Blue Waters Annual Report 2017 – Research Summary Submission
Due: June 27, 2017

Corresponding Author Contact Email and Name:

NAME: Ange-Therese Akono

EMAIL: aakono@illinois.edu

Title: Multi-scale and Multi-Physics Modeling of The Strength of Geopolymer Composites

PI: Ange-Therese Akono, Department of Civil and Environmental Engineering, University of Illinois at Urbana-Champaign

Co-PI(s):

Erman Guleryuz, National Center for Supercomputing Applications, University of Illinois at Urbana-Champaign

Seid Koric, National Center for Supercomputing Applications, Department of Mechanical Science and Engineering, University of Illinois at Urbana-Champaign

Waltraud M. Kriven, Department of Materials Science and Engineering, University of Illinois at Urbana-Champaign

Executive Summary:

Geopolymers are a class of inorganic polymeric, X-ray amorphous materials consisting of alumina, silica, and alkali metal oxides [1]. Geopolymer composites exhibit appealing properties such as high thermal stability [2], small carbon dioxide footprint [3], and high strength, which makes them suitable for many applications including alternative cementitious materials [4], passive cooling systems, passive cooling systems [5], low-level nuclear waste encapsulation [6], and potential novel biomaterials. The research goal is to understand the impact of nanoporosity on the stiffness and strength via molecular dynamics and finite element modeling. Nanoporosity is governed by the monovalent cation and the water content. In contrast, microporosity results from the mixing procedure and from poor bonding between the geopolymer matrix and the filler phase. To increase performance of geopolymer composites several strategies were found: reduce the microporosity, select strong inclusions, increase the volume content of stiff inclusions, or select fillers with a high aspect ratio. These findings pave a new way toward novel, high-performance and multi-functional composites.

Key Challenges:

Geopolymer composites are multi-scale materials as shown in Figure 1. At the molecular, the building units consist of [SiO_4] and [AlO_4]. At the nanometer length-scale, the amorphous aluminosilicate consists of nano-size micelles and nanopores. At the mesoscale, particulate, fibrous or woven reinforcement is embedded I nthe amorphous geopolymer matrix. An important challenge at the molecular level is to reproduce the amorphous and nanoporous structure. Furthermore, describing the interaction between molecules via a physics-based interatomic potential is difficult. At the microscopic scale, formulating the constitutive behavior of the geopolymer matrix has never been done. Finally, at the mesoscale, understanding the influence of particulate interaction and particulate-matrix bonding on the overall strength and stiffness remains a challenge.

Why It Matters:

Geopolymer composites constitute an emerging class of composites with a great potential in civil engineering, mechanical engineering, aerospace, navy, automobile, and biomedical engineering. However, the widespread application and acceptance of geopolymer systems has been so far impeded by many roadblocks such as: (i) the lack of long-term durability data [7], (ii) lack of in-service track record, and (iv) lack of standard geopolymer cements [8]. The computational approach selected based on molecular dynamics and finite elements offer a cost-effective and time-efficient means to accelerate discovery and innovation.

Accomplishments

At the nanoscale, atomic simulations were performed in LAMMPS as shown in Figure 2 b). Various molecular structures were recreated using the Avogadro and Packmol software, for diverse alkali metal cations and Si:Al ratios. The amorphous structure was obtained through a melting and quenching sequence. Afterwards uniaxial tensile and compression tests along with shear tests were simulated to yield the strength behavior and failure micromechanisms. The software OVITO was utilized for visualization. At the mesoscale, the finite element package Abaqus was used., see Figure 2 a) The microstructure was generated from OOF2D and MATLAB based on scanning electron microscopy observations of geopolymer composites. The constitutive behavior of the individual constituents was prescribed based on independent experiments. Tensile tests with prescribed periodic boundary conditions were simulated to yield the effective response.

The strength and stiffness of amorphous geopolymer were correlated to the chemistry and the density. The nanoporosity was found to be a function of the alkali and Si:Al ratio. At the mesoscale, the mechanical resistance of geopolymer composites to permanent deformation was elucidated as a function of the type, nature, size, and shape of the inclusions. Our theoretical and computational framework was validated on 31 different geopolymer-based systems, based on experiments carried out by various researchers over 7 years. To our knowledge, it is the first time that a theoretical upscaling model has been proposed to upscale the constitutive behavior of

geopolymers. In future efforts, we will model the evolution of the fracture toughness of pure geopolymer for various densities via molecular dynamics.

Why Blue Waters?

