Express Polymer Letters Vol.16, No.5 (2022) 476–487
Available online at www.expresspolymlett.com
https://doi.org/10.3144/expresspolymlett.2022.36

Research article

Improving the mechanical, wear and anti-corrosion performance of polyester coating on structural steel by graphite addition

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Received 21 October 2021; accepted in revised form 30 December 2021

Abstract. Organic polymer coatings have been extensively used for painting in industrial applications to prevent the deleterious effects on the metallic components. In this study, electrostatic polyester coatings with two amounts of graphite (5 and 10 wt% Gr) were deposited on structural steel substrate. The effect of Gr addition on the microstructure, mechanical, adhesion and tribological behavior were characterized using SEM, EDS, XRD, nano-indentation and scratch test. The coatings' resistance to corrosion was investigated using electrochemical measurements in 3.5 wt% NaCl solution. According to the results, the Gr particles were homogeneously distributed throughout the polyester coatings. The hardness and elastic modulus of the polyester coatings increased from ~0.23 and ~6.7 to ~0.68 and 10.9 GPa upon increasing the Gr contents from 0 to 10 wt% Gr, respectively. Moreover, the friction coefficient decreased from ~0.5 to ~0.2. The results also showed an attenuation of the severe wear mechanism and a reduction of the plastic deformation. Consequently, the wear resistance improved by three times due to the lubricating role of Gr. All the coatings showed a high adhesion level to the steel substrate with a cohesive critical load higher than L_{c1} = ~7 N and an adhesion strength greater than σ_c = ~215 MPa. The incorporation of Gr allows high protection of the steel against corrosion due to their barrier role.

Keywords: *thermosetting resins, coatings, adhesion, mechanical properties, damage mechanism*

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1. Introduction

Structural S235 steels with comprehensive mechanical performances are widely used on dynamically loaded and marine structures due to their good strength, forming and welding abilities, and low costs [1]. Although uncoated steels perform well in dry environments, they are highly susceptible to adhesive wear and corrosion [2]. Electrostatically deposited epoxy or polyester powder coatings show a better performance in the protection of steel parts against corrosion] [3–4]. Nevertheless, the wear resistance of these thermoset polymers' coatings is low, and the corrosion resistance of steel decreases with the increase of the friction damage of coatings [5].

Like the bulk mechanical responses of polymers, the tribological characteristics of polymers coatings are greatly influenced by temperature, the relative speed of the interacting surfaces [6], the normal load [7], and the environment [8]. Therefore, to deal with these effects, the incorporation of appropriate fillers into the polymers' matrix imparts special characteristics and allows their use as a composite coating for better hardness, excellent corrosion resistance and self-lubrication properties [9].

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Several attempts have been made to improve the wear properties of thermoset polymer coatings by adding proper solid lubricant fillers [5, 10]. In this regard, the frequently used friction and wear reducing fillers, in most polymers, are polytetrafluoroethylene (PTFE), graphite (Gr) and molybdenum disulfide MoS_2 powders [10–11]. Among all fillers, Gr is widely used as an efficient solid lubricant. In fact, under sliding conditions, the shear force can easily break the graphite structure due to the weak van der Waals bonds between the graphite layers. The symmetrical distribution of graphite flakes on the sliding surface reduces direct contact between the composites and steel counterparts [12]. Zouari *et al*. [10] concluded a reduction in friction and wear with the incorporation of Gr fillers on polyester coatings deposited on aluminium substrate. Chang *et al*. [13] found that under standard testing conditions, the specific wear rate of polyetherimide (PEI) filled with graphite was 800 times lower than that of a neat matrix. Similarly, the epoxy matrix filled with graphite and/or carbon nanotubes enhances the tribological properties of the epoxy resin, by reducing the friction coefficient and the wear rate of steel [14]. Friedrich [15] have demonstrated that carbon-based fillers could significantly improve the wear properties of coatings by improving the lubricating effect and strengthening the matrix.

The effect of the incorporating carbon-based fillers on the tribological properties of thermoset coatings remains an open field for further research. Numerous studies have been performed on the tribological properties of carbon/polyester composites coatings [4, 5, 10, 16–18]. However, the influence of graphite addition on the micromechanical, corrosion and scratch behavior of thermoset coatings are still poorly understood [4, 5]. To our knowledge, no study has given much attention to the correlation between the mechanical, corrosion and tribological properties of incorporating Gr fillers into polyester matrix coatings on steel substrate. For this purpose, the current work aims to investigate the effect of graphite addition on the mechanical, adhesion and tribological performance of the polyester coatings. Special importance is attributed to the corrosion resistance related to the barrier effect of graphite fillers.

2. Materials and methods

2.1. Materials

A mild steel sheet (S235) machined to the dimensions of $100\times80\times2$ mm^3 was used as substrates. The polyester powders resin purchased from KC-Kimia society was used both with and without incorporating graphite (Gr) to coat the steel sample substrates. This resin was designed to give good adhesion and high corrosion resistance to metal substrates. The purity of the polyester powder resin was 99.9%. The graphite obtained from Sigma-Aldrich with an average particle size of around ~20 microns was used as a filler in the polyester coatings with three different weight ratios (0, 5, 10 wt% Gr).

2.2. Substrate and polyester/Gr coatings preparation

The application of polyester powder mixed with Gr particles onto a substrate surface using the-electrostatic coating process requires only a simple experimental setup which is detailed in Figure 1. Before depositing the coating, the steel substrate surfaces were cleaned with acetone, then polished with a series of silicon carbide sandpapers. Subsequently, they were roughened by sandblasting to a surface finish $R_a \approx 3$ µm, to promote adhesion between coatings to steel surfaces. Then, the substrates were dried at

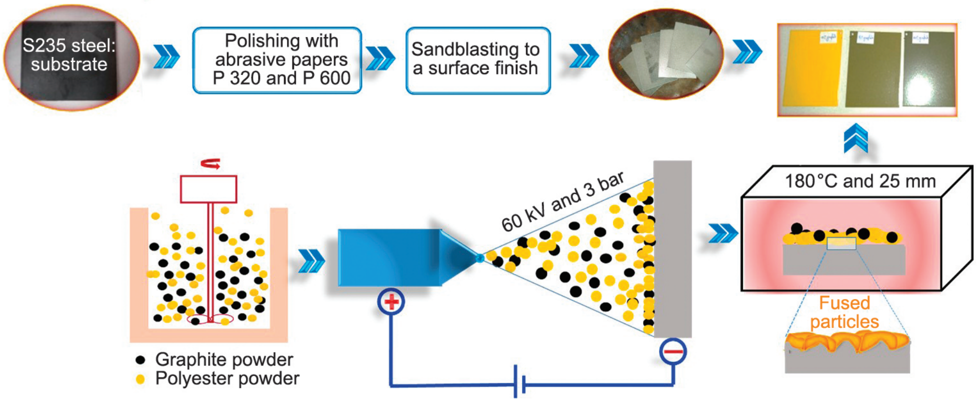


Figure 1. Schematic diagram detailing the electrostatic spray deposition process of polyester/Gr coatings.

