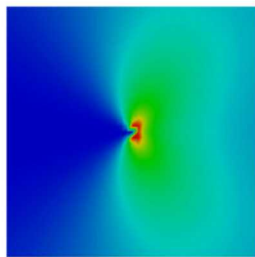
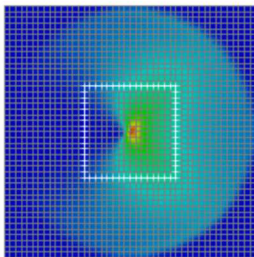


Assessing the fracture strength of geological and related materials via an atomistically based J-integral

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Sandia National Laboratories



atomistic vs. continuum

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Outline

Introduction

Motivation

History

Theory

Configurational forces

Coarse-graining

Results

Basic

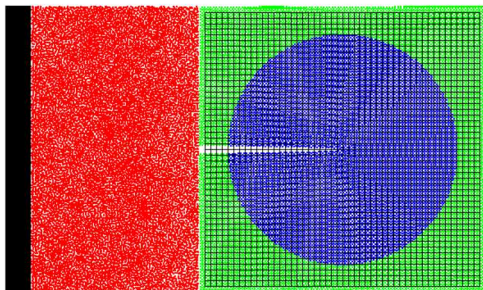
Complex materials

Mechanochemistry

Conclusion

Current Work

Wall regulates pressure in fluid



Green atoms provide far-field loading of the crack and allow access of the pressurized fluid

please ask questions as we go

Technological motivation

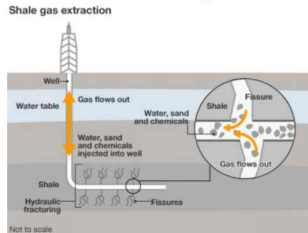
Goals:

- ▶ 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**.

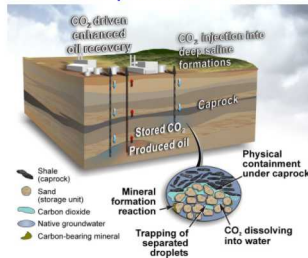
Why Atomistic Simulation?

- ▶ 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**:

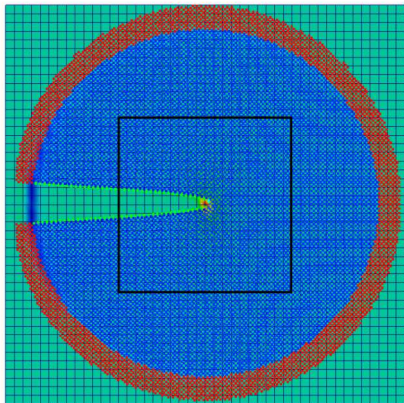
- *consistent* fields from atomistic simulation
 - application of continuum theory
- } → nanoscale results

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

(1) Eshelbian mechanics: J integral

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

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

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

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

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

(1) 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**.

At zero temperature equilibrium, the **J**-integral reduces to

$$\mathbf{J} = \int_{\partial\Omega} \mathcal{S} \mathbf{N} dA = \int_{\partial\Omega} \Psi \mathbf{N} - \mathbf{F}^T \mathbf{P} \mathbf{N} dA = \int_{\partial\Omega} W \mathbf{N} - \mathbf{H}^T \mathbf{P} \mathbf{N} dA$$

where W is the internal energy, $\mathbf{H} = \nabla_{\mathbf{x}} \mathbf{u}$ is the displacement gradient, and \mathbf{N} is the outward unit normal.

(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}_α 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**

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

(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**

$$\Delta\psi = \nabla_{\mathbf{x}}\psi \cdot \Delta\mathbf{X} + \dots = \nabla_{\mathbf{x}}B \cdot \Delta\mathbf{X}$$

via

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

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

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

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

(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

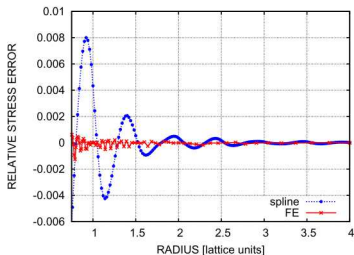
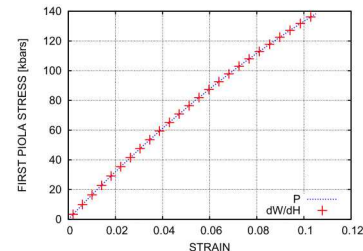
$$\mathbf{u}(\mathbf{X}, t) = \frac{\sum_{\alpha} (\mathbf{x}_{\alpha}(t) - \mathbf{X}_{\alpha}) m_{\alpha} \psi(\mathbf{X}_{\alpha} - \mathbf{X})}{\sum_{\alpha} m_{\alpha} \psi(\mathbf{X}_{\alpha} - \mathbf{X})}$$

