Molecular Dynamics Modeling of Traction Between Polyethylene Fibrils

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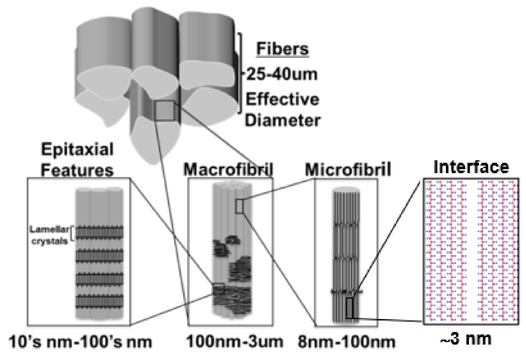
Introduction



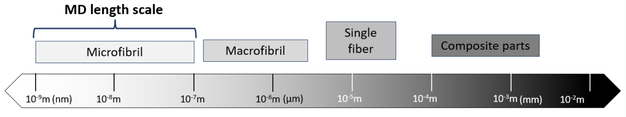
- UHMWPE is widely used in numerous structural applications.
- Atomistic modeling of interfaces between crystal structures provides insight into the behavior of inter-fibril interactions.
- Investigating interface interactions at different temperatures & strain rates is essential for advanced applications such as body armor.

Method

Modeling of fibril containing amorphous and crystalline regions (with and without voids)

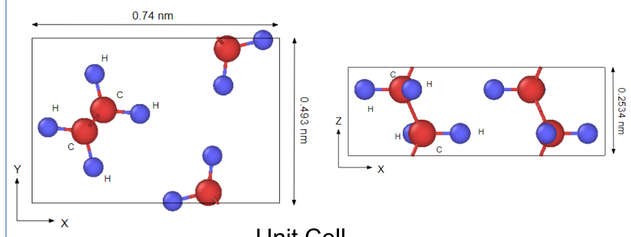
Modeling of the amorphous and crystalline regions in a fibril

Develop fibril interaction traction laws under different temperature and strain rates for bridging length scales for multi-fibril models



- Utilized LAMMPS with AIREBO-M potential to conduct molecular dynamics simulations.
- Postprocessing was done in MATLAB and OVITO.
- A single PE chain consists of 20 monomers (120 atoms). The model contains 240 chains.

Molecular Dynamics Models



E_b = Bulk Energy
E_{100} = (100) Model Energy
E_{010} = (010) Model Energy
$L_{\langle 001\rangle}$ = ⟨001⟩ Length = 10 nm

- Here, "surface energy" is the energy to create two new surfaces after crack propagation, which is the critical fracture energy.

$$\text{Surface Energy (100)} = \frac{(E_{100} - Eb)}{2(L_{\langle 010\rangle} L_{\langle 001\rangle})}$$

$$\text{Surface Energy (010)} = \frac{(E_{010} - Eb)}{2(L_{\langle 100\rangle} L_{\langle 001\rangle})}$$

- Total potential energy consists of three individual components

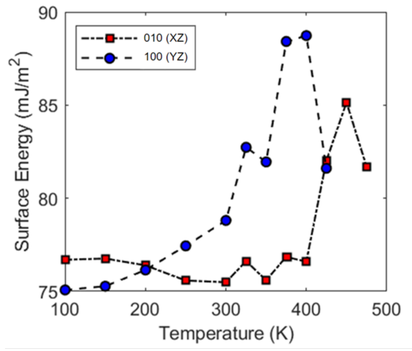
$$E^{\text{AIREBO}} = E^{\text{REBO}} + E^{\text{LJ}} + E^{\text{Torsion}}$$

Model	ΔE^{REBO} (eV)	ΔE^{LJ} (eV)	$\Delta E^{\text{torsion}}$ (eV)
(100) Model	3.0	42.4	3.7
(010) Model	2.5	38.4	1.5

Comparison with Literature

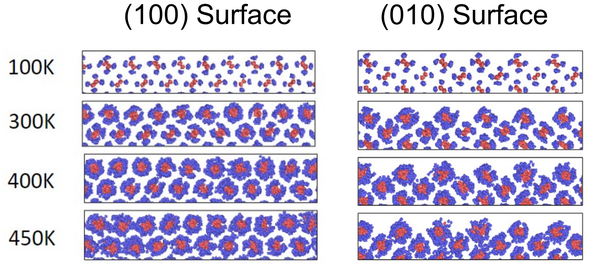
Reference	Surface Energy (mJ/m²)	Remarks
Current Work	***78.8 for (100) surface 75.5 for (010) surface***	***MD simulation with AIREBO-M potential***
Howard et al. *Macromolecules 47.32 (2014)*	145 for (100) surface 105 for (010) surface	MD simulations with quasi-harmonic approximations
Wilhelmi et al. *J. Phys. Chem.* 100.25 (1996)	143 for (100) surface 147 for (010) surface	MD simulations with KDG force field
Yeh et al. *J. Chem. Phys.* 149 (2018)	78 for (100) surface	MD simulations with bonding and debonding processes (AIREBO-M)
Schonhorn et al. J. Phys. Chem. 100 (1996)	*53.6*	*Experiments of single crystalline aggregates*
Owens et al. J. Appl. Polym. Sci. 13, 1741 (1969)	*33.1*	*Experiments of semicrystalline PE*

Surface Energy vs Temperature



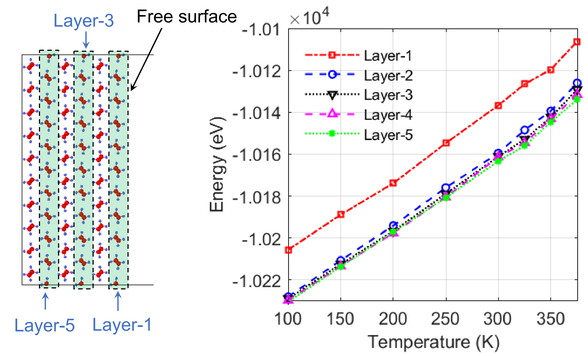
- Simulations reveal that an increase in temperature significantly influences the surface energy of (100) surface, which can be due to the change in entropy (*S*) and enthalpy (*H*): ***G = H - TS***

Surface Morphologies

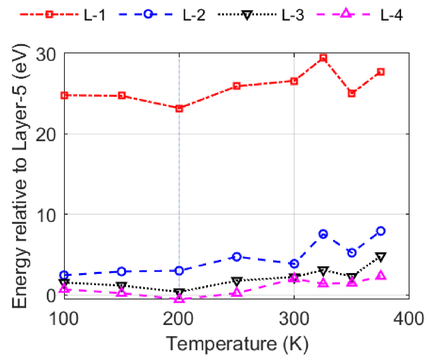


- Increasing temperature leads to higher kinetic energies of chains leading to higher energy conformations.

Energy Change with Thickness (100)



- Layer-1 has significantly higher energy, due to ~50% reduction in the coordination number of Layer-1 atoms.



- After 200 K, subsurface layers demonstrate increased energy compared to Layer-5, indicating that the interior atomic layers progressively absorb energy during the crack propagation at elevated temperatures.

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