Molecular dynamics simulation to better understand environmentally-friendly geopolymer from wastes

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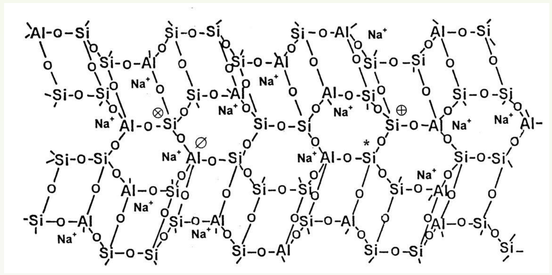
Background

The production of ordinary Portland cement (OPC) consumes extensive amount of energy and emits greenhouse gases such as CO2. Studies have shown that production of 1 ton OPC causes 900 kg of CO2 emission. In order to find an alternative for OPC, researchers have studied the utilization of industrial byproducts such as mine tailings from mining operations, fly ash from coal fired power plants, and concrete waste to produce an environmentally-friendly high strength cementitious material called 'geopolymer'. Geopolymer can not only show comparable performance of OPC but also reduce the carbon footprint on earth because it requires much smaller amount of energy for its production than OPC. The problem of disposing the large amount of industrial wastes like mine tailings, fly ash and concrete waste is solved greatly by using them to produce the geopolymer cement.

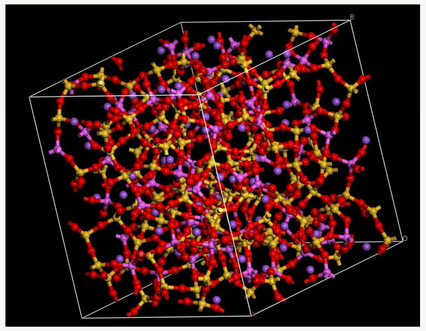
Geopolymer is a new type of "cement" formed by aluminosilicates in reaction with an alkaline solution, having an amorphous to semi-crystalline polymeric network structure with interconnected –Si–O–Al– bonds. Alkali cations such as Na+ act as a charge balancing media for the amorphous aluminosilicate network. Geopolymer not only provides performance comparable to ordinary Portland cement in many applications but shows additional advantages such as rapid development of mechanical strength, good volume stability, excellent adherence to aggregates, superior resistance to chemical attack, and ability to immobilize contaminants.

Research Approach

In this research, Sodium Aluminosilicate glasses (NAS) has been used as a proxy of geopolymer binder phase in order to better understand the main factors that control the properties of geopolymer from an atomic scale.



Geopolymer structure as originally proposed by Joseph Davidovits



NAS glass (representing the geopolymer binder phase) model for Si/Al = 2.0; red is oxygen, blue is sodium, yellow is silicon, purple is aluminum.

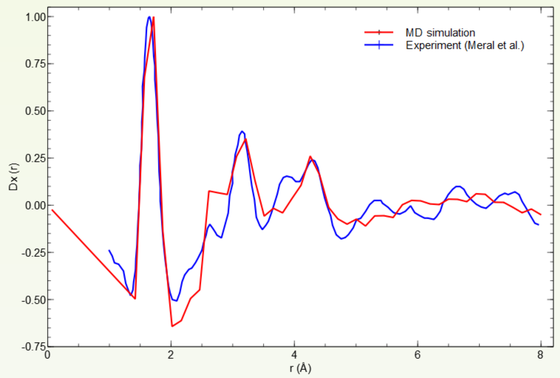
Analysis and Results

Inter-atomic potential model

A pairwise partial ionic charge model in tandem with a Morse potential (Pedone et al. J. Phys. Chem. B. 110 (2006) 11780–11795) was used for representing interatomic interactions. The form of the potential is given as: $U(r) = \frac{z_i z_j e^2}{r} + D_{ij}[\{1 - e^{-a_{ij*(r-r_o)}}\}^2 - 1] + \frac{C_{ij}}{r^{12}}$

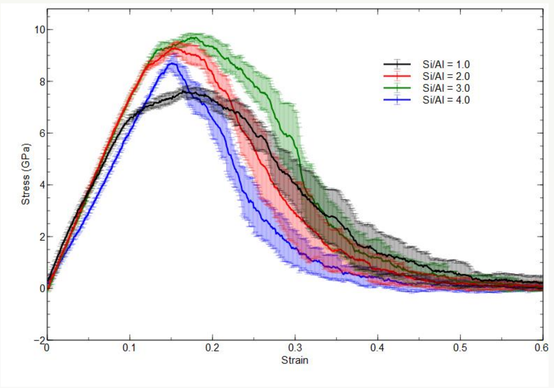
Interaction type (superscripts show the charges)	D_{ij} (eV)	a_{ij} (Å^{-2})	r_o (Å)	C_{ij} (eV Å^{12})
$Na^{0.6}$-$O^{-1.2}$	0.023363	1.763867	3.006315	5.0
$Al^{1.8}$-$O^{-1.2}$	0.361581	1.900442	2.164818	0.9
$Si^{2.4}$-$O^{-1.2}$	0.340554	2.006700	2.100000	1.0
$O^{-1.2}$-$O^{-1.2}$	0.042395	1.379316	3.618701	22.0

X-ray Pair Distribution Function



Simulated and experimental X-ray Pair distribution function shows good match between the simulated NAS glass and geopolymer

