ELECTRODEPOSITION OF NICKEL-COBALT / ALUMINA (Ni-Co/Al$_2$O$_3$) COMPOSITE COATINGS: A CURRENT APPROACH

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ABSTRACT

This paper provides a review of the electrodeposition process that is used to produce nickel-cobalt / alumina (Ni-Co/Al$_2$O$_3$) composite coatings. The electrodeposition techniques, consisting of direct current, pulse current and pulse reverse current, sediment electrodeposition, and CO$_2$ bath, are described. The effects of electrodeposition variables, including applied current density, deposition time, particle concentration and use of surfactants, are also explained. The paper then discusses the limitations of previous studies, such as lack of focus on the effectiveness of this coating in corrosive and erosive environments, and the effects of alumina particles on coating microstructures. It also suggests longer exposure to corrosive media for more significant corrosion study. Further research needs to be conducted in order to improve the corrosion and erosion resistance of Ni-Co/Al$_2$O$_3$ in harsh environments, such as marine conditions.

Keywords: nickel-cobalt / alumina composite coatings, electrodeposition, corrosion and erosion resistance, micro hardness and wear rate, particle concentration.

INTRODUCTION

Electrodeposition is a process of incorporating metals to the subtracts by using the electrochemical method. The electrodeposition technique offers several advantages, such as precisely controlled near room temperature operation, low energy requirements, capability to coat complex component geometries, low cost, and simple scale-up [1]. The ability of the technique to coat various types of materials, such as oxide, nitride, carbide and metallic, in a metal matrix allows it to provide coatings for multiple applications [2]). The advantages of this technique has encouraged researchers to further explore its potentials.

In recent years, there have been increasing interests focused on electrodeposited nickel-cobalt (Ni-Co) and Ni-Co based metal matrix composite (MMC) coatings due to their superior properties, such as higher hardness [3]–[6], improved anti-wear, better corrosion resistance and oxidation resistance [7], as compared with pure Ni and Ni based composite coatings. The properties of Ni-Co or Ni-Co based composite coatings are mainly dependent on the incorporated particles and microstructures of the Ni-Co matrix [7]–[11]. Based on these studies, it was found that the highest nanoparticle concentration in the coating was reported to be less than 30%vol. The average %vol was reported to be just around 10%vol regardless of particle type and matrix. The low concentration of ceramic particles in the coatings have limited their mechanical performances as compared to ceramic coatings.

Among the various inert particles, alumina (Al$_2$O$_3$) particles have many superior properties, such as low price, good chemical stability, and high micro hardness and wear resistance at high temperatures. Hence, Al$_2$O$_3$ coatings are widely used for shipboards and submarines, textile industry and machinery to give protection and support to their structures [12]. The presence of Al$_2$O$_3$ particles in Ni-Co based metal matrix could increase the micro hardness of the coating. These hard ceramic particles provide greater hardness through dispersion strengthening of the coating [13], [14]. However, a lot of factors should be considered to obtain superior coating properties, such as particle characteristics, electrolyte composition including additives, temperature, pH, surfactant type and concentration, current density, hydrodynamics (laminar, mixed and turbulent regimes), and electrode geometry [10].

This paper reviews the electrodeposition techniques that are used to produce Ni-Co/Al$_2$O$_3$ coatings, and the electrodeposition variables that influence the coating performance. It then discusses further research that needs to be conducted in order to improve the corrosion and erosion resistance of Ni-Co/Al$_2$O$_3$ coating in harsh environments, such as marine conditions.

ELECTRODEPOSITION TECHNIQUES

The most commonly used electrodeposition technique is the direct current (DC) technique. This technique uses an electrochemical cell as show in Figure-1, which consists of anode and cathode electrodes, electrolyte, and DC power supply. The currents flow from the anode to the cathode through the electrolyte. During the process, free metal ions in the electrolyte will transfer to the cathode to gain the electrons and form a solid metal [15]. The simple setup of this technique has encouraged researchers to conduct various researches related to this technique as shown in Figure-1.
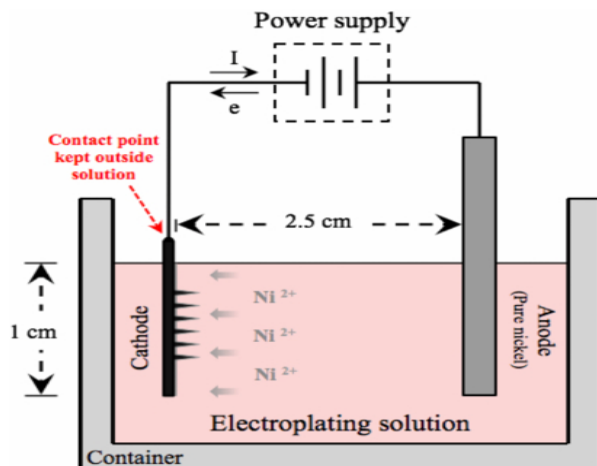
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Figure-1. Schematic diagram of the electrochemical setup for the electrodeposition process [15].

