THE EFFECT OF COATING PROPERTIES ON THE PERFORMANCE OF A-C:H AND TA-C FILMS

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ABSTRACT

DLC films cover a wide range of different carbon based coatings, starting from soft to extremely hard diamond-like carbon films. In this study two different types of DLC films have been studied in respect of their stress and strain characteristics and tribological performance. The coatings are hydrogenated amorphous carbon (a-C:H) coatings deposited by PECVD and tetrahedral amorphous carbon (ta-C) coating deposited by filtered arc technique. In order to evaluate the mechanical behaviour of the coatings under load, 3D FE modelling was carried out in combination with scratch testing. Also the tribological performance was evaluated with pin-on-disc tests using stepwise increasing normal load. The 3D FEM model was developed for calculating the stress and strain distributions of DLC coated systems and to evaluate how coating thickness and elastic properties affect the stress-strain state at crack initiation location. The simulation was compared to the findings to experimental observations in scratch test contact conditions, when the spherical diamond tip was moving with increased load on a coated surface. The coating performance was evaluated with scratch testing to detect the crack generation as well as the coating adhesion. When combining the simulated coating characteristics with empirical observation of coating fracture patterns the coating fracture performance and tolerance to cracking could be evaluated. A major effect of the coating elastic modulus on the stress and fracture behaviour of the coatings was observed. In the tribological testing the both coatings had a low friction performance. In the tribological testing with stepwise increasing load, the critical load for coating delamination was higher for the a-C:H coating, which is in accordance with the results of FE modelling of coating stress state.

Keywords: DLC, ta-C, a-C:H, tribology, modelling
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INTRODUCTION

Diamond-like carbon (DLC) coatings have attracted a great number of researchers since the middle of 1980´s mostly due to their excellent tribological properties [1, 2]. DLC coatings are also intensively used on many sectors of industry, such as computer, automotive and tooling industry [3, 4]. The DLC coatings can be fabricated with different PVD and CVD techniques and depending on the deposition method and the deposition parameters used the material properties of DLC coatings can vary greatly. The structure of DLC coatings is amorphous with a mixture of sp2 and sp3 bonded structures. The coatings can be divided into two major groups, namely the hydrogenated DLC coatings and hydrogen-free DLC coatings. The bond structure and the hydrogen content influence dramatically the properties of the coatings [5].

The amorphous hydrogenated carbon (a-C:H) coatings contain significant amounts of hydrogen and the bond structure is predominantly sp2 bonded. The tetrahedral amorphous carbon (ta-C) coatings on the other had are non-hydrogenated and typically highly sp3 bonded. Therefore the coatings have different basic properties and some differences also in tribological performance. In order to select the suitable coating type for a specific application and to understand their performance it is important to know the stress and strain characteristics of the coatings. In this study we have evaluated the coating performance of a-C:H and ta-C coatings by 3D FEM modeling as well as scratch testing and pin-on-disc testing.

EXPERIMENTAL METHODS

The two DLC coatings studied were the amorphous hydrogenated carbon (a-C:H) and the hydrogen-free tetrahedral amorphous carbon (ta-C). The coatings were deposited on AISI 52100 steel discs that were hardened to 60 HRC and polished to mirror-finish. Prior to coating deposition the discs were sputter cleaned in Argon gas. The ta-C coating was deposited with a T-shape filtered arc device [6]. The a-C:H coating was deposited with the plasma assisted chemical vapour deposition (PACVD) method. The Young´s modulus and the hardness values of the coatings are presented in Table 1. The a-C:H coatings was deposited in three different coatings thicknesses, namely 0.3, 0.6 and 1.0 µm, and the ta-C coating in two thicknesses 0.3 and 0.6 µm, as also shown in Table 1.

The scratch tests were carried out with a Micro-scratch tester by using Rockwell C diamond tip (radius 200 µm) with a loading rate of 5 N/mm and a scratching speed of 10 mm/min. The maximum load was 30 N. The friction force and penetration was measured during scratching and a microscopic examination was carried out after scratching. Two critical loads were determined for the coatings, LC1 presenting the first visible crack generation in the coating and LC2 presenting the first delamination of the coating.

Table 1. The Young´s modulus and hardness values of the two DLC coatings studies with the coatings thickness values used.

