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FABRICATION OF ALUMINUM MATRIX COMPOSITE REINFORCED WITH $Al_{0.5}$CoCrCuFeNi HIGH-ENTROPY ALLOY PARTICLES

The aluminum composite with dispersed high entropy alloy were developed by stir casting involving the powder-in-tube method. First, $Al_{0.5}$CoCrCuFeNi high entropy alloy (HEA) powder was made by mechanical alloying, and the powder was extruded in a tube-type aluminum container to form HEA precursor. The extruded HEA precursor was then dispersed in the aluminum matrix via stir casting. As a result, Fe-Cr-Ni based high-entropy phases was uniformly formed in the aluminum matrix, revealing ~158, 166, 235% enhancement of tensile strength by incorporating 1, 3, and 5 wt% HEA particles, respectively.

Keywords: Aluminum matrix composite; high entropy alloys; reinforcement; powder-in-tube; stir casting

1. Introduction

Aluminum alloys exhibit excellent specific strength, workability, thermal conductivity, and recyclability. Hence, they are now used not only in high-tech industries, such as automotive and aerospace but also in our daily life [1-3]. However, precipitates and second phases in aluminum alloys can easily form at warm and high temperatures, significantly degrading their performance and limiting their range of use [4,5]. Therefore, many researchers have suggested that aluminum matrix composites have better thermal stability than aluminum alloys. However, poor wettability between the aluminum matrix and ceramic or carbon reinforcement has been considered as a technical bottleneck in this field [6-8].

High-entropy alloys (HEAs) that are multicomponent alloys have outstanding mechanical properties and thermal/electrical stabilities comparable to those of ceramic materials though they are crystalline metals because of their metallic bonds [9-11]. These properties are derived from the high entropy and sluggish diffusion rate of HEAs because of the lattice distortion effect caused by random mixing of elements and cocktail effects [12]. Its intrinsically low reactivity, slow diffusion rate, high strength, hardness, and good wetting ability with the metallic matrix makes it an excellent choice for use as a reinforcement in Al matrix composites.

Several studies have been conducted to produce metal matrix composites containing HEA phases, with a majority employing powder metallurgical routes. Chen et al. [13] used vacuum hot pressing to prepare AlCoNiCrFe particle-reinforced copper matrix composites. It has been reported that there is no obvious reaction between the copper matrix and HEA-reinforced particles. Praveen et al. [14] used stir casting to add a cast-manufactured Al-20Cu-10Mg high-entropy alloy to a 2024 aluminum alloy matrix, which improved mechanical properties such as yield strength, tensile strength, and Young's modulus. According to this research, when 15% HEAs were added to 2024 aluminum alloy, yield strength was improved 1.95 times and tensile strength about 1.7 times. However, the HEAs used in this study differed in composition from commonly used HEAs; this type of HEA was used to solve the separation phenomenon caused by the difference in surface energy between the molten aluminum and commonly used HEA particles.

As discussed earlier, most research [13-19] on dispersing HEA particles in a metal matrix is conducted using the powder metallurgical method because the difference in surface energy between the molten metal and the HEA particles makes it is difficult to disperse HEAs in a metal matrix using the casting method. In this study, the feasibility of using the casting method by adding HEAs in the form of a powder in tube (PIT) to uniformly disperse HEA particles in the aluminum matrix was evaluated. Among

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the many compositions of HEA, $Al_{0.5}$CoCrCuFeNi alloy was applied to the experiment. $Al_{0.5}$CoCrCuFeNi alloy has a mixing enthalpy (ΔH_{mix}) of –5 kJ/mol or less to minimize decomposition in molten aluminum during stir casting and partial decomposition of $Al_{0.5}$CoCrCuFeNi alloy can form a reinforcement phase such as $Al_{70}Co_{15}Ni_{15}$, Al_7Cr, Al_2Cu, Al_3Fe. $Al_{0.5}$CoCrCuFeNi alloy was added in the form of a PIT, and high shear stress was applied to uniformly disperse the HEAs in the aluminum matrix. The microstructures of the powders and aluminum matrix composites were evaluated, and tensile tests were conducted to evaluate whether HEAs could act as reinforcements in the aluminum matrix.

