

Editorial

Summary of Publications in the Special Issue: Advances in Corrosion Resistant Coatings

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This editorial provides an outline of publications in the Special Issue on corrosion resistance coatings. It is an updated version of the content summarized in an earlier editorial [1]. The second volume or edition of this Special Issue, “Advances in Corrosion-Resistant Coatings”, is a part of the featured section “Corrosion, Wear and Erosion” in the *Coatings* journal (ISSN 2079-6412). The objective of this Special Issue is to provide a core platform of ideas regarding basic and applied research on corrosion-resistant films and coatings, including the related processing technologies, forms of corrosion, environmental responses, new metallic, non-metallic, and composite coatings, the microstructural examination of various protective coatings, and thin films with useful functions. Sixteen publications are collected in this Special Issue, including one editorial, one communication, one review, and thirteen original research papers. All of them have been cited. An updated summary is given as follows.

Amorphous materials have disordered atomic structures [2]. The absence of grain boundaries and crystal defects allows them to be more corrosion-resistant than conventional materials [3,4]. In the communication, corrosion of an amorphous alloy coating is presented by Wen, Wang, and Ren, as can be found in [5]. The $\text{Al}_{86}\text{Ni}_6\text{Y}_{4.5}\text{Co}_2\text{La}_{1.5}$ amorphous coating was prepared on a ZM5 magnesium alloy substrate by high-velocity air fuel (HVOF) spray. The coating contained 75.8% (in volume) of the amorphous phase in addition to the crystalline phases of $\alpha\text{-Al}$, Al_4NiY , and $\text{Al}_9\text{Ni}_5\text{Y}_3$. The microhardness of the coating reached 420 HV_{0.05}. The coating survived 500 h neutral salt spray tests without apparent corrosion. Moreover, the coating exhibited a very high corrosion potential. Its corrosion current density is two orders of magnitude less than that of the substrate. Such improvements are attributed to the compact coating structure and the passivation film formed on the surface of the substrate.

Corrosion-resistant transition metal oxide coatings for thermoelectric energy conversion are presented in the review [6]. Thermoelectricity permits the direct conversion between heat and electricity, driving the applications of advanced materials and devices for waste heat recovery, solid-state cooling, temperature sensing, and thermal management [7]. Enhancing the energy conversion efficiency of thermoelectric devices (TEDs) requires both the advancement of material composition and structure and innovation in device design. In addition to Bi_2Te_3 -based materials [8], oxide thermoelectric coatings for electricity generation from waste heat have been considered [9,10]. Such a unique function permits thermoelectric units to be used for energy conversion, temperature sensing, and thermal imaging. Transition metallic oxide films and/or coatings have the advantage of corrosion resistant at elevated temperatures in an oxidative environment; they show the capability to scatter phonons. In addition, they possess strong quantum confinement behavior. Therefore, thermoelectric oxide coatings were used as new materials for thermoelectricity.



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They are especially suitable for energy conversion and temperature measurement under harsh service conditions. This review paper provides a comparison on the thermoelectric properties of various oxides and other materials and demonstrates the advantages of transition metal oxides. Typical processing technologies are briefly discussed. These processing technologies are used to make thermoelectric oxide coatings in the form of thin layers, superlattice films, and nanograin powder coatings. Specifically, liquid-phase deposition, chemical vapor deposition, nanocasting, solid-state approaches, physical vapor deposition, and energy beam techniques are mentioned. The micro- and nanostructures and thermal/electrical transport properties of the processed transition metal oxide coatings are presented. Moreover, the concepts of the devices and the applications of oxide coatings for temperature sensing, thermoelectric energy conversion, and thermal imaging are shown. Perspectives of further research on transition metal oxide thermoelectric coatings are described.

Each of the thirteen original research papers represents a direction detailing an aspect of a new coating material, an interesting application, an innovative coating processing technique, or a new characterization or evaluation method. For example, Yang et al. [11] studied the microstructure, wear and corrosion resistance of a CrN coating doped with noble metals, Pt and Ir. The Pt and Ir co-doping approach adopted to enhance the corrosion resistance of the CrN coating was detailed in the article. The CrN coating, deposited using plasma-enhanced magnetron sputtering (PEMS) [12], showed a coherent growth pattern. A relatively high mechanical strength and large grain sizes were obtained. During the corrosion tests on the undoped CrN coating, corrosive fluids penetrated and filled into the locations with defects, resulting in a low corrosion resistance of the coating. Co-doping Pt and Ir atoms suppressed the coherent grain growth and allowed formation of a uniform and smooth surface. Consequently, localized non-coherent lattice growth happened due to the function of the Pt and Ir dopants in the coating. This enhanced the mechanical performance of the doped coating. Multidirectional web-like fractures were observed through the scratch tests on the doped coating. The surface of the dense coating prevented the penetration of corrosive fluids and increased the corrosion resistance of the coating.

