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Original Research Article Characterization of Tribological and Electrochemical Corrosion Behavior of GO Coatings Formed on 6061 Al Alloy Processed by Plasma Electrolytic Oxidation

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ABSTRACT

Plasma electrolytic oxidation (PEO) is an electrochemical surface treatment process that is used to improve the properties of metal parts and components. The coating can provide a range of beneficial properties, such as increased hardness, wear resistance, and corrosion resistance, as well as improved adhesion and durability. Plasma electrolytic oxidation (PEO) improves aluminum alloy properties and adding nanoparticles to the electrolyte can further improve the surface properties, resulting in improved surface hardness, enhanced corrosion resistance, faster and more efficient process, and tailored surface properties. Graphene oxide possesses unique properties such as high mechanical strength, excellent thermal conductivity, and remarkable lubricating abilities, which make it a promising candidate for improving tribological behavior. This study aims to examine the potential advantages of incorporating graphene oxide into coatings produced through the electrolytic plasma oxidation process.

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1. INTRODUCTION

Aluminum alloys are used in a wide variety of applications due to their excellent combination of strength, durability, and light weight. The production of new aluminum alloys is critical to the development of new technologies and the improvement of existing ones. In this article, we will explore the different techniques used to produce aluminum alloys and how they impact the final properties of the alloy [1, 2]. Ceramic coatings are widely used in industry because they provide better protection against wear and corrosion. The process used to create these coatings, called plasma electrolytic oxidation (PEO), involves immersing a metal substrate in a special liquid and applying a high voltage. This creates a plasma discharge that forms a unique oxide layer on the metal surface [3-5]. Plasma electrolytic oxidation (PEO) is a surface treatment that can improve the properties of aluminum alloys. Incorporating nanoparticles into the electrolyte during PEO can further

enhance the surface properties of the aluminum alloy. Here are some of the advantages of PEO containing nanoparticles on aluminum alloy [5]: Incorporating nanoparticles into the PEO process can result in a hard, dense, and wear-resistant surface on the aluminum alloy. The nanoparticles can penetrate the surface of aluminum alloy and improve the surface hardness, which can increase the durability of the material [6]. The incorporation of nanoparticles into the electrolyte during PEO can improve the corrosion resistance of aluminum alloy. This is due to the formation of a compact, uniform, and protective oxide layer that can prevent the penetration of aggressive species into the aluminum alloy [6-8]. PEO containing nanoparticles can result in a faster and more efficient process compared to traditional PEO [9, **10**]. The nanoparticles can enhance the anodic reaction rate and increase the electrical conductivity, resulting in a faster and more uniform coating formation [11]. The addition of nanoparticles can modify the surface properties of the aluminum alloy, such as the surface roughness and hydrophobicity [12]. This can be beneficial for specific applications where tailored surface properties are required. Therefore, PEO containing nanoparticles can improve the surface hardness, enhance corrosion resistance, result in a faster and more efficient process, and allow for tailored surface properties on aluminum alloy [13].

Graphene oxide nanosheets are a type of material that has become increasingly popular in recent years due to their unique properties [14-15]. Graphene oxide is a derivative of graphene, which is a single layer of carbon atoms arranged in a honeycomb lattice. Graphene oxide is essentially graphene with oxygen-containing functional groups attached to the surface of the carbon lattice [16, 17]. Graphene oxide (GO) nanosheets have significant advantages in plasma electrolytic oxidation (PEO) [18, 19]. One advantage is improved corrosion resistance. The use of GO in the PEO electrolyte results in a coating that has higher density, fewer defects, and is more uniform than traditional PEO coatings, leading to improved corrosion resistance [20,21]. In addition, GO nanosheets improve coating adhesion, resulting in more durable and reliable coatings, leading to reduced manufacturing costs in terms of maintenance and repair. Another benefit of using GO in PEO is enhanced electrical conductivity [20-23]. The superior electrical conductivity of GO allows for shorter processing times and more consistent and uniform coatings. GO nanosheets also improve wear resistance and the durability of treated metal surfaces in PEO. Finally, the utilization of GO in PEO can reduce the environmental footprint of the process, as it reduces the need for toxic and hazardous chemicals and improves the process efficiency.

