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Design of low-friction PVD coating systems with enhanced running-in performance – carbon overcoats on TaC/aC coatings

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Abstract

The widespread use of low friction PVD coatings on machine elements is limited by the high costs associated with fulfilling the demands on the surface quality of both the supporting substrate and the counter surface. In this work, an attempt is made at lowering these demands, by adding a sacrificial carbon overcoat to a TaC/aC low friction coating. Both coatings were deposited by planar magnetron DC sputtering, as separate steps in a single PVD-process. Coatings were deposited on substrates of two different surface roughnesses, in order to test the ability of this coating system to function on rougher substrates. Reciprocating ball on disc tests were performed, using balls with two different surface roughnesses. The worn surfaces were investigated using 3-D profilometry and SEM. The ability of the different overcoats to initially reduce the roughness of both the coated surface and the counter surface and to produce stable, low-friction conditions was examined for the different initial roughnesses. The implications for design of efficient run-in coatings for various systems are discussed.

Keywords: Low friction coatings, surface roughness, running in, PVD DLC.

1. Introduction

Carbon based PVD coatings containing nanocrystalline metal carbides (often described as doped DLC coatings) are well-known for their good tribological properties in many applications [1], and are frequently applied to different machine elements, in order to reduce friction and increase wear life. Due to the high costs related to such coatings, their widespread use is however limited. A considerable part of these costs are related to the preparation of the surfaces on which the coatings are deposited, rather than to the coating deposition process itself. If the demands on surface finish could be reduced, low friction coatings would become a viable option for a much larger range of components.

Asperities on the substrate surface are typically inherited by the coating, and cause concentration of contact stresses during use, leading to an increased risk of coating failure, as well as to abrasive wear of the counter surface. If the coating allows for a gentle running in wear of both the coating and the counter surface during early stages of use, where the most protruding asperities are worn down, it is however possible to achieve good adhesion and low wear, even on rough substrates [2].

Addition of a softer coating on top of the coating could potentially be used to make sure that such a gentle run in behaviour is possible. Firstly, a softer overcoat would spread the loads at the asperity level, thus reducing the contact stresses [3, 4] and thereby the risk of coating spallation. Secondly, the overcoat could act as a sacrificial layer, where a larger amount of wear would be acceptable during the run in period. A softer and more ductile coating would be less prone to flaking, and thus able to withstand larger deformations, possibly allowing redistribution of coating material into voids in the surfaces of both the coated surface and the uncoated counter surface.

The initial stage of run in of this type of coatings typically involves formation of a relatively coarse grained tribofilm on both contacting surfaces [5]. For rougher surfaces, the amount of material

transferred during this stage is expected to increase. If the softer overcoat is active during this stage, the wear of the underlying coating is expected to decrease.

In this study, this type of coating system has been tested. As a model system, overcoats of pure carbon of varying thickness were added on top of a previously known low-friction coating, TaC/aC [6].

The topography of carbon based PVD coatings has been studied in several previous studies. The influence of deposition parameters on intrinsic coating topography, and the influence of this topography on the tribological properties have both been investigated [7, 8]. It is important to point out that the roughnesses studied in this work are several orders of magnitude larger than typical intrinsic coating topographies, and that no direct comparison with these studies can be made.

2. Experimental

2.1 Coating substrates

Powder metallurgical high speed steel discs (ASP 2030, hardness 9.5 GPa) were used as substrates. Substrates with two different surface finishes were prepared, hereafter referred to as *smooth* and *rough*. The two types of substrates will be identified by illustrations showing a flat or a pointed disc, respectively, in some of the figures. The smooth substrates were polished to a mirror finish (R_a < 10 nm), whereas the rough substrates were ground against SiC abrasive paper (#120 grit), resulting in an R_a of approximately 0.3 μm. All coatings were also deposited on silicon wafers, used for coating characterisation.

Prior to coating, the substrates were ultrasonically cleaned first in acetone (2*15 min), then alkaline detergent (2*15 min) and finally ethanol (15 min).

2.2 Coating deposition

The deposition was performed in a commercial PVD-coating system (Balzers, BAI640R), equipped with two planar magnetron sputtering sources and a thermionic arc, used for auxiliary ionisation of the chamber.

