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Tribological studies of dual-coating (intermediate hard with top epoxy-graphene-base oil composite layers) on tool steel in dry and lubricated conditions

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ABSTRACT

The main objective of this investigation is to evaluate the tribological performance of dual-coatings of epoxy composites (epoxy with 10 wt% of graphene and 10 wt% of base oil) on different hard intermediate layers (diamond-like-carbon, tungsten carbide and titanium aluminium nitride) deposited on D2 steel. Friction and wear tests were conducted at 10 N and 15 N normal loads, and 0.63 m/s linear sliding speed and the counterface was a 4 mm steel ball. Among all dual-coatings, the combination of DLC intermediate layer and epoxy composite top layer furnished the least coefficient of friction of 0.09 and wear-life exceeded 2×10^5 cycles with no or negligible wear. In comparison, only hard or only soft coating had very limited wear-life.

1. Introduction

Soft tribological coating materials such as polymers can provide low friction but their load bearing capacity is poor, leading to plastic deformation, fracture and wear. Therefore, polymer coatings have limited applications in high load bearing capacity such as in gears and bearings. However, tribological significance of hard coatings is not very good due to higher frictional coefficient, poor bonding strength with substrate because of residual stresses and brittleness [1]. Hence, there is a requirement of low wear and low friction with high load bearing capacity tribological coatings. The load bearing capacity can be enhanced by applying hard film coating between the substrate and a soft coating, thus making a dual (hard and soft) composite coating. By using hard and soft dual-coating, friction is controlled by low shear strength of the top soft layer and the hard intermediate film provides high load bearing support. Many researchers have studied hard coatings followed by solid lubricant coatings [2,3]. Composite dual-coatings of hard (Al_2O_3 or TiO_2) and soft (polymeric such as polytetrafluoroethylene) as top layer coating on soft metals (such as Al and Mg alloy) as substrate were investigated by Gadow and Scherer [4], which provided low coefficient of friction and long wear-life. Minn and Sinha [5] applied composite dual-coatings of DLC as hard film topped with polymeric soft coating of ultra-high molecular weight polyethylene (UHMWPE) on silicon (Si) wafer substrate, providing very long wear life ($> 10^5$ cycles) with consistent low coefficient of friction (CoF) (0.13) in a pin-on-disk tribometer test. Wear-life was further enhanced by more than three times (3×10^5 cycles) and CoF reduced by half (0.06) by coating of perfluoropolyether (PFPE) as top overcoat. The PFPE overcoat provided high lubricity in the mixed lubrication regime [6].

Among many polymers [7,8], the thermoset epoxy exhibited the advantage of high strength thermal resistance and excellent adhesion to many substrates [9,10]. However, epoxy is tribologically very poor. It shows coefficient of friction as high as 0.5–0.7 against common metals and wears out very fast [11]. In our previous work, we have introduced graphene nanoparticles and *in-situ* liquid lubricant to epoxy which drastically improved the tribological properties [12]. The hard coatings that have found usages in tribology are diamond-like-carbon (DLC), tungsten carbide (WC), titanium aluminum nitride (TiAlN) etc. because of their high hardness and low friction (in some cases) [13–19]. Hence, in order to maximize the benefits of both coatings (hard and soft), this study focuses on the dual-coatings consisting of a hard coating topped with a soft coating of epoxy-graphene-base oil composite.

Epoxy/graphene/SN150 composite (hereafter referred as EGnSN) is a very promising coating to be used in relative sliding of components, which acts as a solid lubricant [12]. It has been found that EGnSN coating provides sustained low coefficient of friction (μ = 0.09). The mechanism of lubrication was found to be mixed lubrication due to the presence of *in-situ* lubricant. The load bearing capacity can be further enhanced by applying hard intermediate layer. The hard coatings selected for this study were DLC, WC and TiAlN.

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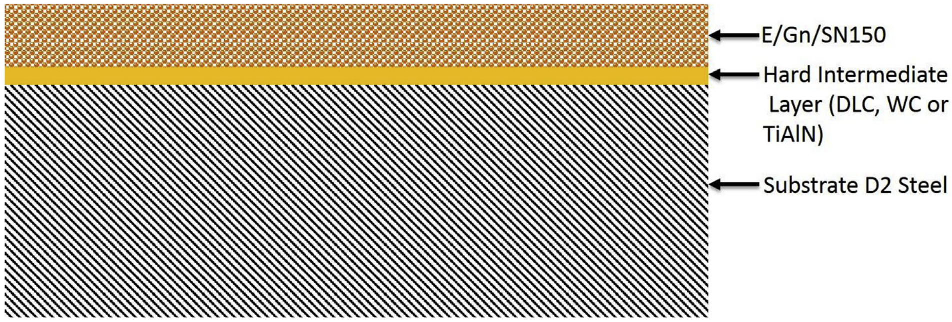

Fig. 1. Schematic of different layers coated on D2 steel substrate for making dual-coating.

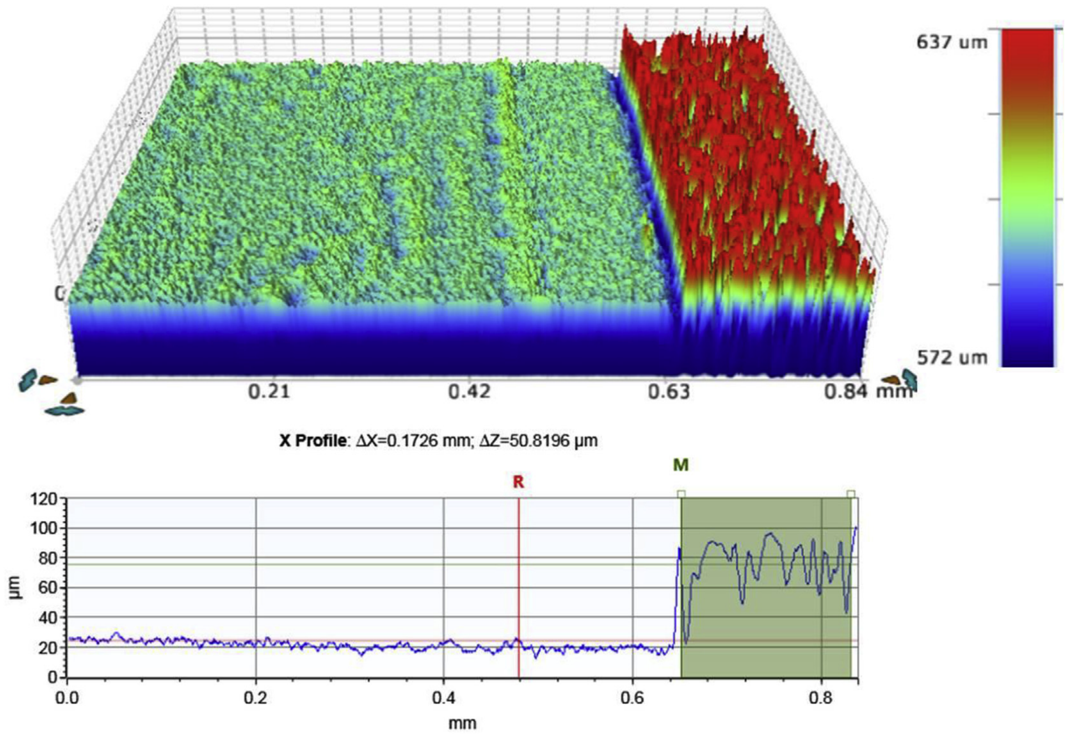

Fig. 2. 3D optical image to measure the coating thickness by step height method.

Table 1
Vickers hardness of hard coatings on D2 steel.

Coating Material	DLC	WC	TiAlN
Hardness (GPa)	11.50 ± 0.75	12.58 ± 0.59	11.86 ± 0.11
Surface Roughness (µm)	0.126 ± 0.029	0.205 ± 0.075	0.134 ± 0.032

Table 2
Vickers hardness of dual-coatings on D2 steel substrate.

Coating Materials	DLC/EGnSN	WC/EGnSN	TiAlN/EGnSN
Hardness (GPa)	0.198 ± 0.023	0.216 ± 0.007	0.205 ± 0.014
Surface Roughness (µm)	0.32 ± 0.015	0.40 ± 0.021	0.37 ± 0.019

The evaluation of the tribological performance of EGnSN coating (50–60 µm thickness) on D2 steel substrate with DLC, WC, or TiAlN film as an intermediate layer of thickness of 6–10 µm is presented. The hard and soft dual-coatings were applied on specimen (D2 steel shaft) and a steel ball (4 mm diameter) was used as the counterface. The test conditions were: Normal load = 10 N and 15 N; Linear sliding speed = 0.63 m/s; Ambient temperature = 25 ± 2 °C and relative humidity = 45 ± 5%. Experiments were carried out in dry sliding and base-oil lubricated sliding conditions.

