Characterization And Modeling Of Glass Chemistry-Structure Relations

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T.R. Zeitler[†], M.T. Brumbach, T.M. Alam, L.J. Criscenti[ɤ], D.F. Susan, J.M. Rodelas , and K.G. Ewsuk

Materials Science and Engineering, [†]Nuclear Energy & Fuel Cycle Programs, and [ɤ]Geochemistry Centers
Sandia National Laboratories, Albuquerque, NM 87185

Objective

- Develop experimentally-validated modeling/simulation tools to predict/control glass and glass interface chemistry-structure-property relations.

Approach

- Characterize & model chemistry-structure relations in a simple 3-component glass.
- Extend the characterization & modeling to more complex glasses and interfaces.
- Test, refine, & validate modeling/simulation by comparison to experiment.

Application/Impact

- Improved performance and reliability glasses and interfaces in glass-to-metal seals, solid oxide fuel cells, glass-bonded ceramics (e.g., debased aluminas, and low temperature cofired ceramic, LTCC, packaging).

Simulation & Characterization Tools

	Property	Computational Modeling	Experimental Approach	Elemental	Chemical State	Structure
STRUCTURE	bulk structure (glass short-range and medium-range order); surface/interface bond structure	classical MD	MAS-NMR		*	*
			FTIR; Raman		*	*
			XANES; EXAFS	*		*
	interface microstructure; devitrification		MAS-NMR		*	*
			XRD			*
			Auger; XPS; SIMS	*	*	*
CHEMISTRY	bulk & interface composition/gradients; diffusion/diffusion profiles	classical MD	Auger; XPS; SIMS	*	*	
			Microprobe; TEM; SEM/EDS	*		
			XRF	*		
	reactivity/reactive sites e.g., hydrolysis of glass bonds	*ab initio* MD; mean force calculations	FTIR / Raman		*	*
			XPS	*	*	*
			NMR		*	*
			NEXAFS	*	*	*
PHYSICAL	glass & composite properties e.g., heat capacity, CTE, viscosity	classical MD; physical models e.g., property & processing	FTIR / Raman		*	
			MAS-NMR		*	*
			Thermal Analysis			
			Wetting/Spreading			

MD Simulation of 25 BaO – X Al_2O_3 – (75-x) SiO_2 Glasses

Using a Pedone* Multicomponent Force Field within The LAMMPS** MD Code

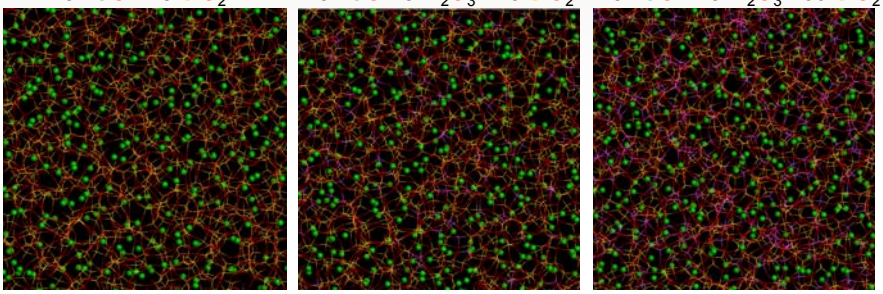
* A Pedone et al., "A new self-consistent empirical interatomic potential model for oxides, silicates, and silica-based glasses", J Phys Chem B, **110**, 11780-11795 (2006).
** S Plimpton, "Fast Parallel Algorithms for Short-Range Molecular-Dynamics, J Comp Phys, **117** [1], 1-19 (1995).

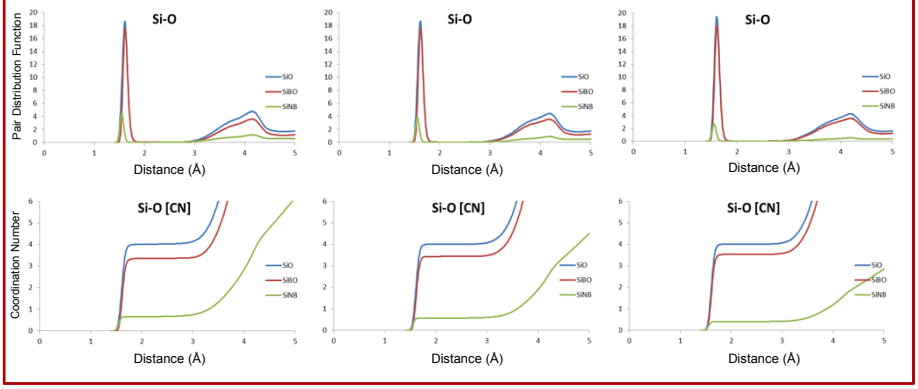
Glass	Oxide Mole %			Density (g/cc)	Number of Atoms (in 1x/4x Simulation)					Box Length (Å)
	BaO	Al_2O_3	SiO_2		Ba	Al	Si	O	Total	
BAS1	25		75	3.52	275/1100	0/0	825/3300	1925/7700	3025/12100	35.108/55.731
BAS2	25	5	70	3.25	275/1100	110/440	770/3380	1980/720	3125/12540	36.354/57.709
BAS3	25	15	60	3.22	275/1100	330/1320	660/2640	2090/8360	3355/13420	37.053/58.818

