Tribological properties of polyimide coating filled with carbon nanotube at elevated temperatures

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Abstract
This work mainly focuses on the development and characterization of polyimide (PI) based composite coatings filled with multi-walled carbon nanotubes (MWCNT) on a steel substrate. The tribological properties of PI/MWCNT composite coatings at different temperatures ranging from room temperature (RT) to 200°C were investigated. Also, the influence of MWCNT and thermal & mechanical properties of PI composites coatings were measured. The addition of MWCNT could reduce the friction coefficient as well as wear volume at elevated temperatures. The PI/3MWCNT composites coatings show better wear-resistant properties as compared to other composites. Also, the PI/MWCNT composites showed improved mechanical properties such as micro-hardness and nanoindentation. Formation of an organic-inorganic structure due to the inclusion of MWCNT, enhance the mechanical properties and reduce the thermo-degradation of composites coatings. The glass transition temperature ($T_g$) of the PI/MWCNT composites was improved as the MWCNT content increases from 0.1 to 5 wt%.

KEYWORDS
carbon nanotube, elevated temperature, PI coatings, tribological properties

1 | INTRODUCTION

Self-lubricating polymer coatings with stable friction and high wear-resistant have been used to replace the traditional liquid lubricants, which could avoid the environmental pollution due to the liquid lubricant leakage.\cite{1,2} Polytetrafluoroethylene (PTFE) is normally used as a self-lubricating coating material in the application of rolling element devices, printing, woven, tobacco and sometimes gymnastic machines.\cite{3} However, the thin layer of PTFE coatings acting as a lubricant is quickly worn away and becomes ineffective at the high-temperature test condition.\cite{4} High temperature breaks the cross-linked network in the coatings, which accelerates the material loss and thus reduces the degradation performance, optical and mechanical characteristics. In this situation, polyimides (PI) coatings can be the best choice for this kind of application due to its outstanding mechanical properties at elevated temperatures.

PI is an important class of super-engineering high-performance plastic,\cite{5} characterized by repeating structure of an imide monomer and well known for their excellent chemical resistant, electrical characteristics, radiation resistance and mechanical properties at high temperature.\cite{6,7} Despite this, PI has high glass transition temperatures ($T_g$) ranging from 310°C to 315°C\cite{7} and
high melting point.\textsuperscript{[8,9]} This is due to the strong intermolecular forces between the polymer chains and the stiffness of the repeating units of aromatic parts in the heterocyclic structure.\textsuperscript{[6,10]} As such, PI composite coatings are often used in a solar cell, spaceship antenna, light emitting diode (LED) display, and mostly in mechanical moving parts like gear, bearing and bushing, and so forth.\textsuperscript{[11]} However, pure PI has high friction and wear rate that limit its utilization in industrial application.\textsuperscript{[12]} Therefore, various nano and micro filler materials are often incorporated into PI to improve the anti-wear and friction properties.

Carbon nanotubes (CNT) are one of the allotropes of carbon.\textsuperscript{[13]} It has ultra-high strength and low weight with highly conductive electrical and thermal properties. It is claimed that the mechanical strength and density of CNT are 100 times and one quarter that of steel, respectively. Also, its thermal conductivity is almost comparable to diamond.\textsuperscript{[14,15]} Because of these excellent properties, CNT is recommended as filler materials in the polymer composites, to increase the load-carrying capacity and tribological performance simultaneously. Because, the thermal conductivity of the polymer can be increased with the addition of CNT, which can lead to a contribution of temperature reduction in sliding contact area.\textsuperscript{[16,17]} Therefore, the internal stress in the contact area reduces which could improve the tribological properties.

Recently, more and more attention has been paid to the investigation of tribological properties of PI composites at elevated temperature. The tribological performance of PI is improved by incorporating carbon fibers into it at high temperatures (180°C-260°C) due to the graphitization of carbon fibers that provides lubricity on the worn surfaces.\textsuperscript{[18–20]} The effect of elevated temperatures in PI composites filled with mesoporous silica (MPS) was investigated.\textsuperscript{[21]} It is mentioned that the specific wear rate of PI/MPS was reduced by one order of magnitude up to 300°C because of the formation of the transfer film of the composite. Some studies showed that the transition of tribological behavior can occur at the temperature where the transfer film could be formed on the contact interface.\textsuperscript{[22–24]} To the best of the author’s knowledge, there was no research carried out for the tribological performance of PI/CNT composites in dry conditions at elevated temperatures.