2. Experimental procedure

2.1. Materials

D2 steel shaft (diameter 40) mm was hardened by heat treatment to a hardness of 2.75 ± 0.05 GPa. The specimen was then coated with different hard coating materials (DLC, WC or TiAlN) by the Physical Vapor Deposition (PVD) (Oerlikon Balzers Coating India Private Limited) process. After that, the soft coating of EGnSN (10 wt% graphene and 10 wt% liquid lubricant SN150 remaining epoxy and hardener) was applied as the top layer [12]. The hard layer coating thickness was 6–10 µm and at the top, soft layer coating thickness was 50–60 µm. Epoxy AY103 with hardener HY951 (Huntsman Advanced Material Pvt. Ltd) was used in the experiments. Graphene nano-particles (sizes < 100 nm) were purchased from Reinste Nano Ventures Pvt. Ltd., India. The base-oil SN150 (Group-I) was provided by Indian Oil Corporation Limited. The viscosity of oil measured was 24.25 cP (at 40 °C). Steel balls (SAE 52100 with hardness of 58 HRC) of 4 mm diameter and roughness 50 nm were employed as the counterface for all

Table 3
Water and oil contact angles of hard coatings and dual-coatings on the D2 steel substrate.

Coating Material	Water Contact Angle (degree)	Oil Contact Angle (degree)
DLC	87.5 ± 0.4	32.5 ± 0.1
WC	89.6 ± 0.7	29.0 ± 1.2
TiAlN	99.9 ± 0.8	20.6 ± 1.7
DLC/EGnSN	89.3 ± 1.1	31.6 ± 1.1
WC/EGnSN	84.6 ± 1.4	34.7 ± 1.8
TiAlN/EGnSN	86.8 ± 1.7	32.8 ± 1.5

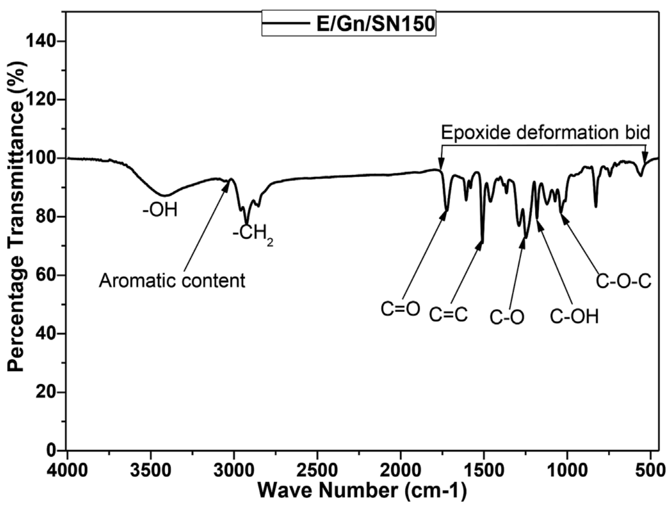

Fig. 3. FTIR of EGnSN composite representing the functional groups and bonding.

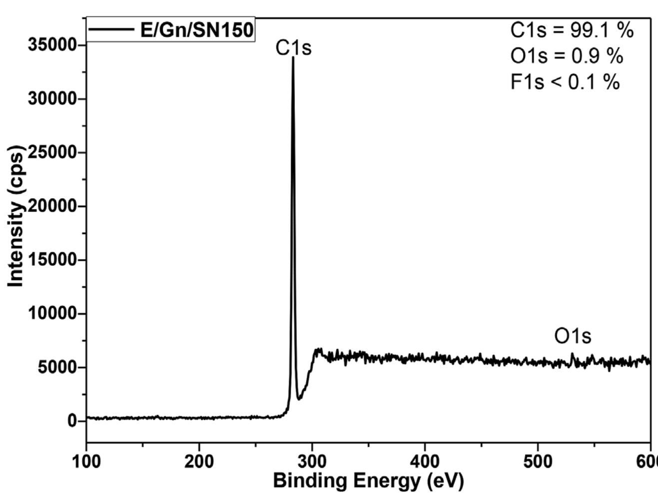

Fig. 4. XPS characterization of EGnSN composite showing the element and their bonding nature.

tribological tests.

2.2. Sample preparation

Before the application of epoxy composite coatings, the hard coating surface was cleaned with acetone, ethyl alcohol, and toluene and then air-dried followed by air plasma cleaning. The filler materials (graphene particles and liquid base oil SN150) were mixed into liquid monomer epoxy, mechanically stirred and homogenized by ultra-sonication. Subsequently, the hardener was mixed, and air bubbles in the mixture were eliminated by using vacuum. EGnSN composite was applied on three different types of samples having intermediate layer of DLC, WC, or TiAlN (Fig. 1). The samples were named as below (all with the same polymer composite coating):

(i) DLC intermediate layer (DLC/EGnSN)
(ii) WC intermediate layer (WC/EGnSN)
(iii) TiAlN intermediate layer (TiAlN/EGnSN)

The polymer composite coating for each specimen was applied on the hard coated steel surface with the help of a customized coating machine. The coated shaft was then rotated at 100 rpm for 4 h for uniformly distributing the composite over the shaft during air-curing [12]. The coating was cured in two stages; STEP 1: keeping at room temperature for 16 h, STEP 2: thermal curing at 80 °C for 4 h. Final thickness of coating was between 50-60 µm as measured using 3D-optical profiler by partially removing the coating from the surface to generate a step height (shown in Fig. 2).

2.3. Surface properties of the hard intermediate coating and dual-coating

Hardness of the hard coatings and the dual-coatings were measured using Vickers hardness tester. Square pyramid diamond tip (taper angle 136°) was used under the normal load of 0.2 N. Two diagonals of the indent was measured and averaged taking five such readings. The hardness value was then calculated using the following formula:

$$HV = \frac{2W\ \sin 68^\circ}{d^2} \tag{1}$$

$$HV = 1.854\frac{W}{d^2} \tag{2}$$

where, W = Normal force, d = Mean diagonal

$$d = \frac{d_1 + d_2}{2} \tag{3}$$

where d_1 and d_2 are diagonals of the indentation mark.

Hardness values of pure epoxy and epoxy-graphene composite (coated on the D2 steel substrate) were measured as 0.215 GPa and 0.235 GPa, respectively. This showed that there was ~9% increase in the hardness for a 10 wt% addition of graphene to epoxy. After adding 10 wt% of the based oil filler to the Epoxy-graphene composite, there is drastic reduction in hardness value which was 0.157 GPa. This is because the presence of *in-situ* liquid lubricant reduced the interfacial friction between the indenter tip and the material which made the indenter go deeper into the material for the same load without the lubricant. Hardness data for the hard coatings and the dual coatings are presented in Table 1 and Table 2, respectively. The observations from Table 1 show that the hardness of all hard coatings were in the range of 11.50–12.58 GPa, which was nearly four times that of D2 steel. WC coating exhibited the maximum value (12.58 GPa) followed by TiAlN (11.86 GPa) and DLC (11.5 GPa) coatings. Similarly, the hardness of epoxy composite (EGnSN) coated on different hard intermediate layers are shown in Table 2. By providing hard intermediate layer (WC), the hardness of EGnSN was enhanced by ~38% as compared to EGnSN coating on D2 steel substrate (hardness = 0.157 GPa) [12]. In general, increasing intermediate layer hardness leads to increase in hardness of epoxy composite. It clearly shows that there is substrate effect and the load bearing capacity of the dual-coatings has increased because of the intermediate hard-coating.

2.4. Surface roughness

Surface roughness of hard coatings (DLC, WC and TiAlN) and epoxy composite coating (EGnSN) on hard intermediate layers (dual coatings) on D2 steel shaft are shown in Tables 1 and 2, which were measured using surface profilometer (Mitutoyo; Surftest SJ-301). These were

