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Lateral size effect of graphene on mechanical properties of aluminum matrix nanolaminated composites

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ABSTRACT

A lateral size effect of reinforcement on mechanical properties is investigated for aluminum matrix nanolaminated composites reinforced with reduced graphene oxide (rGO). rGO sheets with remarkably different lateral size are chosen to cover the critical value estimated by the shear lag model. Tensile tests with loading along the in-plane direction of the nanolaminated structure demonstrate that lateral size has significant influence on mechanical properties. A good balance between strength and ductility is realized by the largest lateral size of rGO.

Graphene has attracted substantial attention for its superior intrinsic mechanical and functional properties [1] in the field of metal matrix composites (MMCs) [2–8]. As compared with traditional hard and brittle ceramic reinforcements in MMCs, graphene is strong but flexible. Moreover, graphene is characterized with an atomic-scale thickness, comparable and even smaller than most precipitates in alloys. These features endue graphene reinforced MMCs with excellent machinability. Additionally, the high aspect ratio of graphene and thus large interfacial contact area with metal matrix lead to high strengthening efficiency in MMCs.

In the earlier time, the major research effort has concentrated upon uniformly dispersing graphene in metal matrix and improving interfacial bonding strength [9,10]. Recently, architecture design [11,12], i.e. adjusting the distribution and orientation of reinforcements without changing in constituents, is being widely accepted as a strategy in enhancing strengthening efficiency or balancing strength and ductility/toughness in composites. In the reported MMCs, while high structural integrity of graphene is always pursued, the lateral size effect of graphene on mechanical properties of MMCs has remained unexplored. The reinforcements of graphene or graphene oxide (GO) are typically obtained by exfoliating graphite or graphite oxide through mechanical shear or ultrasonic, and their lateral sizes are about a few micrometers and are used as prepared in MMCs.

As reported in polymer matrix composites, the mechanical behavior of a two-dimensional (2-D) reinforcement (i.e. ceramic platelet or graphene) can be analyzed by the shear-lag theory [13–15]. It is assumed that there is elastic stress transfer from the matrix to the 2-D reinforcements through a shear stress at the composite interface, and the strain builds up from the edge to the center of the 2-D reinforcements [16]. Therefore, mechanical properties are significantly influenced by the lateral size of 2-D reinforcements. For example, in graphene/polyvinyl alcohol (PVA) composites, it is demonstrated that polymer matrix can be effectively reinforced with graphene provided the flakes have large enough aspect ratio and composite modulus scales with flake size [15].

Herein, the lateral size effect of graphene on the mechanical properties of aluminum matrix composites is investigated by using GO nanosheets with three kinds of lateral size as reinforcement. To ensure the load applied along the in-plane direction of GO nanosheets in composite, a nanolaminated structure is prepared by a modified powder metallurgy [6]. The tensile tests show that the largest lateral size of GO leads to a good balance of strength and ductility in composite, attributing to the synergetic contribution from high strengthening efficiency of graphene with large lateral size and ductile regions of unreinforced Al matrix.

All kinds of GO nanosheet investigated were exfoliated from graphite oxide. The GO with sub-micron lateral size (denoted as S-GO) was produced by exfoliating graphite oxide (used as received from Nanjing XFNano Material Tech Co. Ltd., China) by a high-intensity ultrasonic processing (2 h under a power of 720 W). The GO with larger lateral sizes were prepared by a modified Hummers' method [17], in which large graphite flakes with lateral dimension of 80–180 μm were used as starting material, and milder conditions were applied for oxidization and exfoliation to avoid excessive fragmenting. For preparing GO with

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a middle lateral size (denoted as M-GO), graphite was thoroughly oxidized for a long time of 16 h in ice bath. The as-prepared graphite oxide was then exfoliated by an ultrasonic processing with a lower power (2 h under a power of 360 W) to produce M-GO. To further increase the lateral size of GO, the time for oxidizing graphite was shortened from 16 h to 1.5 h in ice bath. Subsequently, the mixture was heated up to 35 °C and kept stirring for 2 h to exfoliate graphite oxide without using ultrasonic, during which oxidation was continued meanwhile. After centrifugation and washing repeatedly, the GO with a lateral size over 10 μ (denoted as L-GO) was obtained. More details refer to the Supplementary information.

Spherical Al powders (99.99% purity) were ball-milled in ethanol at 352 rpm for 4 h and dried in vacuum to obtain Al flakes. As-obtained Al flakes were stirred with as-prepared GO suspension in ethanol for 1 h and dried in vacuum to get GO/Al composite flakes. After thermal reduction of GO/Al composite flakes at 500 °C for 2 h, uniaxial compaction under 500 MPa pressure and vacuum sintering at 540 °C for 2 h were carried out to fabricate bulk reduced GO/Al (rGO/Al) nanolaminated composites. Completely densified composites were produced by hot rolling at 400 °C by 50% reduction in thickness. According to the lateral sizes of GO nanosheets used (S-GO, M-GO and L-GO), the final rGO/Al composites are designated as S-rGO/Al, M-rGO/Al and L-rGO/Al, respectively. Unreinforced Al matrix was also fabricated by identical processes but without incorporating graphene.

