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Atomistic-scale evaluation of the fracture toughness of silica

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Objectives and Introduction

- Develop a fundamental, atomistic-level understanding of the chemical-mechanical processes that control subcritical cracks in low-permeability geomaterials.
- Link atomic-scale insight (reaction mechanisms, diffusion) to macroscale observables (rate of crack propagation, fracture toughness)
- Address how chemical environment affects mechanical behavior.

Computational Details

Simulation methods:

- Classical molecular dynamics were implemented for atomic level evaluation of stress and energies
- Several forcefields (Tersoff, ClayFF, ReaxFF) were benchmarked for evaluation of elastic constants in silica

Slit cracks:

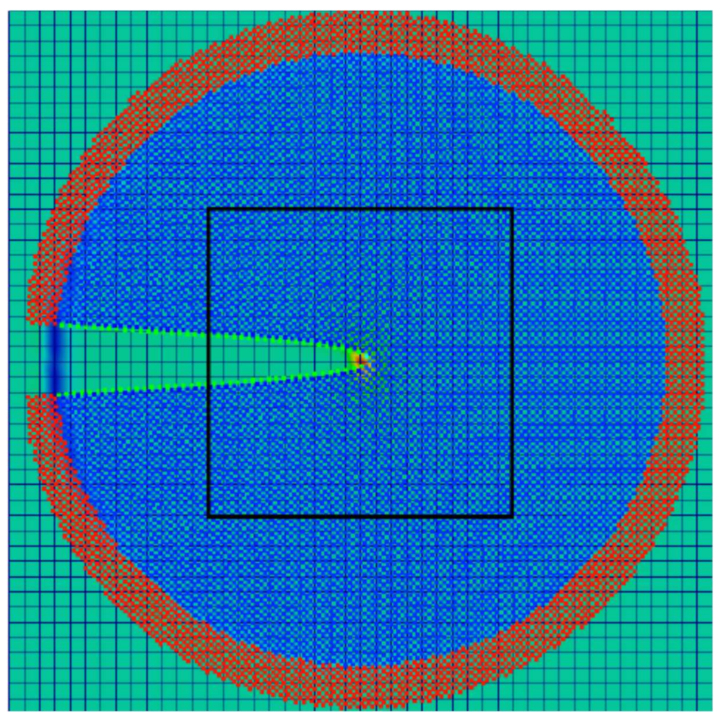
- Develops a singularity at the crack tip for evaluation of the J-integral
- Inserted by breaking bonds along a half plane of atoms
- Cracks are opened in mode I fracture by adjusting boundary atom position and relaxing the interior region

Atomistic-to-Continuum method:

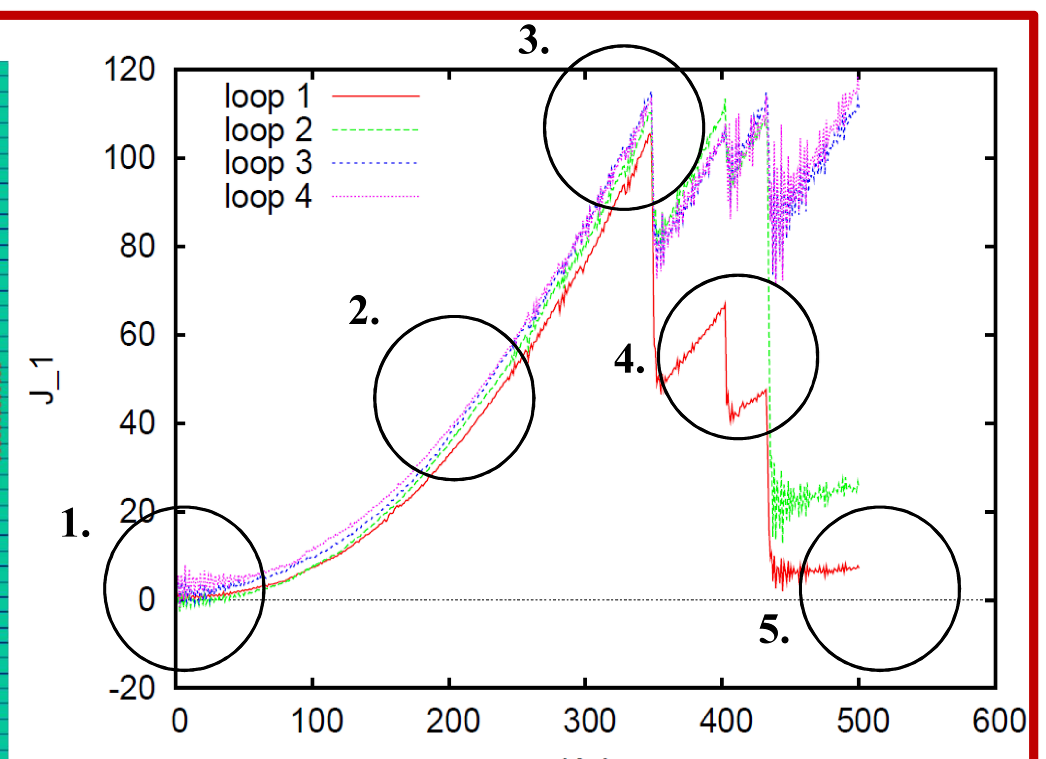
- A far-field continuum displacement boundary is applied on an annulus of atoms.
- Stress, displacement, and energy density fields are coarse-grained on a grid.
- Eshelby stress is formed and the J-integral is evaluated on the contour.

