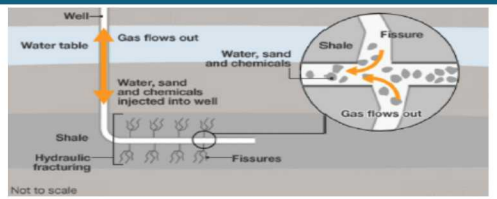
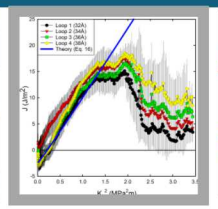
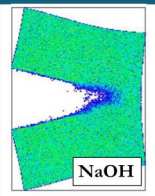
SAND2018-8182C

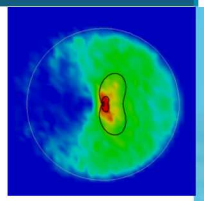
Sandia National Laboratories

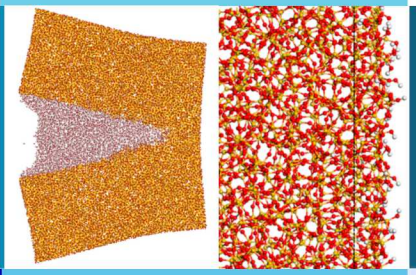
Assessing the Fracture Strength of Geological and Related Materials in Fluid Environments via an Atomistically Based J-integral











13th World Congress on Computational Mechanics, July 22-27, 2018, New York, NY

PRESENTED BY
Reese E. Jones, Louise J. Criscenti, Jessica M. Rimsza, **Jonathan Zimmerman**

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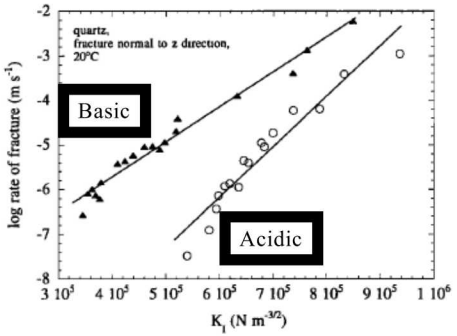
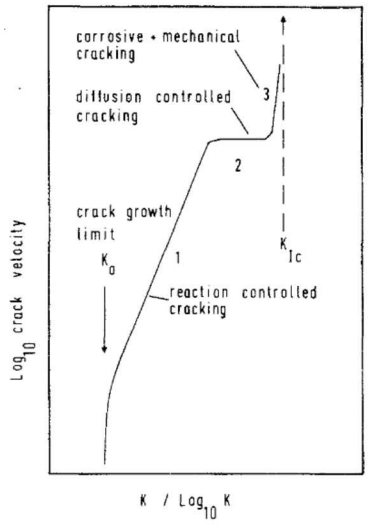
Motivation

- 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)

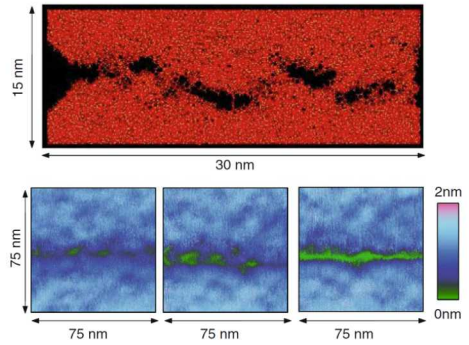


Atkinson, Barry Kean. "A fracture mechanics study of subcritical tensile cracking of quartz in wet environments." *Pure and Applied Geophysics* 117.5 (1979): 1011-1024.

What are the chemical and mechanical aspects of fracture?

Why atomistic simulations?

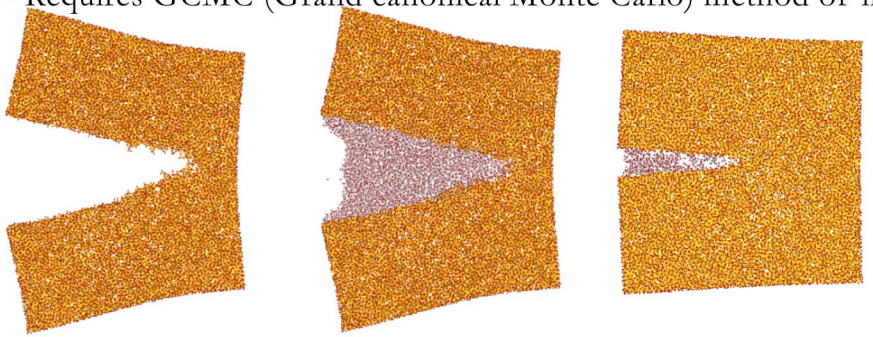
- Cracks start at the atomistic scale by the breaking of bonds at the solid-fluid interface.
- Crack tip formation & crack propagation is influenced by fluid and surface chemistry
- Isolation of chemical and mechanical effects on fracture