2. Experimental procedure

2.1. High entropy alloys

First, Al, Co, Cu, Fe (<45 μm, 99.9%, RND Korea), Cr (<45 μm, 99.99%, RND Korea), and Ni (<45 μm, 99.95%, RND Korea) powders were prepared to fabricate high-entropy powders as starting materials. The $Al_{0.5}$CoCrCu0.5FeNi HEA powder was synthesized by high-energy ball milling using an attrition mill. High-energy ball milling was conducted up to 500 rpm for 72 h in argon atmosphere using stainless steel balls (5 mm) and vials, and the ball-to-powder ratio was 10:1.

2.2. Powder in tube (PIT)

Fig. 1 shows the schematic images of the overall experimental process. Gravity casting was used to fabricate 2-inch pure aluminum billets, and the manufactured HEAs were weighed and filled in the core of the hole drilled 2-inch billet. Following that, an Extruder with 200 tons of hydraulic force or pressure was used to extrude the 2-inch billet into a ϕ20 rod.

2.3. High shear stress stir casting

Aluminum pellets (<1 cm^3, 99.99%, RND Korea) were charged into a chamotte jar and dissolved in high-frequency induction, and PIT was obtained in the molten aluminum. It was held at 700°C and stirred at 300 rpm/30 min before casting.

2.4. Analysis

X-ray diffraction (XRD, Rigaku, miniFlex600) with Cu K_α radiation (0.1542 nm) generated at 40 kV was used to perform a phase analysis. Additionally, X-ray diffraction was used to confirm the formation of the high-entropy powder. The microstructures of the powder and cast alloy were observed using a field emission scanning electron microscope (FE-SEM, FEI, Verios G4 UC). Furthermore, we performed quantitative analysis using energy dispersive spectroscopy (EDS, EDAX, Octane Elect EDS System) to determine the distribution of each atom. The cast materials were processed into tensile test specimens (ASTM E8) to evaluate their mechanical properties (MTS810, MTSC).

