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Steam initiated hydrotalcite conversion coatings: Application to environmental Al alloy surface treatment



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ABSTRACT

A facile process of exploiting high-temperature steam to deposit environmentally friendly hydrotalcite (HT) coatings on Al alloy 6060 was developed in a spray system. Scanning electron microscopy showed the formation of a continuous and conformal coating comprised of a compact mass of crystallites. A range of coating processes based on the formation of HT surface layers has been developed to examine its effect on the coating's thickness and corrosion resistance properties. These varieties include pre-coating cleaning (grid blasting vs. chemical etching), metal species in HT compounds (Al-Zn HT coating vs. Al-Li HT coating), oxidizer additives (K2S2O8, Na₂SO₄, NH₄NO₃, KNO₃), and post-coating treatment (Mg(CH₃COO)₂, Mg(CH₃COO)₂ + Ce(NO₃)₃ + H₂O₂, Mg (CH₃COO)₂ + La(NO₃)₃). Results showed that grid blasting can increase the coating surface area, while chemical etching improves the chemical bonding connection between the coating layer and substrate, and therefore leads to a higher corrosion resistance in the filiform corrosion test. Al-Li HT coatings on the Al alloy are much thicker than the Al-Zn HT coatings that formed from the same procedures. This variation is likely caused by the pH condition of coating solution, which is essential to the co-precipitation of two or more cations in HT structure. The formation of Al-Zn HT coating was enhanced by oxidizer and ammonium salt additions. The thickness of Al-Zn HT coating decreases patterning after the oxidizing and complexing capability of additives in the following order: K₂S₂O₈ > Na₂SO₄ > NH₄NO₃ > KNO₃. Post-coating rinsing by Mg(CH₃COO)₂, in addition to Ce- and La- based salts, is capable of sealing the porous Al-Li HT coatings and increasing the corrosion resistance ability. The Ce modified Al-Li HT coatings show better corrosion protection than the La modified coatings, due to a behaviour of "active corrosion protection" that enables precipitation of insoluble Ce(IV) oxides to reinforce the existed HT coatings.

1. Introduction

Aluminium and its alloys are widely used in the structural and transportation industries due to their low price and superior mechanical properties. The surface of aluminium under normal conditions has a thin oxide film (2.5–10 nm) which provides corrosion resistance. This oxide can be converted or transformed to functional conversion coatings in order to maintain a durable performance. Chromate salts-based anticorrosion pre-treatments and primer pigments have been extensively used to form a conversion coating and to effectively prevent localized corrosion on aluminium alloys [1–3]. However, the toxicity and carcinogenicity of hexavalent chromium is strictly restricted in industry, which requires an urgent development of environmentally

friendly chemical treatment methods. Strictly chemical or electrochemical baths have been used in the present-day for creating the environmental conversion coatings, but with limited industrial application on large components. In contrast, conversion coatings that are initiated by exploiting the oxidative power of steam are not only lowcost but also effortless to operate, such as for onsite repairs as a mobile

The modification process on metal surfaces by steam is comparable to mineral surface coatings in nature, which are formed through the influx of constituents from the dissolving underlying mineral, and from fluids originating in the surrounding soils and sediments [4]. Throughout natural history, patchy to continuous mineral surface coatings at the nano- to micro-meter scale have been formed through

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abiotical chemical reactions and catalysis by micro-organisms [4]. However, the dissolution and precipitation processes resulting from water–rock interaction are intrinsically time-dependent. By utilizing the oxidative power of steam that acts as a carrier and source of energy, the formation of mineral surface coatings occurring in nature can be mimicked, and most importantly, accelerated on the surface of aluminium alloy in association with environmentally friendly additives [5].

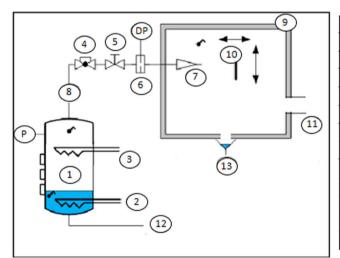
The synthesis of Al-containing minerals, i.e. hydrated aluminium oxides [6-8], aluminium silicate [9-11], and aluminium phosphate [12-14] has been extensively reported. Among these minerals, hydrotalcite (HT), a layered double hydroxide of general formula Mg₆Al₂CO₃(OH)₁₆·4(H₂O) has shown various applications as catalysts, polymer stabilizers, and adsorbents due to its excellent properties, such as high-dimensional stability, reduced gas permeability, optical flame retardancy, and enhanced mechanical performance [15]. HT and HTlike coatings have been successfully employed as anticorrosion pretreatments on Mg-Li [16,17], steel, [18] and aluminium [19] alloys due to their efficiency of scavenging halogen in polymer systems and capturing halide in aqueous solutions. On the other hand, the highly porous and flaky structure enhances its ability to adsorb ions as anticorrosion inhibitors, preventing corrosion from the damage of top paint. This particular structure of HT minerals also functions as an adhesion promoter for the subsequent paint system, thus ensuring a good adhesion between the conversion coating and the paint layer. Successful attempts in synthesizing HT pigment and coating layers by heating ingredient powers and dispersing the products in aqueous solutions have been

described. In this paper, we demonstrate the formation of environmentally friendly HT coatings in a steam-based spray system on Al alloy 6060. A range of variations and their effects, including different pre-coating treatments (grid blasting and chemical etching), different types of HT coatings (Al-Li HT and Al-Zn HT), different oxidizer additives ($K_2S_2O_8$, Na_2SO_4 , NH_4NO_3 , KNO_3), and different sealing solutions ($Mg(CH_3COO)_2$, $Mg(CH_3COO)_2 + Ce(NO_3)_3 + H_2O_2$, $Mg(CH_3COO)_2 + La(NO_3)_3$), were investigated to characterize the resulting coating structure, morphology, and corrosion resistance.

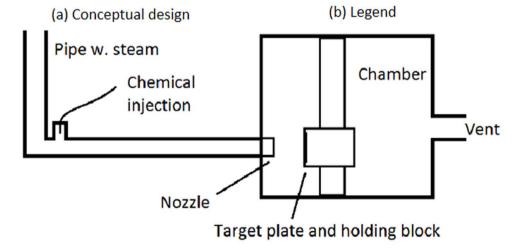
2. Materials and methods

2.1. Experimental procedures

The experiment setup is sketched and illustrated in Fig. 1. The experimental system includes a steam generator consisting of a water tank. The steam generator is connected to a chamber by stainless steel pipes, the end of which acts as an orifice for the steam. This equipment is able to deliver steam with a pressure from 1 to 15 bar, corresponding to temperatures of 100–198 °C. The chamber where the sample is located operates under atmospheric pressure, and is equipped with a sample holder that can be moved sideways relative to the steam. Chemicals were injected into the steam using a pump in the tube above the pipe opening. An atomizer is added to homogenize the distribution of coating on the alloy surface.



Position	Components
1	Pressure vessel
2	Heater
3	Heater
4-5	Shut-off valve
6	Sensor
7	Pipe opening (Nozzle)
8	Super heater
9	Isolated chamber
10	Sample holder
11	Vent
12	Filling valve
13	Drain



(c) Sketch of the last part of the pipe, injection point, chamber and target plate

Fig. 1. The experimental setup for the steam initiating surface modification system.

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