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Laser additive synthesis of high entropy alloy coating on aluminum: Corrosion behavior

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ABSTRACT

High entropy alloy coatings were synthesized on aluminum substrate by laser surface engineering. Dilution from the substrate was minimized with the aid of multi layered coatings. Furthermore, higher laser input energy during processing lead to uniform mixing amongst the components resulting in formation of evenly distributed high entropy alloy phases throughout the matrix. This resulted in enhanced corrosion resistance for the coatings in near neutral NaCl solution.

1. Introduction

High entropy alloys (HEAs) are emerging class of alloys which are based on the presence of multiple components, in equimolar or near equimolar ratio. Multiple components/elements give rise to increasing effects of configurational entropy resulting in stabilization of a randomized solid solution phase [1,2]. HEAs have been reported to posses excellent set of bulk properties such as mechanical strength and ductility [3,4], soft magnetic properties [5]. They also have superior surface properties like high hardness [6,7], better wear performance [8,9] and superior corrosion resistance [7,10,11] making them suitable candidate as coating material. Additionally, considering the complex nature of the system, it is more feasible to use HEAs as coatings rather than bulk so as to take advantage of the properties possessed by theses materials. The field of HEA coatings is new and some of the recent studies have reported synthesis of HEA coatings by various methods like magnetron sputtering [12], electro spark process [13], plasma arc cladding [14], and also by laser processing [15–18]. Laser processing is particularly attractive method which provides quick heating of the coating materials to high temperatures followed by the rapid quenching. Thermal entropy contributions add to the configurational entropy at high temperatures making conditions suitable for HEA formation and rapid quenching ensures retention of the HEA phase at room temperature [18,17]. Ye et al. [15] synthesized AlxFeCoNiCuCr (*x* varying from 1 to 2) HEA alloy on aluminum by laser cladding. The high temperature hardness, abrasion performance, and corrosion response of the coatings synthesized with varying amounts of Al were tested. Optimum set of properties were obtained for intermediate Al content of 1.8 by atomic ratio. Zhang et al. [17] have reported the synthesis of FeNiCoSiCrAlTi HEA coating on Q235 steel substrate using laser processing. It was observed that HEA coating formed for higher concentration of Fe (6FeNiCoSiCrAlTi). The evolution of a simple BCC HEA phase/microstructurewas attributed to the rapid quenching during laser processing. There are many compositions possible under HEA category and one of the well known systems is Al–Fe–Cr–Co–Ni system, possessing attractive set of properties such as mechanical properties [4], high hardness and [6,7] corrosion resistance [7,11] and good elevated temperature properties [19]. Better micro mechanical properties of this system have been predicted by theoretical calculations [20]. Previous investigation from present research group has reported successful synthesis of Al–Fe–Cr–Co–Ni by laser surface engineering [18]. Detailed investigation about microstructure evolution was carried out and influence of processing and thermodynamic parameters on stability of HEA phase were investigated. Current report focuses on corrosion properties of laser synthesized Al–Fe–Cr–Co–Ni coating. Effect of additively produced double layered coatings on dilution from the substrate and subsequent corrosion resistance is reported.

2. Methods and materials

The HEA coatings were synthesized using high power Ytterbium doped Nd-YAG (YLS-3000IPG) continuous wave laser with