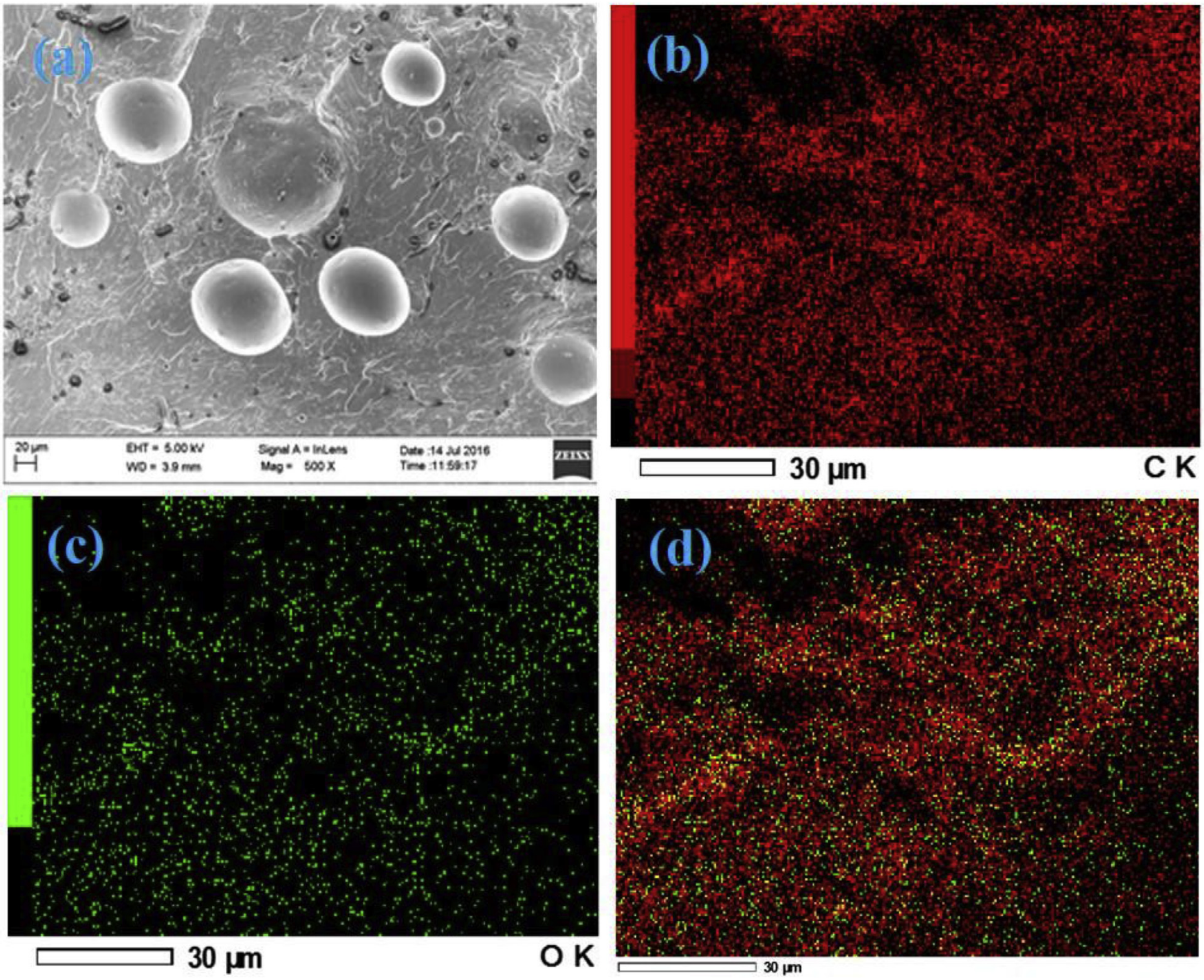

Fig. 5. FESEM and EDS images of cross-section of EGnSN. The average size of SN150 droplets is 50 µm, which were distributed uniformly (a). The images on the right side and below the cross-section show the elemental distribution of carbon and oxygen, which are (b) carbon, (c) oxygen (d) carbon and oxygen. The green colour in (d) shows the presence of oxygen, which is in the epoxy molecules. The red colour of the image shows the presence of carbon due to graphene and epoxy. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

averages of five measurements. Among the hard coatings, WC exhibited the highest surface roughness. Similarly, WC/EGnSN dual-coating exhibited high surface roughness.

2.5. Water and oil contact angle

Wettability of hard coating and dual-coating surfaces were measured for water and oil using contact angle apparatus (optical contact angle systems; OCA 35) and the results are presented in Table 3. Hard and dual-coatings exhibited water contact angles close to 90° hence they were neither hydrophilic nor hydrophobic in nature, except TiAlN coating, which had water contact angle of 99°, making it slightly hydrophobic. However, these coatings were oleophilic since oil contact angles were much lower than 90° (in the range of 30°). The oleophilic surfaces can form oil film easily in the lubricated sliding tests.

2.6. FTIR characterization

Fourier transforms infrared (FTIR) analysis (Thermo Scientific Nicolet) of the composite coating was conducted to understand the functional groups and chemical bonds. Fig. 3 shows the presence of OH group in the range of wave number $\sim$3416-3426 cm^{-1}, indicating moisture (H_2O) on the surface of epoxy and composites. There was epoxide deformation bid which contained a range of wave number $\sim$1576-1631 cm^{-1}, aromatic content at wave numbers 3000–3100, 1590, 1490, 830 cm^{-1} and -CH_2 at wave number 2927 cm^{-1}. Another range of wave number 1607-559 cm^{-1} indicated epoxide deformation bid which contained aromatics, C=C, C=O, O-C, C-OH and C-O-C bonds [20,21].

2.7. XPS characterization

Fig. 4 shows X-ray photoelectron spectroscopy (XPS) characterization of EGnSN sample with the atomic percentages. The atomic percentages are in the form of C1s and O1s scans of the composite. Binding energy spectra of C1s and O1s may contain the bonding of C-C, C-H, C=C, C-O and C=O, and O-H respectively.

2.8. Field Emission Scanning Electron Microscopy of the coating cross-sections

Field Emission Scanning Electron Microscopy (FESEM) (Karl Zeiss, FESEM EIGMA) of cross-section of epoxy composite coating was carried out. The sample was gold-coated using a sputter-coater before imaging. A typical FESEM image of the cross-section of the EGnSN coating is shown in Fig. 5 (a). The globular features were the SN150 oil droplets trapped inside the epoxy matrix during curing. The EDS of the cross-section of the composite was also performed using FESEM, which gives

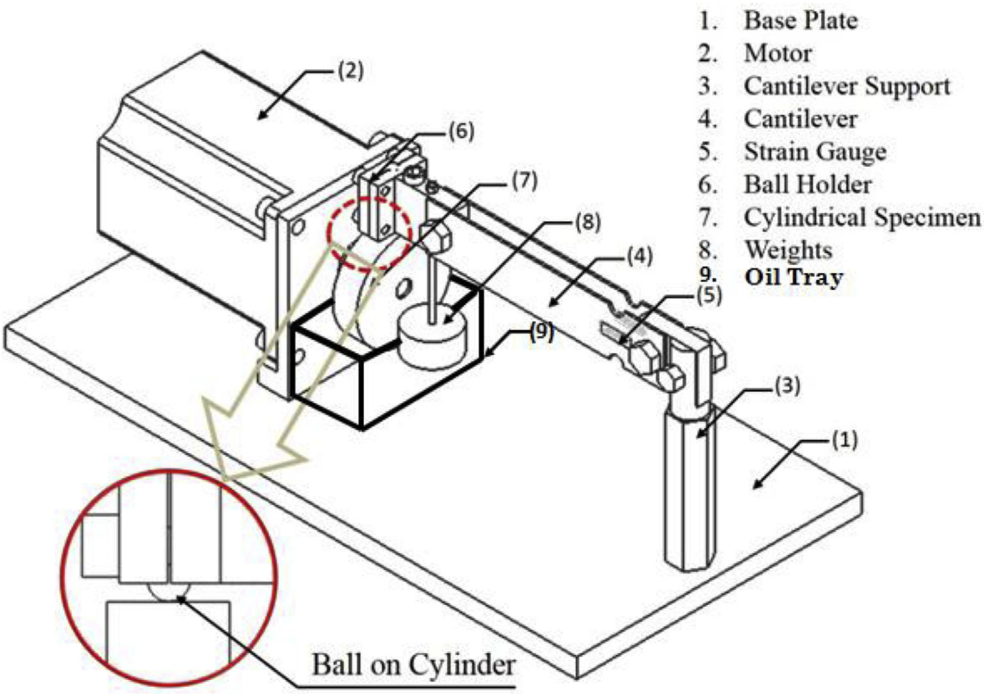

Fig. 6. Schematic of ball-on-cylinder tribometer.

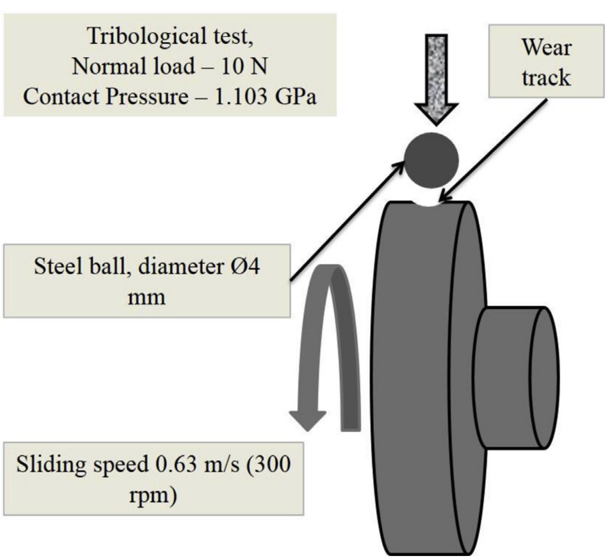

Fig. 7. Schematic of test condition and wear track.

the distribution of constituent atoms (Fig. 5 (b), (c) and (d)) in weight percentages of carbon and oxygen, which were 83.56% and 16.44%, respectively. Graphene distribution with SN150 was homogeneous inside epoxy matrix probably because of the oleophilic nature of graphene with SN150, which mixed well with epoxy.

2.9. Tribological tests

Friction and wear performances were evaluated on a customized tribometer (Ball-on-cylinder type) setup [22] whose schematic is shown in Fig. 6. The outer surface of the cylindrical specimen was coated with dual-coating, as explained earlier. Bearing steel (SAE 52100) ball of diameter 4 mm was employed as the counter surface with applied normal loads of 10 N and 15 N at 0.63 m/s linear sliding speed. The initial and steady-state coefficients of friction were recorded by a strain gauge-based load cell attached to the cantilever arm. The initial coefficient of friction (CoF) was calculated by averaging the values for the first 500 cycles of sliding, and the steady-state CoF was calculated by averaging last 1000 cycles before the test was stopped or the coating failed, whichever happened earlier. Room temperature was maintained at 25 ± 2 °C with relative humidity of 45 ± 5%. The volumetric wear rate was calculated using the data of the average track cross-section and the circumference of the wear track. Worn surface on the cylindrical specimen and the ball surfaces were visualized using an optical microscope. For the lubricated test, an oil tray was placed beneath the rotating cylinder such that a small part of the cylinder was always dipped in the base oil (Fig. 6). Oil got pulled up because of the viscosity of the oil which kept the interface fully supplied with the oil (see Fig. 7).

