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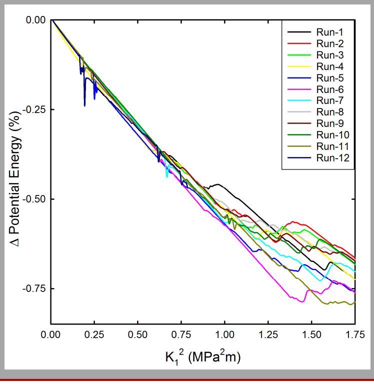

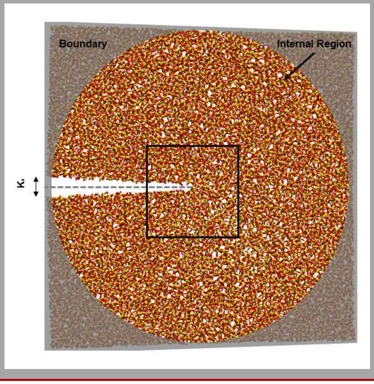

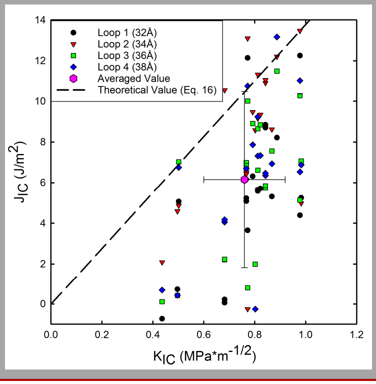

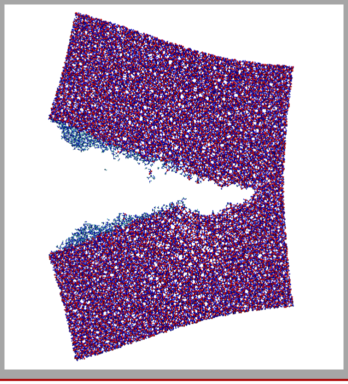

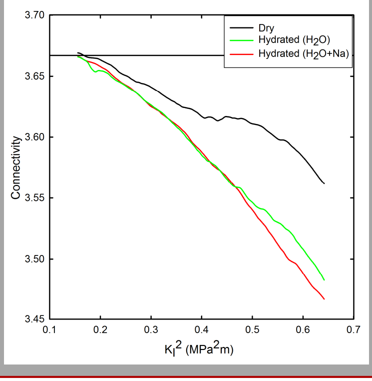

Fracture toughness of amorphous silica from atomistic scale simulations

J.M. Rimsza, R.E. Jones, L.J. Criscenti

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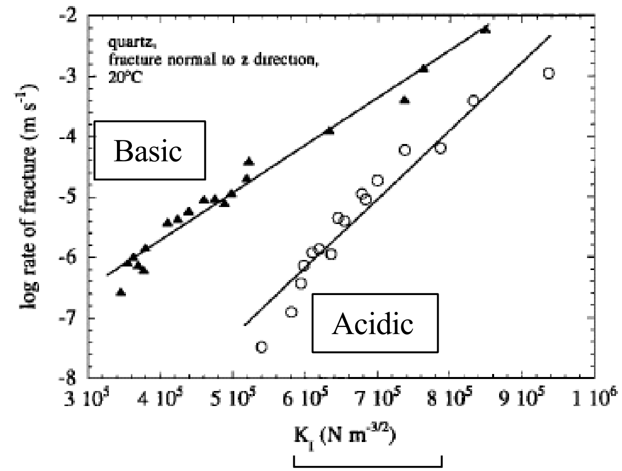
Technical Motivation

- Brittle fracture of silicates affect the stability and reliability of amorphous systems making prediction of the mechanical response difficult

- Develop fundamental understanding of the chemical-mechanical mechanisms that control subcritical cracks in silicates

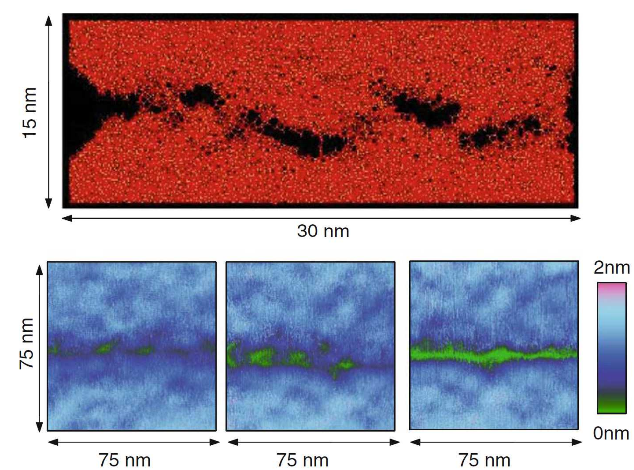
- Link atomic-scale insight to macroscale observables and directly address how chemical environment alter mechanical behavior



K_I = stress intensity factor (N m$^{-3/2}$)

P. M. Dove, *J. Geophys. Res.* (1995)

- Why atomistic simulations?
  - Cracks start at the atomistic scale by the breaking of bonds at the solid-fluid interface.
  - Crack tip formation and propagation is influenced by fluid and surface chemistry (development of surface charges and adsorbed species)



Bonamy, Daniel, et al. *Int. J. Fract.* (2006)

Computational Methods

- Classical molecular dynamics for large scale simulation of silica fracture
- ReaxFF: bond order based forcefield including reactive water and silica bond breakage and formation

