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Microstructure and synergistic-strengthening efficiency of CNTs-SiC_p dual-nano reinforcements in aluminum matrix composites

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ABSTRACT

In this study, reinforcements of carbon nanotubes (CNTs) and silicon carbide particle (SiC_p) in CNTs-SiC_p reinforced aluminum matrix nanocomposites (AMNCs) are studied. The tensile strength of 0.5CNTs-0.5SiC_p/Al increase by 94% compared with pure Al reaching 247 MPa, it also has a lifting of 14% and 56% compared with 1.0CNTs/Al and 1.0SiC_p/Al. CNT-SiC_p reinforcements have the synergistic enhancement effect of 1 + 1 > 2 in tensile strength. It is found that SiC_p as a dispersed particle around CNTs can inhibit and delay the pulling out and peeling of CNTs to further enhance the strengthening effect of CNTs by pinning effect. Between CNTs and Al matrix, there is a nano-scale reaction transition layer which improves the mechanical properties of AMNCs by strengthening the interfacial bonding. The existences of SiC_p inhibit the excessive reaction of the interface, when the tensile strength of AMNCs increases compared with pure Al, the elongation and conductivity have similar maintaining.

1. Introduction

Aluminum matrix composites (AMCs) are composed of Al matrix, and the second phase or the reinforcements by a certain fabrication process. AMCs present a lot of excellent properties, such as high strength, low density, low thermal expansion coefficient, good abrasion resistance, and high thermal conductivity [1–5]. Recently, silicon carbide particles (SiC_p) reinforced AMCs have gained a wide attention among the aerospace and automotive industry, because of its excellent wear resistance, and other comprehensive performance more than the common properties of normal AMCs [6–8]. However, due to the low toughness, the conventional AMCs are hard to present excellent comprehensive performance. Furthermore, low conductivity and lack of functionality also restrict the wide application of AMCs [9–11].

As a new self-assembled monolayer material, carbon nanotubes (CNTs) have been taken as an ideal reinforcement for composites since it has been discovered in 1991 by Iijima [12]. CNT presents some ultra-properties, such as ultrahigh Young's modulus (~1 TPa), ultrahigh strength (~100 GPa) and aspect ratio (up to 1000), and ultrahigh electric and thermal conductibility [13,14]. Due to its excellent mechanical and electrical properties, it has been highly expected that CNTs enhance the mechanical and electrical properties of metals and alloys as a role of potential reinforcement in the field of metal matrix composites [15]. As reported, Deng [16] found that the mechanical properties of 2024Al reinforced with 1.0 wt% CNTs improved observably, the ultimate tensile strength (UTS) reaches 521 MPa, which shows increase by 35.7% compare with 2024Al, at the same time it still maintain a 17.9% elongation rate. Yang [17] prepared CNTs reinforced aluminum matrix nanocomposites (AMNCs) by in-situ growth CNTs on the surface of Al matrix, the UTS of the AMNCs reinforced by CNTs is up to 398 MPa. However, there are many challenges in the study of CNTs reinforced AMNCs. As the small size results in the large Van der Waals force, CNTs tend to bring about agglomeration and lead to poor compatibility in metals. Moreover, it is difficult to control the interfacial reaction between CNTs and Al matrix, and the tendency of the ductility being inversely proportional to the mechanical properties of the composites [18,19]. Thus, there are three main directions in the future research: how to make CNTs uniformly dispersed in Al matrix, how to ensure the structure of CNTs not being damaged during the preparation process and how to determine the appropriate parameters to maximize the overall performance of AMNCs. Aiming at solving those problems belonging to CNTs reinforced AMNCs, our research will focus on the secondary reinforcement nano-SiC_p. As reported, nano-SiC particle is expected to promote

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the dispersion of CNTs as a ball milling medium and to improve the wettability between CNTs and Al [20]. It is also tended to reduce the contact between CNTs and Al matrix, and control the adverse reaction between CNTs and Al matrix. Furthermore, SiC_p can also be used as a reinforcing particle to play the role of dispersion strengthening and synergistically reinforced Al matrix with CNTs [21–23]. In this study, ultrasonic dispersion, planetary ball milling (PBM), spark plasma sintering (SPS) and hot extrusion were used to fabricate CNTs-SiC_p reinforced AMNCs, the processes have been proved to improve the dispersion effect of CNTs and sintering quality of AMNCs in our previous study [24]. The effects of CNTs-SiC_p content and ratio on densification process, ductility, strength and conductivity of composites will be discussed. We expect to explore feasible preparation method and provide experimental reference for the research and development of CNTs-SiC_p dual reinforced AMNCs.

