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Temperature effect on the microstructural development of Al–Ni layered binary couples produced by an unconventional method

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ABSTRACT

In this work, the authors have used an unconventional experimental route that consists of (i) deposition of a nickel coating onto an aluminium substrate and (ii) the subsequent heat treatment of created couple in order to produce intermetallic-layered structures, hypereutectic or bulk aluminide intermetallic alloys. The procedure was conducted by controlling chemical concentrations in the Al-rich corner of Al–Ni binary system. The couple was prepared by high velocity oxyfuel spraying of 99.0 wt.% Ni powder onto the surface of 99.999 wt.% Al sheet. The specimens for heat treatment were manufactured immediately after the spraying. The heat treatment was carried out in a differential thermal analysis apparatus by using the temperature range of 600–1200 °C, thus considering the solid state, transient liquid as well as liquid phase of aluminium, with a constant heating/cooling rate of 5 °C/min in an argon atmosphere. Microstructural development of produced alloys was studied by conventional metallography and scanning electron microscopy. Formed intermetallic layers and compounds were evaluated by using energy dispersive microanalysis and image analysis techniques. The development of a novel ultra-fine eutectic alloy is reported.

1. Introduction

Aluminium and its alloys are the most popular nonferrous materials for a large number of applications in the automotive, aerospace and construction industries, especially because of their excellent oxidation resistance and workability, very low density and high thermal and electrical conductivity [1–4]. The usability of these alloys is, however, often limited by their rather poor mechanical properties (in comparison with iron-based engineering alloys) and, therefore, an extensive effort is invested into the development of new alloys with enhanced properties. The most common methods employed for aluminium alloys and aluminium matrix (nano)composite production are casting and powder metallurgy [5]. Alloying elements such as Zn, Ag, Mg, Li, Ge, Cu, Si, Mn, Cr, and Fe, among others, enable the production of cast or wrought alloys with substantially improved mechanical properties as compared to pure aluminium at the expense of debasement of one or more mentioned physical properties [3]. In the case of wrought aluminium alloys, an additional heat treatment consisting of solution annealing, quenching and ageing is usually applied to increase strength, thus influencing ductility [6,7]. Other alternative methods such as forging, extrusion, superplastic-forming (equal channel angular pressing — ECAP or high pressure torsion — HPT) are also reported on in the relevant literature [7–14]. Some unconventional methods include addition of ceramic, metallic or intermetallic second phase particles with high elastic moduli into the aluminium matrix leading to formation of composites with improved properties [15–18]. As for the cast alloys, if the eutectic reaction exists, it can be properly utilized for strengthening aluminium through formation of intermetallic phases. In general, based on concentrations of individual constituents, the hypo-eutectic, eutectic or hyper-eutectic alloys can be produced [19–21]. It is, however, important that the mixture is allowed to cool down slowly to ensure diffusion, assuring that the alloy will exactly match with the composition dictated by its phase diagram [22].

The Ni–Al binary phase system contains five stable ordered intermetallic compounds, namely $NiAl_3$, Ni_2Al_3, NiAl, Ni_5Al_3 and Ni_3Al. From these, mainly NiAl and Ni_3Al phases receive a major scientific attention due to their use in high-temperature structural applications, such as nickel solid solutions strengthened by Ni_3Al phase or NiAl and/or Ni_3Al-based high-temperature coatings [23–26]. Other three less known stable phases, i.e. $NiAl_3$, Ni_2Al_3 and Ni_5Al_3, have not been considered for high-temperature applications mainly because of their low

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melting temperatures being respectively 854, 1133 and approximately 700 °C. Further limitations stem from their brittle nature at low temperatures. On the other hand, by controlling the dispersion of these phases in the aluminium matrix it is possible to form homogenous or functionally graded metal-intermetallic alloys thereby overcoming aforesaid limitations. Clearly, a new information on these phases (especially on $NiAl_3$ and Ni_2Al_3), such as the formation mechanism, an assessment of their physical and mechanical properties or analysis of their morphology and distribution should lead to better designing of novel aluminium alloys, Ni/Al/Ni interconnections, and/or intermetallic layered structure production [27–31].

Both the formation and microstructural evolution of alloys in the Al-rich corner of the Al–Ni binary system are reported in this study. The unconventional production technique consists of deposition of a nickel coating directly onto an aluminium substrate, which is followed by heat treatment at temperatures below and above the Ni–Al eutectic melting point. The mutual interaction of both constituents was investigated in solid state, transient liquid and liquid phase of Al, respectively. Microstructures of the intermetallic layered structures, hypereutectic and/or bulk intermetallic alloys, created by Al + $NiAl_3$ eutectic, $NiAl_3$, and Ni_2Al_3 primary intermetallic layer/phases are studied and discussed.