60 °C for 24 h. Polyester resin powders were separately mixed with two different amounts of graphite (5 and 10 wt%) in a plastic container rotating on rollers at a rate of 100 rpm for 30 min. Pure powder paint without Gr was used as control (0 wt% Gr).

The coatings were applied using an ITW GEMA powder coating electrostatic gun. The potential of the electrode is 60 kV. A high voltage corona discharge electric field was formed between the spray gun and the workpiece, as shown in Figure 1. When the polyester powder was ejected from the spray gun, it interacted with many electrons to create charged particles. Under the action of electrostatic attraction, these charged particles adsorbed on the positively charged workpiece.

Finally, the applied coatings were cured in a convection oven at 180 °C for 25 min for melting and spreading over the substrate, followed by cooling to ambient temperature. The coatings thickness was measured at about 44±2 µm using an Ecotest plus device gauge.

2.3. Composite polyester/Gr coatings characterization

2.3.1. Structural characterization

The film composition and microstructure of the PS/Gr coatings were evaluated using Scanning Electron Microscopy (SEM (SEM-JEOL JSM 6460LV)) equipped with an energy-dispersive X-ray spectroscopy (EDS) for chemical composition identification. The X-ray spectra were obtained at primary beam energy equal to 20 keV for an acquisition time of 120 s. The study of phase composition of the samples was performed using X-ray diffractometer (Bruker D8 Advance). X-ray pattern data were gathered from 2θ = 10 to 70°.

2.3.2. Nano-indentation testing

The mechanical properties of the polyester-graphite coatings (Hardness and Youngs' modulus) were studied using the nano-indentation technique developed by CSM instrument (Anton Paar). Tests were conducted with a nano-indenter equipped with a diamond Berkovich tip with a nominal angle equal to 65.3° and a nominal radius curvature of 20 nm. The maximum applied load was 10 mN and the penetration depth was set to a value lower than 1/10th of the coating thickness to provide the real film properties and avoid the substrate effect [19]. Fifteen measurements at least were carried out for each coating to ensure the reproducibility and repeatability of the results. The analysis of load-displacement graphs with the Oliver and Pharr method [20] allowed the estimation of the mechanical properties (Young's modulus, hardness of the film).

2.3.3. Adhesion testing

The film adhesion was investigated with a micro scratch tester from CSM instruments (Anton Paar). The indenter was equipped with a Rockwell diamond tip with a radius of 200 µm. The test was performed in a progressive mode in a loading range between 0.3 and 25 N along 3 mm of the length scratch to study the adhesion behavior and to determine the film critical loads. The scratch speed was 10 mm/min.

The tests were carried out in three consecutive steps: pre-scan at 300 mN to determine the initial profile of the samples. Then, progressive scan from 0 to 25 N. During the scratch test, the penetration depth (P_d) was recorded in real-time and post-scan at 100 mN. The SEM was used to study the shape of the residual deformations after scratch tests. Finally, the indenter conducted a post-scan in a constant load of 300 mN, and measured the residual depth (R_d) of the scratch. After the test, the morphology of the scratch was examined by SEM. At least three progressive load scratch tests were carried out for each sample.

2.3.4. Tribological characterization

Tribological tests were performed at a constant normal load using a multi-pass scratch test (an Anton Paar CSM instruments) under a unidirectional loading over a sliding distance (d) of 3 mm. Each test was done at a temperature equal to 22 °C and relative humidity of 46%. The number of passes, the applied load (F_n), and the sliding speed (V) were controlled and fixed during wear testing. The applied normal loads were 1 and 3 N. Tests were carried out during 100 passes for each load condition. Then, wear tracks were examined by Scanning Electron Microscopy (SEM). For each test, the wear volume was calculated by measuring the surface profile on the wear track.

2.3.5. Corrosion testing

To investigate the corrosion resistance of polyester/Gr coated steel, a potentiodynamic polarization test was conducted on the coated specimens in comparison to the uncoated one in 3.5% NaCl aqueous solution using a VoltaLab Electrochemical Equipment PGZ 301. Three electrodes were used for corrosion measurements. Platinium foil was used as an auxiliary counter electrode, saturated calomel electrode (SCE) as a reference, and the specimens acted as working electrodes. The samples were polarized from –1.2 to 0.2 V with a scan rate of 10 mV/s.

3. Results and discussion

3.1. Structural characterization of polyester/Gr coatings

The microstructure of the composites polyester/Gr coatings are shown in Figure 2. The coatings were dense and uniform. It is revealed that the Gr particles were uniformly dispersed in the polyester matrix. The Gr took the form of flake, and the number of Gr particles rose from 5 wt% Gr (Figure 2b) to 10 wt% Gr (Figure 2c). Typical EDS analysis confirmed the presence of oxygen and carbon (Figure 2d). The C content increased with the addition of graphite. This result was proved by the analysis of the chemical composition of the coating's surface using EDS, as shown in Figure 2d. XRD curves show the patterns of polyester and the composite graphite/polyester coatings (Figure 2e). It was found that the composite coatings exhibited a strong diffraction peak at $2\theta = 26.3°$, corresponding to graphite fillers. In addition, the recorded XRD patterns indicated an increase in peaks intensities of graphite from 5 to 10 wt% Gr.

3.2. Mechanical properties of polyester/Gr coatings

Nano-indentation test is useful for evaluating the micromechanical properties of coatings. Figure 3a shows typical load-unload curves of polyester composite coatings prepared with 0, 5, and 10 wt% Gr. The average value and standard deviation of microhardness (H) and elastic modulus (E) of each sample are calculated from 5 indentation tests using the Oliver-Pharr approach [20] and summarized in Table 1. During the loading and the unloading stages, the penetration depth increased as a function of the applied load, and a plastic deformation occurred until the maximum load corresponded to the maximum penetration. All coatings showed an elasto-plastic behavior with a sign of high elastic recovery for an organic film (Figure 3a).

