7th World Tribology Congress, WTC 2022
July 10-15, 2022, Lyon, France

The Effect of Chain Architecture on the Tribology of Polymer Brushes using Coarse-Grained Molecular Dynamics

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Coarse-grained non-equilibrium molecular dynamics (NEMD) simulations were performed on polymer brush bilayers of different topologies to study their tribological behavior under a wide range of compressive pressures and shear rates. The topology of the brushes is varied by changing the grafting density as well as the polymer architecture. The polymers are modelled as bead-spring chains of different architectures including linear, looped, cyclic and bottle-brush. The results of the simulations are compared to scaling laws as well as experimental data focusing on the effect of chain architecture on the kinetic friction coefficient.

Keywords: polymer brush, bead-spring, molecular dynamics, chain architecture, tribology

1. Introduction

Polymer chains that are end-grafted to a flat substrate at sufficiently high grafting density begin to overlap and extend perpendicularly away from the substrate surface due to steric repulsion between monomers on neighbouring chains, thus forming what is known as a "polymer brush" [1]. When two opposing polymer brushes are brought into contact and compressed such that they interpenetrate form a "polymer-brush bilayer". These bilayers show excellent lubricating properties even when subjected to substantial normal loading [2]. An example of such a system is the articular cartilage found in the synovial joints of mammals [3].

2. Methodology

The simulations are performed using the LAMMPS software. The polymer chains are modelled in a coarse-grained way using a Kremer-Grest bread-spring model incorporating Finitely Extensible non-linear Elastic (FENE) bonds and Lennard Jones interactions between the monomers. The solvent is accounted either implicitly using a Langevin (or DPD) thermostat or with explicit solvent particles. The chains are end-grafted randomly to two opposing solid (FCC) substrates creating a polymer brush bilayer. The bilayer is compressed by applying a uniform force to the top substrate fixisng the bottom substrate (see fig. 1). The bilayer is then sheared by applying a constant relative velocity between the substrates. The kinetic friction coefficient is calculated as the ratio between the shear and normal components of the virial stress tensor. Brushes of varying topology, achieved by varying the chain architecture as well as the grafting density, are investigated. Simulations are performed at different compressions and shearing velocities covering several orders of magnitude.

3. Objective

We use NEMD simulations to explain the success of architectures employed in previous experiments [4], as well as to provide design principles for the synthesis of new brushes with improved tribological performance.

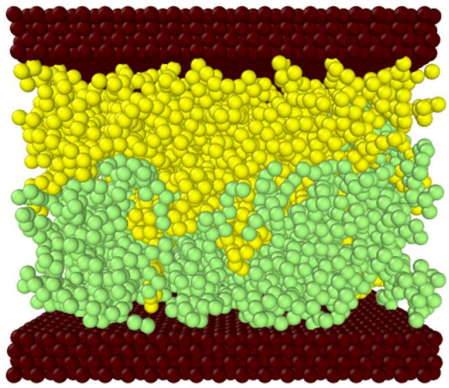

Figure 1. A snapshot from the molecular dynamics simulations the Polymer brush bilayer under compression.

4. References

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