Prior to coating deposition, the substrates were pre-heated to approximately 450ºC for 45 minutes, after which they were argon etched for 15 minutes at a substrate bias of -200 V and a pressure of 0.15 Pa.

The TaC/aC coating was deposited using planar magnetron DC co-sputtering from a carbon target partially covered by tantalum foil, a procedure described elsewhere [9]. The deposition was performed for 120 minutes at a total chamber pressure of 0.4 Pa (achieved by an Ar flow rate of 150 sccm), a magnetron power of 1 kW and a thermionic arc current of 100 A. The substrate bias was set to 0 V (floating potential).

A pure carbon overcoat was deposited on top of the TaC/aC coating (without breaking the vacuum) by planar magnetron DC sputtering from a pure carbon target. The power on the magnetron was 500 W. All other process parameters were identical to those used for the TaC/aC deposition. The duration of the deposition was varied, with the purpose of producing carbon layers of 10, 100 and 1000 nm thickness. In order to achieve this, deposition times of 11, 109 and 1090 minutes were used, calculated from preliminary experiments.

2.3 Coating characterisation

Coating cross sections were studied using HR-SEM (ZEISS 1550), in order to determine their thickness and morphology. Compositional characterisation was performed using SEM/EDS (EDAX LEO 440) at an accelerating voltage of 10 kV. Due to the peak overlap of the TaM and the SiK peaks in the EDS spectra, the analysis was performed on a coating on a smooth steel substrate. Some signal from the underlying substrate was detected; the presented results are normalized values for coating elements and expected contaminants.

The hardness of the two coating layers was determined by nanoindentation (MTS Nano Indenter XP). The indents were performed using a Berkovich diamond indenter, and evaluated using the Oliver-Pharr method [10]. To avoid influence from the underlying material, the indents were made to a maximum depth of 30 nm (less than 10 % of the coating thickness). Measurements were performed on the coating without carbon overcoat and on that with the thickest carbon overcoat, both deposited on smooth steel substrates. The latter measurement is considered to represent the hardness of the carbon overcoat.

The topography of unworn coatings, without carbon overcoat and with the thickest carbon overcoat, was also studied by atomic force microscopy (AFM), using a PSI AXE-50 instrument, operated in non-contact mode. The measurements were performed on coatings deposited on silicon substrates, in order to retrieve information about the intrinsic coating topography.
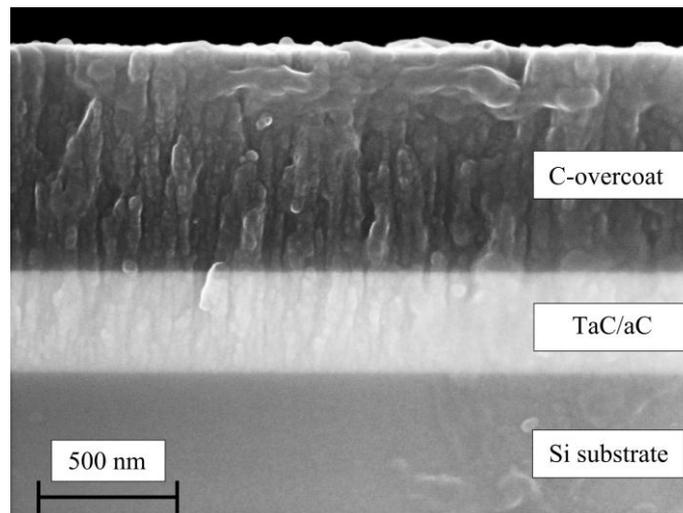

Figure 1. Fracture cross section of the coating with the thickest carbon overcoat (800 nm), deposited on silicon, imaged by HR-SEM. The two layers of the coating are clearly distinguishable.

2.4 Counter surfaces

Ball bearing steel balls (ø6 mm, 100Cr6 steel, hardness 9 GPa) were used as counter surfaces in the tribological tests. The balls were used both as purchased (polished to R_a 0.075 µm) and after intentional roughening. This was performed by placing 60 polished balls on a #120 grit SiC abrasive paper and loading the balls with a 4 kg circular steel plate which was then rotated 1800 revolutions, causing the balls to roll over the abrasive paper. After this treatment, the balls had an

R_a of approximately 0.58 µm. The two types of balls will hereafter be referred to as *smooth* and *rough*. In some of the figures, the type of ball used is indicated by an illustration showing a smooth or pointed ball, respectively.

