A Multiscale Cohesive Zone Model for Rate Dependent Fracture of Interfaces
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Rate-dependent fracture of interfaces

Kinetically Controlled Transfer Printing (Meitl et al., 2006; Feng et al., 2007)

Selective dry transfer of graphene (Na et al., 2015; Xu et al., 2019)
Rate-dependent fracture of a silicon/epoxy interface

Hypothesis: toughness/strength depend on the local separation rate.

A multiscale modeling approach

- Interfacial traction and separation:
  \[ \delta = \lambda_n n r_0 \]
  \[ \sigma = \int (1 - D_n)\rho_0(n)f(\lambda_n)dn \]

- Damage evolution (thermally activated):
  \[ \frac{dD_n}{dt} = \frac{n}{t_0} (1 - D_n)\exp\left(-\frac{E_b}{k_B T}\right) \]

- An energy barrier:
  \[ E_b = E_b(f) \text{ or } E_b(\lambda_n) \]

Bond model (Kuhn segment)

2 bond parameters:
- $r_0 \rightarrow$ equilibrium bond length
- $\varepsilon_0 \rightarrow$ bond energy

3 key features for a simple bond model:
1. The potential energy is minimized at an equilibrium bond length;
2. The potential energy becomes infinitely high as the two atoms approach each other;
3. The potential energy approaches zero as the two atoms are separated far apart from each other.

Chain model

A freely jointed chain with stretchable bonds

\[ \psi(\lambda, \lambda_b) = nU_b(\lambda_b) + nk_B T \left( \beta \mathcal{L}(\beta) + \ln \left( \frac{\beta}{\sinh \beta} \right) \right) \]

\[ \frac{f r_0}{k_B T} = \bar{f}(\lambda, \bar{\varepsilon}) \]

\[ \lambda_b = \frac{r}{r_0} \quad \lambda = \frac{l}{nr_0} \quad \beta = \mathcal{L}^{-1} \left( \frac{\lambda}{\lambda_b} \right) \quad \mathcal{L}(\beta) = \coth(\beta) - \beta^{-1} \]

The chain stretch transitions from linear (Gaussian) to nonlinear and from entropic to enthalpic, with a peak force and a peak stretch.

Kinetics of thermally activated chain scission

- **Bond survival probability**
  \[ \frac{\dot{R}_b}{R_b} = -\frac{1}{t_0} \exp\left(-\frac{E_b}{k_B T}\right) \]

- **Chain survival probability**
  \[ \frac{\dot{R}_{\text{chain}}}{R_{\text{chain}}} = -\frac{n}{t_0} \exp\left(-\frac{E_b}{k_B T}\right) \]

- **Damage evolution**
  \[ D = 1 - R_{\text{chain}} \]
  \[ \frac{dD}{dt} = \frac{n}{t_0} (1 - D) \exp\left(-\frac{E_b}{k_B T}\right) \]

\[ \sigma = Nf = (1 - D)N_0 f(\delta) \]

Assume a constant chain length \((n)\) for now.

A microscopic time scale:
\[ t_0 \sim \frac{\hbar}{k_B T} \sim 10^{-13} \text{ s} \]

What is the activation energy or energy barrier?

Is this relevant?

Energy barrier for chain scission

Transition-state theory: The transition from an equilibrium state of the chain to the state of a broken chain (with one broken bond) may be traversed along a path with the minimum energy barrier in the energy landscape.

\[
E_b = \phi^{*\text{max}} - \phi^{*\text{mi}} \approx \varepsilon_0 \left(1 - 1.664 \hat{f}^{6/7} + \hat{f} + 0.0236 \hat{f}^2\right)
\]

\[
\hat{f} = \frac{f r_0}{\varepsilon_0}
\]

- An external force lowers the energy barrier (nonlinearly) for chain scission.
- The energy barrier becomes zero at the peak force for each chain.
- Reverse transition (healing) is possible under displacement control.

Rate-dependent traction-separation relations

\[ \delta = \dot{\delta} t \]

\[ \sigma = (1 - D)N_0 f(\delta) \]

\[ \frac{dD}{dt} = \frac{n}{t_0} (1 - D) \exp \left( -\frac{E_b}{k_B T} \right) \]

\[ E_b \approx \varepsilon_0 \left( 1 - 1.664 \hat{f}^{6/7} + \hat{f} + 0.0236 \hat{f}^2 \right) \]

Rate-dependent interfacial properties

The initial stiffness is independent of the separation rate.

Both strength and toughness increase with increasing rates.

The predicted toughness is always less than that by the Lake-Thomas model ($\bar{\Gamma} = \bar{\varepsilon}$).

Effect of statistically distributed chain lengths

\[
\lambda_n = \delta / (nr_0)
\]

\[
\sigma = \int (1 - D_n) f(\lambda_n) \rho_0(n) dn
\]

\[
\frac{dD_n}{dt} = \frac{n}{t_0} (1 - D_n) \exp\left(-\frac{E_b}{k_B T}\right)
\]

The shorter chains break earlier, leading to lower strength. The presence of longer chains extends the range of separation.

Effect of chain length distribution on interfacial properties

- Interfacial stiffness: increases slightly, independent of rate
- Interfacial strength: decreases significantly with the relative deviation at fast rate
- Interfacial toughness: less sensitive to the chain length distribution

\[ \bar{\chi} = \frac{\chi}{n_0} \]

Finite element implementation (ABAQUS/UINTER)

Model parameters:
- Bond length \( r_0 \sim 0.5 \text{ nm} \)
- Bond energy \( \varepsilon_0 \sim 1 \text{ eV} \)
- Average chain length \( n_0 \sim 4000 \)
- Chain length deviation \( \chi \sim 230 \)
- Chain areal density \( N_0 \sim 8 \times 10^{17} \text{ m}^{-2} \)
- Microscopic time scale \( t_0 \sim 10^{-13} \text{ s} \)
Summary

- A multiscale, mechanism-based cohesive zone model was proposed for rate-dependent fracture of polymer interfaces, potentially linking the interfacial properties (i.e., stiffness, strength and toughness) to the molecular structures.
- Model parameters were determined by direct comparation to the DCB experiments.
- The predicted traction-separation relation depends on the local separation rate and the loading history (e.g., non-monotonic or cyclic).