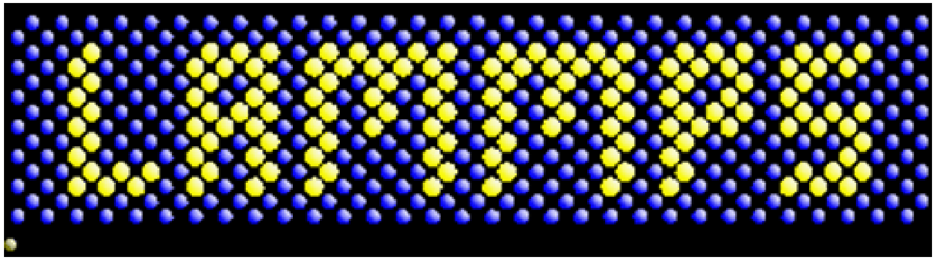
Fogarty, Joseph C., et al. *J. Chem. Phys.* (2010)
Yeon, Jejoon, and Adri CT van Duin. *J. Phys. Chem. C.* (2015)

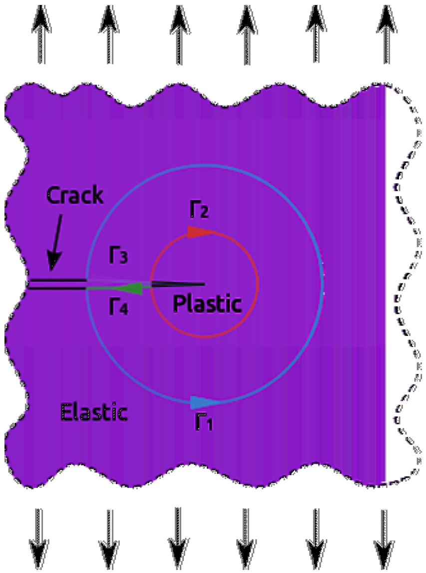
$$E_{Total} = E_{Bond} + E_{Over} + E_{Under} + E_{LP} + E_{Val} + E_{Pen} + E_{Tors} + E_{Conj} + E_{VDW} + E_{Coul} \quad (1)$$

- J-integral and the fracture toughness from the stress, displacement and energy densities of atomistic system coarse grained onto a grid

Jones, Reese E., et al. *J. Phys.: Condens. Matter* (2010)

- Eshelby stress (S) is calculated and the J-integral is evaluated around a loop around the crack tip $J = \int_{\partial\Omega} S \cdot dA$
- Multiple loop sizes confirm that the in-elastic zone is completely enclosed and path independent
- Atoms-to-Continuum (ATC) package allows for calculation of the J-integral and the fracture toughness from atomic stresses and is available as a USER-ATC LAMMPS Package



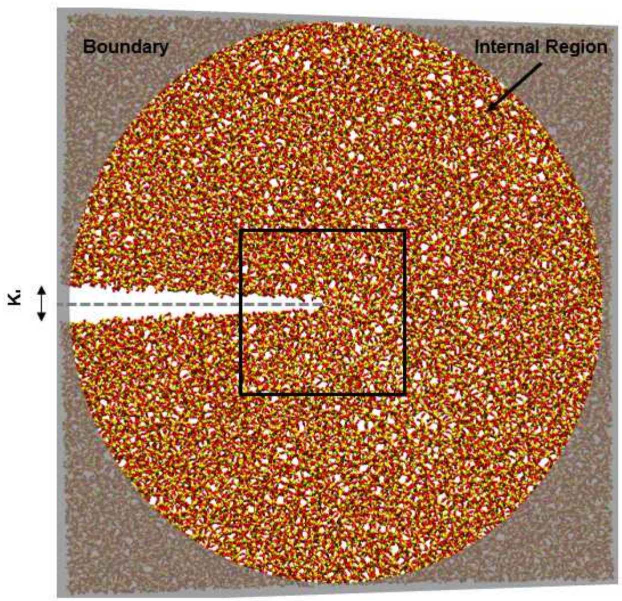


Bhanerje, "J-integral contours for an elastic material" 9/4/2013

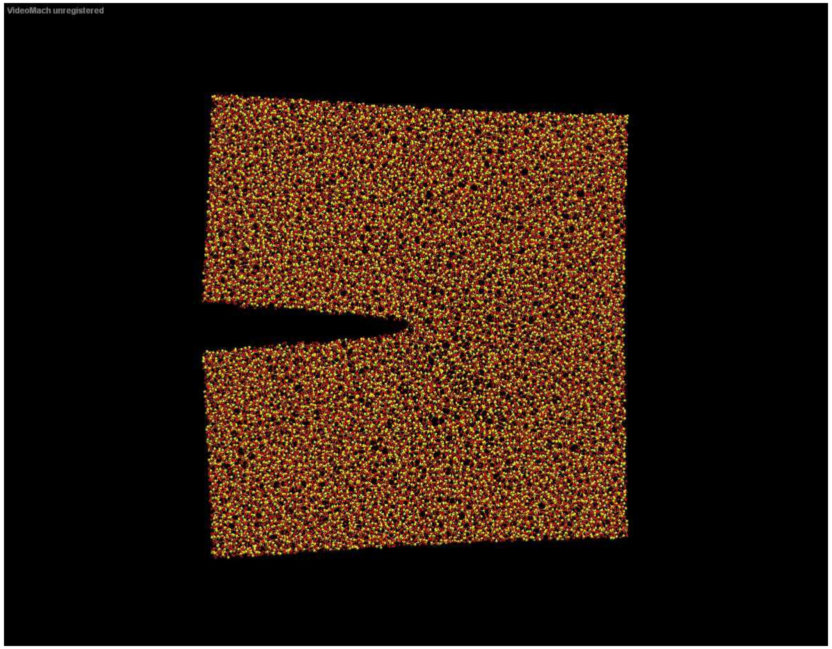
Schematic of J-integral demonstrating the inclusion of the plastic region inside an elastically responding matrix

