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Micro-Arc Oxidation of Magnesium-Based Anodes for Air-Battery Applications: A Review

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Abstract

Magnesium-air batteries are promising low-cost energy systems because of the abundance, high theoretical energy density, and relative safety of magnesium. However, their practical application is limited by severe self-corrosion of the magnesium anode, hydrogen evolution, and the formation of insulating discharge products that increase polarization and reduce anode utilization. This review discusses micro-arc oxidation (MAO), also known as plasma electrolytic oxidation, as a surface-engineering strategy for magnesium-based air-battery anodes. MAO coatings can form ceramic oxide layers that suppress electrolyte penetration, regulate interfacial reactions, and improve corrosion resistance. The review summarizes the working principles of magnesium-air batteries, the formation mechanism and structure of MAO coatings, and the effects of electrolyte additives, electrical parameters, substrate pretreatment, and composite sealing strategies. Remaining challenges, including coating uniformity, long-term stability, self-discharge, process cost, and the balance between protection and discharge activity, are analyzed. The review emphasizes that although MAO shows promise for Mg-air anodes, more battery-specific testing is required to verify its practical effectiveness.

Keywords

Micro-arc Oxidation; Magnesium-air Battery; Magnesium Anode; Corrosion Protection; Composite Coating.

1. Introduction

Magnesium-air (Mg-air) batteries are widely regarded as promising low-cost electrochemical energy systems because magnesium is abundant, has a high theoretical energy density, and offers relatively favorable safety characteristics compared with many lithium-based systems. A typical Mg-air cell uses magnesium or a magnesium alloy as the anode and consumes oxygen from air at the cathode during discharge, giving the system advantages in environmental compatibility and volumetric energy density. These characteristics make Mg-air batteries attractive for portable, wearable, emergency, and other distributed-power scenarios [1]. However, practical development remains constrained by two coupled problems. First, the magnesium anode corrodes spontaneously in aqueous electrolytes, lowering discharge efficiency and causing irreversible loss of active material. Second, discharge often produces insulating or semi-insulating surface films, mainly $Mg(OH)_2$ and related oxide/product layers, which reduce the active reaction interface, increase charge-transfer resistance, and intensify polarization [2].

To address these problems, previous studies have explored alloying, microstructure control, electrolyte additives, gel electrolytes, and surface coatings. Among these strategies, surface engineering is particularly attractive because it directly modifies the anode/electrolyte interface where corrosion and discharge reactions occur. The central challenge is therefore not simply to prevent corrosion, but to control the metal/electrolyte interface without completely blocking the electrochemical reaction. The protective layer should reduce self-corrosion and hydrogen evolution while still allowing sufficient ionic transport, electron transfer through the current path, and controlled product-film evolution during discharge. Micro-arc oxidation (MAO), also known as plasma electrolytic oxidation (PEO), provides a useful surface-engineering route for this purpose. Under high-voltage microdischarge, MAO forms a ceramic oxide layer on reactive metals such as magnesium. By adjusting pore sealing, densification, and composite coating strategies, MAO can combine corrosion shielding with interfacial regulation and thus offers a plausible route for Mg-air anode protection [4].

Research on MAO/PEO of magnesium alloys has mainly focused on corrosion resistance, wear resistance, antibacterial activity, bioactivity, and composite functional coatings. Reviews show that MAO coatings on magnesium are typically composed of MgO and silicate- or phosphate-containing phases, and that their properties depend strongly on electrolyte chemistry, additives, electrical parameters, treatment time, and pulse energy [4]. Dense inner layers generally improve protection, whereas micro-pores and micro-cracks may become pathways for electrolyte penetration or crack initiation under mechanical stress. As a result, coating densification and defect control are recurring themes in the literature [5,6].

Electrochemical studies commonly use potentiodynamic polarization, electrochemical impedance spectroscopy (EIS), immersion tests, salt-spray exposure, and hydrogen-evolution measurements to evaluate coating performance [7]. In parallel, Mg-air battery studies emphasize that the poor reversibility of rechargeable Mg-air systems is closely associated with insulating MgO or $Mg(OH)_2$ product layers, which increase overpotential and polarization [2]. These two research lines are converging. MAO coatings with nano- or micro-porous structures can be further sealed by metal-organic frameworks (MOFs), carbon nanomaterials, nanoparticles, electroless metal layers, or other post-treatments, thereby improving barrier properties and electrochemical stability [8-10]. At the same time, Mg-air anode performance is also governed by substrate microstructure, including grains, orientation, second phases, and heat-treatment state; MAO should therefore be considered not as an isolated coating, but as part of a substrate-coating-electrolyte interface design [11].

