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Invited Review

Deformation treatment and microstructure of graphene-reinforced metal matrix nanocomposites: A review of graphene post-dispersion

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Abstract: Graphene/aluminum (Gr/Al) composites have attracted the attention of researchers all over the world due to their excellent properties. However, graphene agglomerates easily because of the van der Waals force between graphite sheets, thereby affecting the performance of the composites. Decreasing the agglomeration of graphene and dispersing it uniformly in the Al matrix is a key challenge. In the preparation process, predispersion treatment and deformation treatment can play important roles in graphene dispersion. Researchers have conducted a series of research and literature reviews of the graphene predispersion and consolidation of composites. However, they paid less attention to post-deformation processing. This review summarizes different deformation treatments involved in the preparation process of Gr/Al composites and the evolution of the microstructure during the process. Research on deformation parameters is expected to further improve the properties of Gr/Al composites and would provide a deep understanding of the strengthening effect of graphene.

Keywords: graphene/aluminum composites; deformation treatment; dispersion; microstructure

1. Introduction

Aluminum and aluminum alloy materials have been widely used in the aerospace, aviation, automotive, and electronics industries because of their advantages, such as low cost, low density, high specific strength, excellent ductility, and good mechanical properties [1–3]. However, alloy materials can no longer adapt to the needs of the rapid development of modern industrial technology. Therefore, aluminum-based composite materials have emerged to satisfy the increased demand for material strength, stiffness, and wear resistance [4]. Traditional aluminum-based composite materials mainly utilize SiC, Al_2O_3, and carbon fiber as reinforcement. Research on these composite materials is relatively mature and the preparation process is relatively complete. As a result, the performance of traditional aluminum-based composites currently approaches the limit. Therefore, the discovery of new types of reinforcements has become a critical issue at this stage, and it is also the future development trend of aluminum-based composite materials.

In 2004, Novoselov *et al.* [5] utilized a mechanical stripping method to prepare a stable two-dimensional single-layer carbon material called graphene. This material is currently one of the thinnest and hardest materials in the world, and it has extraordinary mechanical properties [6] (a Young's modulus of 1.1 TPa and breaking strength of 125 GPa), good thermal properties [7] (a thermal conductivity of about 5000 $W\cdot m^{-1}\cdot K^{-1}$ and an extremely low thermal expansion coefficient), and excellent electrical properties [8] (electron mobility of 1.5×10^5 $cm^2\cdot V^{-1}\cdot s^{-1}$). These positive characteristics make graphene the most ideal reinforcement for composites, attracting the attention of many researchers.

At present, a large number of studies have been conducted on graphene/polymer composites and graphene/ceramic

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composites, but relatively less research has been performed on graphene/metal composites. Research on graphene/aluminium (Gr/Al) composites was gradually initiated in 2011 only [9–12]. Graphene has a large specific surface area, and it agglomerates easily due to the van der Waals force between sheets during the preparation process [13]. According to the literature, the modified shear lag model can describe the enhancement effect of graphene well, and the enhanced efficiency R of the composite material can be derived from Eq. (1) below:

$$R = \frac{S}{V}\frac{l}{4} - 1 \tag{1}$$

where (S/V) is the specific area of graphene nanoplates (GNPs) and l is the size of GNPs. The agglomeration of graphene results in a decrease in specific surface area, which would directly affect its reinforcing effect. The agglomerated graphene would act as a crack source, causing the material to break early under load. Therefore, the uniform dispersion of graphene is an important issue that needs to be addressed in the preparation of composites [14–16].

The preparation process of Gr/Al composites mainly includes predispersion, consolidation, and deformation treatment. Predispersion treatment and deformation treatment can play important roles in graphene dispersion. At present, research on dispersion is in the early stage, focusing mainly on powder dispersion, and good research progress has been made, as shown in Fig. 1. However, graphene dispersion still needs to be further improved. Therefore, researchers introduced deformation treatment into the manufacturing process of Gr/Al composites and eliminated graphene agglomeration through the shearing effect of the deformation treatment. This method is a new approach for the manufacturing process of Gr/Al composites. Researchers have already made some related applications, but this method still needs to be studied further, which is gradually being recognized.