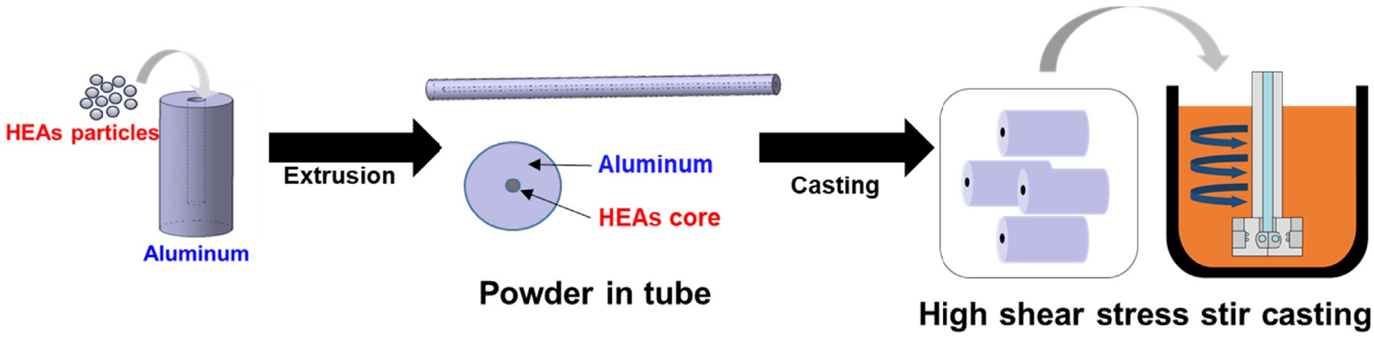

Fig. 1. Schematic image of overall experimental process

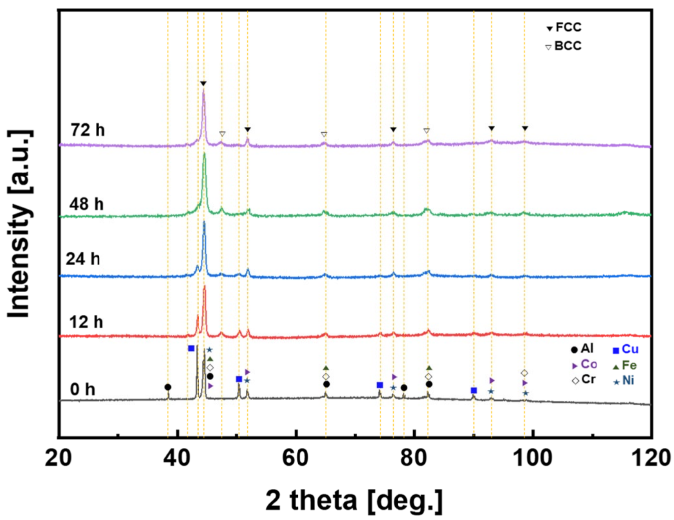

Fig. 2. X-ray diffraction patterns of $Al_{0.5}$CoCrCuFeNi alloys with mechanical alloying time

3. Results and discussion

High-energy ball milling was used to mechanically alloy pure Al, Co, Cr, Cu, Fe, and Ni as starting materials. Fig. 2 shows that the XRD patterns varied according to the duration of mechanical alloying. Solid solution FCC and BCC phases were observed in the patterns obtained from the ball-milled powders, which is typical for Al-containing CoCrCuFeNi alloys [12].

As the ball-milling time increased, there was a decrease in peak intensity, and some peaks became invisible owing to severe lattice distortion [10]. After ball milling for 48 h, peak broadening and only BCC and FCC peaks were observed. So, the optimal milling time is considered to be 48 hours.

Fig. 3 shows the results of the microstructure analysis of the HEA particles synthesized using SEM-EDS. The synthesized HEA particles through ball milling for 48 h was confirmed to have a fine size of approximately 5 µm or less through microstructure analysis; as a result of EDS analysis, it was confirmed that the elements were uniformly dispersed. Based on the data in Figs. 2 and 3, after ball milling for 48 h, we confirmed that multiphase HEAs were well synthesized.

In general, when reinforcement particles are added to molten metal, they float on the surface of the molten metal owing to the difference in surface energy and density [7]. In this study, to minimize this problem, physical contact was induced between the HEA particles and aluminum through extrusion during the manufacturing process of PIT. HEA particles were added in the form of PIT to molten aluminum, and the reinforcement was uniformly dispersed in the aluminum matrix through high shear stress stirring. A composite of Al-1, 3, and 5 wt.% HEA was synthesized using the high shear stress stir casting method. Additionally, to confirm the effect of the PIT method, non-PIT HEA particles were added to the molten aluminum, and the mechanical properties were compared. Fig. 4 shows the microstructure of the Al-5 wt.% HEA composite analyzed by SEM-EDS. SEM images show uniformly dispersed HEA particles in the aluminum matrix without the decomposition of HEAs. Additionally, intermetallic compounds between the HEA elements and aluminum were not identified. However, it was confirmed that Co-rich and Cr-rich regions appeared, and it is expected that analysis of the behavior of Co and Cr in Al–HEA composites through long-term heat treatment will be necessary in the future.