A simple shear lag model has successfully applied to describe mechanical strength of nanolaminated composites [13,18]. The operative failure mode of a 2-D reinforcement in composites is demarcated by a critical aspect ratio (s_c). s_c is equal to the ratio σ_p/τ_y, where σ_p is the tensile stress of reinforcement and τ_y is the shear strength of matrix. The composite fails under the reinforcement pull-out mode for actual aspect ratio (s) smaller than the critical value ($s < s_c$), while under the fracture mode in the case of $s > s_c$ (Fig. 1a). To assess the graphene lateral size effect on mechanical properties of rGO/Al nanolaminated composites, s_c was estimated from mechanical properties of Al matrix and the rGO used. The I_D/I_G value in the Raman data of as-prepared GO nanosheets was measured to be ~0.8 (Supporting information, Fig. S1), and therefore the average tensile strength can be evaluate to be ~30 GPa, according to the relationship established by Zandiatashbar et al. [19]. The shear strength of pure Al is <100 MPa [20]. Therefore, s_c of the GO used was predicted to be ~300. However, the s_c could be larger because of the decrease of defect concentration and thickness of graphene with removing oxygen by thermal reduction [16].

Fig. 1b shows atomic force microscopy (AFM) images of the as-prepared GO nanosheets. The superimposed height profiles along the white line (Fig. 1b) show that the thicknesses of the GO nanosheets were all around 1 nm, consistent with previously reported graphene oxide monolayer [21]. AFM images show that the average lateral sizes of as-prepared S-GO, M-GO and L-GO are about 0.23 μm, 1.1 μm and 15.4 μm, respectively (Supporting information, Fig. S2). The range of obtained lateral size covers the estimated critical aspect ratio.

Spherical Al powders (Fig. 2a) were transformed to Al flakes (Fig. 2b) by ball-milling and then used as building blocks for fabricating the nanolaminated composites (Fig. 2d). The as-obtained Al flakes (Fig. 2b) have a large aspect ratio with a lateral dimension of 60–80 μm and a thickness of 700–800 nm, assuring their alignment during compacting. Fig. 2c shows scanning electron microscopy (SEM) images of the surface of Al flakes after GO absorbing and thermal reduction. The dark parts are the surface covered by rGO sheets, and the coverages are similar to each other and around 60% in all three samples. According to the coverage of rGO and the thickness of Al flakes and rGO nanosheets, the fraction of graphene in the final composites is estimated to be ~0.2 vol%. The lateral sizes of rGO sheets in Fig. 2c are identical to that in Fig. 1b, indicating that GO sheets are fully spread and uniformly dispersed on Al flakes. Next, the rGO/Al flakes self-assembled into laminated green compact under a uniaxial pressure, and final bulk composites were obtained by sintering and rolling. As shown in transmission electron microscopy (TEM) image (Fig. 2e), the microstructure of the composite is characterized by elongated Al grains, forming a laminated structure where each lamella contains predominantly a single grain through its thickness. High-resolution TEM (HR-TEM) image (Fig. 2f)

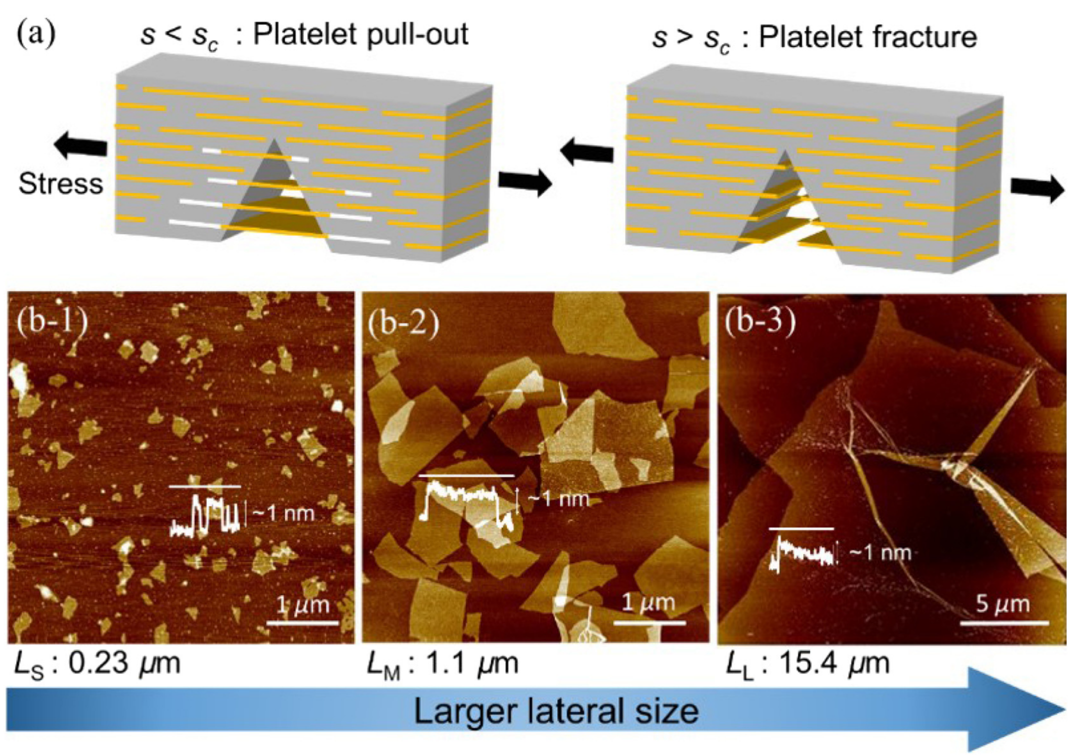

Fig. 1. (a) Classical fracture mechanisms of a platelet reinforced laminated composites. Failure mode depends on the relative aspect ratio of platelets (s) compared with an intrinsic critical value (s_c). (b) AFM images of GO sheets with average lateral sizes of about (b-1) 0.23 μm (S-GO), (b-2) 1.1 μm (M-GO) and (b-3) 15.4 μm (L-GO), respectively. The superimposed height profiles along the white line indicate a sheet thickness of about 1 nm.