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Plasma electrolytic oxidation and its application in tribological and corrosion protections: An overview

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Abstract: Plasma electrolytic oxidation (PEO) plays an important role in the surface finish of metals and alloys by converting the intrinsic surface layer of the metal into an oxide with a certain thickness, microstructure, chemical and thermal stability, and mechanical and functional properties. In a manner similar to other surface conversion-based coating techniques, such as carburization, nitridation, and thermal oxidation, the coating quality of PEO is strongly dependent on the processing conditions and parameters. Rapid progress in the additive manufacturing of metals and alloys in the past decade has increased the requirements for PEO-based surface finish. Recent developments in the PEO process have demonstrated that the characteristics of PEO coatings can also be engineered by incorporating extrinsic elements (other than oxygen) and/or microstructure by engineering the electrolyte and PEO processing strategy. In this overview, we first introduce the mechanism and coating properties of PEO with a focus on recent advances in PEO techniques. Current issues in PEO coatings are presented based on their applications, and existing solutions are discussed in tandem with the material characterization. By presenting the latest approaches, we attempt to provide readers with a perspective on the current state of PEO and an outlook on its future developments.

Keywords: Plasma electrolytic oxidation; anodic oxidation; electrochemical deposition; tribological protection; corrosion and erosion resistant; biocompatible application

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1. Introduction

In recent years, there has been a growing interest in developing advanced surface protection techniques to enhance the durability and performance of metallic materials under challenging operating conditions. One such technique that has gained significant attention is plasma electrolyte oxidation (PEO), also known as microarc oxidation or anodizing spark discharge [1-3]. PEO is a versatile electrochemical conversion coating process that offers unique advantages in terms of tribocorrosion protection for a wide range of metals and alloys. Tribocorrosion, the combined effect of mechanical wear and corrosion, poses a substantial challenge in various industrial applications, including automotive, aerospace, marine, and biomedical sectors [4-8]. The interaction between sliding surfaces coupled with corrosive environments can lead to accelerated material degradation, compromising the reliability and lifespan of critical components. Effective surface protection methods are essential for preventing tribocorrosion of metals and alloys. PEO is a promising solution by providing unique oxide coatings for tribocorrosion protection. This involves subjecting the metal surface to an electrolytic bath and applying a high-voltage electrical potential, resulting in the formation of an adherent oxide layer consisting of species from both the electrolyte and the base material. This unique oxide layer possesses exceptional mechanical, chemical, and tribological properties, making it an ideal candidate for mitigating tribocorrosion [4-8].

There has been frequent mention of association between PEO and another electrochemical technique called anodic metal oxidation (AMO), which has been applied on an industrial scale since 1920s [3, 9-11]. In general, PEO operates at high voltage, typically a few hundred volts, which are sufficient to electrically break down the formed

oxide layer and advance oxidation to the surface of the base material. In comparison, AMO usually operate at voltage lower than 100 V, below the breakdown voltage of the oxide barrier layer, resulting in limited oxide thickness [11-14]. A comparison between PEO and AMO can provide insight into the mechanisms and properties of oxide coatings.

AMO can be processed via two distinct approaches, i.e., barrier and porous approach, under potentiostatic conditions, which exhibit different oxide growth kinetics [15]. In the barrier approach AMO, the oxide growth rate decreases exponentially with time under potentiostatic conditions, resulting in a limited coating thickness. In contrast, in the porous-mode AMO, the anodization reactions are significantly faster at the bottom of the nanopores. Alongside the slight dissolution of the formed oxide into the acidic electrolyte, a static growth rate can be maintained to extend the oxide pores deeper into the base material. A two-step porous-mode AMO process is developed to obtain regular porous oxide structure. This process entails chemical removal of formed oxide layer during the initial anodization in the first step. The remaining shallow craters served as the starting point for the second anodization step [11]. Alternatively, intentional surface craters, such as those created by mold indentation, can be employed to define the initial anodization site [11]. During anodization, the oxide layer at the bottom of the crater, known as the barrier layer, is thinner than the region on the walls. Consequently, a shorter current pass forms at the center of the crater and experiences a higher electrical field. This facilitates the reaction of oxygen ion species (i.e., O^{2-}/OH^{-}) with the base metal through the barrier layer. Conversely, cations can diffuse outward from the oxide/metal interface and be released into the electrolyte from the electrolyte/oxide interface, driven by high electrical field in this region [15]. As a result, the barrier layer at the center of the

crater deepened more rapidly than the surrounding region, leading to the formation of straight nanochannels within the oxide layer induced by the AMO process. The development of a regularly ordered nanoporous oxide layer through AMO is generally influenced by the choice of acidic electrolyte used, such as sulfuric acid, oxalic acid, phosphoric acid, and citric acid, where the anodic oxide is slightly soluble [15].

In contrast to AMO, PEO typically operates in neutral or alkaline electrolytes, where the solubility of anodic oxide is significantly reduced. The working voltage can approach 1000 V, which induces electrical breakdown and microdischarge at the interface between the electrolyte and the initial oxide. This initiates a complex process involving numerous electrochemical reactions as well as physical, metallurgical, and crystallographic change in the oxide layer. Owing to the increased voltage, the growth and breakdown of the oxide occurs randomly at the electrolyte/oxide interface over time. The localized plasma rapidly increases the surrounding temperature, leading to an accelerated melting, oxidation, and ejection of metal and oxide species. Consequently, irregular pore structure is formed in the oxide layer [8, 15-17]. These porous structures exhibit complex architecture that significantly affects the surface integrity, mechanical properties, and fatigue performance of base material [18-20]. However, the introduction of PEO coatings can substantially enhance the tribological properties and corrosion protection of the base material by providing physical and chemical isolation from surrounding environment [21]. The porous nature and microcracks in PEO coatings tend to reduce their hardness and stiffness; thus, their damping capacity is greatly enhanced compared with the completely dense oxide coating. These characteristics significantly enhances the strain tolerance of PEO coatings under external mechanical loads such as

sliding and abrasive forces. Additionally, the inherent nature of the conversion coating results in strong interface adhesion between the PEO layer and the base material, leading to enhanced resistance against spallation of PEO coatings during tribological applications.

The porous structures of oxide layer induced by PEO on Ti- and Mg-based biomaterials offer advantages for enhancing antibacterial and cell-interactive properties for biocompatible applications [22]. Moreover, nano- and microscale particles or ionic species, for example, corrosion inhibitors [23], cell culture promoters (Ca^{2+}/P_i) [24], surface coloring decorators [25], and others [26, 27], can be introduced into the porous oxide layer during the PEO process through the electrolyte with the assistance of microplasma arc. Unlike the static-stable growth of straight oxide pores in the porous-mode AMO process, the oxide growth during the PEO process occurs dynamically throughout the 'grown' oxide layer. This behavior allows the incorporation of additives, such as ceramic particles [28], carbon nanotubes [26], and Ca^{2+}/P_i species [5], not only in the near-surface region, but also deep into the oxide/metal interface. This unique characteristic of the PEO process provides a flexible strategy for modifying the properties of oxide coating, thereby enabling multifunctional applications with enhanced performance.

