

Superlubricity of amorphous carbon surfaces via shear-induced aromatization

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Superlubricity of diamond-like carbon (DLC) is a well-known tribological phenomenon, but the underlying mechanism remains under debate. In this talk, we present recent results of atomistic simulations of DLC surfaces in sliding contact with organic friction modifiers (OFMs), and propose that in situ formation of aromatic nano tribolayers leads to superlubricity of DLC surfaces in boundary lubrication. Understanding the criteria for surface aromatization provides guidelines for controlling mechanochemistry of OFMs and in situ formation of superlubricious nano graphene tribolayers.

Keywords (from 3 to 5 max): diamond-like carbon, superlubricity, molecular dynamics

1. Introduction

Diamond-like carbon (DLC) coatings have been increasingly applied in engineering applications such as combustion engines, bearings and mechanical seals due to their excellent tribological properties. Fontaine et al. [1] reported that highly-hydrogenated DLC (a-C:H) exhibits a super-low friction of 0.003 even in ultrahigh vacuum (UHV). In contrast, non-hydrogenated tetrahedral amorphous carbon (ta-C) yields high friction ($\mu > 0.5$) in UHV. However, under boundary lubrication with organic friction modifiers, the tribological performance of ta-C is superior to that of a-C:H. We showed superlow friction ($\mu < 0.01$) of ta-C boundary-lubricated with glycerol or oleic acid [2].

The key mechanism behind superlow friction of carbon surfaces is surface passivation with hydrogen atoms or hydroxyl groups originating from either environmental species or lubricants. Meanwhile, a significant amount of sp^2 C forms via shear-induced sp^3 -to- sp^2 rehybridization (graphitization). However, the tribolayer is not crystal graphite and contains contaminants. There is no evidence that shows a clear relationship between friction reduction and surface graphitization. Thus, the low-friction mechanism of carbon surfaces remains under debate. In this study, we employ quantum molecular dynamics (QMD) simulations to reveal tribochemical reactions of lubricants with DLC surfaces and underlying low-friction mechanisms of carbon surfaces in boundary lubrication.

2. Results

We carry out QMD simulations of ta-C surfaces in sliding contact with glycerol by using a density-functional tight-binding (DFTB) method. In our QMD simulations, a normal pressure of 5 GPa is imposed to the outer rigid layer of the upper slab. The entire system is thermalized at 300 K. The glycerol molecule anchors to the lower surface after releasing one hydrogen atom from a hydroxyl group. After release of the remaining hydroxyl groups, cross-linking occurs (Fig. 1a) and leads to large shear stress $\sigma \sim 1$ GPa for the first sliding phase (Fig. 1b). Subsequently, glycerol completely decomposes and generates passivating species (hydrogen, hydroxyl, and ether groups). Nevertheless, the two surfaces cold-weld

after 0.1 ns (resulting in an increase in σ). Intermixing and rehybridization near the sliding interface promotes formation of dome-shaped aromatic structures (mainly consisting of planar 6-membered rings) that completely hinder chemical bonding across the interface (Fig. 1c), resulting in ultralow friction $\mu = 0.02$ during the last 0.5 ns of sliding. The result suggests that mechanical mixing of the contaminated sub-surface induces superlubricity of ta-C via surface aromatization (in agreement with our experiments) [2]. In this talk, we will present the detailed mechanism underlying surface aromatization and chemical structures of the nano graphene layer.

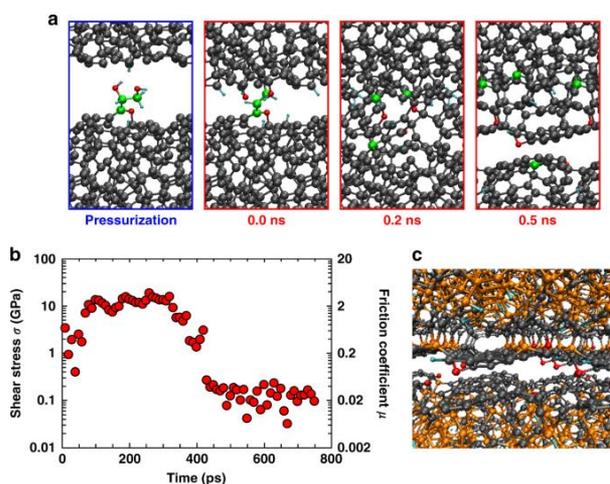


Figure 1: QMD simulation of ta-C surfaces lubricated with glycerol. (a) Snapshots of the sliding simulation. (b) Evolution of the shear stress σ and friction coefficient μ . (c) Shear-induced aromatization of ta-C surfaces.

3. References

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