The corrosion protection mechanism of nitrite-modified CaAl-layered double hydroxide (LDH) in epoxy coatings was investigated by Xue et al. [13]. First, nitrite and molybdate-modified Ca-Al layered double hydroxide (CaAl-LDH) was prepared. Then, the corrosion protection mechanism of the CaAl-LDH intercalated with nitrites in epoxy coatings was studied. Scanning electronic microscopy (SEM) was used to observe the morphology and energy dispersive spectroscopy (EDS) was used to analyze the elemental distribution in the synthesized material. Information on chemical composition was obtained using Fourier transform infrared spectroscopy (FTIR). The structure was characterized by X-ray diffraction (XRD). The SEM and XRD results revealed that the LDH structure was destroyed in the molybdate modification process. Chemical reaction-generated CaMoO_4 , observed as a precipitate, was seen. It has been found that molybdates cannot be loaded into the CaAl-LDH interlayer space to produce an active corrosion inhibition component. The nitrite release curve and the chloride concentration decrease curve were plotted. The anion-exchange reaction was revealed by UV-Vis spectroscopy. The corrosion protection effect of the CaAl-LDH loaded with nitrites towards the carbon steel was evaluated in 0.02 M NaCl solution by electrochemical impedance spectroscopy (EIS). The material in powder form was incorporated into the epoxy coating at a concentration of 2% in weight. The morphology and roughness of the coating were examined by SEM and laser microscopy. The corrosion protection effect was investigated by EIS with an immersion time of 21 days. The fitted coating resistance of the sample with 2% LDH intercalated with nitrites was one order of magnitude higher than that of the sample with 2% LDH. Local electrochemical

impedance spectra (LEIS) were obtained to monitor the corrosion development in micro-corrosion sites. The corrosion product of the scratched area after salt spray exposure was analyzed by EDS and Raman spectroscopy. The corrosion protection mechanism of the CaAl-LDH loaded with nitrites was proposed based on the experimental results.

Cui and Yang [14] studied the structure, mechanical properties, and corrosion behavior of a nanocomposite CrSiN/CrN/Cr coating deposited on the AZ31 magnesium alloy by plasma electrolytic oxidation (PEO). Plasma electrolytic oxidation (PEO) is a new surface treatment process which can create very hard, wear- and corrosion-resistant oxide coatings on metallic substrates, particularly light alloys like aluminum, magnesium, and titanium. It involves submerging the part in an electrolyte bath and applying a high voltage, which generates plasma discharges on the surface. This results in a dense, multi-layered ceramic-like oxide coating. Such a unique technique allows the deposition of coatings in complex shapes. The PEO approach can provide superior properties to those provided by traditional approaches like anodizing [15]. To improve the surface properties of Mg alloys and expand the applications of CrN-based materials, composite CrSiN coatings consisting of amorphous Si_3N_4 and nano CrN phases were deposited on AZ31 based on the theory of fine grain strengthening and multigrain boundaries. The effect of the thickness of the coating on the structure and properties was investigated. The microstructure was analyzed by means of X-ray diffraction (XRD) technique, scanning electron microscopy (SEM) and transmission electron microscopy (TEM). The mechanical properties, adhesion properties, and corrosion resistance were investigated using a nano indenter, a scratch tester, and an electrochemical workstation. The results show that the coating consists of a face-centered cubic CrN phase. Si_3N_4 is amorphous since it could not be observed in the diffraction pattern. The high-resolution transmission electron microscopic (HRTEM) images show a composite structure consisting of amorphous and nanocrystalline phases. With the increase in deposition time and coating thickness, the surface roughness decreased. The number of defects dramatically decreased as well. As a result, the interface had no visible defects. Moreover, the hardness and elastic modulus of the coating increased. The corrosion resistance also increased. The adhesion performance increased first and then decreased. The adhesion between coating and substrate reached the maximum when the sputtering time was 50 min, corresponding to a CrSiN coating thickness of about 0.8 μm .