The yield strength, tensile strength, and elongation of the samples are listed in TABLE 1. The results in TABLE 1 show the average values after five experiments for each sample. To evaluate the effect of the PIT method, Al-1 wt.% HEA composites were prepared in two ways based on the HEA addition method. The tensile strengths of the pure aluminum and Al-1 wt.% HEA (non-PIT) were 50.96 and 60.93 MPa, respectively. From these results, it was confirmed that the addition of HEA in aluminum improved the mechanical properties of pure aluminum, and HEA powder is considered sufficiently applicable as a reinforcement in aluminum matrix composites. Moreover, when HEAs were added in the form of PIT, the tensile strength was 80.62 MPa, confirming that the addition effect of the PIT method was greater than that of the conventional addition method. As the amount of HEA

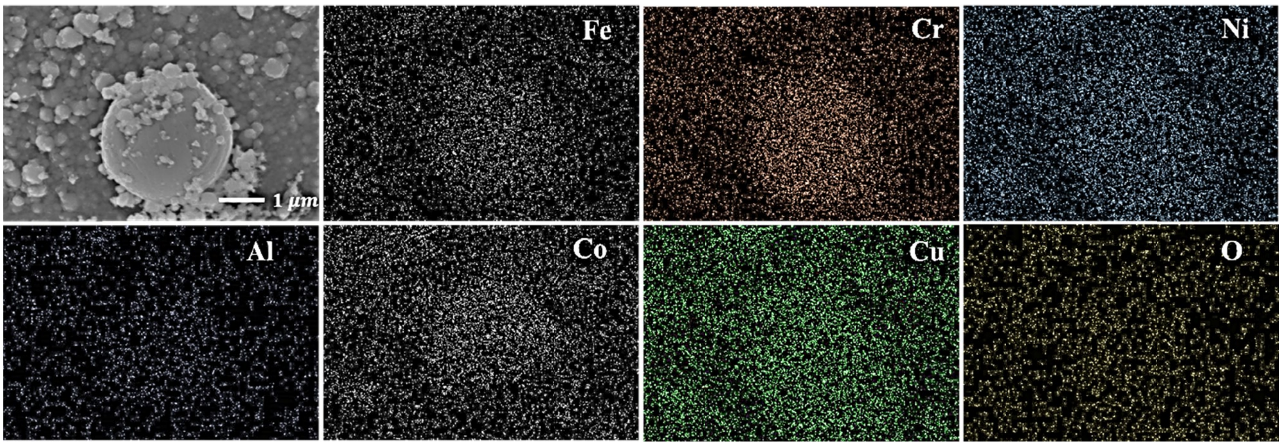

Fig. 3. Microstructures of synthesized HEAs particles through ball milling for 48h and quantitative analysis via SEM-EDS

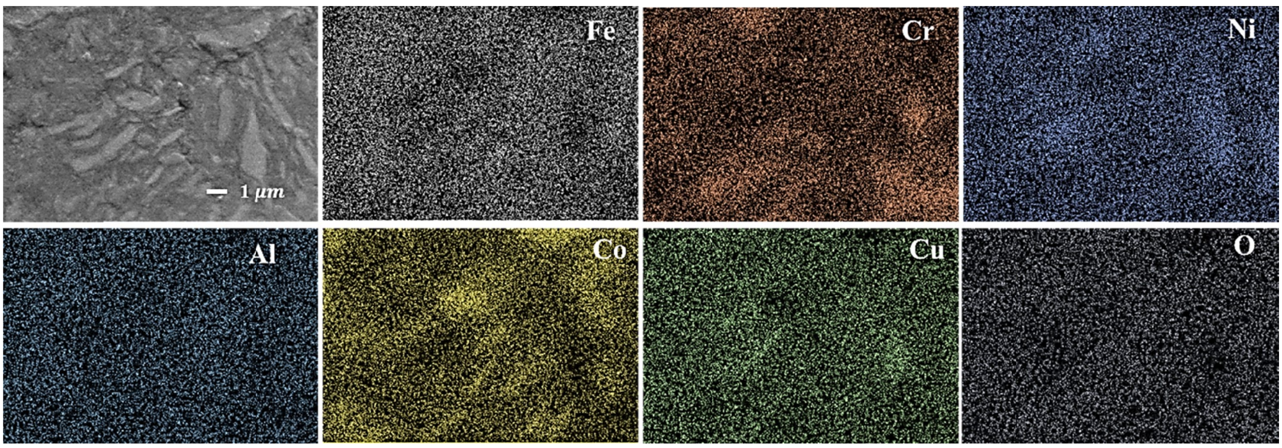

Fig. 4. Microstructures of Al-5 wt.% HEA composite and quantitative analysis via SEM-EDS