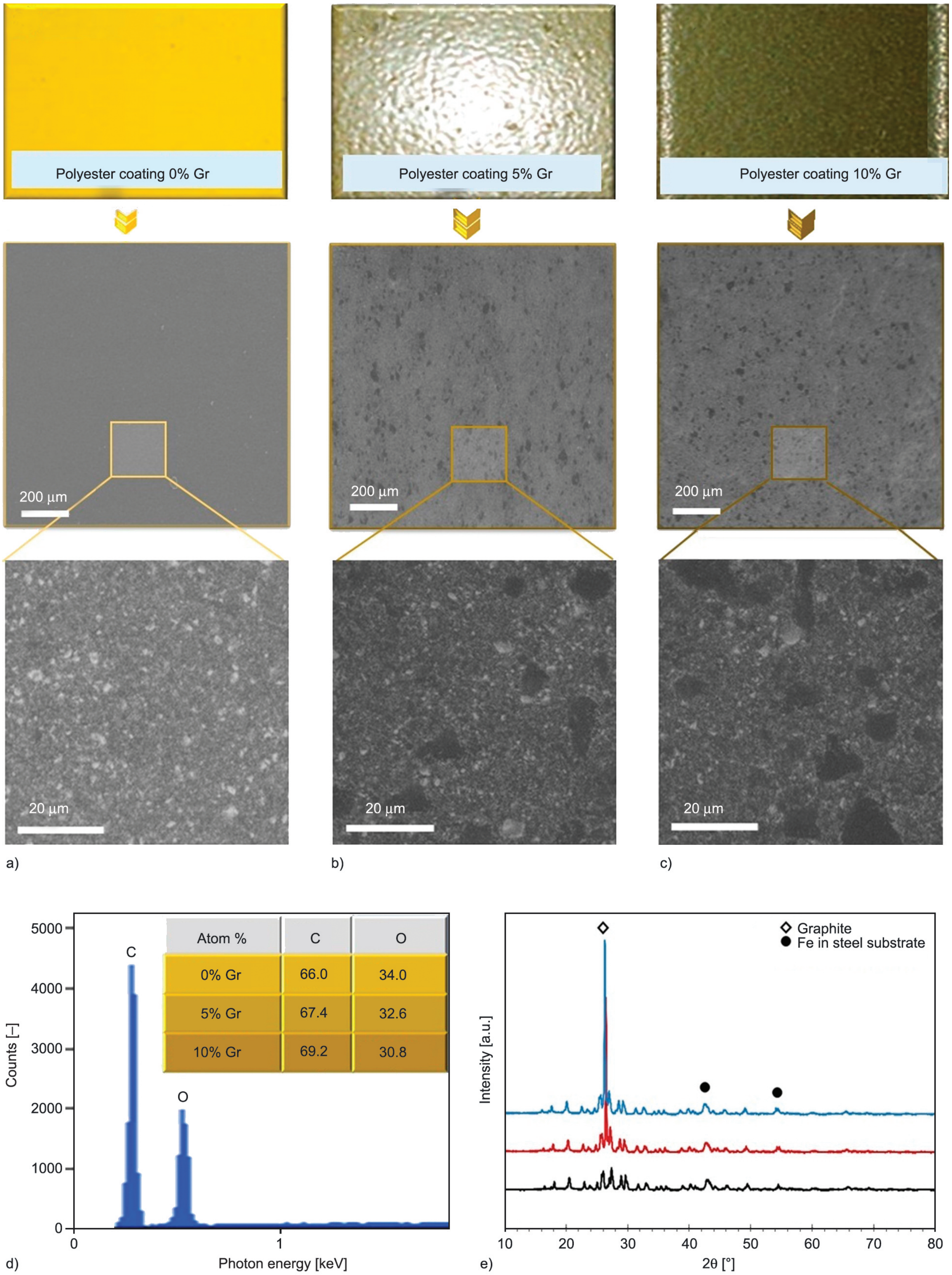
The addition of graphite increased the mechanical properties of the polyester coatings (Figure 3b). The values of hardness H and the elastic modulus E of the samples increased from 235 MPa and $6.76\cdot10^3$ MPa (0% Gr) to 645 MPa and $10.92\cdot10^3$ MPa (10% Gr), respectively (Table 1). These values are consistent with the maximum penetration depth ($h_{\max}$) of indentation on the surface of the 0% Gr-coated sample (1.36 µm) being larger than that of the 10% Gr-coated sample (1.1 µm) (Figure 3a, Table 1). This indicates the improvement of penetration resistance by the addition of Gr. Hence, the addition of graphite-reinforced the polymeric matrix [21].

H/E and H^3/E^2 ratios which were used to assess film resistance to elastic deformation [22], and to predict the resistance to plastic deformation [23], respectively, are illustrated in Figure 3c and Table 1. Results showed that the incorporation of graphite on polyester coating enhanced H/E and H^3/E^2 ratios, which reveals a good resistance to elastic/plastic deformation and good wear resistance.

3.3. Adhesion behavior of polyester/Gr coatings

Scratch tests with progressive load were used to investigate the adhesion behavior of the polyester/Gr composite coatings and to evaluate their scratch resistance (Figure 4). Progression of scratching was accompanied by successive degradations defined by three different critical loads (L_{c1}, L_{c2}, L_{c3}) [24]. The critical loads were determined by combining SEM observations of scratch tracks and the measurements of normal and tangential loads, as well as the penetration and residual depth during the scratch test. The first critical load L_{c1} corresponding to the appearance of the first crack along the scratch pattern was mostly concentrated in its bottom, which indicates a cohesive failure. The second critical load, L_{c2} is the applied normal load at which the extent of fracture events increased both the bottom and the edge of the scratch pattern, which led to the observed repeated tensile cracking (adhesive failure); The third critical load, L_{c3} is the applied normal load at which the coating exhibited a catastrophic failure, and the scratch damage becomes irregular and intense with the removal of the material with partial or complete coating delamination.

Figure 4 reports the trend of tangential force, penetration, and residual depth *vs.* normal force during the scratch test and the related SEM images of the residual scratch tracks of the investigated coatings. A purely elastic deformation took place at low applied scratch loads, no scratch marks were detected along the starting segment of the scratch track for each coating, as shown in Figures 4a, 4b and 4c. By augmenting the normal load, as demonstrated in Figure 4, a sudden slope change in friction load and depth curves related to first cracking phenomena were detected. This cohesive failure (at L_{c1}) is confirmed by SEM



Atom %	C	O
0% Gr	66.0	34.0
5% Gr	67.4	32.6
10% Gr	69.2	30.8

Figure 2. SEM analysis of polyester/Gr coatings: 0 wt% Gr (a), 5 wt% Gr (b), 10 wt% Gr (c); typical EDS spectrum and atomic composition (d), and XRD pattern of coatings (e).