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

Separation of Chemical and Mechanical Impact on Fracture

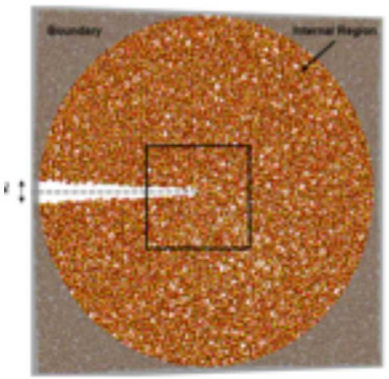
- ReaxFF inter-atomic potential used for all simulations (Fogarty et al., *J. Chem. Phys.*, 2010; Yeon and van Duin. *J. Phys. Chem. C.*, 2015)
- Created 12 configurations: 3 systems 14 x 14 x 2.8 nm^3 (38,400 atoms) annealed at 4000K for 100 ps, cooled to 300K at 5K/ps, alternating NPT/energy min simulations to achieve density of 2.187 g/cm^3. Four (4) orientations relative to crack for each system.
- Investigated three different loading conditions on each configuration to isolate chemical and mechanical effects on fracture
- Protocol: Apply initial loading (0.15 MPa$\sqrt{m}$) and relax fracture tip
  - Mechanical: increase loading (stepwise), relax for 5ps at 300K, repeat
  - Chemical: initial loading held fixed, add water molecules at low density, NVT for 500ps at 300K
  - Chemical-Mechanical: increase loading (stepwise), add in water molecules, relax for 5ps at 300K, repeat
    - Requires GCMC (Grand canonical Monte Carlo) method of inserting water into the fracture to maintain surface wetting

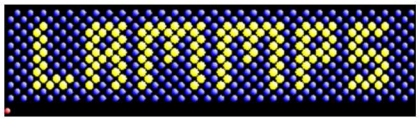


Mechanical
(mechanical loading only)

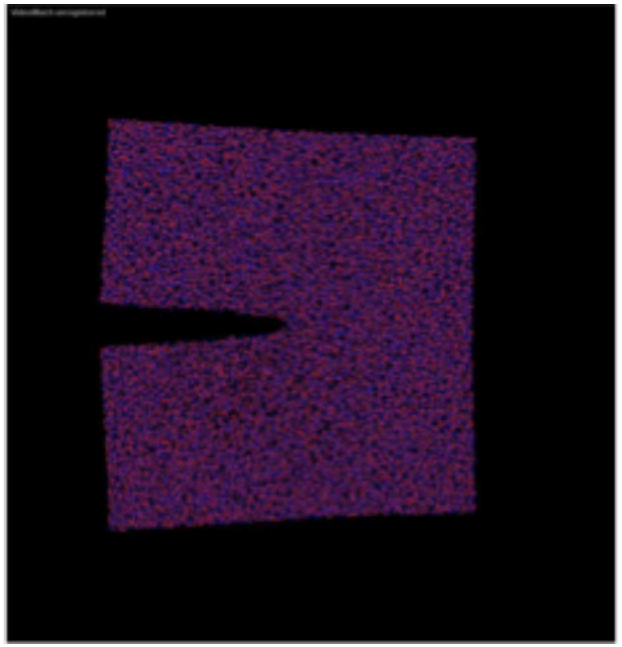
Chemical-Mechanical
(aqueous enviorment and mechanical loading)

Chemical
(aqueous enviorment only)