Mozafarnia et al. [16] used plasma electrolytic oxidation (PEO) to perform surface treatment for generating a thick, adherent coating on valve metals in an environmentally friendly alkaline electrolyte. Specifically, the PEO technique was adopted to modify the surface of the AZ31 magnesium alloy. Composite coatings were formed in a phosphate-based electrolyte containing hydroxyapatite nanoparticles (NPs) and different concentrations (1, 2, 3, and 4 g/L) of TiO_2 NPs. The results showed that the incorporation of TiO_2 NPs in the composite coatings increased the porosity, coating thickness, surface roughness, and surface wettability of the coatings. The corrosion-resistance results of coatings in simulated body fluid (SBF) were obtained based on the tests performed for up to 72 h. All coatings showed superior corrosion resistance compared to the substrate. Among samples containing TiO_2 , the one containing 1 g/L TiO_2 had the highest inner layer resistance and outer resistance and the lowest average friction coefficient. It showed the best wear and corrosion resistance performance. The antibacterial tests showed that the higher the concentration of TiO_2 NPs, the lower the adhesion of bacteria.

High-entropy alloys (HEAs) are complex materials. Unlike traditional alloys, such new materials are made by mixing five or more principal elements in roughly equal proportions. Their properties are not dominated by one or two elements. The unique composition allows HEAs to have superior properties such as high strength, wear resistance, and thermal stability, making them ideal for demanding applications in aerospace and high-temperature

environments. Their enhanced properties stem from certain core effects including high distortion of lattices and slow diffusion, as described in [17]. The corrosion resistance of a laser-clad $\text{Al}_{0.7}\text{FeCoCrNiCu}_x$ high-entropy alloy coating in a marine environment was investigated by Wu and Lu [18]. The $\text{Al}_{0.7}\text{FeCoCrNiCu}_x$ high-entropy alloy coating was prepared by laser melting technology. The 5083-aluminum alloy was the base material. The aging and failure mode of the coating in marine atmospheric environment was interrogated. XRD and SEM were utilized to study the microstructure of the coatings. The laws of influence of Cu on the electrochemical corrosion behavior of the $\text{Al}_{0.7}\text{FeCoCrNiCu}_x$ system high-entropy alloy in a 3.5 wt.% neutral NaCl solution was investigated by using dynamic potential polarization and electrochemical impedance spectroscopy. Neutral salt spray acceleration tests and outdoor atmospheric exposure tests were carried out. The results show that the $\text{Al}_{0.7}\text{FeCoCrNiCu}_x$ ($x = 0$) high-entropy alloy coating has a single BCC phase structure and the $\text{Al}_{0.7}\text{FeCoCrNiCu}_x$ ($x = 0.30, 0.60, 0.80, 1.00$) high-entropy alloy coating consists of both body-centered cubic (BCC) and face-centered cubic (FCC) phases with a typical dendrite morphology. With the increase in Cu content, the self-corrosion potential of $\text{Al}_{0.7}\text{FeCoCrNiCu}_x$ gradually increases and the current density gradually decreases. The electrochemical impedance spectra revealed that the corrosion resistance of $\text{Al}_{0.7}\text{FeCoCrNiCu}_{1.00}$ is the highest. The results of the neutral salt spray acceleration test and the outdoor atmospheric exposure test were used to conduct a comprehensive evaluation of the corrosion resistance of the coating. The corrosion resistance of the $\text{Al}_{0.7}\text{FeCoCrNiCu}_x$ coating increases with the increase in Cu content. The compressive strength and plastic deformation are the best when $x = 0.80$. A neutral salt spray accelerated test showed no corrosion at 5040 h. Even if the coating was broken, it lasted up to 4320 h. In the outdoor atmospheric exposure test, which was conducted 12 months after the coating surface test, no corrosion occurred.

