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In-situ observation of interaction between dislocations and carbon nanotubes in aluminum at elevated temperatures

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ABSTRACT

In this study, in-situ high-temperature transmission electronic microscopy is performed to investigate the formation mechanism of high-fraction (>50%) low-angle grain boundaries (LAGBs) in carbon nanotubes/aluminum (CNTs/Al) composites with balanced strength and ductility. During heating up the low-temperature-sintered composite, we find that dislocation bands first moves and then they are pinned by CNTs to form LAGBs. Abundant dislocations in the Al matrix and large-aspect-ratio CNTs are clarified as two critical factors for producing LAGBs. This study provides new insight into the effect of CNTs on the microstructural and mechanical properties of metal matrix composites.

1. Introduction

In the past decade, increasing attention has been paid to carbon nanotube (CNT)-reinforced aluminum (Al) metal matrix composites (MMCs) because of their promising light weight and excellent mechanical properties [1,2]. Many studies have shown the possibility to achieve high strength over 300 MPa of CNTs/Al composites by using powder metallurgy methods [2]. However, one critical problem of these composites for future applications is the limited ductility of near or less than 5% in tensile elongation [3,4]. To improve plasticity, some studies optimized composite microstructures related to reinforcements, such as CNT dispersion [5], interface [6] and Al_4C_3 amount [7]. However, limit attention has been paid to the matrix microstructure changes induced by CNT additions [1,8].

In metals and alloys, the misorientation angle of grain boundaries is a significant microstructural feature to influence the deformation behavior and mechanical properties [9,10]. Regarding Al alloys, some researches showed that low-angle grain boundaries (LAGBs) can improve the plasticity due to the cross slip mechanism between adjacent grains [11,12]. Recently, Chen et al. [3] reported a large fraction of LAGBs in CNTs/Al composites fabricated at high sintering temperature of 900 K, contributing to the excellent plasticity of tensile elongation larger than 20% as well as tensile strength over 200 MPa. The control of grain features in CNTs/Al composites may break new ground in fabricating high-performance Al MMCs. However, the formation mechanism of the high-fraction LAGBs was still unclear. To this end, in this study we applied in-situ transmission electronic microscopy observations on a low-temperature-sintered CNTs/Al composite to investigate the interaction between dislocations and CNTs at elevated temperatures.

2. Experimental methods

Pure Al powder and multi-walled CNTs (MWCNTs, ~10 nm in diameter and ~1 μm in length, Baytubes C150P, Bayer Material Science Co., Japan) were used as starting materials. To disperse CNTs in Al matrix, flaky Al powders were prepared by high energy balling and then bathed in isopropyl alcohol (IPA) based solution with 1 wt% CNTs in a plastic bottle on a rocking ball milling machine. To further consolidate the composite powders, spark plasma sintering (SPS) and following hot extrusion were applied. SPS (SPS-1030S, SPS Syntex, Japan) was conducted at two sintering temperatures of 800 K and 900 K. Pure Al powders were also processed under the same route as reference materials. Detailed description of the fabrication method can be found elsewhere [3,13].

Crystal grain information of each sample was analyzed using electron backscatter diffraction (EBSD) with a TSL camera (TSL DigiView IV) attached to the field emission scanning electron microscopy (FE-SEM, JEM-6500F, JEOL). The microstructures of CNTs/Al composites were examined by high resolution

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transmission electronic microscopy (HR-TEM, JEM-2010, JEOL, Japan). For in-situ TEM testing, the FIB-TEM sample was then welded on the stage of a semicircle copper plates using tungsten. A Gatan single-tilt (Model 628) heating holder with the copper disk was put inside the chamber of Hitachi H-800 type 200-kV transmission electron microscope. The heating was carried out under a high vacuum with a pressure of lower than 10^{-6} Pa. The CNTs/Al composite sample on the copper plate was slowly heated up to 700 K, 800 K, 850 K and 900 K at a heating rate of ~20 K·min^{-1} and then maintained at each temperature for 5 min. The temperature fluctuation was within ±2 K at 900 K. High-quality bright-field TEM images were captured during the holding stages.

3. Results and discussion

Fig. 1 shows the grain boundary (GB) characteristics obtained by EBSD of pure Al and CNTs/Al composites sintered at 800 K and 900 K. Obviously high-angle GBs (defined as GBs with misorientation angles larger than 15°) are dominant in pure Al sintered at 800 K and 900 K (Fig. 1a) and the CNTs/Al composite sintered at 800 K (Fig. 1b). However, with the CNTs/Al composite sintered at 900 K, the fraction of low-angle GBs (LAGBs, smaller than 15°, red and green lines in Fig. 1c) increases remarkably. On the other hand, the differences in average grain size (1.6–1.7 μm) and texture (⟨1 1 1⟩ dominant) are quite small in the two CNTs/Al composites [13] and thus they have small effect on the mechanical properties. Comparing the mechanical properties of the CNTs/Al composite sintered at 900 K to that of 800 K, there are simultaneous enhancement in tensile strength and ductility (Table 1). The strength improvement mechanism has been clarified as the enhanced bonding conditions of Al-CNTs interface at elevated sintering temperatures in the previous study [13]. The enhancement in interface will contribute to improved load transfer from matrix to CNTs, i.e., increased restriction of matrix deformation, leading to expected reduced ductility [14]. Opposite to prediction, there is remarkable improvement (over 80%) of elongation in the CNTs/Al composite sintered at 900 K compared to that of 800 K (Table 1). Since the effect of grain size and texture has been proved to be small, it is deduced that the ductility improvement is associate to the orientation angles of GBs.