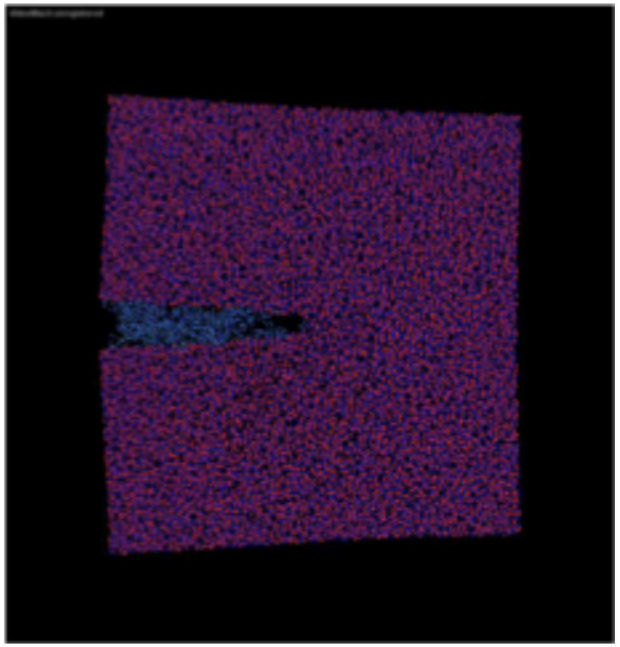




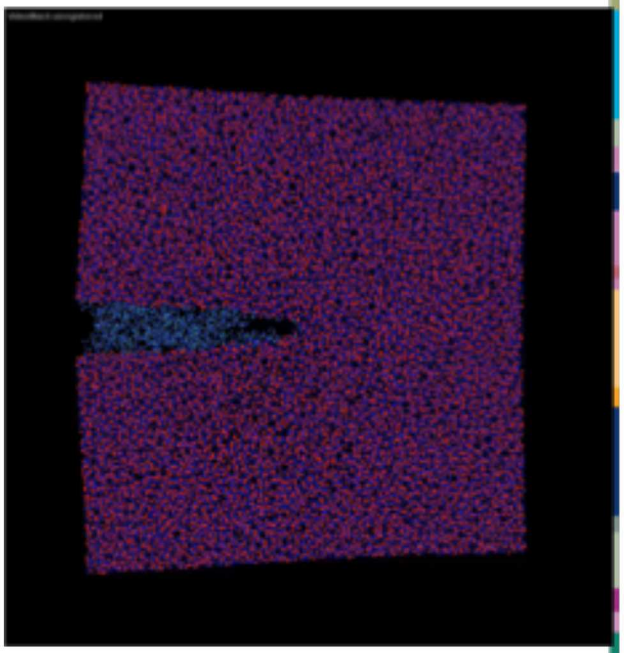
Example Simulations



Mechanical
(mechanical loading only)



Chemical
(aqueous environment)

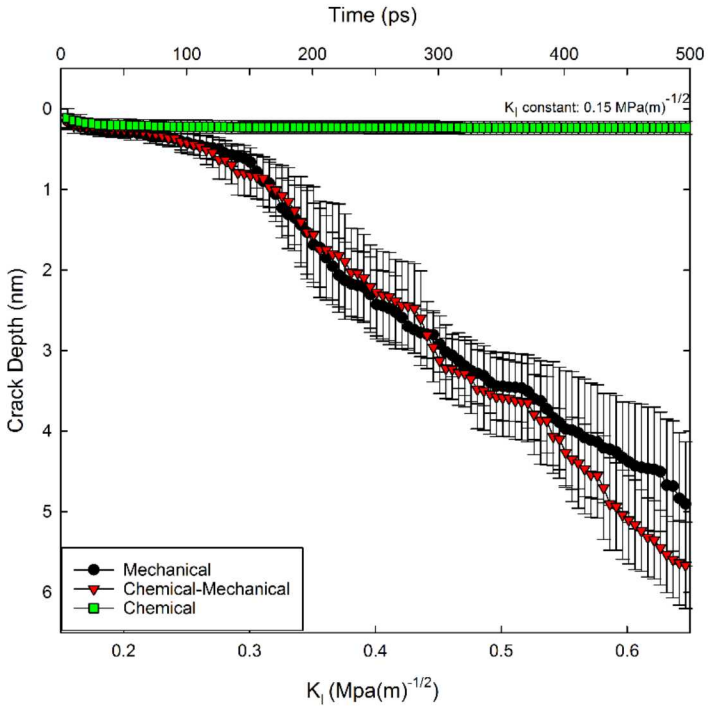


Chemical-Mechanical
(aqueous environment and mechanical loading)

Fracture Depth

- Fracture depth identifies aggregate effect of aqueous environment on fracture
- Chemical-mechanical conditions: longer fracture propagation, larger number of fracture events and slightly shorter average fracture length
- Chemical effects become more prominent as the fracture propagates
- May be altering the conditions for fracture (bond stretching, stress states etc.)
- Chemical impact is more than additive on fracture growth

Crack depth for silica systems in mechanical, chemical, and chemical-mechanical conditions.

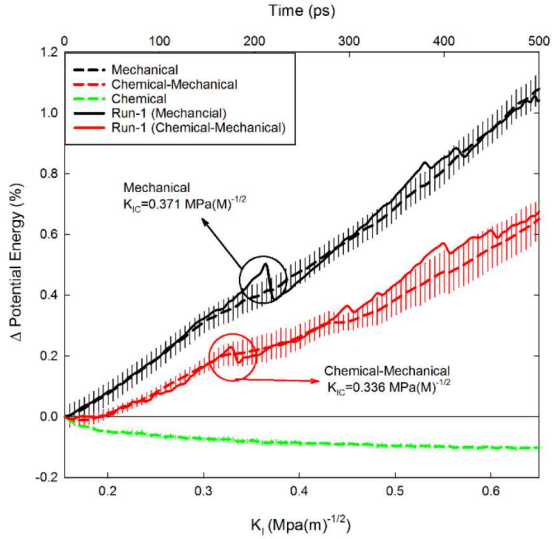


Conditions	Propagation (nm)	Fracture Events* (#)	Average Fracture Length (nm)	Longest Fracture (nm)	Fracture Velocity (m/s)
Mechanical	4.92±0.76	11.50±2.06	0.35±0.08	0.90±0.23	9.85±1.51
Chemical	0.23±0.07	0.50±0.50	0.16±0.08	0.10±0.08	0.47±0.16
Chemical-Mechanical	5.69±0.53	14.83±2.41	0.32±0.06	0.97±0.38	11.38±1.07