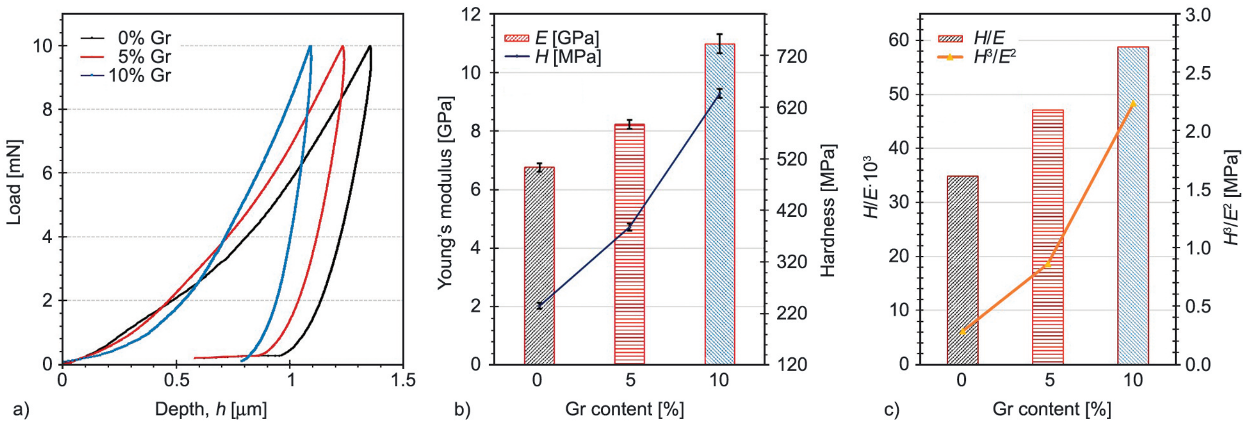


Figure 3. Load-Penetration depth curves of polyester/Gr coatings (a), hardness and elastic modulus (b) and *H*/*E* and H^3/E^2 ratios (c).

Table 1. Nanoindentation values of polyester/Gr coatings.

Sample	*H* [MPa]	*E* [MPa]	*H*/*E*	H^3/E^2 [MPa]	h_{max} [µm]
0% Gr	235.3±5.7	$6.76\cdot10^3\pm0.14$	0.034	0.28	1.36
5% Gr	387.4±6.9	$8.23\cdot10^3\pm0.15$	0.047	0.85	1.23
10% Gr	645.8±8.5	$10.92\cdot10^3\pm0.32$	0.058	2.23	1.10

image (Figure 4a', 4b' and 4c'). When the applied load increased, the track demonstrated a propagation of micro-cracks, which indicates the adhesive failure (at L_{c2}). At the highest critical load L_{c3}, a partial delamination was obtained.

Unfilled polyester coatings showed only cracks and plastic deformation at the end of the scratch track (Figure 4a'), no coating delamination was observed. The adhesive failure took place at L_{c2} equal to 20 N. This result indicates that pure polyester exhibits good adhesion to the steel substrate. The Gr embedded in the polyester matrix reduced the adhesion of composite coatings. The addition of 5 wt% of Gr to the polyester matrix (Figure 4b and 4b') led to the appearance of cracks at a critical force L_{c1} lower than that of the unfilled polyester coatings. In addition, it led to a damage of the coating at the end of the groove L_{c3}. Similar results can be detected with the addition of 10 wt% of Gr to the polyester matrix (Figures 4c and 4c'). Then, a change in the damage mode can be observed by the appearance of parabolic fractures whose density and length increase with the normal force (Figure 4c'). The tensile cracks coatings were relatively large. The critical loads (L_{c1}, L_{c2} and L_{c3}) of polyester coating and its composites are summarized in Table 2.

During the scratch test, when the interfacial shear stresses were accumulated and sufficiently higher than adhesion forces, the delamination of the film occurred [25]. The adhesion strength obtained with the scratch test was calculated using the Equation (1):

$$\sigma_c = \frac{2P_c}{\pi d_c^2}\left[\frac{(4+\vartheta_f)3\pi\mu}{8} - (1-2\vartheta_f)\right] \qquad (1)$$

where P_c is the critical load of spalling, d_c is the critical width of the scratch, μ is the friction coefficient, ϑ_f is the Poisson's ratio of the film. The obtained values of the adhesion strength of coatings are given in Table 2.

For the neat polyester powder coating, only the first two failure events occurred. The addition of Gr to the polyester matrix reduced the critical loads, which could be related to the weakened adhesion at the polyester /graphite interface at high particulate filler content [26]. These results agree with previous work [10]. Despite the reduction of the critical loads of the polyester coating under the addition of Gr, the composite polyester/graphite coatings with high mechanical properties revealed a good and an appropriate adhesion behavior with a cohesive critical load greater than 7 N and an adhesion strength higher than σ_c = ~215 MPa.

3.4. Wear performance of polyester/Gr coatings

A multi-pass sliding scratch test was performed on polyester/Gr coatings to evaluate their friction behavior and wear resistance. The wear tracks obtained