The use of GO in PEO on aluminum alloy 6061 can lead to coatings with improved corrosion

resistance, wear resistance, and mechanical properties. The GO-containing coating has enhanced adhesion, leading to better corrosion resistance and wear resistance. The GO incorporation also improves the mechanical properties of the coating, enhancing its hardness and wear resistance. Moreover, the presence of GO in the PEO process leads to the formation of a more uniform coating, which could improve the consistency of the surface treatment, especially in complex geometries. Overall, the modification of aluminum alloy 6061 with GO by PEO provides a promising route to improve the surface properties of the material. Accordingly, the main approach of this study is to investigate the electrochemical and terbiological properties of aluminum 6061 reinforced with graphene oxide nanosheets.

2. EXPERIMENTAL

2.1. Materials and methods

The materials used in this research are potassium permanganate (Merck, Germany), sulfuric acid 80%, phosphoric acid 90%, and hydrochloric acid 37% (Amertat shimi, Iran). Other consumables are of high purity and any additional preparation. without The µAutolab Type III/FRA2 apparatus with a threeelectrode system was used to accurately assess the corrosion resistance of the coated and uncoated samples. This apparatus allows for precise control and monitoring the of electrochemical reactions occurring at the surface of the working electrodes. The reference electrode used in this setup was Ag/AgCl, which is commonly employed in electrochemical measurements due to its stable potential. It was immersed in a saturated KCl solution to maintain a constant and known reference potential throughout the experiment. The auxiliary electrode, made of platinum, served as a counter electrode. The working electrodes (coated and uncoated samples) were subjected to different

surface treatments or coatings to evaluate their effectiveness in enhancing corrosion resistance. The reference electrode was Ag/AgCl (in saturated KCl solution), auxiliary electrode was platinum, working electrodes were coated and uncoated samples, and corrosion medium was 3.5 wt% NaCl solution, pH 7. In preparation for the electrochemical measurements, the working electrodes were placed in 3.5 wt% NaCl for an interval of 3600 s while being exposed to open circuit potential conditions. Potentiodynamic polarization curves were achieved by using a scan rate of 1 mVS-1 and starting from the corrosion potential (Ecorr) at a voltage of -0.25 V. By applying AC potential with amplitude of 10 mV and varying the frequency range between 100 kHz and 63 MHz, EIS tests were performed at open-circuit potential conditions. Using a spark emission spectrometer (SES) model 4100 Series MP-AES by Agilent Technologies, the chemical composition in the base was measured and is presented in Table 1. Using the 7000 XRD diffractometer from Shimadzu Company (Japan) with an angle ranging from 10 to 110 degrees, we acquired the phase composition of the coatings. To identify the bonds formed at a wavelength of 400 to 4000 cm⁻¹, FTIR analysis was conducted on the acquired coating utilizing an infrared spectrometer from the IRAffinity-1S series produced by Japanese company Shimadzu. The EVO model scanning electron microscope by ZEISS was chosen for its high-resolution imaging capabilities, allowing for detailed examination of the surface morphology and sections of the samples. This microscope utilizes electron beams to scan the sample's surface, providing a magnified view and enabling the analysis of microstructural features. The BRUKER atomic force microscope, ICON model, was selected to assess the topography and wear level of the samples. This instrument uses a sharp probe to scan the sample's surface, measuring forces between the probe and the material. It provides three-dimensional images with nanometer-scale

resolution, allowing for precise characterization of surface roughness and wear patterns. To determine the samples' hardness, a Vickers NOVOTEST TB-MCV-1 metal microhardness tester was employed. This tester applies a load of 500 grams to create an indentation on the sample's surface. By conducting the WTC02 pin/ball on disk wear test using the machine produced by Tajhzih Sanat Nasr Company, under controlled laboratory conditions, the tribological properties of the samples can be evaluated. The applied load, rotation speed during and sliding distance was set at 4 N, 50 rpm, and 150 m. Specific wear rate is calculated according to Equation (1):

Ws=m/(d×F) (1) The specific wear rate, denoted as Ws in the given formula, represents the rate at which mass is worn away and is measured in milligrams. The amount of worn mass, represented by m, is measured in milligrams. In addition, the applied force, denoted as F, is measured in Newtons whereas the distance traveled, represented by d, is measured in meters.