2.5 Tribological testing

The coatings and counter surfaces were tested in unlubricated reciprocating ball-on-disc sliding in ambient air, under a normal load of 5 N. The stroke of the reciprocating motion was 10 mm and the oscillation frequency 5 Hz (corresponding to an average sliding speed of 0.1 m/s). The test duration was 10000 sliding cycles, but some tests were stopped earlier, after showing clear signs of coating failure.

The topography of the wear marks on both ball and coating were studied using white light interference profilometry (WYKO NT 1100). The wear tracks on the coatings and some of the wear scars on the balls were also studied using SEM/EDS.

3. Results

3.1 Coating properties

An SEM image of a fracture cross section of the coating with the thickest carbon overcoat, deposited on silicon, is shown in figure 1. A clear contrast is seen between the two coating layers, due to the large difference in mean atomic mass. Both layers have a columnar microstructure. The transition between the two layers does not visibly change the microstructure; the same grains continue growing across the boundary with no apparent change in structure.

The TaC/aC layer thickness was determined from the image as 350 nm, and the carbon overcoat 800 nm (compared to the intended 1000 nm). SEM images were also taken of the coatings with thinner carbon overcoats, but are not shown here. The thinnest carbon overcoat could not be properly measured in the images, but was instead estimated from the deposition rate determined for the thicker overcoats. The thicknesses of the different carbon overcoats were ~9 nm, 90 nm and 800 nm, respectively.

The AFM measurements performed on unworn coatings deposited on silicon substrates showed a slight increase in surface roughness with the addition of the carbon overcoat. The average roughness within the measured 1x1 µm area was 0.5 nm for the coating without overcoat, compared to 1.4 µm for the coating with the thickest carbon overcoat. Both coatings had roughness of similar character, consisting of round "bumps" with diameters around 50 nm without carbon overcoat and 100 nm for the coating with the thickest carbon overcoat. This is consistent with the columnar coating growth seen in the coating cross section (figure 1).

The composition of the TaC/aC layer, as determined by EDS, was 81 at% C, 16 at% Ta, 2 at% Ar and 1 at% O. The indentation hardness of the TaC/aC layer and the carbon overcoat was 11.2±3.4 GPa and 7.8±1.5 GPa, respectively.
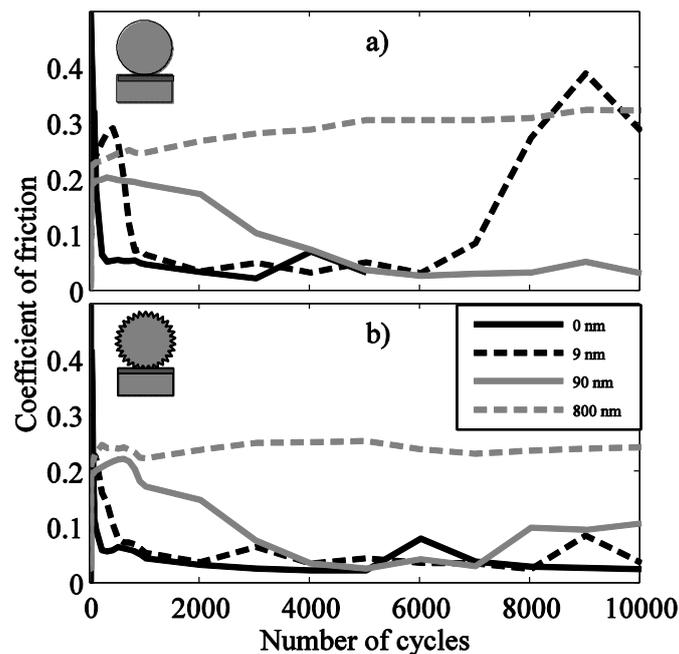

Figure 2. Coefficient of friction as a function of the number of sliding cycles for the coatings deposited on smooth substrates, running against smooth (a) and rough (b) balls.