In the past decade, there has been rapid development in the field of materials research, particularly in the additive manufacturing (AM) of metals and alloys [29-33]. This revolutionary progress, combined with the growing demand for fast and customizable fabrication of medical implants as well as the inherent surface-accessing limitations of AM components with complex geometry [34], has resulted in an increasing need for effective post-AM surface modifications. Consequently, PEO has garnered

increased interest for the treatment of AM metals and components such as Ti and Mg-alloy [35-38]. These alloys often exhibit different structural and mechanical properties than those fabricated by traditional casting and/or wrought methods [26]. The intrinsic structural variation of AM alloys pose challenges for achieving uniform PEO coating with desirable mechanical and tribological properties. In addition, the numerous parameters involved in the PEO setup and process, including electrolyte composition, voltage/current mode and waveform profile, and the incorporation of extrinsic additives, present difficulties in achieving reliable and consistently functional PEO coatings, particularly on AM alloys. Therefore, post-PEO treatments may still be necessary to address specific application requirements and enhance the overall performance of coatings.

In this article, we provide a brief overview of the recent developments in PEO coatings for structural and functional applications. After introducing the basic mechanism and coating properties of PEO along with its recent technical advances, we discuss the current issues associated with PEO coatings and address the tribocorrosion features. Furthermore, we explore existing solutions to these issues, focusing on material characterization, including structural, mechanical and tribological properties, as well as resistance against corrosion, erosion, and bacterial impregnation. By presenting the latest approaches, particularly in antibacterial and biocompatible applications, we aim to provide readers with insights into the current state-of-the-art PEO technique and provide an outlook on its future development.

2. Basic PEO mechanism and coating properties

2.1 Growth mechanism of PEO coating

The PEO process involves subjecting a metal substrate to an electrolytic bath, typically containing an alkaline or acidic solution, and applying a high-voltage electrical potential. This electrical potential creates a localized plasma discharge on the metal surface, leading to the formation of an oxide layer through complex electrochemical reactions. A unique feature of PEO is the generation of microdischarge or spark, which contributes to the growth and densification of oxide layer. The formation of oxide layer in PEO occurs via a series of electrochemical processes. Initially, when an electrical potential was applied, the onset of a thin and porous oxide layer occurred on the metal surface. This layer acts as a barrier, preventing further current flow and limiting the penetration of electrolyte into the underlying substrate. As the process continues, the microdischarge within the plasma discharge region leads to localized melting and vaporization of metal, which combines with oxygen from the electrolyte to form oxides. These oxides deposit on the surface, gradually thickening and densifying the oxide layer.

A typical PEO setup is schematically shown in Fig. 1(a), where the metal plate to be coated and a stainless-steel plate are set as anode and cathode, respectively. Subsequently, they were immersed in an alkaline electrolyte. A power supply is used to apply a voltage between the two electrodes, which should be sufficiently high (up to 1000 V) to generate discharge on the anode. During the PEO process, the current-potential profiles, as well as their variation as a function of time, can be monitored by the U/I probe. A stirrer and cooling system is used to maintain a constant temperature of

electrolyte. Figure 1(b) presents a schematic representing the anodization processes with potentiodynamic scans using high (i), medium (ii), and low (iii) concentration alkaline electrolytes. Similarly, Figure 1(c) presents a typical voltage-time response profile collected during the PEO process in a low-concentration alkaline electrolyte, corresponding to case (ii) in Fig. 1(b).

Figure 1. Schematic illustration of a typical PEO process. (a) Experimental setup, (b) PEO process with a potentiodynamic scan in high (i), medium (ii), and low (iii) concentration alkaline electrolytes, and (c) Process voltage-time response profile of PEO coating, transition from anodization oxidation through microarc oxidation to steady-state microarc oxidation. Scale breaks on the time and voltage axes should be noted. Partly reduced from [39] ©Elsevier 2016.

The comparison in Fig. 1(b) shows that the spark initiation voltage and discharge behaviour strongly depend on the electrolyte concentration. The combination of Figs. 1(b) and 1(c) shows that the PEO process can be defined in three distinct stages. In the first stage, the applied voltage increases linearly at a low current, leading to conventional anodization and the formation of a barrier oxide layer. The growth of oxide layer is controlled by solid-state diffusion of O^{2-} and metallic ions Mn^{n+} under an electric field. Alongside the oxide film growth ($nO^{2-} + 2M^{n+} \rightarrow M_2O_n$) on the surface of the anode, the potential increases because of increased resistance of oxide film. O_2 and H_2 bubbles are released at anode ($O^{2-} + 2e^- \rightarrow O_2$) and cathode ($2H^+ + 2e^- \rightarrow H_2$) surfaces, respectively. The increased voltage leads to an increase in the current density and eventually an electrical breakdown of grown oxide film, giving rise to discharge arc or spark, and the PEO process enters the secondary stage. In the high-concentration electrolyte (case i in

Fig. 1(b)), a further increase in voltage leads to a sharp increase in the current density and arcs, and the PEO process enters the second stage. In medium-concentration electrolyte (case ii in Fig. 1(b)), an increase in the voltage approaching a critical value gradually increases the current density and initiates discharge at the electrolyte/metal interface. However, a further increase in the voltage leads to a sharp increase in the current density and large arc, similar to that in the high-concentration electrolyte, along with O_2 release caused by water oxidation. In contrast, in the low-concentration electrolyte (case iii in Fig. 1(b)), after the anodization stage, only microdischarge appears, which further develops into steady-state microarc oxidation without the presence of large arc, i.e., the third stage of the PEO process.

The involvement of plasma discharge complicates the PEO process, and the oxide growth mechanism is not yet fully understood. However, the investigation of events following discharge can provide some insights to understand these complex phenomena. The discharge energy is absorbed by various phenomena occurred during PEO, including melting and vaporization of substrate, melting of existing oxide coating, initiation and sustaining of plasma, vaporization of water, and electrical heating of electrolyte. By studying an individual discharge on an Al substrate during PEO, the dynamics of most of these transient events were revealed. The typical results are presented in Fig. 2.