2. Material and methods

2.1. Experimental material

An ultra-high purity aluminium (99.999 wt.%) cold rolled sheet of 100 × 20 × 3 mm in size, produced by Al Invest Inc., of Czech Republic was used as a substrate. With the aim to disrupt an oxide layer and to degrease the substrate surface before coating deposition, the Al-sheet was ground with an abrasive paper #600, etched with Tucker solution (45 ml HCl + 15 ml HNO_3 + 15 ml HF + 25 ml H_2O) and washed in acetone. The initial substrate arithmetic average roughness after etching in Tucker solution was $S_a = 2.9 \pm 0.1$ μm. The water-atomized spherical nickel powder, with the nominal composition of 99.0 wt.% Ni and particle size of 20–45 μm, which was purchased from GTV GmbH, Germany, was sprayed onto the aluminium sheet surface using 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 used for HVOF spraying. The average as-sprayed coating has the thickness of 100 ± 20 μm. After spraying, the samples of 2.5 × 2.5 × approx. 0.5 mm in size were cut out from the Ni-coated side of the aluminium sheet. In order to achieve a uniform weight of 10.5 ± 0.3 mg, the aluminium side (opposite to nickel-coated side) was ground with an abrasive paper #1000.

2.2. Experimental procedure

Differential thermal analysis (DTA) measurements were performed using a Setaram SETSYS-1 apparatus and alumina crucibles. Heating/cooling cycle was applied by altering the temperature between T_R = 20 °C and T_1 = 600 °C (solid state treatment), T_2 = 640 °C, T_3 = 655 °C (transient liquid phase treatment), T_4 = 700 °C, T_5 = 721 °C, T_6 = 900 °C, and, T_7 = 1200 °C (liquid phase treatment) respectively, with a constant heating/cooling rate of 5 °C min^{-1}.

Cooling started immediately when the temperature of T_x, (i.e. the temperatures T_1–T_7), had been reached. The measuring cell was emptied and refilled with high purity argon (5 N) three times prior to the measurements. An argon gas flow of 2 L h^{-1} was maintained during all tests. S-type rod PtRh10% thermocouples were used for temperature measurements. Prior to experiments, the thermocouples were calibrated according to the standard [32] by measuring the extrapolated onset temperatures of the melting peaks of Al (660 °C) and Ni (1455 °C) standards. The calibration accuracy was ±1 °C. Since the temperature calibration was not performed during cooling, the undercooling effects are discussed only qualitatively.

2.3. Characterization techniques

The specimens used for microstructural analysis were ground by increasingly finer abrasive papers (up to #4000), subsequently polished with diamond pastes (down to 1 μm) and finally polished using OP–U colloidal silica suspension. Characterization of obtained specimens also included the scanning electron microscope (SEM) Philips XL-30 equipped with the energy dispersive X-ray (EDX) spectroscopy microanalyser by EDAX. The layer thicknesses and the amount of eutectic or individual primary phases were determined based on the SEM images by using the NIS Elements, AR 2.3 image analysis software.

3. Results and discussion

3.1. Initial state

A cross-section image of the initial as-coated state is presented in Fig. 1. The micrographs show a typical microstructure of HVOF-sprayed coatings and of cold-rolled aluminium sheets. The coating shown in Fig. 1(b) consists of nickel splats (around 94%), voids and oxide particles that are located at triple points and in between the splats. The area fraction of voids and oxides estimated by means of image analysis of coating cross-section micrographs is almost 6%. The larger amount of oxides, as compared to 1% commonly reported for the HVOF technique in the literature [33], presumably arose from employed deposition conditions that had to be chosen experimentally

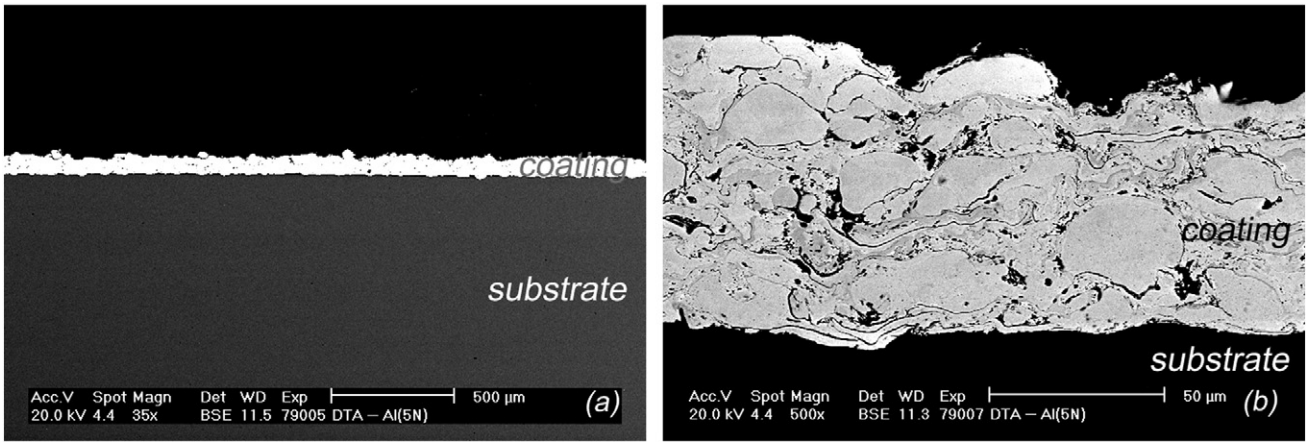

Fig. 1. SEM micrographs of initial-state (a) Ni coating on Al sheet, (b) detail of HVOF sprayed nickel coating–aluminium substrate interface.