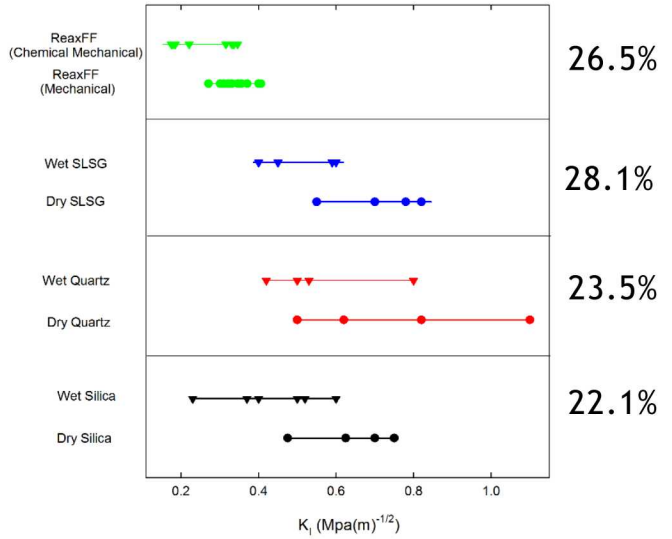
Fracture Toughness

- Identified from variation in the potential energy of the silica during loading
- Earlier fracture of silica in aqueous conditions
- No fracture in chemical-only systems (dissolution)
- K_{IC} is lower than in experimental systems (0.78 MPa√m) due to resolution and temperature effects

Mechanical: 0.339±0.037 MPa√m
Chemical-Mechanical: 0.246±0.074 MPa√M
Reduction in K_{IC}: ~26.5%



Change in potential energy for silica systems in mechanical, chemical, and chemical-mechanical conditions.



Experimental KIC data for amorphous silica, quartz , and soda-lime silicate glasses in dry and aqueous environments compared with current data.

Energy Dissipation

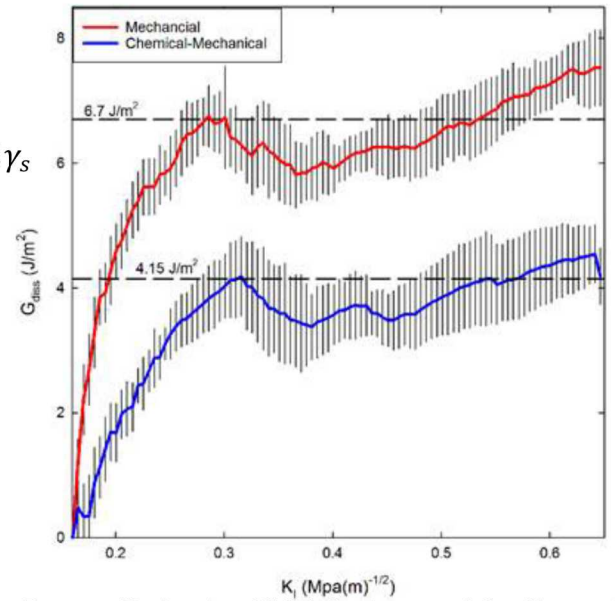
- G is related to both the surface energy and dissipative energy (unrecoverable inelastic character around the fracture tip)

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

- G_{diss} is calculated from energy and surface area of the fracture:

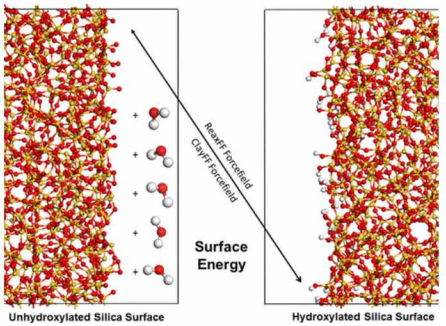
$$G_{diss} = \frac{\Delta U}{\Delta S_A}$$

- Surface energy (γ) = related to hydroxylation of the surface
- Wet fracture results in a lower K_{IC} value and lower G_{IC}, due to lower dissipation energy
- Larger G_{diss} relates to the strain distribution surrounding the fracture tip



Energy dissipation (Gdiss) during crack loading and subsequent crack propagation for silica systems