Simulation set-up

- Amorphous silica, system size: 38400 atoms (143Åx143Åx28Å)
  - Melt and quenched from β-cristobalite system
  - 12 different cracks (three different silica structures with four different crack locations)
- Slit crack is formed by removing neighboring, creating a singular high stress condition
- Boundary atoms are fixed and atomic positions are adjusted to introduce far-field loading as a mode I fracture
- Interior atoms are allowed to freely move by integration with a microcanonical (NVE) ensemble
- Atomic positions adjust to accommodate the added stress
- Stress is introduced iteratively by increasing the crack width



Schematic of silica slit crack, crack width, as well as boundary and internal regions. Atoms: oxygen (red), silicon (yellow)



Opening of silica slit crack
Atoms: oxygen (red), silicon (yellow)

Fracture and Energy Dissipation

- Fracture propagates in distinct steps when the stress at the crack tip exceeds the strength of the material
- Perfectly brittle fracture will have no dissipation energy, with all energy used to propagate the fracture
- Dissipation energy calculated from the total energy of the system and added surface energy

$$\frac{\Delta U}{\Delta S_A} = G_{diss}$$

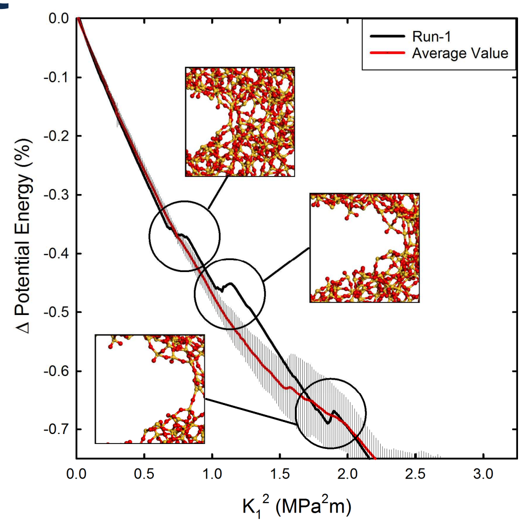
- Local inelastic behavior as energy is introduced into the matrix during loading and not completely dissipated once fracture occurs

$$G = 2\gamma_s + G_{diss}$$

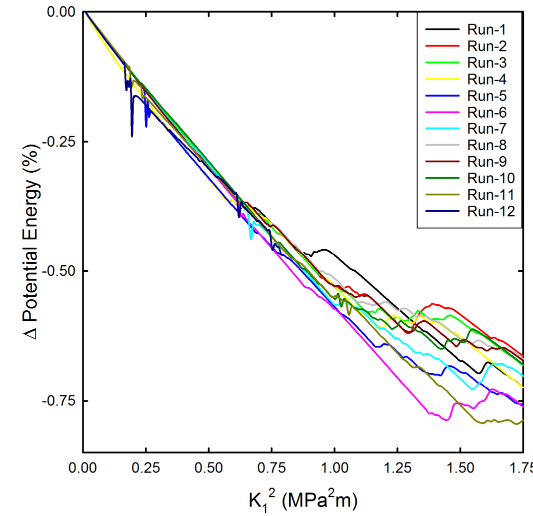
- Surface energies (γ_s): 2-1.2 J/m^2 varying with surface structure and relaxation

Rimsza, J. M. et al. *Langmuir* (2017).

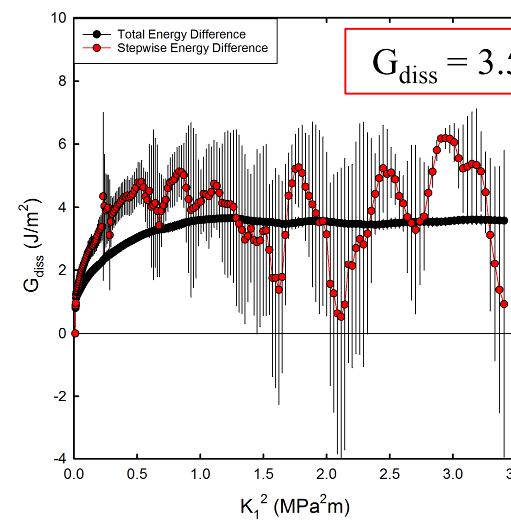
Estimated G values are ~5.9-7.5 J/m^2



Potential energy change with loading for a single simulation (Run-1) and averaged



Potential energy change for individual simulations with loading of 0-1.75 K_I^2 (MPa^{2}m)



G_{diss} = 3.5 J/m^2

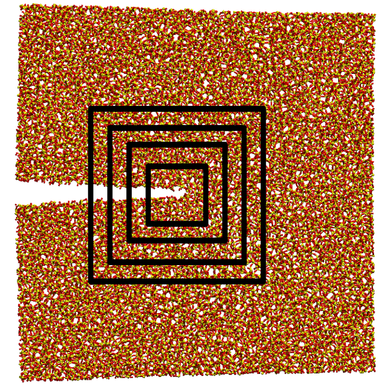
Energy dissipation (G_{diss}) during loading and subsequent crack propagation in amorphous silica

