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AlN nanoparticle-reinforced nanocrystalline Al matrix composites: Fabrication and mechanical properties

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ABSTRACT

To improve the specific strength and stiffness of Al-based composites, AlN/Al nanoparticles were in-situ synthesized by arc plasma evaporation of Al in nitrogen atmosphere and consolidated by hot-pressing to fabricate AlN nanoparticle-reinforced nanocrystalline Al composites (0–39 vol.% AlN). Microstructure characterization shows that AlN nanoparticles homogeneously distribute in the matrix of Al nanocrystalline, which forms atomically bonded interfaces of AlN/Al. The hardness and the elastic modulus of the nanocomposite have been improved dramatically, up to 3.48 GPa and 142 GPa, respectively. Such improvement is believed to result from the grain refinement strengthening and the interface strengthening (load transfer) between the Al matrix and AlN nanoparticles.

1. Introduction

Due to the ultrafine grain size and high interface volume fraction, nanocrystalline (NC) materials with grain sizes typically smaller than 100 nm have unique mechanical properties [1], so the fabrication of NC materials has received considerable attention in recent years [1]. The properties of a lot of traditional materials, in particular metal matrix composites (MMCs), have been significantly improved by introducing nanophases [2–7]. A typical example is Al-based composites, whose hardness, strength, and Young's modulus can be effectively enhanced with addition of small amount of nanoparticles reinforcement [2–6]. Such improvements are attributed to the strengthening effects including dislocations, grain-boundaries (GBs), and sub-structures, etc. [3,4,8]. Based on the above understanding, it is believed that *nanocrystalline matrices* strengthened by *nanosized reinforcement* are expected to have much better microstructural stabilities and performance than NC metals [9,10] because of the concurrence of strengthening by both grain-boundary and nanoparticle reinforcements [11–13]. Koch et al. presented a pioneering review on this subject, revealing remarkable enhancement in hardness and strength [11]. They also pointed out that it is still unknown that whether those mechanisms related to the deformation and strengthening of conventional metal composites are still applicable for the nanocomposites with NC matrix [11]. Therefore, more systematic experimental studies are required.

An open challenge for the fabrication of nanocomposites is how to disperse reinforcing particles homogeneously since nanoparticles are very active and prone to agglomerate. In fact, such agglomeration often occurs in composites fabricated by the powder metallurgy when the reinforcement particles are far smaller than the matrix grains [4,14]. Although the mechanical alloying (or ball milling) is an effective fabrication method, oxide or nitride contaminants are often unavoidable [15], which is disadvantageous for researchers to explore the strengthening mechanism. So it is of great importance to develop effective fabricating techniques of NC composites.

In this study, NC Al composites reinforced by dispersed aluminum nitride (AlN) nanoparticles (0–39%) are fabricated with an in-situ synthesis of AlN/Al mixed nanoparticles, avoiding the mixing of two phases. AlN is selected as the reinforcement due to three reasons, (1) AlN has combined properties of high strength, high thermal conductivity, low coefficient of thermal expansion (CTE), and especially it does not react with the matrix [16]; (2) Al and AlN have a good interfacial adherence without interfacial reaction [17,18]; (3) AlN/Al composite is a promising candidate for electronic packaging materials with high specific strength and stiffness, high thermal conductivity and CTE matched with microelectronic devices [18–23]. In this report, the synthesis and detailed microstructure characterizations are presented, and the hardness and the elastic modulus of the nanocomposites were

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measured, together with a discussion of the strengthening mechanism.

2. Experimental details

The AlN/Al nanoparticles were synthesized in one step via arc discharge plasma evaporation and deposition of elemental aluminum in the N_2/H_2 mixed atmosphere (66.5 kPa). As the cathode target, the Al ingot (99.999% purity) was evaporated by the arc plasma of a constant arc current at 150 A and voltage at 20–30 V. The volume ratio of N_2 to H_2 in the mixed gas varied from 0.1 to 1 so that the phase composition of AlN/Al nanoparticles in the product can be controlled. After collection, the as-deposited AlN/Al nanoparticles were pressed to a cylindrical steel die (ϕ = 8 mm) at room temperature, followed by a hot-press with the following parameters: a heating rate of about 10 °C/min, sintering temperature at 450 °C, holding time for 1 h, applied load at 1 GPa, temperature for applying load at 400 °C and vacuum about 2.0×10^{-4} Pa. The dimension of the final products is 8 mm in diameter and 6–7 mm in height. The relative densities of all the hot-pressed samples are higher than 98% of the theoretical values.

