions. *Cement and Concrete Research* 2008:38(2):226–246. https://doi.org/10.1016/j.cemconres.2007.09.015
[19] Šauman Z. Carbonization of porous concrete and its main binding components. *Cement and Concrete Research* 1971:1(6):645–662. https://doi.org/10.1016/0008-8846(71)90019-6
[20] Shah V., Bishnoi S. Carbonation resistance of cements containing supplementary cementitious materials and its relation to various parameters of concrete. *Construction and Building Materials* 2018:178:219–232. https://doi.org/10.1016/j.conbuildmat.2018.05.162
[21] Neeraj X., Yadav S. Carbon storage by mineral carbonation and industrial applications of CO_2. *Materials Science for Energy Technologies* 2020:3:494–500. https://doi.org/10.1016/j.mset.2020.03.005
[22] Bobicki E. R., Liu Q., Xu Z., Zeng H. Carbon capture and storage using alkaline industrial wastes. *Progress in Energy and Combustion Science* 2012:38(2):302–320. https://doi.org/10.1016/j.pecs.2011.11.002
[23] Teir S., Eloneva S., Fogelholm C.-J., Zevenhoven R. Dissolution of steelmaking slags in acetic acid for precipitated calcium carbonate production. *Energy* 2007:32(4):528–539. https://doi.org/10.1016/j.energy.2006.06.023
[24] Huijgen W. J. J., Witkamp G-J., Comans R. N. J. Mineral CO_2 sequestration by steel slag carbonation. *Environmental Science and Technology* 2005:39(24):9676–9682. https://doi.org/10.1021/es050795f
[25] Li X., Bertos M. F., Hills C. D., Carey P. J., Simon S. Accelerated carbonation of municipal solid waste incineration fly ashes. *Waste Management* 2007:27(9):1200–1206. https://doi.org/10.1016/j.wasman.2006.06.011
[26] Liu J., Liu G., Zhang W., Li Z., Jin H., Xing F. A new approach to CO_2 capture and sequestration: a novel carbon capture artificial aggregates made from biochar and municipal waste incineration bottom ash. *Construction and Building Materials* 2023:398:132472. https://doi.org/10.1016/j.conbuildmat.2023.132472
[27] Baciocchi R., Polettini A., Pomi R., Prigiobbe V., Von Zedwitz V. N., Steinfeld A. CO_2 Sequestration by Direct Gas-Solid Carbonation of Air Pollution Control (APC) Residues. *Energy Fuels* 2006:20(5):1933–1940. https://doi.org/10.1021/ef060135b
[28] Du Y., Pundiene I., Pranckeviciene J., Korjakins A., Kligys M. Mechanical Properties and Lifecycle Assessment of a Green Alkali-Activated Mortar Based on Biomass Wood Ash. *Environmental and Climate Technologies* 2024:28(1):195–207. https://doi.org/10.2478/rtuect-2024-0016
[29] Argalis P. P., Sinka M., Andzs M., Korjakins A., Bajare D. Development of New Bio-Based Building Materials by Utilising Manufacturing Waste. *Environmental and Climate Technologies* 2024:28(1):58–70. https://doi.org/10.2478/rtuect-2024-0006
[30] Lin X., Zhang Y., Liu H., Boczkaj G., Cao Y., Wang Ch. Carbon dioxide sequestration by industrial wastes through mineral carbonation: Current status and perspectives. *Journal of Cleaner Production* 2024:434:140258. https://doi.org/10.1016/j.jclepro.2023.140258
[31] Kasina M., Wendorff-Belon M., Kowalski P., Michalik M. Characterization of incineration residues from wastewater treatment plant in Polish city: a future waste based source of valuable elements? *Journal of Material Cycles and Waste Management* 2019:21(4):885–896. https://doi.org/10.1007/s10163-019-00845-1
[32] Tural H. G., Ozarisoy B., Derogar S., Ince C. Investigating the governing factors influencing the pozzolanic activity through a database approach for the development of sustainable cementitious materials. *Construction and Building* Materials 2024:411(12):134253. https://doi.org/10.1016/j.conbuildmat.2023.134253
[33] Liang C., Le X., Fang W., Zhao J., Fang L., Hou S. The Utilization of Recycled Sewage Sludge Ash as a Supplementary Cementitious Material in Mortar: A Review. *Sustainability* 2022:14(8):4432. https://doi.org/10.3390/su14084432
[34] Lagerblad B. Carbon dioxide uptake during concrete life cycle: State of the art (CBI Report No. FOU 2:2005). Swedish Cement and Concrete Research Institute, 2005.
[35] Cuesta A., Zea-Garcia J. D., Londono-Zuluaga D., De La Torre A.G., Santacruz I., Vallcorba O., Dapiaggi M., Sanfélix S. G., Aranda M. A. G. Multiscale understanding of tricalcium silicate hydration reactions. *Scientific Reports* 2018:8:8544. https://doi.org/10.1038/s41598-018-26943-y
[36] Kasina M., Jarosz K., Stolarczyk M., Göttlicher J., Steininger R., Michalik M. Characteristic of phosphorus rich compounds in the incinerated sewage sludge ashes: a case for sustainable waste management. *Scientific Reports* 2023:13:9137. https://doi.org/10.1038/s41598-023-36407-7
[37] Lu J., Ruan S., Liu Y., Wang T., Zeng Q., Yan D. Morphological characteristics of calcium carbonate crystallization in CO_2 pre-cured aerated concrete. *RSC Advances* 2022:12:14610–14620. https://doi.org/10.1039/D2RA01901A
[38] Kerisit S., Parker S. C. Free energy of adsorption of water and metal ions on the [1014] calcite surface. *Journal of the American Chemical Society* 2004:126(32):10152–10161. https://doi.org/10.1021/ja0487776