3. Results and discussion

3.1. Tribological investigations of the hard coatings

3.1.1. Friction and wear analyses (dry interface)

Dry tests of hard coatings at linear sliding speed of 0.63 m/s and normal load of 10 N were performed for 4×10^4 cycles on DLC and WC coatings, and for 1500 cycles for TiAlN coating since it wore-out the ball counterface very quickly. CoF versus number of cycles for the hard coatings against D2 steel ball are shown in Fig. 8 (a) and worn surfaces of the coatings and the corresponding counterface balls are shown in Fig. 8 (b). Enlarged view of initial coefficient of friction variations are shown in the inset in Fig. 8 (a) for 1500 cycles. The CoF of DLC coating initially increased and then decreased after few cycles as a tribo-film was formed at the interface, which acted as self-lubricating film. DLC hard coating showed lowest steady-state CoF of 0.14 due to formation of amorphous carbon hydroxide [15,16]. Similar trend for CoF was observed for WC coating, which exhibited steady-state CoF of 0.20. Carbon present in the transfer film lowered the CoF depending on the quality of transfer film. The maximum CoF of 0.5–0.6 was observed for TiAlN coating and there was formation of Al_2O_3 (aluminum oxide) tribo-film, which protected the coating, but led to wear of ball counterface [19]. There was negligible wear of the TiAlN coating. The

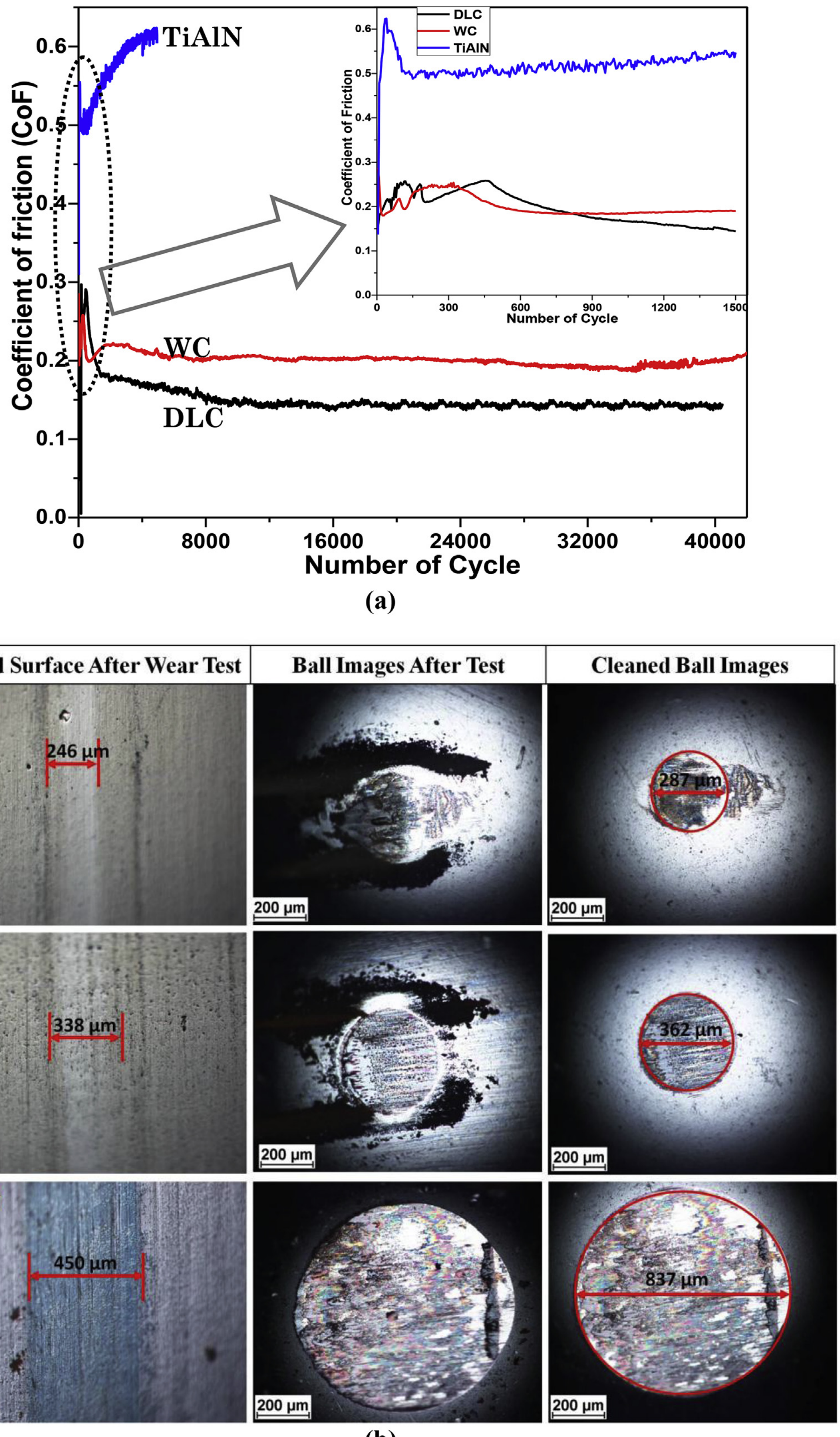

Fig. 8. (a) Variations in CoF with number of sliding cycles, (b) the worn surfaces of coatings and ball after 40,000 cycles with dry interfacial condition.

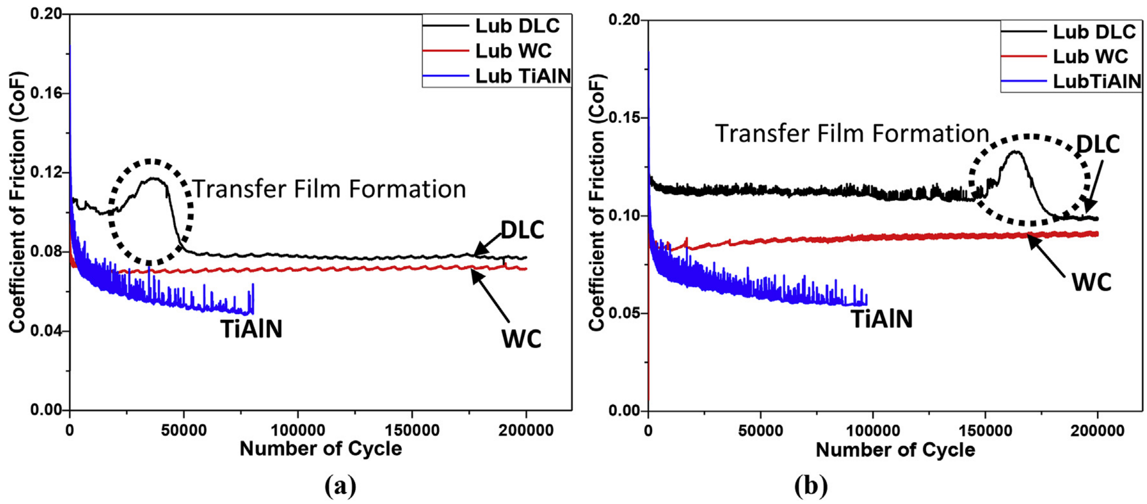

Fig. 9. Variations in CoF with number of cycles for (a) 10 N and (b) 15 N normal load under base oil lubricated condition.

transfer film or wear debris were visually observed to be of rusty colour due to transfer and oxidation of iron from the ball counterface. Coating surface wear track and ball counterface wear scars are shown in Fig. 8 (b). Wear of the steel ball occurred in all cases and the size of the wear scar was largest for TiAlN and smallest for DLC. Wear trend followed the friction trend closely. The coefficient of friction of ball against TiAlN coating was the highest, which caused maximum wear of the ball as explained earlier. In terms of hardness, these coatings exhibited comparable hardness. Friction, which is a result of the interface that is formed after initial few sliding, determines the wear process.

3.1.2. Friction and wear analyses (base oil (SN150) lubricated interface)

Experiments on hard coatings (DLC, WC and TiAlN) over D2 steel at linear sliding speed of 0.63 m/s and normal loads of 10 N and 15 N were performed for 2×10^5 cycles under base oil lubrication interfacial condition. The variations in CoF with the number of cycles are shown in Fig. 9 (a) and (b) at 10 N and 15 N normal loads, respectively. The CoF for DLC coating was initially slightly higher (0.10) which further increased to 0.12 (marked by dotted ovals in Fig. 9). As the running-in wear of DLC took place, a thin transfer film formed on the wear track, which reduced the CoF to 0.08 at normal load of 10 N. Similar trend was obtained at 15 N normal load as the CoF increased from 0.11 to 0.13 and then decreased to 0.10. The transfer film on DLC coatings were formed at different number of cycles at two loads of 10 N and 15 N as shown in Fig. 9 (a) and (b). The CoF for WC coating was found to be low, 0.075 and 0.09, at the normal loads of 10 N and 15 N, respectively, and the CoF remained nearly constant throughout these tests. The CoF for TiAlN coating was initially observed to be highest (0.19) which reduced to the lowest (0.06) amongst all coatings at 10 N and 15 N normal load. The optical images of worn surfaces of coatings and ball counterface just after sliding test and after cleaning are shown in Fig. 10 (a) and 10 (b). From the optical images, it could be observed that DLC coating surface exhibited maximum wear (deeper wear track), and least wear on the ball counterface. However, TiAlN coating surface exhibited the lowest wear of coating and highest wear of the ball counterface amongst all three coatings at both loads of 10 N and 15 N. Wide wear track in the case of TiAlN coating was because of widening of the wear scar on the ball surface which increased the contact area (with occasional scratches) but reduced the contact stress. Lower contact stress can lead to thicker fluid film according to the Stribeck curve which is the likely reason for the drastic drop in the CoF at higher sliding cycles for TiAlN coating. On the coating surface, it only showed the initial deformation and wear of the asperities but no gross wear to the coating.