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 movement of the crack tip
- Silica fracture occurs in steps, with an initial high J-integral value for amorphous silica (point 3)
- Additional fracture events (point 4 and 5) occur at lower J-integral values

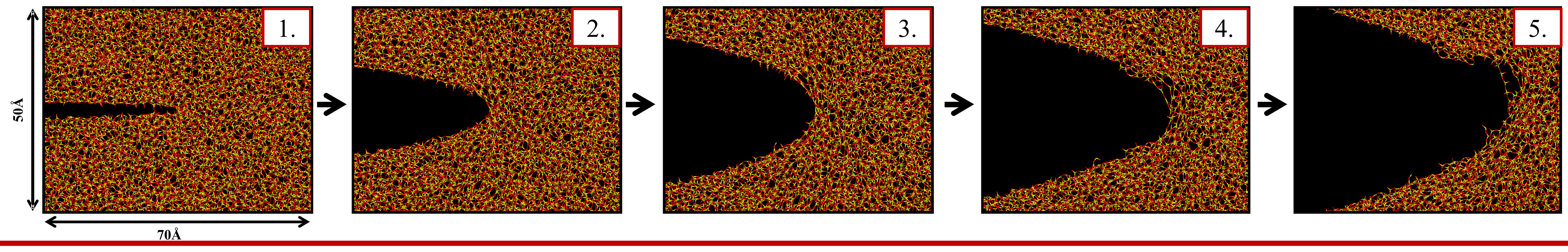


Energy field of silica slit crack



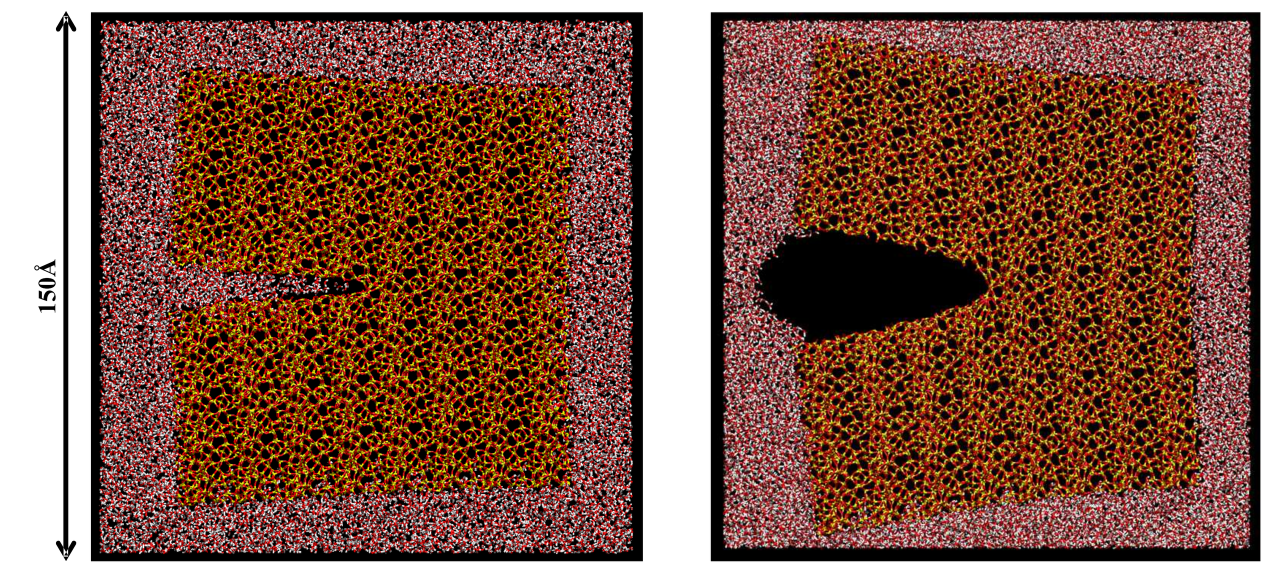
J-integral and fracture toughness (K_{1C}) during crack opening and propagation