[39] Kusiorowski R., Gerle A., Kujawa M., Antonovič V., Boris R. Comparative Analysis of Cement-Asbestos Materials from Poland and Lithuania: Thermal Decomposition, Mineralogical Characterisation, and Implications for Recycling. *Environmental and Climate Technologies* 2025:29(1):940–959. https://doi.org/10.2478/rtuect-2025-0063

[40] Plusquellec G., Geiker M. R., Lindgård J., Duchesne J., Fournier B., De Weerdt K. Determination of the pH and the free alkali metal content in the pore solution of concrete: Review and experimental comparison. *Cement and Concrete Research* 2017:96:13–26. https://doi.org/10.1016/j.cemconres.2017.03.002

[41] Rodriguez-Navarro C., Ilić T., Ruiz-Agudo E., Elert K. Carbonation mechanisms and kinetics of lime-based binders: An overview. *Cement and Concrete Research* 2023:173:107301. https://doi.org/10.1016/j.cemconres.2023.107301

[42] Abu-Eishah S. I., Anabtawi M. J. J., Isaac S. L. Upgrading of carbonaceous phosphate rocks by direct carbonation with CO_2–water solutions. *Chemical Engineering and Processing: Process Intensification* 2024:43(8):1085–1094. https://doi.org/10.1016/j.cep.2003.11.001

[43] Nielsen M. R., Sand K. K., Rodriguez-Blanco J. D., Bovet N., Generosi J., Dalby K. N., Stipp S. L. S. Inhibition of Calcite Growth: Combined Effects of Mg^{2+} and SO_4^{2-}. *Crystal Growth & Design* 2016:16(11):6199–6207. https://doi.org/10.1021/acs.cgd.6b00536

[44] Zhang Y., Dawe R. A. Influence of Mg^{2+} on the kinetics of calcite precipitation and calcite crystal morphology. *Chemical Geology* 2000:163(1–4):129–138. https://doi.org/10.1016/S0009-2541(99)00097-2

[45] Lin S., Chen P., Xiang W., Hu C., Li F., Liu J., Ding Y. Exploring the Effect of Moisture on CO_2 Diffusion and Particle Cementation in Carbonated Steel Slag. *Applied Sciences* 2024:14(9):3631. https://doi.org/10.3390/app14093631

[46] Verbeck G. Carbonation of hydrated Portland cement. *ASTM Special Technical Publication* 1958:205:17–3. https://doi.org/10.1520/STP39460S

[47] Leemann A., Moro F. Carbonation of concrete: the role of CO_2 concentration, relative humidity and CO2 buffer capacity. *Materials and Structures* 2017:50:30. https://doi.org/10.1617/s11527-016-0917-2

[48] Rutkowska G., Żółtowski M., Rusakov K., Pawluk K., Andrzejak J., Żółtowski B. The Influence of Fly Ash from Sewage Sludge on the Concrete Carbonation Course. *Buildings* 2023:13(7):1838. https://doi.org/10.3390/buildings13071838

[49] Qiu Q. A state-of-the-art review on the carbonation process in cementitious materials: Fundamentals and characterization techniques. *Construction and Building Materials* 2020:247:118503. https://doi.org/10.1016/j.conbuildmat.2020.118503

[50] Snehal K., Das B. B. Pozzolanic reactivity and drying shrinkage characteristics of optimized blended cementitious composites comprising of Nano-Silica particles. *Construction and Building Materials* 2022:316:125796. https://doi.org/10.1016/j.conbuildmat.2021.125796

[51] Drouet E., Poyet S., Le Bescop P., Torrenti J.-M., Bourbon X. Carbonation of hardened cement pastes: Influence of temperature. *Cement and Concrete Research* 2018:115:445–459. https://doi.org/10.1016/j.cemconres.2018.09.019

[52] Chen Y., Liu P., Yu Z. Effects of Environmental Factors on Concrete Carbonation Depth and Compressive Strength. *Materials* 2018:11(11):2167. https://doi.org/10.3390/ma11112167

[53] Pham S. T., Prince W. Effects of Carbonation on the Microstructure of Cement Materials: Influence of Measuring Methods and of Types of Cement. *International Journal of Concrete Structures and Materials* 2014:8(4):327–333. https://doi.org/10.1007/s40069-014-0079-y

[54] Wang J., Hu Z., Chen Y., Huang J., Ma Y., Zhu W., Liu J. Effect of Ca/Si and Al/Si on micromechanical properties of C(-A)-S-H. *Cement and Concrete Research* 2022:157:106811. https://doi.org/10.1016/j.cemconres.2022.106811