As shown in Fig. 2(a), once the discharge is ignited, a small portion of the released energy is used by ionization to create and sustain the plasma with a lifetime on the order of 100 μs. Soon after the generation of plasma, the discharge energy is absorbed by the electrolyte, leading to the vaporization of water and the formation of vapor bubbles. The bubbles expand to a maximum radius, shrink, and eventually collapse [see

Fig. 2(b)]. The lifetime of vapor bubble is approximately 160 μs. With the shrinkage of vapor bubble, the surrounding radicals and chemicals in the electrolyte are pulled towards the growth frontier of coating. The collapse of vapor bubble generates jet flow and shock waves, which enable efficient incorporation of chemicals and additives into PEO coating through the electrolyte. As shown in Fig. 2(a), after plasma ignition for approximately 100 μs, the majority of the discharge energy is absorbed by the electrolyte.

Figure 2. Semi-quantitative estimation of energy evolution for an individual discharge during typical PEO process. (a) Energy evolution and (b) bubble evolution. Reproduced from [40] ©Elsevier 2015.

Although the PEO process can be understood from the perspective of time domain by analyzing the voltage and/or energy evolution, as shown in Figs. 1 and 2. In practice, the three typical PEO stages occur simultaneously and dynamically at different locations on the workpiece. The dielectric breakdown incudes millions of short-lived microdischarge uniformly spreads on the surface of the workpiece, creating discharge channels for the direct ejection of molten metals (i.e., Al, Ti, or Mg), which are then oxidized, hydrolyzed, and precipitated on the substrate. Chemical, electrochemical, thermodynamic, and plasma-chemical reactions occur at the discharge site, resulting in structural, compositional, and morphological modification of PEO coating. The dielectric breakdown of the oxide layer results in a sharp increase in the electric current and local temperature (10^3–10^4 K). As a result, water and gas vapor are produced with pressure build-up (~10^2 MPa), leading to water oxidation and O_2 evolution. Phase change in the

grown oxide occurring at high temperature, for example, transformation of Al_2O_3 from γ- to α-phase, can further increase the hardness and anti-wear properties of PEO coatings.

PEO coatings can be divided into three layers: the outer, the middle, and the bottom layers. The outer layer is typically porous and rough and can be removed by mechanical grinding and polishing for specific applications. In comparison, depending on the processing parameters, the middle and bottom PEO layers, which are sometimes indistinguishable from each other except for the relative spatial sequence with the bottom one near the coating/substrate interface, can be optimized to be pore- and crack-free. Varying the applied current, for example, alternating current, direct current, unipolar current, bipolar current, etc., could significantly modify the inner layers of PEO coatings. Recent studies adjusting the periodic PEO current waveforms revealed that a proper increase in the negatively pulsed current can remarkably reduce porosity of the middle and bottom PEO layers. A typical example of the effect of the applied current waveforms on the PEO process of an Al-alloy substrate is shown in Fig. 3 [41], where the aqueous electrolyte consisted of KOH and Na_2SiO_3 with a conductivity of 2.3 ~ 2.8 mS/cm and a pH value of 11.7. The electrolyte temperature was maintained below 30 °C during the PEO coating process and the positive current density was maintained at 380 mA/cm^2.

Figure 3. Effect of bipolar current ratios on the deposition of aluminium oxide coatings on 2214-T6 Al alloy. (a) Current waveforms of a single period with positive current I_p (above the horizontal dashed line) and negative current I_n (below the horizontal dashed line); (b) and (c) Intensity evolutions of the Al emission line at λ =396.2 nm as a function of PEO process time with R = 1.57 and 0.89,

respectively; (d) and (e) Side-view photographs taken at different PEO stages for R = 1.57 and 0.89, respectively; and (f) and (g) Cross-sectional SEM images taken from PEO coatings on Al alloy processed at R = 1.57 for 140 min and R = 0.89 for 90 min, respectively. Reproduced from Ref. [41] ©Elsevier 2007.

Figure 3(a) shows the current mode with waveform profile as a function of time within a single period, where the positive-to-negative charge quantity ratio R is defined by $R = \frac{\int_0^{T_1+T_2+T_3} I_p \cdot dt}{\int_{T_1+T_2+T_3}^{T} I_n \cdot dt}$. Increased negative charge is manifested by a reduced R value. Presented in Figs. 3(b) and 3(c) are featured Al emission intensity at 396.2 nm as a function of time measured by an optical emission spectroscopy (i.e., a combination of a monochromator and a coupled charge detector) from the surface of the Al-alloy coupon during the PEO process with R = 1.57 and 0.89, respectively. Similarly, as presented in Figs. 3(d) and 3(e), photographs were recorded from the side edge of the Al-alloy coupon by a digital camera at different PEO stages with R = 1.57 and 0.89, respectively. The combination of Figs. 3(b)-3(c) and 3(d)-3(e) reveals that the discharge plasma damping is faster at a reduced R. Additionally, the sparking spots were smaller when the R value was reduced. Cross-sectional scanning electron microscopy (SEM) images shown in Figs. 3(f) and 3(g) were taken from the PEO coating processed at R = 1.57 for 140 min and R = 0.89 for 90 min, respectively. Their comparison provides direct evidence for reduced porosity in the middle and bottom PEO coating layers of the process with a reduced R value, which is strongly correlated with reduced spark spot size and fast plasma damping rate.

Detailed microstructural studies of PEO coatings by Sharma et al. on Al alloy substrates revealed that the outer layer consists of sintered Al_2O_3 and SiO_2 with high porosity and amorphous content [42], while the middle layer (also known as the intermediate layer) and the inner layer (usually very thin and similar to the barrier layer formed during anodization [43]) contain well-crystallized α-phase Al_2O_3 with lower porosity [42]. It is worth mentioning that the phase composition of PEO coating largely depends on the process parameters, particularly the process voltage and time. Our recent studies revealed that the potential bipolar PEO process of Al alloys using NaOH and Na_2SiO_3 electrolytes gave rise to single-phase γ-Al_2O_3 coatings with a thickness of up to 20 μm. An increase in the coating thickness, i.e., by increasing the process time and voltage, to approximately 30 μm resulted in the formation of α-Al_2O_3 alongside the growth of γ-Al_2O_3. The single-phase γ-Al_2O_3 coatings formed by the PEO process on Al alloy are consistent with those observed by Shore et al. on AA6060-T6 with a coating thickness smaller than 30 μm [44]. The inclusion of SiO_2 in the outer PEO layer provided direct evidence for the incorporation of Si species from the electrolyte, i.e., the Na_2SiO_3 salt solute, into the coating. This tends to modify the functions of the outer layer, such as, the absorption and adhesion of the bonding media [44]. The dense middle and inner layers can physically isolate the base material from the working environment, providing excellent corrosion resistance. The porous outer layer, the dense immediate and inner layers, together with the well-adhered coating/substrate interface of the PEO process, contributes to enhanced thermomechanical and tribological properties in addition to the corrosion resistance of the PEO coatings. It is believed that optimal combination of

thermomechanical, tribological, and anticorrosion properties can be obtained by engineering the thickness of outer and intermediate layers within the PEO coating.