2.2. Method

The use of an alloy as the anode and a cylinder as the cathode in this system suggests that there is an electrochemical process taking place. The specific choice of materials for the anode and cathode would depend on the desired reaction and the properties of the alloy being used. The electrolyte solution, which consists of Na₂SiO₃ (5 g), CaF₂ (3 g), and KOH (2 g) dissolved in distilled water (1000 mL) with a pH of 12.55, plays a crucial role in facilitating the electrochemical reaction. Each component likely serves a specific purpose in enhancing the efficiency or selectivity of the process. After stirring and sonication (for 4 h), the solution was carefully transferred into a clean glass container to remove any impurities or non-dissolved particles. This ensured а homogeneous mixture, free from any clumps or sedimentation. Next, the container was placed on

a hot plate and heated to a specific temperature, depending on the experiment requirements. The heating process helped in further dissolving any remaining solid particles and facilitated the formation of a clear and transparent solution. The PEO-800300 device, made by Mirab Sanat, was selected for its ability to coat the 6061 Al alloy sample effectively. It uses a pulsed plasma oxidation process, subjecting the sample to highenergy plasma discharges in a controlled environment. Fig 1(a and b) provides details about the design and features of the device. To carry out the coating experiment, carefully prepared aluminum alloy samples with dimensions of 40x5x40 mm were used. During the pulsed plasma oxidation process, the aluminum alloy sample served as the base metal or anode (as illustrated in Fig 1 (c)). To apply a coating to the aluminum alloy sample, the PEO-800300 model pulsed plasma oxidation device, manufactured by Mirab Sanat Company, was utilized (as depicted in Fig 1 (a and b)). The base metal used for the samples was aluminum alloy 6061, from which samples with dimensions of 40x5x40 mm were prepared (as shown in Fig 1(c). A direct current source with a maximum potential of 700 volts and a current of 50 amperes was employed, along with a pulser

designed based on an isolated gate bipolar transistor, which had the capability to convert direct current to rectifying bipolar and provide the necessary energy for coating. Table 1 presents the device specifications included a pulse current source for electrolytic plasma oxidation, with the ability to apply positive and negative voltage and controllable current. The surface preparation of the samples involved polishing the surfaces with sandpaper, followed by exposing the sample to radiation in a solution of distilled water and acetone for 30 minutes in an ultrasonic device, to remove any organic pollutants.

2.3. Graphene oxide synthesis by improved Hummers method

In this method, which aims to produce stable graphite oxide, potassium permanganate, and phosphoric and sulfuric acids are used. Initially, sulfuric acid and phosphoric acid are combined in a 9:1 ratio, followed by the gradual addition of graphite powder while stirring until a homogeneous black mixture is achieved. Subsequently, the mixture is subjected to an ice bath, and potassium permanganate is slowly added while stirring for 15 minutes, with the

					P-100-110				
Characteristic			Pulsed plasma oxidation						
Output type			Constant voltage and current intensity						
Flow type			Bipolar pulse						
Voltage				-300 to +800 volts					
Frequency				50-2000 Hz					
Duty cycle			15%-85%						
Output power			10 kw						
input voltage			3 phase + zero 380 volts						
Table 2. Chemical composition of aluminum alloy 6061 (mass percentage)									
Fe	Cu	Si	Zn	Mn	Mg	Cr	Ti	Al	
0.16	0.19	0.71	0.04	0.02	0.94	0.08	0.03	Bal.	

Table 1. Technical specifications of the pulsed plasma oxidation device



Fig1. (a) Schematic image of plasma electrolytic oxidation, (b) The plasma electrolytic oxidation device, and (c) Dimensions of aluminum alloy prepared for plasma electrolytic oxidation

temperature maintained below 20 °C. After complete dissolution of permanganate, the mixture is transferred to an incubator at 50 °C and stirred for 12 hours, and then the mixture is cooled to room temperature and ice is added to it. Following a brief period of ice addition, hydrogen peroxide (30% by weight) was gradually introduced to the mixture while stirring to halt the oxidation process and diminish the residual permanganate. This process resulted in a light brown/orange coloration. Subsequently, the mixture was subjected to centrifugation, and the supernatant was discarded. The sediment was washed twice with hydrochloric acid, and then with water. Dialysis against water was conducted for one week to eliminate metal ions, sulfate, and chlorine. The aqueous dispersion or sol of graphene oxide, which was obtained through the modified Hamers method, involved the oxidation of graphite, and yielded a single layer with a thickness ranging from 1.4 to 0.7 nm in an

aqueous polar solvent with a concentration of 0.9% by weight. Formula 2 was utilized to determine the percentage of weight change. Specifically, M1 represented the weight of the sample prior to coating, M2 represented the sample weight after coating, and $\%\Delta W$ represented the percentage of sample weight changes.