	E [GPa]	H [GPa]	Thickness [µm]
ta-C	352.6 ± 14.6	66.8 ± 3.2	0.3 0.6
a-C:H	212.4 ± 5.1	25.4 ± 0.7	0.3 0.6 1.0

For the tribological evaluation of the coatings the pin-on-disc tests were carried out. The alumina balls (diameter 10 mm) were used as counter parts sliding against the coated discs with and step-wise increasing load. The first load was 2 N which was increasing to 5 N and further to 10 N, 15 N and finally to 20 N after 200 m sliding distance on each step. The Herzian contact pressure was thus increased from about 0.6 GPa to 1.4 GPa. The total sliding distance was thus 1000 m. The sliding speed was 0.15 ms^{-1} and the tests were carried out in controlled laboratory atmosphere with the humidity of 50±5 %RH and temperature of 21±2C.

MODELLING THE COATED TRIBOCONTACT

A three dimensional finite element model was used for the calculation of the stresses and strains in the coating substrate system and for identifying the stress concentrations occurring at the spot of the first crack. The scratch test experiment was discretised using the inherent symmetry of the geometry and introducing a finite element mesh where mesh sizing is of the order of the smallest assessed coating thickness. A finite strain and sliding framework was utilized, the substrate behavior being considered elastic-plastic while the coatings as well as the contacting diamond tip were modeled using a linear-elastic constitutive law. Coating properties were extracted from nanoindentation results as shown in Table 1. Bilinear brick elements were used in constructing the mesh and analyses were carried out using the Abaqus general purpose finite element package. The volume of the finite element slit taken to describe the scratch test configuration was $12 \times 4 \times 1$ mm^3 (length, width, thickness). The finite element mesh including the diamond tip is presented in Fig.1, the direction of sliding being from left to right.

Three different coating thicknesses were modeled for both coatings, for ta-C 0.3, 0.6 and 2.0 µm thick coatings and for a-C:H 0.3, 0.6 and 1.0 µm ones. The purpose of these analyses was to evaluate how coating thickness and elastic properties affect the stress-strain state at crack initiation locations and compare the findings to experimental observations. The numerical results are presented from an identical point of sliding comparable to crack initiation critical load in order to permit direct comparison.

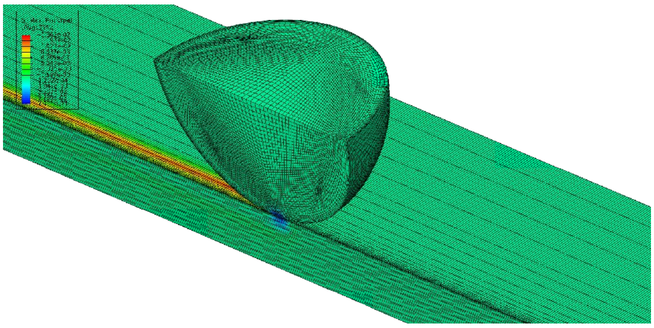


Figure 1. Finite element mesh of a 0.3 µm thick ta-C coating.

RESULTS

Scratch test results

The critical load values LC1 and LC2 determined for a-C:H and ta-C coatings are presented for the coatings in Fig.2. The ta-C coating provided higher LC1 values compared to a-C:H coating which means that cracks are generated in a-C:H coating with lower critical load. The cracks appeared as angular cracks on the scratch groove edge. The LC1 value increased for both coatings as the coating thickness increased from 0.3 mm to 0.6 µm. When the coating thickness increased further from 0.6 µm to 1 µm for the a-C:H coating, the LC1 value decreased. The LC2 value which is related to coating delamination and represents the adhesion characteristics of the coating, was higher for the a-C:H coating compared to ta-C. Also the LC2 values increased for the both coatings as the coating thickness increased from 0.3 µm to 0.6 µm and again decreased for the a-C:H coating as the coating thickness increased from 0.6 µm to 1 µm.

Modelling and simulation

The first principal stress distributions of various ta-C and a-C:H coatings are presented in Figs. 3-6. Results are provided for different coatings thicknesses for both studied coatings to highlight the differences arising from combinations of differing modulus of elasticity and coating thickness. The thin ta-C coatings in particular produce a stress-state which can be characterized to be of membrane character in a sense that the coating is highly and evenly stressed in comparison to the substrate in the present loading conditions. For thicker ta-C, the distribution differs in character and significantly higher stresses can be found at the scratch edge. The behavior of a-C:H differs from that of the ta-C films in the fact that the stress field gradient within the contact region and its trailing edge is less steep, and as such the differences in stress state between the plane of symmetry and scratch edge smaller.