The morphology and microstructure of AlN/Al mixed nanoparticles and as-prepared bulk nanocomposites were observed using transmission electron microscopy (TEM, JEOL-2010) at 200 kV. For TEM observations, thin specimen of each composite was mechanically ground and polished to a thickness of about 50 μm, and then the disc was thinned by dimpling and ion-beam milling (the ion-beam voltage was about 4 kV). Phase composition (weight fractions W) of the nanocomposite powders and bulk samples was characterized by X-ray diffraction (XRD, Regaku D/max-2400). The volume fraction V is calculated through the density ρ (AlN, 3.26 g/cm^3, Al, 2.7 g/cm^3) by the following equation,

$$\frac{W/\rho_{AlN}}{W/\rho_{AlN} + (1-W)/\rho_{Al}} \quad (1)$$

The oxygen content of the powders was measured using nitrogen/oxygen determinator (LECO TC-436). The micro-hardness of the AlN/Al composites was measured on a MVK-H3 testing machine with a load of 100 g and a loading time of 15 s. The data given are average values of eight testing points. All values of the elastic modulus were obtained on a Nanoindenter XP^T with a diamond Berkovich tip with a maximum applied load of 50 mN. The surfaces of samples were machine-polished using 0.5 μm paste before indentation tests. Four tests were carried out for each sample to obtain the average value. The elastic modulus was calculated from the load–displacement curves achieved from the indentation tests based on the method employed by Oliver and Pharr [24].

3. Results and discussion

3.1. Characterization of AlN/Al nanoparticles

The XRD pattern and phase composition of nanoparticles synthesized in different atmospheres are shown in Fig. 1. Only two phases are found in the XRD pattern (Fig. 1a), suggesting that no new phase or impurity was generated during the synthesis, except for a thin amorphous oxide film on the surface of Al particles detected in TEM observations. The oxygen content of the powders produced in the atmosphere of 100% H_2 and 100% N_2 are 3.1% and 3.4%, respectively. The mixed nanoparticles with different phase compositions can be obtained by controlling the partial pressure of N_2, and the amount of AlN increases monotonically with the increase of N_2 partial pressure (Fig. 1b). From the typical bright-field TEM micrograph (Fig. 2a), AlN/Al nanoparticles have two typical shapes: hexagonal and spherical. The faceted particles are AlN single crystals and a part of the spherical particles correspond to single-crystals Al nanoparticles, as shown in the selected-area electron diffraction patterns in Fig. 2b and c. Some spherical particles were observed to be the composite particles of AlN and Al. As shown in Fig. 2d and e, there exists an orientation relationship between AlN and Al: $(\bar{1}\ 3\ 1)_{Al} || (1\ 1\ 2)_{AlN}$. The formation of AlN/Al composite particles may follow such process that the Al ingot was firstly evaporated to gas phase and partially reacted with nitrogen to form AlN and Al clusters, and then nucleated and crystallized. At the low-temperature area, AlN firstly solidified for its much higher melting point than Al, and then parts of Al deposited on the surface of AlN particles by random collision. In addition, the above synthesis method has a yield of decades of gram per hour, which is sufficient to fabricate bulk samples for measurement of mechanical and physical properties. The average particle sizes of Al and AlN in the mixed nanoparticles with different volume fractions are shown in Table 1. The particle size of Al in the as-prepared nanoparticles reduces with the increase of the amount of AlN, suggesting that the second phase can constrain the growth of Al nanoparticles, which plays an important role on the grain refinement of the matrix, as discussed in the following part.

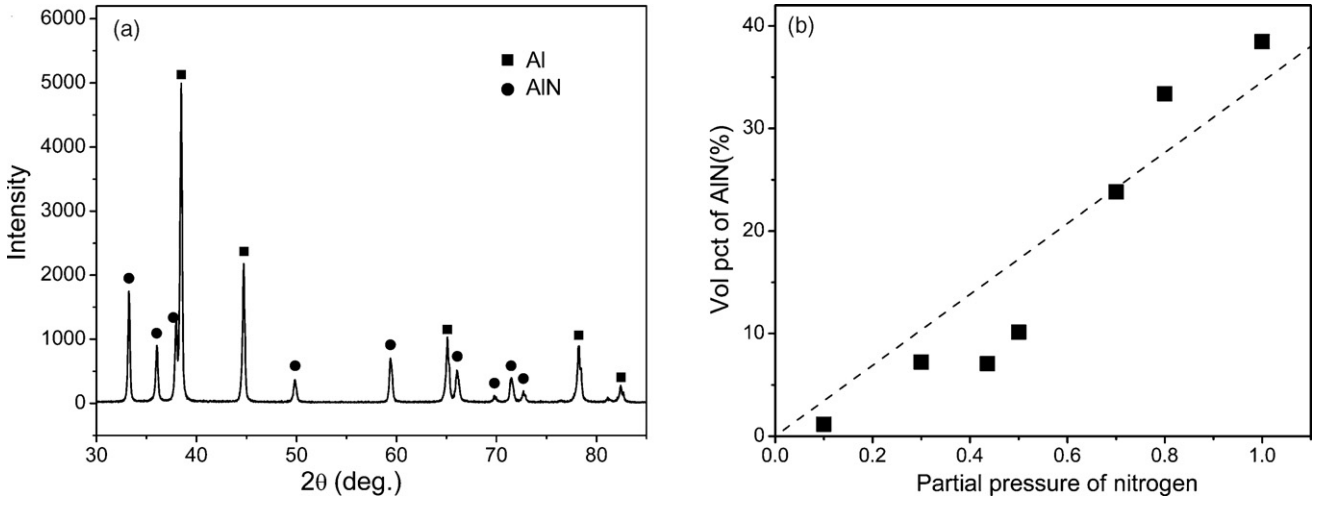

Fig. 1. (a) Typical XRD pattern of AlN/Al mixed nanoparticles, (b) variation of the volume fractions of AlN with the nitrogen partial pressure.