

Achieving superlow friction with hydrogenated amorphous carbon: some key requirements

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Abstract

Among diamond-like carbon (DLC) coatings, hydrogenated amorphous carbon (a-C:H) films are of special interest, since some of them may exhibit coefficient of friction in the millirange, known as “superlow friction”. This paper will review some key requirements for achieving this specific friction regime.

At first, the formation of a carbonaceous transfer film on the steel counterface seems necessary for friction reduction. Oxide layers apparently slow down this build-up, as a-C:H surface reacts preferentially with metallic iron.

Secondly, the formation and evolution of the transfer film on the sliding counterface seems to control the frictional behavior. By performing tribological experiments under different controlled environment, it is possible to have some clues on the evolution of the transfer film and on its relation with the friction level observed. Phenomena of loss and restoration of superlow friction are for instance very helpful.

Finally, all hydrogenated amorphous carbon films do not lead to superlow friction under vacuum. The role of hydrogen content is known to be critical, but it appears to be strongly dependent on deposition process. Some mechanical properties of the films can also be correlated with friction reduction, like viscoplasticity. All these results suggest that surface rheological properties are of paramount importance in the achievement of superlow friction with a-C:H films.

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1. Introduction

Since the first deposition of diamond-like carbon (DLC) material as thin film by Aisenberg and Chabot [1] in the early 1970s, a wide range of deposition methods have been developed for DLC synthesis, especially during the eighties. Due to intense research and development efforts, some of these materials are now suitable for industrial applications [2] and they are widely commercially available since the 1990s.

The variety of deposition techniques and the large available range of process parameters are both leading to many compositions and structures. It is thus possible to control the physical and mechanical properties of a DLC film by an appropriate choice of the deposition technique and its parameters. Diamond-like carbon has then become a generic

term comprising many amorphous carbon forms as thin coatings obtained by chemical or physical vapor deposition (PVD) techniques. Recent reviews describe the various structures and properties of DLC coatings and their applications [3].

PVD processes using graphite or carbonaceous targets will lead to amorphous carbon a-C and at high sp³ content to tetrahedrally coordinated amorphous carbon ta-C, with very low hydrogen content (less than ~10 at.%). Plasma-assisted chemical vapor deposition (CVD) processes using gaseous precursors will lead to hydrogenated amorphous carbon (a-C:H) and tetrahedrally coordinated hydrogenated amorphous carbon (ta-C:H), with significant hydrogen content (more than ~15 at.%). These different compositions correspond to different structures and properties.

The present paper will focus on a peculiar tribological property of some DLC films: superlubricity. Relying both on previous studies and on new results, we will discuss the

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different phenomena involved in the achievement of super-low friction with hydrogenated amorphous carbon.

2. Background on friction reduction with DLC coatings

DLC films may exhibit many different tribological behaviors due to the great variety of their compositions and structures, as reported elsewhere [4,5]. Friction coefficients are reported between less than 0.01 up to 1, depending both on testing conditions and on composition. In ambient air, friction coefficients range generally between 0.05 and 0.3, whereas in inert environment (ultra-high vacuum [UHV], dry N₂ or dry argon), they can reach either ultra-low values (down to 0.01) or high values (more than 0.5). Furthermore, the composition of DLC films strongly affects their tribological dependence to environment. In ambient air, the friction coefficient increases with increasing relative humidity (RH) for hydrogenated amorphous carbon (a-C:H) [6,7], whereas it decreases with increasing RH for pure amorphous carbon (a-C or ta-C) [8]. The lowest friction coefficient values have been observed for hydrogenated DLC films, in inert environment. Friction coefficients lower than 0.01 and down to 0.001 [9,10] have indeed been reported. With values lower than 0.01, this vanishing of sliding resistance is often named “superlubricity” or “superlow friction”.

These very low friction coefficient values are not achievable with any DLC film: according to the literature, only hydrogenated amorphous carbon (a-C:H) allow such friction reduction under vacuum or inert environment. Nevertheless, not all hydrogenated amorphous carbon films lead to superlow friction. When slid in high vacuum, the friction coefficient of these films stabilizes either at very low or very high values, as reported by many authors. Miyake et al. [11] showed indeed that DLC with high hydrogen content (not quantified) leads to friction coefficients near 0.01 in vacuum, whereas films with low hydrogen content reach values higher than 0.35. Zaidi et al. [12] and Le Huu et al. [13] observed that hydrogen is responsible for an ultra-low friction level, and acts as a lubricating source when present in the near-surface region. In 1997, Donnet and Grill [14] have shown the existence of a threshold in hydrogen content of DLC films between high (>0.5) and low (<0.02) friction coefficients in UHV. This threshold lied between 34 and 40 at.% of hydrogen for films deposited by plasma-enhanced chemical vapor deposition (PECVD). These authors also showed that superlow friction is achievable by a suitable combination of hydrogen content and fraction of hydrogen bonded to carbon, both being controlled by deposition parameters.

The question then arises to explain the mechanisms involved in such a friction reduction.

At first, it is well established that a carbon-rich transfer film is build up on many metallic or ceramic counterfaces when friction and wear are low [4,5]. In such a case, friction

occurs between two surfaces coated with hydrogenated amorphous carbon, which might nevertheless be different from the original film. Several authors have reported the conversion of sp³ to sp² sites [15] and even “graphitization” [16,17] of the amorphous structure, as observed on the wear debris by electron energy loss spectroscopy (EELS) and also on transfer layer by Raman spectroscopy. Hydrogen may also be released from the film during friction [18]. All these friction-induced modifications of the amorphous network have a strong influence on the tribological behavior of the DLC material.

Secondly, the strong difference observed under ultra-high vacuum between high and low friction films could be explained by surface interactions between the carbon-covered counterfaces. For higher hydrogen content in the film, hydrogen atoms are covering the surface, leading to weak Van der Waals interactions between the sliding surfaces, whereas for lower hydrogen content there are not enough hydrogen atoms to shield the strong interactions between the π -orbitals of sp² carbon double bonds [19]. Moreover, we have shown in previous work that the high friction under ultra-high vacuum can be altered by the introduction of gaseous hydrogen, which enabled the achievement of super-low friction, probably by promoting tribochemical reactions between hydrogen and the a-C:H surface [20,21]. Thus, the lubricating role of hydrogen on a-C:H surface is now commonly accepted, with weak Van der Waals interactions accounting for the superlow friction values [10,22].

3. Experimental

3.1. Sample preparation

The experiments reported in this paper were performed on several sets of samples. A first set of samples, labeled “PECVD”, was deposited on Si (100) substrates by a d.c. plasma-enhanced chemical vapor deposition (d.c.-PECVD) process from acetylene (C₂H₂) or cyclohexane precursors (C₆H₁₂) at different bias voltages. Details on the process can be found in Ref. [14] and on these samples in Ref. [23]. Two of these coatings, labeled AC8 and AC5, obtained respectively at –800 V under 13 Pa of acetylene and –500 V under 26 Pa of acetylene, were more specifically studied. Their main characteristics are summarized in Table 1.

Table 1
General characteristics and properties of two model a-C:H samples

	AC8	AC5
Hydrogen content (at.%)	34%	40%
C sp ² :C sp ³ ratio	70:30	65:35
H bonded to C (NMR)	93%	98%
Hardness H_0 (GPa)	13	11
Viscoplastic exponent x	0.014	0.068
Stabilized coefficient of friction in ultra-high vacuum	0.53	0.003

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