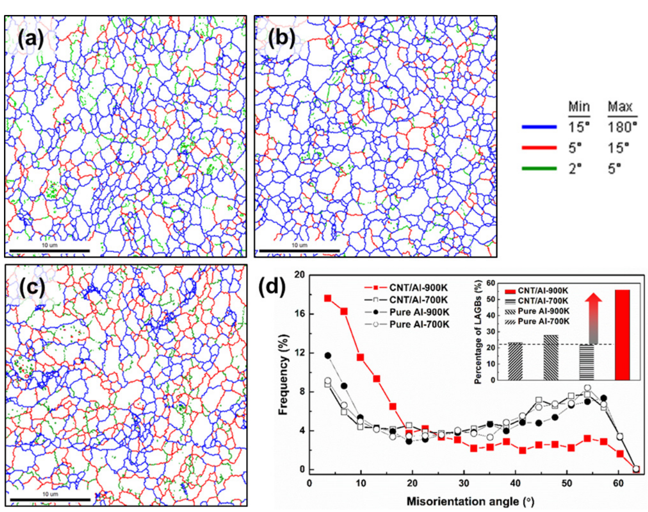

Fig. 1. Misorientation angle maps of grain boundaries in pure Al sintered at 900 K (a), CNTs/Al at 800 K (b), and CNTs/Al at 900 K (c). (d) is a summary of misorientation angle frequency. Inset of (d) shows the fraction of low-angle grain boundaries (LAGBs, <15°). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

The detailed information of misorientation angle is further examined by its frequency distribution in Al and CNTs/Al materials. It is observed that in the CNTs/Al composite sintered at 900 K, the frequencies of misorientation angles less than 20° increase noticeably; contrarily those of angles larger than 30° reduce much (Fig. 1d). As a result, the length fraction of LAGBs occupies 0.59 in CNTs/Al sintered at 900 K, while the value of 0.2–0.3 in the pure Al and CNTs/Al sintered at 800 K (inset of Fig. 1d, Table 1). Therefore, the sintering temperature plays a significant role in determining the GB features of CNTs/Al composites. It is also noted that the pure Al sintered at 900 K possesses a slightly increased value of 0.30 compared with that of pure Al sintered at 800 K, which is associate to the in-situ formed alumina nanoparticles [15].

The grain and dislocation features are examined by TEM in the CNTs/Al composites. In the composite sintered at 800 K, large quantity of dislocations as well as dislocation bands are observed (Fig. S1a). The formation of a high density of dislocations is attributed to the stored strains in the prepared flaky Al powders that experienced severe plastic deformation in the high-energy ball milling process [16]. With the CNTs/Al composite sintered at 900 K, except for a high dislocation density, the entangled dislocations and the LAGB are also detected (Fig. S1b).

To understand the formation mechanism of LAGBs, in-situ high-temperature TEM study is performed on the TEM sample of the CNTs/Al composite sintered at 800 K (Fig. S1a). Well-dispersed and unreacted CNTs are observed in Al matrix at the initial state before heating-up (inset of Fig. 2a). There is no obvious microstructure change at temperature of 700 K (Fig. 2b) and 800 K (Fig. 2c). As the temperature is increased to 850 K, it is seen that a dislocation band (as black arrows indicated in Fig. 2c) moves ahead to the position as a red arrow indicated in Fig. 2d. It is seen that the dislocation is pinned by CNTs, as a white arrow indicated in Fig. 2d. The grey contracts surrounding CNTs should be a result of strain at Al-CNT interface because of the large difference of the thermal expansion coefficients between Al (~10^{-6} K^{-1}) and CNTs (23.6×10^{-6} K^{-1}) [17]. Even at higher temperature of 900 K, the dislocation no long moves forward and it is steady at the place contacting with CNTs (Fig. 2e). After cooling down, a LAGB with misorientaiton angle of ~4° forms as revealed in Fig. S2.

Fig. 3 shows the formation scheme of the LAGBs based on the in-situ study and microstructural observations. With Al, as the process temperature is increased, the dislocation immigration is activated under the energy input. As a result, many dislocations prefer to sink at GBs, which corresponds to the commonly observed recovery process of metals at high annealing temperatures [18]. However, in CNTs/Al composites, because of the existence of long

Table 1
Tensile properties and grain features in Al and CNTs/Al composites consolidated at different temperatures.

