

## Abstract

Aluminum coatings are widely used for corrosion resistance, high-temperature oxidation resistance, and a bright aesthetic appearance. The aluminum electrodeposition involving an ionic liquid-based electrolyte is preferred because of its extensive electrochemical window, minimal vapor pressure, non-flammability and a greener alternative to the existing organic solvent system. Aluminum deposition employing an ionic liquid-based electrolyte on mild steel has been used as a protective coating for enhancing corrosion resistance. Nevertheless, the adherence of aluminum to the mild steel has been a concern since the iron oxide layer forms on its surface before the deposition. Conventional in-situ electrochemical etching prior to electrodeposition improves adhesion, but it leads to electrolyte contamination from substrate dissolution. This leads to electrolyte degradation and the formation of an impure coating. To overcome these shortcomings, plasma treatment is proposed, and its feasibility as a surface modification technique is investigated. The effects of surface treatment, both plasma and electrochemical, on deposit morphology, roughness, crystal orientation, micro-hardness, and corrosion resistance are discussed. Plasma modification has led to the development of a finer, denser, and smoother aluminum coating. The aluminum deposition on plasma-modified steel exhibited higher micro-hardness with an increase of 32.4 % and lower corrosion current with a reduction of 65.3 % in comparison to that on the electrochemically etched sample. These improved performances establish plasma modification as the superior alternative to the electrochemical etching for aluminum electrodeposition on steel.

Plasma modification of the steel substrate enabled the deposition of a well-adherent aluminum coating. Since aluminum electrodeposition from the chloroaluminate solution is complex, careful optimization of process parameters is necessary to ensure the development of high-quality aluminum deposition in the form of dense, uniform, and compact polyhedral growth with consistent excellent surface coverage. Among the process variables, applied potential,

stirring speed and temperature of the electrolyte were chosen as the critical factors due to their dominating role in governing the nucleation, growth, kinetics of ions, and transport of ions, particularly in the context of aluminum electrodeposition from decane-shielded chloroaluminate electrolyte. After conducting a series of experiments, the optimal values of these parameters were found. The optimal value of applied potential, stirring speed, and electrolyte temperature are -400 mV, 250 rpm, and 25 °C, respectively. With these process parameters, a high-quality aluminum coating was developed.

Two major shortcomings of the pure aluminum coating are its reactivity towards the chlorinated aqueous solution and its low hardness, which are addressed by incorporating functionalized graphene nanoplatelets into the matrix of aluminum coating. The performance of the developed coating was assessed in terms of coating morphology, topography, thickness, crystal orientation, micro-hardness, and corrosion, and compared with that of pure aluminum coating. The composite coating exhibited an increase of 38.18 % in micro-hardness and an increase of 141.7 % in polarization resistance as compared to the pure aluminum coating. Reinforcement of functionalized graphene nanoplatelets successfully shifted the corrosion potential to a more positive potential from -824 mV to -767 mV. These improvements highlight that the aluminum-based composite coating can be a strong alternative to pure aluminum coating on steel.

The development and characterization of pure and composite coatings were done successfully on the flat mild steel sample. For broadening the horizon of its real-world applications, the optimized parameters were effectively transferred to the wire geometry, aimed at potential deployment in aerospace fasteners, industrial fittings, and welding wire. Continuous and adequately adhered aluminum-based coatings were successfully deposited on the mild steel welding wire. This successful transfer suggests the suitability of this process for real-world industrial environments necessitating continuous coating along an extended wire.

**Keywords:** Electrodeposition, Composite coating, Graphene nanoplatelets, Corrosion