Investigation of Sol-Gel Synthesized Phosphate-based Geopolymers: a ReaxFF study

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Abstract: Geopolymers have gained widespread interest due to their diverse applications, such as coatings, fire-resistant materials, and composites for infrastructure reinforcement. These materials are regarded as environmentally friendly alternatives to conventional Portland cement, due to their lower carbon dioxide emissions, and comparable mechanical strength. The mechanical strength of geopolymers is attributed to $AlPO_4$ phases formed in their matrix. Our collaborator, Dr. Gadikota's group at Cornell University prepared geopolymers experimentally using a sol-gel approach by mixing Si_2Al_2Ox with phosphoric acid (H_3PO_4) to obtain the geopolymers with a composition having Si:Al:P ratio of 1:1:1. Understanding the evolution of Si-Al linkages during the synthesis of geopolymers, and formation of Al-P linkages are essential to develop predict controls over the chemistry of these geopolymers. To obtain an atomistic scale understanding of this evolution process, we are employing molecular dynamics simulations using the ReaxFF reactive force field. In these ReaxFF simulations, a system with a periodic boundary condition consisting of $Al_2Si_2O_7$ and Phosphoric acid has been modeled. We extended a previously developed LiTiAlPOH ReaxFF parameter set with Si-parameters and trained our force field against a set of Quantum Mechanics data. To overcome the AlPO phase formation timescale, we performed Replica Exchange Molecular Dynamics (REMD) simulations. Analyzing the results of the REMD simulations shows an increasing trend in radial distribution function (RDF) between Aluminum-Phosphorus atoms which indicate the formation of Al-O-P linkages and subsequently the formation of $AlPO_4$. RDF analysis also reveals the Si phase segregation. Comparing the RDF and Angle distribution of the silicon cluster formed in REMD simulations with the most stable silicon oxides shows it to be close in structure to Quartz and Cristobalite. The findings from this study contribute to a deeper understanding of how the composition of geopolymers influences their properties, paving the way for the development of next-generation construction materials.