TMS 2017, 146th Annual Meeting, San Diego, 26 Feb-2 Mar 2017

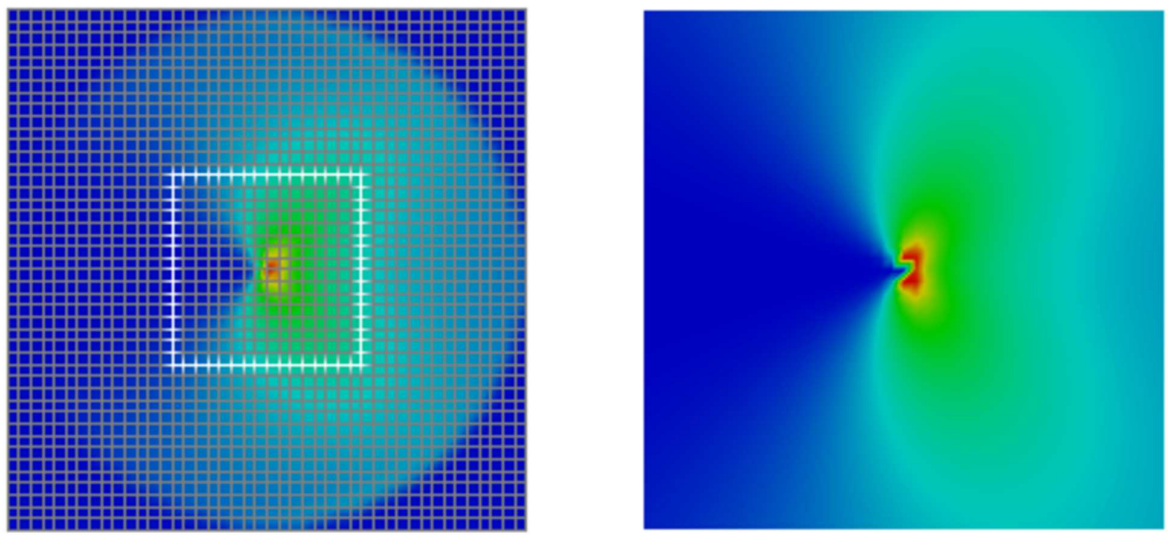
Environmentally Assisted Cracking: Theory and Practice Stress Corrosion Cracking II, Tuesday 4:00pm

SAND2017-2575C

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

Reese Jones, Louise Criscenti and Jessica Rimsza

Sandia National Laboratories



atomistic vs. continuum

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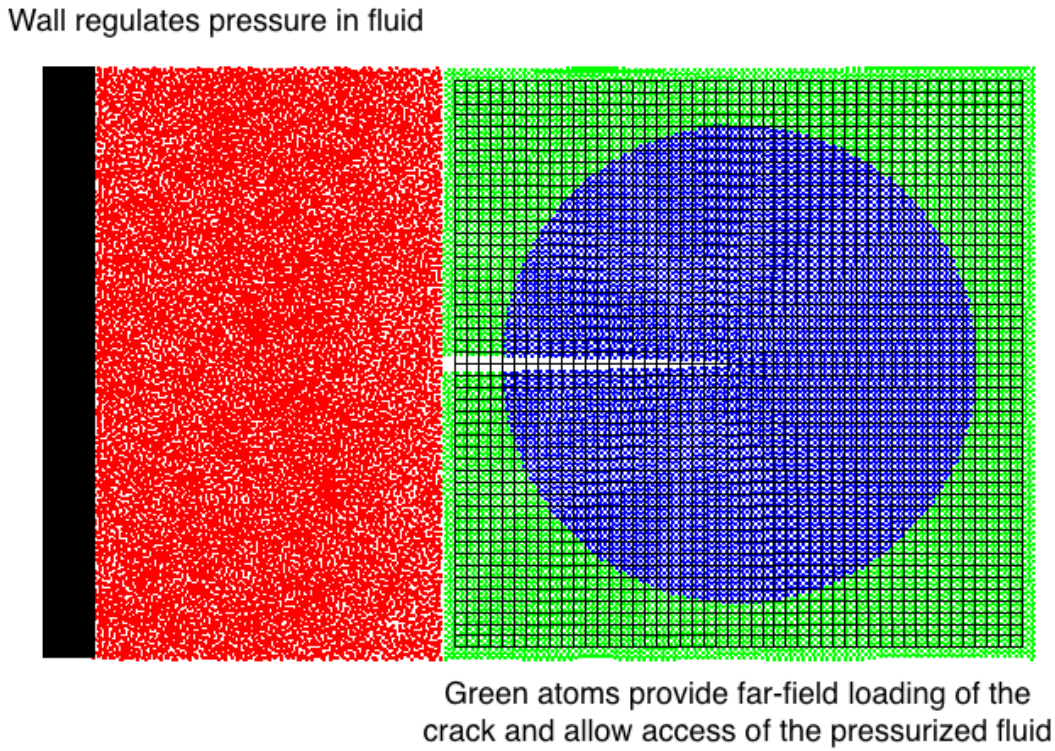
Outline

- Introduction
  - Motivation
  - History
- Theory
  - Configurational forces
  - Coarse-graining
- Results
  - Consistency
  - Basic Lennard-Jones
  - Structured Tersoff
  - Reacting ReaxFF
- Conclusion



please ask questions as we go

Technological motivation

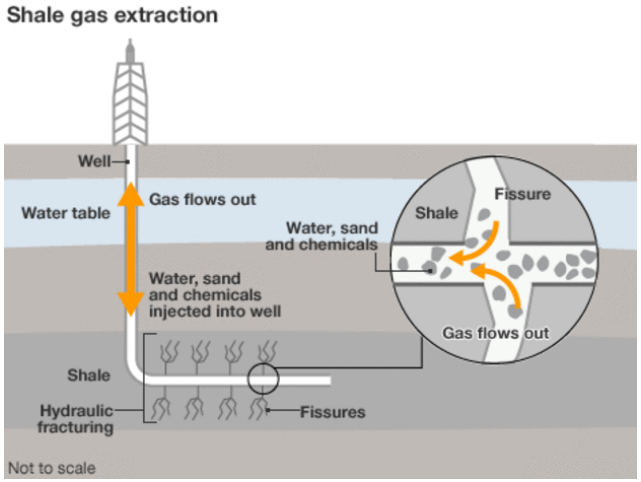
Goals:

- Develop fundamental understanding of the *chemical-mechanical* mechanisms that **control subcritical cracks** in low-permeability geomaterials
- Link atomic-scale insight to macroscale observables and directly address how chemical environment affects mechanical behavior.

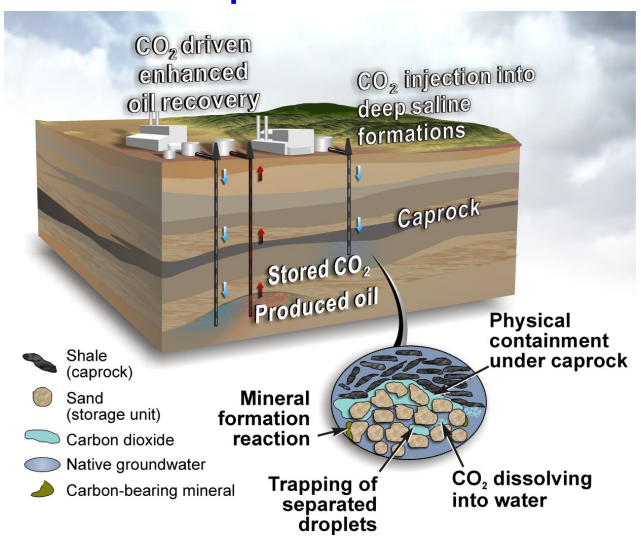
Why Atomistic Simulation?

- Cracks start at the *atomistic scale* by the *breaking of bonds* at the rock-fluid interface.
- Crack tip formation & crack propagation is influenced by fluid and surface *chemistry* (e.g., development of surface charge & adsorbed species along fracture surface).

Fracking



Sequestration



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

Approach

Generally speaking, this work is based on the **simple premise**:

- *consistent* fields from atomistic simulation
- application of continuum theory

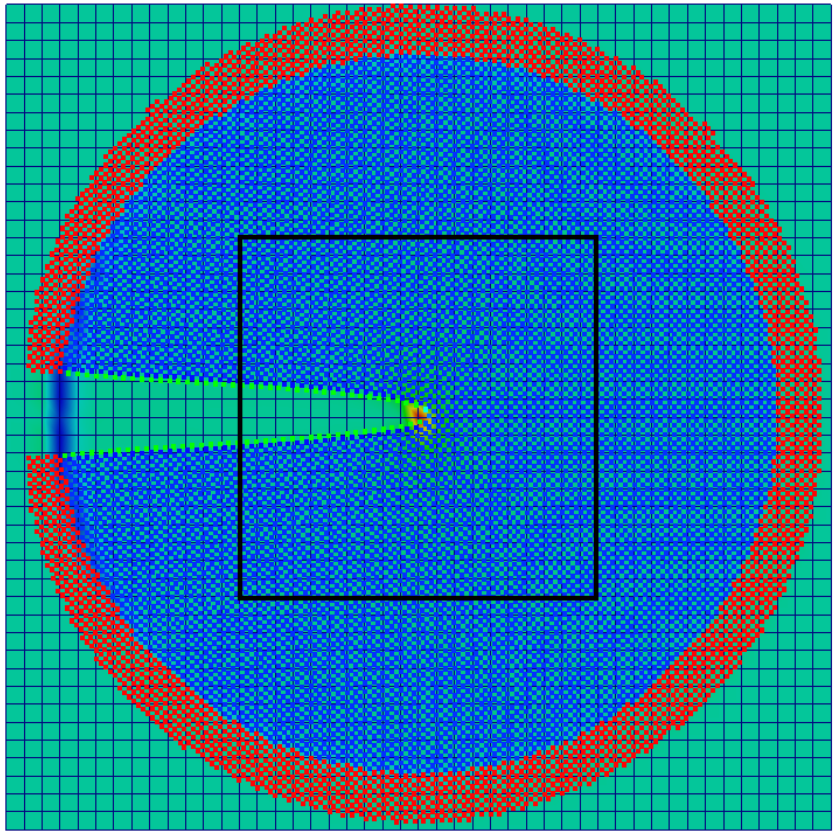
$\Big\} \rightarrow$ nanoscale results

i.e. first, connect atomistic data to continuum fields in a manner and a **scale** consistent with the conservation balances; then, use continuum theory to analyze the process.

There are many technologically relevant examples of the validity of this approach down to the length-scale of nanometers and hundreds of atoms.

In particular, atomistic simulation is particularly suited to the analysis of configurational forces since they typically have atomic details.

Method: 1-way coupling



1. A slit is inserted through bonds crossing a half plane
2. A far-field continuum displacement solution is applied on an annulus of atoms (red)
3. stress, displacement, and energy density fields are coarse-grained on a grid (green)
4. Eshelby stress is formed and the J-integral is evaluated on the contour (**black**)

Ingredients

- atomistics provides a first-principles description of the crack surface and other defects structure and energetics
- continuum provides far-field surrogate model and theory for interpretation

Background

(1) **Connection between particle & continuum mechanics**:

- [Irving&Kirkwood 1950] **Correspondence of particle trajectories & continuum fields through the balance laws**
- [Hardy 1982] Extension of I&K to smooth kernels
- [Zimmerman 2010] Referential/material frame extension well-suited to solids

(2) **Theory of configurational forces**:

- [Eshelby 1951] **Foundations of configurational forces**
- [Rice 1968] Application to fracture
- [Wu 2001] Application to diffusion

(3) **Application to atomic systems**:

- [Jones 2010] Path-independent, $T = 0$ atomistic **J**-integral
- [Jones 2010] Finite T, quasistatic **J**-integral
- [Zimmerman 2013] Application to ductile metals

Eshelbian mechanics: J integral

The energy release rate *relative* to a process with motion $\boldsymbol{\chi}_t$, $\mathbf{F} = \partial_{\mathbf{X}} \boldsymbol{\chi}_t$, and a *fixed reference configuration* $\mathbf{X}$ is:

$$\dot{\Pi}(\boldsymbol{\varphi}_t, \boldsymbol{\chi}_t, \mathbf{F}) - \dot{\Pi}(\mathbf{X}, \boldsymbol{\chi}_t, \mathbf{F}) = \int \partial_{\mathbf{X}} \Psi \cdot \dot{\boldsymbol{\varphi}}_t \, dV$$

where $\boldsymbol{\varphi}_t(\mathbf{X})$ is a map describing the *configurational change* in the reference configuration, and

$$\partial_{\mathbf{X}} \Psi = \boldsymbol{\nabla}_{\mathbf{X}} \Psi(\mathbf{X}, \mathbf{F}(\mathbf{X})) - \partial_{\mathbf{X}} W(\mathbf{X}, \mathbf{F}) : \boldsymbol{\nabla}_{\mathbf{X}} \mathbf{F} = \boldsymbol{\nabla}_{\mathbf{X}} \cdot \mathcal{S}$$

is *work conjugate* to the configurational change $\boldsymbol{\varphi}_t$.