3.2 Friction measurements

The results of the friction test are shown for the smooth substrates in figure 2 and for the rough substrates in figure 3. Each figure shows the results for both smooth (a) and rough balls (b) running against the two types of substrates. For each combination of ball and substrate, results for the four

different carbon overcoat thicknesses are shown. Some of the tests were ended prematurely, after showing obvious signs of coating failure.

For the smooth substrates, the initial coefficient of friction was relatively high, particularly for the coatings without carbon overcoat, where the friction during the first few cycles exceeded 0.5 for both types of balls (a fact difficult to distinguish in the figures). The friction decreased rapidly during the first hundreds of cycles, to a level of approximately 0.05, in some cases below 0.03. The length of this running-in phase increased with the carbon overcoat thickness. For a given overcoat thickness, it was shorter for the rough balls than for the smooth. These observations are valid for all coatings except those with the thickest carbon overcoat, where the friction remained high for the duration of the tests.
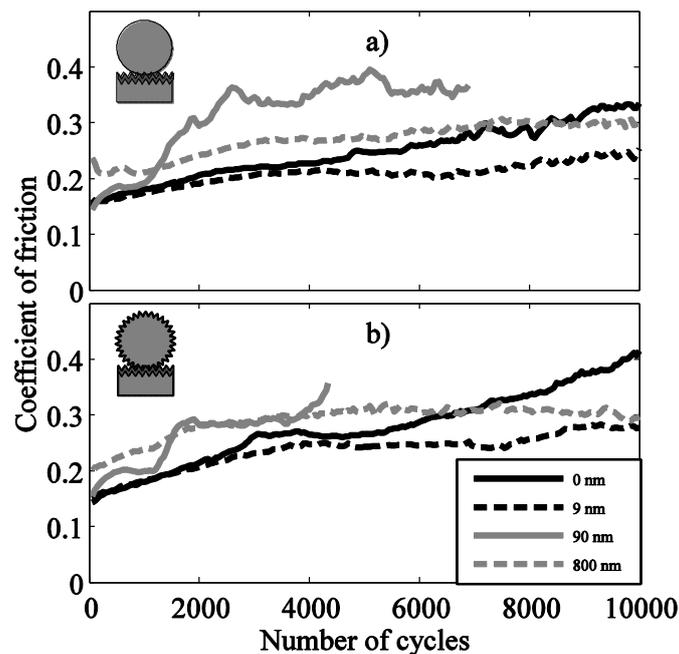

Figure 3. Coefficient of friction as a function of the number of sliding cycles for the coatings deposited on rough substrates, running against smooth (a) and rough (b) balls.

The friction behaviour of the coatings on rough substrates was entirely different. The initial friction was slightly lower than for the smooth substrates (around 0.15 for most of the tests), but no friction decrease was seen during the test. Instead, the friction slowly increased during the entire test, to a

final level around 0.3. The tests on the coating with 90 nm carbon overcoat were ended prematurely for both types of balls.

3.3 Wear scars on balls and coatings

The topographies of the balls after testing, measured by white light interference profilometry, are shown in figures 4 (for the smooth substrates) and 5 (for the rough substrates). Each line shows the profile of a ball, taken across the centre of the wear scar. The initial roughness difference between the smooth and rough balls is clearly seen in the unworn parts of the balls. At the top of each figure, a line corresponding to the shape of a perfect sphere is included as a reference.
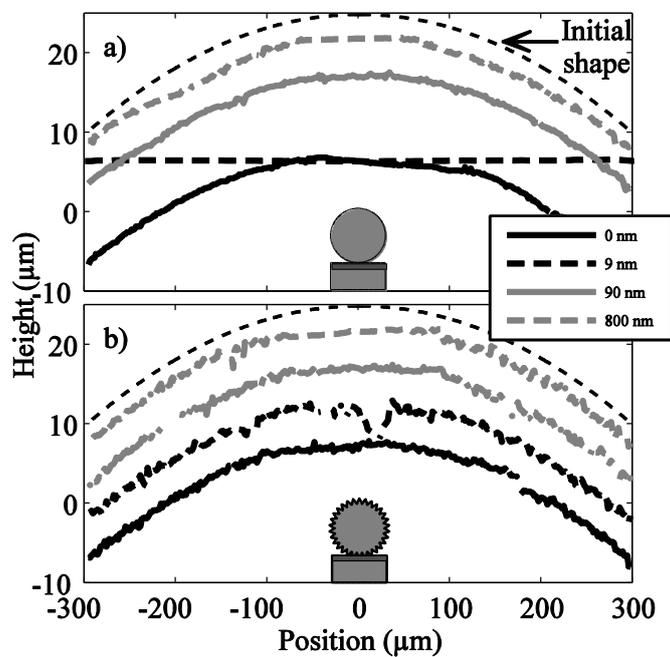