2.2 Unique properties and functions of PEO coating

Owing to their unique structural characteristics, including a porous outer layer and dense intermediate and inner layers, as well as a well-adhered coating/substrate interface, PEO coatings exhibit unique mechanical properties. Typical hardness of PEO coatings is in the range of 1000–1800 HV on Al alloy substrates, which is significantly greater than stainless steel (i.e., ~500 HV), and over 400 HV on Mg- and Ti-alloy substrates owing to a combination of crystallization (of oxides) and incorporation of elements from the electrolyte into the ceramic coating. Physically, a combination of high hardness and compliance is necessary to obtain the comprehensive wear resistance of a coating system. The porous outer layer of typical PEO coatings allows for a certain level of deformation under external loading and/or thermal expansion without creating undue stress at the coating/substrate interface. The combination of extreme hardness and porous outer-layer-induced compliance of PEO coatings makes them important for anti-wear applications [45]. Similarly, the unique microstructure of PEO coatings gives rise to high fracture toughness, i.e., with reduced potential for cracking under external loading, and thus enhanced tribological performance [46-49].

Compared with traditional anodization processes in acidic electrolyte, PEO processes in aqueous electrolytes are more eco-friendly. Furthermore, PEO is much less sensitive than traditional anodization to the surface and chemical composition of the substrate material. These advantages of PEO process make it promising to replace

traditional anodization for producing adhesion coating on Al-, Mg-, and Ti-alloys for bonding applications [44, 50-54]. To develop PEO coatings for adhesion applications, i.e., to host bonding media, for example, epoxy adhesives, at the joint connection of separated components, Shore et al. studied PEO coatings on AA6060-T6 using $Na_4P_2O_7$ and $Na_4P_2O_7 + Na_2SiO_3$ electrolytes [44]. The PEO coatings, with thickness in the range 15–25 μm, are dominated by single-phase γ-Al_2O_3, regardless of the nature of electrolyte. In a similar study, Yaakop et al. found that an electrolyte containing $(P_2O_7)^{4-}$ anions tends to promote the formation of α-Al_2O_3 through the formation of $AlPO_4$ [55]. An increase in the coating thickness to ~26 μm resulted in the uniform distribution of α-Al_2O_3 throughout the PEO coating. In comparison, the incorporation of SiO_3^{2-} anions into the electrolyte tends to increase the coating rate, porosity, and surface roughness, as well as delay the formation of α-Al_2O_3 [55]. It has been suggested that the delayed formation of α-Al_2O_3 is due to increased porosity and roughness, and thus, enhanced electrolytic cooling at the reaction regions within the discharge channels [55]. The scratch test indicated that both the critical load and failure load monotonically increased with coating thickness. In contrast, the lap-shear strength of coatings decreased monotonically with coating thickness. The thinner PEO coatings succeeded in the lap-shear testing, whereas the thicker PEO coatings failed cohesively within the oxide layer rather than at the interface, and the fracture typically propagated through the dense inner oxide region [44].

The well-adhered coating/substrate interface, excellent thermal conduction performance of Al_2O_3 (especially α-Al_2O_3), and intrinsic dielectric and electrical insulation characteristics of PEO coatings make them promising for high-power electronic applications [55]. Even-increasing developments in high-power electronic

systems with enhanced functions and/or performances for electric vehicles and avionic applications are pushing the requirements for effective thermal dissipation at both the device and system levels [56, 57]. The device-level thermal dissipation is largely dependent on the application and performance of thermal interface materials (TIMs) that promote heat transfer from the device to the heat sink on the printed circuit board (PCB), while the system-level thermal dissipation is partly promoted by applying a heat-conductive wedge-lock between the PCB and its housing chassis. In both cases, the thermal interface resistance, and thermal conduction resistance, electrical insulation, and compliance and tolerance to mechanical pressing play an important role in determining the heat dissipation efficiency. Such specific requirements, along with weight reduction for electrical vehicles and avionic applications, motivated investigations on PEO process, especially on Al alloy substrates [55]. Araújo et al. studied the PEO process on an Al alloy substrate using a Na_2SiO_3 electrolyte and obtained a high thermal conductivity for the PEO coating (~290 W/m·K), which is nearly one order of magnitude greater than that of Al_2O_3 and AlN [58]. The electrical resistance of obtained PEO coating reached as high as 4.6×10^{12} Ω·cm [58]. The combination of high electrical resistance and high thermal conductivity of PEO coating sheds light on its applications for heat dissipation in high-power electronics.

3. PEO coatings for tribological and corrosion protections

In addition to their unique properties and functions for bonding and microelectronic applications, PEO coatings have been extensively studied for many applications, ranging from biomedical, automation, aerospace, chemistry, and energy to infrastructure.

Excellent review articles have been published on the process parameters, microstructure, and resultant performance, as well as their relationship with a unique class of PEO coatings [3, 6-9, 59]. In this section, we do not repeat the basic material features of PEO coatings but concentrate on the most recent observations and problems encountered with respect to their tribological and corrosion protection on various alloy substrates.

3.1 PEO coatings on Al-, Ti-, and Mg-based alloys

The layered ceramic structure and adhered interface of PEO coatings on the base material makes them attractive for many protective applications where wear resistance to sliding and/or abrasive is desired. The adhered interface and porosity-induced toughness of PEO coating system contributes to its high resistance to spallation upon external loading. By engineering the electrolyte, which is typically dominated by sodium silicate and/or sodium phosphate with additives of NaOH, KOH, and/or nano-/micro-scale particles, the processing mode (single- or dual-polar bias), processing parameters (i.e., waveforms), processing time, hardness, porosity, and, therefore, the wear resistance of the PEO coating can be enhanced. However, the wear and PEO processes involve complex interactions and electrochemical reactions, respectively. There is a great challenge to build up the relationship among the PEO process, microstructure of PEO coating, and wear behavior of coating system.