J-Integral Calculation

- Calculated via the AtC method through coarse graining energy, displacement, and stress
  Jones, Reese E., et al. *J. Phys.: Condens. Matter* (2010)
- J_{IC} value = J-integral when the crack begins propagating

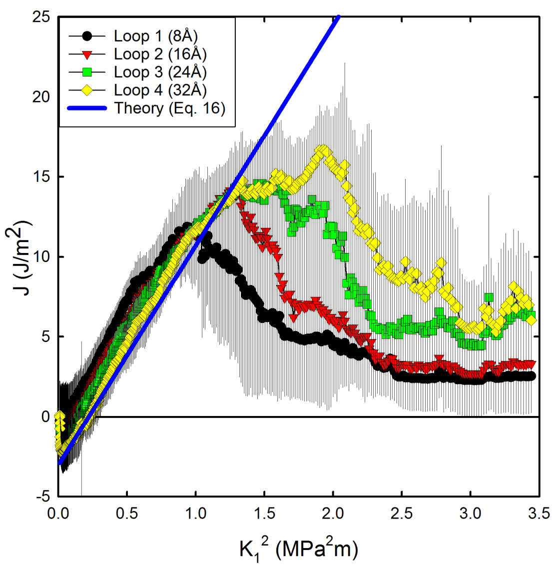
$$J_{IC} = \frac{K_{IC}^2}{E^*} \qquad E^* = \frac{2\mu}{1-\nu}$$

- J-integral converges at loop sizes of ~3 nm approximates the size of an inelastic zone

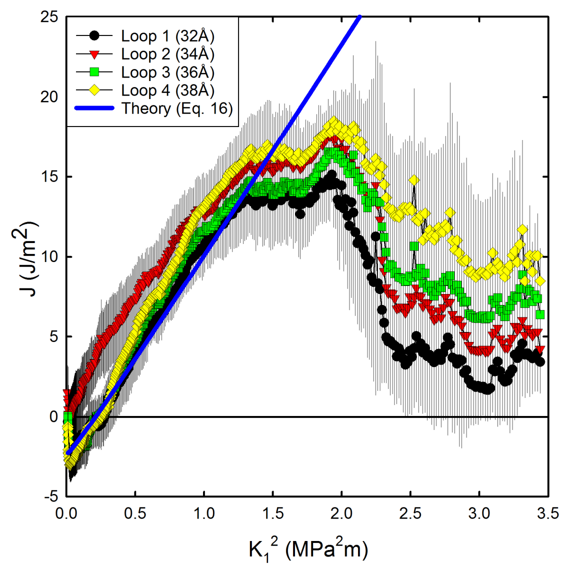


Schematic of increasing loop sizes for J-integral convergence test

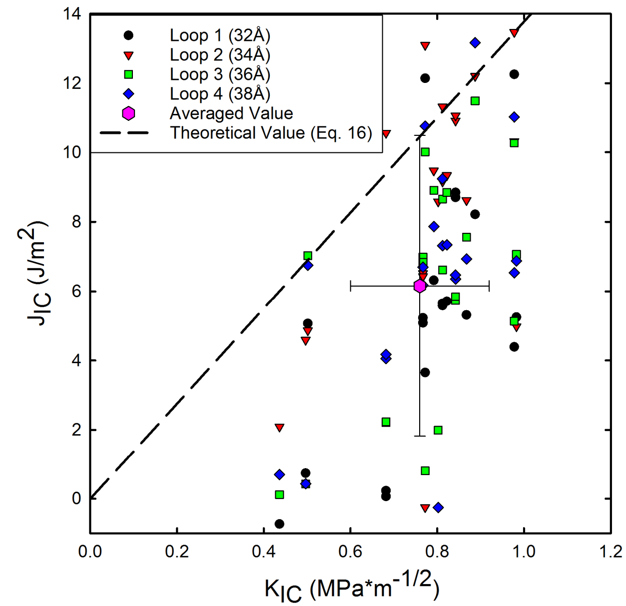
K_{IC}=0.76±0.16 MPa√m
J_{IC}=6.16±4.34 J/m²



J-integral with increasing loop sizes



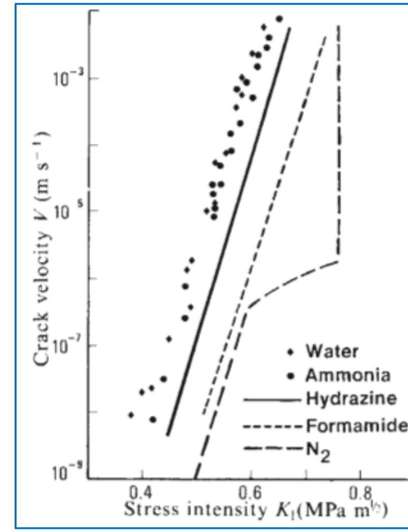
J-integrals with converged loop sizes



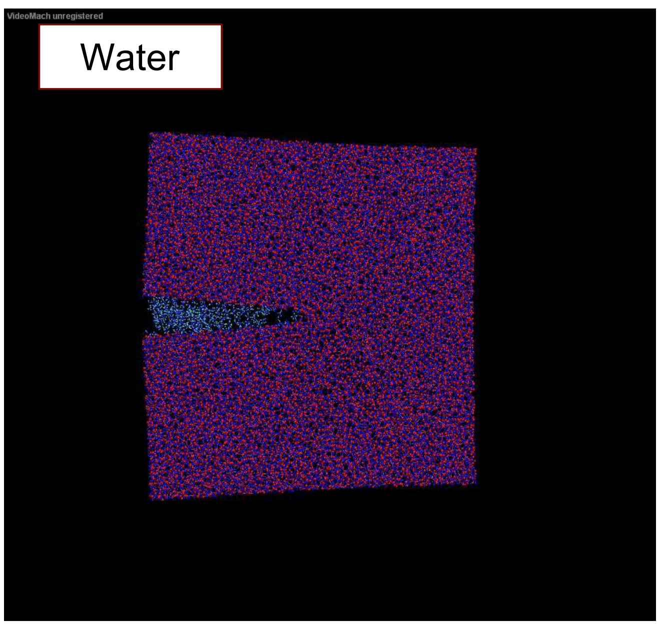
K_{IC} and J_{IC} values for the initiation of crack propagation in amorphous silica