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

- ▶ **stress**

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

Results: Consistency and Accuracy



The coarse-grained fields satisfy

$$\mathbf{P} = \nabla_{\mathbf{F}} \Psi$$

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

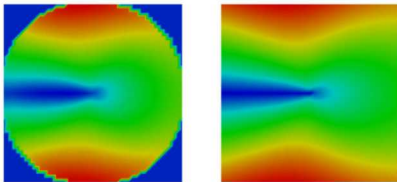
Also our use of *partition of unity*/FE kernels

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

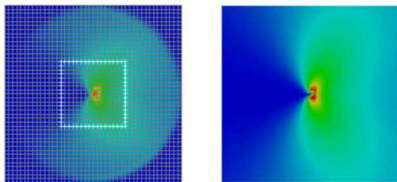
(and interpolation) can lead to faster and more *accurate* estimates than with traditional kernels.

Idealized crack with displacement boundary conditions

Hardy vs. LEFM

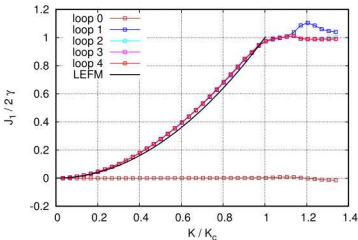


displacement

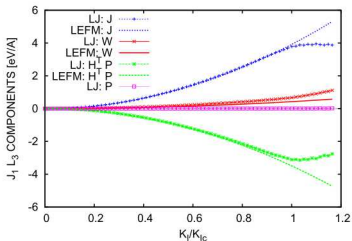


stress

- path independence



- expected limit: 2γ at $K = K_c$



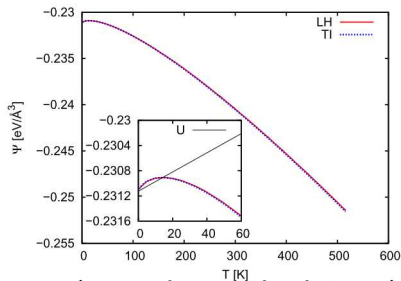
- components W & $H^T P$ match theory

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).

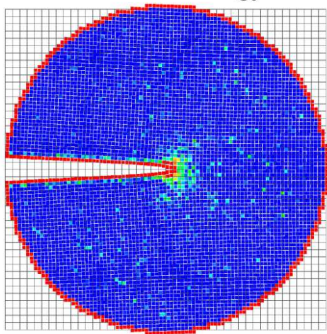


Heating of system under tension, notice internal U vs free energy

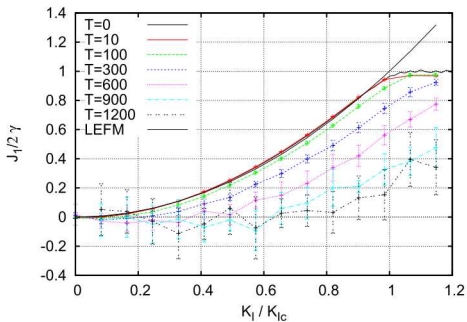
We verified the harmonic model was sufficiently accurate using thermodynamic integration $\Psi_1 - \Psi_0 = \int_0^1 \mathbf{P} \cdot d\mathbf{F} + \int_0^1 S dT$ over a variety of (homogenous) loading conditions.

Quasi-static, finite temperature crack

Potential energy



J-integral for various temperatures

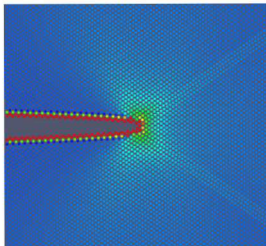
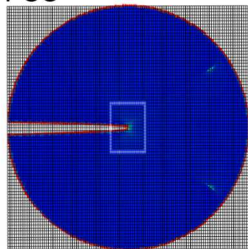


Significant variations due to thermal, surface and crack tip energies are apparent.