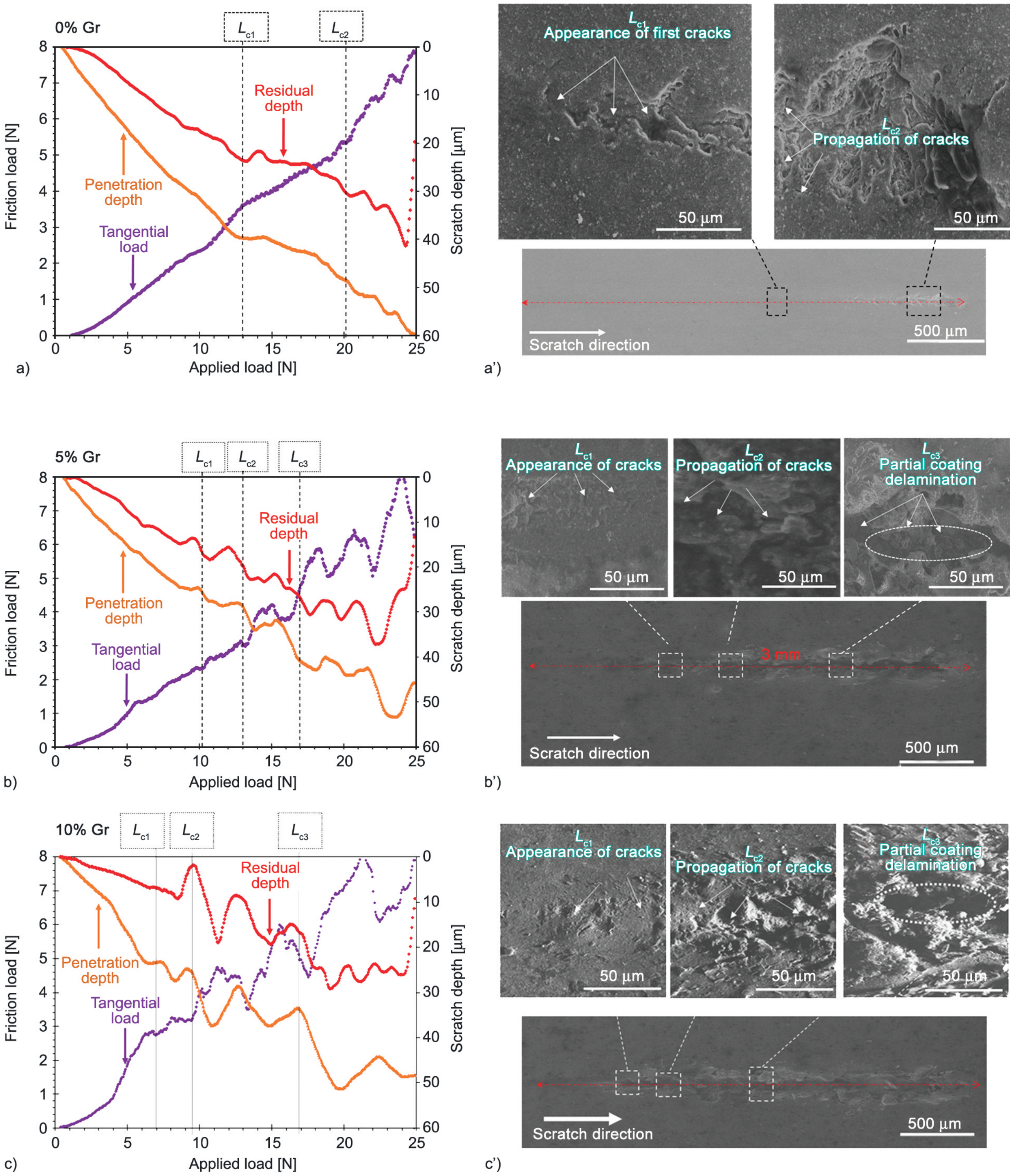


Figure 4. Variation of the tangential load, penetration and residual depths of coatings versus applied load (a, b, c) SEM micrographs of scratch tracks of the coatings (a' b' c').

Table 2. Values of measured critical loads of different polyester/Gr coatings.

Samples	Critical loads [N]			σ_c [MPa]
	When first cracks appeared, L_{c1}	Propagation of cracks, L_{c2}	Delamination, L_{c3}	
0% Gr	13.00±0.17	20.1±0.30	–	944±18
5% Gr	10.10±0.20	13.0±0.10	17±0.25	373±11
10% Gr	7.00±0.13	9.4±0.14	16.7±0.19	215±9

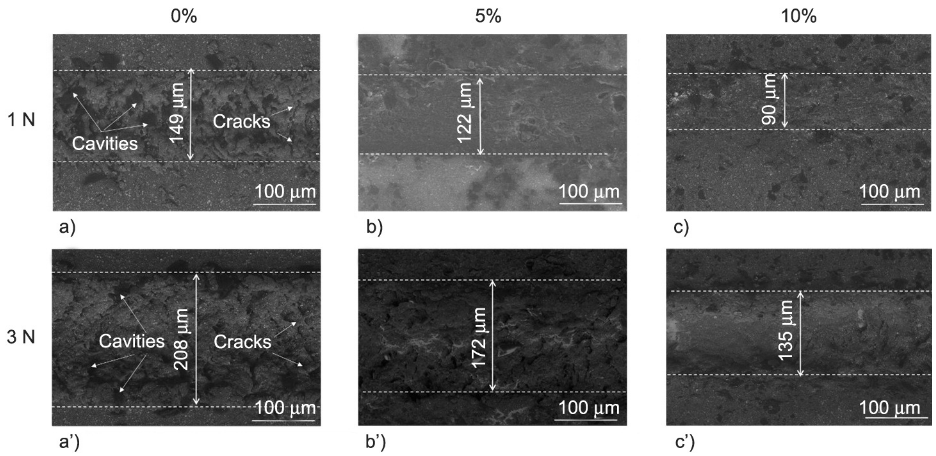


Figure 5. SEM images of the wear track profiles of the polyester/Gr (0, 5 and 10 wt%) coatings at the applied load 1 and 3 N after 100 cycles.

under 100 sliding cycles at constant loads of 1 and 3 N were examined using SEM. The applied load was lower than the cohesive failure load (L_{c1}) of the coatings.

Figure 5 illustrates the SEM images of the damage present on the surfaces of the coatings using loads of 1 and 3 N. For the unfilled polyester coating (Figure 5a and 5a'), the SEM observation revealed significant plastic deformation in the track center and the appearance of extensive cracks located at the edge and the bottom of the wear pattern. In addition, it can be noticed in Figure 5 that the wear track of the 0% Gr coated sample was the widest, and its average width was 149 and 208 µm for 1 and 3 N, respectively.

After the incorporation of 5 wt% Gr, the asperities of the softer surface were easily deformed, and the crack's magnitude located along the wear track decreased (Figure 5b and 5b'). The severity of the initial wear mechanism attenuated in polyester coatings with the increase of the Gr fillers content. After the addition of 10 wt% of Gr (Figure 5c and 5c'), the composite coating became stiff. Hence, the widths of the wear tracks decreased by up to 35% and reached a value of 90 and 135 µm for 1 and 3 N, respectively. Similar results were found by Shah *et al.* [27], authors have demonstrated that the addition of graphene enhances the frictional and wear properties of epoxy resin.

The possible wear mechanism of polyester/Gr coatings deposited on steel substrates is shown in a schematic diagram in Figure 6. Two different scratch failure mechanisms can be proposed and schematically described. Under normal load, the unfilled polyester coatings were easily plastically deformed and susceptible to cracking. This phenomenon led to the formation of large cracks and cavities inducing chipping, which led to adhesive wear mechanisms. This