The previous literature review mainly reported on the predispersion and consolidation process of graphene–metal matrix (including aluminum) composites and mainly focused on the enhancement in mechanical properties of materials [17–21]. By contrast, the present review mainly focuses on the post-treatment (deformation treatment) of graphene aluminum composite and the microstructure evolution of the material, which was previously not reported systematically.

The deformation treatment and microstructure of graphene-reinforced nanocomposites are discussed in the following sections.

2. Deformation treatment and strengthening mechanism

A variety of deformation treatments are used in the preparation of Gr/Al and carbon nanotubes/Al (CNTs/Al) composites, including hot extrusion, rolling, friction stir processing (FSP), high-pressure torsion (HPT), equal channel angular process (ECAP) and drawing, as shown in Fig. 2. During the deformation treatment, the shear deformation of matrix plastic flow can open the agglomeration of GNPs. Moreover, severe plastic deformation may cause the GNPs to bend and twist, and it may even cause separation between GNP layers, thereby reducing the thickness of GNPs. The opening of graphene agglomeration and the reduction of GNP thickness are conducive to performance improvement according to the shear lag model.

2.1. Hot extrusion

Hot extrusion is the most commonly utilized deformation treatment process [22]. During the hot extrusion process, the material is subjected to a three-way stress constraint state so that the plasticity is fully exerted and the formability of the material can be better guaranteed [23].

Zhou *et al.* [24] used graphene oxide (GO) as a raw material. They also mixed reinforcement with aluminum powder by using stirring and ultrasonic dispersion and then prepared Gr/Al composite by using the spark plasma sintering (SPS) process. Subsequently, the composite was hot-extruded at an extrusion temperature of 773 K with an extrusion ratio of 20:1. The GO has many functional groups on the surface, which is easier to disperse than graphene. More importantly, few-layered reduced graphene oxide (RGO) can be obtained by simple thermal reduction. An alumina–graphene–alumina sandwich structure was observed in the SPS composite because of the presence of alumina. After extrusion, the deformation of the aluminum matrix damaged the structure of graphene and caused the graphene to rearrange along the direction of the plastic flow, as shown in Fig. 3. At the same time, the plastic flow broke the alumina layer, which caused direct contact between GO and aluminum. Researchers pointed out that graphene brought a significant strengthening effect through the load transfer mechanism.

Yang *et al.* [25] reported a liquid synthesis method that mixed graphene with aluminum powder by ball milling. They then prepared a Gr/Al composite by using the pressure infiltration process and performed hot extrusion at 450°C at an extrusion ratio of 11:1. The grain size of the Al matrix was greatly refined, and significant orientation of the Al grains was found in the material after extrusion. The content of the dimples and tearing ridges in the fracture of the extruded composite material increased significantly. Meanwhile, the phenomenon of graphene pull-out and bridging was observed. The addition of 0.54wt% graphene increased the yield strength of the composite by 116%; after extrusion, the yield strength increased by 228%.

One-dimensional or two-dimensional reinforcements were aligned by extrusion, and the interface between reinforcement and matrix was improved so that the reinforcing effect was fully exerted. This finding was also observed in other systems [26–28].