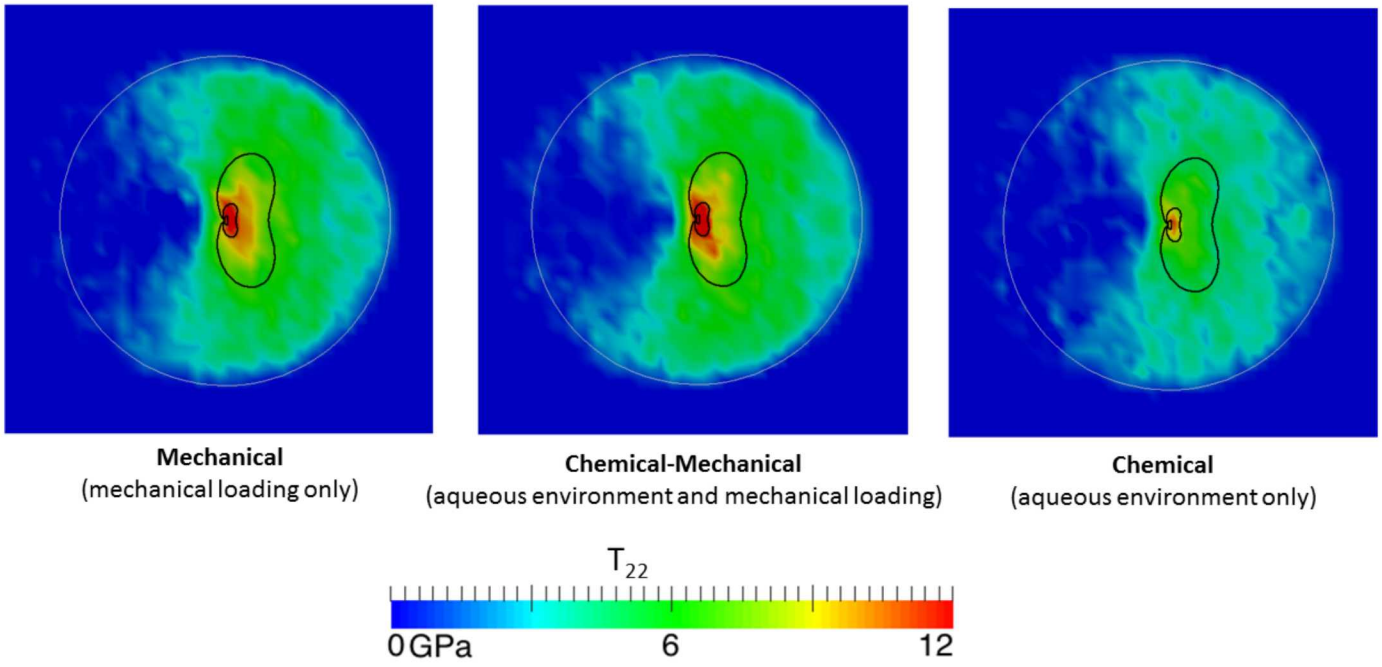
	KIC (MPa√m)	GIC (J/m²)	Gdiss (J/m²)	Si-OH (#/nm²)	γ (J/m²)
Mechanical	0.339±0.037	8.90	6.70	0.0	1.1
Chemical-Mechanical	0.246±0.074	4.95	4.15	3.1	0.4



Stress Distribution

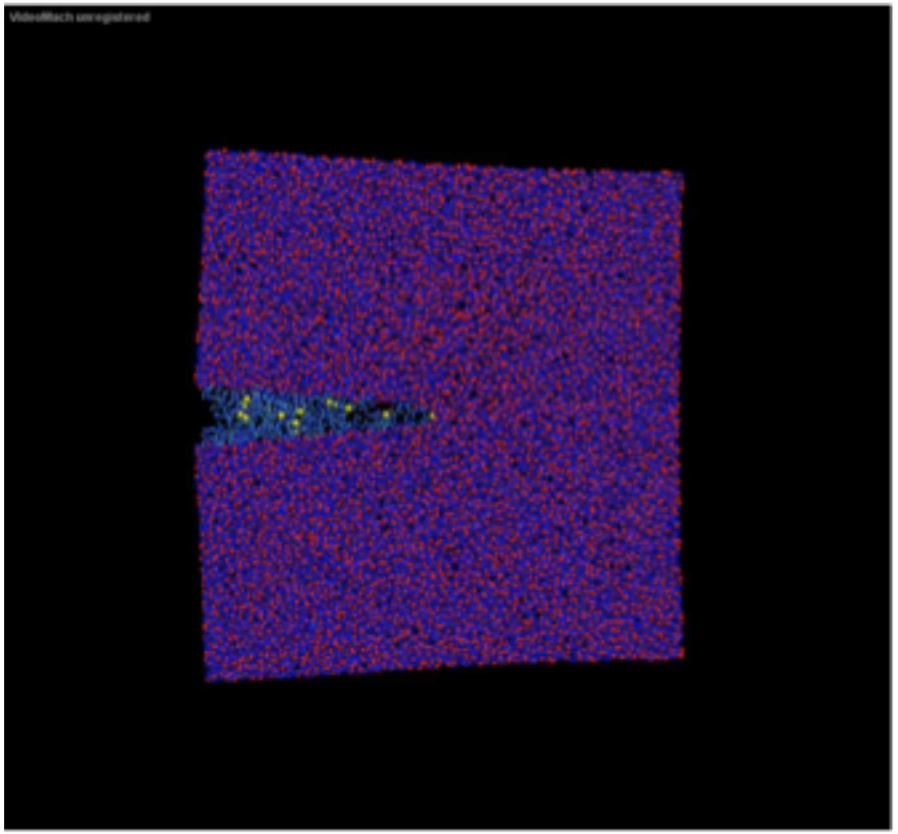
- Stresses from the atomistic simulations were coarse grained and averaged over the twelve replicates to describe the stress states surrounding the fracture tip