Al alloy has been widely applied in equipment for semiconductor manufacturing, automobile, aerospace, and consumable electronics, etc. Surface treatment is a high-value-added process that is critical to the parts and equipment performance of Al alloy components. To evaluate the tribological properties of PEO coatings, in comparison with

traditional hard-anodization coatings, formed on Al alloy (AA6061-T6) substrates, Krishna et al. studied the effect of normal load on abrasion wear using a dry-sand rubber wheel system, and studied the effect of impact velocity and angle on erosion wear of protective coatings using a solid-particle method [60]. The PEO coatings were grown in KOH (4g/l) and Na_2SiO_3 (2g/l) aqueous electrolyte under 0.3 A/cm^2 and 150~200 V for 28 min. Their abrasive tests revealed that PEO coatings can reduce the wear rate of Al alloy substrate by a factor of 12–30, which is up to 15 times better than that of the protective coating formed by traditional hard-anodization [60]. However, their erosion test revealed that the PEO coating did not enhance the wear resistance, while the hard-anodization coatings adversely increased the wear rate of the Al alloy substrate by a factor of 10. Mechanically, the abrasive wear of Al alloy substrate was dominated by grooving; the cylindrical micropores of hard-anodization coating caused chipping of coating alongside grooving, and, in comparison, macro cracks of PEO coating accumulated damage and removed the coating in flacks [60]. In contrast to the abrasive wear, lip formation and shear driven by the localized plastic flow of the alloy substrate dominated its erosion wear. The erosion rate of protective coatings largely depends on the impact angle. Cratering, shear deformation, and fracture-assisted chipping can occur on hard-anodized coating, whereas lateral fracture dominates the erosion wear of the PEO coating [60].

Owing to their low density and high strength at high temperatures, Ti-based alloys play an important role in addition to Ni-based superalloys in aerospace applications such as, fan blades for aerospace turbine engines [61]. In addition to aerospace applications, Ti-alloy, e.g., Ti-6Al-4V, is also a biocompatible material owing to the natural formation

of a titania coating [8]. Zhong et al. studied PEO coatings on TiZr-based alloys (Ti30Zr5Al3V) to improve their wear resistance [62]. By varying the applied voltage from 300 to 500 V and processing times from 60 to 1800 s, similar composition of PEO coatings was obtained, including anatase, rutile, srilankite, and Al_2O_3 crystallographic phase structures, along with increased amorphous compositions with increased applied voltage and processing time. Tribological tests revealed that the PEO coating on Ti30Zr5Al3V alloy possessed a higher coefficient of friction but a lower wear rate. Both the applied voltage and processing time influenced the wear resistance of PEO coatings [62]. By changing the electrolyte composition, including $NaAlO_2$, Na_3PO_4, NaH_2PO_2, and/or Na_2SiO_3, PEO coatings were obtained by C. Yang et al. on a Ti-alloy (TC4) exhibited varied properties [63]. Generally, the PEO coating processed in the NaH_2PO_2-based electrolyte exhibited excellent corrosion resistance, whereas those processed in the $NaAlO_2$-based electrolyte showed superior tribological performance. A composite PEO coating produced in a mixed electrolyte containing NaH_2PO_2 and $NaAlO_2$ exhibited higher wear and corrosion resistance than those obtained in a single-component electrolyte [63]. These findings shed light on the electrolyte engineering of high-quality PEO coatings.

In contrast to the biomedical application of Ti alloy, which has the advantage of being able to form biocompatible titania coatings, Mg alloy is biocompatible and has been proposed for temporary bio-implant applications to promote the recovery of fractured bone while degrading itself after completing its biofunctions. However, rapid biocorrosion, corrosion-induced release hydrogen gas, and premature loss of biomechanical functions adversely influences the applications of Mg-based alloys as

biodegradable implant materials [64]. Moreover, accelerated biocorrosion might occur in Mg alloys upon fretting wear, i.e., tribocorrosion, which limits the application of Mg alloys as junction implants. To engineer the surface of Mg alloys and improve their biofunctions, da Silva Rodrigues et al. studied the effect of PEO coatings, consisting of Si and Mg oxides, on the corrosion resistance and tribological behavior of ZK30 [64]. They found that the corrosion resistance of the Mg alloy was enhanced by the PEO coating, and corrosion-induced hydrogen release was mitigated. Meanwhile, the PEO-induced surface topography and roughness increases the coefficient of friction and decreases the material removal rate of the Mg alloy [64]. In addition to the PEO processing parameters, the composition of Mg alloy substrate could also affect the corrosion resistance of PEO coatings obtained under identical conditions. For example, Hussein et al. found that PEO coatings processed by a bipolar mode on AZ91D exhibited better corrosion resistance than those produced on AJ62 and AM60B under the same conditions [65].

3.2 PEO coatings on Fe-based steel alloys

Along with its development on Al-, Ti-, and Mg-based alloys, PEO has also been developed on Fe-based steel alloys. By employing electrolytes containing calcium and phosphorus, i.e., calcium acetate (10 g/l) and beta glycerophosphate (1 g/l), Alves de Menezes et al. studied the PEO coating of hydroxyapatite layers on stainless steel substrates [66]. By varying the PEO processing parameters, including the current polarity and profile, as well as the processing time (i.e., in the range of 15 to 90 s), hydroxyapatite coatings were successfully obtained on cylindrical stainless-steel substrates with various thicknesses, porosities, topographies, roughness, and compositions of Ca and P inclusions. Figure 4 summarizes the typical results reported by Alves de Menezes et al. on PEO

coating of hydroxyapatite on stainless steel substrates using cathodic and anodic configuration schematically shown in Figs. 4(a) and 4(b), respectively [66].

The SEM images shown in Figs. 4(a_1)-4(a_4) and 4(b_1)-4(b_4) were obtained for PEO coatings processed using the cathodic and anodic modes, respectively [66]. They exhibited significantly different topographies, because of different coating mechanisms, which, together with the coating thickness [Fig. 4©] and contact angle [Fig. 4(d)], could influence their biofunctions [66-68]. In the cathodic PEO coating mode [Fig. 4(a)], the microarc occurred directly on the substrate, leading to both substrate oxidation and the inclusion of species from the electrolyte into the coatings. In comparison, the anodic PEO coating mode led to dominant electrolytic reactions and the deposition of extrinsic materials on stainless-steel substrate [66, 69]. Studies on chemical composition of PEO coatings revealed that Ca incorporation into coatings deposited by the anodic mode were much higher than those deposited by the cathodic mode while P incorporation, much lower than that of Ca (with the Ca/P atomic ratios up to 78.6), did not exhibit apparent differences between two modes [66]. These observations, together with microscale cracks formed in PEO coatings [indicated by the arrows in Fig. 4(b_1)], suggest that more work is required to optimize the PEO process for coating hydroxyapatite layers on stainless steel substrates for their biocompatible applications.

Figure 4. PEO coating of hydroxyapatite on cylindric stainless steel substrate in (a) Cathodic mode, (b) Anodic mode. (a_1)-(a_4) SEM images taken from PEO coatings in cathodic mode for 15, 30, 60, and 90 s, respectively; (b_1)-(b_4) SEM images taken from PEO coatings in anodic mode for 15, 30, 60, and 90 s,

respectively; and (c) and (d) processing time-dependent coating thickness and contact angle, respectively. The arrows indicate the formation of microscale cracks in the anodic PEO coating. Reproduced from Ref. [66] ©Elsevier 2022.