Some studies have also been conducted on the electrode arrangement in the bath. The conventional electrodeposition technique positions the electrodes vertically. However, a few researchers have suggested positioning the electrodes horizontally, with the cathode placed below to the anode. This type of arrangement is known as the sediment electrodepositing technique [16]. This technique would allow the particles to settle down easily as sediment on the cathode surface with the support of gravitational and electrophoretic forces. Figure-2 shows a schematic image of the applied force during the deposition process. The gravitational and electrophoretic forces act vertically in the same direction, while the in conventional technique, the electrophoretic force is disturbed by the gravitational force in the perpendicular direction. Working with Ni-SiC coating, they found that the percentage of SiC content in the coating increased as compared to the conventional technique. Feng *et al*. [17] reported that $Ni-Al_2O_3$ coatings produced using this technique provide better micro hardness and lower wear rate than $Ni-Al_2O_3$ coatings produced using the conventional technique. Bakhit *et al*.[18], who investigated the effect of the electrodeposition technique on micro hardness and corrosion properties, also found that sediment electrodeposition increases the SiC content in the Ni-Co matrix and improves the micro hardness of nanocomposite coatings [19].

Other techniques frequently used for the electrodeposition process include pulse current and reverse pulse current. In the pulse current technique, the current supply or potential alternates between two different values, while the pulse reverse current technique uses an alternating current between the anodic and cathodic current densities. These techniques are used to control the electron transfer through the electrolyte by controlling the current distribution. Regulating the current would remove the negatively charged layer that is constructed during the plating process [20]. Chang *et al*. [21] found that Ni-Co/Al_2O_3 coatings produced using this technique are

uniform, compact and possess finer grain. Similar findings were reported by Mohan *et al*. [22] for $Ni/nanoAl_2O_3$ coatings and Nemes *et al*. [23] for Zn/CeO_2 coatings. In addition, Bahrololoom & Sani [24] reported that pulse current provides better mechanical properties to Ni-Al coatings. They also investigated the relationship between frequency and duty cycle of pulse currents with the micro hardness and wear behaviour of the coatings. They found the hardness of the coating increased when low duty cycle and frequency were used. These results are in good agreement with the findings of studies conducted using $Zn-TiO_2$ coatings [25], [26].
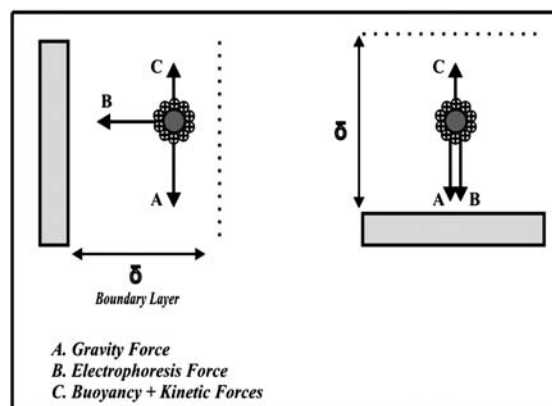

Figure-2. Schematic diagram of the applied forces for an immersed particle in the (L) conventional and (R) sediment electrodepositing techniques [19].

Zheng & An [27] fabricated $Zn-Ni/Al_2O_3$ coatings using an ultrasound horn in the electrolyte to produce ultrasonic streaming and cavitation bubbles. They found that increasing the ultrasonic power from 0 to 0.7 W/cm^2 would increase the Al_2O_3 content in the coating from 4.5 to 8.9 wt.% and could reduce the agglomeration of the Al_2O_3 particles. The presence of ultrasonic streaming is believed to catalyse the movement of the Al_2O_3 nanoparticles towards the cathode, while the cavitation bubbles deagglomerate the nanoparticle. However, as the ultrasonic power is further increased, the coating performance reduces, with high porosity and reduced Al_2O_3 content. Bakhit & Akbari [2] also used this method in their research involving deposition of Ni-Co/SiC composite coatings. Other researchers also agree with the advantages provided by the ultrasonic method [8], [28]–[30].