3.2. Tribological investigations of dual-coatings

Tribological test of dual-coatings (hard intermediate and soft top layer) were performed under dry and lubricated conditions at ambient environmental conditions, at 0.63 m/s (300 rpm) linear sliding speed, and 10 N and 15 N normal loads, and the results are shown in Fig. 11 (a), (b) and 12 (a), (b), respectively. Fig. 11 (a) and (b) show the initial and steady-state CoF of EGnSN with DLC, WC and TiAlN intermediate layers under dry and lubricated interface conditions at 10 N normal load and 0.63 m/s linear sliding speed. Dual-coated D2 steel exhibited lower coefficient of friction compared to only hard coated surface because of self-lubricating properties of EGnSN. Shear stresses developed between these contacting surfaces were much lower than that for bare D2 steel. Initial CoF of all dual coatings was constant (0.10) in dry conditions, whereas under lubricated conditions, epoxy composite coating with intermediate layer of TiAlN demonstrated lowest CoF (0.07) followed by DLC (0.08). However, at 15 N normal load under dry and lubricated conditions, epoxy composite coating with DLC intermediate layer exhibited the lowest initial and steady-state CoF, which was constant throughout at ~0.07 (see Fig. 12 (a), (b)). The CoF further reduced under lubricated condition due to reduction in the shear stress (mixed lubrication).

The wear life of dual-coating was much higher than that of the hard coatings under dry as well as base oil lubricated condition at 10 N and 15 N normal loads and linear sliding speed of 0.63 m/s. As seen in Fig. 13 (a) and (b), all the dual-coatings had nearly stable CoF and the coatings did not fail at 2×10^5 cycles, up to which the tests were conducted. DLC as intermediate hard layer was the most effective coating since it provided stable and low CoF for dry and lubricated interfacial conditions. Interestingly, the dry interface of the dual-coating with DLC intermediate layer furnished CoF of 0.06, which was lower than that of the lubricated interface in the steady-state regime. At 10 N normal load and dry interface condition, there were no observable wear of DLC/EGnSN and WC/EGnSN dual-coatings or the ball counterface surface (seen after wiping the ball), but very little wear of TiAlN/EGnSN coating and corresponding ball counterface surface was observed, as seen in Fig. 14 (a). At higher load of 15 N (Fig. 14 (b)), there was wear to WC/EGnSN and TiAlN/EGnSN dual-coatings and the corresponding balls, however DLC/EGnSN dual-coating and the corresponding ball did not show any sign of wear mark. At 15 N normal load, the DLC/EGnSN dual-coating showed much lower CoF compared to other dual-coatings, both for dry and lubricated interfaces. Lower friction resulted in lower wear of coating as well as the ball counterface. It was found that wear was higher for the ball counterface, when liquid lubricant was used compared to dry interface. This could be because of

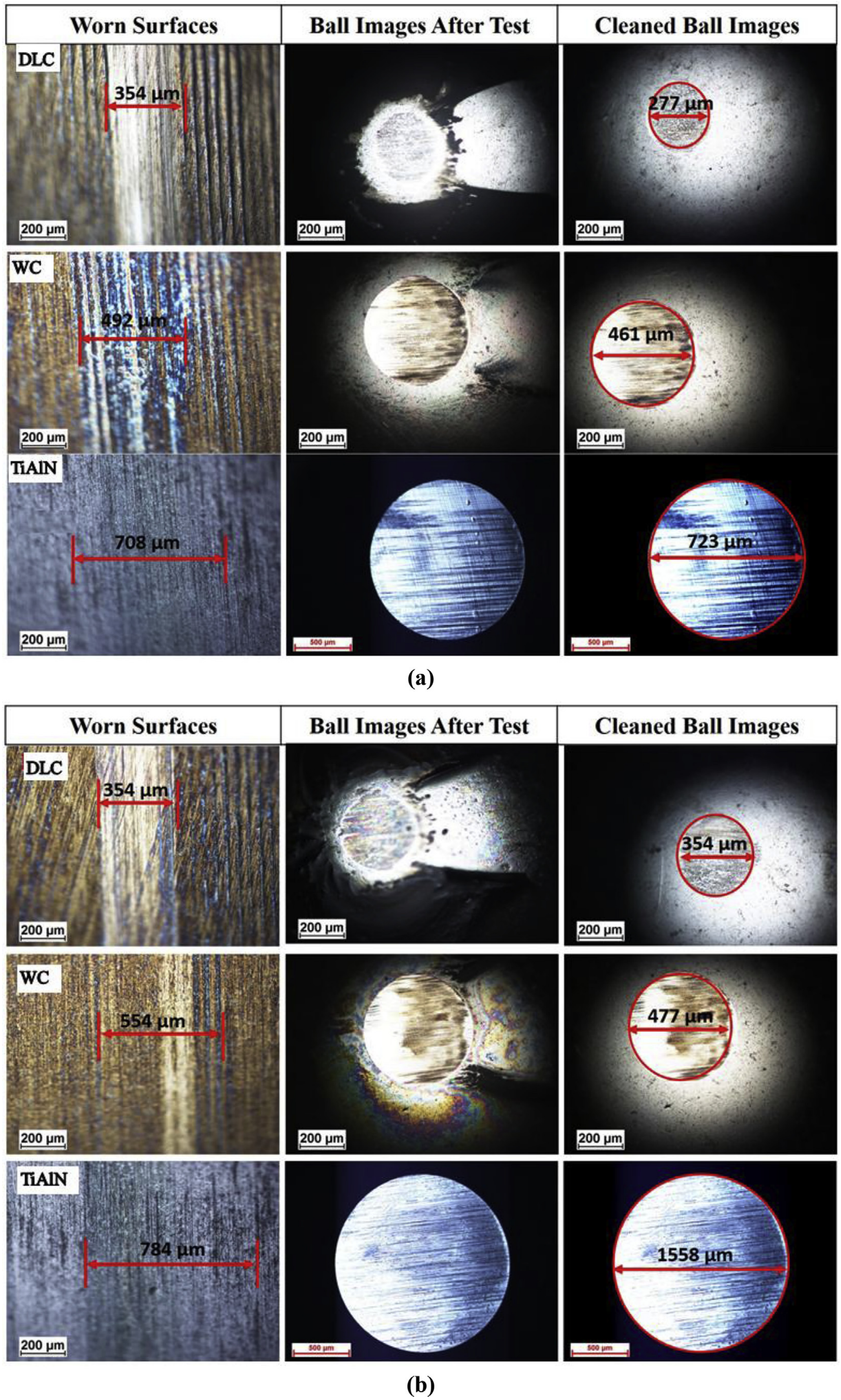

Fig. 10. Worn surfaces and ball images after test under base oil lubricated condition for (a) 10 N and (b) 15 N normal loads. The wear track width and wear scar diameters are shown by dimension lines.

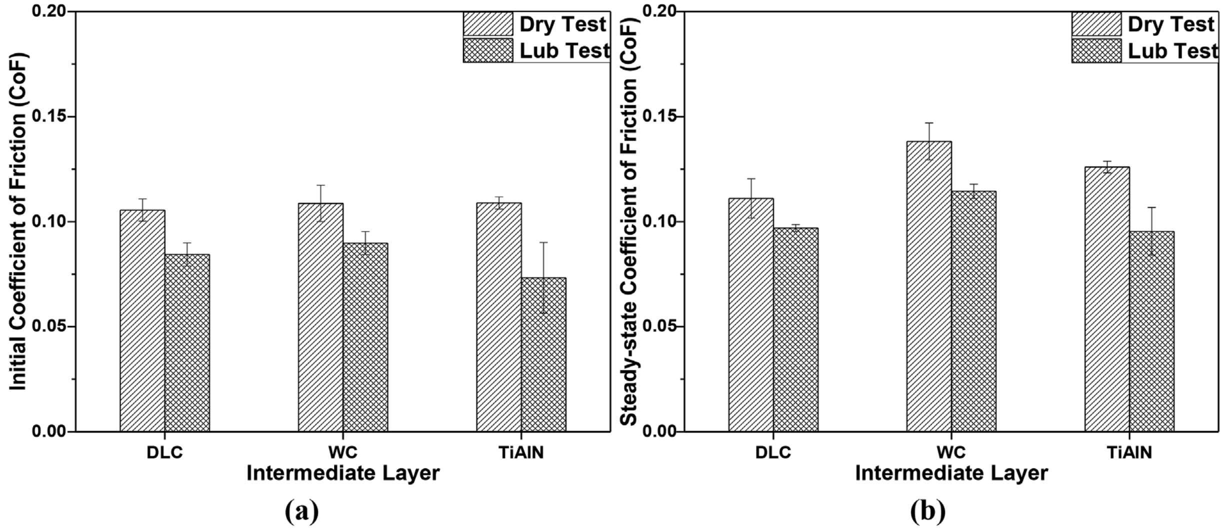

Fig. 11. (a) Initial and (b) steady-state CoF of EGnSN coating with different hard intermediate coating layers (dual coatings) under dry and lubricated test conditions at 10 N load and 0.63 m/s sliding speed.