In this work, the various weight percentages of CNT were incorporated into PI to enhance the mechanical and thermal properties of PI/CNT composites. In addition, the tribological properties of the PI composites at elevated temperatures were also studied. The wear mechanism of the PI composites was investigated by scanning electron microscopy (SEM). It is proposed that the present study can contribute to the extensive use of MWCNT in PI coatings at elevated temperatures.

2 | EXPERIMENTAL

2.1 | Materials

The aromatic polyimide precursors 4, 4 oxydianiline (ODA) with 97% purity (molar mass: 200.24 g/mol); N, N-dimethylacetamide (DMAc) (molar mass: 87.12 g/mol); and polar aprotic solvent pyromellitic dihydride (PMDA) with 97% purity (molar mass: 218.12 g/mol) were purchased from Sigma-Aldrich. Also, the filler material multi-walled carbon nanotubes (MWCNT) with an average of 3 to 15 μm length, 9 to 16 nm diameter, and purity 95% was brought from Nanjing XFNANO Materials Tech Co., Ltd, China.

2.2 | Preparation of PI and PI/MWCNT composites

Initially, 0.20 g of 4, 4-oxydianiline (ODA) was added in 2 mL of N, N-dimethylacetamide (DMAc) aprotic solvents and stirred until the ODA totally dissolved. Then 0.21 g of polyimide precursor pyromellitic dihydride (PMDA) was mixed into the previously made solution and stirred magnetically in room temperature around 6 h in an ice bath.\textsuperscript{[25,26]} The resulting solution known as polyamic acid (PAA) looked like a yellow or orange color. In a similar way, for the PI composites, various weight percentages (wt%) of MWCNT were added into the first solution (a mixture of ODA and DMAc) and sonicated for about 1 h until it was correctly dispersed. The following steps were the same as Polyimide PAA and subsequent PI/MWCNT PAA was black in color.

The PAA solution was poured on steel disc (100CR6 ESU hardened, Ø24 mm × 7.9 mm, and surface roughness [Ra] 120 nm, supplied by Optimal Instruments Prüftechnik GmbH, Germany, and were washed with acetone and ethanol before using) and followed lab wound technique to make smoother coatings. The coatings were then placed in a vacuum chamber at 80°C for 10 h to remove the moisture and solvents. The cross-linking of the PI with MWCNT was done by thermal imidization which follows: the resulting coatings from vacuum oven were heated in an oven at 100°C and hold for 1 h, heated from 100°C to 200°C and hold for 1 h, and finally heated from 200°C to 300°C and hold for another 1 h. Finally, the coatings were cooled at room temperature and the resulting coating was 30 to 50 μm thickness ready for the tribological test. Five different coatings were made as PI, PI/0.1MWCNT, PI/0.5MWCNT, PI/1MWCNT, PI/3MWCNT, and PI/5MWCNT where the numerical number indicates the weight percentage of MWCNT.
2.3 | Characterization

To determine the thermal stability of PI and PI/MWCNT composites, the thermal gravimetric analysis (TGA) study was performed by a TGA, TA instrument Q500 thermal analyzer under a nitrogen atmosphere from 34°C to 900°C with a heating rate of 10°C/min. The differential scanning calorimetry (DSC) was performed using NETZSCH instrument (STA449) in N₂ atmosphere at a fixed heating rate of 10°C min⁻¹, from 26°C to 450°C to measure the glass transition temperature (Tg) of the PI and MWCNT contents PI composites.

Micro-hardness of prepared PI and PI composites coatings were performed by the Matsuzawa MXT-α micro-hardness tester. For each specimen, five tests were conducted under the load of 10 g and lasting for 10 s. Vickers method and indenter were considered for the measurement.