(a)

(b)

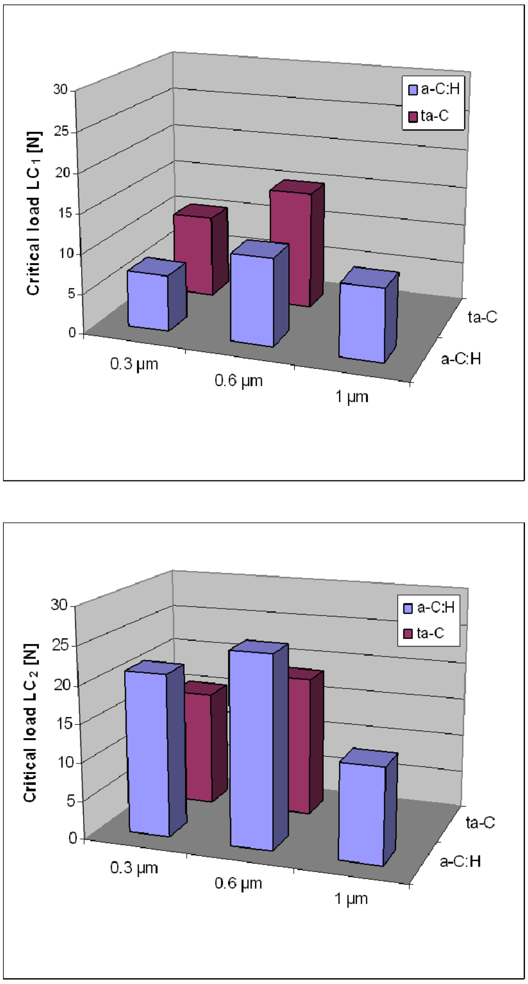


Figure 2. (a) The critical loads LC1 and (b) LC2 of the a-C:H and ta-C coatings determined by micro-scratch testing.

The coupling of material elasticity to coating thickness in yielding the system response can be identified with ease. The mismatch between the a-C:H films and the substrate is smaller, which results in an overall smoother response over the studied range of thicknesses. The increase in coating thickness is in this respect secondary to the elastic properties of the coating and its mismatch with the substrate material. The ta-C films, however, exhibit an overall higher stress-state with greater sensitivity to the studied parameters. This is further demonstrated in Fig. 7 and Fig. 8, where a measure of bending to membrane stresses is used to demonstrate how the stress field is affected by the coating thickness. For ta-C, the thinnest 0.3 µm coating for the specified loading conditions exhibits a stress state with a large gradient between the symmetry plane and the edge of the scratch, which when compared to the 0.6 µm coating indicates that for this specified loading condition a thicker coating behaves in a more suitable manner. The 0.6 µm coating has a stress-state somewhat smaller in magnitude and its local gradient is smaller. The 2.0 µm thick ta-C coating has a greater load bearing capacity, which results in a smaller state of tensile stress, but the thick coating combined to a large modulus of elasticity promotes an uneven distribution of stress, which can be seen as a large maximum value at the contact edge in Fig. 7. A larger value of stress or its gradient does not necessarily mean a weaker coating performance, but if the different film thicknesses were to have similar defects contributing to initiation of failure, such local values can result in crack initiation. As such, it can be stated that the utilization of coating properties for this combination of modulus of elasticity and thickness can be unfavorable. Similar results for a-C:H films in Fig. 8 demonstrate the significance of elasticity and mismatch to substrate in the development of the local stresses within the coatings. In terms of elasticity the a-C:H film is nearly evenly matched with the substrate, and this smoothens the stress distribution within the film as a function of coating thickness. The behavior differs significantly from the ta-C coatings and the sensitivity to coating thickness is far smaller. Due to smaller mismatching the stress state remains far more uniform, which can be taken as a measure of how effectively the coating material is utilized in the coating to substrate system. Since the sensitivity and local values are smaller in magnitude, such a coating can yield a somewhat more damage tolerant response with respect to crack initiation due to the more uniform distribution of stresses, but this does limit the load bearing abilities of the system and possibilities in tailoring and selecting an optimal coating thickness for a specific state of loading. The a-C:H film similarly results in a more lenient stress state in terms of loss of adhesion, but as a result is unable to protect the substrate as well as the ta-C due to lesser load bearing ability.

