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

Atomic-scale insights into the relationship between friction and surface passivation: the case of hydrogenated and fluorinated carbon

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A stable surface passivation is indispensable to the outstanding friction properties of diamond and diamond-like carbon. To understand how friction depends on the atomic-scale properties of the passivation species, we develop a quantum-mechanical-based interatomic force field for non-reactive interactions in dry hydrogenated and fluorinated carbon. Molecular dynamics simulations reveal a linear correlation between friction and corrugation of the contact potential energy. At odds with common assumptions, the energy corrugation is almost exclusively determined by the steric properties of the passivating atoms and not by their charge. We discuss the implications of these results for diamond, diamond-like carbon and PTFE.

Keywords (from 3 to 5 max): friction, chemical passivation, diamond, DLC, fluorine

1. Introduction

The outstanding friction properties of diamond and diamond-like carbon (DLC) coatings in boundary lubrication rely on a stable passivation of surface dangling bonds. However, the relationship between friction and the atomic-scale properties of the passivation species in still not clear. For instance, while hydrogen is the simplest passivation of a carbon dangling bond, fluorine can also be used as a monoatomic termination, providing an even higher chemical stability. However, whether and under which conditions a substitution of hydrogen with fluorine can be beneficial to friction is still an open question. Moreover, which of the chemical differences between H and F atoms and their bonds to carbon are responsible for the change in friction has not been unequivocally understood yet and is a controversial topic in the literature [1].

2. Results and discussion

To shed light on this problem, we develop a density-functional-theory-based, non-reactive force field that describes the relevant properties of hydrogen- and fluorine-terminated diamond and DLC tribological interfaces [1]. Molecular dynamics (MD) and nudged-elastic band (NEB) simulations reveal that the frictional stress at these interfaces correlates with the corrugation of the contact potential energy (Figure 1), thus ruling out a significant role of the mass of the terminating species on friction. Furthermore, the corrugation of the contact potential energy is almost exclusively determined by steric factors, while electrostatic interactions only play a minor role. This result is at odds with common assumptions made in the literature about the role played by electrostatic repulsion in the friction of fluorinated carbon surfaces. However, it is in agreement with the rationalization of polar hydrophobicity in fluorocarbons, which relies on the short-rangeness of the electrostatic field normal to the surface [2].

These results have potential implications for friction of technologically relevant carbon-based materials. In particular, friction between atomically flat diamond surfaces is controlled by the density of terminations, by the C−H and C−F bond lengths and by the H and F atomic radii. For DLC/DLC sliding interfaces, the intrinsic atomic-scale surface roughness plays an additional role. While surface fluorination decreases friction of incommensurate diamond contacts, it can negatively affect the friction performance of carbon surfaces that are disordered and not atomically flat. How these results apply to surfaces that are passivated by hydrocarbon and fluorocarbon molecules, as well as to PTFE, is under investigation and will be discussed at the end of the presentation.

This work provides a general framework to understand the impact of chemical structure of surfaces on friction and to generate design rules for optimally terminated low-friction systems.

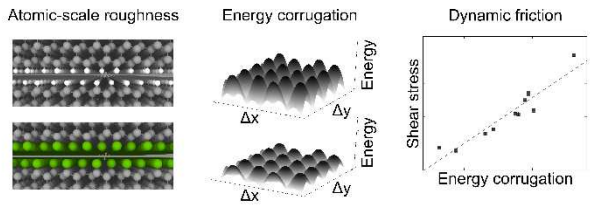

Figure 1: The atomic-scale roughness of H- and F-passivated diamond and DLC surfaces determines the corrugation of the contact potential energy surfaces and, in turn, the shear stress of the tribological interfaces.

3. References

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