Figure 4. Wear scars on smooth (a) and rough (b) balls, tested against coatings on smooth substrates. The thin dashed line describes the shape of an unworn ball. Please note that the test with a smooth ball on a coating without carbon overcoat was ended prematurely.

The balls tested against smooth coating substrates show very little wear, with the only exception being the smooth ball tested against the 9 nm carbon overcoat. Here, the coating failed after approximately 6000 cycles, leading to a drastic increase in friction. The increase was however not large enough to cause a premature stop, so the test was continued for the planned duration.
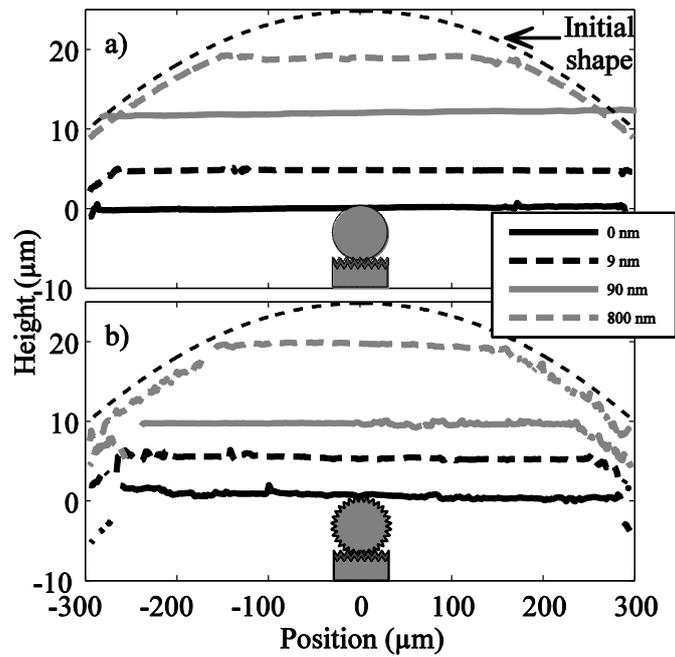
Figure 5. *Wear scars on smooth (a) and rough (b) balls, tested against coatings on rough substrates. The thin dashed line describes the shape of an unworn ball. Please note that both tests on the coating with 90 nm carbon overcoat were ended prematurely.*

Surface profiles for the wear tracks corresponding to the balls in figure 4 are shown in figures 6 (for the smooth balls) and 7 (for the rough balls). The profiles were measured across the wear track, at mid-stroke. The thickness of the two layers in each of the coatings is indicated in the surface profiles. Please note that this is not a measured distribution of the material from the two layers, but merely an illustration of the relationships between wear depths and coating thicknesses.

Again, please note that the test with a smooth ball on a coating without carbon overcoat deposited on a smooth substrate was ended prematurely (uppermost profile in figure 6). The test with a smooth ball on a smooth substrate with 9 nm carbon overcoat resulted in a much larger wear track than the other tests, confirming the findings from the balls.
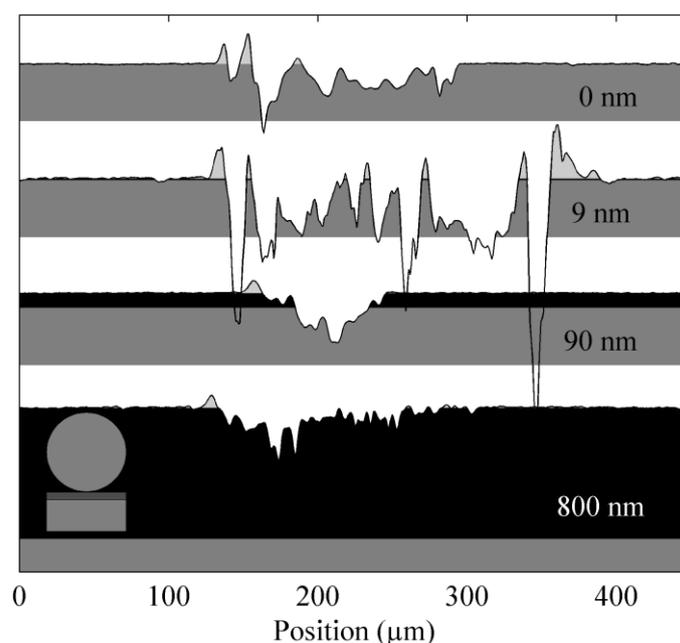
Figure 6. Surface profiles across wear scars on coatings on smooth substrate, tested against smooth balls. The profiles show the average depth in a 100 μm wide band at mid stroke. The thickness of the carbon overcoats and the TaC/aC layers are indicated with black and dark gray, respectively. Features protruding above the level of the unworn coating are indicated by light gray shading. Please note that the test on the coating without carbon overcoat was ended prematurely.