The Blue Waters platform was essential to carry-out our molecular dynamics simulations as well as our finite element simulations. Standard desktop work stations do not have the required memory and computational power to resolve the complex systems involved. For molecular dynamics, it was crucial to be able to simulate large systems in a timely fashion. In the case of finite element simulations, the non-linearity of the equations due to friction, contact and plastic flow, made it impossible to obtain results using a desktop workstation. In both cases, the Blue Waters supercomputer with high-performance computing has enabled us to obtain novel results that bring new insights into the origins of strength and toughness in geopolymer composites.

Next Generation Work:

At the molecular scale, further simulations will be carried out to elucidate the resistance to fracture propagation of geopolymer as a function of the monovalent cation and Si:Al ratio. At the mesoscale, Abaqus Explicit will be employed to perform 3-D simulations.

References

[1] Davidovits, J., Properties of geopolymer cements. In *First international conference on alkaline cements and concretes* 1, (1994), pp. 131-149.

[2] Barbosa, V.F. and MacKenzie, K.J., Synthesis and thermal behaviour of potassium sialate geopolymers. *Mater. Lett.*, 57:9 (2003), pp.1477-1482.

[3] McLellan, B.C., Williams, R.P., Lay, J., Van Riessen, A. and Corder, G.D., 2011. Costs and carbon emissions for geopolymer pastes in comparison to ordinary portland cement. *Journal Cleaner Product.*, 19: 9 (2011), pp.1080-1090.

[4] Moon, J., Bae, S., Celik, K., Yoon, S., Kim, K.H., Kim, K.S. and Monteiro, P.J. Characterization of natural pozzolan-based geopolymeric binders, *Cem. Concr. Comp., 53* (2014), pp.97-104.

[5] Sukontasukkul, P., Nontiyutsirikul, N., Songpiriyakij, S., Sakai, K. and Chindaprasirt, P. Use of phase change material to improve thermal properties of lightweight geopolymer panel, *Mater. Struct.*, 49:11 (2016), pp.4637-4645.

[6] Davidovits, J., Environmentally driven geopolymer cement applications. In *Proceedings of 2002 Geopolymer Conference,* Melbourne. Australia, October, 2002.

[7] Sakulich, A.R., 2011. Reinforced geopolymer composites for enhanced material greenness and durability, *Sustain. Cit. Soc.*, 1:4 (2011), pp.195-210.

[8] Davidovits, J. Geopolymer cement. *A review. Geopolymer Institute, Technical papers*, 21 (2013), pp.1-11.

Publications and Data Sets

Kataruka, A., S., Koric, W., Kriven, A.-T., Akono, Strength Properties of Particulate Potassium-Based Geopolymer Composites. *ASCE Engineering Mechanics Institute Meeting*, San Diego, CA, June 4th-7th, 2017.

Cui, Y., E., Guleryuz, A.-T., Akono, Investigation of the Mechanical Properties of geopolymer binders using molecular dynamics simulations. *ASCE Engineering Mechanics Institute Meeting*, San Diego, CA, June 4th-7th, 2017.

Kataruka, A., E., Guleryuz, S., Koric, W., Kriven, A.-T., Akono, Representative Elementary Volume Modeling of Geopolymer Composites. *14th U.S. National Congress on Computational Mechanics*, Montreal, QC, Canada, July 17th -20th, 2017

Cui, Y., E., Guleryuz, S., Koric, W., Kriven, A.-T., Akono, Molecular dynamics study on the mechanical and fracture properties of geopolymer binders. *8th Advances in Cement-Based Materials American Ceramic Society*, Atlanta, GA, June 26th-28th, 2017.

Kataruka, A., E., Guleryuz, S., Koric, W., Kriven, A.-T., Akono, A Parametric Study of Strength Properties of Particulate Potassium-Based Geopolymer Composites. *8th Advances in Cement-Based Materials American Ceramic Society*, Atlanta, GA June 26th-28th, 2017.

Kataruka, A., S. Koric, E. Guleryuz, W. M. Kriven, and A.-T. Akono, Multiscale Strength Homogenization of Geopolymer Composites. *Cement and Concrete Composites*. In Preparation.

Cui, Y., E. Guleryuz, W. M. Kriven, S. Koric, and A.-T. Akono, Elastic and strength properties of geopolymer precursor via atomistic modeling. *Physical Review B*. In Preparation.

