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Original Research Paper

# Microstructure and property characterization of Al-based composites reinforced with CuZrAl particles fabricated by mechanical alloying and spark plasma sintering

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ABSTRACT

In the present work, CuZrAl metallic glass particles were synthesized by mechanical alloying method. High relative density Al-based composites (ABCs) reinforced with different volume fraction of CuZrAl particles have been fabricated by spark plasma sintering (SPS) technique. The microstructures, mechanical properties and corrosion resistance in seawater solution of the ABCs were investigated. The sintered products are all composed of fcc-Al, $Al_3Zr$ and $CuAl_2$ phases. For CuZrAl addition, bright and network precipitates are clearly observed in the Al matrix. On account of the interdiffusion of Al and Cu atoms between matrix and reinforcement, the ABCs present the good interfacial bonding. Compared with SPS-ed pure Al bulk, ABCs possess the excellent mechanical properties. It is mainly ascribed to the second phase strengthening, continuously distributed precipitates, high relative density or bonding interface, and grain refinement strengthening. Thereinto, combined with a degree of plastic strain, the composite with 20 vol% CuZrAl reinforcement reveals the best micro-hardness (290 HV), and the highest yield strength and fracture strength of 408 and 459 MPa, respectively. Moreover, the ABCs bear the better pitting resistance with wide passive region in seawater solution.

## 1. Introduction

Particulate-reinforced metal matrix composites (MMCs) have been attracted considerable attention for several decades [1]. Among this, Al-based MMCs as advanced engineering materials have been widely used in aerospace, defense and automotive applications fields. They possess the excellent performance characteristics, such as high strength, low density, high elastic modulus, good fatigue resistance and wear resistance [2,3].

For conventional MMCs, ceramic particles as the most widely used reinforcements display a certain degree of disadvantages, such as the poor wetting with metal matrix and the tendency to agglomerate and form clusters [4]. These drawbacks can adversely affect the final mechanical properties and corrosion behaviors of the composites [5]. Metallic glasses have attracted much attention due to their high strength and hardness, good corrosion resistance, and excellent functional performance [6]. It has been reported that metallic glass reinforcements effectively improved the mechanical properties of Al-based [7–10], Fe-based [11] and Ti-based [12] metal matrix composites.

The metallic glasses used as the reinforcements possess a series of superiority compared with ceramic reinforcements. It suggests that metallic glasses are expected to promote the atomic diffusion at the reinforcement/matrix interfaces and induce the similar coefficient of thermal expansion between matrix and reinforcement [10,13]. The latter could reduce the internal stresses produced in the cooling process [14].

Among different processing routes for the fabrication of MMCs, the powder metallurgy through solid-state sintering is particularly suitable for the composite preparation owing to the excellent control over the microstructure, particle size, volume fraction of matrix and reinforcement [15]. The high thermo-efficiency and quick heating-up of powder particles are provided by the spark plasma sintering (SPS) technique. The good self-purification of powder particle surface enabled a fast sintering forming at a relatively low temperature [16]. Moreover, the high sintering speed and low sintering temperature could effectively restrain the grain growth during heating process [17].

In present study, we used as-milled CuZrAl glassy particles as the reinforcement in pure Al matrix, which were fabricated by

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mechanical alloying (MA) method. The homogeneous and continuous distribution of the reinforcement particles can induce the better mechanical properties. Several reports have been exhibited that the contents of additive reinforcements in Al-based composites were greater than or equal to the volume fraction of 10 vol% [8,10,15]. Therefore, different volume fractions of 10, 20, and 30 vol% of the CuZrAl glassy powders were selected in this study. These mixed samples are denoted as Al10, Al20, and Al30 for the pure Al powders with 10, 20 and 30 vol% CuZrAl, respectively. Then the Al-based composites (ABCs) reinforced with CuZrAl glass particles were consolidated by SPS technique. The microstructures, micro-hardness (HV), compression properties as well as corrosion resistance in seawater solution were investigated. The CuZrAl reinforcement and Al matrix all possess Al element with different concentration, which promotes the diffusion of Al atoms between the matrix and reinforcement interface. It is beneficial to enhance the adequate compatibility during the sintering process. In this scenario, the SPS-ed CuZrAl/Al MMCs would be expected to be obtained with the high sintering quality and excellent mechanical properties and corrosion resistance.

## 2. Experimental procedures

The powders of Cu Zr (>99.5 wt% purity, ≤200 mesh) and Al (>99.9 wt% purity, ≤200 mesh) were mechanically alloyed to prepare equiatomic CuZrAl (at.%) glass particles. This progress is carried out using a high-energy planetary ball mill (Fritsch P6) at a rotation speed of 300 rpm (revolutions per minute) in an argon atmosphere. The chromium steel vial and 304 L balls were used and the ball-to-powder weight ratio is 15:1. In order to avoid over heating in the vials, the milling procedure was interrupted each 20 min and halted for 10 min. Beyond that, the milling process was interrupted at various time to obtain the as-milled samples for characterization. The pre-sintered powders were obtained by the homogeneous mixing with pure Al powder and CuZrAl glass particles milled after 120 h (10, 20 and 30 vol%) through 10 h.