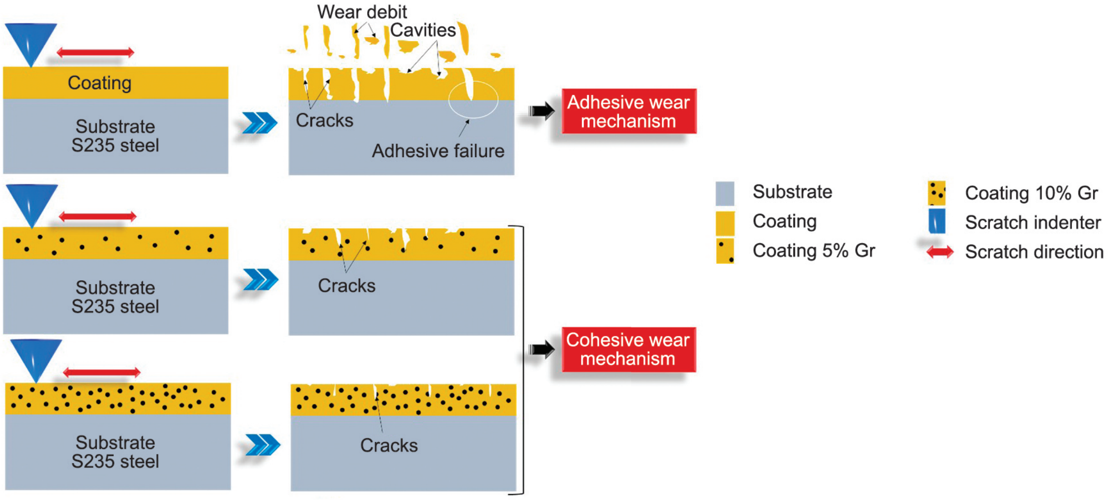


Figure 6. Schematic representation of the possible wear mechanisms of polyester/Gr coatings after wear test.

wear process evolves in the formation of adhesion junction, its growth and fracture [28].

The addition of Gr fillers to polyester coatings acted as a lubricating layer to protect the substrates from serious wear. As a result, the SEM image showed limited cracks and some cavities that produce only cohesive wear.

The results of wear tests in terms of static friction coefficient and wear volume are illustrated in Figure 7. The evolution of the static friction coefficients of the coatings is presented as a function of the number of cycles for 1 and 3 N in Figure 7a and Figure 7b, respectively. The presence of Gr caused the decrease of the average static friction coefficient. The addition of 5 wt% Gr to the polyester matrix reduced the static friction coefficient from 0.485 to 0.232 at the two applied loads. A similar result was obtained for the 10% Gr coatings at the applied load of 1 N. For a load of 3 N, a drop in the static friction coefficient from 0.485 in the case of net polyester to a value of 0.3 was noticed when the mass fraction of Gr was 10%. After 100 sliding cycles, the evolution of the static friction coefficient and wear volume of the thermosetting composites powder coatings vs. loads of 1 and 3 N is reported in Figure 7c and Figure 7d, respectively. It can be noticed that the incorporation of Gr particles to the polyester matrix resulted in a more significant static friction coefficient and wear volume reduction. Consequently, the Gr enhanced the wear resistance of the polyester coating. The volume loss of the polyester coating decreased with the increase of the amount of Gr. The composite coated samples were able to distribute and absorb the load since their surfaces were flexed. Therefore, they exhibited self-lubrication and low wear. which is in perfect agreement with the results of the SEM of wear track (Figure 5).

Consequently, it can be concluded that the surfaces acquired self-lubricating properties leading to an increase in the wear resistance of the polyester coating. This finding shows a perfect agreement with the previous hardness results (Figure 3). As a hard phase in the soft polymer matrix, graphite powders can reduce the plastic deformation of the coating.

3.5. Anti-corrosion performance of polyester/Gr coatings

The dynamic potential polarization curves of different coatings immersed in 3.5 wt% NaCl solution for 1 hour are shown in Figure 8, and the specific data (Table 3) are obtained by Tafel fitting of these curves. The corrosion resistance of the coating can be described in detail by the electrodynamic polarization curve, and the lower polarization current indicates the better corrosion resistance [29].

The corrosion potentials (E_{corr}) and the corrosion current densities (i_{corr}) values of the steel were –0.739 V *vs*. SCE and 1620 μA·cm^{-2}, respectively.

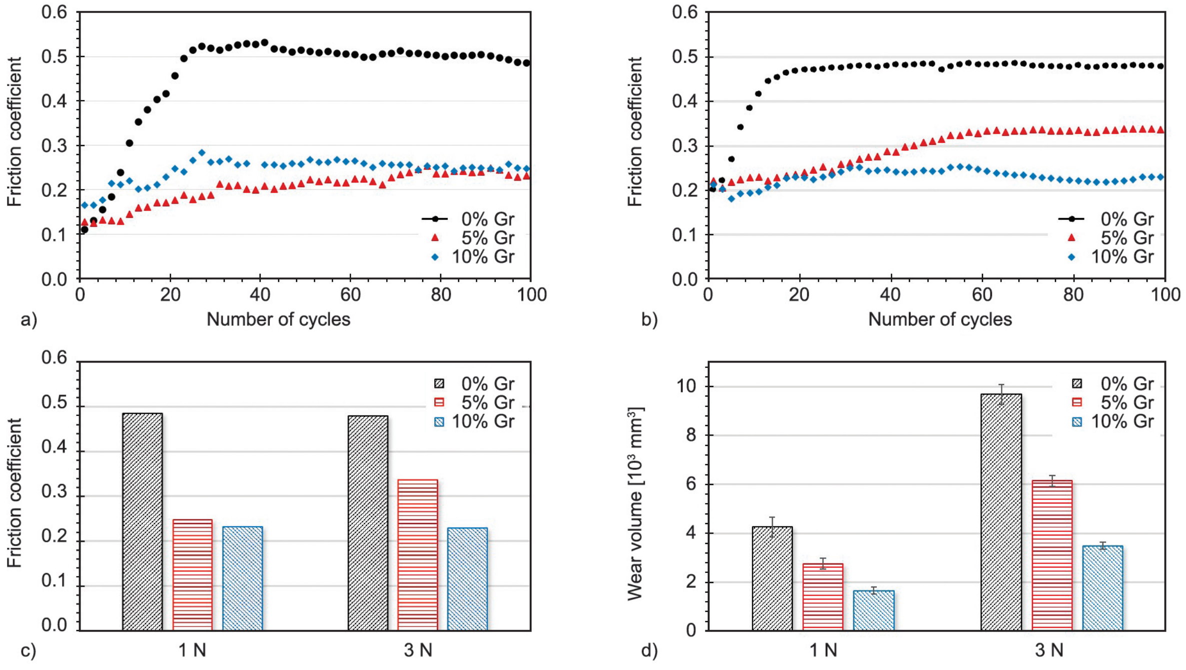


Figure 7. Static friction coefficient plot for a load of 1 N (a) and 3 N (b) of polyester/Gr coatings, Bar graph of static friction coefficient (c) and wear volume (d) at load 1 and 3 N after 100 passes of polyester/Gr coatings.