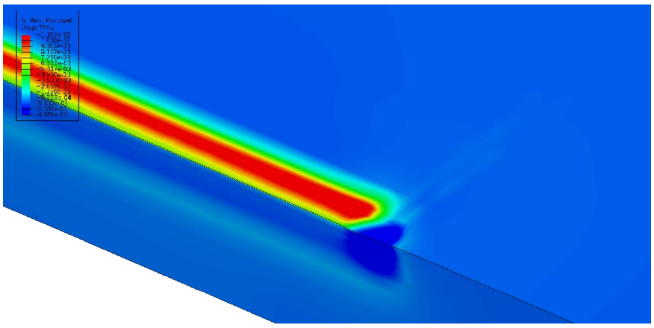


Figure 3. First principal stress distribution in a 0.3 µm thick ta-C coating.

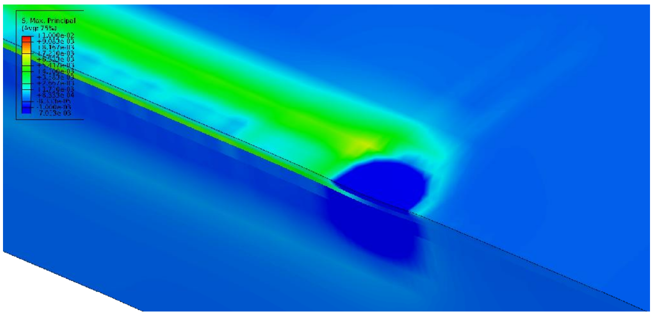


Figure 4. First principal stress distribution in a 2.0 µm thick ta-C coating.

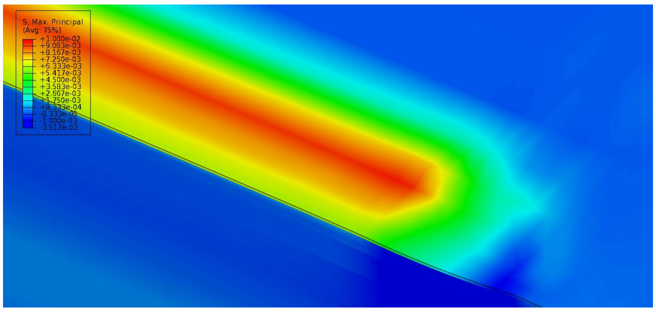


Figure 5. First principal stress distribution in a 0.3 µm thick a-C:H coating.

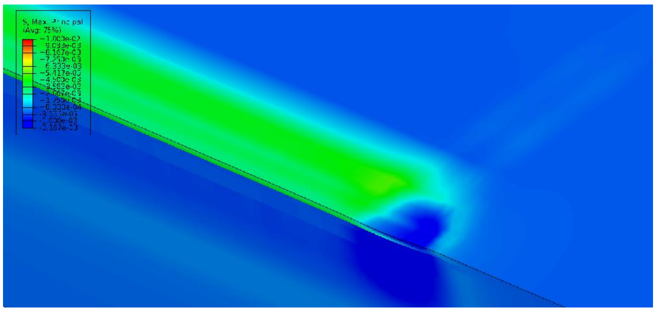


Figure 6. First principal stress distribution in a 1.0 µm thick a-C:H coating.

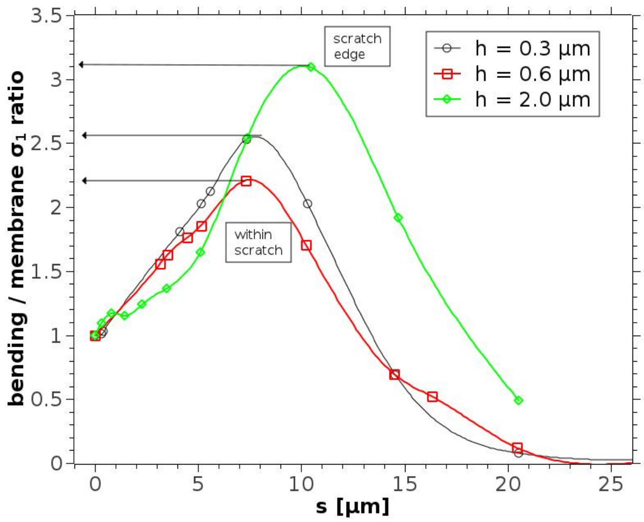


Figure 7. Characteristics of first principal stress field in computed ta-C coatings. "s" is the distance from plane of symmetry at the coating surface at the locale of maximum principal stress.