The '*extrinsic*' PEO coating, i.e., with the coating species deposited from the electrolyte directly onto the arc-activated surface of substrate, has also been developed for applications in marine and offshore industry, e.g., coating low carbon steel (LCS) pipelines [70-72]. By introducing Al nanoparticles into sodium silicate electrolyte for direct PEO process on LCS, Yang et al. found that Al nanoparticles can be incorporated into an oxide coating by forming alumina. The incorporation of alumina reduced the composition of iron oxide within the PEO coating on LCS, which significantly enhanced the tribological properties [see Figs. 5(a) and 5(b)] as well as the corrosion resistance [see Figs. 5(c) and 5(d)] of the LCS substrates [71].

Figure 5. PEO coating on low carbon steel substrate using aqueous sodium silicate electrolyte with and without introducing Al nanoparticles (NPs). (a) Comparison of friction coefficients; (b) Comparison of the wear-induced surface profiles; (c) Comparison of the electrochemical corrosion current densities; and (d) Comparison of the salt spray evolutions of the PEO coatings. Reproduced from Ref. [71] ©Elsevier 2027.

It is worth mentioning that such an '*extrinsic*' PEO coating might be further developed by coating the inner walls of hollow three-dimensional (3D) structures, e.g., pipelines [73], to enhance their tribological properties. This concept, as well as its variations, including plasma electrolytic nitridation, carburization, polishing, hardening, etc. [74-77], has attracted increased interest [75, 78], driven by rapid developments in additive manufacturing [79, 80], e.g., the direct energy deposition of geometrically complex components with increased integrity, in the past decades [81-83]. Hybrid coating methods [84], analogous to the '*extrinsic*' PEO process, but introducing the coating species by pre-deposition, e.g., dip-coating, physical vapor deposition, or laser cladding of Al or Ti layers [14, 70, 85-88], on the targeted substrate, have also been explored. These techniques, together with the variants of plasma electrolytic processes, provide more innovative strategies for coating protection with novel functions and/or enhanced qualities, not only on Al-, Ti-, and Mg-based light metal alloys but also on non-valve metals such as Fe-based steels and, Ta- and, Zr-alloys [48, 66, 70, 73, 89-91].

4. Electrolyte additives for PEO coatings

Additives can be introduced into the electrolyte to engineer the coating functions and/or properties via the PEO process. The most frequently employed electrolytic additives are particles at the nano-and/or micro scale. In general, the particles may fill the pores and/or cracks formed in the coatings during the PEO process, which in turn reduces the porosity, increases the hardness, and enhances the tribological properties and corrosion resistance of coatings. Depending on the selected materials of particles, the dielectric properties (e.g., the breakdown voltage and thermal conduction coefficient) of coatings can also be engineered.

4.1 Soluble electrolytic additives

Fluoride salts such as NH_4F and NaF have been studied in the PEO process as electrolytic additives; they are completely dissolved in the aqueous electrolyte to provide fluoride species. For example, Hao et al. found that the porosity of a PEO coating on a Ta-12W substrate in a traditional silicate-phosphate electrolyte can be reduced to less than 1% by adding NH_4F [90]. Large-pore structure can be effectively removed by optimizing the incorporation of NH_4F additives, leading to pancake-like surface structure consisting of sealed pores and pore-free features. Both features were associated with promoted gas release and bubble breakage in the later PEO stage, i.e., the bubble breakage resulted in the formation of pore-free pancake-like surface features, whereas the dielectric breakdown at the periphery of gas bubbles resulted in sealing of pores [90].

NaF additives (8 g/l) were introduced by Mohedano et al. into an aqueous Na_3PO_4 (10g/l)- KOH (1g/l)- CaO (2.9 g/l) electrolyte for processing bioactive PEO coatings on Mg-Ca alloy substrates [92]. They observed that the introduction of NaF significantly increased the coating rate, i.e., from ~6 µm thick by 600 s to ~50 µm thick by 420 s and ~70 µm thick by 540 s, through the formation of $Ca_5(PO_4)_3OH$, $Ca_5(PO_4)_3F$, and $Mg_2(PO_4)F$. Electrochemical corrosion tests in a simulated body fluid (SBF) at 37 °C revealed that the protection performance of the PEO coatings was significantly enhanced by introducing the NaF additives, i.e., the corrosion potentials shifted in the positive direction (i.e., nobler side) by over 0.2 V and the corrosion current density was reduced by over three orders of magnitude. It was also found that the fluoride ions released from the PEO coatings could effectively influence the degradation rate and enhance the bioactivity of Mg-Ca alloys for biomedical applications [92].

4.2 Insoluble electrolytic additives

In contrast to soluble additives, which are completely dissolved in aqueous electrolytes to provide ionic species for the PEO process, insoluble additives have also been widely studied for the engineering of PEO coatings. Some insoluble additives, such as, nano- or micro-scale Al particles [71], might be melted and oxidized during the PEO process, i.e., involving both physical and chemical reactions. In comparison, other insoluble additives with higher melting point, e.g., ceramic particles such as SiC [93] and, Al_2O_3, are predominantly involved in the physical activities during the PEO process.

The physical incorporation of nano- or micro-scale ceramic particles into PEO coatings through additives in the electrolyte tends to seal the pores and microcracks, increase the hardness, enhance the wear and corrosion resistance, and modify the dielectric and thermal conduction properties. It has been reported that the incorporation of SiC nanoparticles into electrolyte of PEO process results in enhanced wear and corrosion resistances, and thus, the resistance against tribocorrosion of the PEO coatings on Mg-based alloys [93]. The introduction of ZnO nanoparticles into the electrolyte resulted in reduced porosity, thickness, and surface roughness of the PEO coatings, which exhibited enhanced corrosion resistance and bioactivity owing to hindered ion penetration and promoted the formation of calcium phosphate [94]. The incorporation of nano- and micro-scale SiO_2 particles apparently influences the thickness, roughness, hardness, wear resistance, and corrosion resistance of PEO coatings [95]. Physically, both the particle size and melting point play a key role in their incorporation into the PEO coatings and, in turn, the coating performance. Table 1 summarizes typical electrolytic additives and their influence on PEO coatings.

Table 1: Typical electrolyte additives and their influence on PEO coatings.