$\mathcal{S}$ is the Eshelby stress and the **J**-integral is the resultant force

$$\mathbf{J} = \int_{\partial\Omega} \mathcal{S}\mathbf{N} \, dA = \partial_{\mathbf{X}} \Pi$$

conjugate to the motion of the crack tip $\dot{\boldsymbol{\varphi}}_t$.

Eshelbian mechanics: Eshelby stress

The Eshelby stress can be defined explicitly as

$$\mathcal{S} = \Psi \mathbf{I} - \mathbf{F}^T \mathbf{P}$$

where

- Ψ is the (Helmholtz) free energy,
- $\mathbf{F} = \nabla_{\mathbf{X}} \chi$ is the deformation gradient,
- $\mathbf{P}$ is the 1st Piola-Kirchhoff stress

notice these are an **energy-conjugate triplet**.

Each of these fields must be estimated **accurately** and **consistently** from atomic data in order to obtain a **reliable** estimate of **fracture toughness** through the **J**-integral

Consistent fields

We adapt the Irving-Kirkwood/Hardy formalism:

- **energy density**:

$$W(\mathbf{X},t) = \sum_{\alpha} \phi_\alpha(t)\,\psi(\mathbf{X}-\mathbf{X}_\alpha) - \overbrace{\sum_{\alpha} \phi_\alpha^X \psi(\mathbf{X}-\mathbf{X}_\alpha)}^{W_0(\mathbf{X})}$$

where $\phi_\alpha^X = \phi_\alpha(\{\mathbf{X}_\beta\})$ is PE in a *fixed reference configuration* $\{\mathbf{X}^\beta\}$ (perfect lattice w/ bonds deleted along crack).

- **displacement gradient** :

$$\mathbf{H} = \nabla_{\mathbf{X}}\mathbf{u} = \sum_I \mathbf{u}_I(t)\nabla_{\mathbf{X}} N_I(\mathbf{X}) \ .$$

using *interpolation* $\mathbf{u} = \sum_I \mathbf{u}(\mathbf{X}_I,t)N_I(\mathbf{X})$ of displacement

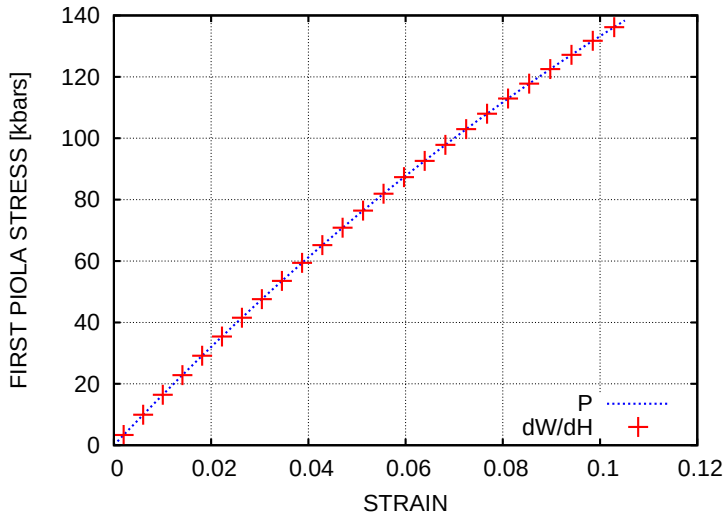
$$\mathbf{u}(\mathbf{X},t) = \frac{\sum_\alpha (\mathbf{x}_\alpha(t)-\mathbf{X}_\alpha)\, m_\alpha \psi(\mathbf{X}_\alpha - \mathbf{X})}{\sum_\alpha m_\alpha \psi(\mathbf{X}_\alpha - \mathbf{X})}$$

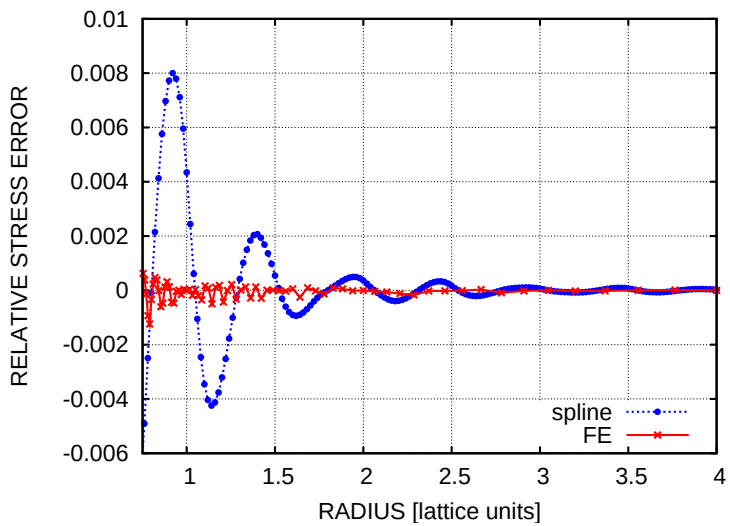
defined *mass-weighted* consistent w/ momentum density.

- **stress** via the I-K procedure:

$$\mathbf{P}(\mathbf{X},t) = -\sum_{\alpha<\beta} \mathbf{f}_{\alpha\beta}(t) \otimes \mathbf{X}_{\alpha\beta} B_{\alpha\beta}(\mathbf{X})$$

Results: Consistency and Accuracy





The coarse-grained fields satisfy

$$\mathbf{P} = \boldsymbol{\nabla}_{\mathbf{F}} \Psi$$

i.e. there is (thermodynamic) **consistency** between the energy, stress and deformation measures ($\mathbf{P}$ –, $\boldsymbol{\nabla}_{\mathbf{F}} \Psi$ +)

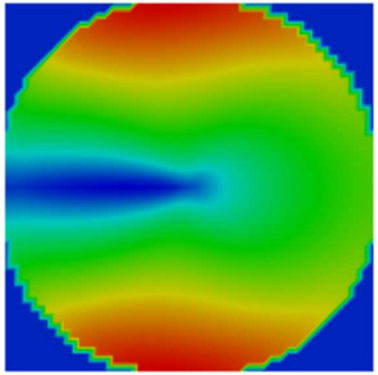
Also our use of *partition of unity*/FE kernels

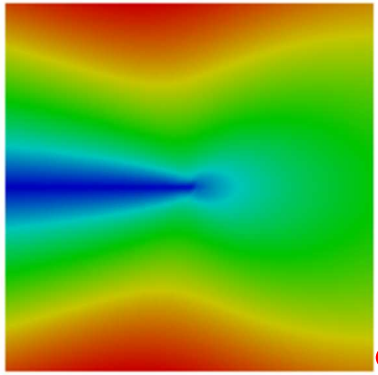
$$\psi_\alpha(\mathbf{X}_I) = N_I(\mathbf{x}_\alpha)$$

(and interpolation) can lead to faster and more *accurate* estimates than with traditional kernels.

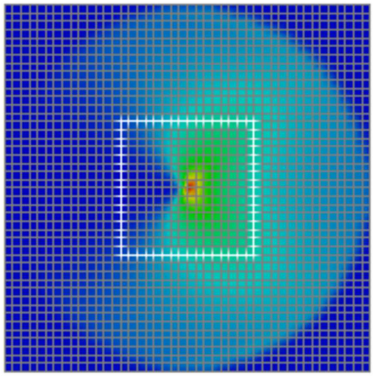
Idealized crack with displacement boundary conditions