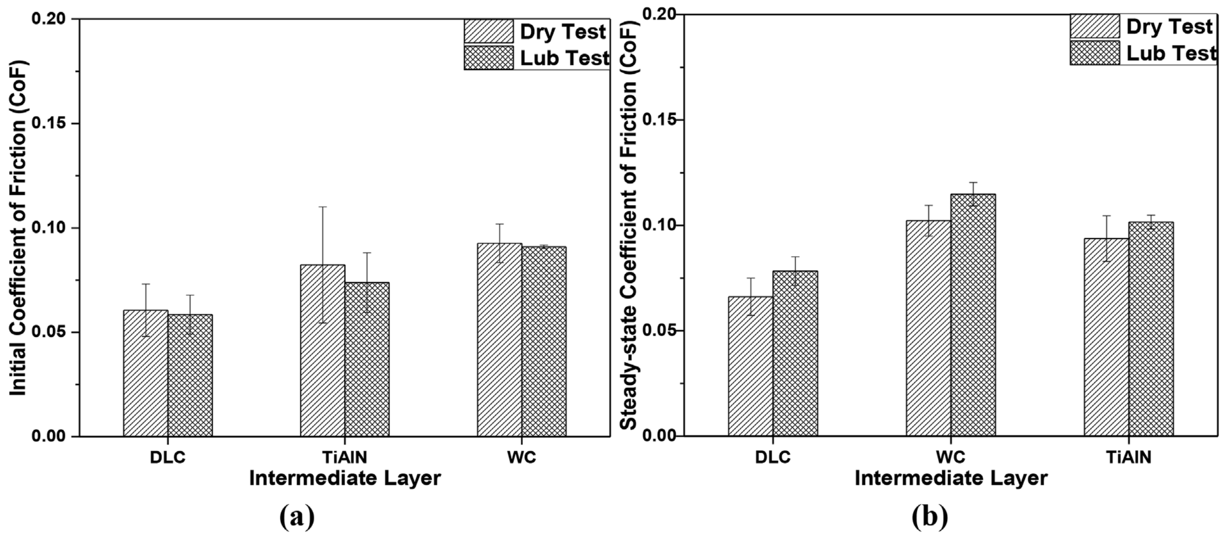

Fig. 12. (a) Initial and (b) steady-state CoF of EGnSN coating with different hard intermediate coating layers (dual coatings) under dry and lubricated interfaces at 15 N normal load and 0.63 m/s linear sliding speed.

the re-circulation of initial wear debris to the interface along with the lubricant which led to higher wear. The wear of DLC/EGnSN dual-coating was minimal in comparison.

3.3. Polymer film transfer mechanism

Transfer film of polymer on the counterface surface has great influence on the wear and friction characteristics. The transfer film formed during sliding between polymer film and ball counterface and its mechanism was understood. As hard steel ball slid over the soft epoxy composite film, there was transfer film formation on top of the ball surface. Hence, after the initial running-in, sliding between the transfer film and the soft coating eventually occurred. Hence, there was lower interfacial shear stress leading to lower CoF.

Since, the polymer coating contained *in-situ* lubricating oil (SN150) in the matrix, this oil was present on the ball surfaces in the form of transfer film, which acted as lubricant. These lubrication regimes were either boundary or mixed lubrication regimes, as discussed by Kumar et al. [12]. The material transfer was studied by taking optical images of the wear tracks of the DLC/EGnSN, WC/EGnSN and TiAlN/EGnSN dual-coatings and the balls. The test conditions were fixed at 0.63 m/s (300 rpm) linear sliding speed and 10 N and 15 N normal load for 2×10^5 cycles. The transfer film of epoxy composite consisted of oil droplets and asperities at the top of the contacting surfaces, which was sheared by ball counterface at the beginning of sliding. The thickness of transfer film was directly proportional to the track width of the coatings. It was clear that DLC/EGnSN dual coatings exhibited the lowest transfer film and wear. However, TiAlN as intermediate layer contributed large amount of materials to the transfer film, as shown in Fig. 14 (a).

Similar tribological behavior was observed at 15 N normal load and 0.63 m/s linear sliding speed for the dual coatings (DLC/EGnSN, WC/EGnSN and TiAlN/EGnSN) as shown in Fig. 14 (b). Under base oil lubricated test, the transfer films were removed leading to wear of ball counterface as shown in Fig. 15 (a) and (b) at 10 N and 15 N normal load, respectively. The transfer film formation by a polymeric material is hampered by the presence of a liquid at the interface.

Thus, the dry friction mechanism for dual-coatings is dominated by the transfer film formed on the counterface which should be thin with smeared liquid lubricant droplets for very low coefficient of friction.

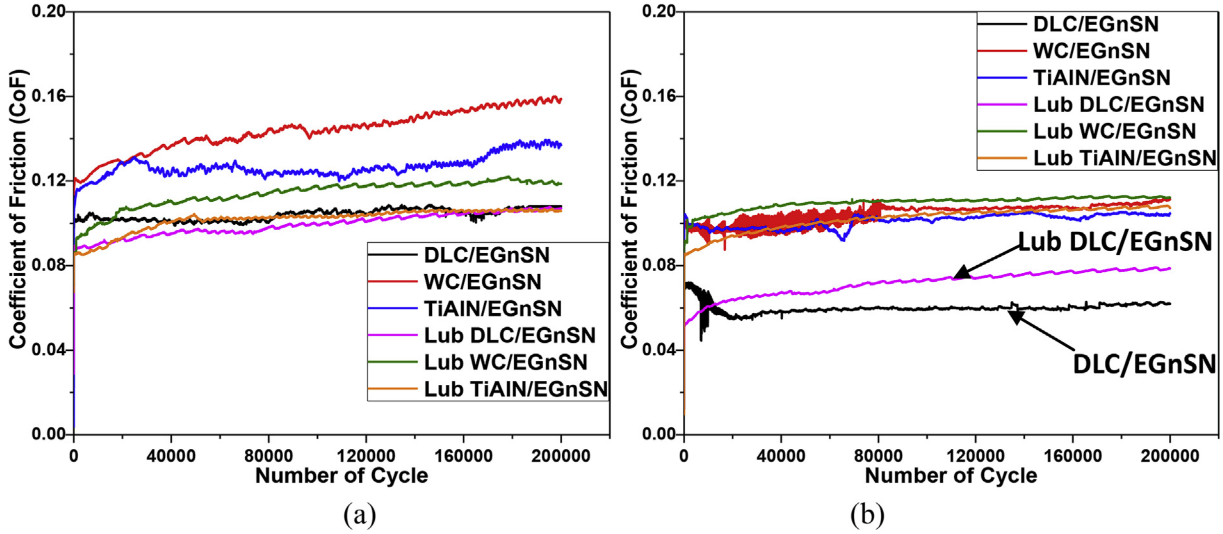

Fig. 13. Variations in CoF of epoxy/graphene/SN150 coating for different hard intermediate layers under dry and lubricated test condition at (a) 10 N and (b) 15 N normal loads and 0.63 m/s linear sliding speed. As shown in Figs. 11 and 12, epoxy/graphene/SN150 coating with DLC intermediate layer exhibited lowest coefficient of friction for both loads (10 and 15 N) under dry as well as lubricated condition.

The soft coating should have better adhesion with the intermediate layer which was the case with DLC because of hydrophilic nature of DLC. The presence of graphene and liquid lubricant filler in epoxy helped the polymer coating to be mechanically strong and lubricating in nature. Wear performance greatly depends upon the initial and steady-state friction. Wear is negligible for lower load but at higher load, wear of the soft coating takes place by abrasion by the asperities on the counterface and material transfer to the counterface. In the externally lubricated sliding condition, there is no transfer film formation and hence, there is abrasive interaction between the soft coating and the counterface in the mixed-lubrication condition. The coefficient of friction is low because of the presence of *in situ* and external lubricants, however, wear is high for the counterface because of the abrasive polishing actions in the presence of debris materials which re-circulate to the contact area.

3.4. Normal load effect on friction

Friction and wear properties of dual-coatings were tested at 10 N and 15 N normal loads and at 0.63 m/s linear sliding speed to understand the load effect for 2×10^5 cycles. Friction data are shown in Fig. 16. The CoF of all dual-coatings reduced with increasing normal load from 10 N to 15 N but there was sharp reduction in CoF (from 0.11 to 0.08) for DLC/EGnSN dual-coating under dry test condition due to larger lubricant transfer to the interface. There was little increase in CoF for WC/EGnSN and TiAlN/EGnSN dual coatings as load increased under lubricated condition. It indicated that in the mixed lubrication regime, as the normal load increased, the asperity to asperity contact also increased. This reduced the role of liquid lubricant at the interface.

