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Nanoscale sliding friction phenomena at the interface of diamond-like carbon and tungsten

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Abstract

Macroscopic tribometry is linked to classical atomistic simulations in order to improve understanding of the nanoscale interfacial processes during sliding of hydrogenated DLC (a-C:H) against a metal (W) in dry and lubricated conditions. Experimentally, using an online tribometer, wear and roughness measurements are performed after each sliding cycle, which are then correlated with the frictional resistance. Ex situ analysis is also performed on the worn surfaces (i.e. plates and counterfaces) using X-ray photoelectron spectroscopy, Auger electron spectroscopy and cross-sectional transmission electron microscopy imaging of the near-surface region. Then, in order to elucidate the atomistic level processes that contribute to the observed microstructural evolution in the experiments, classical molecular dynamics are performed, employing a bond order potential for the tungsten–carbon–hydrogen system. Macroscopic tribometry shows that dry sliding of a-C:H against W results in higher frictional resistance and significantly more material transfer compared with lubricated conditions. Similarly, the molecular dynamic simulations exhibit higher average shear stresses and clear material transfer for dry conditions compared with simulations with hexadecane as a lubricant. In the lubricated simulations, the lower shear stress and the absence of a material transfer are attributed to hexadecane monolayers that are partially tethered to the a-C:H surface and significantly reduce adhesion and mechanical mixing between the sliding partners.

Keywords: Mechanical mixing; Third-body; Transferfilm; Diamond-like carbon; Hexadecane

1. Introduction

Nowadays, it is widely accepted that nanoscale phenomena (i.e. the third bodies) of sliding couples determine the friction and wear performance of a macroscopic system. Understanding these processes can lead to optimal tribological performance by developing novel materials and optimizing the sliding conditions (e.g. contact pressure, sliding velocity). However, studying third bodies can be quite challenging, owing to the confined nature of the sliding interfaces and the dimensions of third bodies. Many of the interfacial processes occurring throughout sliding can consist of dynamic events such as plowing, inter-film shearing and phase transformations [1], making it difficult to find any conclusive correlations between the interfacial processes and the friction and wear behavior. Therefore,

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in recent decades, researchers have focused on developing new techniques to monitor the buried interfaces during sliding [2,3] of metals, coatings and various solid lubricants (e.g. DLC, MoS_2, chameleon coatings).

Diamond-like carbon (DLC) is a good example of a coating where the friction and endurance life are governed by the interfacial processes [4–14] and thus has been intensively studied using the in situ methodology within the contact [12,15–18]. In addition to in situ observation of the contact, most of these studies have also used spectroscopy to identify the tribochemical changes that occur at the sliding interfaces with DLC [5,11,16]. While this methodology has provided essential insights into the friction and wear mechanisms of DLC, there still remains the uncertainty of whether these interfacial processes translate directly onto tribocouples without a transparent counterbody.

In addition to in situ macroscopic tribology on DLC, other work has also focused on the small-scale mechanical and chemical factors that influence tribological behavior, using nanotribological setups [12,19–21]. With an experimental setup using environmental transmission electron microscopy (TEM), M'ndange-Pfupfu et al. [19] simulated a single asperity contact of tungsten sliding against DLC in various environmental conditions (i.e. wet N_2 and H_2 gas). This approach is powerful owing to the possibility of providing important insights on single asperity contacts, which can help explain the microscopic mechanisms. However, bridging the gap between nano- and macrotribology is not always straightforward, specifically owing to the significant increase in normal load and contact area.

Recently, the present authors made an effort to combine various experimental techniques and classical atomistic simulations in order not to restrict the selection of the counterface materials to something transparent [22–24]. Molecular dynamics is a powerful tool for studying the mechanisms that underlie friction and wear at the nanoscale (see for instance Refs. [25–27]). In a previous study, focus was laid on third body formation during dry-sliding of a metal/ceramics tribocouple by linking atomistic simulations to macroscopic tribometry in order to provide a better understanding of interfacial processes for a WC/W pairing [23]. It was observed that the increase in the friction coefficient is attributed to mechanical mixing and amorphization within the WC. A subsequent study used the same approach and the same tribocouple in order to evaluate the topographical changes of dry and lubricated sliding contacts and to identify the different velocity accommodation modes that lead to fluctuations in the friction [24].

The goal of the present study is to provide an improved understanding of the nanoscale interfacial processes and behavior during sliding of a DLC coating against a metal, for dry and lubricated conditions. Although increasingly used in industrial application [28,29], a mechanistic understanding of friction and third body formation of such tribo pairs is still lacking. Owing to the availability of a suitable interatomic potential, a-C:H (a hydrogenated DLC) and tungsten are chosen as first body materials and hexadecane as lubricant for the combined experimental/theoretical investigation. A "real time" study of the topographical behavior is conducted using an on-line tribometer, while the structural and chemical changes within the "mixed material" are analyzed ex situ by means of X-ray photoelectron spectroscopy (XPS), Auger electron spectroscopy (AES) and TEM. In addition, molecular dynamics simulations of the sliding couples are performed (i.e. to observe the shear accommodation) and compared with the chemical changes near the surfaces. Overall, the results demonstrate an excellent correlation between the evolution of the roughness and the friction behavior throughout the sliding procedure. Molecular dynamics simulations also reflect the main observations found in the ex situ analysis and provide further insights into the microscopic processes.

2. Methods

2.1. Experimental procedure

Reciprocating sliding tests are performed using an "online" tribometer in order to monitor topographical changes. This tribometer contains a force sensor, digital holographic microscope (LyncéeTec SA DHM R2100, Switzerland) and an atomic force microscope (Bruker AXS Microanalysis GmbH, Germany). The force sensor consists of three SKL 1417-IR (Tetra GMBH, Germany) fiber optic sensors. A 20× objective lens is used with the holographic microscope in order to measure the wear and roughness after each cycle in the case of dry sliding, and a 50× immersion lens in the case of lubricated sliding. The experiments are performed in dry and lubricated conditions (i.e. hexadecane as lubricant) on a 99.9 wt.% tungsten plate. A cross-sectional TEM image of the W plate is shown in the Supplementary material. a-C:H coated WC–Co (94% WC and 6% Co, obtained from Spherotech GmbH) spheres are used as counterfaces. The a-C:H coating is deposited on WC spheres using the chemical vapor deposition (CVD) technique and is up to 1.4 μm thick. The hardness and the Young's modulus of the a-C:H is ~1800 HV and 130 GPa, respectively. A cross-sectional image produced by a FIB cut of the a-C:H coated WC–Co sphere is shown in the Supplementary material. The coating consists of three layers: a Si layer on the WC; an amorphous Si–C–H layer; and the a-C:H layer on the top. The Si–C–H layer contains ~40 at.% Si and the a-C:H layer on top contains between 25 and 30 at.% H.

The sliding experiments are performed with a sliding velocity of 5 mm s^{-1} and an initial normal load of 2 N. Wear and roughness measurements are performed after each cycle and correlated with the friction behavior, which was recorded at the position of the holographic microscope. More details on the experimental setup can be found elsewhere [23]. Ex situ analysis is performed on the worn surfaces (i.e. plates and counterfaces) using XPS (Axis Nova, Kratos Analytical), TEM (EM 400, by Philips), Raman spectroscopy (LabRAM, Horiba, Jobin Yvon)