particles added increased, the yield, tensile strength increased and elongation decreased owing to the tradeoff between strength and elongation. In the case of Al-5 wt.% HEA composite, it was confirmed that the yield strength and tensile strength were more than doubled compared to pure aluminum. The improvement of mechanical properties is considered that grain refinement of aluminum matrix was presented by the pinning effect through the added HEAs particles. Through this study, it was confirmed that HEA particles can sufficiently act as reinforcing materials in aluminum composites, and it is expected that HEA particles can be applied to high-strength aluminum alloys.

TABLE 1

Mechanical properties of pure aluminum and Al-1, 3, 5 wt.% HEAs composite

Sample	PIT	Yield strength (MPa)	Tensile strength (MPa)	Elongation (%)
Pure Aluminum	—	29.9	50.96	36.35
Al-1 wt.% HEA	X	38.71	60.93	30.97
Al-1 wt.% HEA	O	44.84	80.62	24.85
Al-3 wt.% HEA	O	49.92	84.62	20.26
Al-5 wt.% HEA	O	67.27	119.77	19.35

4. Conclusion

A high-entropy alloy of $Al_{0.5}$CoCrCuFeNi was prepared using a mechanical alloying method and dispersed in an aluminum matrix to evaluate the feasibility of using HEAs as reinforcements in the aluminum matrix. Mechanical alloying reduced the intensity of the XRD peaks during the high-energy milling of Al, Co, Cr, Cu, Fe, and Ni powder for up to 48 h, and peak broadening occurred owing to decreased crystalline size and lattice distortion effect. After ball milling for 48 h, we confirmed that multiple elements were uniformly mixed and that they formed only the FCC and BCC phases. To effectively add HEA particles during the casting process, it was applied in the form of a PIT. There was a difference in mechanical properties depending on whether the PIT method was applied; when the PIT method was applied, it was confirmed that the mechanical properties improved by approximately 30%. The fabricated HEA particles were successfully dispersed in the aluminum matrix by the high shear stress stir casting method, and it was confirmed that the HEA particles were uniformly dispersed in the aluminum matrix without decomposition or formation of intermetallic compounds. In the case of Al-5 wt.% HEA composite, it was confirmed that the yield strength and tensile strength were more than doubled compared to pure aluminum.

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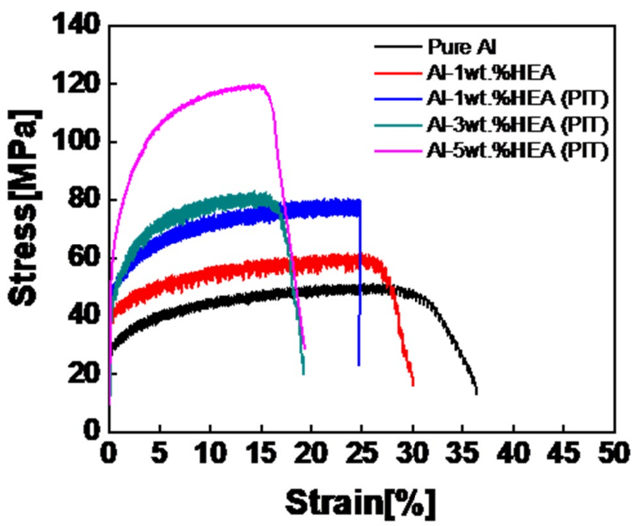

Supplementary Fig. 1. Stress-strain curves of Al-HEA composite