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NANO-TRIBOLOGY OF A POLYTETRAFLUOROETHYLENE TRANSFER FILMS USING MOLECULAR DYNAMICS SIMULATION AND MICROTRIBOMETRY

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ABSTRACT

Polytetrafluoroethylene (PTFE) is a well known solid lubricant and polymer nanocomposites based on PTFE are considered to be promising materials for tribological applications in space. Like other polymer materials, many properties of PTFE depend on morphology. In this study, molecular dynamics (MD) simulations are performed to examine the effect of chain configuration on the frictional behavior of PTFE at the molecular level and compared to microtribological studies on aligned transfer films of PTFE.

INTRODUCTION

Mechanical devices for space applications need to be able to operate reliably in an extreme range of environments. They are produced and tested on earth, launched, and then operated in space. The reliability of the moving mechanical assemblies is one of the most critical issues in attempting to prolong service life. These components experience an extreme environmental range, and nanostructured polymer composite materials are candidate materials for improved performance. In particular, PTFE either as filler or a matrix is being explored. The C-F bond in PTFE provides both thermal and oxidative stability, and the strong interchain interactions in PTFE confer resistance to almost all solvents [1]. The intrinsically poor wear resistance of PTFE is improved by incorporation with nanoscopic fillers. For example, Sawyer *et al.* created a nanocomposite of PTFE with alumina that has a reduction in wear rate of over 2 orders of magnitude compared with unfilled PTFE [2].

In these nanocomposite systems PTFE transfer films provide a low friction coefficient and increased wear resistance. It is hypothesized that the molecular structure of the transfer film has a considerable effect on its tribological behavior. In this study, PTFE films are in sliding contact with one another in classical MD simulations and microtribometry. Our goal is to examine the effect of chain configuration on the frictional behavior of PTFE films at the molecular level.

EXPERIMENTAL METHODS

Classical MD simulations numerically integrate Newton's equations of motion with a third-order Nordsieck predictor corrector [3] using a timestep of 0.2 fs. Short-range interatomic forces are calculated using the C-H-F reactive empirical bond order (REBO) potential [4] based on Brenner's second generation REBO potential for hydrocarbon systems [5]. Long-range van der Waals interactions are also included in the form of a Lennard-Jones (LJ) potential to calculate interchain interaction [3]. The LJ potential is only active at distances greater than the covalent bond lengths.

A schematic representation of the initial system configuration is shown in Figure 1. Each PTFE film used in the simulations contains seven layers of chains for a total thickness of 40 Å and a sliding surface area of 45 Å × 45 Å. The bottom layer of the lower film is fixed and the top layer of upper film moves as a rigid unit, thereby allowing compression and sliding. The two layers next to the fixed or moving rigid regions are thermostated with Langevin frictional and stochastic forces to maintain the system temperature at 300 K [3]. The rest of the atoms in the system have no constraints and are designated as 'active'. Periodic boundary conditions are also applied within the plane of the surface to mimic an infinite interface. Once the system is equilibrated at 300 K, the two films are compressed by moving the upper film towards lower film. At the desired degree of compression (order 1 GPa), the system is equilibrated and the films are slid against each other by moving the upper film parallel to the lower film.

Oriented transfer films of PTFE are created on a steel foil under reciprocated sliding. These transfer films are approximately 2 µm in thickness. The foils and transfer films are folded into a crossed-cylinder contact geometry oriented at 90° as shown in Figure 2. The normal load is 40 mN (the contact pressures are probably in the kPa range) and the sliding speed is 1 mm/s. The reciprocating path length is 0.6mm. Friction coefficient is continuously monitored and recorded using a low force flexure and computer data acquisition.

PRELIMINARY RESULTS AND DISCUSSION

To examine the effect of chain configuration on the frictional behavior of the PTFE films at the molecular level, three systems with different chain configurations are examined: (i) the chains in both films are perpendicular to the sliding direction (perpendicular configuration), (ii) the chains in both films are parallel to the sliding direction (parallel configuration), and (iii) the chains in one film are parallel to the sliding direction while they are perpendicular in the other film (mixed configuration). Additionally the effect of small oligomeric fluorocarbon molecules in the interface is examined.