This review summarizes the application logic of MAO for magnesium-based air-battery anodes. It focuses on the working principles of Mg-air batteries, the coating structure generated by MAO, the relationship between coating defects and corrosion/electrochemical behavior, the effects of electrolyte additives and electrical parameters, and the remaining challenges for battery-oriented use. The discussion is condensed around mechanisms, evidence, and practical constraints. However, direct studies on MAO-coated magnesium anodes in complete Mg-air batteries remain limited, and many conclusions must be inferred from corrosion, coating, and interface studies. Therefore, a critical review is needed to connect MAO coating mechanisms with Mg-air battery requirements.

2. Fundamentals and Structure of Magnesium-Air Batteries

In neutral or alkaline aqueous electrolytes, the main anodic reaction can be written as $Mg \rightarrow Mg^{2+} + 2e^-$, followed by the formation of $Mg(OH)_2$ through hydrolysis or precipitation. The cathodic process involves oxygen reduction, such as $O_2 + 2H_2O + 4e^- \rightarrow 4OH^-$ in alkaline media. A parasitic reaction, $Mg + 2H_2O \rightarrow Mg(OH)_2 + H_2$, competes with useful discharge and causes self-corrosion. The discharge of a Mg-air battery involves anodic dissolution of magnesium and cathodic oxygen reduction. At the anode, magnesium loses electrons and forms Mg^{2+} or related surface products; at the air cathode, oxygen reduction closes the external circuit. In aqueous or near-neutral electrolytes, the local pH, ion migration, and product solubility strongly influence the surface reaction pathway. Because magnesium is highly active, self-corrosion occurs in parallel with useful discharge, consuming active material and reducing anodic efficiency. Hydrogen evolution is a particularly important side reaction because it wastes electrochemical activity and can accelerate degradation of the anode/electrolyte interface.

A second limitation is the formation of passivating product layers. $Mg(OH)_2$ and related oxide or peroxide products may form dense or semi-dense films on the anode surface. These layers may temporarily suppress corrosion, but they also block mass transport and increase interfacial impedance, resulting in voltage decay, higher overpotential, and poor reversibility. For rechargeable Mg-air batteries, insulating MgO or $Mg(OH)_2$ product layers are regarded as a major origin of high polarization and low cycling efficiency [2]. This explains why an ideal protection strategy must balance two conflicting requirements: it must suppress self-corrosion and hydrogen evolution, but it must not form an impermeable layer that disables discharge.

A practical Mg-air cell normally includes a magnesium or magnesium-alloy anode, an air cathode with a carbon-based catalytic layer, current collector and gas-diffusion structure, an electrolyte, and a separator or mechanical support. Pure magnesium and Mg alloys such as AZ-series, Mg-Ca, and Mg-Zn systems have been used to balance reactivity, mechanical processing, and corrosion behavior. The anode microstructure matters because grain size, orientation, twins, dislocations, and second phases can change localized corrosion and hydrogen evolution. Recent work on microstructure design for advanced Mg-air anodes therefore argues that substrate engineering is as important as electrolyte or cathode optimization [12].

Electrolyte composition and additives also affect product-film formation. Some additives regulate pH, complex Mg^{2+}, or modify $Mg(OH)_2$ deposition, thereby improving voltage stability and reducing polarization [13]. In this coupled environment, MAO acts as a solid interface-engineering method. It changes surface chemistry, roughness, pore distribution, and the continuity of defects, and it may also guide how discharge products deposit or detach. If the coating suppresses uncontrolled electrolyte penetration while retaining controlled reaction pathways, it can contribute to higher anodic efficiency, more stable discharge voltage, lower self-discharge, and better cycling stability.

The main performance indices of Mg-air batteries include anodic utilization efficiency, discharge voltage, specific capacity, energy density, polarization behavior, self-discharge rate, and cycle stability. In flexible or wearable configurations, additional requirements such as mechanical integrity and deformation tolerance are important. Dual-layer gel electrolyte designs, for example, have been reported to mitigate both magnesium corrosion and dense passivation-film formation, enabling high specific capacity and energy density in flexible fiber-shaped cells [14]. This example highlights a key point for MAO design: coating protection must be coordinated with electrolyte and cell architecture instead of being evaluated only by static corrosion resistance

3. Micro-Arc Oxidation of Magnesium: Principles and Coating Formation

MAO/PEO is based on dielectric breakdown and localized plasma discharge between a metal substrate and an electrolyte under high voltage. On magnesium alloys, MAO is usually performed in alkaline silicate, phosphate, or other electrolyte systems. The process produces a ceramic coating mainly containing MgO, and depending on electrolyte chemistry, phases such as Mg_2SiO_4 or other phosphate/silicate compounds may also form. The coating grows through repeated discharge, melting, oxidation, ejection, and rapid solidification. The resulting surface is typically porous, while the inner region is more compact and strongly bonded to the substrate [15].