A nanoindentation test of PI coatings was carried out for local or grain hardness measurement by means of Berkovich indenter in Nanotest Vantage, UK by slowly increasing the load up to 5 mN and after holding for a certain time the load was gradually released. For each sample, nine different tests were conducted and the average value of nine tests was considered in this paper.

The friction test was carried out in reciprocating type ball on disc SRV-III (Schwingung Reibung Verschleiß) machine. In the SRV-III machine, the thermocouple is integrated inside the machine and directly connected to the base where the sample is kept. The temperature is automatically controlled though it takes few minutes to get temperature stable from machine starting time. The constant controlled temperature shows in the display of the machine with other parameters.

A contact schematic diagram of the SRV machine is shown in Figure 1. The PI coatings surface was used as the lower specimens and commercially available 100Cr6 steel, Ra 20 nm with a diameter of 5 mm provided by SKF, Sweden was used as the upper specimens. The sliding was performed with a constant load of 10 N, reciprocating sliding frequency 5 Hz, linear stroke length 1000 μm and duration of 30 minutes for all the coatings. Every sample was tested for five different temperatures RT, 50°C, 100°C, 150°C, and 200°C. The steel balls were cleaned each time with ethanol to remove the dust, dirt from the surfaces. A computer is connected to the SRV tester recorded the friction curve automatically. The wear scar or wear volume produced on polymer coatings surface by steel ball during friction test was measured in a 3D optical surface profiler called Zygo New View™ 7300. Three repeated friction and wear tests were carried out for each run and an average of this value was taken into calculation.

The worn surface of the PI and PI/3MWCNT composite coatings surface were platinum coated to make conductive and then observed on a high-resolution bench-top scanning electron microscope (SEM) (JCM-6000 Neoscope, Jeol) at different magnifications. Only 3 wt% of MWCNT with PI coatings has been examined for SEM as it was showing high wear-resistant properties than any other composite coatings.

3 | RESULTS AND DISCUSSION

3.1 | Mechanical property and thermal stability

The thermal stability of PI composite coatings with various MWCNT contents is shown in Figure 2. It is shown that all the TG curves of polymer composite coatings follow the same change trend in the temperature range of 34°C to 350°C. But further increased of temperatures, the weight of the samples is slowly decreased due to the volatilization of some organic matter of PI matrix. This is taken place in the temperature range of 350°C to 550°C. When the temperature is increased from 550°C to 700°C, the weight loss rises sharply with this temperature range. The breakdown of the PI chains occurred at these temperatures due to the thermal decomposition of the PI main chain. The degradation rate of the PI and PI composite coatings after 700°C is slow. The differential thermal gravimetry (DTG) graph in Figure 2B shows the maximum degradation temperature at which the sample losses its maximum weight which is occurred between 585°C to 600°C. There is a slight difference between this degradation temperature like PI and PI/0.1MWCNT losses their maximum weight at 595°C, PI/0.5MWCNT and PI/5MWCNT at 591°C; PI/1MWCNT and PI/3MWCNT at 585°C. The slight variation of these degradation temperatures may be the reasons for integration and dispersion of CNT into PI.
However, the thermal stability of PI/MWCNTs composites is larger than that of pure PI at high temperatures ranging from 600°C to 900°C. The red circled mark indicating the high-temperature stability of composite coatings and this is inserted in the inset of Figure 2A in an enlarged view. At 60% weight residue, the corresponding temperatures for the PI, PI/0.5MWCNT, and PI/1MWCNTs are almost the same as 730°C; while for PI/0.1MWCNTs and PI/5MWCNT are 685°C and 765°C, respectively. The maximum temperature stability of PI composite is 887°C for PI/3MWCNT. Moreover, the weight losses of pure PI and 0.1%, 0.5%, 1%, 3%, 5% MWCNT content PI at 900°C are 55%, 53%, 56%, 56%, 60%, and 57%, respectively. Again, the PI/3MWCNT composite shows the high residual composition at 900°C as compared to others and proving the ability to withstand high temperature. So, MWCNT could enhance the heat resistant properties of PI/MWCNT composite coatings. Since the thermal resistance of the composite coatings depends on the proper dispersion and bond collaboration of MWCNT,[27,28] it can be concluded that the 3MWCNT is properly cross-linked and well dispersed with the PI chain as its’ showing high thermal stability.