2. Experimental process

2.1. Materials and methods

AMNCs reinforced with CNTs (Showa Denkon, Purity > 99 wt%, diameter ~ 100 nm, length 10 ~ 50 μm, aspect ratio 100 ~ 500, density 2.0 g/cm^3), nano SiC_p (Purity > 99.9 wt%, diameter ~ 30 nm) and CNTs-SiC_p were prepared at identical condition, respectively. CNTs or SiC_p reinforced AMNCs and pure Al were chosen as reference materials. As a necessary pre-experiment, thermodynamic characteristics of CNTs and Al were studied by DSC. The fabrication processes of CNTs/Al, SiC_p/Al and CNTs-SiC_p/Al mainly consist of three steps. The first step was to disperse reinforcements in Al powder (Purity > 99 wt%, diameter ~ 20 μm), in which ultrasonic dispersion (UD) and low speed planetary ball milling (PBM) process were conducted. The powder mixture of CNTs and SiC_p were dispersed by ultrasonic for 1 h in ethanol solution and then were dried. The total volume fraction of reinforcements in AMNCs were 0.5, 1.0, 1.5 vol%. Then, 60 g powder mixtures were mixed with 2 wt% ethanol which was used as process control agent (PCA), 240 g ZrO_2 milling balls at a diameter of 10 mm and 60 g ZrO_2 milling balls at a diameter of 5 mm were used as media. The PBM was carried out for a net milling time of 4 h at 200 rpm with argon gas protection. After that, the powder mixtures were filled into a graphite die (30 mm in diameter) in a glove box under the protection of argon gas. The second step was to consolidate the powder mixtures by SPS at 630 °C for 60 min, in the pressure 30 MPa applied on the sample under a vacuum of ~5 Pa. The third step was hot-extrusion. The as-sintered billet was finally sent to hot-extrusion in order to obtain a relatively high density, finer grain and CNTs well-dispersed materials [24,25]. Before hot extrusion, the sintered samples were preheated to 400 °C and hold for 5 min under an argon gas atmosphere. The billet was then immediately put into a steel container and extruded through a die with inner diameter of 7 mm. The extrusion ratio and the ram speed were 18:1 and 3 mm/s, respectively. Reference materials were also processed under the same route.

2.2. Characterization

To obtain the thermodynamic characteristics of CNTs and Al, samples were analyzed by a Differential Scanning Calorimetry (DSC, 200-F3, Netzsch, Germany). The electrical conductivity of AMNCs was measured by a portable eddy current tester (Sigma-2008B1, TIAN YAN, China). The mechanical properties, including yield strength, tensile strength and elongation, were obtained by a universal testing machine (HT-2402, HUANG HE, China). The morphologies of raw Al powder, raw CNTs, raw SiC powder, powder mixtures and fracture surface of AMNCs tensile samples were observed by a field emission scanning electronic microscope (FESEM, JSM−6700F, JEOL, Japan). The electron backscatter diffraction (EBSD) technique which was per-attached to the using TSL camera (TSL DigiView IV; EDAX) attached to the FE-SEM operating, 20 kV was used to analyze the crystal orientation containing grain size and phase information.

The high resolution and magnification micrographs of CNTs and CNTs-SiC_p/Al interface in AMNCs were observed by a transmission electronic microscope (TEM, JEM-2010, JEOL, Japan). The TEM samples were fabricated by using a focused ion beam (FIB) system (FB-2000S, HITACHI, Japan) with a sample thickness of ~100 nm. To confirm the phase structures in extruded AMNCs, X-ray diffraction (XRD-7000, SHIMADZU, Japan) was used. Cu Ka was used as the radiation source and the scanning speed was 8°/min. The crystallographic structure of raw CNTs and the CNTs in composites after processing were examined by a microscopic laser Raman spectrometer (ARAMIS, HORIBA, Japan).

3. Results and discussion

3.1. Morphologies of the powders and thermodynamic analysis

SEM micrographs of raw powder and powder mixture are showed in Fig. 1. The morphology of raw Al is nearly spherical shown in Fig. 1a. It is obtained that the diameter of CNT is about 100 nm and the aspect ratio is 100 ~ 500 from Fig. 1b. And the spherical tips of CNTs are also shown in Fig. 1b. Fig. 1c shows that the diameter of raw nano-SiC_p is about 30 nm. Fig. 1(d–f) show a mixture of CNTs-SiC_p powder after UD and PBM. It is shown that Al powder displays flake-shape after experiencing some plastic deformation. The morphology and the EDS results of powder mixture reveal that CNTs and SiC_p are dispersed uniformly on Al flake.

Interface is one of the most important aspects in the study of CNTs reinforced AMNCs [26,27], and thermodynamics is the key to the study of interfacial reaction. In Peng's and Gang's researches [28,29], the thermodynamics of Al matrix and CNTs are as follows:

$$4Al(s) + 3C(s) = Al_4C_3(s) \quad (1)$$

$$\Delta G_r^\theta = -28.93\ln T + 1.121 \times 10^2 T^2 + 1.155 \times 10^6 T^{-1} + 5.153 \times 10^{-6} T^3 + 131.33T - 222379.8 \quad (2)$$

$$4Al(s) + 3SiC(s) = Al_4C_3(s) + 3Si(s) \quad (3)$$

$$\Delta G_r^\theta = -2650 + 25.7767T \quad (4)$$

$$I = 1.5689 \times 10^6 \sqrt{texp\left(-\frac{246007.1}{RT}\right)} \quad (5)$$

where ΔG and T are Gibbs free energy change of the reaction and the temperature (K) respectively, I is the amount of the formation of Al_4C_3, t is sintering time, and R is gas constant (8.314). Thermodynamic Eqs. (1) and (2) reveal that Gibbs free energy change is less than zero when the temperature is between 25 and 660 °C which is near to the melting point of Al, it means that the reaction between graphite and Al can be activated at temperatures below the melting point of Al, and the interfacial reaction between CNTs and Al is difficult to be hindered. Thermodynamic Eq. (4) shows that Gibbs free energy of Eq. (3) is more than zero when the temperature is between 25 and 660 °C. Therefore, in the preparation of AMNCs by powder metallurgy, SiC_p can be stable in Al matrix, but CNTs will inevitably react with Al to generate Al_4C_3 brittle phase. SiC_p is