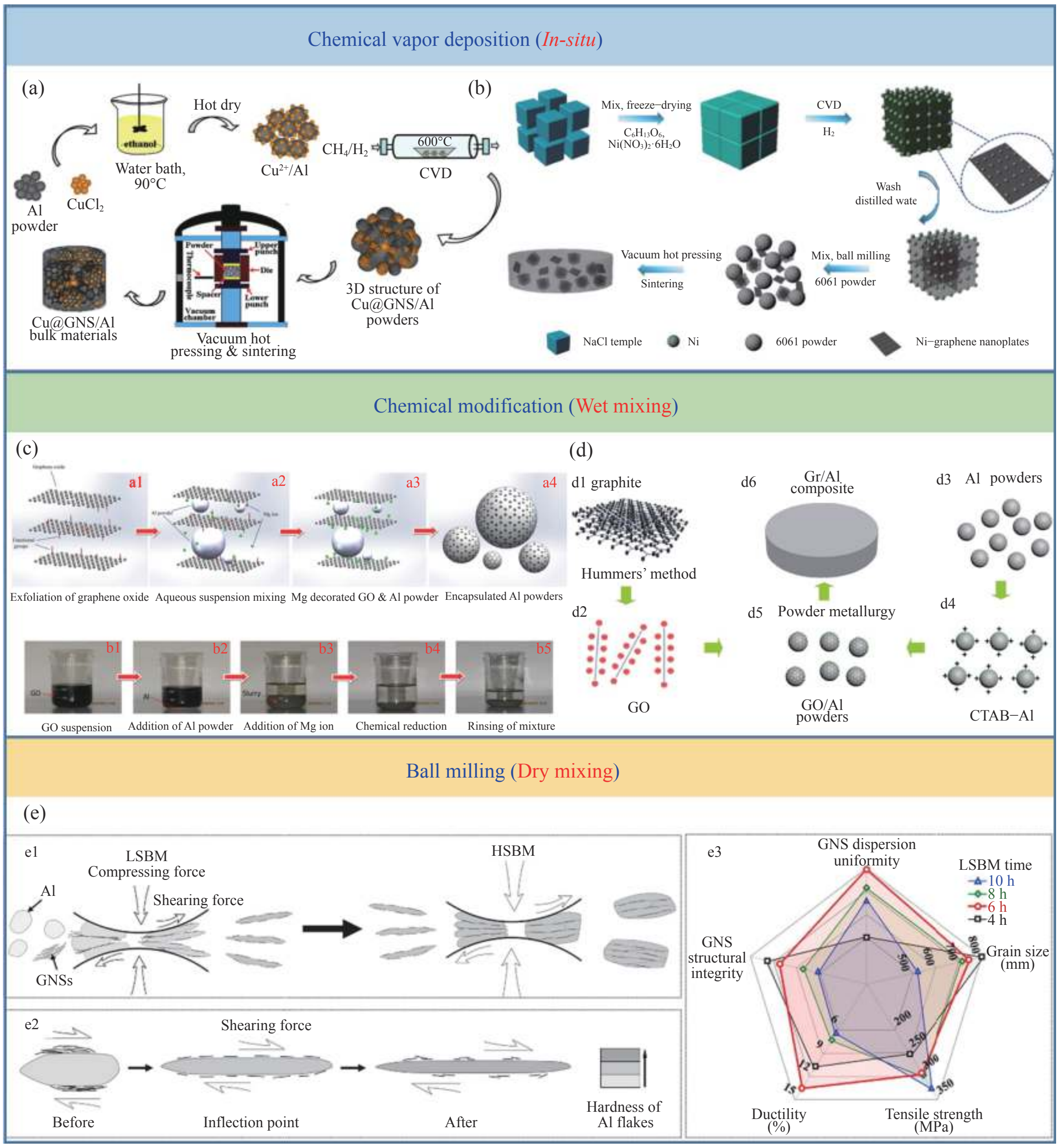

Fig. 1. Examples of graphene raw materials used in the literature and typical predispersion methods: (a, b) the process of graphene dispersion by chemical vapor deposition (CVD); (c, d) the process of graphene dispersion by wet mixing, the raw material is graphene oxide (GO), a1–a4 is the schematic diagram of chemical modification, while b1–b5 is the image of actual operation in (c), and the chemical modifier is ctabcetyltriethylammnonium bromide (CTAB) in (d); (e) the process of graphene dispersion by dry mixing (LSBM: low speed ball milling; HSBM: high speed ball milling), and the raw material is graphene nanosheets (GNSs). (a) Reprinted from *Mater. Sci. Eng. A*, 709, X.H. Liu, J.J. Li, J.W. Sha, E.Z. Liu, Q.Y. Li, C.N. He, C.S. Shi, and N.Q. Zhao, *In-situ* synthesis of graphene nanosheets coated copper for preparing reinforced aluminum matrix composites, 65-71, Copyright 2018, with permission from Elsevier. (b) Reprinted from *Mater. Sci. Eng. A*, 699, G. Liu, N.Q. Zhao, C.S. Shi, E.Z. Liu, F. He, L.Y. Ma, Q.Y. Li, J.J. Li, and C.N. He, *In-situ* synthesis of graphene decorated with nickel nanoparticles for fabricating reinforced 6061Al matrix composites, 185-193, Copyright 2017, with permission from Elsevier. (c) Reprinted from *Mater. Des.*, 94, X. Gao, H.Y. Yue, E.J. Guo, H. Zhang, X.Y. Lin, L.H. Yao, and B. Wang, Preparation and tensile properties of homogeneously dispersed graphene reinforced aluminum matrix composites, 54-60, Copyright 2016, with permission from Elsevier. (d) Reprinted from *J. Alloys Compd.*, 704, J.M. Ju, G.F. Wang, and K.H. Sim, Facile synthesis of graphene reinforced Al matrix composites with improved dispersion of graphene and enhanced mechanical properties, 585-592, Copyright 2017, with permission from Elsevier. (e) Reprinted from *Composites*, *Part A*, 111, Y.Y. Jiang, Z.Q. Tan, R. Xu, G.L. Fan, D.B. Xiong, Q. Guo, Y.S. Su, Z.Q. Li, and Di Zhang, Tailoring the structure and mechanical properties of graphene nanosheet/aluminum composites by flake powder metallurgy via shift-speed ball milling, 73-82, Copyright 2018, with permission from Elsevier.