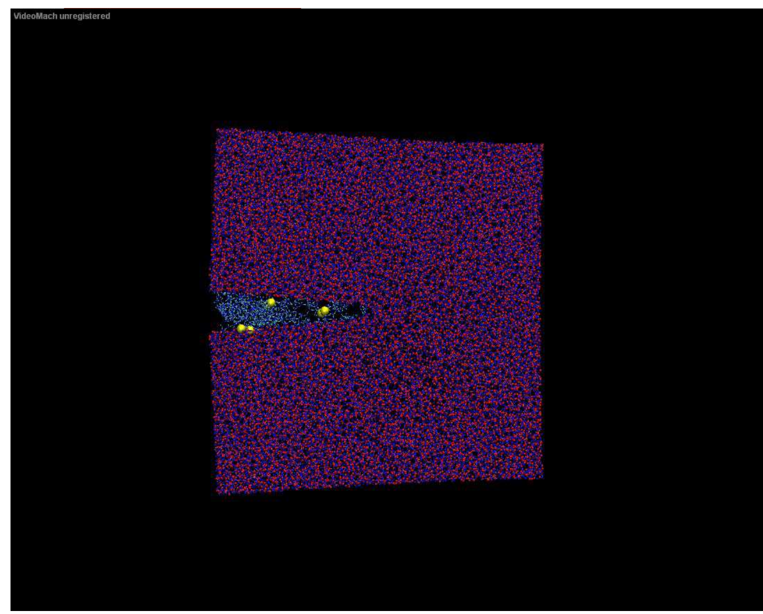
Fracture: Aqueous Conditions

- Coupled chemical-mechanical processes inside the crack affects the propagation rate of cracks in silicates
- Method:
  - Introduce thin film of water wetting the crack surfaces
  - Iterative loading of the crack with silica and water relaxation
  - Water molecules added via GCMC to maintain surface wetting
  - Can vary the fluid composition to investigate the effect of dissolved salts on fracture (dissolved NaCl, pH)
  - System size: 38400-42000 atoms



T.A. Michalske & S.W. Freiman, *Nature* (1982)

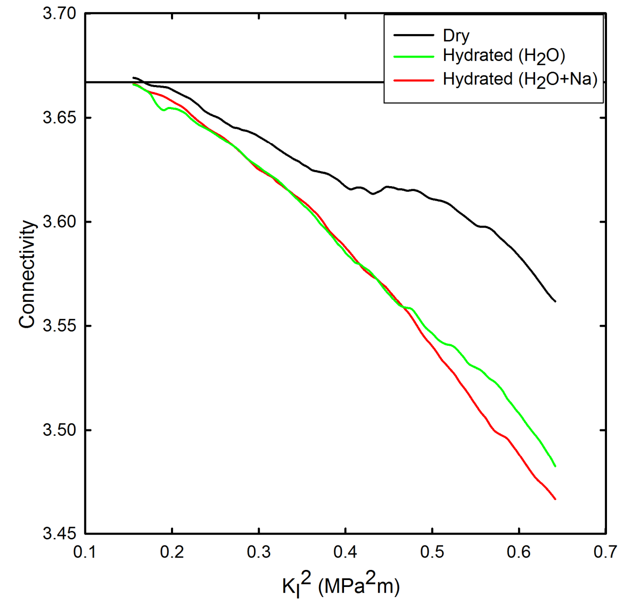




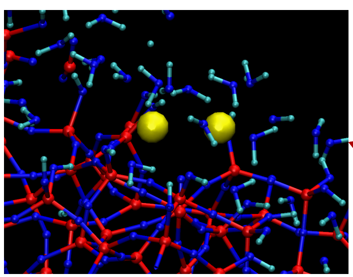
Opening of silica slit crack in a hydrated environment
Atoms: oxygen (red), silicon (purple), hydrogen (blue), sodium (yellow)

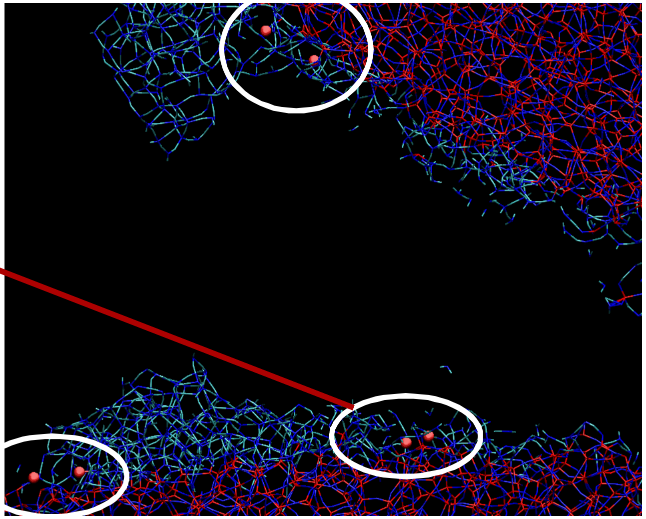
Connectivity and Na Environment

- Connectivity:
  - Water is facilitating Si-O bond breakage
  - Increased surface healing in in dry systems
  - Sodium impacts recovery of surface connectivity
  - Investigation of local environments of sodium and impact on fracture is ongoing
- Na Environment:
  - Sodium does not migrate to the crack tip, but may be impacting the composition of the solution, altering fracture properties



