Assessing the fracture strength of geological and related materials via an atomistically based J-integral

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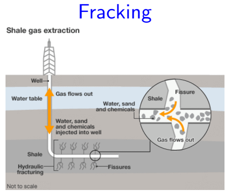
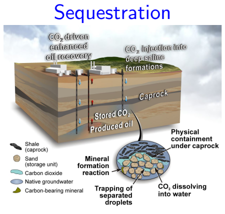
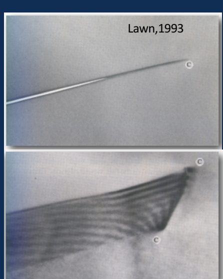
Fracking



Sequestration



large-scale problems with mechanical and chemical forces acting on cracks



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Objectives

- Develop a fundamental understanding of the **chemo-mechanical processes** that control subcritical cracks in low-permeability geomaterials.
- Link atomic-scale insight (reaction mechanisms, diffusion, sterics) to macroscale observables (rate of crack propagation, fracture toughness)

Method

Simulation:

- Classical molecular statics/dynamics were used to obtain an atomic level evaluation of stress and energies
- Several forcefields for silica, both non-reacting (Tersoff, ClayFF) and reacting (ReaxFF), were evaluated (elastic constants, surface energies)

Slit crack:

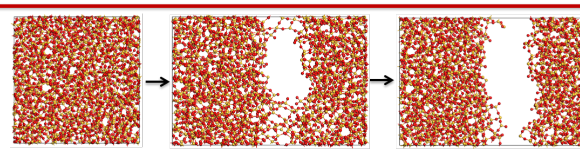
- Bonds along a half plane are severed *a priori*
- The singularity at the crack tip that is mollified by the local atomic relaxation
- The cracks is opened in mode I fracture by adjusting boundary atom positions and relaxing the **interior region** **(see figure below)**

Coarse-graining method:

- A continuum solution to the elastic crack is applied as a far-field displacement boundary condition on an **annulus** of atoms.
- Stress, displacement, and energy density fields are coarse-grained on a **grid**.
- Eshelby stress field is formed and the J-integral is evaluated on the **contour**.

Forcefield Benchmarking

- Multiple classical forcefields have been parametrized for Si/O/H systems (Tersoff, ReaxFF, ClayFF etc.)
- Features of the forcefield determine how silica will fracture, e.g. the forcefields predict different elastic moduli & surface energies, as well as surface and bulk structures
- We are developing a formalism to add chemistry to the J-integral to evaluate fracture toughness in wet and reacting environments and using bulk and flat surface configurations to obtain expectations

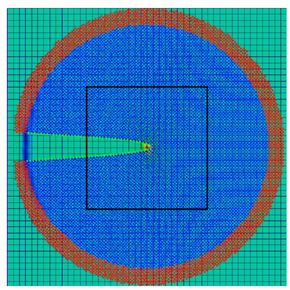


ε=0 Dense Silica → ε~0.35 Void nucleation and fracture initiation → ε~0.4 Fracture

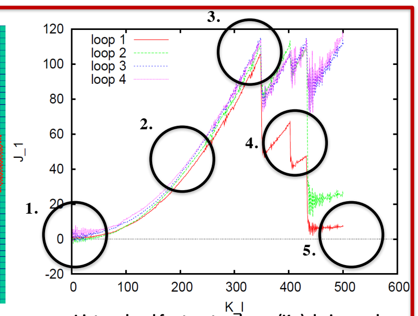
ClayFF Elastic Constants	ReaxFF Elastic Constants	Experimental Elastic Constants
E=108.5±0.6 GPa	E=76.1±0.3 GPa	E=73.1 GPa
K=65.6 ±0.5 GPa	K=55.2 ±5.2 GPa	K=36.7 GPa
G=47.1±0.9 GPa	G=21.8±0.8 GPa	G=31.2 GPa
v=0.21±0.02	v=0.32±0.02	v=0.17

Fracture of Dry Silica

- Crack opening by adjustment of boundary atoms and subsequent relaxation of internal silica allowed for crack propagation (system size ~25,000 atoms)
- Significant strain needs to develop at the crack tip (point 1 and 2) in order to allow for Si-O bond breakage and propagation of the crack tip
- Silica fracture occurs in steps, with an initial high J-integral value indicating the theoretical fracture toughness for amorphous silica (point 3)
- Additional fracture events (point 4 and 5) occur at lower J-integral values

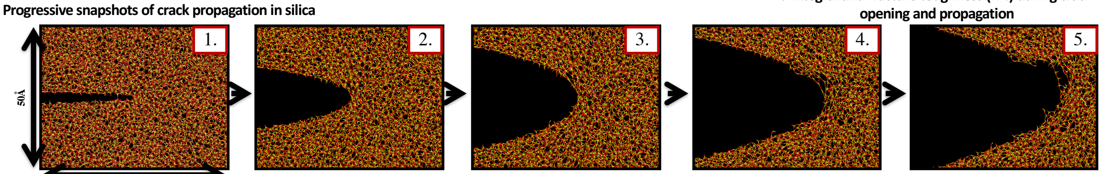


Energy field of silica slit crack



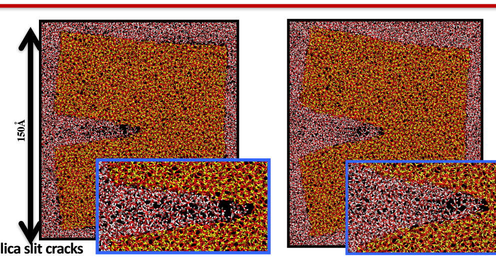
J-integral and fracture toughness (K_{Ic}) during crack opening and propagation

Progressive snapshots of crack propagation in silica



Fracture of Silica infiltrated with water

- There are kinetic bottlenecks for observing water infiltration over feasible computational time scales.
- High temperature and pressures accelerate water infiltration
- We used staggered process: (1) open the crack and allow the system to relax, (2) run dynamics to allow for water diffusion (~1ns)
- Water infiltrates a slit crack while hydroxylating the silica fracture surfaces
- Shown: Tersoff Force Field + TIP3P Water (system size ~60,000 atoms).



Snapshots of water infiltration of silica slit cracks

Summary

- We used atomically perfect slit cracks in bulk silica for comparison with experimental measurements of silica fracture toughness
- Via upscaling of atomistic stress and strain to calculate the J-integral we were able to connect atomistic data to continuum theory and estimates.
- Water introduced into the crack adheres to the crack faces but does not completely infiltrate the crack tip and has substantial effects on the J-integral

Current Work

- Test chemo-mechanical J-integral with wet, reacting cracks
- Add salts to water and investigate how dissolved cations alter the fracture surface and impacts fracture toughness

References

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