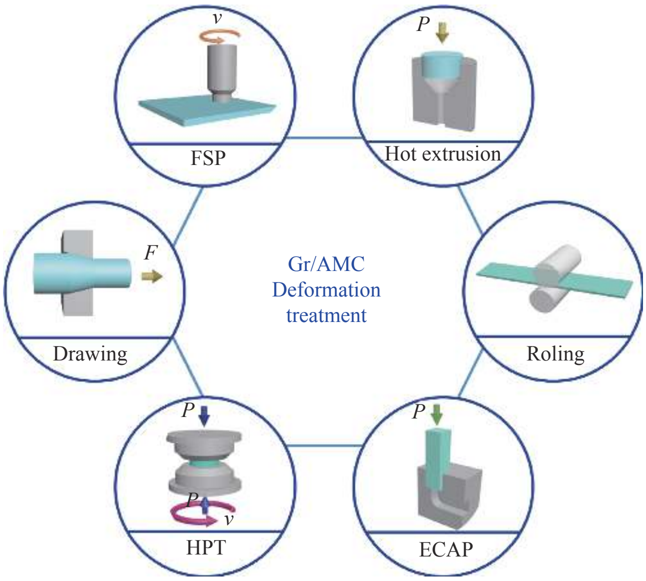

Fig. 2. Deformation treatment used in preparation of graphene reinforced aluminum matrix composites (Gr/AMC). *F* is the drawing force; *P* is the pressure; *v* is the linear speed of rotation.