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

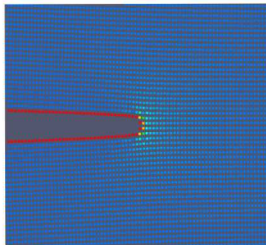
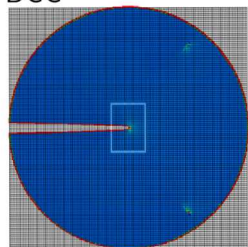
Ductile fracture: EAM Au

FCC



BCC displays significantly more blunting than FCC.

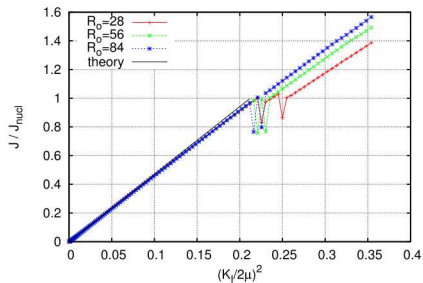
BCC



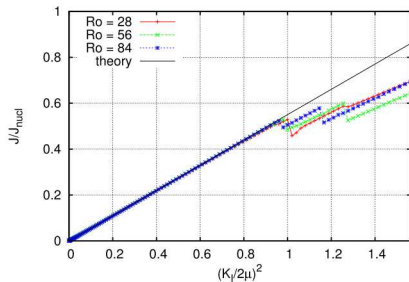
Both emit two dislocations that propagate to the boundary of the atomic system.

Ductile fracture

FCC



BCC

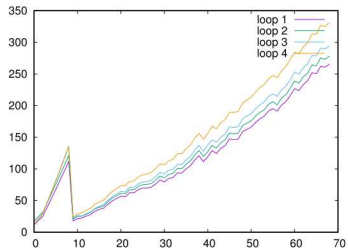
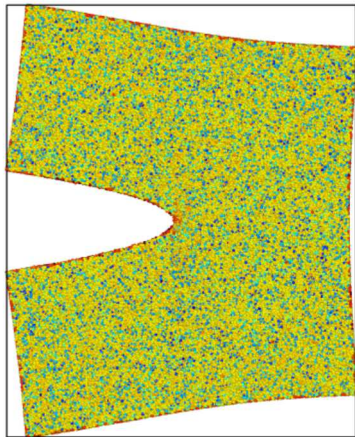


The J-integrals of both systems show evidence of two distinct emissions.

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.

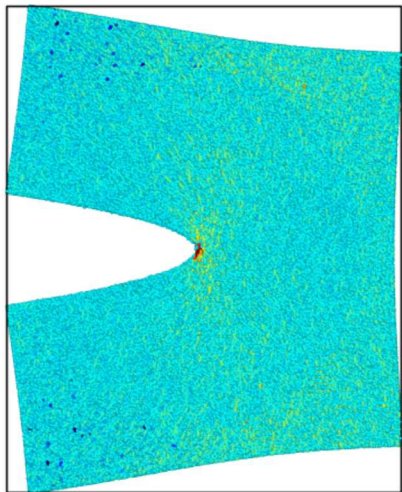
The graphs also show **system size independence** for the pre-emission J integral i.e. integration contours are sufficiently far from the plastic zone.

