

## Carbon utilization technologies in cement-based materials. Effect of carbonation on fresh mixtures and on recycled cementitious fines

### Summary

This doctoral thesis presents an in-depth scientific investigation into two innovative carbon utilization strategies within the cement and concrete industry, illustrated in Figure 1. Both approaches contribute to broader Carbon Capture, Utilization, and Storage (CCUS) technologies, addressing distinct challenges and mechanisms related to CO<sub>2</sub> sequestration, cementitious reactivity, and material performance.

The first line of research explores carbonation during the mixing stage of cement-based materials, aiming to incorporate CO<sub>2</sub> at the earliest point of concrete production. The goal is to induce carbonation concurrently with hydration, enabling early-stage CO<sub>2</sub> sequestration. The research evaluates physicochemical transformations under these conditions, focusing on the hydration-carbonation interplay and its consequences on performance. Results show that introducing CO<sub>2</sub> during mixing significantly alters hydration kinetics. The gas accelerates cement dissolution, especially in finer particles, causing high heat release and unbalanced constituent consumption. These effects inhibit orderly C-S-H growth. Thermogravimetric analysis and X-ray diffraction confirm early calcium carbonate polymorph formation, disrupting typical hydration pathways. While this strategy enhances carbon uptake, it may compromise early strength and microstructure unless controlled. Still, it shows promise for reducing carbon footprint during mixing and offers insights to optimize mix design for reactivity, durability, and sustainability.

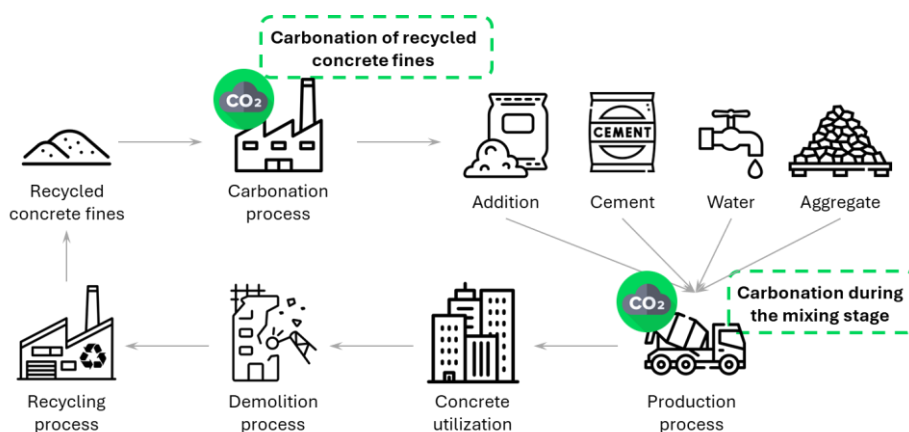


Figure 1: Carbon utilization strategies studied within the concrete cycle

The second research axis focuses on carbonating recycled concrete fines (RCF) for use as supplementary cementitious materials (SCMs). This approach utilizes two waste streams: CO<sub>2</sub> emissions and concrete residues. RCF are subjected to controlled accelerated carbonation to transform their mineralogy and improve reactivity. Experimental campaigns characterize carbonation kinetics and mechanisms. Rapid initial carbonation consumes calcium hydroxide and other phases, followed by slower, diffusion-controlled reactions involving C-S-H. A key advancement is a short-duration carbonation process, reducing treatment from days to hours while maintaining performance. Optimization of CO<sub>2</sub> levels, water content, particle size, and layer thickness was critical. Carbonated RCF were then used as partial clinker replacements. Compared to uncarbonated RCF and limestone fillers, carbonated RCF improved strength at later ages due to better packing, refined microstructure, and possible latent hydraulic activity.

Overall, this thesis significantly contributes to sustainable construction materials, demonstrating the technical feasibility, scientific basis, and environmental value of two novel CO<sub>2</sub> utilization strategies in cement systems. It advances understanding of carbonation under varied conditions and provides a foundation for scaling and industrial adoption. The work supports a shift toward carbon circularity in cement production, positioning concrete as a potential carbon sink.

### Keywords

Carbon Capture Utilization and Storage; Carbon Utilization Strategy; Cement-Based Materials; Carbonation Reaction; Carbon Absorption Capacity.



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