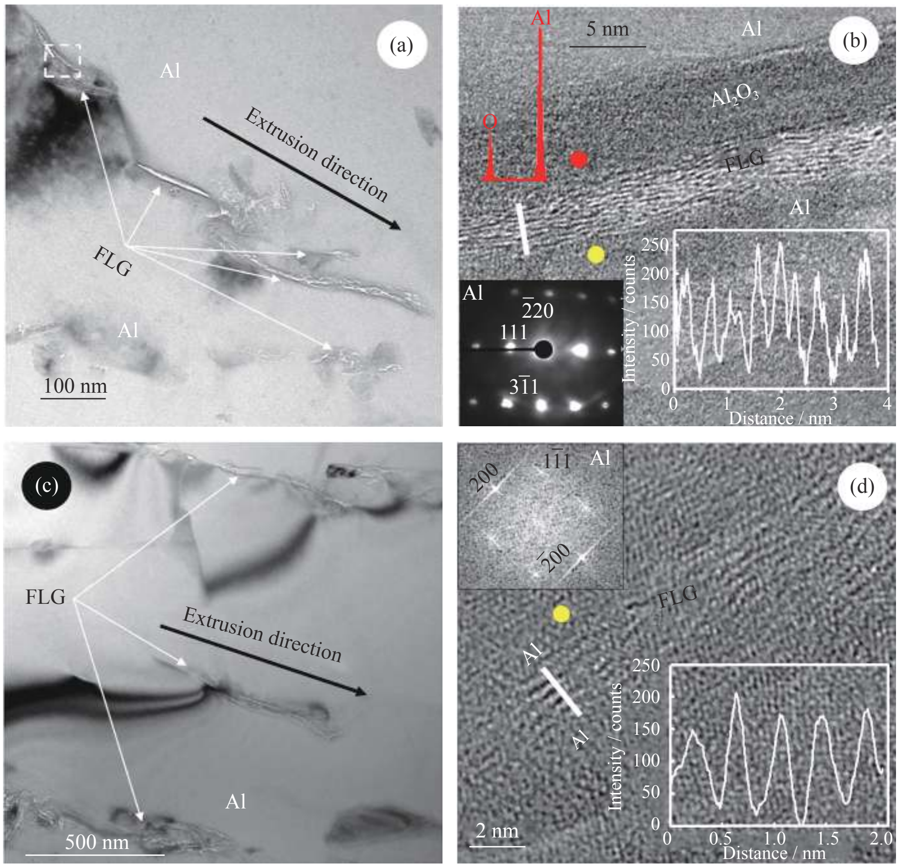

Fig. 3. Transmission electron microscope (TEM) (a, c) and high resolution transmission electron microscope (HRTEM) (b, d) images of the hot-extruded 0.4vol% few-layer graphene/Al (FLG/Al) composite. (b) is taken from the position of the white square in (a). Insets in (b) show the EDS analysis of Al_2O_3-layer taken from the red spot, the SAED pattern of Al matrix taken from the yellow spot and the profile plot along the white line. Insets in (d) show the fast Fourier transform pattern of the Al matrix taken from the yellow spot and the profile plot along the white line. Reprinted from *Composites, Part A*, 112, W.W. Zhou, Y.C. Fan, X.P. Feng, K. Kikuchi, N. Nomura, and A. Kawasaki, Creation of individual few-layer graphene incorporated in an aluminum matrix, 168-177, Copyright 2018, with permission from Elsevier.

2.2. Rolling

Rolling is another common method used in deformation treatment [29]. Shin *et al.* [13] first used a planetary mill to mix exfoliated graphene and aluminum powder at a low speed (100 r/min) and then shifted to a high speed (500 r/min) to promote further dispersion of graphene. Finally, the mixed powder was packed into a copper tube, compacted, and hot-rolled to a thickness of 1 mm on a plate at a rolling reduction of 12% per pass. When the composite was loaded, the few-layered graphene (FLG) between the aluminum grains limited the plastic deformation of the grains, and local deformation bands were generated by shear force. The restriction also led to a decrease in the plastic strain-to-failure of the composite. The mechanical properties of the FLG/Al composite also improved greatly due to the good dispersion of graphene. The addition of only 0.7vol% of graphene caused the tensile strength of the composite to reach 440 MPa, which is 71.8% higher than that of the matrix, as shown in Fig. 4. The differences between GNPs and multi-walled carbon nanotubes (MWCNTs) were explained through comparative experiments and theoretical model analysis. Although GNPs and MWCNTs have similar molecular structures, FLG has a larger contact area to transfer stress between the reinforcement and the matrix compared with MWCNTs. The increase in specific surface area is the major factor in improving the mechanical performance of the composite.

Li *et al.* [30] fabricated reduced graphene oxide/Al (RGO/Al) composite with a bioinspired nanolaminated structure [31–32]. Flake Al powders were prepared by ball milling and mixed with a GO suspension. Al powders and GO were combined by adsorption, then filtered and dried to obtain a GO/Al mixed powder. The powders were thermally reduced in a tube furnace under H_2/Ar flow to obtain RGO/Al powders, which were then made into a composite by vacuum hot pressing and sintering. The sintered composite was rolled 14 times at 350°C, and the rolling reduction of each pass was 5%. Finally, a composite with a lamellar structure was obtained, as shown in Fig. 5. The performance of the composite improved significantly, and the elongation was better than the matrix. Researchers point out that the advantage of the layered structure is that the reinforcement RGO is oriented in the stretching direction and can form a good interface bond to fully exert the capacity of loading transfer. The design of bioinspired structure provides new ideas for the preparation of Gr/Al composites.