3.5. SEM analysis

After the wear test of dual-coatings, SEM images of the wear tracks were taken and analysed. The wear track on the surface of DLC/EGnSN coating with DLC intermediate layer is found to be due to initial running-in only with no further wear. No scratch or material removal was observed and the surface of the wear track was quite similar in topography to the surface outside of the wear track. There were some scratches found on the surfaces of WC/EGnSN and TiAlN/EGnSN dual-coatings which still showed negligible wear, as shown in Fig. 17 (a, b and c) (after 2×10^5 cycles).

4. Conclusions

Hard coatings of DLC, WC and TiAlN were applied on D2 steel and their coefficient of friction and wear life were evaluated under dry and lubricated interfacial conditions at 10 N and 15 N normal loads and 0.063 m/s linear sliding speed. After that, epoxy composite coating (with 10 wt% of graphene) filled with liquid lubricant (10 wt% of SN150) was applied over the hard coated steel samples (dual-coatings) and tribological investigations were carried out under dry and lubricated (with external lubrication) interface condition in sliding against the counterface of 4 mm steel (SAE 52100) ball. The wear life of all dual-coatings were more than 2×10^5 cycles. The following conclusions were drawn from the experimental results and alalysis:

1. Amongst the three hard coatings, DLC coating showed the lowest coefficient of friction (0.14) under dry condition at 10 N normal load, whereas under lubricated condition, WC coating showed the lowest CoF of 0.075 and 0.09 at normal loads of 10 N and 15 N, respectively.
2. Amongst all dual-coatings at 10 N load, the CoF of the dual-coating with DLC intermediate layer was the lowest (0.09) in dry condition. In lubricated condition, it was lowest for TiAlN as an intermediate layer (0.07).
3. After 2×10^5 cycles of sliding, there was no wear or negligible wear on dual-coating with DLC intermediate layer or on the ball counterface under dry condition, but minor scratches were seen on the ball counterface under base oil lubricated condition at 10 N and 15 N normal loads.

It is thus concluded from the current results that DLC/epoxy composite (with 10 wt% graphene and 10 wt% base oil SN150) dual-coating is suitable for tribological contact conditions, in dry and externally lubricated conditions for machine components under extreme conditions.

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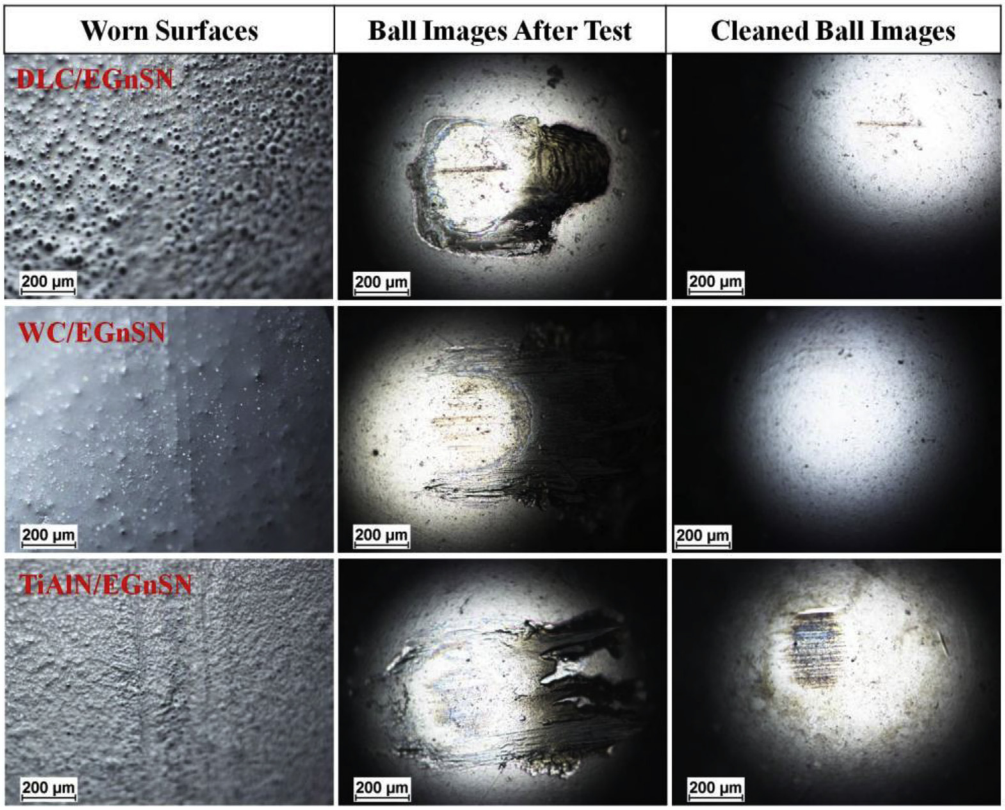
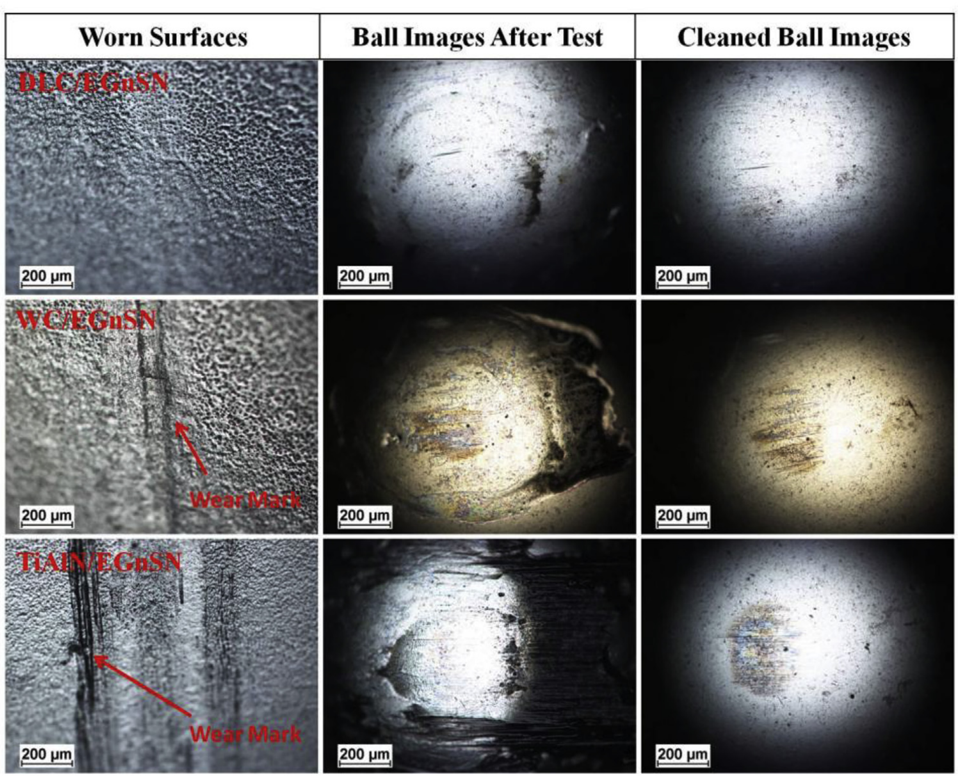
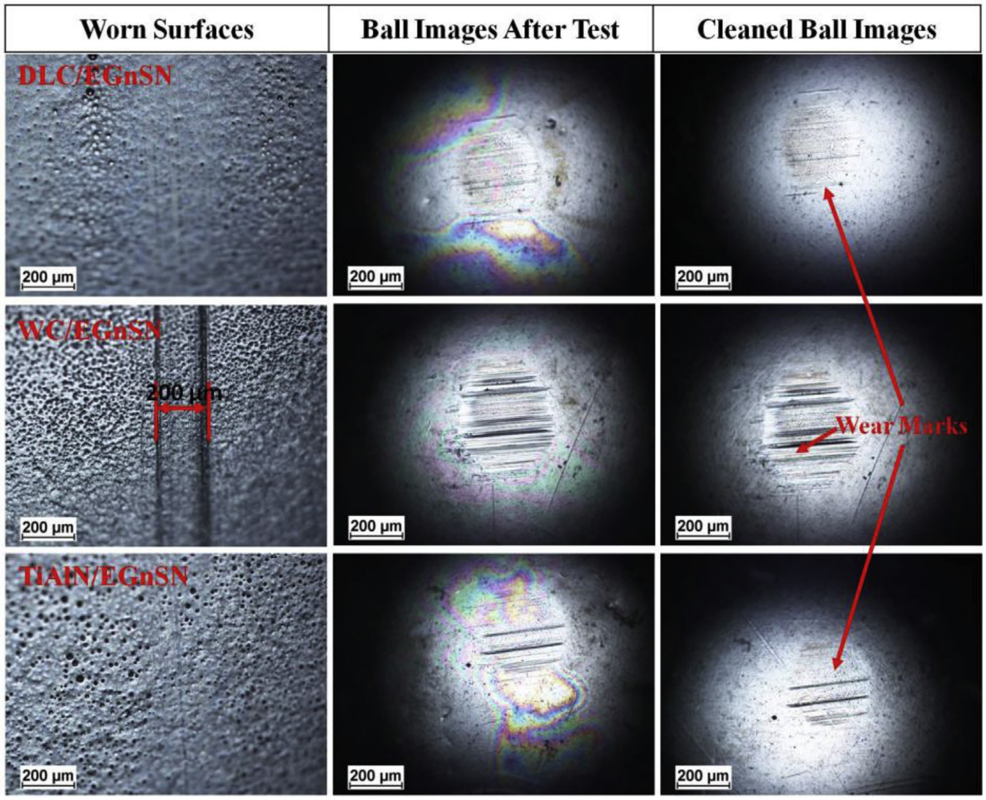