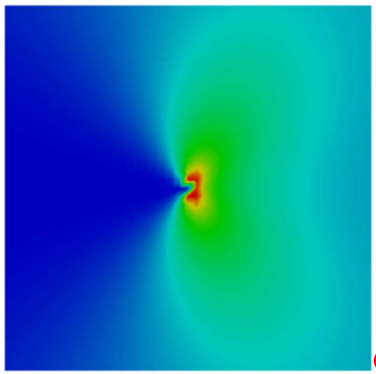
IK/Hardy vs. LEFM





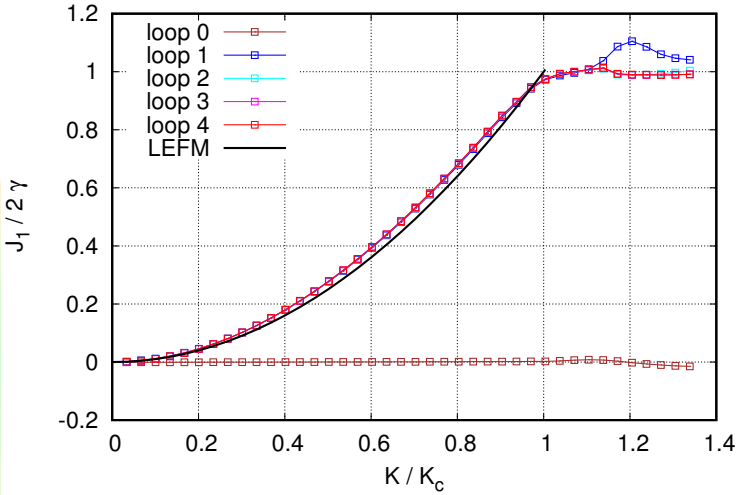
displacement



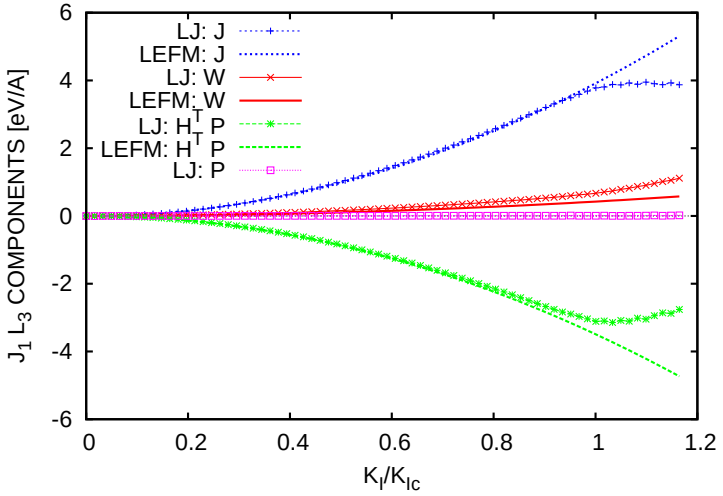


stress

- path independence

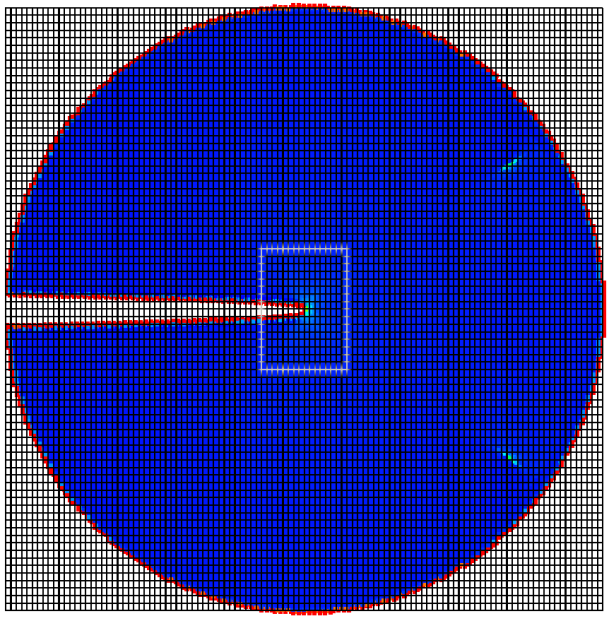


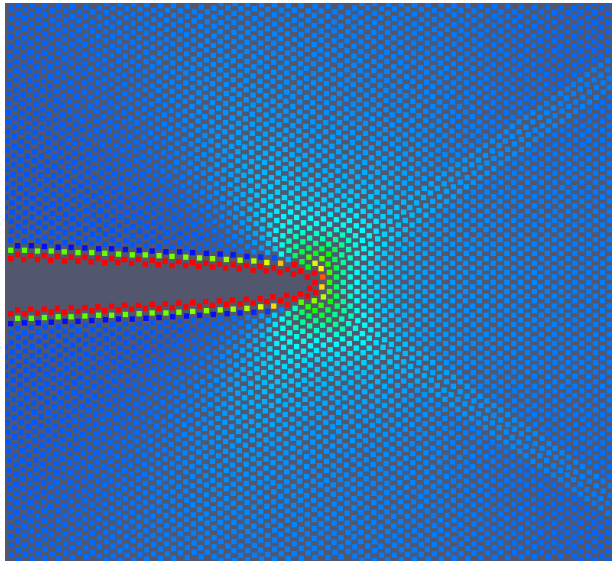
- expected limit: 2γ at $K = K_c$

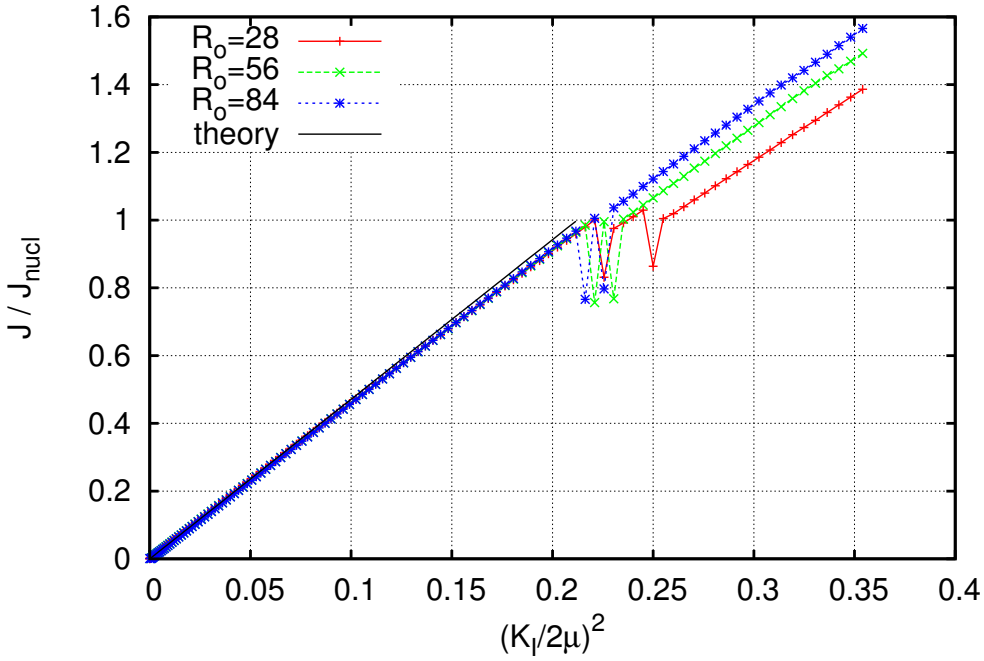


- components W & $\mathbf{H}^T\mathbf{P}$ match theory

Applications







The method has been applied to:

- ductile fracture
- finite temperature effects
- shear modes
- amorphous materials

Mechanochemistry of fracture

The combination of mechanical & chemical "forces" result in a configurational force driving crack forward. Traditionally mechanical effects are accounted for on **J** side, and the chemical effects on the K side

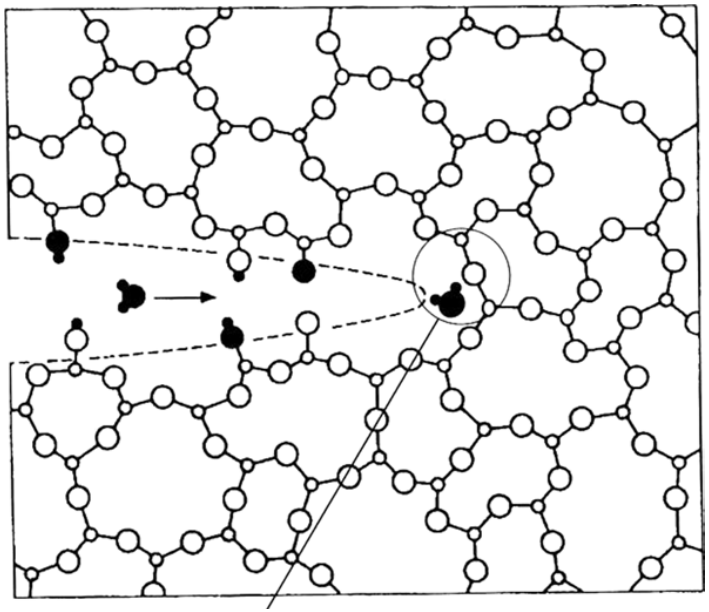
$$\partial_{\mathbf{X}}\Pi \cdot a\mathbf{E}_1 \equiv \mathbf{J}_{\text{crit}}(a) \cdot \mathbf{E}_1 = \frac{K_{\text{crit}}^2}{E^*} a = 2\gamma a$$

since Π is the free energy of the **solid**, and *fluid* effects, e.g. changes of surface energy, are consider external/surface effects.

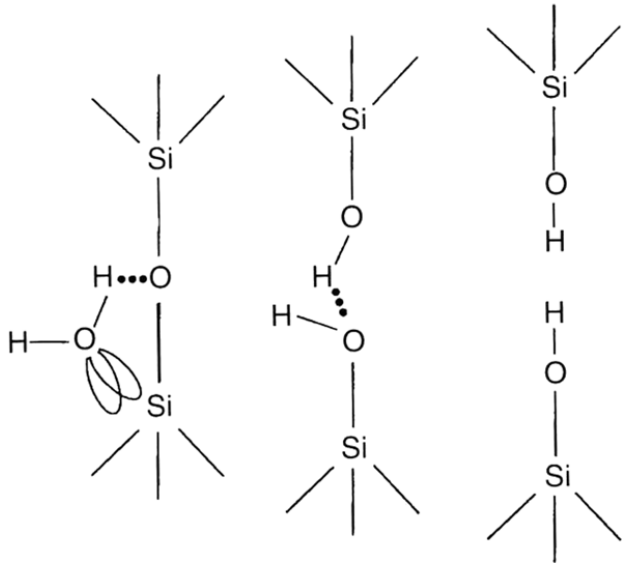
A molecular level model captures:

- crack advance discrete jumps due to atomic structure
- **surface energy** : reconstruction, reaction/binding to ambient species, [Orowan 1944]
  (crack different environment than flat, relaxed surface)
- access to tip & **steric** effects: e.g. cusp/sharp vs parabolic/blunt crack, driving pressure, temperature
- **time-scales** & sequencing: breaking/forming bonds, diffusion

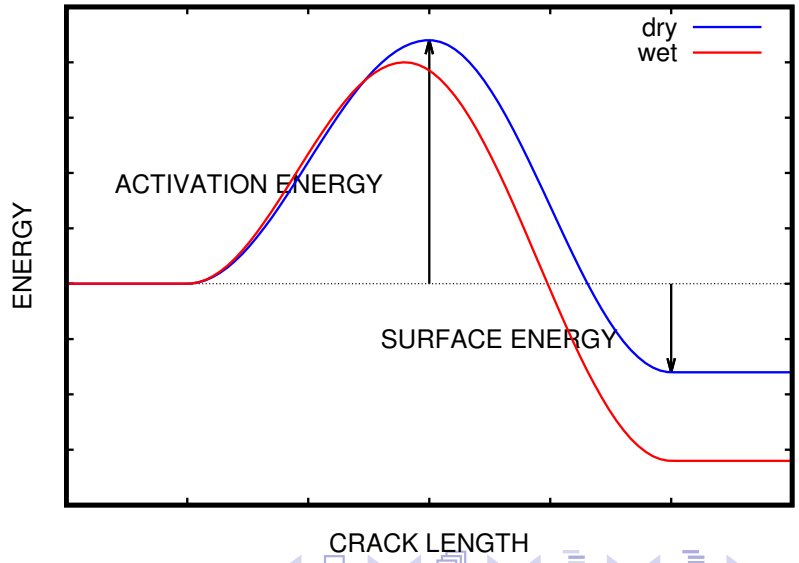
Mechanochemistry of atomic fracture



[Michalske 1983]



- opening due to far-field mechanical loading
- species diffusion driven by concentration & pressure, subject to steric & hydrophobicity effects
- reactions near/at tip lower bond breaking barrier & decrease surface energy



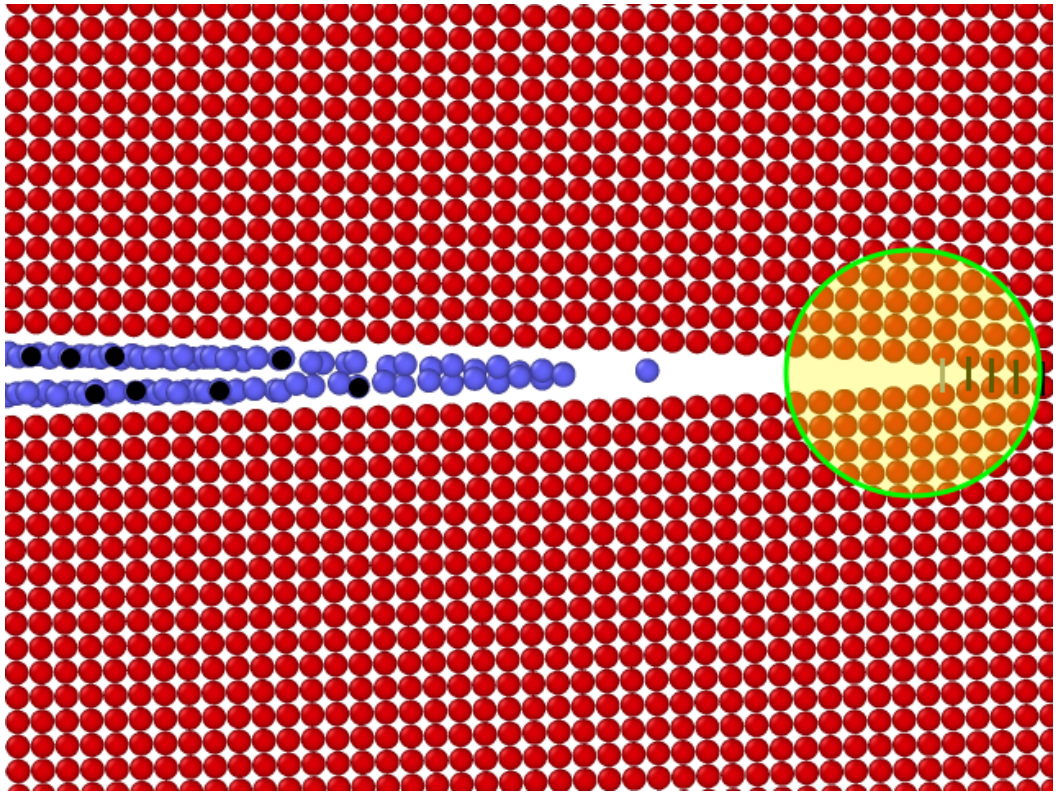
Extension of methodology to chemistry

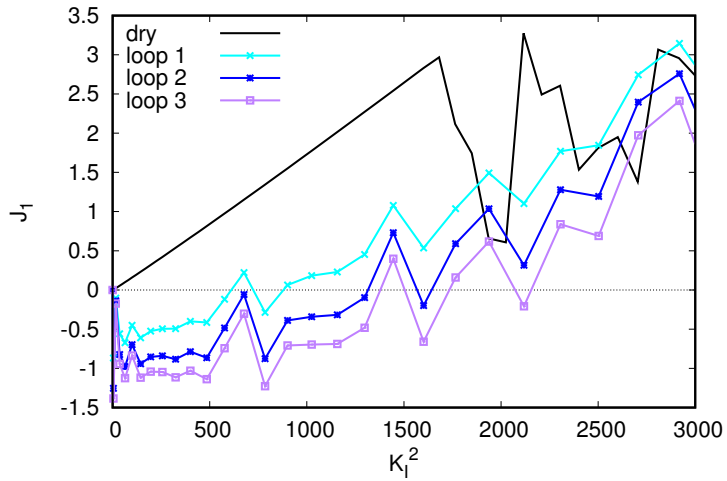
Levels of models

1. **Lennard-Jones solid & fluid**
   - simple, unified potential (Lorenz-Berthiot mixing)
   - easy to interpret & parameterize e.g. hydrophobicity, steric effects
   - useful in discovery of similarity relations
2. **Tersoff silica + SPC water**
   - polyatomic
   - atomic structure effects - crystalline, amorphous
3. **ReaxFF silica + water**
   - complex, unified potential
   - attention to reactions i.e. molecules break & reform bonds, e.g. $H_2O \leftrightarrow OH^- + H^+$
   - add pH and salt concentration effects
   - potential short coming reaction erodes sharp tip

Using a Lennard-Jones system to explore:

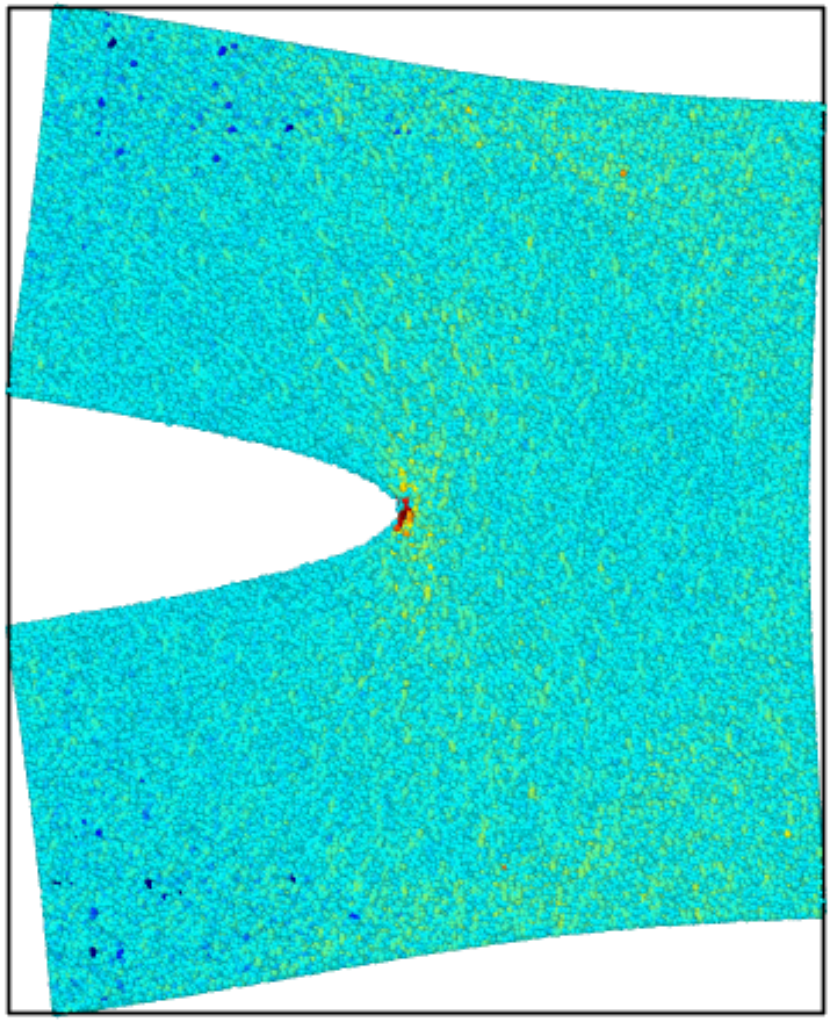
- **pressure**: function of density, acts on the crack faces & diffusion to tip
- **steric effects**: controlled by radius σ of fluid, particle size vs crack tip radius
- **hydrophobicity effects**: controlled by solid-fluid energy ϵ





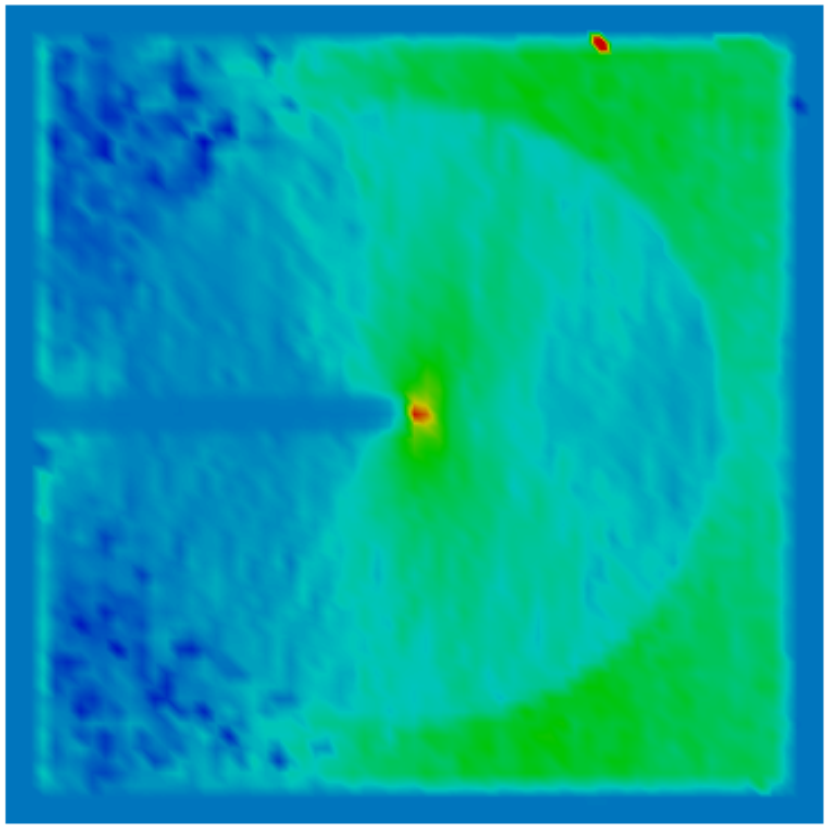
- solid
- fluid
- surface-bound fluid

Tersoff: silica glass



atomic

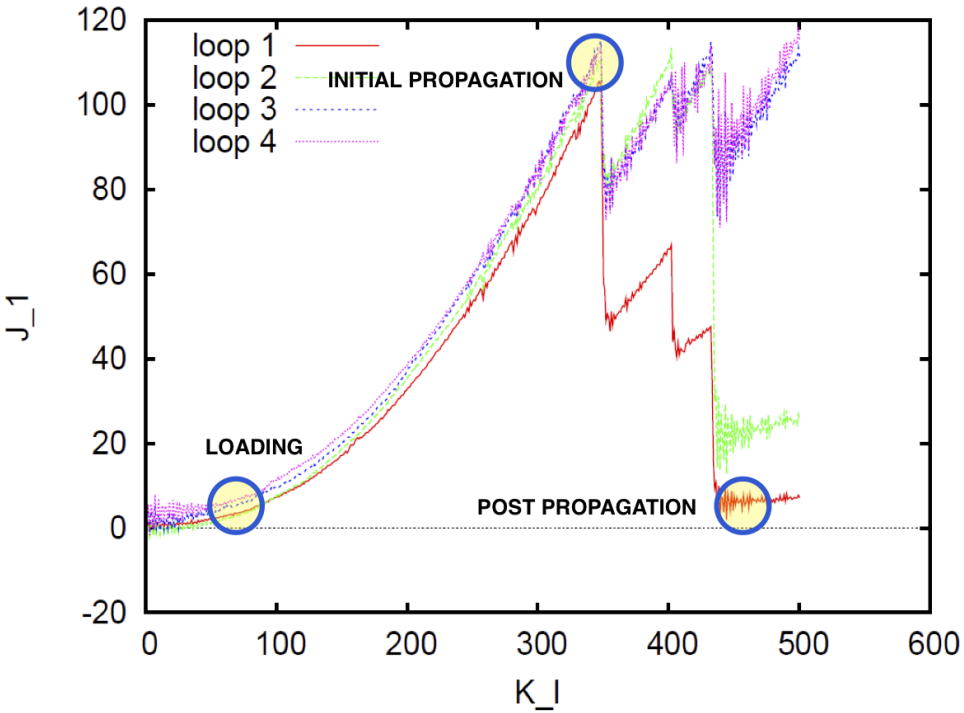
22-stress

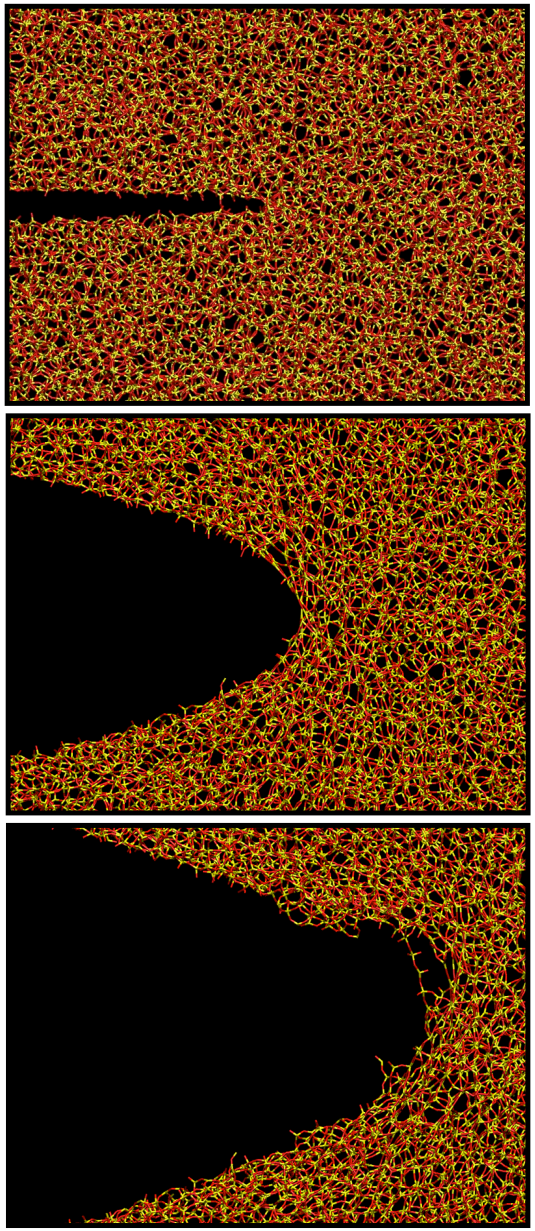


coarse-grained

appears to be extremely ductile

Crack progression in a (dry) ReaxFF system





- similar ductile behavior
- classical $J \sim K^2$ dependence
- propagation deviates from slit crack

Current work

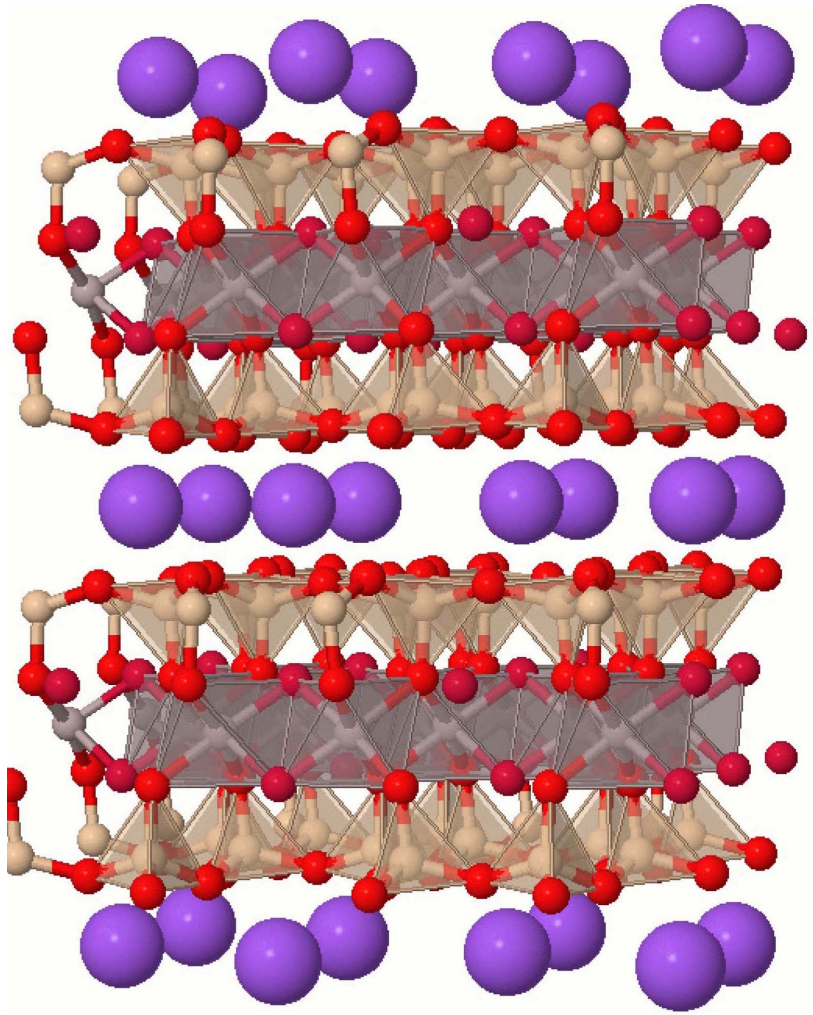
This atomistic-continuum approach

- shows path-independence
- good agreement with theory
- rational implementation
- extensible to many types of fracture

We are currently working on:

- reacting fluids
- pH effects
- mineral & clay structures

using potentials like ReaxFF.



montmorillonite

rjones@sandia.gov

additional slides

Current work

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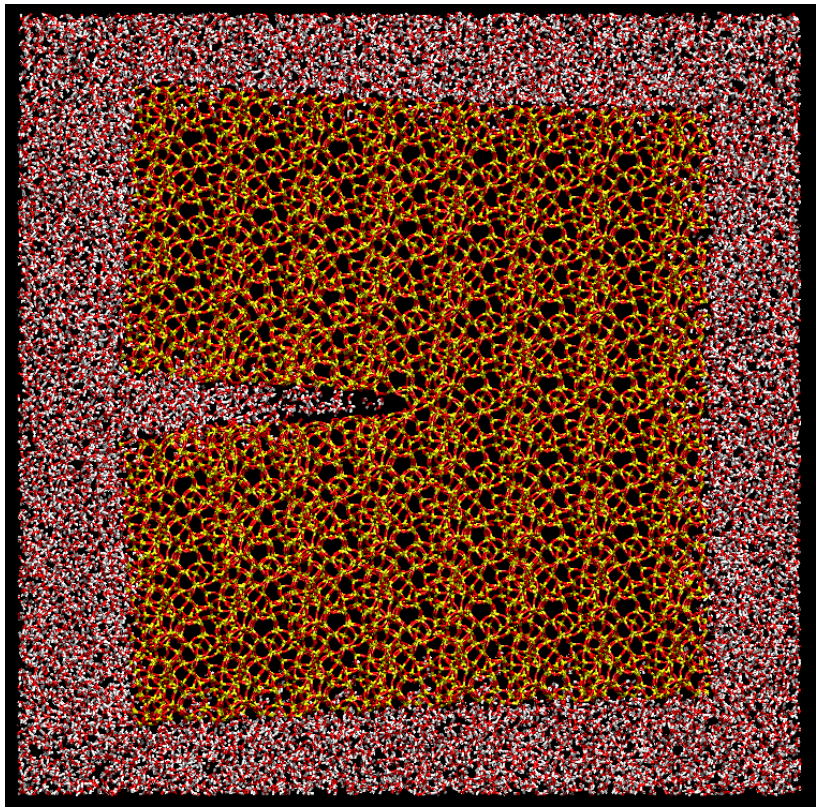

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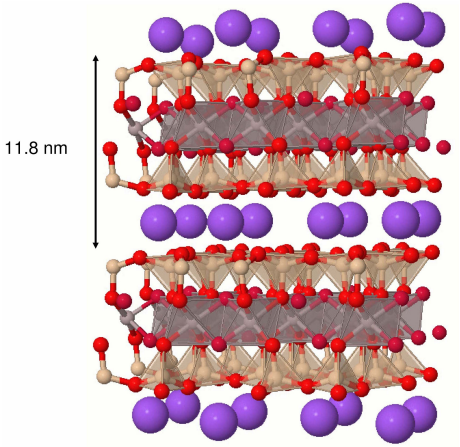

- reacting fluids
- pH effects
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using potentials like ReaxFF.

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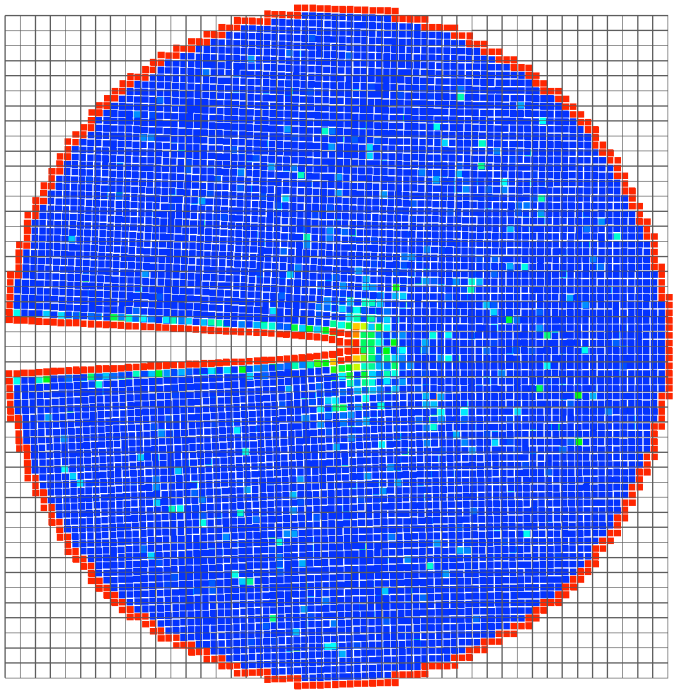
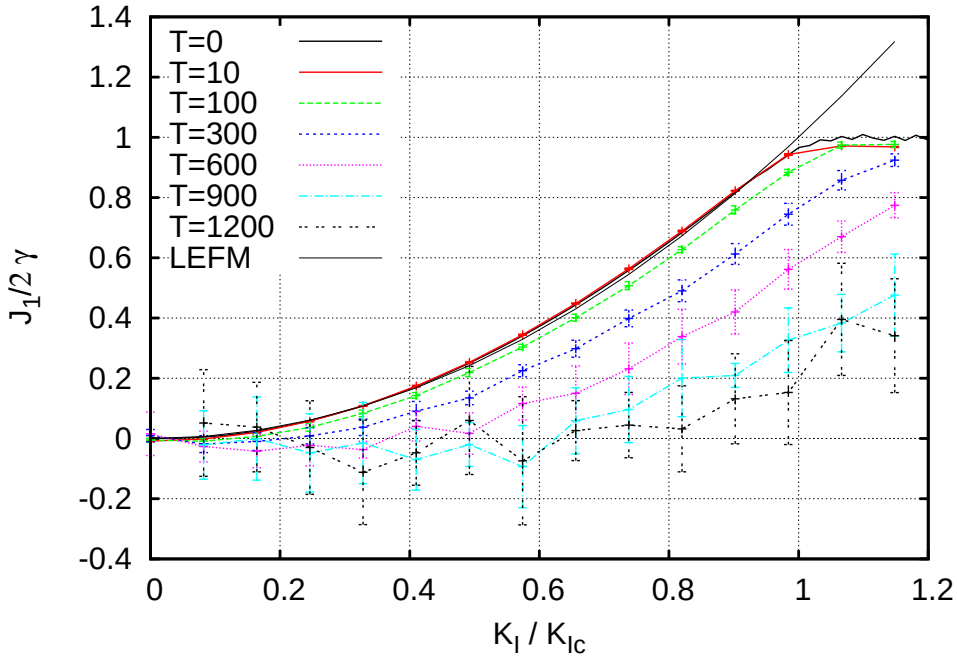
Quasi-static, finite temperature crack

Potential energy



J-integral for various temperatures

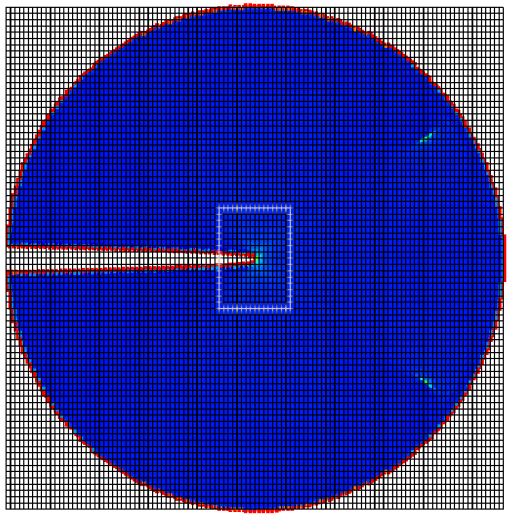


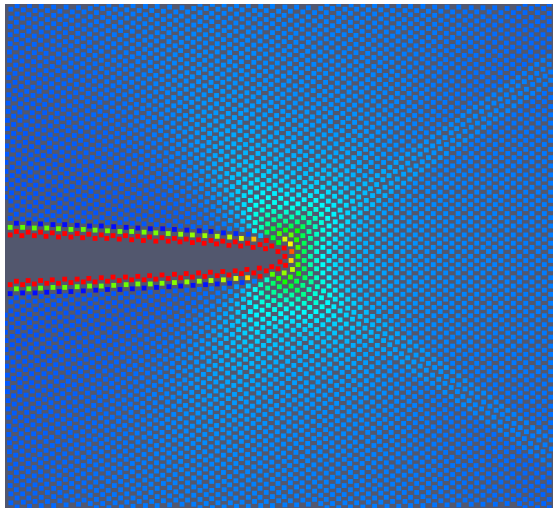
Significant variations due to thermal, surface and crack tip energies are apparent.

The J-integral is similar in shape at finite temperatures but shows delayed fracture due to thermal expansion.

Ductile fracture: EAM Au

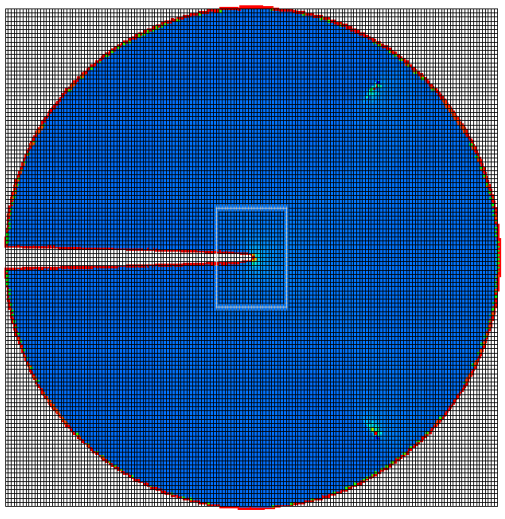
FCC

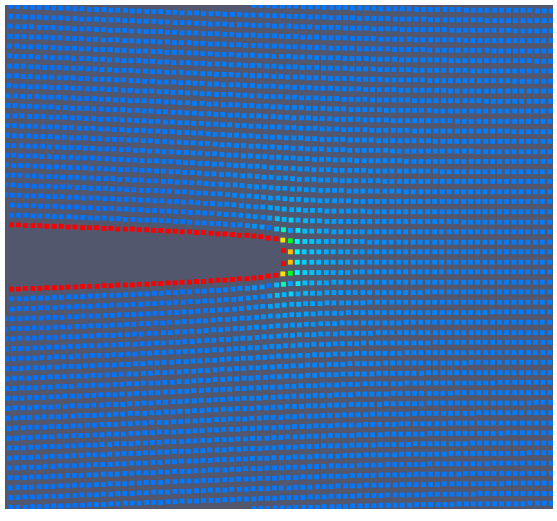




BCC displays significantly more blunting than FCC.

BCC

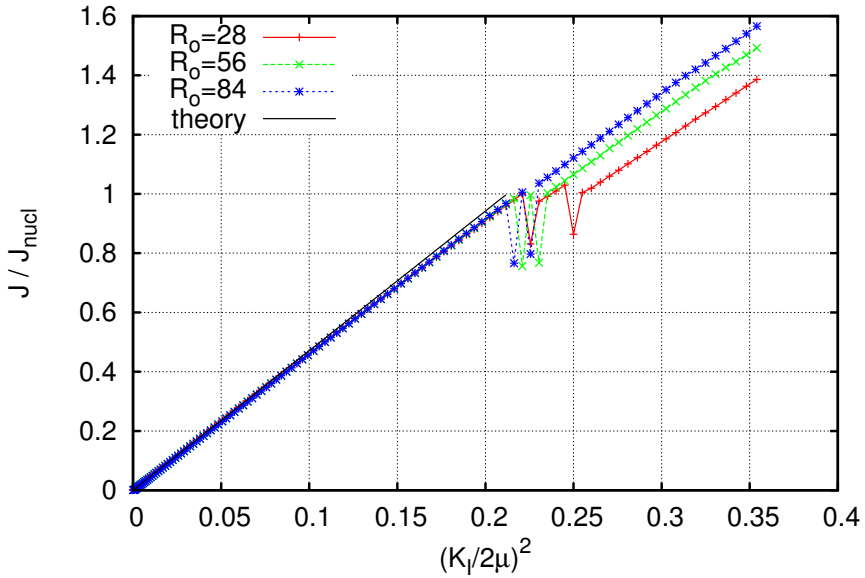




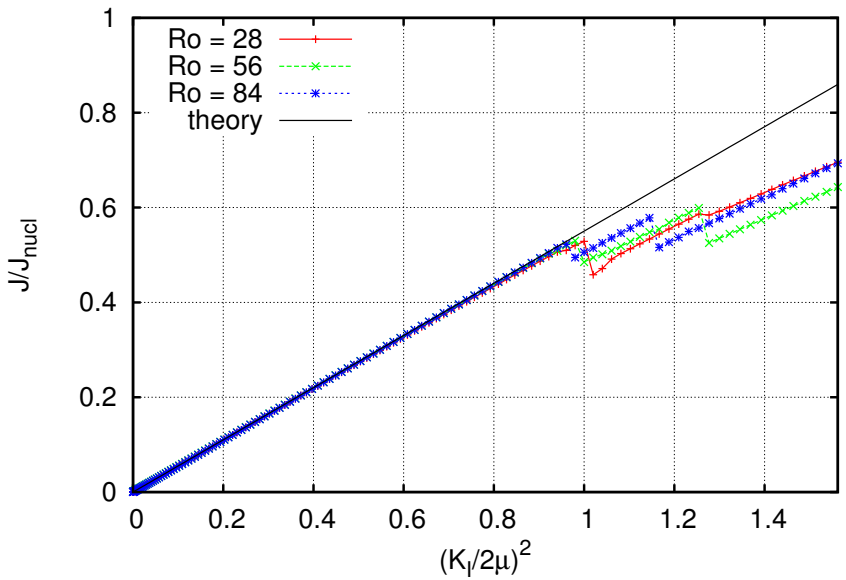
Both emit two dislocations that propagate to the boundary of the atomic system.