Another technique that been discussed among the researchers is the addition of CO_2 in the electrolyte. CO_2 is emulsified in the electrolyte with pressure of more than more than 10 MPa to increase the solubility of H_2 produced during the electrodeposition process. The presence of CO_2 in the water forms carbonic acid, which increases the H^+ content in the solution [31]. As a result, the coatings produced have better hardness and low porosity, as reported by Chung *et al*.[32] and Chiu *et al*.[33].

ELECTRODEPOSITION VARIABLES

In addition to the electrodeposition techniques, the mechanical properties of the produced coatings are influenced by several deposition variables, which include applied current density, deposition duration, particle concentrations and particle type [14].

Current density

Wu *et al*. [34] studied the effect of current density on Ni-Co/Al_2O_3 coatings. The highest volume percentage of Al_2O_3 was obtained at current density of 3 A/dm^2. The study on Ni-Al_2O_3 coatings conducted by Goral *et al*. [35] found that micro hardness increases when current density is increased from 2 to 6 A/dm^2. Simultaneously, this parameter also provides more uniform particle distribution. Current density also gives the same effect to Ni-SiC coatings, as reported by Bakhit *et al*. [18]. The optimum current density used to obtain the highest particle content in the coating was 3 A/dm^2. In contrast, Sun *et al*. [36] reported that the lowest current density used (6.5 A/dm^2) provided a slightly higher Al content as compared to current density of 34 A/dm^2, but the difference was less than 1%.

Deposition period

Farrokhzad *et al*. [14] studied the effect of deposition time on the coating characteristics of Ni/Al_2O_3-TiO_2. They found that the coating prepared at 120 min of deposition time was thicker than the coating prepared at 90 min of deposition time. However, duration time had no significant effect on the micro hardness of the coatings. While not many papers discuss the effect of deposition time, some researchers prefer to vary the duration in order to get the suitable thickness of coatings for their studies [2], [18], [28].

Particle concentration

Another approach used to improve coating properties is by varying the ceramic particle concentration in the electrolyte. Farrokhzad *et al*. [14] reported that molarities of Al_2O_3 and TiO_2 particles is proportional to the particle volume in the coating. However, Gul *et al*. [37] found that the increase of volume of Al_2O_3 in the coating is not comparable to the increase of concentration of Al_2O_3 particles in the electrolyte; a six times increment of Al_2O_3 in the electrolyte only increased the Al_2O_3 concentration in the coating by less than three times. Nguyen *et al*. [38] also reported similar results; by increasing the fly ash concentration in the electrolyte from 20 to 50 g/l, it could produce coatings with 2.5% higher particle concentration. The concentration of matrix particles in the electrolyte also influences the ceramic particles in the coatings. Kuo *et al*. [8] increased the concentration of Ni^{2+} (by changing the concentration of $Ni(NH_2SO_3)_2$) in the electrolyte. As result, they managed to increase the Al_2O_3 particles in the coating from 8.4 to 26.8 vol%. Conversely, low Ni^{2+} concentrations would encourage H^+ reduction, which produces coatings with high porosity.

Additive

The presence of a surfactant could develop positive charges on the particle surface to prevent agglomeration and improve particles stability in the electrolyte. Chen *et al*. [39] used hexadecylpyridinium bromide (HPB) as a surfactant in Ni-Al_2O_3 coatings. The increase of HBP to 300 mg/l enhanced Al_2O_3 content by up to 14.7 vol%. Similar results were also reported by Gul *et al*. [37] and Narasimman *et al*. [16] by using SiC particles. Surfactants are also used in the sediment electrodeposition technique to decrease electrolyte viscosity and thus, gravitational force and diffusion coefficient improve the deposition process [29]. Chiu *et al*. [33] used an octa (ethylene oxide) dodecyl ether ($C_{12}EO_8$) surfactant to emulsify the electrolyte in presence of CO_2. The emulsified electrolyte increases the solubility of H_2. Hence, the formation of H_2 gases bubble could be minimised to produce a smoother surface [31]. Commonly used surfactants are hexadecylpyridinium bromide (HBP) [29], [37], [39], tetramethyl ammonium hydroxide (TMAH) [16], polyglycol [40], $H(CH_2CH_2O)_8O$–$(CH2)12H$ [32], cetyltrimethyl ammonium bromide (CTAB) [8] and sodium dodecyl sulfate (SDS) [2], [41].

pH

Electrodeposition composite coatings usually deposited from acidic bath. The pH range from 3 to 5 commonly used due to the optimum parameters of Ni-Co coating [7], [42] which shows a high current efficiency, bright and smooth deposit with fine grain size [43], [44]. In contrast, the alkaline bath had reported gave higher alumina concentration in the coating [45]. Bund & Thieming [45] also describe that surface charge of alumina particle was dependence to pH of the bath. In the acidic bath, surface charge of the alumina particle is positive while in the alkaline bath it would be negative. The magnitude of the surface charge of particle in contact with the electrolyte affect the stability of the particle dispersions and ability to adsorb ion from the solution [46].