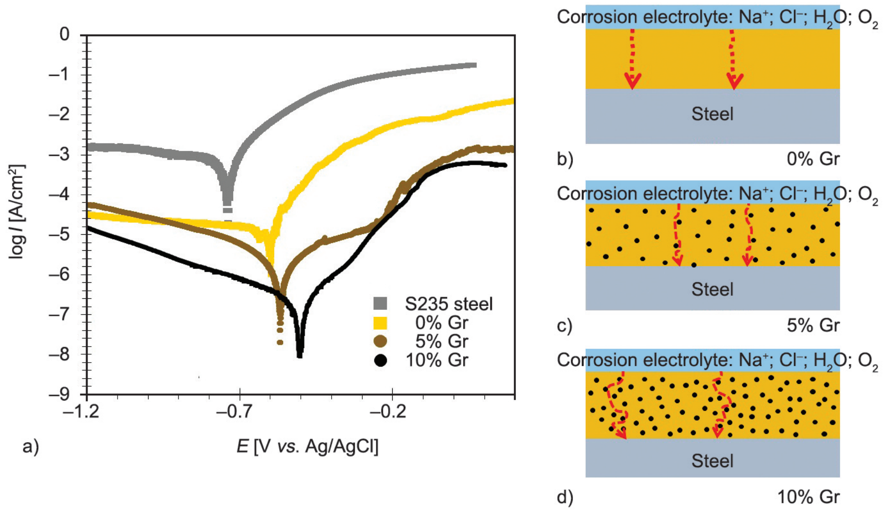


Figure 8. Potentiodynamic polarization curves of steel and coatings in 3.5 wt% NaCl solution (a) and the corrosion protection mechanism of (b) 0 wt% Gr, (c) 5 wt% Gr and (d) 10 wt% Gr.

Table 3. Electrochemical parameters for steel (substrate) and polyester/Gr coatings obtained from polarization curves.

Coatings	E_{corr} [V]	i_{corr} [µA/cm²]	V_{corr} [µm/y]
Uncoated steel (substrate)	0.739	1620.000	7890.00
Coated steel with Polyester/0 wt% Gr	0.603	13.100	95.00
Coated mild steel with Polyester/5 wt% Gr	0.567	1.200	12.12
Coated mild steel with Polyester/10 wt% Gr	0.492	0.129	1.45

The deposition of PS coatings enhanced E_{corr} to –0.603 V and reduced i_{corr} to 13.1 µA·cm^{-2}. The coatings with 5 and 10 wt% Gr significantly enhanced E_{corr} to –0.567 and –0.492 V, and reduced i_{corr} to 1.2 and 0.129 µA·cm^{-2}, respectively. This indicates that the graphite particles dispersed in the polyester coating could significantly retard the penetration of corrosive ions to the metal matrix, thus greatly improving the corrosion resistance. Therefore, the coating with 10 wt% of graphite is of the lowest corrosion velocity (1.45 µm·y^{-1}).

Figures 8b–8d illustrate the corrosion protection mechanism of unfilled and filled polyester coatings by Gr particles. For pure polyester coating (Figure 8b), corrosive electrolytes can easily pass through the micropores onto the steel surface and form corrosion products. Thus, reducing the corrosion resistance of the coating.

The graphite can slightly block the penetration of corrosive ions to the steel surface due to stacked structure and the large particle size (Figure 8c and 8d). Consequently, the barrier effect was greatly enhanced by the graphite addition for better corrosion resistance.

4. Conclusions

In this study, protective polyester-based composite coatings were developed and applied on structural steel S235 substrate using the electrostatic powder spraying process. The influence of graphite content (5 and 10 wt%) on the mechanical, adhesion, tribological and corrosion performance of polyester coatings was investigated by nano-indentation, scratch test, SEM and Cyclic Voltammetry. The following conclusions are drawn from the experimental results:

– The composite coatings exhibited a compact structure consisting of graphite uniformly dispersed in polyester matrix.
– The nano-indentation test showed that the graphite content enhances the hardness and the Young's modulus of polyester coating.
– Good resistance to adhesive and cohesive failure was found for the polyester coatings and their composites filled with graphite. The incorporation of graphite until 10% reduces the critical loads of polyester coatings, but the adhesion was satisfied.
– The coated steel surfaces acquired self-lubricating properties due to the addition of graphite, leading

to an increase in the wear resistance and a significant reduction of the static friction coefficient.
- After the wear tests, the severity of wear mechanisms of polyester coatings with the incorporating graphite was attenuated and passed from adhesive wear to cohesive wear.
- The corrosion protection ability of the polyester coatings was significantly improved by the incorporation of graphite due to its barrier role.

Acknowledgements

The authors would like to thank 'Mr. Yassine Zoughlami' the general manager of society 'Peinture industrielle en poudre epoxy' of Marsa Tunisia for the successful deposition of polyester/graphite coatings on steel substrates and for his technical assistance.

References

[1] Aguiari M., Palombo M., Rizzo C. M.: Performance characterization of high-strength steel and quenched and tempered steels and their joints for structural applications. Weld World, **65**, 289–300 (2021). https://doi.org/10.1007/s40194-020-01019-6

[2] Nguyen D. T., To H. T. X., Gervasi J., Paint Y., Gonon M., Olivier M. G.: Corrosion inhibition of carbon steel by hydrotalcites modified with different organic carboxylic acids for organic coatings. Progress in Organic Coatings, **124**, 256–266 (2018). https://doi.org/10.1016/j.porgcoat.2017.12.006

[3] Ding J., ur Rahman. O., Peng W., Dou H., Yu H.: A novel hydroxyl epoxy phosphate monomer enhancing the anticorrosive performance of waterborne graphene/epoxy coatings. Applied Surface Science, **427**, 981–991 (2018). https://doi.org/10.1016/j.apsusc.2017.08.224

[4] Alhumade H., Yu A., Elkamel A., Simon L., Abdala A.: Enhanced protective properties and UV stability of epoxy/graphene nanocomposite coating on stainless steel. Express Polymer Letters, **10**, 1034–1046 (2016). https://doi.org/10.3144/expresspolymlett.2016.96

[5] Luévano-Cabrales O. L., Alvarez-Vera M., Hdz-García H. M., Muñoz-Arroyo R., Mtz-Enriquez A. I., Acevedo-Dávila J. L., Hernandez-Rodriguez M. A. L.: Effect of graphene oxide on wear resistance of polyester resin electrostatically deposited on steel sheets. Wear, **426**, 296–301 (2019). https://doi.org/10.1016/j.wear.2019.01.083

[6] Cho D-H., Bhushan B.: Friction and wear of various polymer pairs used for label and wiper in labeling machine. Tribology International, **98**, 10–19 (2016). https://doi.org/10.1016/j.triboint.2016.02.019