(a)

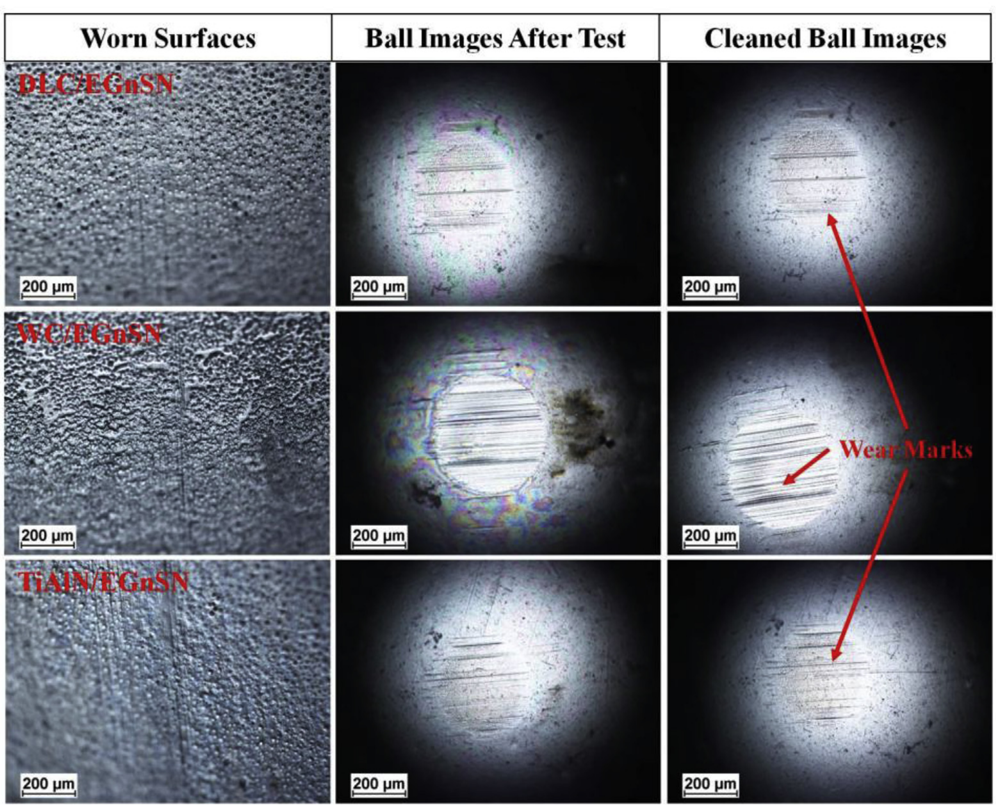


(b)

Fig. 14. Worn surfaces of the dual coatings and the corresponding ball images after test at (a) 10 N and (b) 15 N normal loads under dry test condition.



(a)



(b)

Fig. 15. Worn surfaces of the dual coatings and the corresponding ball images after test at (a) 10 N and (b) 15 N normal load under externally lubricated condition.

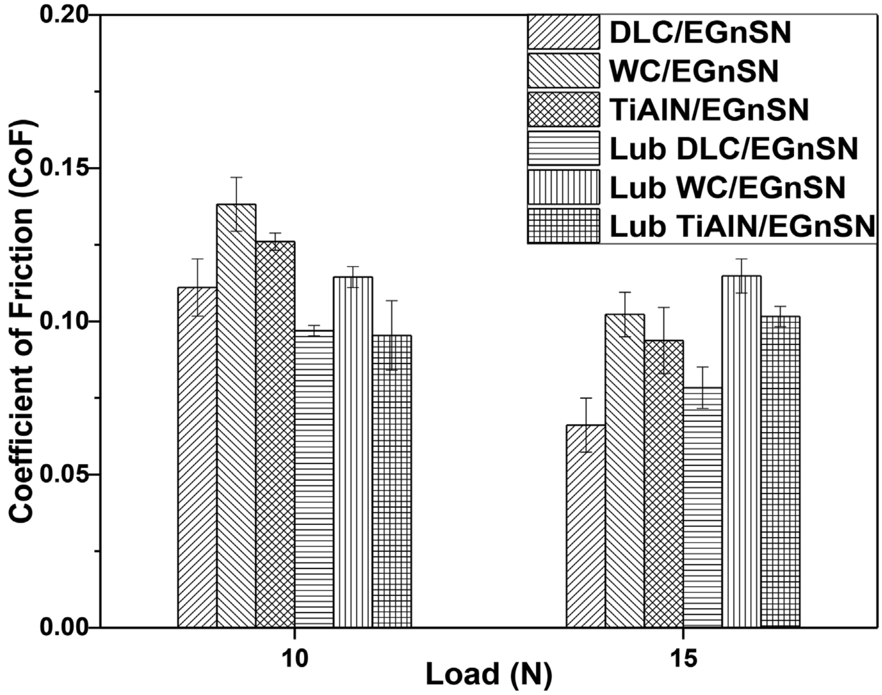


Fig. 16. CoF for dual-coatings at 10 and 15 N normal load under dry and lubricated test conditions.

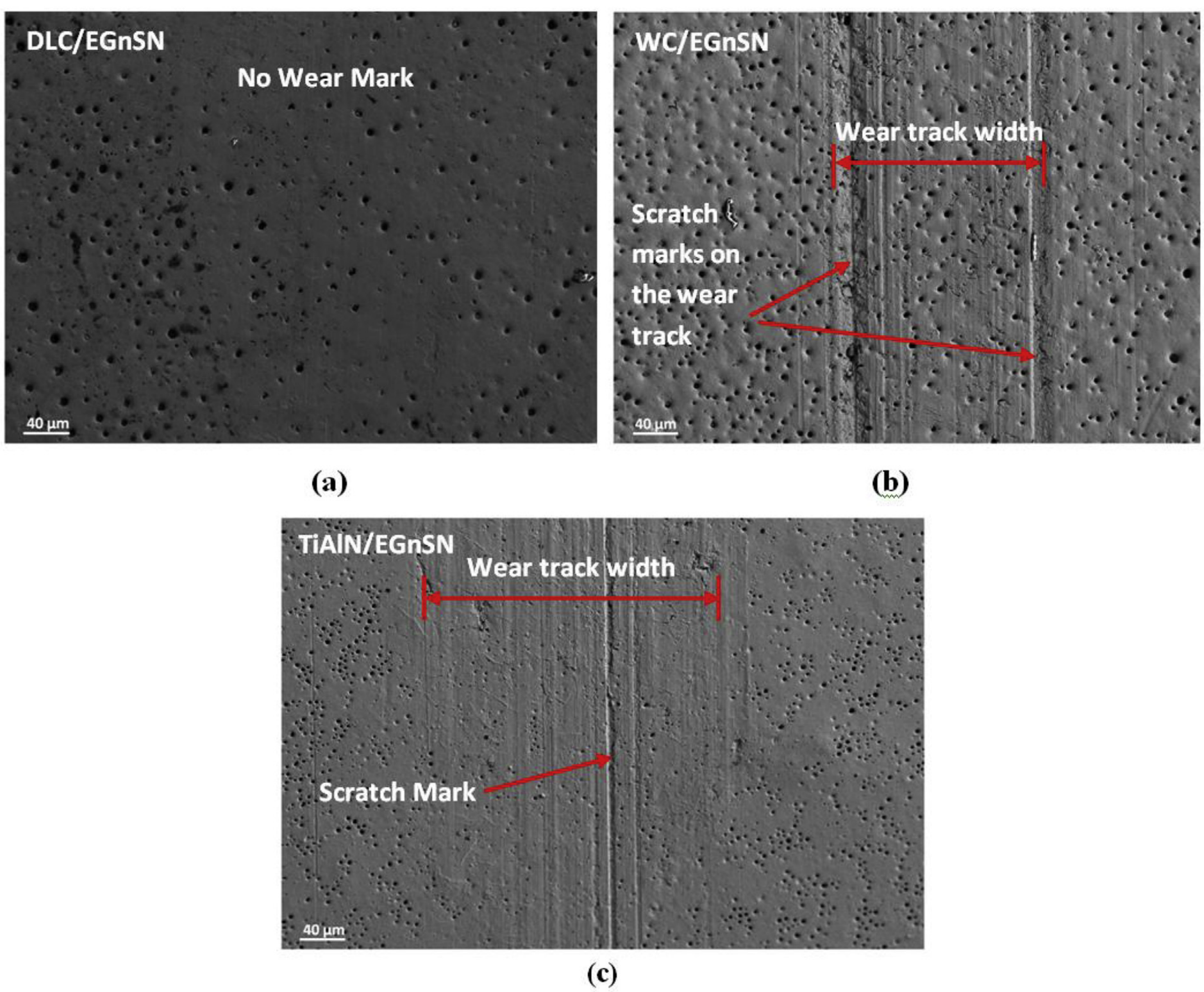


Fig. 17. SEM images of worn surfaces of dual-coatings (a) DLC/EGnSN (b) WC/EGnSN and (c) TiAlN/EGnSN after 2×10^5 cycle.

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