Ductile fracture

FCC



BCC

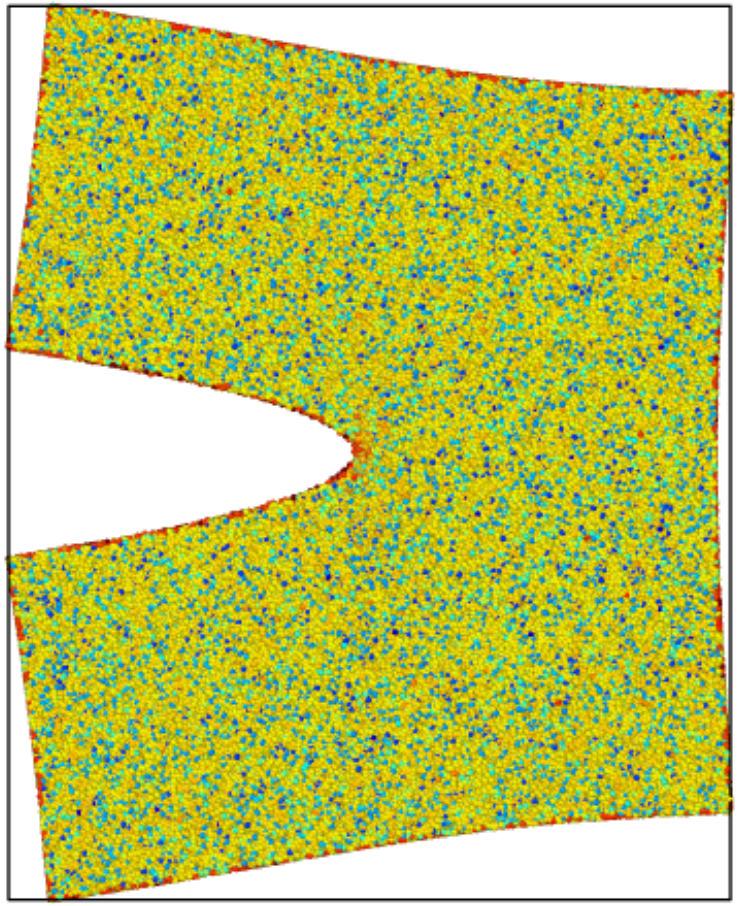


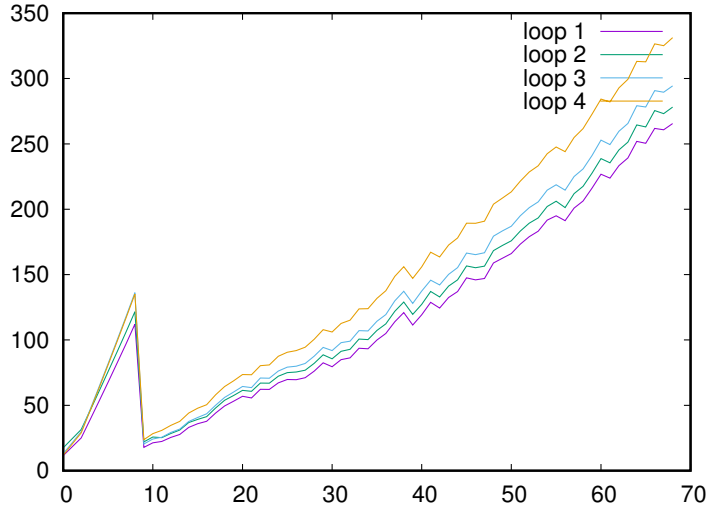
The J-integrals of both systems show evidence of two distinct emissions.

The limit of the elastic regime of the FCC system is nearly ideal with recovery after the dislocation emission, whereas the BCC system yields later, at a lower J value and does not recover after first emission.

The graphs also show **system size independence** for the pre-emission J integral i.e. integration contours are sufficiently far from the plastic zone.

Amorphous materials: silica glass

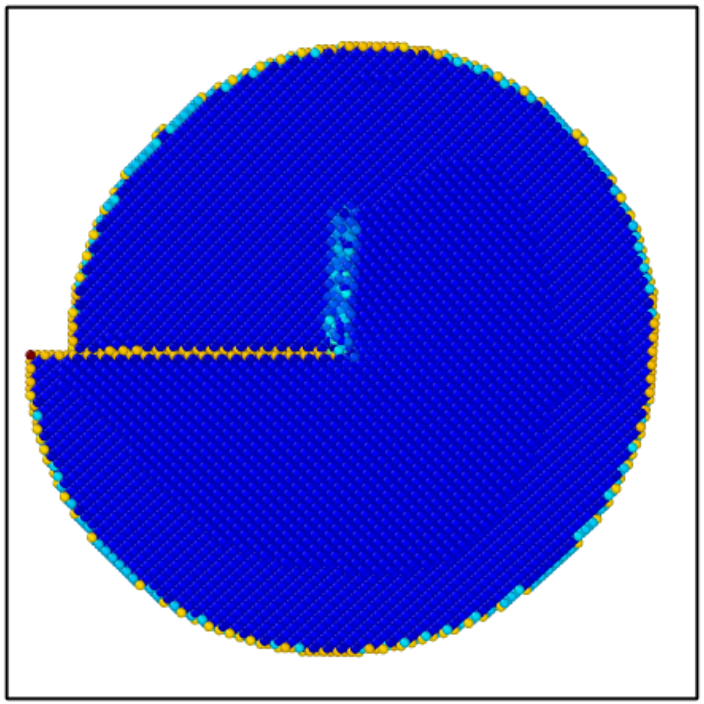




- appears to be very ductile
- likely has a large/larger non-continuum zone

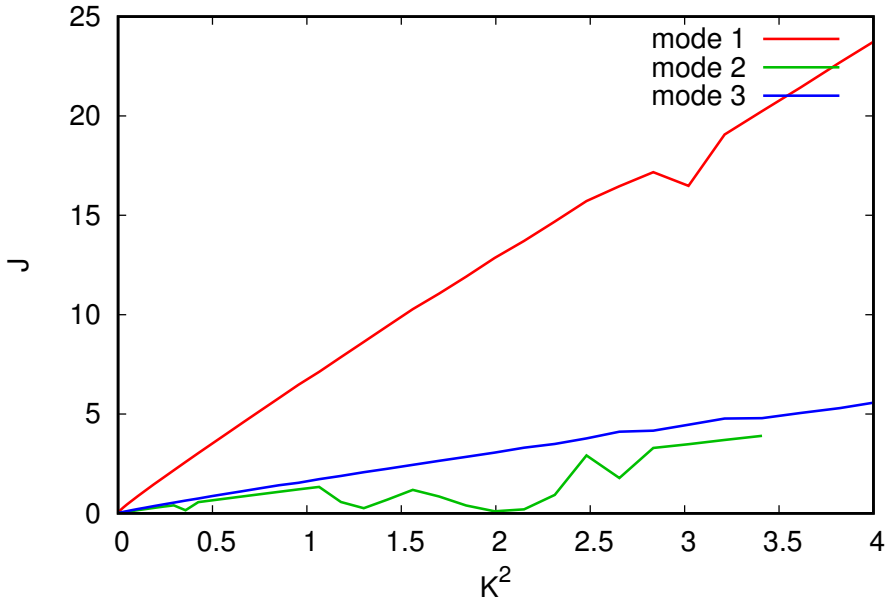
Mode dependence: silicon

Mode 2



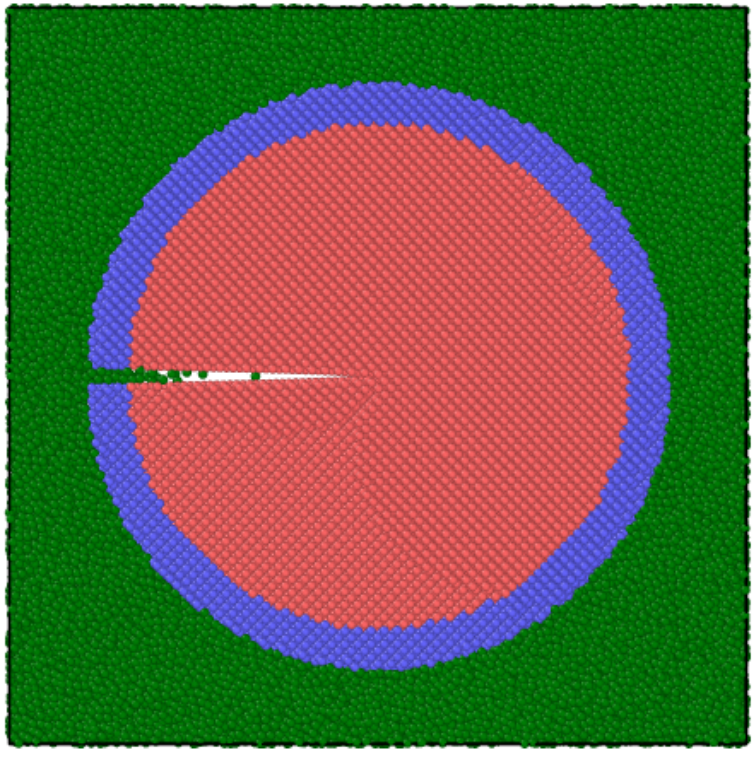
dislocation emission

EDIP: 100 orientation



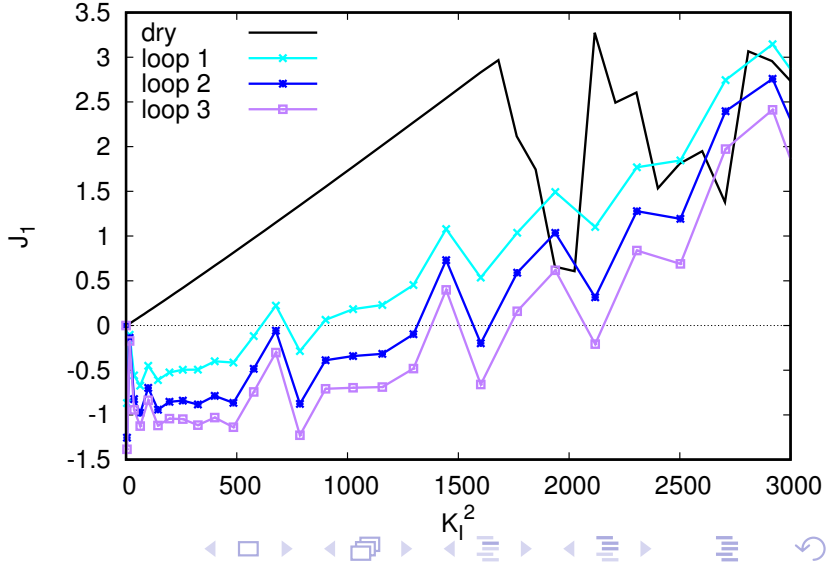
Mode 1, 2, 3

Fluid infiltration: beginning of chemistry...