The DSC analysis of the pure PI and MWCNT incorporated PI composites are shown in Figure 3. The endothermic peak corresponds to the glass transition temperature ($T_g$) of the PI, which is around 305°C. According to Figure 3, the $T_g$ of 0.1%, 0.5%, 1%, 3%, and 5% MWCNT contents PI composites are 309°C, 310°C, 311°C, 320°C, and 340°C, respectively. The findings suggests that the MWCNT helped to form secondary network structure along with primary crosslink structure of the polymers, whereas MWCNT immobilize the polymer chains at high temperature.[29] So, the increasing wt% of MWCNT into PI increases the $T_g$ of PI/MWCNT composites. $T_m$ indicates the melting point of the polymer, which remains same for all the composites.

Figure 4 shows the micro-hardness of PI composite coatings filled with various weight percentages of MWCNT. It is shown that the hardness is gradually increasing with MWCNT contents ranging from 0.0% to 5%. At the low percentages of MWCNT such as 0.1%, 0.5%, and 1% the hardness’s are 19.42, 20.56, and 21 Hv, respectively while at higher percentage the hardness is also high approximately 22.38 Hv at 5 wt% MWCNT. The addition of 5 wt% MWCNT into the PI matrix increases the micro-hardness of PI composite coating around 22% than pure PI. This micro-hardness result is similar to the previous report.[28] The reinforcing agent MWCNT enhances the adhesive strength into the PI matrix. Also, the micro-hardness of composite coatings increases due to the proper dispersion of MWCNT into the PI matrix as well as the development of a three-dimensional network.[30] Higher the micro-hardness increases the load-carrying capacity of the composite coatings.[27]
In this work, PI/3MWCNT shows greater weight residue at high temperature providing higher thermal stability. In addition, PI/3MWCNT shows better wear-resistant property than other composite coatings described in Section 3.2.1. Here, we choose PI/3MWCNT as the PI/MWCNT composite coating representative for the microscopy analysis. Table 1 shows the mechanical properties of the PI and PI filled MWCNT composites examined by nanoindentation. It can be shown that the value of hardness of the polymer composites increases with increasing MWCNT. The PI/5MWCNT composite hardness is about 18% higher than the pure PI. Also, the penetration depth of PI/5MWCNT polymer composite is 980 nm lower than that of pure PI (1074 nm). Moreover, the permanent deformation of the pure PI is bigger than the composite polymer. Besides these, by seeing plastic and elastic work, it can be proved that the PI/MWCNT composite resists the indentation more that is, inherent superior mechanical properties than the pure PI. Therefore, the addition of MWCNT into the PI increases the micro-hardness of the PI composites, which ultimately contributes to carrying the higher load.

Figure 5 shows the nanoindentation curve of pure PI and PI/MWCNT composite coatings. It can be seen that the load on the coating surface slowly increases to a maximum load of 5 mN, which could lead to deformation of the surface. The pure PI has a higher displacement or penetration than PI/MWCNT composites as shown in Figure 5. The main reason for the escalation of hardness is the inclusion of MWCNTs in PI, which actually contributes to cross-link the polymer matrix with it and resulting active and strong interface between CNT & polymer.[31] Moreover, it is also shown that the polymer composites have a wider elastic region than pure PI during the unloading period that is, MWCNT enhanced the elastic properties of the PI/MWCNT composite. Just before the end of unloading, there is a sudden recovery of materials in the curve for both PI and PI/MWCNT, which is occurred due to the viscoelastic behavior and thermal contraction of the polymer.