Connectivity of the system with loading for silica in three different environments (dry, hydrated, hydrated with sodium)





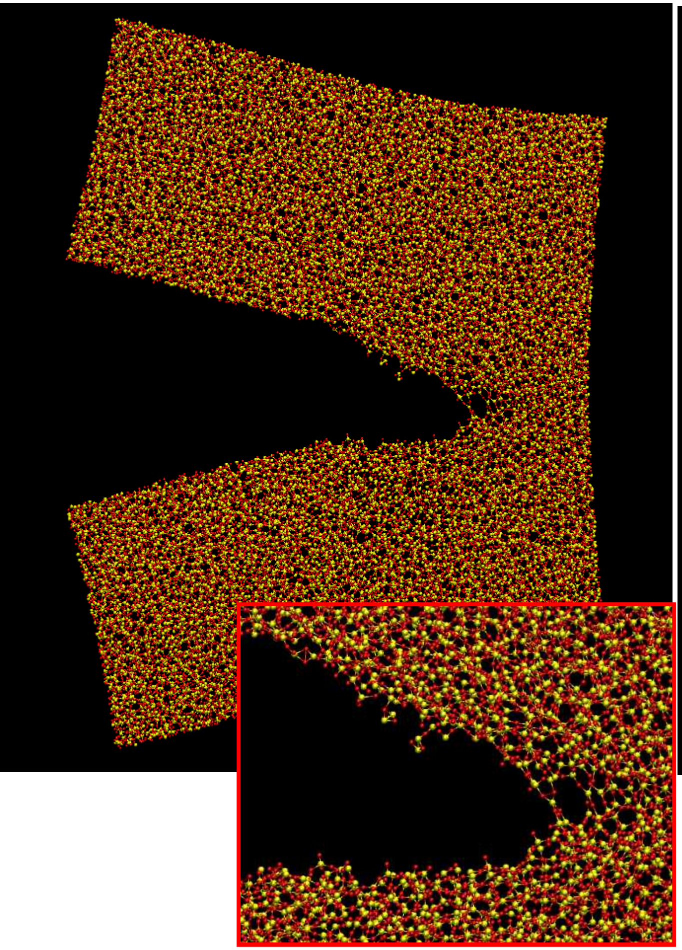
Na Environment

Coordination Number	5
Charge	0.83-0.92
Na-O distance	2.64±0.18 Å

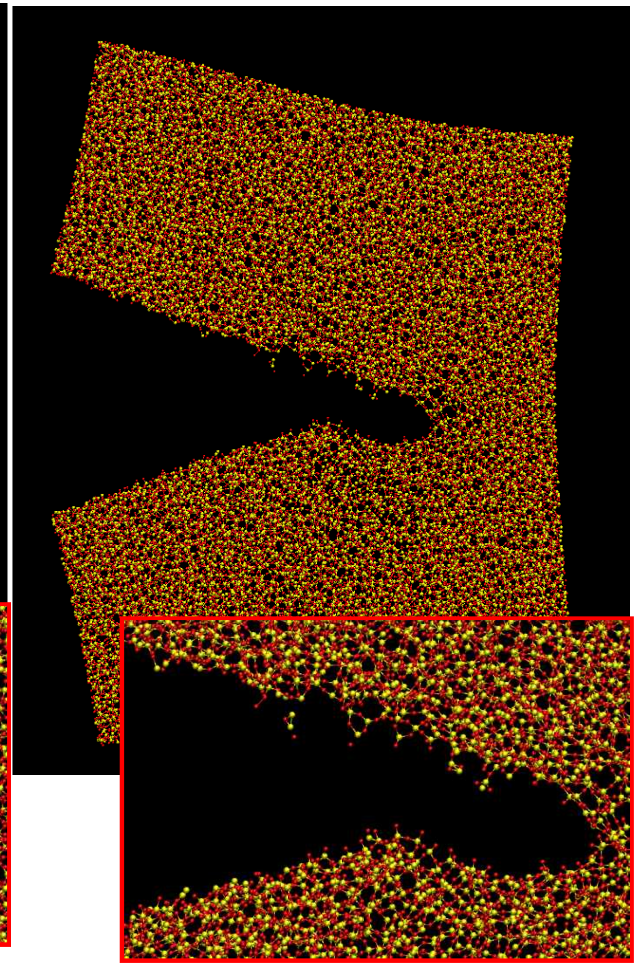
Crack Tip Shape

- Narrowing of crack tip with hydration and introduction of cations
- Possible capillary effects altering surface structure and stability during fracture