Sample	Yield strength (MPa)	Tensile strength (MPa)	Elongation (%)	Grain size (μm)	Texture	Length fraction of LAGBs
Al-SPS800K	122	172	21.1	1.92	⟨1 1 1⟩	0.21
Al-SPS900K	121	166	24.1	2.34	⟨1 1 1⟩	0.31
CNTs/Al-SPS800K	127	186	11.2	1.67	⟨1 1 1⟩	0.20
CNTs/Al-SPS900K	150	212	20.4	1.75	⟨1 1 1⟩	0.59

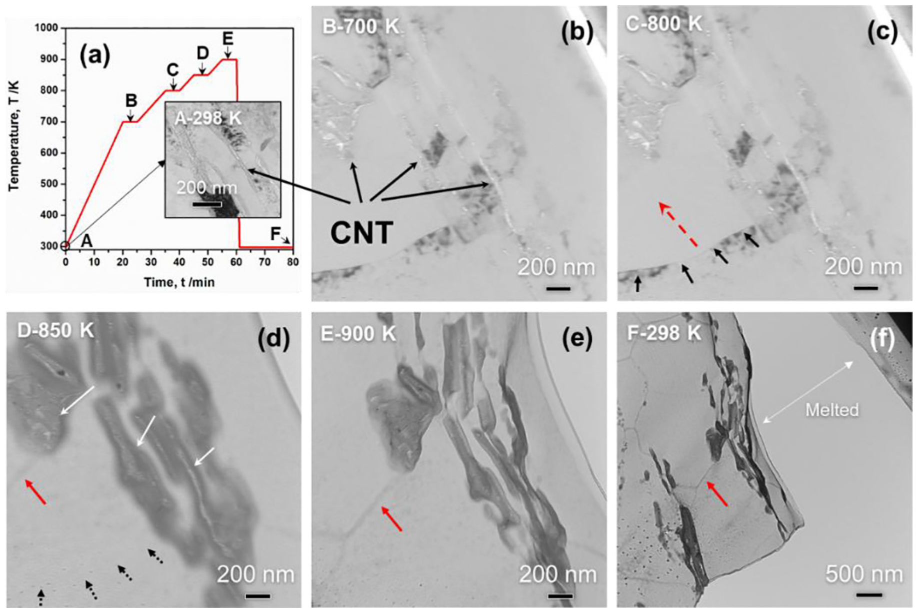

Fig. 2. In-situ TEM observation on CNTs/Al composite at different temperatures. (a) Target temperature-time curve. (b-f) Recorded TEM images during the stages in (a) as marked by corresponding captial letters.

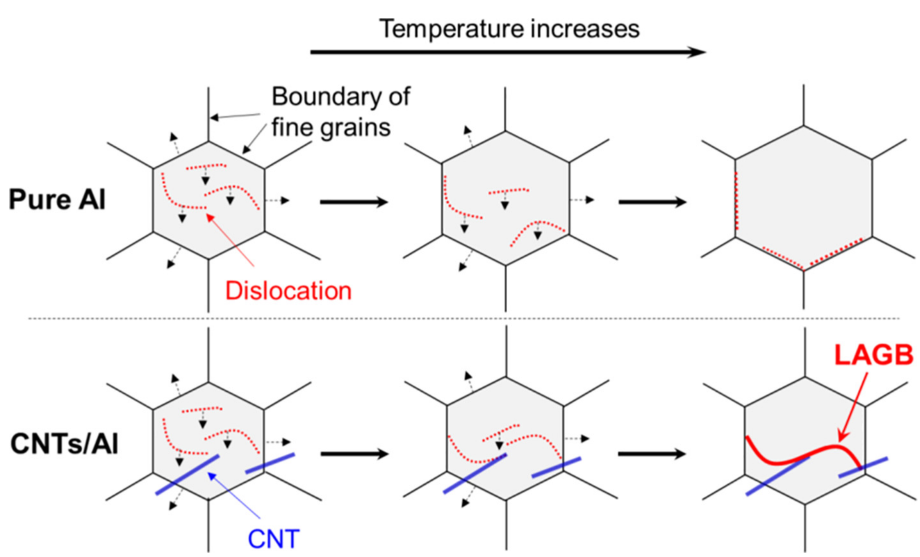

Fig. 3. Formation scheme of a CNT-induced low angle grain boundary in Al MMCs with abundant dislocations at elevated temperatures.

CNTs, the movement of dislocations is restricted as they encounter the CNTs. The dislocations finally settle down (Fig. 3). As more CNTs or CNT bands stop there, LAGBs will form in the area adjoining to CNTs. From the viewpoint of the whole sample, large fraction of LAGBs are thus observed (Fig. 1d).

4. Conclusions

This study investigates the formation mechanism of high fraction LAGBs at elevated temperatures in Al MMCs. From in-situ high-temperature TEM observations on CNTs/Al composite sintered at 800 K, the abundant CNTs can be trapped by long CNTs and evolve to LAGBs. The abundant dislocations and long CNTs are internal factors and high temperature is the external one to produce high fraction LAGBs and consequently outstanding tensile properties. Our study provides new insight into designing high performance MMCs reinforced with large-aspect ratio CNTs or graphene.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.matlet.2020.127323.

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