Stress fields for silica systems in mechanical and chemical-mechanical (K_I=0.2 MPa$\sqrt{m}$) conditions, and chemical-only condition (K_I=0.15 MPa$\sqrt{m}$).



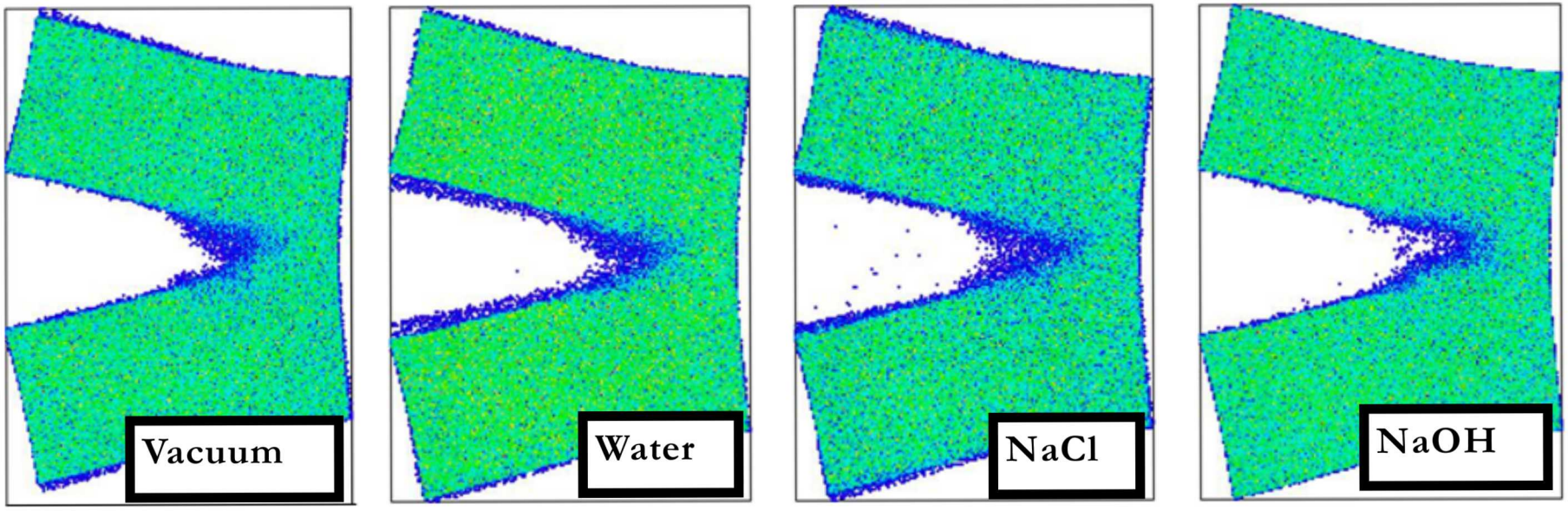
Aqueous Solutions with Chemical Complexity – Preliminary Results

- NaCl is added at the entrance of the slit crack at each step – results in a concentration of ~1M over the course of the simulation
- All other computational details remain constant
- Video: pink/purple – silica, blue – water, yellow – NaCl



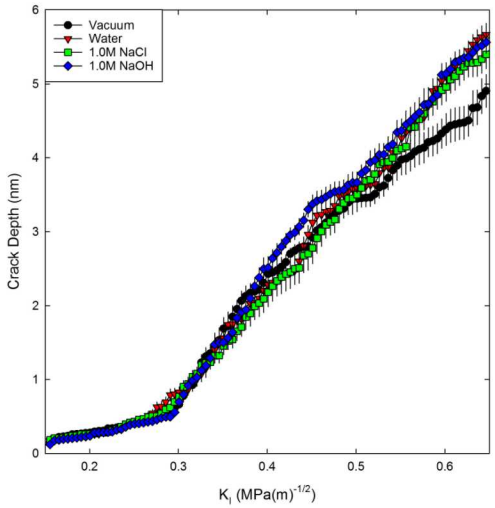
Addition of NaCl/NaOH affects Crack Tip Morphology