In general, the wear tracks are smaller for the thicker carbon overcoats. No clear difference is seen between wear tracks caused by the two different types of balls.

For the coatings on rough substrates, the depth of the wear tracks was small compared to the initial topography, making the wear tracks impossible to distinguish properly using the white light interference profilometer. As an alternative method for evaluation of coating wear, EDS analysis was performed on the wear tracks for the coatings with 800 nm carbon overcoat. EDS intensity maps of the TaM peak intensity were constructed for areas covering the entire width of the wear tracks, including regions of unworn coating. At the accelerating voltage used (10 kV), no TaM signal from the TaC/aC coating layer is expected, since the incoming electrons will not penetrate the carbon overcoat. As the carbon overcoat is worn, the TaM peak intensity is expected to increase, as more and more electrons reach the TaC/aC layer with high enough remaining energy to excite TaM photons.
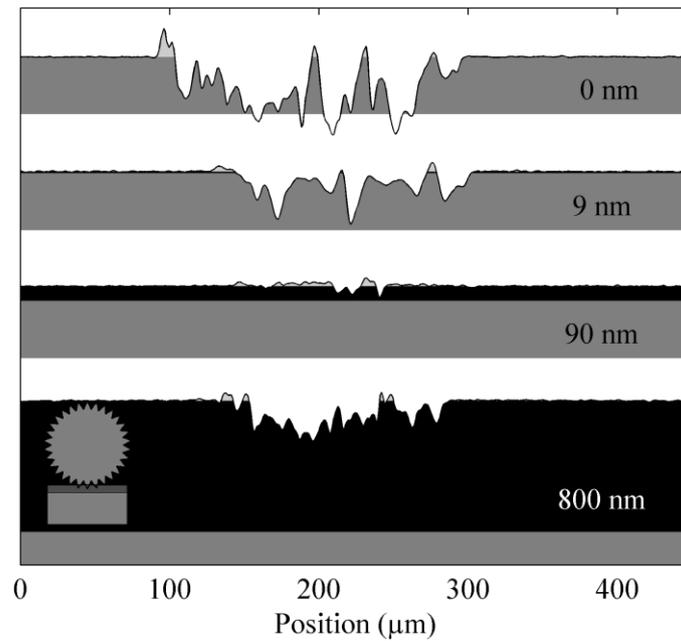
Figure 7. Surface profiles across wear scars on coatings on smooth substrate, tested against rough balls. The profiles show the average depth in a 100 μm wide band at mid stroke. The thickness of the carbon overcoats and the TaC/aC layers are indicated.