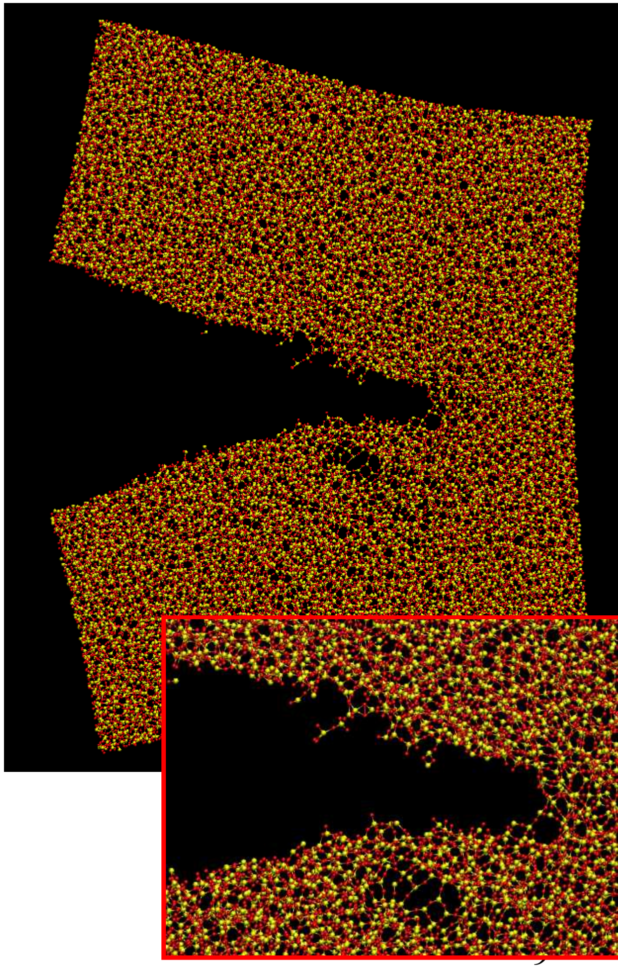
Dry



Hydrated (H_2O)

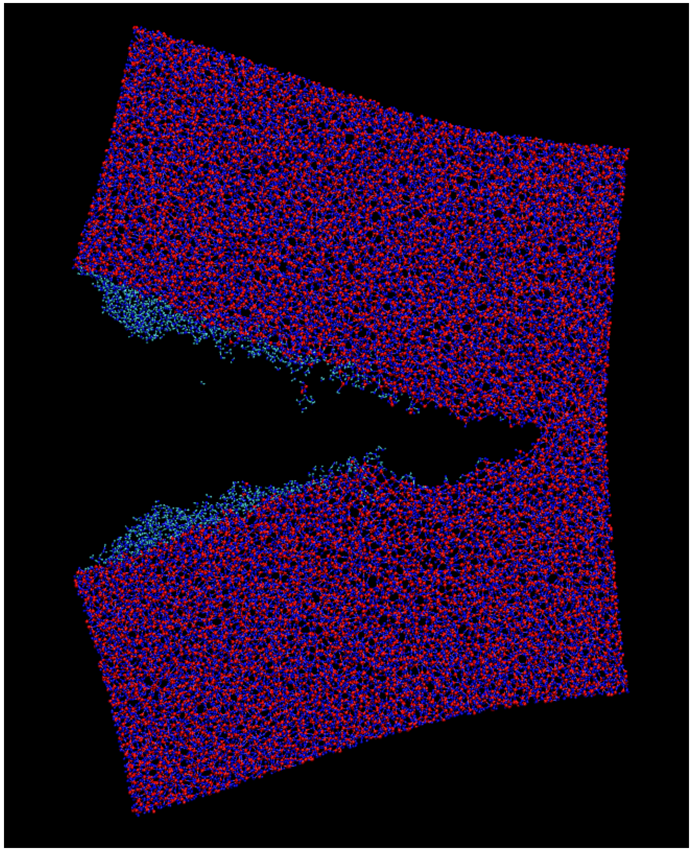


Hydrated (H_2O+Na)



Conclusions

- Dry silica fracture:
  - Development of localized high stress inelastic region prior to fracture
  - Inelastic zone estimated with a radius between 3-3.2 nm
  - K_{IC} of ~0.78 MPa√m consistent with experimental results
- Atoms-to-Continuum methods appropriate for evaluating macroscopic fracture properties from atomistic scale information
- Hydrated silica fracture:
  - Decreasing connectivity of the structure during crack propagation
  - Changing morphology of the crack tip



Snapshot of fracture silica in a hydrated environment

Current Work

- Reacting fluids and pH effects on silica fracture
  - Varying brine compositions
  - Changing levels of surface wetting
  - Fracture properties
- Layered mineral and clay structures

Acknowledgements

This work was fully supported by the Laboratory Directed Research and Development (LDRD) program of Sandia National Laboratories.

Back Up Slides

http://thediagram.com/12_3/thethreemodes.html

Zhu, X, and J.A. Joyce. *Eng. Fract. Mech.* (2012)

Introduction to fracture mechanics

- Cracks propagate when the stress at the crack tip exceeds the strength of the material (Griffith criterion)

$$\sigma = \left(\frac{2E\gamma}{\pi a}\right)^{\frac{1}{2}}$$

A.A. Griffith, *Phil. Trans. R. Soc. A* (1921)

