

## Exploring amorphous aluminosilicates at the nanoscale

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Understanding the formation mechanisms and phase stability of amorphous aluminosilicates is extremely important for a range of geological and industrial processes including zeolites, glasses and low-CO<sub>2</sub> geopolymer cements. However, due to their disordered nature at the atomic length scale it is difficult to elucidate the exact structural rearrangements occurring during formation. Here, two state-of-the-art theoretical and experimental approaches will be outlined, and their suitability for studying amorphous aluminosilicates will be discussed.

Novel modeling and simulation methods across length scales are emerging in the research community, yet it remains difficult to span length scales without a significant compromise in accuracy. Coarse-grained Monte Carlo simulations used in conjunction with quantum chemical thermodynamic calculations is a relatively new methodology capable of spanning the atomic and nanoscale. Here, it will be applied to modeling (i) silicate speciation (oligomerization) as a function of concentration at high pH, and (ii) the geopolymerization reaction, revealing new structural insight on the formation mechanisms taking place. The advantages and disadvantages of using this multiscale simulation methodology will be outlined and compared with conventional simulation approaches.

Experimental pair distribution function analysis is a powerful tool capable of elucidating the local structural motifs present in amorphous materials. This technique is well-suited for studying the structural arrangements in amorphous aluminosilicates, including glasses and cements. Here, by utilizing pair distribution function analysis it will be shown that the atomic structure of calcium-aluminum-silicate-hydrate gel (found in alkali-activated slag cements) is intrinsically different to calcium-silicate-hydrate gel known to exist in ordinary Portland cement (OPC)-based concrete. This fundamental difference (amorphous versus nanocrystalline) draws into question the suitability of OPC-based approaches (thermodynamic modeling, atomic ordering, phase formation) for studying alkali-activated cement systems.