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Effect of Inorganic Content on the Performance of Anticorrosive Hybrid Sol-Gel Coated EN AW-6063 Alloy

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Abstract. The organic-inorganic hybrid sol-gel films have been reported as an effective anti-corrosion and environmentally friendly alternative to Cr(VI) pre-treatment for aluminium alloys. The sol-gel process used to obtain these coatings allows the variation of the different synthesis parameters to achieve coatings with optimized properties. In this work, hybrid films with different Zr/Si ratios were synthesized from glycidoxypropyltrimethoxysilane (GPTMS) and zirconium n-propoxide (TPOZ) precursors. Electrochemical Impedance Spectroscopy (EIS) was used to evaluate the corrosion behaviour of coated aluminium specimens in 0.5 M NaCl solution. The morphology and chemical structure of the hybrid coatings prepared were studied by Scanning Electron Microscopy (SEM) and Energy Dispersive Spectroscopy (EDS), Fourier Transformed Infrared Spectroscopy (FTIR) and Thermo Gravimetric Analysis (TGA). It was found that increasing Zr/Si ratio leads to a more cross linked inorganic network, resulting in higher initial coatings resistance, but may turn coatings more hydrophilic, prone to rapid degradation in water, due to a less connected organic network. Consequently, the best anticorrosive performance derives from the balance between the two opposite trends and it was achieved with Zr/Si molar ratio of 0.25.

Introduction

The organic-inorganic hybrid sol-gel films have been reported as an effective anti-corrosion and environmentally friendly alternative to Cr(VI) pre-treatment for aluminium alloys [1,2]. The hybrid materials based in organofunctional silane alkoxides that allow polymerization of the organic groups, originate hybrid nanostructured networks interconnecting organic and inorganic materials, with a resultant synergic effect on the mechanical and chemical properties achieved. The organic components impart flexibility, density and functional compatibility with organic coatings, while inorganic components impart hardness, resistance to abrasion and improve adhesion to the metal substrate [1-4]. Reinforcement of this hybrid polymer network can be achieved through incorporation of oxide nanoparticles, what contributes to the reduction of coating crack ability and porosity, improving its anticorrosive properties [1-3,5-10]. Moreover, the incorporated inorganic nanoparticles can be used as nanoreservoirs for storage and controllable release of inhibitor species [1,3,8–10]. Nanoparticles introduction can be done by addition of nanopowders into sol-gel system [1,3,5], but also can be achieved by *in situ* synthesis during the sol-gel process, through controlled hydrolysis/condensation of metal alkoxides [1,6-8].

In previous works, Zheludkevich et al. [6-8] developed epoxysilane-based hybrid coatings with incorporated zirconia nanoparticles by the *in situ* method, showing good corrosion protective properties for aluminium alloys. In those works, it was found that metallic alkoxide concentration and its hydrolysis conditions (time, temperature and complexing agent), influenced the coatings corrosion behaviour. There are few more studies concerning the influence of Zr content in hybrid sol-gel coatings corrosion properties [12], being more focused on its influence on structure and