Figure 8 presents the variation of the EDS intensity across the wear tracks, along with SEM images of the corresponding surfaces (taken at a higher acceleration voltage, 15 kV). The lines were constructed by column-wise averaging of the intensity in the EDS maps and thus represent the average TaM intensities over the entire height of the images. To enable direct comparison between the different wear tracks, the EDS intensities are normalized to the same level in the unworn parts of the coatings.
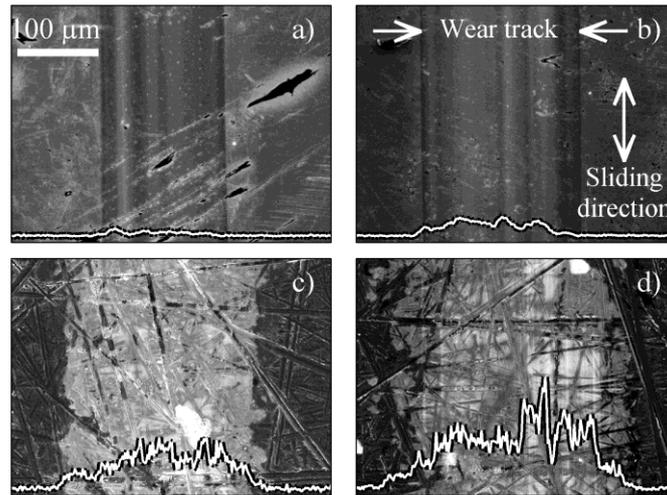
Figure 8. SEM images and EDX data for wear tracks on coatings with 800 nm carbon overcoat, on smooth (a, b) and rough substrates (c, d), tested against smooth (a, c) and rough balls (b, d). The line in each image shows the intensity of the TaM peak in the EDS spectra across the wear track.

The SEM images and the EDS data show a clear difference between the wear of the coatings on the two types of substrates and between the two types of balls. The coating on a smooth substrate tested against a smooth ball shows a small TaM intensity increase in the wear track (figure 8a). When a rough ball was used (figure 8b), the increase is considerably larger. For the rough substrates (figures 8c and d), the intensity increase is even more apparent, indicating more extensive wear of the carbon overcoat. Again, the test with a rough ball (figure 8d) shows a further increase in the intensity variation.
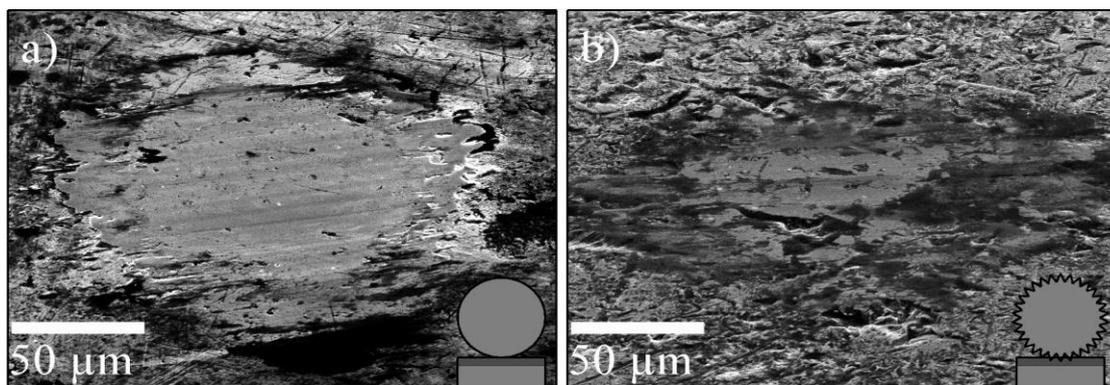

Figure 9. SEM images of wear scars on balls tested against the coating with 800 nm carbon overcoat, on smooth steel substrate. The images show a smooth (a) and a rough (b) ball, corresponding to the wear tracks shown in figure 8a and 8b. The topography of the ball and the substrate against which it was tested is indicated by the illustration in the lower right corner of each image.