Additives (g/l)	Electrolyte (g/l)	Substrate	Observed effects	Ref.
NH_4F (1~8)	Na_2SiO_3 (10); Na_3PO_4 (10); NaOH (2)	Ta-12W	Reduced porosity (< 1%); bubble breakage and pore-filling	[90]
NaF (8)	Na_3PO_4 (10); KOH (1); CaO (2.9)	Mg-1Ca	Increased coating rate and Ca/P-inclusion; bioactive coating by releasing fluoride ions	[92]
KF (10)	Na_3SiO_3 (8); NaOH (5); $NaH_2PO_4 \cdot 2H_2O$ (4/0)		Increased wear resistance, surface hardness, elastic modulus, and corrosion resistance	[96]
Al (2)	$Na_2SiO_3 \cdot 9H_2O$ (30); $NaH_2PO_2 \cdot H_2O$ (1.2)	Low carbon steel	Increased alumina and reduced iron oxide; enhanced wear and corrosion resistance	[71]
SiC (2)	$Na_2SiO_3 \cdot 9H_2O$ (12); $(NaPO_3)_6$ (15)	Mg-alloy (AZ31)	Increased coating rate; uniform SiC partial inclusions; increased wear and corrosion resistance	[93]
SiC (5)	Na_3PO_4 (4); KOH (1.5)	Mg-alloy (AZ31)	Increased microhardness and wear/corrosion resistance;	[97]
	$NaAlO_2$ (2); Na_2SiO_3 (2); KOH (1.5)		Larger hardness and wear/corrosion resistance increasements	
ZnO (1.5~4.5)	$K_3PO_4 \cdot 3H_2O$ (5); KOH (2)	Mg-alloy (AZ91)	Reduced surface porosity; increased corrosion resistance; $Ca_3(PO_4)_2$ layer	[94]
SiO_2 (5)	Na_3PO_4 (20); KOH (1)	Mg-alloy (AM50)	Nanoparticles filled into discharge channels and open pores; microparticles entered the surface; increased wear resistance and slight reduced corrosion resistance	[95]
SiO_2 (5, 10, 15)	Na_3PO_4 (12); NaOH (4)	Al-alloy (7075)	Porosity reduced by 30%; increased corrosion resistance; optimized addition of 10 g/l; increased wear resistance and tribological properties	[98]
Monoclinic ZrO (2)	$Na_6P_6O_{18}$ (15)	Al metal	Zirconia nanoparticles in near-surface cavities and in coatings; Zr-rich boundaries of micro cellular structure in	[99]

			Al-rich coating	
	$Na_2SiO_3 \cdot 5H_2O$ (5.6); KOH (2.8)		Zirconia nanoparticles entrained Si-rich region on coating surface; γ-Al_2O_3 phase promoted	
α-Al_2O_3 (1.5)	Na_2SiO_3 (10); $(NaPO_3)_6$ (6); $NaAlO_2$ (1.5)		Increased coating rate; increased hardness; reduced porosity and roughness	[100]
Cr_2O_3 (1.5)			Larger increased coating rate; increased hardness; slightly reduced porosity and roughness	
h-BN (1.5)			The largest increased coating rate; increased hardness; reduced porosity and roughness; more effectively enhanced weak resistance than those by α-Al_2O_3 and Cr_2O_3	
WC (5)	Na_3PO_4 (4); KOH (1.5)	Mg-alloy (AZ31B)	Reduced mean pore diameters by ~15% and surface porosity by ~6%; friction coefficient reduced by ~45%; increased hardness and wear resistance	[101]
α-Al_2O_3 (2, 10)	Na_2SiO_3 (10.5); KOH (2.8)	Al-alloy (6082-T6)	Increased α-Al_2O_3 in the coatings and more in the outer layer; similar friction coefficient to hard chrome; increased hardness and wear resistance	[102]
Hydroxyapatite (10)	Na_2HPO_4 (6)	Ti-6Al-4V	Porous network with embedded hydroxyapatite nanoparticles; higher corrosion resistance	[103]
Hydroxyapatite (5~20)	Na_3PO_4 (10); KOH (2)	Mg metal	Reduced thickness and porosity; increased corrosion resistance; precipitation and deposition of CaP further enhanced the corrosion resistance by sealing effect	[104]
Graphene (2)	Na_2SiO_3 (11.65); NaOH (1.42); $Na_6O_{18}P_6$ (6); $C_3H_8O_3$ (2.5)	Al-alloy (D16T)	Graphene embedded in the coating; flatter, denser surface; increased hardness and thickness; increased corrosion resistance	[105]
MoS_2 (15); $CH_3(CH_2)_{11}N(CH_3)_3Cl$	$HCONH_2$ (900);	Ti-6Al-4V	MoS_2-Ti(C, N)-TiO_2 coating; slightly increased hardness;	[106]

(8)	KCl (10)		enhanced tribological properties	
h-BN (2, 8)	Na_2SiO_3 (20); $(NaPO_3)_6$ (8); NaOH (2)	Ti-6Al-4V	Rutile TiO_2, anatase TiO_2, and h-BN composites; reduced porosity; increased thickness and roughness; better density, interface adhesion, and wear resistance with 2 g/l h-BN	[107]
Clay (5)	$NaAlO_2$ (12); Na_3PO_4 (8); KOH (1)	Mg-alloy (AM50)	Bigger closed pores and rougher surface; higher wear rate and friction coefficient at higher load; increased amorphous and corrosion resistance	[108]
Clay (5)	Na_3PO_4 (10); KOH (1)	Mg-alloy (AM50)	Induced amorphous coatings and better corrosion resistance; crystallinity, thickness, and porosity increased at higher current density	[109]

5. Advances in the development of PEO

Extensive investigations have been carried out to develop PEO techniques in the past decades, addressing the processing parameters, including power mode and waveform profiles, selecting and engineering the concentration of electrolytes, and choosing and modifying electrolytic additives. This, together with the green and sustainable characteristics of the PEO process, has led to its entry into industrial applications, particularly for protective coatings on Al- and Ti-based alloy components. Similar to many other coating technologies, the PEO process is also designed to be fit-for-purpose, which drives the need for advanced PEO developments tailored to specific applications. Alongside the ever-increasing development of additive manufacturing with enhanced

integrity, the importance of advanced PEO processes has grown because of their potential to coat complex and hollow metallic structures in a conformal-like manner.

Hybrid techniques, developed by combining coating processes with pre- and/or post-coating treatments [19, 20, 32, 81, 110], have also been introduced into PEO technology. Laser texturing of the substrate and MoS_2-deposition have been introduced by Qin et al. on a Ti-6Al-4V substrate before and after the PEO process, respectively [83]; the former was used to form micro-dimple arrays, while the later was serviced as solid lubricate [67, 111-113]. The hybrid process significantly enhanced the wear resistance and tribological performance of Ti-6Al-4V alloy substrate, in which the micro-dimple arrays acted as reservoir for MoS_2 solid lubricants, as well as sinks for wear debris [83, 114]. It is worth mentioning that surface crater structures can also be formed by peening or micromachining, while self-lubrication can be engineered into PEO coatings through electrolyte modifications [98, 115-118]. Polymer coatings have also been developed as post-PEO treatment methods to enhance the performance of PEO coatings, which exhibit significant enhancement in the corrosion and tribocorrosion resistances of PEO coatings on Mg alloy substrates under bioactive conditions [119, 120].