Coating growth is highly sensitive to electrical parameters. Current density, treatment time, pulse frequency, duty cycle, and pulse energy determine the number, intensity, and distribution of microdischarge channels. Changes in discharge behavior affect coating thickness, pore size, micro-crack density, and the ratio between porous and compact layers. Studies on AZ31B magnesium alloy show that lower current density can produce a more uniform and dense structure, while longer treatment time increases thickness and salt-spray protection. Ultra-audio-frequency pulsed MAO can delay arc initiation, reduce pore size, improve surface uniformity, raise corrosion potential, and lower corrosion current density by several orders of magnitude; MgO and Mg_2SiO_4 are usually the main phases in such silicate systems.

PEO coatings often display a multilayer or gradient structure. Comparative studies on AZ31 and AZ91 show that coatings may evolve from a two-layer structure into three layers with increasing treatment time: an outer layer, an inner layer, and an anodic layer. The formation of these layers is associated with vertical discharge pores, horizontal pore interfaces, local electrolyte heating, etching reactions, and inner-layer growth or dissolution. AZ91 can form a denser protective structure under certain conditions because its aluminum content changes inner-layer formation and dissolution. However, once corrosion is initiated, the corrosion propagation behavior may differ from AZ31, indicating that the substrate continues to influence coating performance after the coating has formed.

The coating defects that are tolerated in ordinary corrosion protection may become critical in battery operation. Micro-pores and micro-cracks can serve as paths for electrolyte penetration, local corrosion, hydrogen evolution, and crack initiation. In corrosion-fatigue conditions, these defects may reduce service life even when static corrosion resistance improves. Battery anodes can experience local stress, repeated deposition/dissolution, swelling, and interfacial stress from product layers. Therefore, MAO coating design for Mg-air batteries must evaluate both electrochemical and mechanical stability.

Process innovations also influence coating composition and defect chemistry. Laser-assisted PEO, for example, can modify discharge evolution, increase the proportion of corrosion-resistant Mg_2SiO_4, reduce oxygen-vacancy defects and micro-cracks, and improve corrosion resistance [16]. Low-energy PEO studies further suggest that electrolyte chemistry can control discharge behavior and coating growth, which is important for lowering the energy cost of large-scale MAO production [17]. Overall, battery-oriented MAO requires careful control of pore connectivity, compact-layer continuity, phase composition, and interfacial adhesion.

4. Potential Application of MAO Magnesium-Based Materials in Mg-Air Batteries

Because MAO is mainly applied to magnesium substrates, the following discussion focuses on the anode side of Mg-air batteries rather than air-cathode catalysts or oxygen-reduction mechanisms. The most direct contribution of MAO to Mg-air batteries is anode protection. By forming a ceramic barrier between the electrolyte and the active substrate, MAO can reduce direct attack by solution species, suppress localized corrosion, and lower hydrogen evolution. Although many MAO studies are conducted in corrosion or biomedical environments rather than complete battery cells, their methods and conclusions are highly relevant because Mg-air anodes face the same essential problem: aqueous electrolyte penetration into a reactive magnesium surface. For example, tannic acid modified MAO coatings on AZ31 magnesium alloy have been reported to produce thicker coatings with smaller pores and fewer micro-cracks than unmodified MAO, leading to better corrosion resistance and antibacterial performance. The battery relevance lies in the mechanism: reducing pore size and eliminating connected defects should limit aggressive ion penetration and lower local corrosion and hydrogen evolution. Similarly, MAO/Ni-P duplex coatings show that the MAO pretreatment and electroless Ni-P sealing can markedly influence pore uniformity, roughness, and corrosion rate in 3.5 wt.% NaCl [18]. Such duplex concepts are useful for Mg-air systems because cycling electrolytes can repeatedly enter and leave coating defects; sealing and composite layers increase the tortuosity of penetration paths and delay substrate exposure. However, improved corrosion resistance does not necessarily guarantee better battery performance, because an overly compact coating may also increase interfacial resistance and reduce discharge activity. Therefore, these coating strategies should be further evaluated by discharge voltage, anodic efficiency, hydrogen evolution, and impedance evolution in actual Mg-air cells.

Surface morphology also affects electrochemical behavior. The porous ceramic layer may increase apparent surface area and wettability, which can be beneficial for discharge kinetics, but excessive porosity creates corrosion channels. A useful coating therefore needs controlled porosity rather than simple maximum densification. MAO/graphene oxide (GO) composite coatings provide an illustrative case. GO sheets can cover and seal micro-pores in the MAO layer, producing a smoother multilayer structure with more uniform element distribution and substantially higher corrosion resistance than single MAO coatings, although coating adhesion still needs improvement. In a Mg-air anode, such nanoscale sealing may reduce self-corrosion while retaining limited ionic transport, but this effect depends strongly on coating thickness, pore connectivity, adhesion, and electrolyte composition.