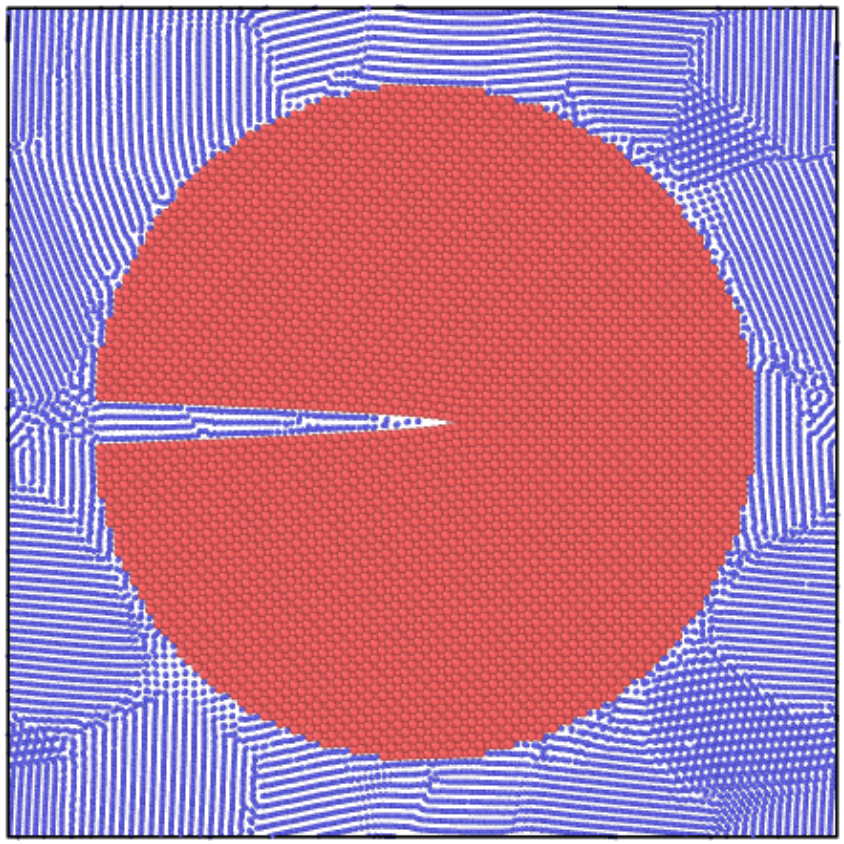


- Access to crack tip is limited by steric & diffusion consideration
- [Orowan 1944] surface adsorption can reduce surface energy
- pressure can do work on the leaves of crack and reduce the effective J

We see suppression of fracture and path dependence due to increased contribution by fluid exposure. The J integral needs to augmented to handle significant surface energy and stresses.



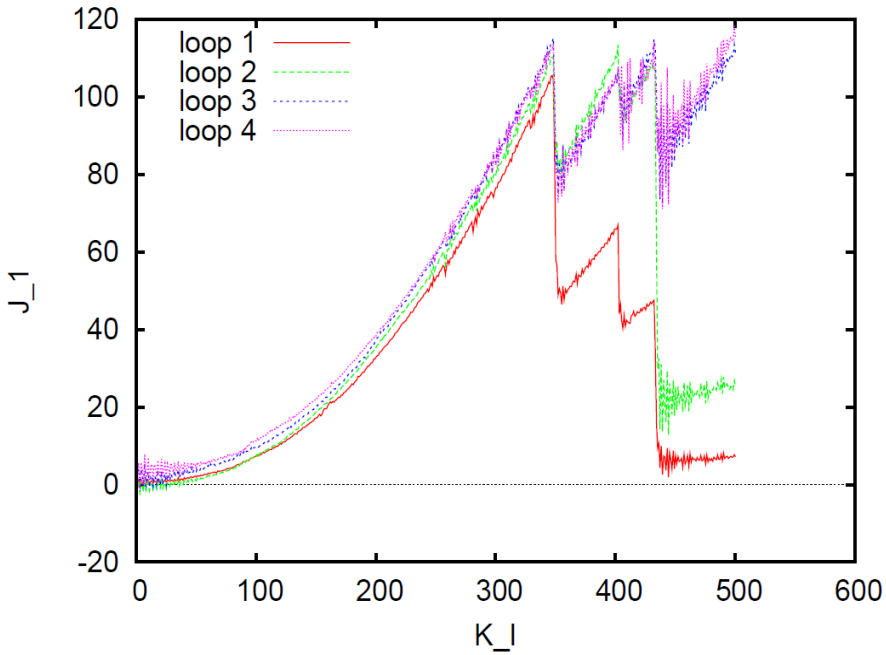
Lennard-Jones system

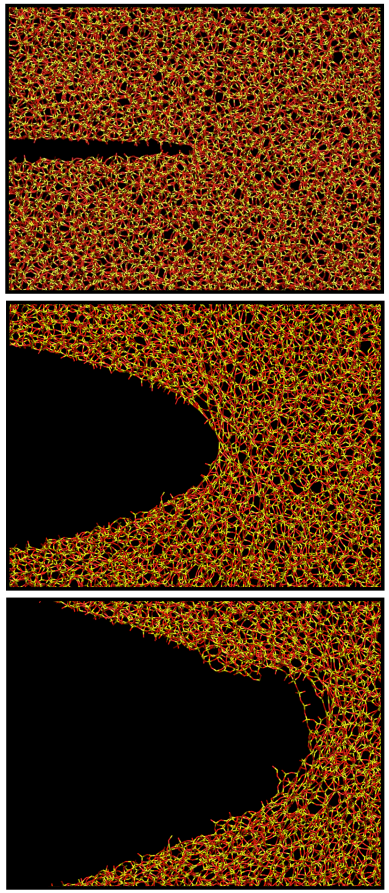


Using a cheap, simple to interpret system to explore

- pressure
- steric effects
- hydrophobicity effects

Crack progression



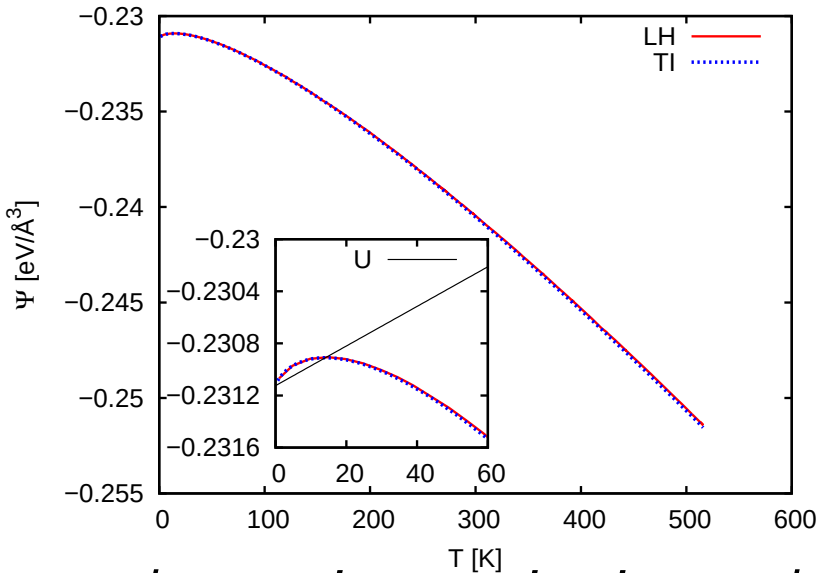


Finite temperature: free energy

For finite temperatures we need to use a *quasi-harmonic model* to estimate the free energy

$$\Psi_{QH} = \Phi_0 + \frac{k_B T}{V} \log \prod_{i=1}^{n} \frac{\hbar \omega_i}{k_B T}$$

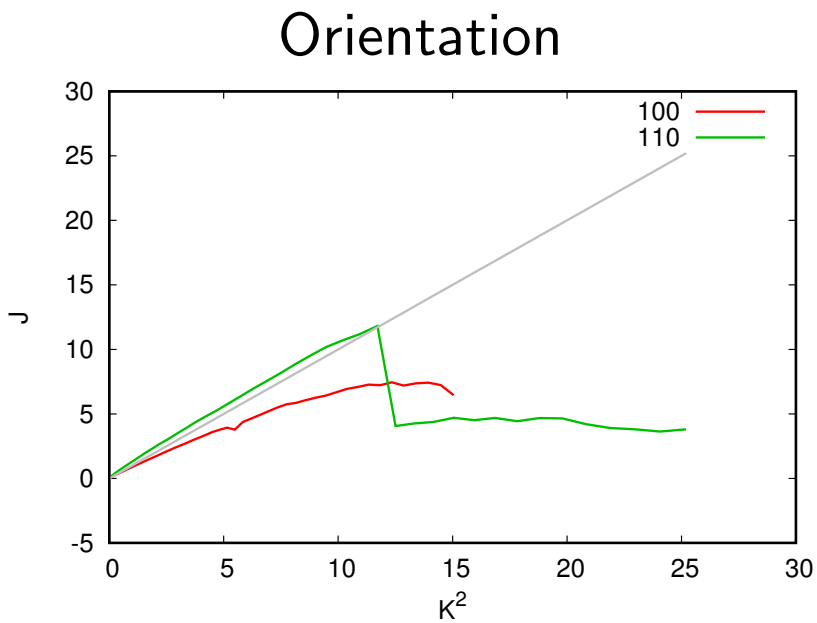
(via an estimate of the entropy/partition function).

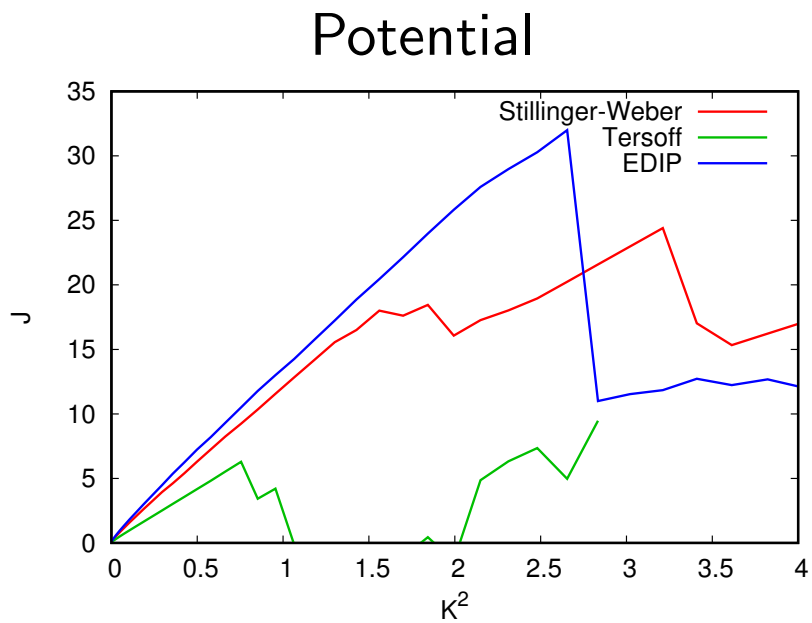


Heating of system under tension, notice internal U vs free energy

We verified the harmonic model was sufficiently accurate using thermodynamic integration $\Psi_1 - \Psi_0 = \int_0^1 \mathbf{P} \cdot \mathrm{d}\mathbf{F} + \int_0^1 S \, \mathrm{d}\, T$ over a variety of (homogenous) loading conditions.

Orientation & Potential dependence





(2) Consistent fields

The estimation of three energy-conjugate fields are necessary.

- **energy density**:

$$W(\mathbf{X},t) = \sum_{\alpha} \phi_{\alpha}(t)\,\psi(\mathbf{X}-\mathbf{X}_{\alpha}) - \overbrace{\sum_{\alpha} \phi_{\alpha}^{X}\psi(\mathbf{X}-\mathbf{X}_{\alpha})}^{W_0(\mathbf{X})}$$

  where $\phi_{\alpha}^{X} = \phi_{\alpha}(\{\mathbf{X}_{\beta}\})$ is PE in a *fixed reference configuration* $\{\mathbf{X}^{\beta}\}$ (perfect lattice w/ bonds deleted along crack).

- **displacement gradient** :

$$\mathbf{H} = \nabla_{\mathbf{X}}\mathbf{u} = \sum_{I} \mathbf{u}_{I}(t)\nabla_{\mathbf{X}} N_{I}(\mathbf{X}) \ .$$

  using *interpolation* $\mathbf{u} = \sum_{I} \mathbf{u}(\mathbf{X}_{I},t)N_{I}(\mathbf{X})$ of displacement

$$\mathbf{u}(\mathbf{X},t) = \frac{\sum_{\alpha}\left(\mathbf{x}_{\alpha}(t)-\mathbf{X}_{\alpha}\right) m_{\alpha}\psi(\mathbf{X}_{\alpha}-\mathbf{X})}{\sum_{\alpha} m_{\alpha}\psi(\mathbf{X}_{\alpha}-\mathbf{X})}$$

  defined *mass-weighted* consistent w/ momentum density.

