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Influence of test methodology and probe geometry on nanoscale fatigue failure of diamond-like carbon film

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ABSTRACT

The aim of this paper is to investigate the mechanism of nanoscale fatigue using nano-impact and multiple-loading cycle nanoindentation tests, and compare it to previously reported findings of nanoscale fatigue using integrated stiffness and depth sensing approach. Two different film loading mechanisms, loading history and indenter shapes are compared to comprehend the influence of test methodology on the nanoscale fatigue failure mechanisms of a DLC film. An amorphous 100 nm thick DLC film was deposited on a 500 μm silicon substrate using sputtering of graphite target in pure argon atmosphere. Nano-impact and multiple-load cycle indentations were performed in the load range of 100 μN to 1000 μN and 0.1 mN to 100 mN, respectively. Both test types were conducted using conical and Berkovich indenters. Results indicate that for the case of a conical indenter, the combination of nano-impact and multiple-loading cycle nanoindentation tests provides information on the life and failure mechanism of the DLC film, which is comparable to the previously reported findings using the integrated stiffness and depth sensing approach. However, the comparison of results is sensitive to the applied load, loading mechanism, test-type and probe geometry. The loading mechanism and load history are therefore critical which also lead to two different definitions of film failure. The choice of exact test methodology, load and probe geometry should therefore be dictated by the in-service tribological conditions, and where necessary both test methodologies can be used to provide better insights of failure mechanism. Molecular dynamics (MD) simulations of the elastic response of nanoindentation are reported, which indicate that the elastic modulus of the film measured using MD simulation was higher than that experimentally measured. This difference is attributed to the factors related to the presence of material defects, crystal structure, residual stress, indenter geometry and loading/unloading rate differences between the MD and experimental results.

1. Introduction

Hydrogen free diamond-like-carbon (DLC) thin films are frequently used as protective coatings on magnetic and optical storage discs, solar panels, optical windows, medical implants and micro/nano-electromechanical (MEMS/NEMS) devices [1]. As these applications exhibit tribological aspects, there is a growing interest on advancing the understanding of the nanomechanical behaviour of these films. The mechanical properties of these films are strongly dependent on the scale of measurement [2]. This consideration has motivated research on the nanoindentation behaviour of a variety of work piece and tool material combinations [3].

Previous experimental investigations have attempted to understand nanoscale contact fatigue behaviour of engineering materials using various commercially available and bespoke instruments and indenter shapes (e.g. [4–16]). These investigations take advantage of the features in the real time force–displacement (P–h), displacement–time (h–t) or stiffness curves recorded during repeated loading/unloading of bulk materials and thin films. Test methodologies and data analysis techniques adapted in previous investigations can be categorised in three main areas. The first one is depth sensing as indicated by Beake et al. [5–9] where a sudden increase in contact depth vs. time or number of impacts indicates the failure of component or film. The second one is area based calculations as indicated by Bouzakis et al. [10,11] where the fracture ratio of failed area and undamaged film indicates film failure. The third one is contact stiffness based evaluations as indicated by Bhushan and Li [4,13,17] where failure is defined as the change in contact stiffness of probe. A more recent development is the use of in-situ transmission electron microscopy (TEM) for nano-fatigue investigations by Wang et al. [15] where phase transformations in thin carbon films were investigated. A different approach was adapted by Liou et al. [18] for a 545 nm thick SiO_2 film on a Si wafer, where oscillating loads were used to evaluate the work required to delaminate the film. Other studies relating to TiN and AlN films [16,19] also considered the mechanism of thick film degradation (micrometre thickness range)

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under cyclic indentation loading, whereas Yonezu et al. [20] considered similar evaluation via incorporating acoustic emission investigation.

However, the literature lacks back-to-back comparison of coatings tested using different loading mechanisms with a view to ascertain failure mechanism of DLC coatings. Recently, authors reported nanoscale fatigue evaluation of 100 nm thick DLC coatings using an integrated stiffness and depth sensing approach and concluded a five-stage failure mechanism of these films [21]. The aim of the current paper is to ascertain the scientific merit of testing the same coating, using a different loading mechanism and instrument-type, with a view to comprehend the influence of test methodology on the mechanisms of DLC coating failure. To provide a back-to-back analysis, same DLC film was used in the current investigation as was reported previously [21]. In the current investigation, a P–h and h–t approach was adapted to experimentally investigate the mechanisms of film failure using i) nano-impact (low-cycle fatigue) failure and ii) multiple-load cycle nanoindentation (very low-cycle fatigue) of the DLC film. Furthermore, some molecular dynamics (MD) results are presented to consider the elastic level response of the coating substrate system. Elastic modulus thus evaluated is compared with the experimental findings of modulus using the nanoindentation system.