![FIGURE 4](https://example.com/figure4.png) **FIGURE 4** Effect of MWCNT content on micro-hardness of PI coatings (10 g load applied for 10 s). MWCNT, multi-walled carbon nanotubes; PI, polyimide [Color figure can be viewed at wileyonlinelibrary.com]

![FIGURE 5](https://example.com/figure5.png) **FIGURE 5** Nanoindentation curve of pure PI and PI/MWCNT composites. MWCNT, multi-walled carbon nanotubes; PI, polyimide [Color figure can be viewed at wileyonlinelibrary.com]

| Type           | Max. depth (nm) | Plastic depth (nm) | Hardness (MPa) | Plastic work (nJ) | Elastic work (nJ) |
|----------------|-----------------|--------------------|---------------|------------------|------------------|
| PI             | 1074            | 898                | 230           | 1.39             | 0.85             |
| PI/0.1MWCNT    | 1043            | 872                | 240           | 1.39             | 0.83             |
| PI/0.5MWCNT    | 1018            | 845                | 255           | 1.31             | 0.84             |
| PI/1MWCNT      | 1014            | 834                | 261           | 1.37             | 0.86             |
| PI/3MWCNT      | 1001            | 824                | 267           | 1.23             | 0.85             |
| PI/5MWCNT      | 1001            | 804                | 280           | 1.15             | 0.85             |

Abbreviations: MWCNT, multi-walled carbon nanotubes; PI, polyimide.
3.2  |  Tribological behavior

3.2.1  |  Friction and wear volume

Figure 6 illustrates the friction coefficient of PI and PI/MWCNT composite coatings at different temperatures. It can be seen that the friction coefficient of neat PI at room temperature is 0.817, and then increases with temperature rises. But at 150°C, the friction coefficient decreases to the value of the room temperature (RT) and again it becomes high 0.874 at 200°C. Overall, the friction coefficients of pure PI at higher temperatures are relatively larger than RT. PI is a viscoelastic material and its friction mainly depends on the adhesion between the steel and PI surface area. At the proper heat and shear stress, the PI chain is lessened due to the rubbing in the sliding direction and the resulting decrease in friction.[32] Therefore, the pure PI has a lower friction value at 150°C. But further increased the temperature, decline the hardness of the coatings which enlarge the contact area and causes severe wear on the contact interface[27] as shown in Figures 7 and 8. As a result, the friction coefficient of PI again increases at 200°C. For PI/0.1MWCNT, the friction coefficient is continuously increasing (0.755 to 0.849) from RT to 200°C but lower than pure PI. From Figure 6, it is also seen that the friction coefficient of some of the PI composite coatings at RT is higher, indicating the CNT does not provide lubricity at this low temperature. Further increasing loading of CNT into PI reduces the friction at elevated temperatures. Overall, the average lowest friction coefficient of the PI composite is observed at 50°C and 100°C. The lowest variation of friction coefficient can be seen in PI/3MWCNT composites from RT to 150°C in Figure 6. At high loading 5 wt% CNT, the friction pattern of the composite coating is lower than pure and other composites both at RT and elevated temperatures. At the RT, the friction curve has higher fluctuation with a lot of spikes but with increasing temperature, the curve becomes stable with little variation. Also, spikes start disappearing from 50°C. However, with the further increased temperature like 200°C, the friction coefficient again starts to become unstable and high. This is due to the thermoplastic behavior of PI and contact geometry.[18] However, the addition of CNT in PI slowly decreases the friction with the gradual rise of test temperatures. This friction reduction is due to the rolling of embedded particles generated from the PI/MWCNT surfaces at high temperatures because the viscous polymer cannot hold it at this condition.[27,33] Alternatively, PI/MWCNT could form a transfer film on the counterpart.

**FIGURE 6** Friction coefficient of A, PI; B, PI/0.1MWCNT; C, PI/0.5MWCNT; D, PI/1MWCNT; E, PI/3MWCNT; and F, PI/5MWCNT at different temperatures. MWCNT, multiwalled carbon nanotubes; PI, polyimide [Color figure can be viewed at wileyonlinelibrary.com]
steel ball\cite{34} and hence, the friction coefficient as well as wear volume reduce as in Figures 6 and 7, respectively.