$$\%\Delta W = (M2 - M1)/M1 \times 100$$
 (2)

3. RESULTS AND DISCUSSION

3.1. Studying the changes of flow-time diagram

The present study aimed to investigate the changes in the flow-time diagram of plasma oxidized alloy samples in the electrolyte with and without graphene oxide. (Fig 2) displays the current-time diagram for the samples with a current density of 7.2 A/dm². The evolution of the current during the formation of anodic film in the constant potential state includes three distinct regions. Region 1 of the diagram



Fig 2.Voltage-time curve of PEO coating in a solution containing 0 and 25 g/L of graphene oxide at a current density of 7.2 A/dm²

represents the anodizing region, where a sharp increase in current density is observed, and a thin passive film with high resistance begins to form on the surface. Before dielectric breakdown and plasma formation, aluminum releases some oxygen bubbles from the surface. In this area, no spark is observed, and the outer surface of the metal is covered with a thin layer of insulation, indicating the normal anodizing process. Over time, the cell voltage reaches a voltage called the breakdown voltage.

At the beginning of region 2, due to the luminescence phenomenon, tiny sparks appeared on the surface of the anode, and current fluctuations caused the formation of a porous layer. In this step, cell voltage increases continuously and with a lower slope. The breakdown current of the anode film is obtained when the current values reach a critical level to create an avalanche of electric current. After passing through the normal anodizing process, the increase of electric field in the created layer and the sound emission indicate a more intense release of oxygen gas in the anode. In this region, assuming that the electrolyte resistance is constant during the process, the system resistance can be considered equivalent to the coating resistance. Therefore, the decrease in current density in the electrolyte containing graphene oxide is due to the increase in coating resistance. This is due to the higher electrical conductivity of the electrolyte containing graphene oxide, as the increase in electrical conductivity causes a sharp decrease in the output current density. In region 3, despite the coating resistance causing the anode current to reach approximately 7.2 A/dm², ignition continues to occur in a constant and uniform manner due to the cathodic current. The size of the sparks decreases as a result of the increase in voltage, energy, and their density on the metal surface. The breakdown voltage of the sample coated with graphene oxide is lower than that of the sample without graphene oxide, which may be attributed to the increase in electrolyte conductivity. The final cell voltage of the graphene oxide-coated samples is lower than that of the uncoated samples, potentially due to the reduction of spark energy in the presence of graphene oxide. A higher final voltage corresponds to a coating with higher porosity and lower electrical resistance. The coatings

thickness is reduced due to the reduction of spark energy and the final cell voltage. In this area, the ignition intensity of the graphene oxide-10 sample is higher than that of the plasmaoxidized alloy without graphene oxide. In a general analysis, it can be posited that graphene oxide in the electrolyte impedes the absorption of anions on the surface of the sample, thereby preventing the occurrence of a critical voltage or current between the substrate and the electrolyte. In other words, the final reactions are more intense, and the presence of graphene oxide particles increases the ions consumption, leading to a reduction of ions in the electrolyte. Consequently, the intensity of the current increases significantly, necessitating more current to maintain the balance of the electric field. Meanwhile, the occurrence of electron avalanche improves the dielectric breakdown. Overall, the presence of graphene oxide can obstruct the tiny pores in the areas where the melt caused by critical sparks emerges, preventing the loss of the coating.

3.2. Study of the morphology of the samples

The transmission electron microscopic image of graphene oxide is presented in (Fig 3a), while the scanning electron microscope images of the samples surface are demonstrated in (Fig 3b). The morphology of the alloy surface is characterized by parallel lines and small pits, which were created during the surface preparation stage using sandpaper. Microscopic images of the sample surface without graphene oxide after 100 and 300 seconds are indicated in Fig 3(c and d), revealing a smooth surface with scattered micro-holes of different sizes (pancake structure) surrounded by relatively smooth quasi-circular regions of the refrozen coating. The morphology of the outer layer remains unchanged over time, with only the holes becoming smaller and more numerous. (Fig 3e and f) illustrates the surface morphology of the sample in the solution containing 10 and 25 g/L