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a wavelength of 1064 nm and laser beam diameter of 0.6 mm. Energy inputs of 21 J mm^{-2} and 25 J mm^{-2} were obtained with combination of input power of 1000 W and 1200 W with fill spacing/ laser track overlap of 0.4 and 0.3 mm respectively. The beam scanning speed was kept constant at 100 mm/s. Al,Fe,Co,Cr, and Ni elemental powders (with average particle sizes ranging from 1 to 3 μm) with a purity of 99.99% in equiatomic ratio were mixed with a water soluble organic binder (LISI W 15853 obtained from Warren Paint and Color Company, Nashville, TN, USA) to form a thick slurry. The slurry was then uniformly deposited (average thickness ~300 μm) onto 1100 Al substrate using air-spray technique. The substrates were thoroughly cleaned in acetone and dried prior to deposition of the powders. The laser parameters were selected based on prior experience after many trials in order to obtain a uniform and sound coating. Although these parameters are not the most optimum, primary purpose of this preliminary study was to produce HEA coatings using laser processing and study their corrosion behavior. Focus towards exploration of large number of materials and laser process parameters along with the computational modeling for optimization of these laser processing parameters is being given in ongoing work and details would be reported in the future publications. Energy input of 21 J mm^{-2} was used to produce single layered coating. Additional layer was deposited with same procedure as described above and scanned with 21 J mm^{-2} and 25 J mm^{-2} laser energy input to obtain double layered coating under two different conditions. All the coatings were processed using Ar as the shielding gas so as to avoid/minimize undesirable effects such as oxidation. The synthesized coatings were further prepared for microstructural characterization in the section perpendicular to the laser scanning direction. Sample preparation was done using standard metallography methods. Aqua-regia solution was used as etching reagent to reveal the microstructures. X ray diffraction (XRD) analysis was carried out for phase detection with Cu Kαradiation (wavelength = 0.154 nm) using step size of 0.025°, and a scan speed of 2 degree/minute) (Rigaku Altima). Scanning electron microscope (SEM) equipped with energy dispersive spectroscopy (EDS) (ESEM quanta) was used for microstructural analysis. Potentiodynamic polarization tests were performed using SP-300 modular research grade potentiostat/galvanostat (Bio-logic USA, Knoxville, TN) equipped with 250 ml standard three electrode cell employing saturated calomel electrode (SCE) as reference, platinum mesh as counter electrode and sample as the working electrode. In the present study, 3.5 wt% NaCl solution of pH of 6.9 ± 0.2, prepared from laboratory grade NaCl (Sigma Aldrich) and ultra-pure deionized water was used as an electrolyte. All the tests were carried out at room temperature. The area of 1 cm^2 of coated samples and untreated Al 1100 substrate material was tested for corrosion response. Both the coated samples and untreated Al 1100 substrate samples were lightly polished to same surface roughness (~4 μm). Before commencing polarization scan, the open circuit potential (OCP) was stabilized for 1 h. The scan was initiated at the scan rate of 0.5 mV/s and by sweeping the potential from −100 mV with respect to OCP. The polarization scans were terminated at 1 mA/cm^2 for all the conditions. Values of corrosion potential (E_{corr}) and corrosion current (i_{corr}) were extracted from the Tafel plots. Samples were cleaned with de-ionized water and ethanol prior to observation of corroded surface under SEM.

3. Results and discussion

XRD analysis revealed the presence of HEA phases (BCC1 and BCC2), Al_3Ni, and $FeAl_3$ compounds for all the processing conditions (Fig. 1). The formation of two HEA phases with slightly different lattice parameters (BCC1: 2.81 Å and, BCC2: 2.77 Å) and compound phases have been discussed in detail elsewhere [18] with the aid of various thermodynamic parameters related to HEA formation explored in open literature [21,22].

Back scattered electron (BSE) SEM analysis of cross section of coated samples revealed formation of coating with the phases of gray and bright contrast (Fig. 2a–c) in various proportions. Previous investigation on this coating system with the aid of transmission electron microscopy has indicated that the bright contrast phase consisted of mixture of HEA phases [18] which is also confirmed by XRD analysis (Fig. 1). With single layer of coating (21 J mm^{-2} as energy input), the gray contrast phase was present near the surface of the coating and bright contrasted phase underneath (Fig. 2a). When the combination of additional coating layer with the same energy input (21 J mm^{-2}) was employed, even distribution of gray contrasted phase throughout the coating occurred (Fig. 2b). This type of occurrence could be attributed to melting of Al substrate during the processing which resulted in dilution effects leading to Al enrichment within the coating, giving rise to gray contrast, considering Al being the lightest amongst the elements used in this study. With double layer and higher energy input (25 J mm^{-2}), uniform mixing of the coating components occurred thereby abating the dilution effect with formation of the bright contrasted phase throughout the coating (Fig. 2c). Effect of dilution was also realized in the morphology of the phase. The lower Al content in case of sample processed with lower energy input (21 J mm^{-2}) and single layer led to formation of predominately thick primary columnar dendritic morphology (~2 μm thickness) (Fig. 2d). Furthermore in

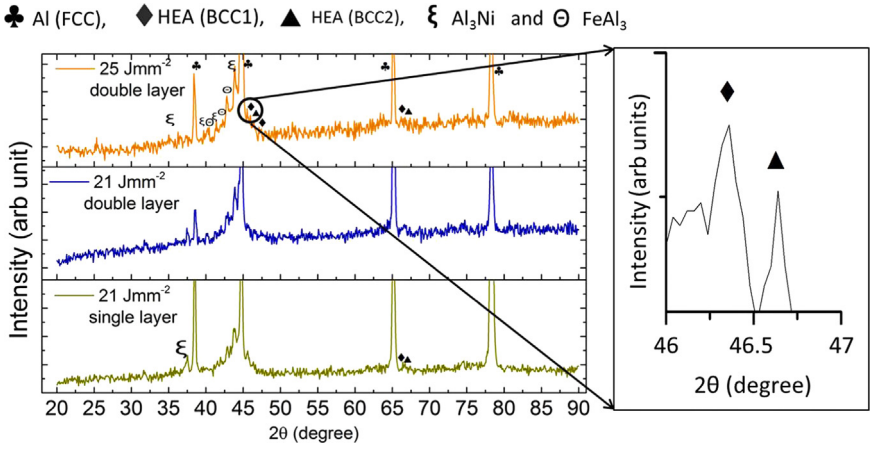

Fig. 1. XRD pattern for coatings synthesized using various laser processing conditions.