Images:

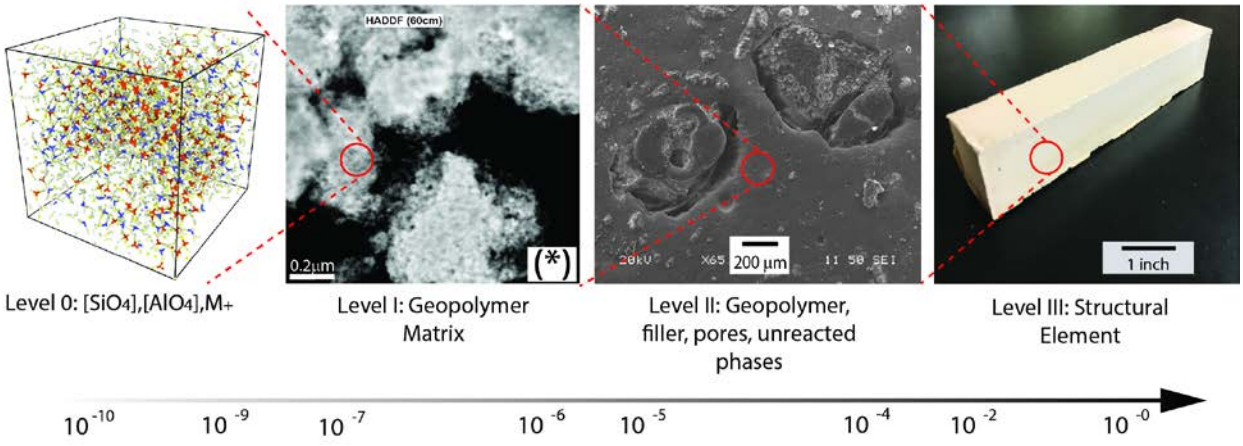

Figure 1: multi-scale nature of geopolymer composites. Our study spans levels 0 at the nanometer length-scale to level III at the macroscopic level.

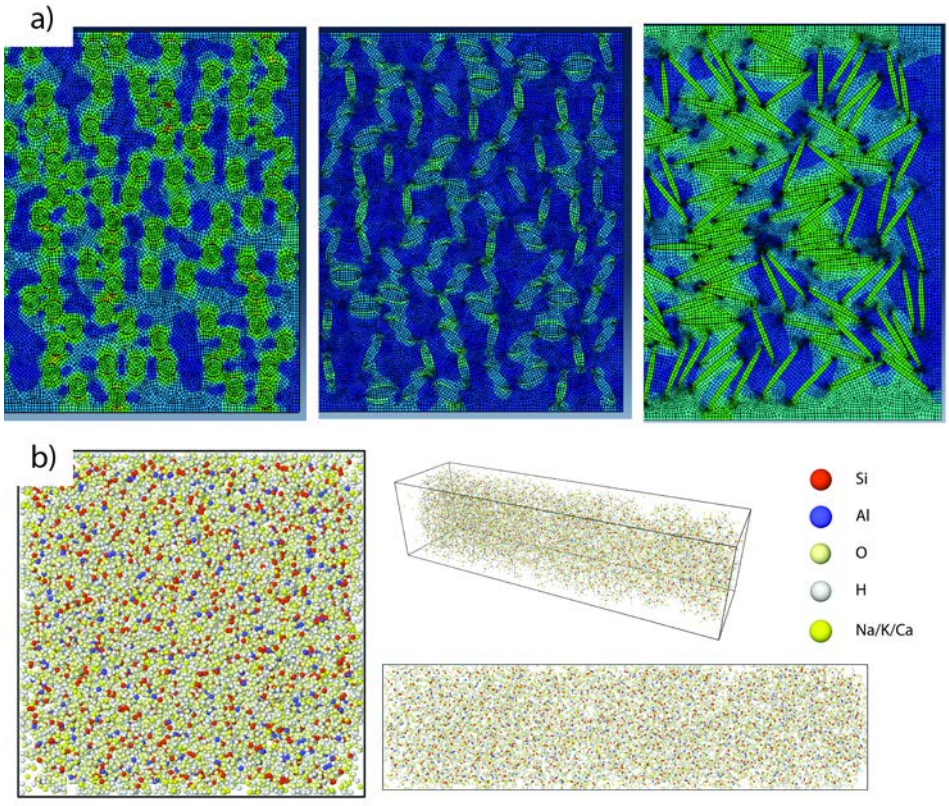

Figure 2: a) FEM model at mesoscale showing the stress distribution as a function of the filler aspect ratio. b) Molecular dynamics modelling of inorganic polysialate disiloxo.