

# Composition-property relationships of geopolymer materials for lunar in-situ resource utilization and sustainable construction applications

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There is a global dependence on cement and concrete to develop and maintain the modern world and the built environment. Behind only water, concrete is the most widely consumed material in the world, with a yearly production of ~4 billion metric tons of cement. With global manufacturing of concrete accounting for 8-9% of global CO<sub>2</sub> emissions, there is a need for alternative construction materials. Approximately 240,000 miles away, distinct yet related challenges exist for construction on the Moon. With a global surge in initiatives explicitly intended to establish a sustained human presence on the lunar surface (e.g., the Artemis Program from NASA), significant engineering challenges arise in the development of lunar infrastructure, including landing surfaces, radiation shielding, and storage facilities. Traditional terrestrial construction methods are not feasible due to chemical incompatibility of lunar regolith, payload limitations and cost, and the harsh environment of the lunar surface. Thus, extensive in-situ resource utilization (ISRU) will be required for viable lunar construction materials. Geopolymer materials present opportunities for both these terrestrial and extraterrestrial challenges.

Geopolymers are a class of inorganic aluminosilicate alkali-activated materials (AAMs), synthesized from an aluminosilicate precursor and an alkaline solution, that react to form solid cement-like binders. These materials can be synthesized from aluminosilicate sources including calcined clays, industrial waste products such as fly ash, as well as lunar regolith. The formation of these materials can result in a reduction of CO<sub>2</sub> emissions up to 80% compared to traditional cements and have excellent mechanical performance, thermal stability, and acid resistance. Relevant for lunar applications, unlike traditional cements, the geopolymerization process is net-water neutral and water is not incorporated in the final binder structure.

Variations in precursor composition, overall formulation, and processing can significantly influence the reaction kinetics, resulting chemistry, and final material properties of geopolymer materials. A deeper understanding of these relationships connecting the properties to the composition and processing is therefore critical for designing geopolymers for targeted applications. Therefore, the overarching goal of this dissertation is to advance the domain knowledge of geopolymer materials through the development of quantitative relationships between composition, processing, and resulting properties relevant for sustainable construction alternatives and lunar ISRU. This work integrates experimental investigations and machine learning approaches to improve our understanding and predictive capabilities of these materials to accelerate terrestrial and space-based applications. To couple learnings for both the relevant applications in lunar ISRU and terrestrial construction, the aluminosilicate source of Black Point One (BP-1) is the primary focus of this work as it is both a lunar regolith simulant and classified as a terrestrial basalt. Other lunar regolith simulants and metakaolin are investigated as well, albeit to a lesser extent.

Aim 1 of this dissertation establishes composition-property relationships for how the overall chemical composition impacts the observed chemistry and material properties of geopolymers formed from aluminosilicate sources relevant to the lunar environment. This is accomplished through a

comprehensive study of BP-1 geopolymer's chemical structure and material properties, an additional investigation of another lunar regolith simulant (LHS-1), and curation of a dataset of the compressive strength of lunar regolith simulant geopolymer from the literature. Establishing relationships between the composition and resulting properties of geopolymers allows for the rational design and optimization of materials for lunar ISRU applications.

Aim 2 of this dissertation establishes relationships between parameters involved in the construction of the final geopolymer material that, independent of the chemical composition of the aluminosilicate source and activating solution, influence the resulting properties. This was accomplished through three independent studies. First, the impact of BP-1 particle size on geopolymer kinetics and properties was characterized, identifying the Sauter mean diameter as the best particle size descriptor. Second, microwave curing of BP-1 geopolymers was evaluated against traditional curing protocols, drastically accelerating strength development. Finally, in collaboration with the Air Force Research Laboratory, the effect of a superplasticizer dosage on phosphate-based geopolymer rheology, chemistry, and properties was quantified. Understanding how these additional parameters influence material properties can complement the relationships with the chemical composition to further optimize properties.

Aim 3 of this dissertation advances the development of geopolymer materials towards lunar applications and establishes predictive frameworks for geopolymer properties. Multi-fidelity neural networks (MFNNs) are developed and effectively demonstrated for modeling complex fluid rheology, with minimal experimental data requirements. This framework is successfully applied to the steady-state behavior of geopolymer materials. Next, building on the results of Aim 1 and Aim 2, a predictive machine learning framework is developed for the compressive strength of lunar regolith simulant geopolymers from their composition and processing conditions. Finally, four geopolymers (two lunar regolith simulant geopolymers, one Martian regolith simulant geopolymer, and a metakaolin geopolymer) were affixed to the exterior of the International Space Station (ISS) as a part of the Materials on the ISS Experiment (MISSE) 20 mission. The durability of these materials after exposure to low-Earth orbit for six months demonstrates the viability of this class of materials for lunar applications. Overall, the efforts from this dissertation link experimental understanding with predictive modeling to guide the intelligent design of geopolymer materials for applications as sustainable construction materials and in lunar ISRU.