In the work conducted by Chen et al. [19], the oxidative corrosion of Ti_2AlNb -based alloys during alternate high temperature–salt spray exposure was investigated. Specifically, the corrosion damage mechanisms of Ti_2AlNb -based alloys under high temperature, salt spray and coupled high temperature–salt spray conditions were studied. The alloys were analyzed in detail macroscopically and microscopically by means of XRD, SEM, and EDS. The results indicated that the surface oxide layer of Ti_2AlNb -based alloys is dense and complete. The thickness of the surface layer is only 3 μm after oxidation at 650 $^\circ\text{C}$ for 400 h. The formation of a passivation film resulted in almost no damage to the Ti_2AlNb -based alloys after 50 cycles of salt spray testing at room temperature. The tests showed that Ti_2AlNb alloys were erosion-resistant at 650 $^\circ\text{C}$ in salt spray. However, the alloys had an oxide layer thickness of up to 30 μm , and corrosion pits on the surface were observed after 50 cycles of corrosion under alternating high temperature–salt spray conditions. The Cl_2 produced by the mixed salt eutectic reaction acted as a catalytic carrier to accelerate the volatilization of the chloride in the oxide layer. Re-oxidation of the substrate was observed. In addition, the growth of unprotected corrosion products (Na_2TiO_3 , NaNbO_3 and AlNbO_4) caused changes in the internal structure of the oxide layer, suppressing the surface densification and causing severe damage to the alloy surface.

In the paper written by Zhang et al. [20], the effects of NH_4HF_2 , H_3PO_4 , phytic acid (IP6), and EDTA- ZnNa_2 concentration on corrosion resistance and the Zn amount in micro-arc oxidation (MAO) coatings were revealed by an orthogonal experiment. The influencing order of four factors on coating corrosion resistance is EDTA- $\text{ZnNa}_2 > \text{NH}_4\text{HF}_2 > \text{IP6} > \text{H}_3\text{PO}_4$, while the Zn amount decreased in the order EDTA- $\text{ZnNa}_2 > \text{NH}_4\text{HF}_2 > \text{H}_3\text{PO}_4 > \text{IP6}$. The fabricated Zn-containing coatings exhibit excellent corrosion resistance, and their i_{corr} values are two orders of magnitude lower than those of the WE43 substrate, while the highest Zn amount is 4.12 wt.%. P and F compete to take part in coating formation, and Zn ions

enter anodic coatings by diffusion. The coating corrosion resistance is jointly determined by surface characteristics, which may provide the important theoretical foundation for fabricating Zn-containing coatings with high corrosion resistance.

Liu et al. [21] investigated the role of a Cr transition layer in the hot-salt corrosion behavior of an Al-Si alloy coating. Hot-salt corrosion experiments were performed at 650 °C and corrosion kinetic curves were plotted. The weight gain of the Al-Si alloy coating increased to 0.89 mg/cm² at 100 h and then decreased steadily to 0.77 mg/cm² at 200 h. The weight of the Al-Si alloy-coated samples with the addition of a Cr transition layer increased immediately to 0.79 mg/cm² at 20 h and then gradually increased to 0.85 mg/cm² at 200 h. This Cr diffusion promoted the preferential creation of an Al₂O₃ layer, which effectively hindered the upward diffusion of Fe and resulted in the production of a Cr₂O₃-SiO₂ layer, which impeded the multi-scale salt mixture's penetration. Cr diffusion also caused a notable seal-healing effect, which healed the micro-pores. These oxidation and degradation reactions were considerably repressed by the high barrier properties of these oxide layers and the dense surface, resulting in an increased hot-salt corrosion resistance of the Al-Si alloy coating. The current findings provide a feasible strategy for the design of a diffusion barrier layer of a thermal protective coating on martensitic stainless steel.

Titanium and its alloys have been increasingly used in the aeronautical industry. It is important that they have high corrosion resistance to acidic agents and/or chlorine ions. Generally, Ti and its alloys are better than other alloys in terms of electrochemical properties because of the formation of the protective TiO₂ coating on their surfaces. In the work presented by Jáquez-Muñoz et al. [22], the corrosion behaviors of anodized titanium and its alloys with a TiO₂ protective coating are investigated. First, the paper demonstrated the procedures for the formation of the protective oxide coating. Then, the electrochemical corrosion performances of Ti and Ti alloys with the compositions of Ti-6Al-2Sn-4Zr-2Mo and Ti-6Al-4V were outlined. Anodization of pure Ti and the two alloys was performed in both diluted H₃PO₄ and H₂SO₄ (1.0 M) solutions at a current density of 250 A/m². Corrosion tests on the anodized specimens were carried out in 3.5 wt.% NaCl and 3.5 wt.% H₂SO₄ solutions at ambient temperature. SEM (scanning electron microscopy) was used to reveal the morphology of the anodized surfaces, and the electrochemical responses of the anodized coatings were examined. The results show that the Ti alloys treated in the H₃PO₄ electrolyte demonstrate electrochemical behavior associated with a uniform passive film being exposed to the 3.5 wt.% NaCl solution. The Ti alloy containing more beta-phase stabilizers produced a less uniform oxidized coating.