[7] Bjerremand C., Larsen J., Hinge M.: Temperature- and time dependency on high friction poly(styrene-*co*-butyl methacrylate) coated paper. Tribology International, **103**, 261–265 (2016). https://doi.org/10.1016/j.triboint.2016.07.006

[8] Laux K. A., Jean-Fulcrand A., Sue H. J., Bremner T., Wong J. S. S.: The influence of surface properties on sliding contact temperature and friction for polyetheretherketone (PEEK). Polymer, **103**, 397–404 (2016). https://doi.org/10.1016/j.polymer.2016.09.064

[9] Vinayagamoorthy R.: Friction and wear characteristics of fibre-reinforced plastic composites. Journal of Thermoplastic Composite Materials, **33**, 828–850 (2020). https://doi.org/10.1177/0892705718815529

[10] Zouari M., Kharrat M., Dammak M.: Wear and friction analysis of polyester coatings with solid lubricant. Surface and Coatings Technology, **204**, 2593–2599 (2010). https://doi.org/10.1016/j.surfcoat.2010.02.001

[11] Larsen T. Ø., Andersen T. L., Thorning B., Horsewell A., Vigild M. E.: Changes in the tribological behavior of an epoxy resin by incorporating CuO nanoparticles and PTFE microparticles. Wear, **265**, 203–213 (2008). https://doi.org/10.1016/j.wear.2007.10.003

[12] Suresha B., Chandramohan G., Renukappa M. N., Siddaramaiah: Mechanical and tribological properties of glass–epoxy composites with and without graphite particulate filler. Journal of Applied Polymer Science, **103**, 2472–2480 (2007). https://doi.org/10.1002/app.25413

[13] Chang L., Zhang Z., Zhang H., Friedrich K.: Effect of nanoparticles on the tribological behaviour of short carbon fibre reinforced poly(etherimide) composites. Tribology International, **38**, 966–973 (2005). https://doi.org/10.1016/j.triboint.2005.07.026

[14] Sakka M. M., Antar Z., Elleuch K., Feller J. F.: Tribological response of an epoxy matrix filled with graphite and/or carbon nanotubes. Friction, **5**, 171–182 (2017). https://doi.org/10.1007/s40544-017-0144-z

[15] Friedrich K.: Polymer composites for tribological applications. Advanced Industrial and Engineering Polymer Research, **1**, 3–39 (2018). https://doi.org/10.1016/j.aiepr.2018.05.001

[16] Bora C., Bharali P., Baglari S., Dolui S. K., Konwar B. K.: Strong and conductive reduced graphene oxide/polyester resin composite films with improved mechanical strength, thermal stability and its antibacterial activity. Composite Science and Technology, **87**, 1–7 (2013). https://doi.org/10.1016/j.compscitech.2013.07.025

[17] Avilés M. D., Saurín N., Carrión F. J., Arias-Pardilla J., Martínez-Mateo I., Sanes J., Bermúdez M. D.: Epoxy resin coatings modified by ionic liquid. Study of abrasion resistance. Express Polymer Letters, **13**, 303–310 (2019). https://doi.org/10.3144/expresspolymlett.2019.26

[18] Katiyar J. K., Sinha S. K., Kumar A.: Friction and wear durability study of epoxy-based polymer (SU-8) composite coatings with talc and graphite as fillers. Wear, **362**, 199–208 (2016). https://doi.org/10.1016/j.wear.2016.06.011

[19] Khlifi K., Dhiflaoui H., Aissa C. B., Barhoumi N., Larbi A. B. C.: Friction and wear behavior of a physical vapor deposition coating studied using a micro-scratch technique. Journal of Engineering Materials and Technology, **144**, 021002 (2022). https://doi.org/10.1115/1.4052239

[20] Oliver W. C., Pharr G. M.: An improved technique for determining hardness and elastic modulus using load and displacement sensing indentation experiments. Journal of Materials Research, **7**, 1564–1583 (1992). https://doi.org/10.1557/JMR.1992.1564

[21] Baptista R., Mendão A., Rodrigues F., Figueiredo-Pina C. G., Guedes M., Marat-Mendes R.: Effect of high graphite filler contents on the mechanical and tribological failure behavior of epoxy matrix composites. Theoretical and Applied Fracture Mechanics, **85**, 113–124 (2016). https://doi.org/10.1016/j.tafmec.2016.08.013

[22] Affonso L. O. A.: Machinery failure analysis handbook: Sustain your operations and maximize uptime. Gulf Publishing Company, Houston (2008).

[23] Tsui T. Y., Pharr G. M., Oliver W. C., Bhatia C. S., White R. L., Anders S., Brown I. G.: Nanoindentation and nanoscratching of hard carbon coatings for magnetic disks. MRS Online Proceedings Library, **383**, 447–452 (1995). https://doi.org/10.1557/PROC-383-447

[24] Barletta M., Gisario A., Rubino G.: Scratch response of high-performance thermoset and thermoplastic powders deposited by the electrostatic spray and 'hot dipping' fluidised bed coating methods: The role of the contact condition. Surface and Coatings Technology, **205**, 5186–5198 (2011). https://doi.org/10.1016/j.surfcoat.2011.05.032

[25] Huang Y-C., Chang S-Y., Chang C-H.: Effect of residual stresses on mechanical properties and interface adhesion strength of SiN thin films. Thin Solid Films, **517**, 4857–4861 (2009). https://doi.org/10.1016/j.tsf.2009.03.043

[26] Hu H., Zhao S., Sun G., Zhong Y., You B.: Evaluation of scratch resistance of functionalized graphene oxide/polysiloxane nanocomposite coatings. Progress in Organic Coatings, **117**, 118–129 (2018). https://doi.org/10.1016/j.porgcoat.2018.01.002

[27] Shah R., Datashvili T., Cai T., Wahrmund J., Menard B., Menard K. P., Brostow W., Perez J.: Effects of functionalised reduced graphene oxide on frictional and wear properties of epoxy resin. Materials Research Innovations,**19**, 97–106 (2015). https://doi.org/10.1179/1433075X14Y.0000000220

[28] Myshkin N. K., Petrokovets M. I., Kovalev A. V.: Tribology of polymers: Adhesion, friction, wear, and mass-transfer. Tribology International, **38**, 910–921 (2005). https://doi.org/10.1016/j.triboint.2005.07.016

[29] Kumar A., Ghosh P. K., Yadav K. L., Kumar K.: Thermo-mechanical and anti-corrosive properties of MWCNT/epoxy nanocomposite fabricated by innovative dispersion technique. Composites Part B: Engineering, **113**, 291–299 (2017). https://doi.org/10.1016/j.compositesb.2017.01.046