Consolidation of the mixing powders was performed using SPS technique. High strength heat-resistant graphite punch and die with an inner diameter of 10 mm were used. For the tested samples, SPS was done at sintering temperature of 773 K and sintering pressure of 80 MPa for 30 min. The sintered samples were cylindrical shape with a dimension of $\phi 10 \times 16$ mm.

The phase constitution of the tested samples was identified by X-ray diffraction (XRD, Rigaku D8 Advance) using Cu $K_\alpha$ radiation ($\lambda = 0.15406$ nm). The thermal properties were investigated by a differential scanning calorimeter (DSC, Mettler-Toledo) at a heating rate of 20 K $\text{min}^{-1}$ under a continuous flow of purified argon. The microstructure and chemical composition were investigated by FESEM (QUANTA FEG 250) coupled with energy dispersive spectrometry (EDS). The HV of the SPS-ed samples was measured with Vickers hardness tester with a load of 200 g and a duration time of 15 s. The obtained HV values are the average of 15 indentations for each sample. The density of these sintered samples was determined according to Archimedes' principle method with distilled water as the suspending medium using an electronic analytical balance. The compression treatments of the samples were tested in a universal material testing machine (MTS). The compression speed was controlled at strain rate of $1 \times 10^{-3}$ $\text{s}^{-1}$. The samples for the compression tests were cylindrical shape with a dimension of $\phi 10 \times 15$ mm.

The corrosion behaviors of the SPS-ed samples were conducted by an electrochemical polarization measurement using the electrochemical workstation (LK2005A). All studies were performed in 3.5% NaCl solution using a three-electrode cell with a platinum mesh as a counter-electrode. Bulk sample and saturated calomel electrode (SCE) act as a working electrode and reference electrode, respectively. Prior to electrochemical measurements, all samples were ground with 2000-grit SiC papers.

## 3. Results

Fig. 1 illustrates XRD patterns of phase evolution for the CuZrAl particles milled at different times. The XRD pattern of mixed powders milled for 0 h contains Cu, Zr and Al diffraction peaks. Partial Cu and Zr phases still exist and Al phase disappear when the milling time reaches 5 h. Increasing milling time to 10 h, it shows a broad halo overlapped with a Zr phase peak at 2θ = 36.7. A single broad halo is observed without any crystalline peaks at 30 h of milling time, indicating the existence of glass phase. Prolonging milling time from 60 to 120 h, the glass phase still exists, indicating that the glassy phase possesses the better mechanical stability.

The DSC curve of the as-milled CuZrAl glassy particles at 120 h milling time is given in Fig. 2. One exothermic peak is clearly found, presenting the crystallization of amorphous phase. The onset crystallization temperature ($T_x$) and crystallization peak temperature ($T_p$) are 972 and 990 K, respectively, and the melting point ($T_m$) is up to 1547 K.

Fig. 3 displays FESEM patterns of as-milled CuZrAl alloy powders after different milling times. Compared to the initial mixing powders (Fig. 3(a)), the size of powder particles after the milling time of 10 h becomes larger, and there are many particles with the size above 50 μm, which are ascribed to the severe cold welding and agglomeration of small particles (Fig. 3(b)). With prolonging the milling time to 60 h (Fig. 3(c)), the size of CuZrAl powder particles exhibits an obvious refinement and reaches about 10 μm with a uniformly distribution, which may be due to their brittle and easy fracture characteristics. When the milling time is extended to 120 h, the particles are further reduced in size and present the flake shape, as shown in Fig. 3(d).

Fig. 4 presents the micrograph and the corresponding element mappings of Al20 powders after mechanical mixing of 10 h. According to the element mappings, it is observed that Cu and Zr elements exhibit a uniform dispersion state. Therefore, it indicates that CuZrAl particles uniformly distribute in Al matrix after the appropriate mixing time, which is beneficial to sintering quality during SPS process. XRD patterns of the SPS-ed pure Al bulk and ABCs reinforced with different volume fraction of CuZrAl are shown in Fig. 5. The inset is the photograph of SPS-ed Al30, presenting the typical metallic luster. The XRD pattern of the Al10 (Fig. 5(a)) shows fcc-Al phase along with $Al_3Zr$ ($D0_{23}$, a = b =

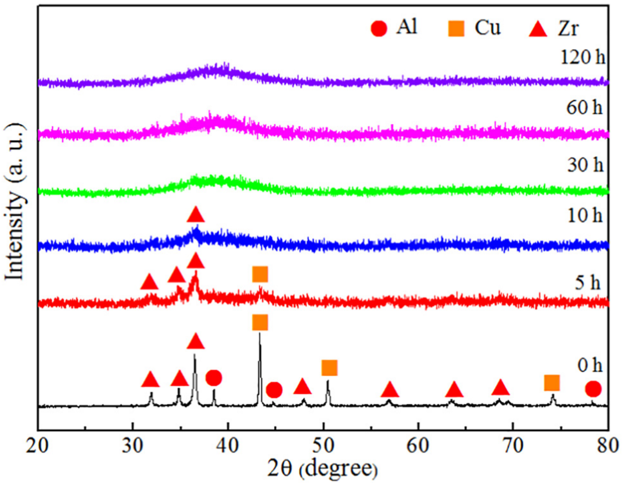

**Fig. 1.** XRD patterns of as-milled CuZrAl alloy powders after different milling time.

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