In the paper published by Samad et al. [23], the mechanical, thermal, nanomechanical, and electrochemical properties of epoxy coatings containing varying amounts of ZnO NPs (nanoparticles) were studied. Epoxy coatings were examined after complete curing for seven days. The dispersion of ZnO NPs in the epoxy matrix was examined by SEM (scanning electron microscopy) followed by FTIR (Fourier-transformed infrared spectroscopy) to assess the effect of the addition of ZnO NPs on the curing of epoxy. DSC (differential scanning calorimetry) was applied to study the thermal properties of the composite coatings. The electrochemical (anticorrosion) properties of the coatings were tested by immersing the prepared composite coatings into a solution containing 3.5% NaCl. The results reveal that the addition of the ZnO NPs is effective at lower loading ranges, and that higher loadings of ZnO NPs lead to particle agglomeration. In the higher NP loading ranges, the curing of epoxy is adversely affected, and an insufficient curing state is observed. The lower degree of curing adversely influences the thermal, electrochemical, and mechanical properties. The upper boundary for the ZnO nanoparticle incorporation is found to be 2%.

Burduhos-Nergis et al. [24] investigated how to enhance the corrosion properties of carbon steels used for manufacturing carabiners, considering the issue of worker safety.

Carabiners are considered crucial components in safety systems; they are used to connect various other elements within systems, building the connections between systems and anchor points. Therefore, to maximize safety, the materials used to make carabiners must have high corrosion resistance in various environments. This paper is a comprehensive study that aims to improve the corrosion properties of carbon steels for carabiners. Previous studies have proven that the corrosion resistance of carbon steels in various corrosive environments could be improved by depositing different types of phosphate coatings and other coatings. This work concentrated on evaluating the galvanic corrosion of different galvanic couples (duralumin-coated specimens, aluminum–bronze-coated specimens, and carbon–steel-coated specimens) tested in three different corrosive media. For the first time, comprehensive studies were carried out on the galvanic corrosion of potential materials for the manufacturing of carabiners. It is confirmed that the specimen coated with the zinc phosphate layer exhibits the best performance in the corrosive media, including fire-extinguishing fluids and saltwater.

The research published by Wu et al. in [25] introduced work on using transition-metal nitride coatings for protecting electronic connector devices in marine environments. Such electronic devices should have high electrical conductivity and good corrosion resistance. This study synthesized a novel CrN–Pt coating with a dense growth texture. Pt addition induced a pronounced increase in electrical conductivity and corrosion resistance. The resistivity decreased from 0.0149 Ohm·cm in the Cr–N coating to 0.000472 Ohm·cm in the CrN–Pt coating, while the corrosion current density decreased from 24 nA/cm² in the Cr–N coating to 6.3 nA/cm² in the CrN–Pt coating. The results of the above studies confirm that Pt doping has significant advantages in improving the electrical conductivity and corrosion resistance of nitride coatings for potential applications in the marine environment.

The corrosion behavior of the AA2055 aluminum–lithium alloy anodized in a sulfuric acid (H₂SO₄) bath at the current density of 0.19 and 1 A·cm^{−2} was evaluated by Samaniego-Gómez et al. [26]. The sealing solution used contains water (H₂O) and sodium dichromate (Na₂Cr₂O₇). Anodized samples were exposed to a 10 vol.% H₂SO₄ solution and the electrochemical technique used was electrochemical impedance spectroscopy. Scanning electron microscopy and X-ray photoelectron spectroscopy were applied for observing the morphology and thickness of the anodizing coating. The Na₂Cr₂O₇ sealing solution tends to increase the charge transfer resistance and produces a more homogeneous and compact passive oxide layer. Such an anodized layer increases the corrosion inhibition protection to the AA2055. SEM observations indicated that the morphology and thickness of the anodic films formed on AA2055 aluminum–lithium alloy anodized are ideal using the above-mentioned current density conditions.

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