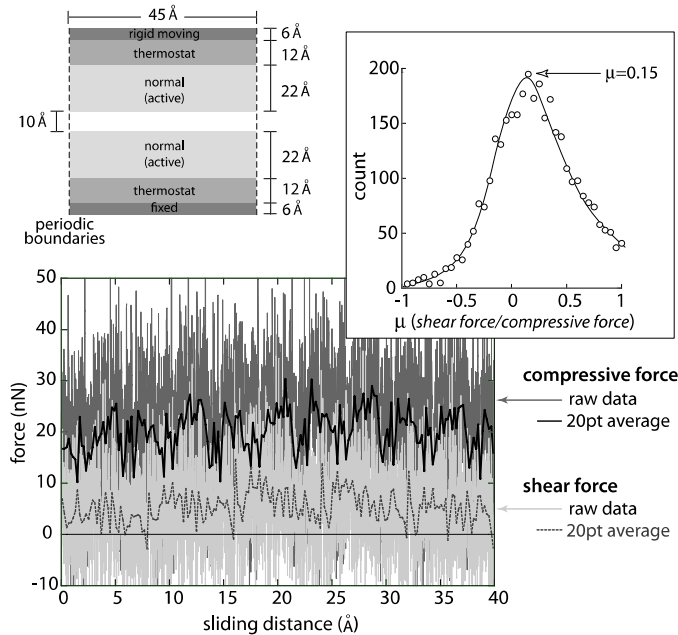


Figure 1 The experimental setup (upper left) and results. The histogram of friction coefficient (upper right) is calculated from the raw data. The sliding speed is 10 m/s. The transfer films are in the perpendicular configuration.

Figure 1 shows the compressive force and shear force during the sliding process in the perpendicular configuration. The rate of sliding of the upper film against the lower film is 10m/s. The most probable friction coefficient in this configuration is about $\mu=0.15$ based on the distribution of calculated friction coefficient from the raw data (ratios of the 20 point average shear and compressive forces give $\mu=0.27$). Figure 2 shows the friction coefficient recorded using microtribometry in the mixed configuration. The average friction coefficient was substantially lower ($\mu<0.04$) for the duration of the experiment. Low friction coefficients ($\mu<0.10$) are seen for PTFE under slow speed contacts in open air; friction coefficients under MPa of contact pressure and m/s sliding speed are typically closer to $\mu=0.2$.

The simulation results are obtained from a well-contacted nanoscale region, which makes it difficult to make a direct comparison to microscale and macroscale tribological measurements. The ideal morphological configurations studied here, and to be explored in future work, may be representative of nanoscale elemental domains in real systems (although GPa contact pressure are unlikely). It is nearly impossible to extract a fundamental understanding of the tribological interface from the microscale experimental measurements. Thus a

complementary approach that compares trends between the two systems is followed. The simulation studies can provide detailed insight into the tribological behavior of PTFE at the molecular level and help improve the design of nanocomposite materials that function through modification of these transfer film interfaces.

The dynamics of the transfer films during sliding are responsible for the frictional behavior. Different morphological configurations and the addition of small oligomeric molecules are both expected to show different deformation behavior and frictional coefficients.

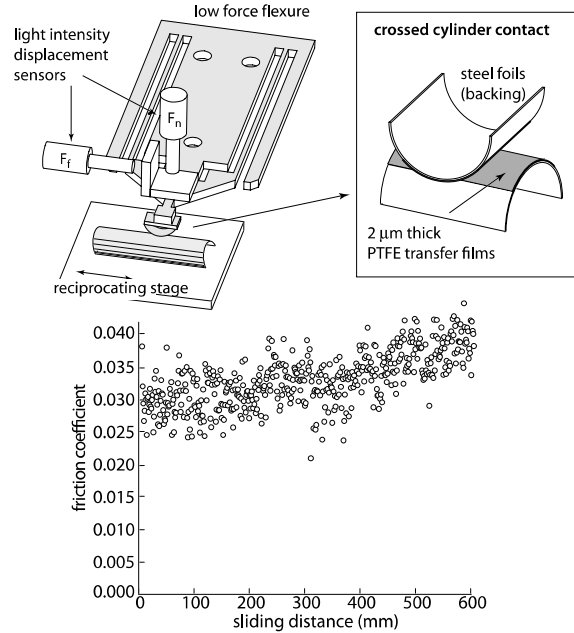


Figure 2 The experimental setup (upper left) and results from a crossed cylinder contact (mixed configuration). The normal load is 40 mN and the sliding speed is 1 mm/s.

CLOSURE

This work reports on preliminary efforts to model PTFE transfer films using classical molecular dynamics techniques and experimentally examine the microtribological behavior of self-mated PTFE transfer films.

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REFERENCES

1. H. R. Allcock, F. W. Lampe, and J. E. Mark, Contemporary polymer chemistry, 3rd ed. (Pearson/Prentice Hall, Upper Saddle River, N.J., 2003).
2. W. G. Sawyer, K. D. Freudenberg, P. Bhimaraj, and L. S. Schadler, *Wear* **254**, 573 (2003).
3. M. P. Allen and D. J. Tildesley, Computer simulation of liquids (Oxford University Press, New York, 1987).
4. I. Jang and S. B. Sinnott, *Journal of Physical Chemistry B* **108**, 18993 (2004).
5. D. W. Brenner, O. A. Shenderova, J. A. Harrison, S. J. Stuart, B. Ni, and S. B. Sinnott, *Journal of Physics-Condensed Matter* **14**, 783 (2002).