Particle incorporation and carbon-based modification can further tune the MAO surface. Carbon-sphere additions in the electrolyte change coating thickness, pore size, open porosity, hardness, friction behavior, and corrosion resistance; the formation of SiC-related phases has been associated with improved hardness and more stable friction coefficients [19]. Graphene additions to PEO electrolytes can reduce the number and size of pores and cracks, increase microhardness, and lower corrosion current density by orders of magnitude [20]. Although wear resistance is not the main metric of a Mg-air battery, mechanical robustness matters because assembly pressure, electrolyte flow, and repeated surface-product formation can expand coating defects during service. Corrosion and mechanical property data on PEO-coated AZ80 also indicate that coating protection should be assessed together with mechanical integrity [21].

Coating architecture also influences cycling stability. The porous-outer/compact-inner configuration can redistribute electrolyte access and reaction sites. When the compact layer is too thin or discontinuous, corrosion protection is weak; when the coating is too thick or too impermeable, discharge may be hindered. Studies of AZ31 and AZ91 indicate that significant corrosion resistance is associated with sufficient coating thickness and the formation of a compact inner layer. Battery use therefore requires a balance between barrier protection and electrochemical accessibility.

Pore sealing is one of the most promising strategies for improving long-term stability. ZIF-67 films grown on MAO coatings can uniformly seal pores, increase the tortuosity of corrosive-medium penetration, and improve corrosion resistance on AZ31 magnesium alloy. MAO/LDH self-healing coatings provide another route: low-temperature in-situ grown Mg-Al layered double hydroxide can cover the MAO coating, fill defects, and lower corrosion current density by several orders of magnitude, with a wide passive region appearing in anodic polarization [22]. This self-healing or re-passivation idea is especially relevant to Mg-air batteries, where local damage and product-film disruption repeatedly occur during cycling.

A further requirement is that coating performance must be interpreted together with the substrate. The efficiency of PEO on magnesium alloys containing Gd and Y depends strongly on metallurgical state. T4-treated substrates can produce thicker coatings with lower porosity and better corrosion resistance, whereas precipitates under T6 conditions may induce stronger cathodic microdischarges and damage coating growth. For Mg-air batteries, substrate microstructure also controls galvanic coupling, hydrogen evolution, and utilization efficiency. Thus, MAO should be integrated with alloy composition, heat treatment, and electrolyte formulation.

5. Conclusion

MAO provides a meaningful surface-engineering strategy for magnesium-based air-battery anodes because it can form a ceramic oxide boundary that reduces self-corrosion and hydrogen evolution while offering a tunable interface for product-film evolution. The major limitations of Mg-air batteries--spontaneous corrosion, hydrogen evolution, passivating discharge products, and polarization--are directly related to the same surface processes that MAO can modify. The effectiveness of MAO depends on pore structure, compact-layer continuity, phase composition, defect density, and adhesion to the magnesium substrate. The reviewed studies show that MAO/PEO coatings on magnesium alloys normally possess multilayer structures whose thickness and density depend on electrical parameters, treatment time, electrolyte chemistry, and substrate metallurgical state. Additives such as tannic acid can reduce pores and micro-cracks and improve corrosion resistance. Carbon materials, particles, and composite sealing layers can further improve barrier performance, as demonstrated by GO, graphene, carbon-sphere, TiC, MOF, Ni-P, and LDH-based strategies. Pulse-parameter control and low-energy processing are also important for reducing defect size, improving uniformity, and lowering cost. For air-battery use, however, MAO should not be treated as a simple passive coating. Its role is to mediate a dynamic substrate-coating-electrolyte interface. Excessively dense coatings may suppress useful discharge, whereas highly porous coatings may accelerate self-corrosion. The practical goal is a controlled, stable, and selectively permeable interface that balances corrosion inhibition with electrochemical activity. This goal requires the joint optimization of substrate microstructure, MAO process, pore sealing, electrolyte additives, and cell architecture.

The remaining barriers are coating uniformity, long-term stability, defect-induced failure, self-discharge, and process cost. Realistic evaluation should combine corrosion tests with battery-level measurements, including hydrogen evolution, discharge polarization, impedance evolution, cycling or intermittent discharge, and post-mortem analysis of coating and product layers. With suitable densification, composite sealing, and electrolyte coordination, MAO has the potential to improve magnesium anode utilization and reliability in Mg-air batteries, but further battery-specific evidence is still needed before its advantages can be fully translated into engineering applications.

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