



Transformation of iron containing constituent intermetallic particles during hydrothermal treatment

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ABSTRACT

Aluminium alloys AA3102 and AA9108 were treated with high temperature steam, which resulted in the formation of an oxide layer of average thickness of 300–400 nm. Hydrothermal steam treatment resulted in the removal or oxidation of Al (Fe) Mn and Al (Fe-Si) Mn type intermetallic particles present in the alloys. Furthermore, electron energy loss spectroscopy analysis revealed that during the steam treatment, the Fe enriched areas of the Al (Fe-Si) Mn type intermetallic particles were transformed into Fe₂O₃ and Fe₃O₄ phases, while energy-dispersive X-ray spectroscopy line profile measurements by scanning transmission electron microscope showed that Mn and Si were leached out and incorporated into the surrounding oxide layer. Further, the part of intermetallic phase was transformed into polycrystalline material.

1. Introduction

Aluminium and its alloys are widely used in the aerospace, architectural, and automotive industries due to its light weight, good mechanical properties, corrosion resistance, and recyclability [1–3]. During the manufacturing process, elemental additives are added to increase the mechanical properties of the aluminium alloys [4–7]. However, the amounts of iron, silicon, and manganese play an important role in establishing the mechanical properties of the aluminium alloys [8]. Iron as an impurity, is always present in the alloys made from commercially pure base material. The solid solubility of iron in aluminium is very low with the result that most iron present leads to the formation of the intermetallic compounds. The chemical composition of such intermetallic phases strongly depends on other impurities or alloying elements present in the alloy system [9]. Further, Manganese is the most common alloying addition, which is used to neutralize the effect of iron by preventing the formation of FeAl₃ or FeSi rich phases [10]. In most manganese based aluminium alloys, iron can replace large amounts of manganese in the intermetallics without changing the crystalline structure i.e. (FeMn)Al₆ and (FeMn)₃Si₂Al₁₅ [8,9]. Furthermore, in the Si based the alloy systems, mainly FeSiAl₅ intermetallic compound is usually present, while number of others have reported the formation of Al-Fe-Si type intermetallics [11–13]. However, the replacement of FeSiAl₅ phase by manganese based intermetallic compounds is of great interest due to the corrosion resistance properties. It

is reported that manganese addition to the intermetallics reduces the potential difference between the iron-bearing intermetallic particles and the matrix [10,14].

In general, the presence of these intermetallic particles in aluminium matrix results in the heterogeneous microstructure, which has a high impact on the corrosion resistance properties of aluminium alloys [15–19]. Thus, aluminium alloys essentially require pre-treatment (well known as conversion coatings) to maintain its corrosion performance with or without polymer coatings such as paints [20–26]. However, the formation of various conversion coatings on commercial aluminium alloys such as AA3000, AA2000 and AA7000 series is complicated due to the presence of variety of intermetallic particles [27–32]. For instance, the formation of chromate conversion coating on Cu enriched intermetallic particles resulted in the formation of thinner oxide layer over the copper rich particles when compared to the aluminium matrix [28]. Further, no Cr (III)-Cr (VI) mixed oxides over AlCuMg, Al₂CuMg, and Al₅CuMg₄, which is usually present over aluminium matrix, were observed, while thicker films were observed on Al₃Mg intermetallic particles [33,34]. Cerium based conversion coatings is one of the most promising chromate free pre-treatment which have been studied extensively [35]. A number of investigations [36,37] revealed that the preferential precipitation of the Ce oxide layer occurs at the top of Cu-containing intermetallic particles, which was due to the cathodic nature of these intermetallic particles. However, the Cu free cathodic intermetallic particles i.e. (Fe, Cr)₃SiAl₁₂, FeNiAl₉, Al₆-(Mn,Fe,Cr) did not

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exhibit the rapid deposition of cerium oxide layer [27,36–39]. Similarly, permanganate conversion coatings appear to have some promising properties as on aluminium alloys [40–42]. During the formation of permanganate conversion coating on AA2024, the deposition of MnO₂ oxide layer was observed over cathodic intermetallic particles, which was considerably thicker than on the aluminium matrix [43]. Thus, it is evident that the composition and chemical nature of intermetallic particles significantly influence the formation of various conversion coating, which in general influences the corrosion performance of aluminium alloys [27,31,34,44–46].

A number of studies [47–53] have been reported on the hydrothermal treatment of aluminium alloys, which results in the formation of aluminium hydroxide layer. However, these studies do not deal with the fundamental role of intermetallic particles in the growth and corrosion resistance properties of hydrothermally grown films. Our earlier work [54–57] of hydrothermal treatment of aluminium alloys with steam showed that the growth of aluminium hydroxide was thicker on Al-Fe-Si intermetallic particles when compared to the aluminium matrix and resulted in the partial oxidation of the intermetallic phases. Further, in the presence of acidic chemistries, the intermetallic particles were the preferential sites for initiation of oxide growth [56]. However, these studies do not deal with the change in the microstructure of specific types of intermetallic particles before and after the steam treatment. The later investigations showed that the presence of oxide at the intermetallic may significantly affect the adhesion performance of the commercially applied powder coating [58]. Thus, the purpose of present study is to investigate chemical and microstructure changes to the intermetallic particles under hydrothermal conditions. Therefore, the growth of the aluminium hydroxide films on AA3102 and AA9108 was investigated; where the effect of steam treatment on Al(Fe)Mn and Al(Fe-Si)Mn type of intermetallic particles were studied in detail. The detailed microstructural characterization of the produced coatings has been carried out using glow discharge optical emission spectroscopy (GD-OES) and, scanning electron microscopy (SEM) and transmission electron microscopy (TEM) including energy dispersive X-ray spectroscopy (EDS) and electron energy loss spectroscopy (EELS).