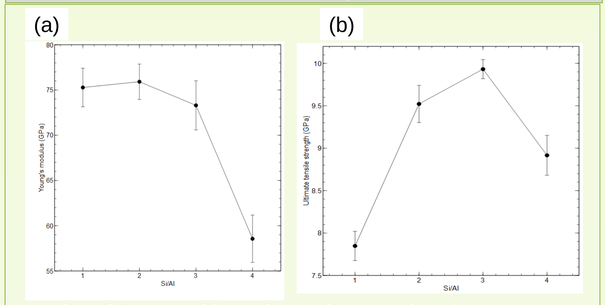
Uniaxial Stress vs. Strain



Uniaxial stress vs. strain for different Si/Al ratio; error bars shows the standard deviation of 12 different simulations

Results (Cont.)

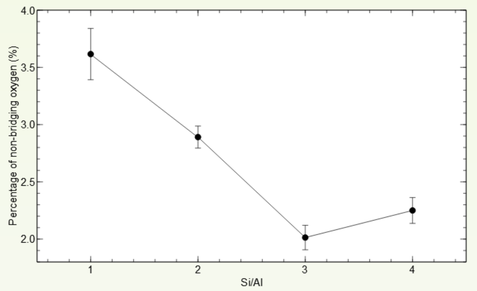
Mechanical Properties



Mechanical Properties of the Na-aluminosilicate geopolymer binder; a) Young's modulus vs. Si/Al ratio b) Ultimate tensile strength (UTS) vs. Si/Al ratio

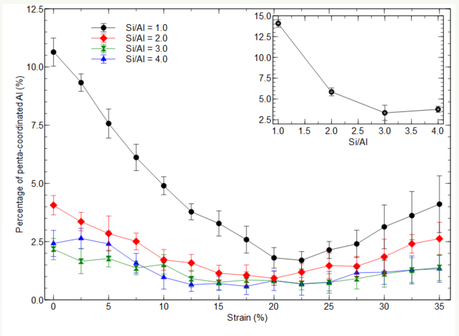
The trends in the Young's modulus, UTS and failure behavior can be explained by the structure associated at the respective Si/Al ratios.

Effect of Non-bridging oxygen



Percentage of non-bridging oxygen (NBO) versus Si/Al ratio at zero strain condition.

Effect of Penta-coordinated Al

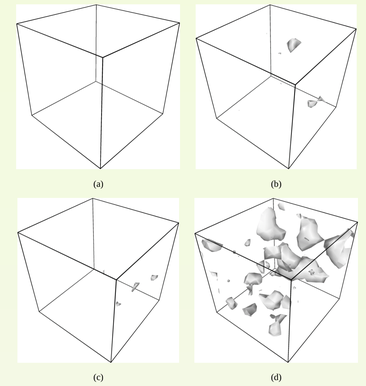


Percentage of penta-coordinated Al evolution as a function of strain for different Si/Al ratios. (The inset shows the percentage of penta-coordinated Al at zero-strain; Si/Al=1 has the highest and Si/Al=3 has the lowest percentage of such penta-coordinated Al.)

NBO works as a weak point of the Si and Al tetrahedral network resulting in the lowering of Young's modulus and UTS of the glass structure. The intrinsic bulk modulus of pristine alumina and NBO together determines the Young's modulus of the system. While the presence of penta-coordinated Al and NBO both are responsible for the lowering of UTS.

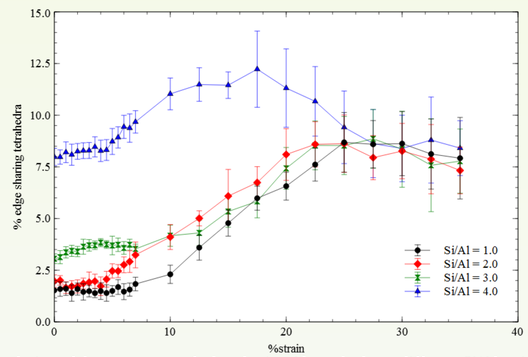
Results (Cont.)

Effect of void sizes on the Mechanical Properties



Visual representation of void sizes using a probe radius of 3 Å at 0% strain: a) Si/Al =1; b) Si/Al =2; c) Si/Al =3; and d) Si/Al =4 (Si/Al = 4 includes voids up to 10 Å in diameter).

Effect of edge sharing tetrahedra



Evolution of the percentage of edge sharing Al tetrahedra at different Si/Al ratios.

The synergistic interplay of void size and edge sharing tetrahedrons plays role in the observed brittle failure mechanism of the NAS glass. As the material is strained, the voids grow further, promoting the formation of more edge-sharing tetrahedra, with the Si/Al = 4 glass having the largest percentage of edge-sharing tetrahedra for all strains prior to failure.

Conclusions

- ❖ The mechanical properties of geopolymer binder phase is significantly influenced by the associated Si/Al ratio.
- ❖ The optimum Si/Al ratio for highest mechanical strength is observed at an Si/Al ratio of 2-3 consistent with experimental observations.
- ❖ The presence of NBO, Penta coordinated Al, edge sharing Al tetrahedra can all lead to the lowering of tensile strength and Young's modulus of the system.
- ❖ Increasing void size and edge sharing tetrahedra promotes the brittle failure of the system.