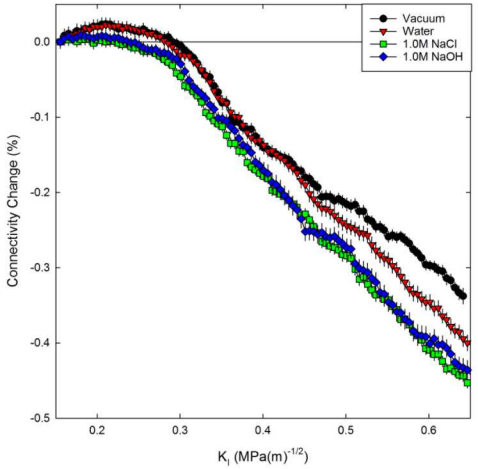
- Changes in the fracture tip morphology with water composition
- Addition of NaCl causes an increased number of dissolution events (formation of Q0)

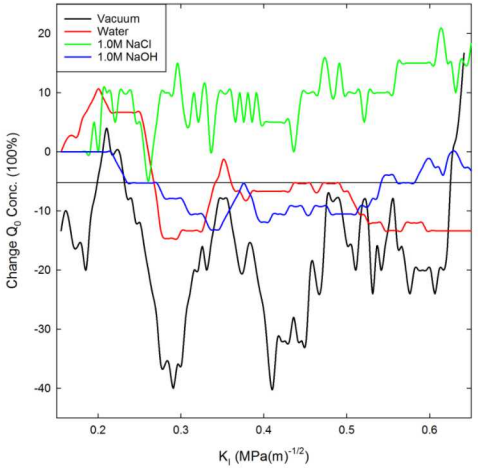


Addition of NaCl/NaOH affects Connectivity and Dissolution Events

- Water/NaCl/NaOH solutions show the same crack depth with vacuum conditions being shorter
- Connectivity within wet/vacuum systems overlap until $K_I = 0.45$ MPa$\sqrt{m}$ then diverge; NaCl/NaOH systems are separate (but overlap heavily)
- Addition of NaCl causes an increased number of dissolution events (formation of Q0)
- Q1/Q0 has NaCl as the strongest effect on connectivity – causing "dissolution" events
- Connectivity for NaCl/NaOH is similar, possibility of different types of mechanisms?



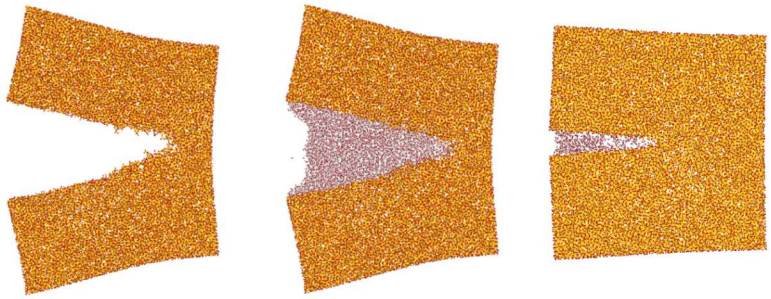




Conclusions

Atomistic simulations of silica fracture in aqueous environments were used to isolate the chemical and mechanical effects of fracture

- Chemical-mechanical systems exhibited increased fracture growth due to higher number of fracture events (and possibly lower threshold for fracture)
- Fracture toughness was decreased by ~25% between vacuum and water conditions, consistent with reported experimental data
- G_{IC} (strain energy release rate) was decreased by ~50% in chemical-mechanical systems due to decreased dissipation energy and surface energy
- Stress fields indicate relaxation of the process zone surrounding the fracture in aqueous conditions, suggesting that the strain effects are even more localized at the fracture tip
- Thresholds for fracture may be decreased in the presence of water, even on extremely local distance and time scales, and chemical effects are not additive