DISCUSSION

Composite coating performance has improved over time in terms of mechanical properties and corrosion resistance. However, some improvements are needed with the intention of making this coating reliable for harsh environments. Applications in marine environments expose materials to corrosive conditions and erosive medium. Thus, the use of materials that combine erosion and corrosion resistance with high mechanical strength is a requirement in industrials applications [47].

While the improvement of Ni-Co/Al_2O_3 coating properties has shown good achievement to proof its ability as a protective coating, there is a lack of studies on the effectiveness of this coating in erosive environments. Most

researches conducted separate between corrosion and erosion effects. Rayes *et al*. [47] describe the erosion-corrosion as an acceleration of the deterioration rate of metal due to the combined effect of erosion and electrochemical attacks. The combined effect could enhance the damages not only to the coating, but also to the substrate through the combined interaction between the two processes. Erosion could strip the coating and catalyse the corrosion attack. On the other hand, corrosion occurs first to catalyse the erosion process. This mechanism leads the greater damage to the coating and substrate [47]–[49].

The presence of ceramic particles in the metal matrix also gives different results among the researchers in terms of their corrosion behaviour. Some of the researchers claim that ceramic particles could enhance the corrosion resistance of the coatings [18], while others report different findings [33], [36]. They verified that the corrosion behaviour of the coatings is not only influenced by the embedded particles, but also other factors. The coating microstructure plays a big role in providing corrosion protection [50]. The presence of permeable defects, such as pores, cracks and grain boundary gaps, could allow the electrolyte to penetrate and interact with the metal surface, which catalyses the corrosion attack [51]. The interaction between the electrolyte and substrate exhibits galvanic corrosion either to the coating or to the substract itself depending on nobility of the materials. That is the reason why several researchers have tried to reduce the porosity in their coatings [52]–[54]. However, previous studies on Ni-Co/Al$_2$O$_3$ coatings did not discuss this effect very well.

The penetration of the electrolyte through the permeable defects is sluggish due to the small space or capillary. As a result, electrochemistry impedance spectroscopy (EIS) tests for corrosion could give different values over the time and require a longer testing period. Nevertheless, most of the corrosion studies were conducted in a day or two, which believed not be sufficient to model the real process.

Another issue rising from the literature is the low concentration of the ceramic particles in the coatings. Kuo *et al*.[8] found that lower electrolyte concentrations tend to decrease the agglomerated Al$_2$O$_3$ particle size. The Al$_2$O$_3$ particles would disperse effectively in the electrolyte solution. As a result, it would create more opportunity for loose Al$_2$O$_3$ particles to attach onto the electrodes or substrate [8]. The increase of Al$_2$O$_3$ particles could provide better mechanical performance, especially micro hardness and wear resistance [55]. Kuo *et al*. [8] also changed the Ni^{2+} concentration in order to control the electrolyte concentration, as compared to researchers who used Al$_2$O$_3$ concentration as the focus of their study [14], [28], [30]. However, the effect of Ni^{2+} concentration with the presence other metallic elements, such as Co^{2+}, and inert particle, such as Al$_2$O$_3$, has not been studied. Other than that, Al$_2$O$_3$ nanoparticle could adsorb Ni^{2+} well as reported by Srivastava et.al. [56]. They were using Al$_2$O$_3$ as an adsorbent in the aqueous solution to remove the heavy metal. The adsorption of Ni^{2+} to the Al$_2$O$_3$ seems can be use as a carrier in order to deposit the nanoparticle in the coating. However, further studies need to identify the right pH value and the effect of the other anion or cation to the Al$_2$O$_3$ surface charge.

CONCLUSIONS

This paper has reviewed several electrodeposition techniques that have been used to produce Ni-Co/Al$_2$O$_3$ composite coatings. Direct current is the most commonly used technique due to its simple setup. Other techniques that can be used are pulse current and pulse reverse current, sediment electrodeposition, and CO$_2$ bath. All the techniques discussed have their own advantages to improve the coating properties. In addition, process variables, including applied current density, deposition time, particle concentration and use of surfactants, also give great effect to the coating performance.

Although the electrodeposition of composite coatings has been established, some improvements are still needed with the intention of making this coating reliable for severe environments. For this purpose, a corrosion-erosion study for Ni-Co/Al$_2$O$_3$ coatings should be conducted. Furthermore, increase of Al$_2$O$_3$ content is a key to produce coatings with excellence performance.

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