MICROSTRUCTURE DEVELOPMENT AND PHASE EVOLUTION OF NICKEL COATED ALUMINIUM AND HYDRONALIUM SUBSTRATES

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Abstract

In this work, the microstructural changes of nickel coated aluminium and hydronalium substrates were investigated after the heat treatment above the melting point of pure aluminium. The 99.0Ni powder was used to produce the coating using high velocity oxyfuel spraying on 5 mm thick 99.5Al and AlMg3 sheets surface. As-sprayed samples were annealed in the temperature range of 660 - 800°C in ambient atmosphere. The dwell time for isothermal exposure was set from several minutes up to several hours depending on used temperature. Resulting microstructures were investigated by means of metallography and light and scanning electron microscopy techniques. Employed heat treatment allowed to form two different (binary and ternary) hypereutectic alloys. The main differences resulted from different initial substrate microstructures and chemical composition. Compare to pure aluminium substrate microstructure, hydronalium alloy contained a number of nucleation sites in the form of eutectic and/or intermetallic phases and also, probably due to the higher diffusion mobility of aluminium, exhibited the faster dissolution of as-sprayed nickel coating region.

Keywords: Aluminium, Hydronalium, Thermal Spraying, Heat Treatment, Electron Microscopy - Scanning

1. INTRODUCTION

Aluminium and its alloys provide attractive properties like very low density (≤ 2.7 g/cm\(^3\)), excellent oxidation resistance in ambient and low acid environments and very good workability, which predetermine their extensive use across the aerospace, aircraft, automotive, food-processing, building, etc. industries and in common human life [1,2]. However, in spite of aforementioned positives, the usability of aluminium alloys is often limited by their rather poor mechanical resistance in comparison to other metals, which led the technicians and scientists to the development of several conventional and unconventional methods aimed at improving their mechanical properties. The most common methods are based on alloying of aluminium by elements such as Zn, Ag, Mg, Li, Ge, Cu, Si, Mn, Cr and Fe during casting, enabling the production of aluminium alloys with substantially enhanced mechanical properties as compared to pure aluminium, or additional heat treatment processes of wrought alloys consisting of solution annealing, quenching and aging to increase the alloy strength influencing their ductility [3,4]. As examples of alternative methods, the forging, extrusion, superplastic-forming (equal channel angular pressing - ECAP or high pressure torsion - HPT) are also cited in the relevant literature [4-9]. Other unconventional methods include adding of ceramics, metallic or intermetallic second phase particles with high elastic moduli into the aluminium matrix leading to the formation of a composite alloy with improved properties [10-12].

In this paper, a new unconventional method consisting of forming a sacrificial nickel coating on aluminium substrates and its subsequent annealing at temperatures above the melting point of aluminium has been used to produce bulk hypereutectic aluminium alloys. The differences in formation processes and in resultant microstructures of two different nickel coated alloys (commercially pure aluminium and hydronalium) were
studied by means of available materials science techniques, i.e. metallography, light microscopy, scanning electron microscopy and energy dispersive microanalysis.

2. MATERIAL AND METHODS

Sheets of commercially pure 99.5 wt% Al and AlMg3 alloy (Al Invest Inc., Czech Republic) were used as the substrate. The sheets surface were ground with abrasive paper #600, etched with the Tucker solution (45 ml HCl + 15 ml HNO₃ + 15 ml HF + 25 ml H₂O) and washed with acetone in an ultrasonic cleaning bath before coating deposition. The 99.0 wt% Ni powder (GTV GmbH, Germany) with a nominal particle size of 20-45 μm was sprayed onto the sheets surface by means of the High Velocity OxyFuel technique (HVOF). The carrier gas mixture of propane (40 L/min), oxygen (160 L/min) and nitrogen (3.2 L/min) was set up for HVOF spraying. The average as-sprayed coating thickness was 150 μm. After the spraying, the sheets were sectioned into samples of 80 × 15 × 5 mm in size. The samples were annealed at the temperatures of 660, 680, 700 and 800°C for different dwell timed (2.5 min up to 5 h) in ambient atmosphere.

Specimens were grounded by increasingly finer abrasive papers (up to #4000) and subsequently polished with diamond pastes (down to 1 μm) and with the OP-U colloidal silica final suspension (Co. Struers GmbH). Microscopical investigations were accomplished by means of the light microscope (LM) Olympus GX-50 and the scanning electron microscope (SEM) Philips XL-30 equipped with the energy dispersive X-ray (EDX) spectroscopy microanalyser.

3. RESULTS AND DISCUSSION

An example of the specimens in an initial state, i.e. after the nickel coating deposition and without the heat treatment, are shown in Fig. 1. The microstructure exhibited a typical drawing for thermally sprayed coatings and cold-rolled aluminium sheet, Fig 1a, and hydronalium alloy, Fig 1b. As-sprayed coating consists of 92-94% nickel flattened particles “splat”, voids and oxide particles that are located at triple points and in between the splats. Amount of voids and oxides was of approximately 6-8%. In comparison to the literature, the amount of voids and oxides is rather higher than typically presented value of 1-2% for the HVOF technique [13]. The microstructure of the aluminium and hydronalium substrates is typical for cold-rolled sheets, i.e. crystal grains oriented parallel to the rolling direction and a certain small amount of impurities based on Si, Fe and Mg compounds. The interface between the aluminium or hydronalium alloy sheets substrate and nickel coating is free of apparent mutual diffusion interactions.
Although the nickel coated Al and AlMg3 substrates are in initial microstructure quite similar, the substantially different response and micro-scale changes between these two systems were observed during/after the heat treatment process, see Fig. 2. While the aluminium alloys formed from Ni coated Al (binary system) show almost no difference in dimension, except the rounded edges of the heat treated samples shown in Fig. 2a, the alloys produced from Ni coated AlMg (ternary system) were significantly different in size and shape, see Fig. 2b.