Amorphous materials: silica glass

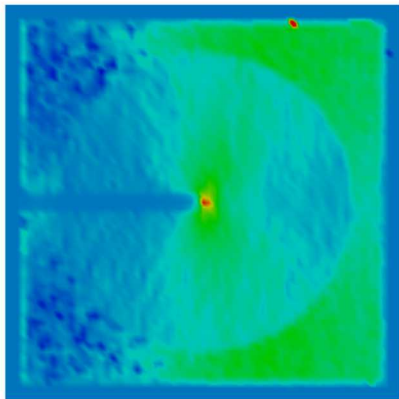


- ▶ appears to be very ductile
- ▶ likely has a large/larger non-continuum zone

Amorphous materials: silica glass



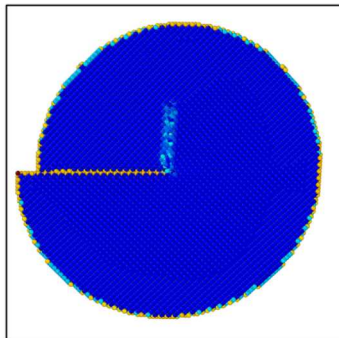
atomic



coarse-grained

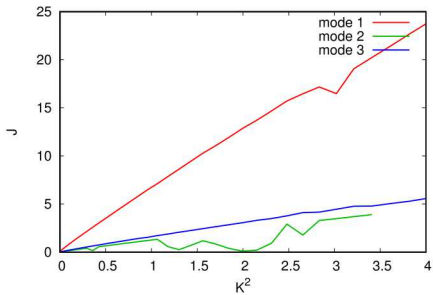
Mode dependence: silicon

Mode 2



dislocation emission

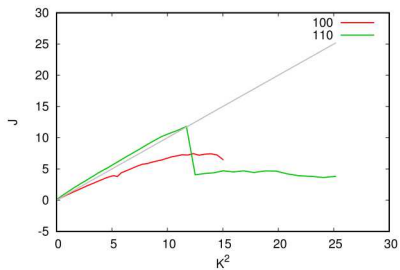
EDIP: 100 orientation



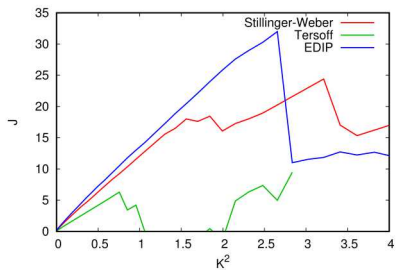
Mode 1, 2, 3

Orientation & Potential dependence

Orientation

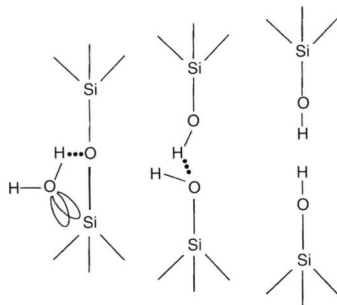
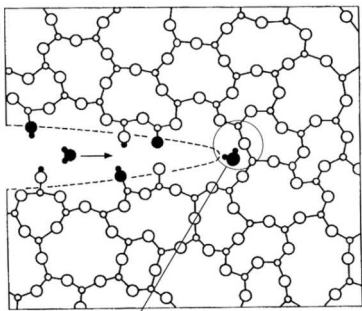


Potential



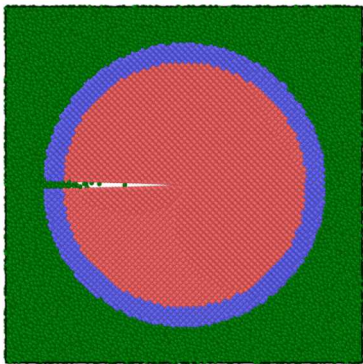
Chemistry

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



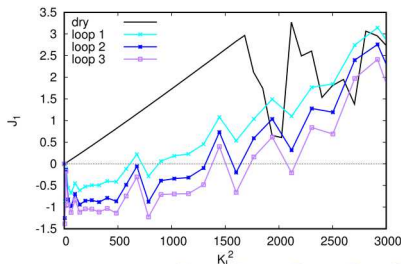
[MICHALSKE 1983]

Fluid infiltration



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

- ▶ 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

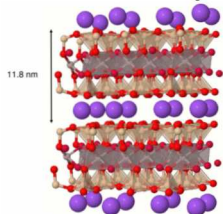


Current work

This atomistic-continuum approach

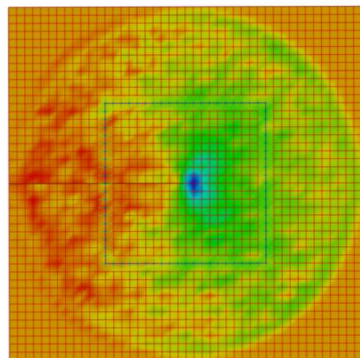
- ▶ shows path-independence
- ▶ good agreement with theory
- ▶ rational implementation
- ▶ extensible to many types of fracture

We are currently working on:



- ▶ reacting fluids
- ▶ pH effects
- ▶ mineral & clay structures

using potentials like REAXFF.



Eshelby stress 22 (eV/Å³)



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