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Elevated temperature tribological behavior of non-hydrogenated diamond-like carbon coatings against 319 aluminum alloy

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Abstract

Diamond-like carbon (DLC) coatings are promising candidates for dry machining of aluminum alloys since in ambient conditions aluminum has much less tendency to adhere to the DLC coating surfaces compared to other hard coatings such as TiN, TiAlN and CrN. In an attempt to better understand the tribological properties of the DLC coatings for cutting tool coating applications, non-hydrogenated DLC coatings were produced by magnetron sputtering and their elevated temperature friction and wear behavior were studied. DLC coated M2 tool steel discs were tested against 319 Al pins using a high-temperature tribometer. Counterface materials of tungsten carbide (WC) and sapphire (Al₂O₃) balls were also tested for comparison. Tests were done at 25, 120, 300 and 400 °C in air. The wear resistance of the DLC coating has been found to be poor at elevated temperatures. The mechanisms that provide the high wear resistance of the coating in ambient temperature cease to operate at temperatures as low as 120 °C. Annealing at elevated temperatures in air causes oxidation of the coating. Although much softer than WC and sapphire, the 319 Al alloy causes the most severe wear to the DLC coating. A two- and three-body abrasive wear model is proposed to explain this surprising observation.

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1. Introduction

DLC coatings have been attracting scientific and industrial attention because of their low coefficient of friction (COF) and wear rates. They have been used in a variety of areas like hard discs, bearings, seals, forming and cutting tools. In ambient conditions, aluminum does not tend to adhere to DLC surfaces [1]. This property of DLC makes it very promising in applications like dry drilling of aluminum alloys where the adhesion of aluminum chips to the drill surface is a major problem. In dry drilling of aluminum alloys, the chips that are formed become adhered to the surfaces of the steel or carbide drills and fill up drill

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flutes, eventually causing drill failure. In order to reduce the severity of the material transfer and adhesion, drill surfaces can be coated with a DLC coating.

Studies so far have shown that the tribological behavior of DLC coatings heavily depends on their bond structure, namely the ratio of sp² to sp³ bonding [2], hydrogen content [3–7], dopant content [8,9] as well as loading conditions and the test atmosphere [10–15]. Specifically, the water vapor in the test environment has been found to affect the friction and wear behavior significantly [16–22]. The DLC films with no hydrogen in their structure showed low COF and low wear rates in the presence of water vapor [14,15,21]. On the other hand, introduction of water vapor into the test chamber increased both COF and wear rate of hydrogenated DLC films [14,15].

Most tribological contacts generate frictional heat. In machining operations, much higher heat is released due to extensive plastic deformation during material removal (i.e.

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chip formation). Therefore, the temperature increase on the surface of the tool is particularly significant in machining processes. The tool coating should be able to retain its properties at elevated temperatures.

In the literature, there are several studies on the elevated temperature behavior of DLC coatings [23-28]. These studies indicate that DLC coatings do not perform well at high temperatures. Vanhulsel et al. [24] studied the wear behavior of plasma assisted chemical vapor deposition (PACVD) produced a-C:H coatings (containing 35% H) against corundum balls using a low-amplitude oscillatory test machine. They found that starting at 100 °C, the COF decreased from around 0.13 at room temperature to 0.07 at 300 °C and the wear scars became larger as the temperature was increased. Their room temperature test on a previously annealed sample (16 h at 300 °C) gave similar results to an unannealed sample. Therefore the authors concluded that the tribological behavior of a-C:H coatings would change only when they are exposed to load and heat simultaneously and structural changes (sp³-sp² change and/or dehydrogenation) are confined only to the top surface layer.

Krumpiegl et al. [25] tested three different DLC coatings (a-C, a-C:H and Ti-doped Ti-C:H) against M2 steel at elevated temperatures (up to 450 °C) and under vacuum (10^{-3} Pa) and observed high COF (0.6) and wear rates. The authors observed a drastic drop in hardness of the coatings after heating them to 450 °C in ambient air. This was thought to be related with the heavy oxidation of the coating.

Liu et al. [26] studied the high-temperature (up to 400 °C) tribological behavior of DLC films, which were grown using CH₄ gas in an rf-PACVD system against alumina balls in a reciprocating tester. The wear rate increased more than 10³ times when the temperature was increased from 200 °C to 300 °C, and the films started to peel off from the Si substrate above 300 °C.

Using thermogravimetric and differential thermal analyses and Raman spectroscopy, Wang et al. [27] investigated the high-temperature behavior of DLC films produced by combined PVD and PACVD processes. The authors found that DLC films disintegrated at 350 °C due to graphitic transformation and heavy oxidation.

Bremond et al. [28] performed elevated temperature pinon-disc tests (up to 400 °C) on a-C:H (10% hydrogen estimated) coatings deposited on 100C6 (AISI 52100 equivalent) steel discs by PACVD technique. They concluded that the failure of the DLC coating was due to the combined effect of the oxidation of the coating and the softening of the substrate.

The effect of vacuum annealing (up to 590 °C) on the friction and wear behavior of DLC films produced by PACVD method at various temperatures and bias was studied by Grill et al. [23]. In humid air the authors did not find a difference in the COF values between the as-deposited films and the ones annealed at different temperatures. However, the DLC films deposited at higher temperatures and biases showed higher wear resistance after annealing.

Gupta et al. [29] studied the friction and wear behavior of diamond films at 25 and 500 °C in ambient air against alumina balls. The COF of the polished film increased from 0.11 at 25 °C to 0.28 at 500 °C. The authors suggested that the increase in friction might be due to either desorption of the hydrogen from the diamond surface, or formation of tribological layers on alumina surface and/or to the increased fracture of alumina.

There are a few concrete conclusions that can be drawn from the studies reviewed above:

- 1) DLC coatings with significant amount of sp² bonding can maintain low friction at elevated temperatures only if they include a significant amount of hydrogen in their structures
- 2) Increasing the test temperature increases the wear rate. However, the COF does not show a consistent trend.
- 3) There is a temperature limit above which the DLC coatings graphitize and oxidize heavily. This temperature depends on the deposition method, deposition parameters, structure and composition of the coating.

Most of the studies mentioned above were performed on the hydrogen containing DLC coatings produced from hydrocarbon precursors. The coatings investigated in this study were sputtered from graphite and chromium targets using argon as the working gas. This process resulted in non-hydrogenated DLC coatings since the hydrogen content of the films does not exceed a few atomic percent.

Some aspects of the tribological behavior of the non-hydrogenated DLC coating studied here were previously reported by Camino et al. [30], Yang et al. [31,32] and Zeng et al. [33]. The coatings showed low COF and wear rate in humid air but very high wear rate in dry nitrogen similar to the behavior of graphite. Raman spectra [30] showed that the coating had mostly sp² type (graphitic) bonding, but it was still possible to reach hardness values ranging from 1600 HV to 3300 HV by increasing bias voltage from 20 V to 90 V [31]. Yang et al. [32] showed that the coating is composed of small graphite-like domains (1–3 nm) cross-linked to each other causing this high hardness.

To our knowledge, there is no work in the open literature on the tribological behavior of non-hydrogenated DLC coatings against an aluminum alloy at elevated temperatures. Therefore in this study, non-hydrogenated DLC coatings produced by magnetron sputtering were tested at 25, 120, 300 and 400 °C to investigate their friction and wear behavior against 319 Al alloy.

2. Experimental

2.1. Production of DLC coatings

DLC coatings of $2.06\pm0.13~\mu m$ thickness were deposited on M2 tool steel discs (58–62 Rc) using a Teer UDP 550

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