2.3. Friction stir processing

FSP [33] is a solid-state metal working technique derived from friction stir welding. The process is similar to fric-

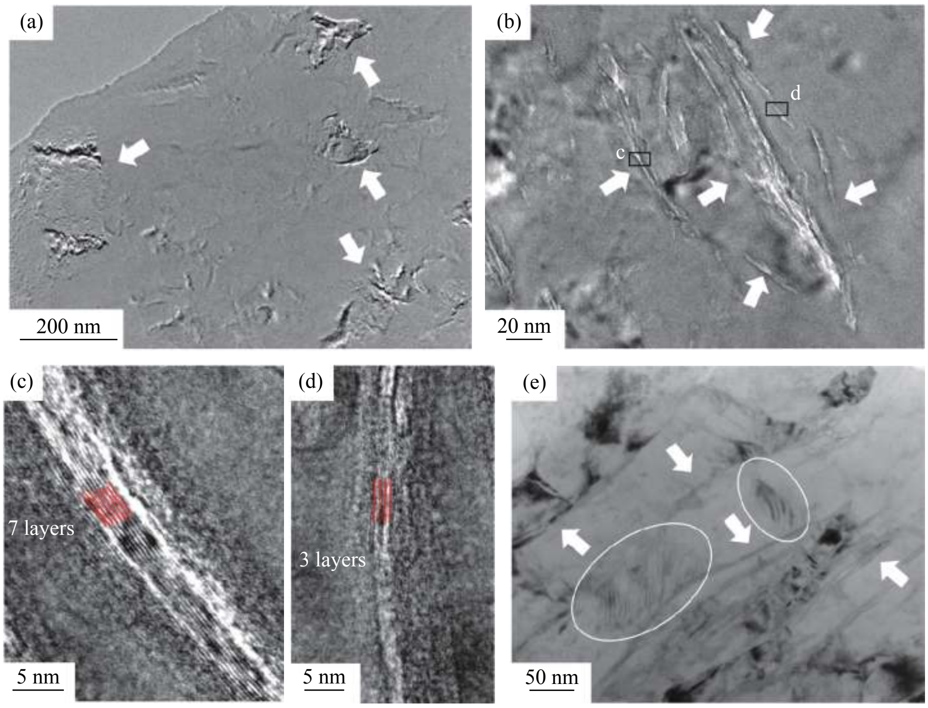

Fig. 4. TEM images of the graphenes in the hot-rolled Al/0.3vol% composite observed on the (a) RD–TD plane and (b) ND–RD plane (graphene is marked by white arrows) (RD: rolling direction; TD: transverse direction; ND: normal direction). (c, d) magnification TEM images of the graphenes in the hot-rolled Al/0.3vol% composite (FLGs are marked by red lines). (e) highly-deformed regions (marked by circles) between the graphenes (marked by white arrows) after 6% deformation. Reprinted from *Carbon*, 82, S.E. Shin, H.J. Choi, J.H. Shin, and D.H. Bae, Strengthening behavior of few-layered graphene/aluminum composites, 143-151, Copyright 2015, with permission from Elsevier.

