

Ph.D. DISSERTATION DEFENSE

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Title: Bioinspired Cementitious Composites with Superior Strength, Toughness, and Multifunctionality

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ABSTRACT

Traditional cementitious materials exhibit excellent compressive strength but suffer from low flexural strength and toughness. In addition, their high carbon emissions and thermal conductivity contribute to energy-intensive and environmentally unsustainable construction practices. To address these limitations, this research draws inspiration from natural materials, aiming to replicate their unique structure to improve the flexural strength, toughness, sustainability, and multifunctionality of cementitious systems. This dissertation is organized into two main research thrusts.

First, this work focuses on engineering calcium silicate hydrate (C-S-H), the primary hydration product of cement, as well as cement itself into brick-and-mortar architectures, inspired by the natural microstructure of nacre. Various polymers, including poly(acrylamide-co-acrylic acid) (P-co-P), polyvinyl alcohol (PVA), and cellulose nanocrystals (CNCs), were investigated for their ability to disperse and toughen both C-S-H and cement particles. Fabrication strategies such as vacuum filtration, freeze casting and freeze drying were developed to assemble these polymer-reinforced phases into hierarchical brick-and-mortar structures. By constructing such organic and inorganic hybrids, the resulting composites exhibit significantly improved flexural strength and fracture toughness.

Second, this research aims to advance the sustainability and multifunctionality of cementitious materials by emulating the porous architectures in natural biominerals, such as sea urchin spine and cuttlebones. To achieve this, a series of cement aerogels with both hierarchical porous and foam-like structures were developed through the crosslinking of PVA or sodium alginate (SA) with cement particles. These organic-inorganic hybrid porous architectures confer several advantages, including increased surface area, reduced cement consumption, and enhanced pathways for CO₂ diffusion and mineralization. As a result, the fabricated aerogels demonstrate a unique combination of low density (0.042–0.50 g/cm³), ultra-low thermal conductivity (0.037–0.051 W/(m·K)), CO₂ capture and in-situ mineralization capability, and improved flexural and compressive strength. Collectively, these attributes position the developed materials as strong candidates for next-generation sustainable construction application.