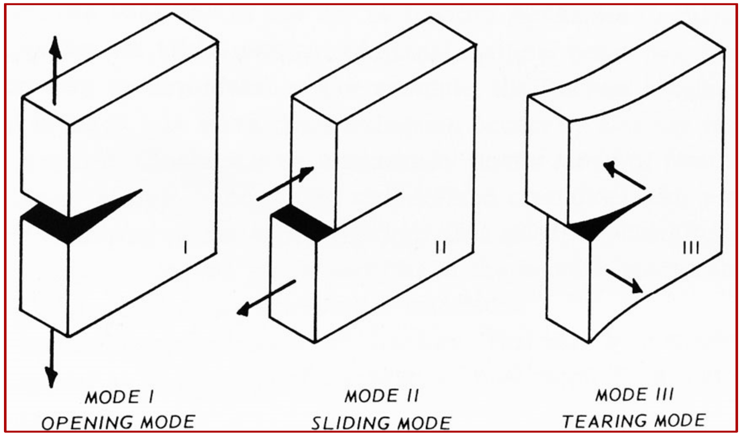
- Fracture toughness (K): the energy required to propagate a crack in a material

- Strain energy release rate (G): energy dissipated during fracture per unit of surface area

$$G = \frac{K_c^2(1-\nu)^2}{E} = 2\gamma_s \qquad \text{(brittle material)}$$

$$G = G_{diss} + 2\gamma_s \qquad \text{(material with inelastic behavior)}$$

- J-integral: method of calculating G for monotonic loading through a path independent contour integral
  - For a linear elastic (non-yielding) material G=J
  - Bulk material property
  - In Mode I: $J_{IC} = G_{IC} = K_{IC}^2\left(\frac{1-\nu^2}{E}\right)$
    - E: elastic modulus
    - ν: poission's ratio
- Modes of loading
  - Mode 1: Opening
  - Mode 2: Sliding
  - More 3: Tearing



http://thediagram.com/12_3/thethreemodes.html

Zhu, X, and J.A. Joyce. *Eng. Fract. Mech.* (2012)

Crack Tip Blunting

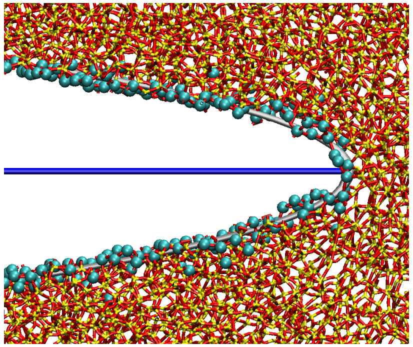
- Crack tip blunting indicates an even stress distribution and the development of local inelastic behavior
- Strength of a material is proportional to the radius of curvature of the crack tip (ρ):

$$\sigma_{max} \propto \sqrt{\frac{1}{\rho}}$$

C.E. Inglis, Spie Milestone series MS (1997)

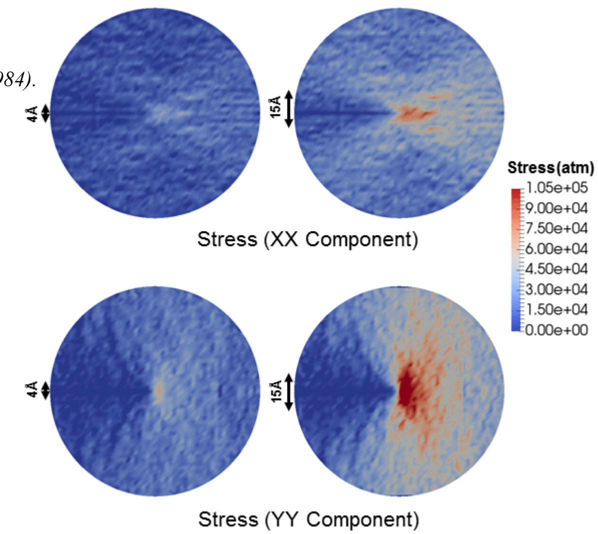
- To calculate ρ the internal atoms of the crack tip are fit with a parabolic function, from which ρ is calculated: *A. Howard, Calculus: With analytic geometry, John Wiley 1988.*

$$\rho = \frac{(1 + f'(x)^2)^{\frac{3}{2}}}{f''(x)}$$

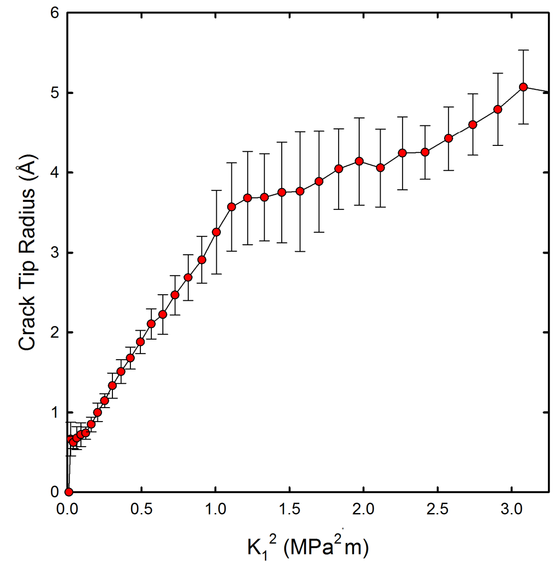


Silica structure (red and yellow) atoms in quadratic fit (turquoise), parabola estimating the shape of the crack tip (silver), and horizontal axis (blue)

- Radius of curvature: 0.1Å-5Å, experimental value: 15Å *Bando et al. J. Amer. Cera. Soc. 67.3 (1984).*
- Increasing ρ with increasing load, changing distribution of stress around the crack tip
- Stress fields indicate increasing stress around the crack tip prior to fracture, due to incorporation of stress around the crack tip
- Highlights the size of the inelastic zone and some development of a high stress conditions ahead of the crack tip



Stress fields calculated from individual atomic stresses. Averaged over twelve different trajectories.



Radius of curvature and crack length during brittle fracture in silica . Error bars are the standard deviation