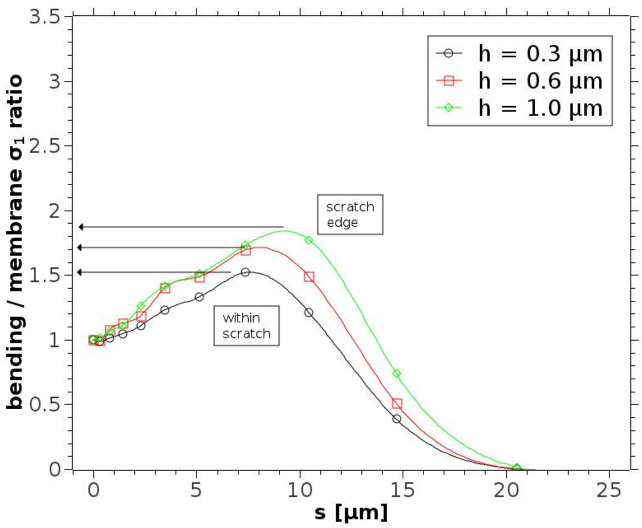


Figure 8. Characteristics of first principal stress field in computed a-C:H coatings. "s" is the distance from plane of symmetry at the coating surface at the locale of maximum principal stress.

Tribological testing

The coefficient of friction measured during pin-on-disc tests is presented in Fig. 9. For the a-C:H coating the friction coefficient decreased as the normal force was increased and the final values were in the range of 0.05 to 0.07 depending on the coatings thickness. The friction coefficient of 0.3 µm thick ta-C coating was about 0.2 in the beginning of test, but as the load was increased up to 15 N the coating was partly delaminated causing sudden increase in friction coefficient. However, after some sliding the friction started to reduce again approaching the level 0f 0.1 thus showing accommodating behavior of the tribocontact. The 0.6 µm thick ta-C coating had an unstable friction performance during the entire test and as the normal load was increased to 15 N, the friction coefficient increased and remained in the range of 0.17 to 0.2.

The wear surfaces were studied by optical microscopy and by SEM. In Fig. 10 are presented the SEM images of the wear track surfaces of different coatings and coating thicknesses. The wear surface of the 0.3 µm thin a-C:H coating was smooth after the test. On the other hand the thicker a-C:H coatings had some groove formations in the wear surfaces as presented in Fig. 10. Some crack generations were also observed in the thicker a-C:H coatings, particularly the 1 µm thick a-C:H coating had several crack on the wear surface around the groove formations. The 0.3 µm thin ta-C coating was partly delaminated and partly remaining intact in the substrate surface showing fragmented appearance on the wear surface. However, the 0.6 µm thick ta-C coating showed smooth appearance after the test. The counter surfaces were also studied by OM and typically some tribofilm formations were generated on the ball wear surface. The wear volume of the counter part was $1x10^{-5}$ mm^3 or less for the a-C:H films and $5x10^{-4}$ mm^3 for the ta-C coatings.

More detailed microscopic examinations were carried out of the selected coatings. The Fig. 11 presents the wear surface of the 0.3 µm thick ta-C coating showing the fragmented film on the substrate surface. The groove formation on the wear surface of the 1.0 µm thick a-C:H coating is presented in Fig. 12(a). A perpendicular crack formation was observed in the groove area of the coating which is presented in Fig. 12(b).

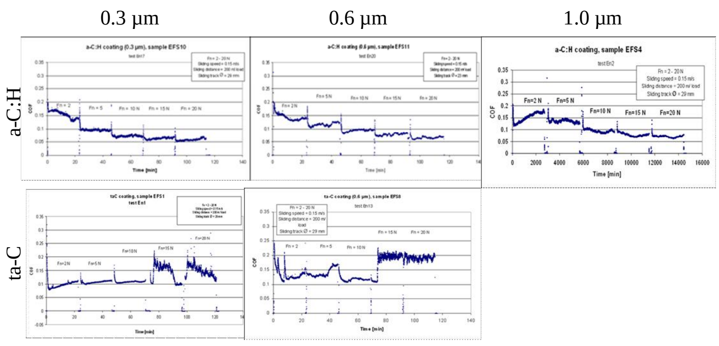


Figure 9. The coefficient of friction during pin-on-disc testing of a-C:H and ta-C coatings. The normal load was increased step-wise from 5 to 20 N after 200 m of sliding