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The balls tested against the 800 nm carbon overcoat on smooth substrate, were also studied in the SEM. The wear scars on these balls can be found in figure 9, where figure 9a shows the smooth ball (corresponding to the lowermost wear profile in figure 6 and the wear track in figure 8a) and figure 9b shows the rough ball (corresponding the lowermost wear profile in figure 7 and to the wear track in figure 8b). The images reveal rather similar and mild wear on both balls. Some residual roughness can be seen in the wear scar on the rough ball (figure 9b), in the form of small pits, similar to those seen outside of the wear scar. The protrusions seen in the unworn parts of the ball are however worn down, leaving a rather flat plateau, with some pits.
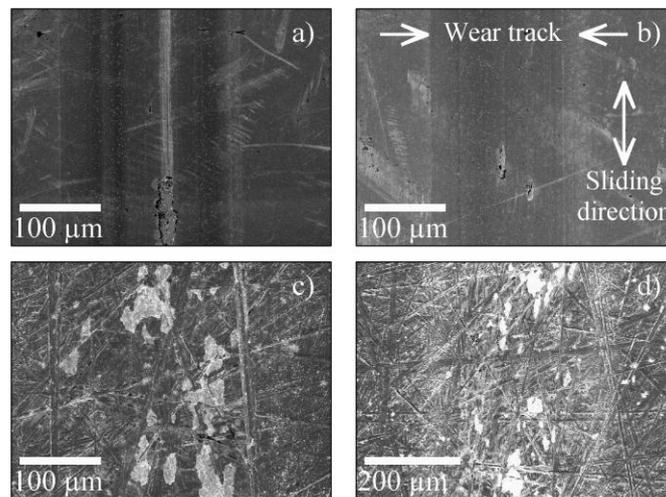

Figure 10. SEM images of wear tracks on coatings without carbon overcoat, on smooth (a, b) and rough substrates (c, d), tested against smooth (a, c) and rough balls (b, d). Please note that image d was taken at a lower magnification than the other images. The wear tracks on rough substrates (c,d) are from shorter tests (1000 cycles). Coating spallation is seen in the centres of image a and b, and as bright areas in c and d.

SEM images of wear tracks on the coatings without carbon overcoat are shown in figure 10. The images of coatings on rough substrate (figures 10c and d) show wear tracks from test that were ended already after 1000 cycles, and do not correspond to the friction data and ball wear scars presented earlier. As mentioned, the test with a smooth ball on a smooth substrate was ended prematurely.

All images in figure 10 show signs of coating failure; in some areas, the coating has spalled off, leaving the substrate surface exposed.

4. Discussion

One observation from the presented friction data for the smooth substrates is that the carbon overcoat is not, in itself, a low friction coating. It appears that the friction remains at a relatively high level until the carbon overcoat has been worn through. It is also clear that this high friction is not associated with high wear. As an example, the coating on a smooth substrate, with 800 nm carbon overcoat, tested against a rough ball, has an approximately sixfold coefficient of friction, compared to the same coating without a carbon overcoat (figure 2b). Despite this, the coating without carbon overcoat was worn approximately three times as much (figure 7).

Having said this, it is important to point out that a low wear rate of the carbon overcoat does not equal a good result. The primary purpose of the carbon overcoats used in this work is for them to be worn in a controlled way, thereby gently smoothing the counter surface and avoiding excessive wear of the underlying TaC/aC coating layer during the early stages of sliding wear. With this in mind, one should note that wear of the carbon overcoat itself is not necessarily to be considered as a problem.

A more important aspect is the wear of the counter surfaces, in this case the balls. In the tests with smooth substrates, differences regarding the wear of the balls are difficult to find, since the wear volumes are very small for almost all cases. For the rough substrates, however, significant differences can be seen. Here, the smallest wear volumes are found for the two balls (smooth and rough) tested against the thickest carbon overcoats, a strong indication that the overcoat may actually work as intended.

As already mentioned for the coating wear, one may point out that the high friction of the carbon overcoat does not lead to increased wear of the balls. One may also note that the roughness of the balls seems to have a rather limited impact on the wear of both the coating and on the ball itself. The ball wear scars shown in figure 9 (showing balls of different roughness tested against the same coating) reveal very little difference in wear.

Further experiments must be conducted in order to determine if the overcoat actually makes it possible to reach a low friction state even on rough substrates. This is a critical point, which must be established before this overcoat concept can be considered as a truly promising solution. The fact that the thickest carbon overcoats are not worn through after the tests, on neither the smooth nor the rough substrates, is encouraging for these future investigations.

5. Conclusions

- Addition of a carbon overcoat to the studied TaC/aC coating has a large beneficial impact on the wear of both coating and counter surface.

- Increased substrate surface roughness leads to increased counter surface wear.

- The roughness of the counter surface (ball) did not have any major impact on the wear of neither the coating nor the ball itself.

- The carbon overcoat increases friction during early stages of use, but reaches the same steady state friction level as a coating without overcoat when the overcoat is worn through.

- The effect on friction and wear in long-term tests has not been investigated, and needs further attention.

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