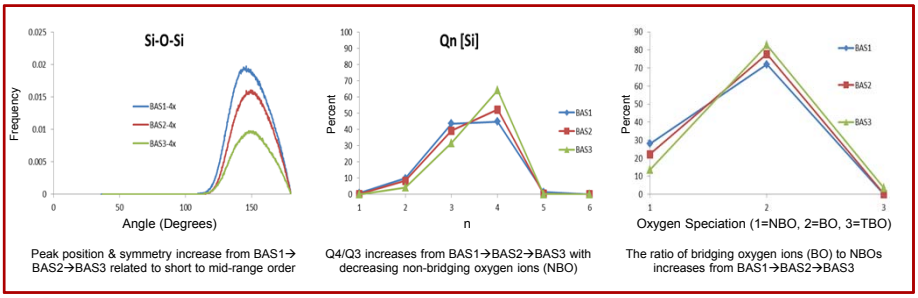
BAS1
25 BaO - 75 SiO_2

BAS2
25 BaO - 5 Al_2O_3 - 70 SiO_2

BAS3
25 BaO - 15 Al_2O_3 - 60 SiO_2





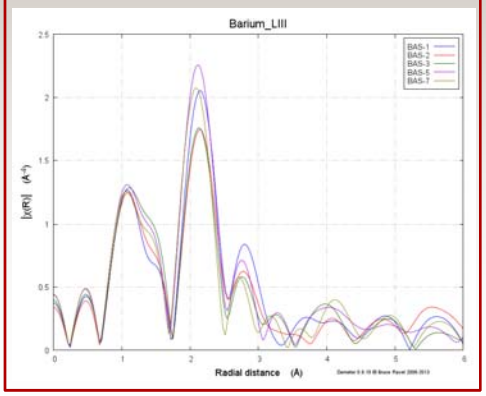


Peak position & symmetry increase from BAS1→BAS2→BAS3 related to short to mid-range order

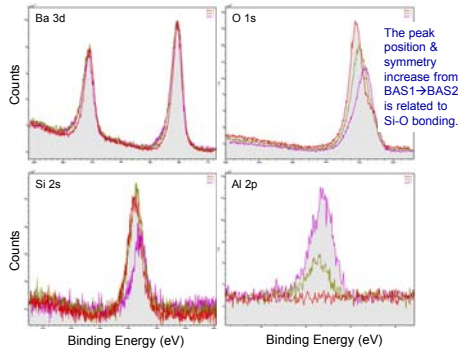
Q4/Q3 increases from BAS1→BAS2→BAS3 with decreasing non-bridging oxygen ions (NBO)

The ratio of bridging oxygen ions (BO) to NBOs increases from BAS1→BAS2→BAS3

EXAFS Radial Distribution Functions



X-ray Photoelectron Spectroscopy



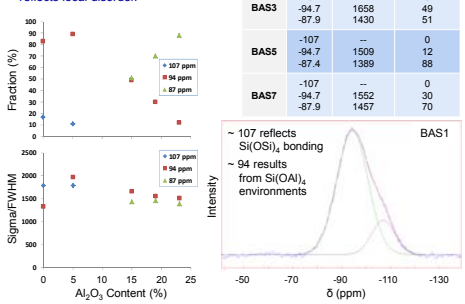
The peak position & symmetry increase from BAS1→BAS2 is related to Si-O bonding.

^{29}Si MAS NMR Spectra

The chemical shift reflects the change in the Si-O bond length related to the local glass network modifier concentration.

The peak/line width (FWHM) reflects local disorder.

Sample	δ (ppm)	FWHM (Hz)	Fraction(%)
BAS1	-107.1 -94.5	1792 1326	17 83
BAS2	-107.1 -94.7	1792 1971	11 89
BAS3	-107 -94.7 -87.9	-- 1658 1430	0 49 51
BAS5	-107 -94.7 -87.4	-- 1509 1389	0 12 88
BAS7	-107 -94.7 -87.9	-- 1552 1457	0 30 70



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