Figure 2 – macrographs of as-sprayed and heat treated samples at the temperature of 800°C for 30 min (a) nickel coated 99.5 commercially pure aluminium and (b) nickel coated AlMg3 hydronalium alloy

The microstructures of nickel coated commercially pure aluminium after the heat treatment are shown in Fig. 3. In comparison with the small-volume samples, where the formation of Al-Al₃Ni eutectic occurred immediately after the appropriate temperature had been reached [14], the large-volume samples undergoes several time-dependent sequences. At first, similarly to annealing below the melting point [15], the two-phase islands consisting of Al₃Ni₂ (adjacent to nickel coating) and Al₃Ni (adjacent to aluminium or silumin substrate) start to form at the initial coating-substrate interface. With increasing dwell-time, these islands are converted into the continual layer and, if the temperature is higher that the melting point, the thin layer consisting of Al-Al₃Ni eutectic is produced within the substrate just below the Al₃Ni continual layer, see Fig 3a. With the increasing dwell, this layer grows until the whole substrate thickness is converted and/or the nickel coating is completely dissolve, see Fig. 3b. In general, at the temperature of 660°C, predominantly coarse eutectic is formed. With temperature increased up to 700°C, the coarse blocks of eutectic dissolve into the rod-like form and needle-like primary Al₃Ni particles, see Fig. 3c. For even higher temperatures, up to 800°C, the time necessary to produce the eutectic is significantly shortened as shown in Fig. 3d. The prolonged dwell-time is clearly related to negligible coarsening of intermetallic phase in eutectic [16]. The initial nickel coating thickness, i.e. the total nickel quantity, also influence the hypereutectic, eutectic and/or hypoeutectic formations. In particular, the formation mechanism of eutectic is not so easy to capture as it is in a homogeneously nickel distributed cast aluminium alloy [17], mainly due to the fact that nickel diffuse from coating area to the substrate through a much longer distances. Moreover, using high-purity aluminium thin substrates, the specific conditions enabling the formation of ultrafine eutectic were found [18], which has never been observed in commercially pure aluminium substrate, probably due to the amount of impurities hindering formation of homogeneous eutectic. Slight changes in the dimensions of samples, in this case due to the rounded edges, are probably also accompanied by a highly-stable natural alumina layer formation, which holds the molten aluminium inside and enable nickel diffusion.
If hydronalium is utilized as a nickel coated substrate, the mechanism of eutectic formation is somewhat different. In comparison to above discussed commercially pure aluminium, only the islands of single phase similar to stoichiometry of the Al₃Ni with a small amount (up to 3 at%) of Mg, start to form at the initial coating-substrate interface. These islands do not convert into the continual layer and the coarse blocks of Al₃Ni phase appears instead. In the next stage, the thin layer consisting of Al-Al₃Ni eutectic doped by Mg is produced within the substrate just below the Al₃Ni like continual layer. This layer features the coarse randomly oriented eutectic, see Fig 4a. Based on the dwell-time, the layer grows through the substrate thickness and a finer eutectic is formed, see Fig. 4b. When studying the remaining part of the coating, the disruption by the matrix phase was observed. At longer dwell and higher temperatures, this effect coupled with the Mg content reduced the stability of protective natural alumina layer formation. Further increase of the temperature up to 700°C resulted in eutectic coarsening and long needles of primary Al₃Ni doped by Mg phase being formed, see Fig. 4c. At the temperature of 800°C, only a short time is necessary to the nickel coating dissolution as shown in Fig. 4d.
Figure 4 – SEM micrographs of nickel coated hydronalium alloys after isothermal heat treatment at the temperature of (a) 660 °C / 60 min, (b) 660 °C / 300 min, (c) 700 °C / 30 min and (d) 800°C / 30 min

4. CONCLUSIONS

The effect of temperature level and isothermal dwell of the heat treatment on the eutectic formation in the nickel coated aluminium and hydronalium alloy substrates was studied. The dissimilar microstructures were produced under the different temperatures and dwells. The formation mechanism of resulting alloys slightly differs. The main difference is of single instead of the two-phase islands formation across the initial coating-substrate interface, if the hydronalium alloy substrate is utilized and no islands or a continual Al$_3$Ni$_2$ layer is formed. Magnesium contained in an aluminium matrix always participate on Al$_3$Ni like phase formation and negatively influences the stable protective alumina layer formation. Nevertheless, for both substrates, the higher temperatures seems to have a negligible influence on resulting alloys production. The undesirable effect of higher temperatures is related to the needle-like primary phase formation and to eutectic coarsening.

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