2. Experimental work

2.1. Test specimen

The material used for the substrate was a commercially available four inch diameter and 500 μm thick silicon wafer with crystal orientation (100). An amorphous DLC film of 100 nm thickness was deposited on the silicon wafer in pure argon (Ar) atmosphere using sputtering of graphite target without intentional substrate heating. The substrate holder was rotated throughout the process in order to ensure uniform deposition of the film. The substrate to target distance was 100 mm and flow rate of argon gas was 15 sccm at a pressure of 5 mTorr. The base pressure of the chamber was maintained at 2×10^{-3} mTorr. The RF plasma power was 150 W. The deposition rate was kept at 12.5 nm/min and the deposition duration was adjusted to achieve a film thickness of 100 nm [22]. Raman scattering of the DLC film was performed through Raman spectroscopy (Renishaw System 3000) with He–Ne laser (wavelength of 514.5 nm). The wafer curvature before and after film deposition was measured using a profilometer and the residual stress (σ) was calculated from the change in the radius of curvature (R_1 and R_2) of the wafer's bi-layer structure using Stoney's equation [23]:

$$\sigma = \frac{E_s}{(1-\upsilon_s)}\frac{t_s^2}{t_f}\left[\frac{1}{R_2}-\frac{1}{R_1}\right] \quad (1)$$

where E_s is the Young's modulus of the Si-substrate (130 GPa), υ_s is the Poisson's ratio of the Si-substrate (0.28) and t_s and t_f are the thicknesses of the Si-substrate and the thin film respectively. As suggested previously, Stoney's equation was applied without any correction [23] since the ratio of $t_f/t_s \leq 0.1$.

2.2. Nanoindentation test

The evaluation of hardness (H) and elastic modulus (E_s) of the thin film requires careful assessment of the test parameters. This is due to the fact that measurements may sometime represent a combined property of the film and the substrate rather than the film alone. A general rule of penetration depth of 1/10th of the film thickness has been recommended by Haanappel et al. [24] to avoid plastic deformation of the film. In the current investigation, the nanoindentation tests were done in depth control mode of 10 nm, which corresponded to a penetration depth of 1/10th of the film thickness.

Nanoindentation hardness and modulus measurements were performed using a calibrated TriboIndenter® system (Triboscope, Hysitron Inc., Minneapolis, USA) with a standard Berkovich tip. The measurements were taken at room temperature (~20 °C) in depth control of 10 nm. The indentation procedures were programmed as three segments of trapezoidal shape. The first segment increased the load to a maximum value with a loading rate of 200 mN/s, following a 5-second holding segment at the maximum load. The third segment retrieved the indenter tip from the sample with an unloading rate of 200 mN/s. The P–h profiles were analysed using a standard method with the area function for the Berkovich indenter, whereas the modulus and hardness were analysed according to the Oliver and Pharr method [25]. For calculation, the elastic modulus (E_i) and Poisson's ratio (ν_i) of the diamond indenter were considered as 1140 GPa and 0.07 respectively, and the Poisson's ratio of the DLC thin film (ν_s) was considered as 0.22 [26].

2.3. Low cycle nano-fatigue tests

Nano-fatigue tests were conducted using a calibrated NanoTest™ system (Micro Materials Limited, UK). Fig. 1a shows the schematic of the loading mechanism for the NanoTest™ system. The loading mechanism comprises a pendulum which rotates around a pivot and loaded electromagnetically. The test sample is mounted vertically and the test probe displacement is measured with a parallel plate capacitor with sub-nm resolution. Further details of the working mechanism of the NanoTest™ system can be seen elsewhere [5–7]. The NanoTest™ system was also equipped with an optical microscope (OM) and an integrated atomic force microscope (AFM, Nanosurf® Nanite, SPM S50, Liestal, Switzerland) directly linked by an automated positioning system.

Two different types of low cycle nano-fatigue tests were conducted in this investigation on the 100 nm thick DLC film using the NanoTest ™ system. These were (i) nano-impact tests and (ii) multiple-loading cycle nanoindentation tests. Both of these test-types were conducted using the Berkovich indenter probe with a negative rake angle of 65.3°, and also a conical indenter with 60° apex angle and 10 μm tip radius. The former was used to promote stress concentration and drive fracture in the film, whereas the stress field using the conical indenter did not promote stress concentrations in the contact region due to probe geometry.

(i) Nano-impact tests: Nano-impact experiments were conducted using the pendulum impulse impact option of the NanoTest™ system. A solenoid connected to a time relay was used to generate a repetitive indenter impact on the sample surface. The indenter was accelerated from a distance of 10 μm from the surface to produce each impact. For Berkovich nanoindenter, the impact loads applied were 100 μN, 250 μN and 1000 μN, whereas, for conical nanoindenter, the impact loads applied were 100 μN, 250 μN, 500 μN and 1000 μN. Nano-impact tests constituted linear loading of the specimen to full load in 1 s, followed by an immediate release of full (100%) load in 1 s without holding the load at its peak. Each test was conducted for a total of 1000 fatigue cycles on the same position of the specimen surface. Five repeat tests were done for each load. The evolution of surface impact response was recorded in-situ by monitoring the changes in the position of the indenter (depth vs. time). The failure was defined as the sudden change in depth amplitude with time or number of impacts. After the impact testing, the residual impression was characterised using integrated AFM. Further details of the testing instrument and measurement procedures are comprehensively described elsewhere [5–10].

Recently, authors also reported the findings of nano-fatigue tests on these 100 nm thick DLC coatings using a different loading mechanism and loading history using a calibrated TriboIndenter® system equipped with a standard Berkovich indenter [21]. The results of this previous investigation are compared here with the current investigation. The loading mechanism of the TriboIndenter® system was different to the