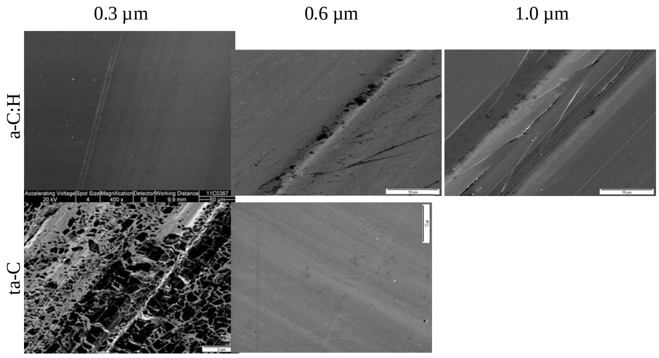


Figure 10. The wear surface of a-C:H and ta-C coatings after the pin-on-disc tests carried out with increasing normal load from 5 up to 20 N.

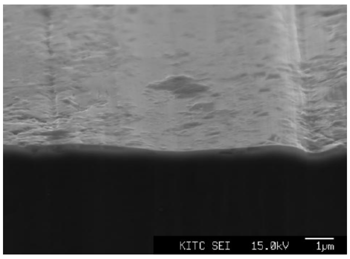
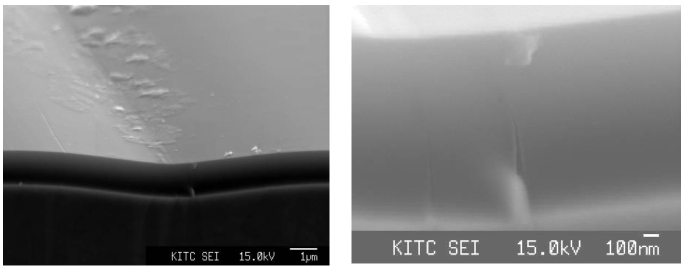


Figure 11. The wear surface of the 0.3 µm thick ta-C coating after the pin-on-disc test.



(a) (b)

Figure 12. (a) The wear surface of the 1.0 µm thick a-C:H coating and (b) a close-up picture of the coating cross section.

DISCUSSION

These two coatings are both DLC films, yet their properties differ from each other. The modelling results indicate that the stress-state of the thin ta-C coating had a membrane type behavior meaning that the coating is highly and evenly stressed. When studying the coating thickness effect by comparing the bending stresses on the scratch groove edge and the membrane stresses it was observed that the ta-C coating had an overall higher stress state compared to a-C:H. Comparing the modeling results of two coating thicknesses of ta-C, the thicker coating 0.6 µm seemed to have a more favourable behavior to 0.3 µm coating in the scratching contact conditions. This is actually in accordance with the LC1 critical load results, since the 0.6 µm thick ta-C coating had a higher value compared to 0.3 µm thick one. In scratch testing the contact is highly loaded and the coated system experiences a combination of elastic and plastic deformation. In pin-on-disc testing the contact is less concentrated and the contact pressure is lower. In this study the load in pin- on-disc testing was increased from 0.6 GPa up. Similar performance as described above was observed also in this case, since the 0.3 µm thick ta-C coating was partly delaminated when the normal load was 15 N representing the Herzian contact pressure of about 1.3 GPa. However, when the coating thickness was 0.6 µm, the coating could withstand the load without breaking through the whole load range up to 20 N normal load.

For a-C:H coating the stress field gradient within the contact region and in the trailing edge was less steep. The mismatch between the a-C:H coating and the substrate was smaller over the film thickness range studied which could be observed as low values in the bending to membrane stress stage graph. In tribological testing the a-C:H coating had a low friction performance with all coating thicknesses. Some groove formations were observed when testing the 0.6 µm and 1 µm thick films that were probably caused by the wear debris from the alumina ball. Related to groove formations also some crack formations could be found in the a-C:H coating, particularly for the 1 µm thick coating. In SEM studies cracks formed perpendicular through the coating thickness were observed. Despite the crack formations the coating was still intact with the substrate without delamination.

The modelling and experimental evaluation of the DLC coatings showed that similar features of coating performance can be found both in scratch testing and tribological evaluation. The stress and strain modelling of coated systems can give valuable information and increase the understanding of coated system performance. The modelling can emphasize the importance of coating property and coating thickness combination on the actual performance of coated system.

CONCLUSIONS

The ta-C coating experiences higher stress-state compared to a-C:H coating. It is also more prone to coating thickness effects. For the a-C:H coating the mismatch between the coating and the substrate was smaller over the film thickness range studied leading to lower stresses.

ACKNOWLEDGEMENT

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