Progressive snapshots of crack propagation in silica



Amorphous Silica and Water

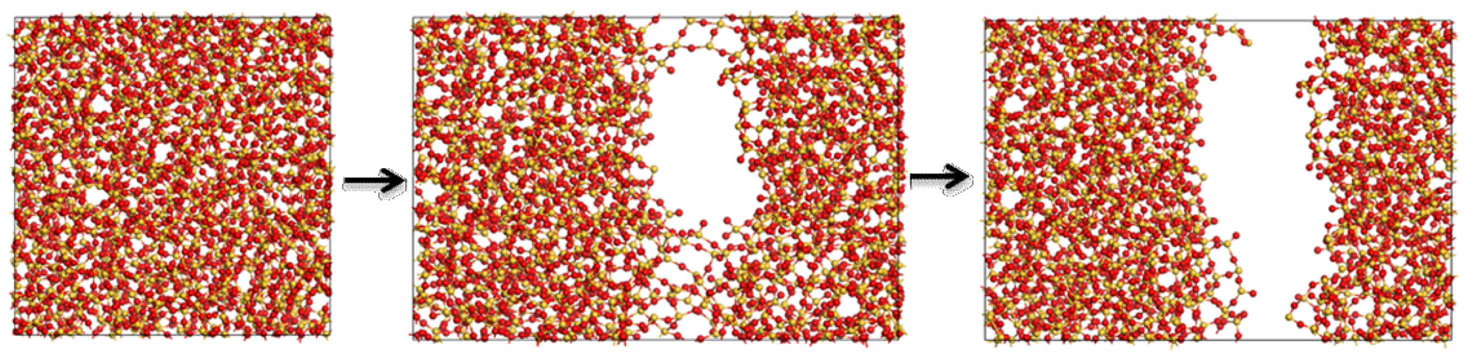
- Water should infiltrate a slit crack while hydroxylating the silica fracture surfaces
- A coupled process is implemented by opening the crack a set amount and allowing the system to relax for an extended period of simulation time (1 ns), resulting in water infiltration.
- Kinetic or mechanistic limitations exist for water infiltration at computational time frames.
- High temperature and pressures are required to force water infiltration at efficient time scales.
- Examples here are with Tersoff Force Field + TIP3P Water (system size ~60,000 atoms).



Snapshots of water infiltration of silica slit cracks

Silica Forcefield Benchmarking

- Multiple classical forcefields have been parametrized for Si/O/H systems (Tersoff, ReaxFF, ClayFF etc.)
- Features of the forcefield describe how silica will fracture in dry and wet conditions
- Enthalpy of reactions, velocities, stress, strain, surface energies etc., are used to develop algorithm to add chemistry to the J-integral for evolution of fracture toughness
- Elastic constants, surface energy values, and structural parameters were evaluated for three different forcefields



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

Conclusions

- Slit cracks have been introduced into bulk silica for investigation of silica fracture toughness
- Upscaling of atomistic stress and strain through the atomistic-to-continuum method has been used to calculate the J-integral
- Introduction of water into the slit crack occurred through coupled relaxation of the silica for water diffusion into the crack space
- Benchmarking of silica forcefields was also performed on three forcefields (Tersoff, ReaxFF, ClayFF) for integration with J-integral calculation

Future Work

- Develop computational approach to use additional forcefields (ReaxFF, ClayFF) to generate cracks with water
- Add salts to water and investigate how dissolved cations alter the fracture surface and impacts fracture toughness

References

[1] Reese E Jones et al. (2016) Multiscale Materials Modeling for Nanomechanics. 223-259 [2] Yeon, J. and. Adri CT van Duin. (2015) The Journal of Physical Chemistry C 120.1 305-317. [3] Cygan, Randall T et al. (2004) The Journal of Physical Chemistry B 108.4 1255-1266. [4] Munetoh, Shinji, et al. (2007) Computational Materials Science 39.2 334-339.

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