Figure 7 illustrates the wear volume of PI and various weight percentages of CNT incorporated PI composites from RT to 200°C. For PI coating, the wear volume reduces and then increases with the increasing temperature, which is similar to the previous report.\cite{19,22} The minimum wear volume of the pure PI coating is $1.42 \times 10^{-3} \text{ mm}^3$ at 50°C. Also, the graph clearly depicts that the PI without CNT has the highest wear volume $4.82 \times 10^{-3} \text{ mm}^3$ observed at 200°C, which is almost 3-fold of RT. A similar trend of wear volume can be observed for the PI/MWCNT composite coatings from RT to high temperatures. The addition of 0.1 wt% CNT has no or little effect of wear-resistant in the PI composites and even higher wear than pure PI at 50°C, 100°C, and 150°C. This may be attributed to the low content of MWCNT into PI coating. When the CNT content increases to 0.5 wt%, 1.0 wt%, 3.0 wt%, and 5.0 wt%, the wear volume is significantly reduced from the neat PI coating. The lowest wear volume observed at 50°C for PI/5MWCNT is $0.46 \times 10^{-3} \text{ mm}^3$ but it sharply rises with further increased temperatures. Higher weight percentage and large surface area of CNT could agglomerate and increase the wear volume.\cite{35} Also, it is clearly distinguishable from Figure 7 that the 3.0 wt% of MWCNT has

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure7.png}
\caption{Wear volume of worn coating surfaces of PI and PI/MWCNT. MWCNT, multi-walled carbon nanotubes; PI, polyimide [Color figure can be viewed at wileyonlinelibrary.com]}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure8.png}
\caption{3D wear scar of A, PI; B, PI/0.1MWCNT; C, PI/0.5MWCNT; D, PI/1MWCNT; E, PI/3MWCNT; F, PI/5MWCNT where 1, 2, 3, 4, and 5 denotes RT, 50°C, 100°C, 150°C, and 200°C, respectively. MWCNT, multi-walled carbon nanotubes; PI, polyimide [Color figure can be viewed at wileyonlinelibrary.com]}
\end{figure}
a low variation of wear and overall higher wear-resistant than other coating compositions. Also at elevated temperatures 50°C, 100°C, and 150°C the wear-resistant phenomena of the PI/3MWCNT composite coatings are superior and the best loading among five composites. Thus, the CNT increases the wear resistance property of PI coatings under higher temperatures. The incorporation of CNT into PI could form an organic-inorganic hybrid structure that inhibits the heat flow to the PI matrix. This helps to decline the thermal degradation of the composite structure which is observed in Figure 2. Therefore, the wear-resistant property of PI composite coatings is significantly improved.

The 3D optical photo of the worn surfaces of the neat PI and PI composite coatings are shown in Figure 8. The neat PI coating surfaces slowly wear out and with increasing temperatures, the volume of the scar also increases. The black color of 3D wear scar of the pure PI coating at 200°C in Figure 8A5 indicates the damage of the coating at high temperature, which specifies that the further increased test temperature will degrade the coating due to high friction and will result in higher wear volume. The wear track of the worn surfaces becomes smaller and shallower from 0 to 3 wt% MWCNT. It is interesting that PI/3MWCNT has a small wear track up to 150°C while PI/5MWCNT has up to 100°C. From the 3D optical photo of the worn coating surfaces in Figure 8, it can be seen that the addition of MWCNT could increase the temperature range of PI composite coating in the tribological application, which could be due to the higher Tg of PI/MWCNT as shown in Figure 3. Therefore, the worn surfaces of composite coating still remain undamaged at high temperature. In summary, Figure 8 is basically the physical representation of the wear volume of Figure 7 and shows the change of shape of the wear tract of PI and PI composites coating over increasing temperatures.

