Structure-property relationships of alkali-activated geopolymer materials for sustainable construction applications

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Alkali-activated materials (AAMs), also known as geopolymers, are a class of inorganic binders studied for use as sustainable construction materials. Traditional cement production has an immense carbon footprint, approaching 1 ton of CO_2 emitted per ton of cement produced, with over 4 billion tons of cement produced annually (~8% of anthropogenic CO_2 production). Alternatively, geopolymers are synthesized via in-situ resource utilization (ISRU) of locally available materials including clays, fly ash, and slag, resulting in a CO_2 emissions reduction of 30-80% compared to traditional cements. Geopolymers chemically transform from a reacting colloidal suspension to a final composite material with excellent compressive strength and resistance to chemical and thermal degradation. Targeted formulation and processing of geopolymers in the initial paste state can have enormous effects on the early-age rheology and final material properties of this class of materials. Thus, this dissertation applies principles from polymer and colloid science to mechanistically design geopolymer materials for targeted applications.

A primary challenge hindering the widespread adoption of geopolymer technology is a limited understanding of how chemical composition and processing history influence early-age rheological properties and ultimate mechanical performance. The need to tailor early-age material properties is particularly pronounced in emerging additive manufacturing techniques for construction, which require precise control of rheological properties throughout the processing life cycle in addition to sufficient final compressive strengths. The goal of this thesis to is to experimentally connect formulation chemistry, nano-to-microscale structure, and material properties of geopolymers as a necessary step toward the design of sustainable, low-CO_2 construction materials. A combination of materials characterization techniques including rheology, small-angle scattering, and NMR measure structure-property relationships of model geopolymer binders and compare results with scaling theories from the polymer gel literature. Results also inform ongoing collaborative projects with machine-learning experts at Northeastern University and multi-scale simulations of colloidal gels in partnership with researchers at Georgetown University.

In Aim 1 of this thesis, the material property development and reaction kinetics of a model geopolymer are experimentally measured using a combination of rheological techniques, solid-state NMR, and isothermal calorimetry. NMR and calorimetry results provide a time-dependent extent of reaction and identify the chemical structures formed as the binder develops mechanical strength. A semi-empirical model is constructed to quantitatively connect temporal rheological properties to the amount of reaction product formed in the binder. Following successful characterization of early-age metakaolin geopolymer curing in the quiescent state, the rheological response to applied shear is then measured. Critical to inform processing design for this class of materials, the measured critical gel point of the geopolymer defines the “open time” or workability window of the binder. While the application of extended shear processing significantly impacts rheological properties prior to the critical gel point, final material properties are maintained if processing ends before the measured gel point.

For geopolymers which contain sodium as the primary cation, the main reaction product responsible for connectivity in the binder is a sodium aluminosilicate gel (N-A-S-H gel). In Aim 2, N-A-S-H gels are synthesized from dilute sodium silicate and sodium aluminate solutions as a model system to interrogate the initial stages of the geopolymer polycondensation reaction. In collaboration with scientists at the NIST Center for Neutron Research, a new small-angle x-ray scattering (SAXS) sample environment

is adapted to measure the early-age structure of a reacting aluminosilicate gel from timescales of milliseconds to hours. Paired with rheological measurements of gelation, this technique provides a complete picture of the nucleation and growth mechanisms of the geopolymer gel as a function of composition. Master curves for rheological property development and the volume fraction of fractal aggregates unify the behavior of a wide gel composition space using simple scaling arguments. The evolving N-A-S-H gel structure is also connected to temporal rheological properties via the Shih scaling model and the Avrami kinetic equation, which universally describe the N-A-S-H gel reaction across a wide composition space.

Finally, in Aim 3, geopolymers are designed for potential lunar construction applications via the in-situ resource utilization of the aluminosilicate regolith present on the lunar surface. The compressive strength and chemical structure (^{29}Si and ^{27}Al NMR) of Black Point 1 lunar regolith simulant geopolymers are measured, defining relationships between composition and material properties. Four high-strength samples are flown to the International Space Station as part of the 20th Materials International Space Station Experiment (MISSE) for exposure to low-earth orbit conditions for a period of six months. Combined mechanical testing and imaging analyses of samples exposed to low-earth orbit prove the durability of this class of materials for long-term lunar construction and confirm a material Technology Readiness Level of 5. In summary, the novel quantitative relationships discovered in this dissertation enable mechanistic design of sustainable geopolymer binders with targeted final material properties for both terrestrial and lunar use.