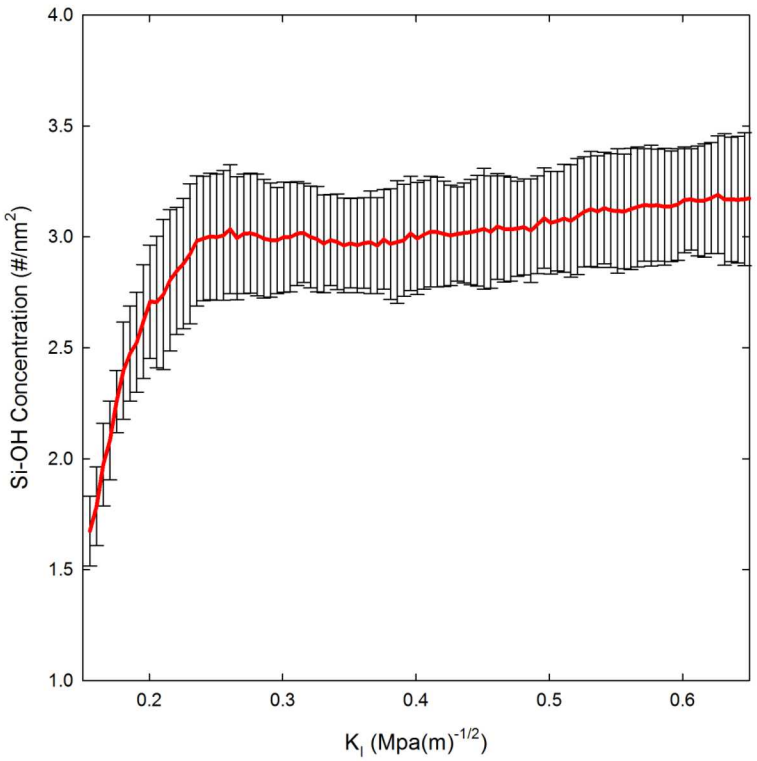
Acknowledgements

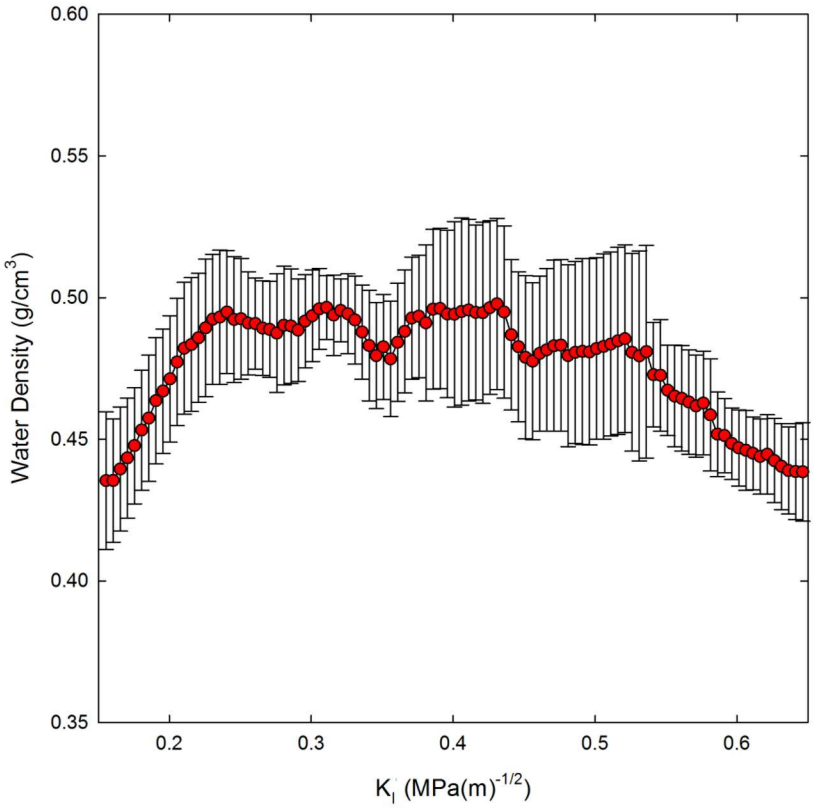
This work was fully supported by the Laboratory Directed Research and Development (LDRD) program of Sandia National Laboratories. Special thanks to Mark Wilson (SNL) for LAMMPS support.

Publications

- J.M. Rimsza, R.E. Jones and L.J. Criscenti, "Surface Structure and Stability of Partially Hydroxylated Silica Surfaces", *Langmuir*, **33**, pp. 3882-3891, 2017.
- J.M. Rimsza, R.E. Jones and L.J. Criscenti, "Crack propagation in silica from reactive classical molecular dynamics simulations", *Journal of the American Ceramic Society*, **101**, pp. 1488-1499, 2018.
- J.M. Rimsza, R.E. Jones and L.J. Criscenti, "Interaction of NaOH solutions with silica surfaces", *Journal of Colloid and Interface Science*, **516**, pp. 128-137, 2018.
- R.E. Jones, J.M. Rimsza and L.J. Criscenti, "An atomic-scale evaluation of the fracture toughness of silica glass", *Journal of Physics: Condensed Matter*, **30**, p. 245901, 2018.
- J.M. Rimsza, R.E. Jones and L.J. Criscenti, "Chemical effects on subcritical fracture in silica from molecular dynamics simulations", In Review, 2018.

Back-Up Data


Si-OH Concentration (#/nm^2)
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.2
0.3
0.4
0.5
0.6
K_I (Mpa(m)$^{-1/2}$)


Water Density (g/cm^3)
0.60
0.55
0.50
0.45
0.40
0.35
0.2
0.3
0.4
0.5
0.6
K_I (MPa(m)$^{-1/2}$)