graphene oxide after 300 seconds of the process. The morphology of the created surface is completely volcanic, with samples containing graphene oxide completely covering the holes surface. The high current density during the coating process causes stronger arcs on the sample surface, changing the structure from pancake to volcano. The surface morphology of the coatings is characterized by microholes, which are typical of plasma electrolytic oxidation coatings. The very high temperature resulting discharge phenomenon from the causes aluminum to melt from the substrate, and the resulting melt is oxidized by the oxygen in the electrolyte, turning into aluminum oxide while leaving the discharge channels due to the strong electric field. The rapid freezing of molten aluminum oxide in contact with the electrolyte that comes out of the discharge channels causes the formation of pancake structures and volcanic craters. Fig 4 depicts the graph related to the percentage of surface porosity and the average diameter of the holes of the coated samples. The percentage of porosity and the diameter of the holes created in the samples without graphene oxide after 100 and 300 seconds and the solution samples containing 10 and 25 grams per liter of graphene oxide are shown.

3.3. Study of cross-section of samples

The present study investigates the crosssectional characteristics of samples coated with graphene oxide solution at concentrations of 15 and 25 g/L. Fig 5 displays the microstructure of the coated samples, revealing an increase in density and a reduction in internal pore diameter compared to oxide coating. The cross-sectional image also reveals two types of surface holes, namely Type 1 holes that do not reach the joint metal-coating and are caused by trapped gases on the coating surface, and deep surface pits that have more depth than Type 2 pits, but do not reach the cover-substrate joint. The latter type of pits is considered the most destructive, as they

extend from the cover surface to the sub layer. The growth of the coating is controlled by the penetration of oxygen and the exit of molten aluminum through the discharge channels, resulting in the freezing of the molten aluminum on the surface. Electrophoretic absorption of graphene oxide and the formation of molten aluminum on the anode surface lead to higher compression and reduced dimensions of the discharge channels in the sample with graphene oxide compared to the sample without it. The decrease in the number and size of sparks during the coating process due to the decrease in part temperature has reduced the dissolution of oxygen and, consequently, the number of gas holes in the coated cross-section. Graphene oxide has filled the holes and reduced the crosssectional porosity. The weight of the coating in the sample without graphene oxide is 0.4 ± 0.02 , while the sample containing graphene oxide (25 g/L) has a weight of 0.8 ± 0.02 g. The addition of graphene oxide in the silicate electrolyte has significantly increased the weight of the coating, indicating that the additive presence has partially filled the pores and thickened the coating. *3.4. Study of samples in terms of phase*

The diffraction pattern depicted in Fig 6 reveals that the predominant phases present in the coatings are forsterite and periclase. This observation suggests that the formation of aluminum oxide is consistent with the anodizing process. The X-ray diffraction (XRD) patterns of



Fig 3.(a) TEM image of produced graphene oxide, SEM image of (b) aluminum 6061, (c) aluminum 6061 modified by plasma electrolytic oxidation (c) aluminum 6061 modified by plasma electrolytic oxidation after 100 seconds of the process, (d) alloy modified by electrolytic oxidation in a solution containing graphene oxide after 300 seconds, (e) alloy modified by electrolytic oxidation in a solution containing graphene oxide with a concentration of 15 g/L, and (f) alloy modified by electrolytic oxidation of 25 g/l



Fig 4.Porosity chart and average size of holes in samples without graphene oxide after 100 and 300 seconds and solution samples containing 10 and 25 g/l of graphene oxide



Fig 5.Image of the cross section of samples coated with 15 and 25 g/l graphene oxide solution

PEO layers, produced using optimal concentrations of potassium hydroxide (KOH) and sodium silicate (Na_2SiO_3) , as well as uncoated alloy, are displayed in Fig 8. The thin and porous nature of the PEO layers allows for easy penetration of X-rays, resulting in the detection of strong diffraction peaks of the aluminum (Al) substrate in all samples. The XRD patterns reveal that the primary phases of the PEO layers are alpha-Al₂O₃ and gamma-Al₂O₃. Furthermore, the coated samples exhibit the presence of mullite (Al_2O_3 nSiO₂), a well-known ceramic phase with excellent thermal and chemical stability. The alpha-Al₂O₃ phase possesses a trigonal crystal structure, high stability, and a high melting point of 2050 C. It is