The conformal-like coating of PEO process has attracted increased interest in surface engineering and protection of additively manufactured metal alloys [79, 121, 122]. Although additive manufacturing, such as the 3D-printing of metal alloys, has entered industrial applications, differences in microstructure and properties exist between additively manufactured alloys and their counterparts produced by traditional methods, such as casting, arc-melting, and wrought techniques [123-125]. For example, the relatively low wear resistance of Ti alloys manufactured via selective laser melting (LSM)

hinders their application. PEO is proposed as a promising method to enhance the wear resistance of additively manufactured Ti alloys by forming dense oxide coatings [121]. By comparing the PEO process on LSM- and traditionally cast-AlSi10Mg alloys, Pezzato et al. found that a continuous oxide coating can be formed by PEO on SLM-AlSi10Mg; however, the morphology of the PEO coatings was strongly influenced by the microstructure of the substrate [79, 122]. The lower porosity of LSM alloys resulted in more homogeneous PEO coatings with reduced porosity and, therefore, enhanced corrosion resistance [79].

High- and medium-entropy alloys (HEA and MEA) have attracted increasing interest in recent years because of their unique crystallographic and metallurgical characteristics [31, 61]. The presence of multi-principal elements in HEA and MEA might result in competing oxidation reactions and near-surface localized atomic diffusion during the PEO process [126, 127]. Zhang et al. studied PEO process on TiNbZr MEA by varying the applied voltage and obtained homogeneous coating matrix consisted of amorphous Ti-, Nb-, and Zr-oxide originated from the substrate and amorphous CaO, $Ca(OH)_2$, SiO_2 and $CaSiO_3$ originated from the electrolyte [126]. An increased process voltage led to the formation of tetragonal-ZrO_2 nanocrystals scattered in amorphous matrix of the coating, which enhanced the wear and corrosion resistance. However, a further increase in the process voltage increased the crystallinity with major tetragonal-ZrO_2 and minor rutile-TiO2, as well as nano-sized or amorphous regions, leading to a decreased protection performance [126]. In a similar PEO process, the application of a relatively low processing voltage (150 V) resulted in amorphous oxide coatings on the TiNbTaZrHf HEA, involving multiple metal elements from the HEA substrate and Ca, Si,

Na, and O from the electrolyte [127]. The PEO coatings exhibited excellent corrosion and wear resistance. However, increased processing voltage (≥ 200 V) led to $Hf_{0.5}Zr_{0.5}O_2$ nanocrystals, which were homogeneously scattered in the amorphous coating matrix, leading to degraded wear and corrosion resistance [127].

Along with extensive experimental studies of PEO process, theoretical investigations also play a significant role in providing insights into the fundamentals and advancements of PEO technology [128-131]. These investigations explored the relationship among various aspects, including the processing conditions, such as, the electrolyte and additives, as well as the processing parameters; the properties of the coatings, such as microstructure, composition, phase constituents, hardness, and friction coefficient; and the performance of the coatings, including corrosion and tribocorrosion resistance, as well as wear resistance, under different conditions. For example, molecular dynamics (MD) simulations could provide insights into the interface states of graphite inclusions from the electrolyte and the induced self-lubricating mechanism of PEO coatings [132]. The phase composition of γ-Al_2O_3, α-Al_2O_3, graphite, and amorphous alumina can also be estimated and/or interpreted in terms of the PEO conditions on a pure Al substrate [132].

Finally, recent developments in machine learning and artificial intelligence alongside automation in experimental data collection have shown great potential in accelerating the development of the PEO process [133-136]. To predict the coating thickness using the PEO process, machine learning and neural network models have been developed to identify the most important factors for determining the specified coating properties and their relationship with respect to the PEO parameters [133]. A

combination of accelerated electrochemical tests and artificial neural networks was recently proposed by Finke et al. to predict the salt spray test results of PEO coatings on Al-alloys [136]. A mathematical model of PEO process can be built through parametric identification based on PEO experiments, which can be further used to develop an intelligent hardware-software system aimed at the production of protective coatings by the PEO process [135, 137].

6. Conclusions and outlook

As an eco-friendly surface modification and ceramic coating technique, PEO has been extensively investigated in the past decade owing to its promising applications in the production of protective coatings on metallic components in various industrial sectors. The PEO process typically involves physical chemistry, specifically associated with electro-, thermo-, and plasma dynamics, leading to various electrochemical reactions simultaneously occurring on the surface of the metal substrates in the electrolyte under iterative ultrashort-pulsed plasma conditions. The metal ions from the substrate, oxygen and other elemental species from the electrolyte react on the substrate surface to form a thick, porous oxide layer. As the PEO process continues, the oxide layer becomes thicker and dense, leading to improved hardness, wear and corrosion resistances and, therefore, enhanced protection performance. Although majority of PEO investigations and commercialized PEO processes have focused on valve alloys such as Al-, Ti-, and Mg-based alloys, recent developments in the PEO process have expended to non-valve alloys such as low-carbon steel, stainless, steels, Zr-, and Ni-based alloys.

The structure of PEO coatings is strongly dependent on processing parameters, including power application mode, waveform profiles, electrolyte compositions, and application of electrolytic additives. Surface structure and elemental compositions may influence PEO coatings, especially at the initial coating stage, which leads to increased interest in the PEO process on HEA and MEA substrates as well as hybrid processes by introducing substrate treatment before the PEO process. The conformal-like coating of PEO process has also attracted increased interest in additive manufacturing, where 3D surfaces and/or hollow structures poses great challenge to traditional coating technologies such as physical/chemical vapor deposition and thermal/cold spray.

Recently, theoretical studies of PEO have also been conducted in connection with experimental observations, addressing the coating mechanisms with insight into the relaxation kinetics of plasma and the role of negative currents in the waveform profiles. Molecular dynamics simulations have been applied to understand the interface states of micro- and nanoscale inclusions, originating from electrolyte, within PEO coatings, as well as their effects on resulting corrosion and tribological properties. Machine learning and artificial intelligence techniques have also been applied to recent advances in PEO technologies. Through a combination of accelerated experimental data collection and artificial neural networks, the protective performance of PEO coatings can be theoretically predicted. Parametric identification from experiments can be applied for the mathematical modelling of PEO process, which can be further applied for developing intelligent hardware-software systems for advanced PEO processes. The emerging intelligent PEO technology, which still has concern, could be the best bet for PEO coatings.

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