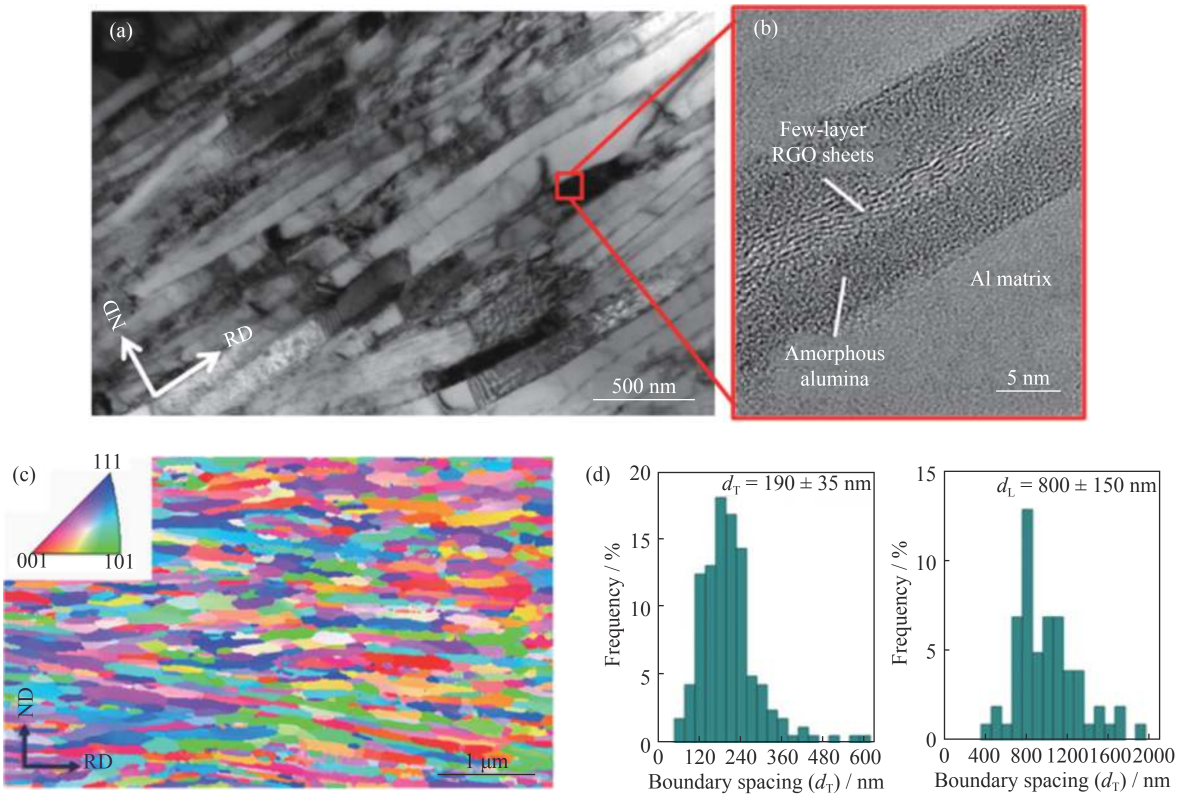

Fig. 5. Microstructural characterization of RGO/Al nanolaminated composites with RGO concentration of 1.50vol%: (a) cross-sectional TEM image of the hot-rolled composites; (b) HRTEM image of the interface of RGO and Al matrix; (c) EBSD image of the ND–RD cross section, from which the spacing of lamellar boundaries parallel to ND (d_T) and the spacing of interconnecting boundaries parallel to RD (d_L) were estimated; (d) distribution of the boundary spacing of the elongated Al grains, at least 150 boundaries were measured and statistically averaged. Reprinted with permission from Z. Li, Q. Guo, Z.Q. Li, G.L. Fan, D.B. Xiong, Y.S. Su, J. Zhang, and D. Zhang, *Nano Lett.*, 15, 8077-8083(2015) [30]. Copyright 2015 American Chemical Society.

tion stir welding, in which a rotary tool is inserted into the material for mechanical stirring, and the material is locally melted by frictional heat generation, thereby ensuring that the reinforcement can be uniformly dispersed in the matrix to obtain a material with a uniform structure. This method can be used simultaneously for deformation treatment of composite materials and also for preparation of composite materials.

Zhang *et al.* [34] used ultrasonic-assisted mechanical stirring for a 1.0wt%GNPs/2009Al mixed slurry, which was then dried at 40°C for 20 h to obtain GNPs/2009Al powders. Afterwards, the powders were pressed and sintered to a composite. Finally, the green composites were subjected to FSP at a tool rotation rate of 2000 r/min and a travel speed of 100 mm/min. The effects of frictional stirring on the dispersion of GNPs and the mechanical properties of composites were investigated. Results show that a large amount of parallel-aligned GNPs agglomerates were present in the sintered composite, while after two passes, almost no agglomeration of GNPs is observed in the microstructure of the material, as shown in Figs. 6(a) and 6(b). When the number of passes was increased to four, the distribution of the GNPs became more uniform and the grains were further refined, thereby improving the strength and toughness of the composite, as shown in Fig. 6(a). After two passes, the composite material achieved the highest mechanical properties. After four passes, the size of the GNPs was greatly reduced, thereby weakening the loading transfer effect and decreasing the performance of the composite.