noteworthy that the meta-stable c-Al2O3 phase transforms into alpha-Al2O3 within the temperature range of 800 to 1200 C. The diffraction pattern of the coated samples reveals the presence of Al_2SiO_5 (aluminum silicate), indicating a significant amount of SiO₄ in the film. Sodium silicate undergoes facile hydrolysis in aqueous solution, leading to the formation of Si $(OH)_4$ and hydrated SiO₂ due to the high heat generated by the arc phenomenon. The PEO process involves exposure to high temperatures, causing both SiO_2 and Al_2O_3 to melt. Upon extinguishing of the micro-arcs, the electrolyte cools the molten mixture, leading to a phase transformation resulting in a mixture of Al₂SiO₅ and Al_2O_3 phases.



Fig 6.XRD spectrum of uncoated and coated samples with 15 and 25 g/L graphene oxide solution

Notably, the silicate-based coating GO-0 does not exhibit any discernible phase indicative of the GO presence. However, upon the addition of graphene oxide, the intensity of several peaks in the aluminum form in the sample without graphene oxide transforms into Al₂O₃ and Al₂SiO₅ phases. Furthermore, the intensity of these peaks increases with increasing the GO concentrations. *3.5. FT-IR analysis study*

To validate the X-ray diffraction (XRD) analyses and to identify the bonds between oxygen, carbon, and silicon in the coatings, Fourier transform infrared (FTIR) analysis was conducted on the samples, as depicted in Fig 7. It is well-established that the positions of peaks in an infrared spectrum are indicative of the molecular composition and structure. The absorption bands obtained from the infrared spectrum are consistent with the findings of previous researchers. In the GO-15 sample, the peaks of carboxyl and hydroxyl vibrations were observed in the range of 3040 to 3810 cm⁻¹. The absorption bands in the range of 1600 to 1650 cm⁻¹ correspond to the O=C ketonic species, while the peaks of the bands presented from 1355 to 1420 cm⁻¹ are related to the H-C bond. The observed peaks in the range of 1044 to 1092

cm⁻¹ are associated with the asymmetric and stretching vibration of C-O-Si and Si-O-Si groups. The peaks observed in the 0-GO sample are primarily H-Si and O-Si, and the absorption band at 510 cm⁻¹ indicates the presence of Al_2O_3 composition. The peaks of graphene oxide (GO) powder are related to C=C, -CH, and O-C bonds. Concerning the presence of carbon peaks in the GO-15 sample, it can be inferred that the graphene oxide particles were effectively coated during the plasma electrolytic oxidation (PEO) process.

3.6. Microhardness study

The hardness of the base metal was measured to be 85 ± 0.1 , while the GO-0, GO-15, and GO-25 samples exhibited hardness values of 422 ± 0.2 , 552 ± 0.4 , and [value missing for GO-25]. The results indicated that the addition of GO to the electrolyte led to an increase in the coating hardness. This finding was supported by Fig 7, which showed that the incorporation of GO resulted in the formation of hard phases such as Al_2O_3 and Al_2SiO_5 , and a fine structure with fewer structural defects. Furthermore, the compression and density of the cross-sections of the coatings



Fig 7.FTIR diagram of uncoated and coated samples with 15 and 25 g/L graphene oxide solution

demonstrated that the hardness increased with the density of the coating, resulting in a higher hardness of the coated samples compared to the uncoated sample. In terms of abrasion behavior, the coefficient of friction was measured for a distance of 100 meters. The reference sample (6061 Al alloy) exhibited the highest average friction coefficient of approximately 0.25. The fluctuations in this sample were observed to occur in two stages. The first stage, which lasted for approximately 70 meters, exhibited a large range of fluctuations due to the roughness and unevenness of the surface. The second stage, which continued from 75 meters to the end, showed a lower range of fluctuations and was attributed to the accumulation of particles resulting from wear.

3.7. Abrasion behavior study

The diagram presented sudden fluctuations and valleys in a partial form, which may have occurred due to the removal of a piece of the surface. Specifically, in the first stage, the coefficient of friction was smooth on the surface, and partial wear created a groove on the surface.

As the procedure continued, adhesive wear overcame sliding, resulting in an increase in the friction coefficient. Overall, the findings of this study suggest that the incorporation of GO in the electrolyte can enhance the hardness and abrasion resistance of base metal coatings.