3.2.2 Wear morphology analysis

Figure 9A shows the SEM micrograph of wear track of pure PI at 50 μm magnification and at five different temperatures ranging from RT to 200°C. At RT and 50°C, the coating surfaces are quite smooth and considered with many furrows which indicates the dominant nature of abrasive wear mechanism. At 100°C, both of furrows and spalling points are observed on the worn surface, designating the abrasive and fatigue wear mechanism. Moreover, severe cracks and peel of the materials from the surface of the coating occurred, perpendicular to the sliding direction, representing fatigue wear at 150°C and 200°C temperatures. The result of this SEM micrograph of pure PI coatings at high temperatures is similar to the previous reference paper. At RT, there is a crack or scar on the coating surface, which means the surface itself does not resist the load applied to it. However, with the increasing temperature like at 50°C and 100°C the surface becomes smoother and appears very low scar on the worn surface as compared to other coating surfaces. It can be explained in the way that these temperatures help to relax the PI chain and branches which helps to reduce friction as well as wear. However, with increasing temperatures like at 150°C and 200°C the coating hardness decreases and that leads to severe wear or removal of stock of materials as we can see the on coating surfaces at these temperatures. Also, at these two temperatures, more wear debris or lots of separated chunk of materials.

![Figure 9](image-url)

**Figure 9** SEM micrograph of worn surfaces of (A) PI coatings; and (B) PI/3MWCNT coatings; (where 1, 2, 3, 4, and 5 denotes RT, 50°C, 100°C, 150°C, and 200°C, respectively). The arrows depict the sliding direction. MWCNT, multi-walled carbon nanotubes; PI, polyimide
appearing on the worn surfaces. This is also the reason for the increase of friction coefficients at high temperatures as shown in Figure 6.

To understand the effect of MWCNT on the tribological behavior of the composite coatings at elevated temperatures, the SEM micrograph of PI/3MWCNT coatings is shown in Figure 9B. Compared with the SEM photographs of pure PI coating surfaces as in Figure 9A, it is observed that the PI/3MWCNT coating surfaces are smoother. Addition of MWCNT with proper dispersion into PI forms PI-MWCNT structure, which helps to increase the load capacities. At RT, the presence of the small grooves on the worn surface indicates the detachment of a fragment from the surfaces. This fragment could attach to the mating parts like ball which describe as adhesive wear. [28] Also, at elevated temperatures from 50°C to 150°C, there is no major crack and breaking off materials from the worn surfaces, which will lead to stable and low wear volume. However, the adhesive wear becomes more intense in this condition and increases with increasing temperature. At these temperatures and load, a layer of microscopic materials from the composite coating surfaces could transfer to the contact interfaces which ultimately produces a lubricating layer and helped to resist severe wear. [21,37] Further increased temperature like 200°C, the crack or furrow appeared on the worn surface representing abrasive wear of the PI/MWCNT composite. [21] But the intensity of the CNT content PI wear mechanism is not severe as pure PI wear at high temperature. So, adding the CNT could provide great lubricity, enhance the load-carrying capacity and improve the wear resistance of PI composites. [38,39] Therefore, CNT filled in PI composites can withstand the larger shear force at elevated temperatures.

4 | CONCLUSIONS

To sum up, the thermal stability, mechanical and tribological properties of PI/MWCNT composites at elevated temperatures (RT, 50°C, 100°C, 150°C, and 200°C) have been investigated in this work. The gradual addition of MWCNT into PI coatings increases the micro-hardness of the composite coatings. PI/5MWCNT displays 22.22% more micro-hardness than pure PI. At 60% weight residue, the composite coatings PI/3MWCNT show higher thermal stability that is, 887°C than other composites in TG analysis. In addition, the glass transition temperature of the polymer increases after the addition of MWCNT, which is around 12% higher than pure PI. The friction coefficient and wear volume of the PI composites coatings significantly reduced by the incorporation of CNT. Under elevated temperatures situations, the CNT-reinforced composite coatings show low friction and better wear resistance than neat PI coatings. The wear volume of the polymer composites is reduced as the CNT content increases. In the meantime, the friction coefficient of the PI coatings filled with CNT slightly decreases but exhibits a greater reduction of wear volume with the increase of test temperature. The addition of MWCNT could increase the temperature range of PI composite coating in the tribological application. Such behavior is mainly due to the formation of organic-inorganic structure which enhances the mechanical properties and reduces the thermolysis of composites coatings, so the wear resistance is significantly improved at elevated temperatures.

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