

## Carbonation-activated microstructural refinement in GUL-GGBFS blended mortars: shrinkage mitigation and strength enhancement

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With climate change intensifying, carbon capture, utilization, and storage (CCUS) has become essential for environmental protection. Among CCUS methods, carbonation curing ( $3\% \pm 0.5\%$   $\text{CO}_2$ ) offers a promising path to carbon-neutral concrete by absorbing and reacting  $\text{CO}_2$  with cement and hydration products, forming stable carbonates and increasing concrete strength. This study investigates the carbonation behavior of low carbon cementitious composites with 0 to 80% Ground granulated blast furnace slag (GGBFS) replacement of General Use Limestone (GUL) cement. Compressive strength and shrinkage were first evaluated to provide key insights into macroscopic properties. The carbonation depth, carbonation rate, and degree of carbonation (DOC) were then measured to assess carbonation performance of mixtures containing 0% to 80% GGBFS. The mineralogical composition and microstructural evolution of the GGBFS composites were characterized using thermogravimetric analysis (TG), quantitative X-ray diffraction (XRD), X-ray computed tomography (XCT), and dynamic vapor sorption (DVS) to clarify the mechanisms governing carbonation-induced property changes and improve control of cementitious material properties.

Key Takeaways:

- 1. Effect of drying and carbonation on compressive strength of GGBFS specimens (Fig. 1)**- Under drying conditions ( $50 \pm 2\%$  RH,  $23 \pm 2$  °C) (0-90 days), specimens with GGBFS exhibited 7.7% to 65.6% lower compressive strength than those without GGBFS, as the slower hydration of GGBFS delayed the formation of strength-contributing hydrates. However, accelerated carbonation mitigated this reduction by promoting a secondary compressive strength gain of 18.3% to 46.9%. The combination of dolomite and calcite occurred during carbonation also induces a beneficial effect, contributing to a refined pore structure and enhanced mechanical strength.
- 2) Effect of GGBFS content on drying and carbonation shrinkage (Fig. 2)**-Increasing the GGBFS content from 20% to 60% reduced drying shrinkage by 3% to 7% compared to the mix without GGBFS. At high replacement levels (80%), however, with limited early hydration, more free water was left in the system and evaporated led to increased drying shrinkage. Although accelerated carbonation increased shrinkage by refining capillary pores, incorporating 40% to 80% GGBFS still helped mitigate carbonation-induced shrinkage by 20.3%. Overall, 40% of GGBFS replacement provided the most favorable balance, maintaining volume stability under both drying and carbonation conditions.
- 3) Influence of GGBFS content on carbonation behavior (Fig. 3)**-Carbonation densified the pore structure, partially restricting further  $\text{CO}_2$  ingress. However, the presence of GGBFS contributed to higher carbonation depth, carbonation rate, and DOC compared to the samples with 0% GGBFS. This carbonation increase is attributed to the reduced cement fraction limited the

formation of C-S-H and  $\text{Ca}(\text{OH})_2$ , while the pozzolanic reaction further depleted  $\text{Ca}(\text{OH})_2$ , thereby lowering buffering capacity and accelerating C-S-H decalcification.

**4) 3D characterization of carbonation-induced cracks by XCT (Fig. 4)**-XCT analysis revealed the formation of cracks in specimens with high GGBFS contents (60% and 80%) during accelerated carbonation. Unlike conventional linear fractures observed in 2D image, the 3D models reconstructed from the XCT data showed that these cracks extended as spatial defects within the specimens. The presence of such extensive cracking negatively impacted mechanical integrity, increased maintenance and repair costs, and undermines sustainability of the cementitious materials.

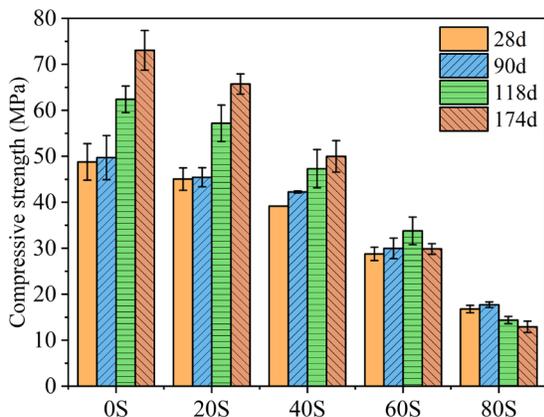


Fig. 1. Compressive strengths of mortar cubes with varying GGBFS (0S to 80S) subjected to drying (28d, 90d) and accelerated carbonation (118d, 174d).

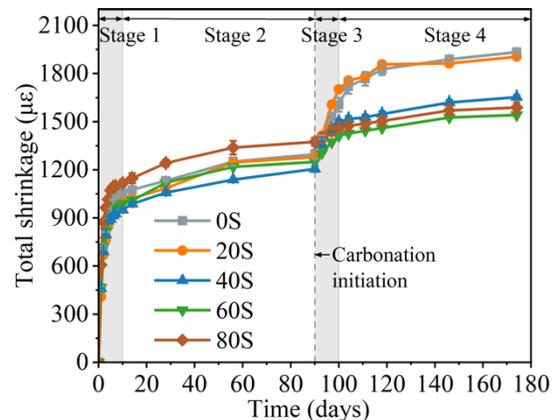


Fig. 2. Shrinkage of specimens with 0% (0S) to 80% (80S) GGBFS subjected to drying (0-90 days) and accelerated carbonation (90-174 days).

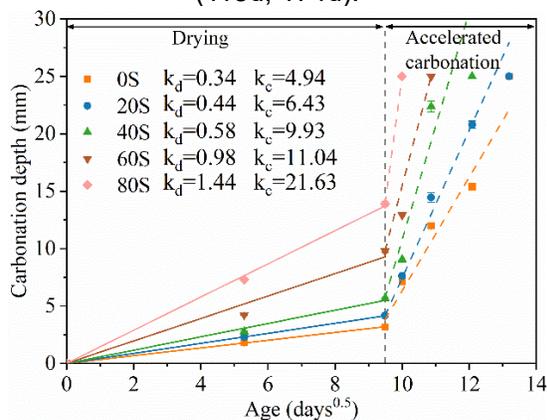


Fig. 3. Carbonation rate of specimens with 0% to 80% GGBFS subjected to drying and accelerated carbonation conditions.

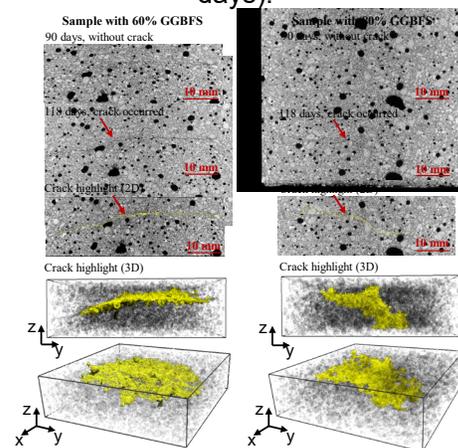


Fig. 4. Micro tomographic crack images of dry and carbonated samples: (a) 60S; (b) 80S.