Fig 8. The graph of the friction coefficient according to the distance traveled of uncoated and coated samples with 15 and 25 g/L graphene oxide solution

The present study investigates the variation range of friction coefficient in coated samples, which is found to be 0.15 and 0.07, respectively. The samples are divided into two areas, with the first area being short and the second area being entered after traveling a distance of 2 to 8 meters. The small extent of the first area indicates low roughness and a relatively smooth surface with friction. The steel pin initially scratches the surface protrusions of the coating, leading to a rapid increase in the friction coefficient until the protrusions disappear. The reduction of adhesion between two surfaces logically reduces the coefficient of friction compared to the uncoated sample. It is noteworthy that GO-15 and GO-25 samples exhibit self-lubricating properties due to the presence of graphene oxide. The relatively high friction coefficient for the uncoated sample is attributed to the loss and destruction of the porous outer layer caused by the effects of hard phases such as hard Al₂O₃, Al₂SiO₅, and SiO₂, and the weakness of this layer, which breaks easily.

The surface of the coated samples reveals that the lubrication effect of the graphene oxide additive fills the pores, resulting in a denser composite coating than the PEO coating without the addition of graphene oxide. Following abrasion and a reduction of shear force, a lower friction coefficient is observed in composite samples. Graphene oxide, either in single layer or multi-layer form, easily merges with molten oxide in PEO electrolyte, and with the formation of graphene oxide in surface cracks and pores, it leads to an increase in the solid lubrication properties of the coating .Table 3 presents the weight loss data of the samples after the wear test. The results indicate that the sample without graphene has the highest weight loss, while the GO-15 sample has the lowest weight loss. The weight loss of the samples after wear is primarily dependent on the surface roughness and the chemical composition of the coating, with a lower weight loss observed in the coating with a smoother surface and higher hardness.

Sample code	Sample	Pin
6061 Al GO-0	2.8	3.6
6061 Al GO-15	1.1	-0.4
6061 Al GO-25	0.6	-1.2

Table 3. Weight reduction of uncoated and coated samples after wear test

The inclusion of graphene oxide has been found to significantly enhance surface roughness and hardness, as well as increase wear resistance. The uncoated sample (6061 Al GO-25) exhibited a high degree of weight loss due to its low hardness and wear resistance, despite the fact that the abrasive pin's weight increased as a result of the accumulation of separated aluminum particles on its surface. The weight loss of the coated samples varied slightly, indicating an improvement in wear resistance. Notably, the weight of the pins decreased due to the surface's hardness, increased wear resistance, and eventual tearing of the pin's surface. It should be noted that the small weight loss observed in the coated samples does not imply an absence of weight loss; rather, it is compensated for by the weight increase resulting from the accumulation of iron particles worn from the pin on the sample's surface. In addition, the weight reduction or high wear of the steel pin suggests that the composite coatings are resistant to the steel surface and cause severe wear. The specific wear rate is directly proportional to the friction coefficient, and samples with lower friction coefficients exhibit lower specific wear rates. The wear rates of GO-0, GO-15, and GO-25 were measured as 4.1±0.2, 4.2±0.3, and 1.1±0.1, respectively. The GO-25 sample exhibited the lowest specific wear rate due to its high hardness, low friction coefficient, and the formation of a composite containing carbon in the aluminum oxide structure, which lubricated the surface more effectively.

4. CONCLUSION

In conclusion, the addition of graphene oxide to the silicate base coating electrolyte has resulted in an increase in electrical conductivity and applied current density. The surface morphology of the coated samples exhibited a volcanic crater structure, while the uncoated samples had a pancake structure. The coating thickness was not significantly affected by the addition of graphene oxide, but it did fill the pores. The Pertwix diffraction pattern of the coated samples clearly indicated the presence of major phases such as Al₂O₃, Al₂SiO₅, and SiO₂. Furthermore, a peak affected by graphene oxide was observed at an angle of approximately 10 degrees for the samples coated with graphene oxide. The IR-FT results revealed that the uncoated sample mainly presented H-Si and O-Si peaks, while the coated samples showed the presence of C=C, H-C, and O-C carbon bonds, indicating the successful coverage of graphene oxide particles during the PEO process. The wear behavior of the coated samples at 100 meters demonstrated a decrease in friction coefficient compared to the reference sample, and a significant increase in wear resistance due to the enhanced hardness and lubrication properties of graphene oxide in the coating.

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