- **stress**

$$\mathbf{P}(\mathbf{X},t) = -\sum_{\alpha<\beta} \mathbf{f}_{\alpha\beta}(t) \otimes \mathbf{X}_{\alpha\beta} B_{\alpha\beta}(\mathbf{X})$$

Eshelbian mechanics: J integral

The energy release rate *relative* to a process with motion $\boldsymbol{\chi}_t$, $\mathbf{F} = \partial_{\mathbf{X}} \boldsymbol{\chi}_t$, and a *fixed reference configuration* $\mathbf{X}$ is:

$$\dot{\Pi}(\boldsymbol{\varphi}_t, \boldsymbol{\chi}_t, \mathbf{F}) - \dot{\Pi}(\mathbf{X}, \boldsymbol{\chi}_t, \mathbf{F}) = \int \partial_{\mathbf{X}} \Psi \cdot \dot{\boldsymbol{\varphi}}_t \, dV$$

where $\boldsymbol{\varphi}_t(\mathbf{X})$ is a map describing the *configurational change* in the reference configuration, and

$$\partial_{\mathbf{X}} \Psi = \boldsymbol{\nabla}_{\mathbf{X}} \Psi(\mathbf{X}, \mathbf{F}(\mathbf{X})) - \partial_{\mathbf{X}} W(\mathbf{X}, \mathbf{F}) : \boldsymbol{\nabla}_{\mathbf{X}} \mathbf{F} = \boldsymbol{\nabla}_{\mathbf{X}} \cdot \mathcal{S}$$

is *work conjugate* to the configurational change $\boldsymbol{\varphi}_t$.

$\mathcal{S}$ is the Eshelby stress and the **J**-integral is the resultant force

$$\mathbf{J} = \int_{\partial\Omega} \mathcal{S}\mathbf{N} \, dA = \partial_{\mathbf{X}} \Pi$$

conjugate to the motion of the crack tip $\dot{\boldsymbol{\varphi}}_t$.

(2) Consistent coarse-grained fields: momentum

In order to construct the necessary fields, take Newton's law

$$m_\alpha \ddot{\mathbf{u}}_\alpha = \mathbf{f}_\alpha$$

with mass m_α, displacement $\mathbf{u}_\alpha$ and force $\mathbf{f}_\alpha = \partial_{\mathbf{x}_\alpha} \Phi$, and localize with kernel $\psi = \psi(\mathbf{X})$ where $\psi_\alpha \equiv \psi(\mathbf{X}_\alpha - \mathbf{X})$ is an averaging weight

$$\sum_\alpha m_\alpha \ddot{\mathbf{u}}_\alpha \, \psi_\alpha = \sum_\alpha \mathbf{f}_\alpha \, \psi_\alpha$$

The left-hand side can be identified with a change in **momentum**

$$\mathbf{p}(\mathbf{X}, t) = \sum_\alpha m_\alpha \dot{\mathbf{u}}_\alpha(t) \, \psi_\alpha$$

and the right-hand side with a divergence of **stress**

$$\boldsymbol{\nabla}_\mathbf{X} \cdot \mathbf{P} = \sum_\alpha \mathbf{f}_\alpha \, \psi_\alpha = \sum_{\alpha\beta} \psi_\alpha \mathbf{f}_{\alpha\beta} = \frac{1}{2} \sum_{\alpha\beta} (\psi_\alpha - \psi_\beta) \mathbf{f}_{\alpha\beta} = \dots$$

(2) Irving-Kirkwood/Hardy formalism

To obtain the stress from its divergence, instead of a truncated expansion for $\Delta\psi = \psi_\alpha - \psi_\beta$, define an **exact relation**

$$\Delta\psi = \boldsymbol{\nabla}_{\mathbf{X}}\psi \cdot \Delta\mathbf{X} + \ldots = \boldsymbol{\nabla}_{\mathbf{X}} B \cdot \Delta\mathbf{X}$$

via

$$B_{\alpha\beta}(\mathbf{X}) = \int_0^1 \psi\left(\lambda(\mathbf{X}_\alpha - \mathbf{X}) + (1-\lambda)(\mathbf{X}_\beta - \mathbf{X})\right)\, d\lambda$$

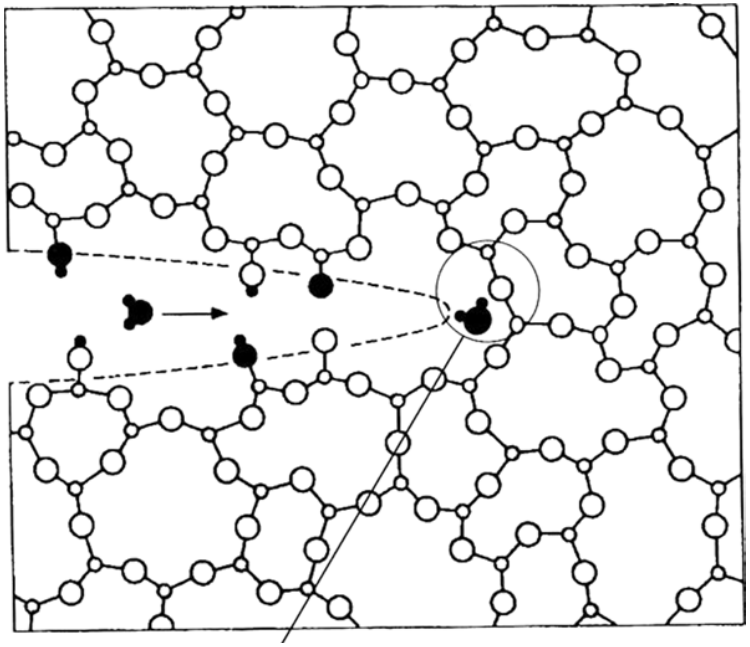
to obtain a stress (recall $\boldsymbol{\nabla}_{\mathbf{X}} \cdot \mathbf{P} = \frac{1}{2}\sum_{\alpha,\beta} \mathbf{f}_{\alpha\beta}\, \boldsymbol{\nabla}_{\mathbf{X}} B_{\alpha\beta} \cdot \mathbf{X}_{\alpha\beta}$)

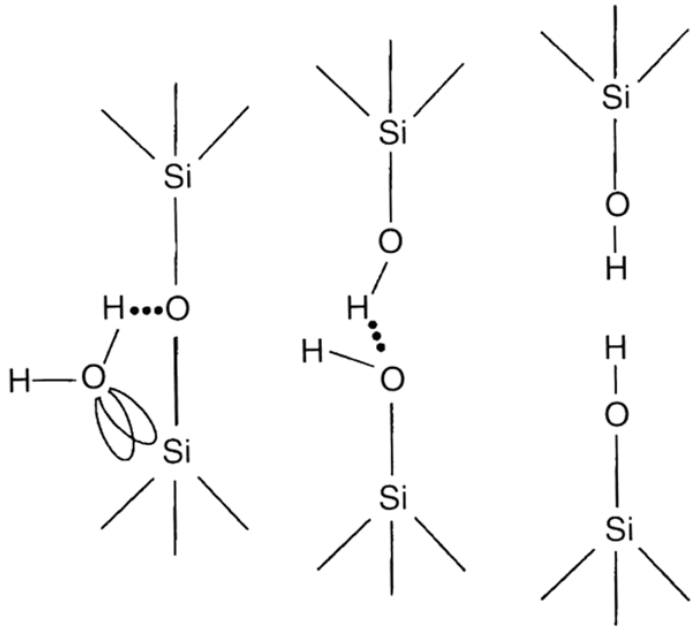
$$\mathbf{P}(\mathbf{X}, t) = -\frac{1}{2}\sum_{\alpha,\beta} \mathbf{f}_{\alpha\beta}(t) \otimes \mathbf{X}_{\alpha\beta}\, B_{\alpha\beta}(\mathbf{X})$$

which is unique up to a solenoidal field & is similar to the **virial**.

Chemistry

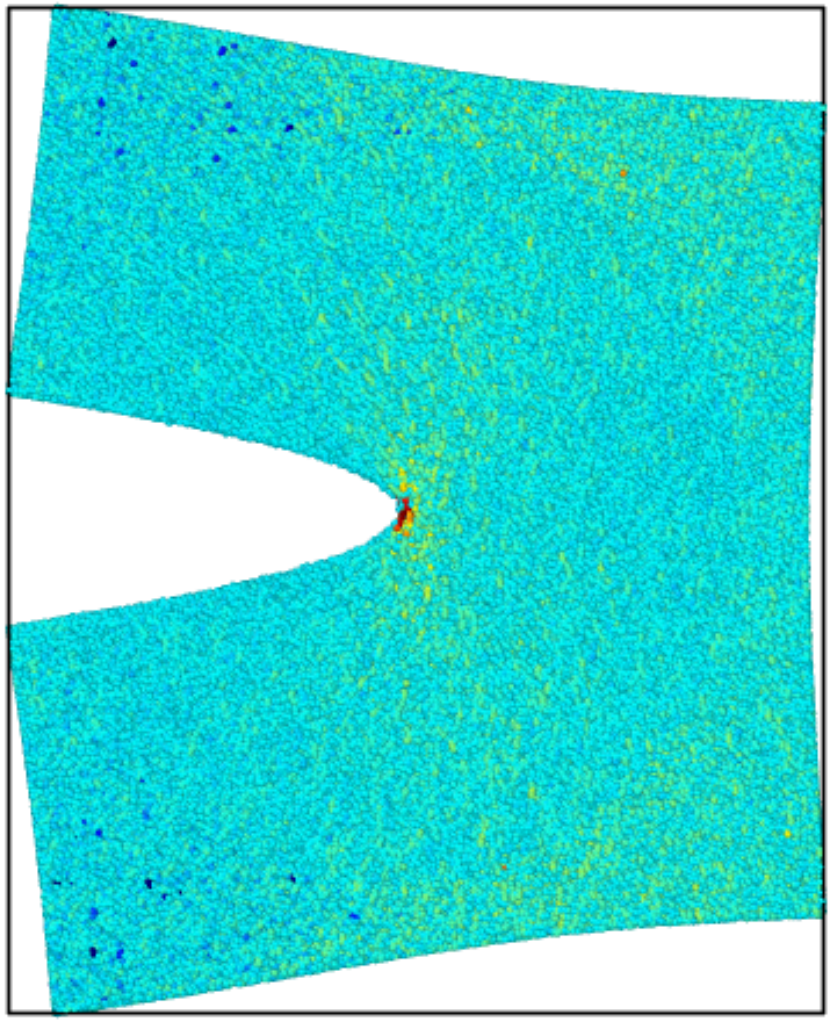
The interaction of the crack and a fluid can change the apparent fracture toughness. Chemical as well as mechanical influences can drive fracture.



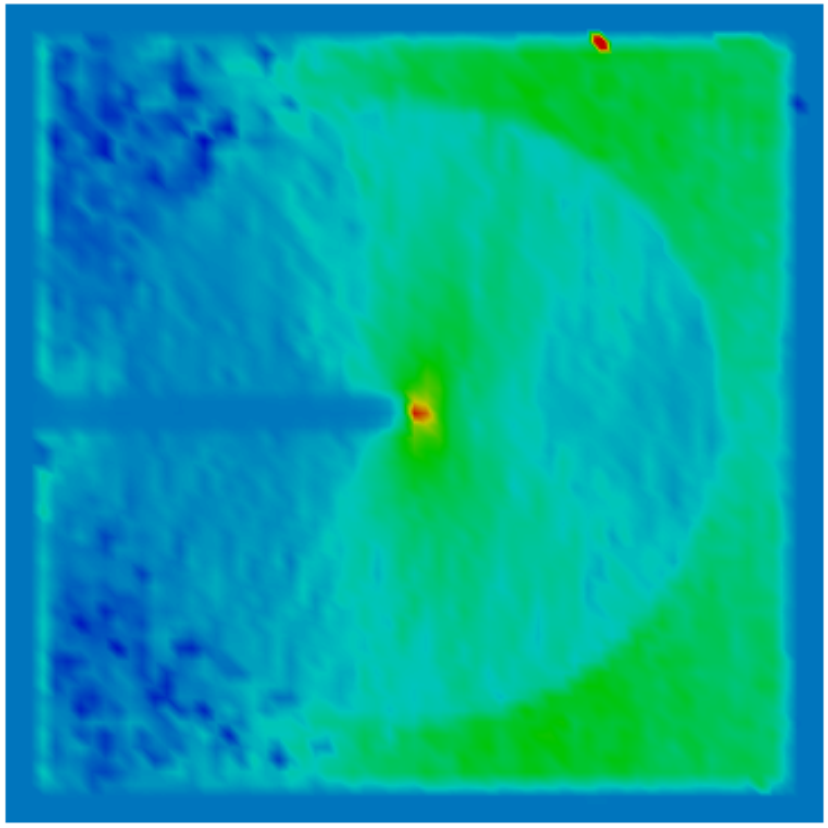


[Michalske 1983]

Amorphous materials: silica glass



atomic



coarse-grained