2. Experimental methods

2.1. Material

Commercial aluminium alloys AA3102 and AA9108 were used as the substrate for the investigations. Chemical composition of the alloys in as-received condition is shown in Table 1. Prior to the hydrothermal surface treatment, the samples were cut into 20 mm × 15 mm test coupons and ultrasonically cleaned for 5 min, by using analytical grade acetone as a solvent. After being cleaned in acetone, they were rinsed in deionised water and dried using an air gun.

2.2. Sample preparation

In order to identify various intermetallic particles and to locate the analysis area before and after the steam treatment, a set of microhardness indents were made using a Vickers micro hardness indenter (FutureTech FM 700 microhardness tester) with a load of 50 g. Using these indentation marks, the same intermetallics particles could be analysed before and after the hydrothermal steam treatment. Prior to the steam treatment, the samples were degreased for 1 min in 5 wt%

Table 1
Chemical composition of alloys in wt% (remaining percentage Al), measured by GD-OES.

Alloy	Si	Fe	Cu	Mn	Mg	Cr	Zn	Ti
AA3102	0.40	0.70	0.10	0.40	–	0.10	0.30	0.10
AA9108	0.20	0.30	0.20	1.00	0.05	0.10	0.20	0.15

commercial Alficlean (weakly alkaline detergent pH = 7.5–8) aqueous solution at 30 °C followed by rinsing in distilled water and dried in ambient air.

2.3. Steam treatment

The samples were exposed to high temperature steam, generated from deionised water in an autoclave (All American Pressure Canners, USA) at 107 °C and 130 kPa vapour pressure, for a period of 30, 60, 120, 240 and 600 s, respectively. After the steam treatment, the samples were rinsed with deionised water and dried in ambient air.

2.4. Microstructural characterization

2.4.1. Glow discharge optical emission spectroscopy (GD-OES)

After the steam treatment, the thickness and relative chemical composition of the surface layers were analysed using GD-OES (GD-2 profiler, Horiba Jobin YVON). The instrument is equipped with a radio frequency generator, a standard discharge source with an anode of 4 mm internal diameter, a monochromator and polychromator optical spectrometers, and Quantum XP software. Calibration of the GD-OES profiler was carried out at optimised discharge condition of 850 Pa pressure and RF power of 40 W by sputtering the sample surface for a specific time and then measuring the depth of the resulting sputtered crater using a surface profilometer.

2.4.2. Scanning electron microscopy (SEM)

The morphology and composition of intermetallic particles present at the surfaces of aluminium alloys before and after the steam treatment were investigated using field emission gun (FEG) SEM (Quanta 200 FEG MKII, FEI) with an Oxford Instrument INCA EDS analyser. The EDS analysis has been performed with an acceleration voltage of 15–20 keV and Cu calibration.

2.4.3. Transmission electron microscopy (TEM), electron energy loss (EELS) and energy dispersive X-ray (EDS) spectroscopies

Thin film lamella from the steam treated surfaces were prepared using in-situ lift-out technique using a dual beam focused ion beam SEM (FIB-SEM) (Quanta 200 3D DualBeam, FEI) and were further thinned down to approximately 120 nm thickness for electron transparency in a FIB-SEM (Helios Nanolab DualBeam, FEI). A FEI Tecnai G2 T20 TEM was used at 200 keV to investigate the prepared lamellae. The EELS spectra were acquired with a dispersion of 0.05 eV/channel and an energy resolution of 1 eV in TEM diffraction mode. The EELS spectra were processed using Digital Micrograph software from Gatan. EDS was performed at 200 kV using the Tecnai TEM equipped with an Oxford Instruments X-MaxN 80 T silicon drift detector. The EDS spectra were processed using TIA software from FEI Company.

3. Results and discussion

3.1. Microstructural characterization

3.1.1. Glow discharge optical emission spectroscopy (GD-OES) analysis

Fig. 1 shows a typical GD-OES depth profile and an average oxide thickness on AA3102 and AA9108 after steam treatment for 30, 60, 120, 240, and 600 s. The X-axis in Fig. 1(a) shows the sputter time converted into sputter depth using regulated sputter rate vs. depth data for aluminium oxide and aluminium substrate. This data was obtained by calibrating sputter depth of the craters using GD-OES for a specific time. The average sputter rates for the steam generated oxide and aluminium substrate at a given pressure (850 Pa) and RF power (40 W) were 30 nm/s and 65 nm/s respectively. The concentration of elements shown by the GD-OES profile were relatively compared, a decrease in the intensity of oxygen (O) counts is related to an increase in the intensity of aluminium, indicating the interface between the steam

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