F. Khodabakhshi *et al.* [35] reported a study on the addition of GNPs as a reinforcing agent into the Al matrix by directly using the FSP to prepare a 3vol%GNPs/Al–Mg composite. A groove was initially machined on the Al–Mg alloy sheet and filled with graphene powders. Capping pass was first performed at 1250 r/min and a movement speed of 25 mm/min by using a 12 mm friction head, and then five passes were performed on the composite material by using an 18mm friction head at a speed of 1200 r/min and a movement speed of 100 mm/min. The structure of GNPs was not significantly damaged during processing, but some agglomeration of GNPs in the composite still occurred. Compared with the matrix, the composite's yield strength was enhanced by more than three times, and it had an elongation of more than 20%. The researcher indicates that producing a strong interfacial bonding can enable more effective stress transfer from the matrix

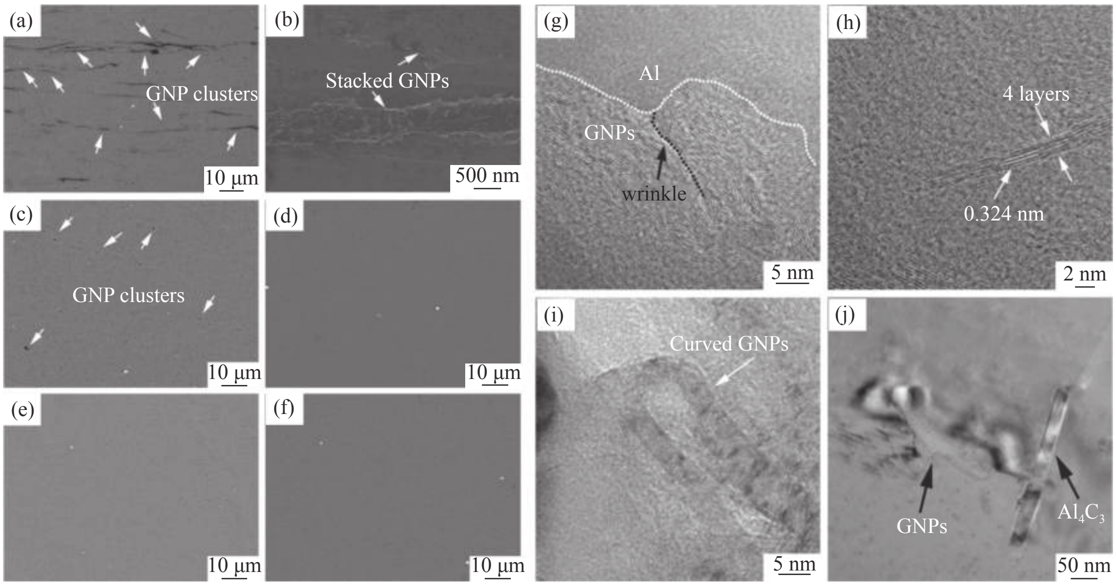

Fig. 6. SEM images showing GNP distributions in T4-treated 1wt%GNP/2009Al composites: (a, b) forged, (c) 1-pass FSP, (d) 2-pass FSP, (e) 3-pass FSP, and (f) 4-pass FSP. HRTEM images of 2-pass FSP composite showing (g) GNP–Al interface and wrinkled GNP, (h) FLG, (i) curved GNPs, and (j) TEM image of Al_4C_3 in Al matrix near GNPs. Reprinted from *Carbon*, 135, Z.W. Zhang, Z.Y. Liu, B.L. Xiao, D.R. Ni, and Z.Y. Ma, High efficiency dispersal and strengthening of graphene reinforced aluminum alloy composites fabricated by powder metallurgy combined with friction stir processing, 215-223, Copyright 2018, with permission from Elsevier.

and subsequently much higher strengthening effect, while FSP was the most effectual method in the dispersion of GNPs.

2.4. High-pressure torsion

HPT is a typical method of severe plastic deformation with obvious fine-grained effects [36–37]. It involves placing the sample between the upper and lower indenters, apply a ~10 GPa pressure along the height direction of the sample, and rotating the lower indenter at the same time. In the process, the sample is subjected to longitudinal compression and friction, thereby promoting its severe shear plastic deformation. The advantage of this method is that Gr/Al composites can be prepared at room temperature or less than 0.4T_m (T_m is the melting point of the material), avoiding harmful interface reactions through thermodynamic control and enabling the further dispersal of graphene in the composite.

To evaluate the use of HPT processing in the fabrication of Gr/Al composites to develop a comprehensive understanding of the microstructural developments in both the Al matrix and the graphene, Huang *et al.* [38] poured 5wt% GNPs and aluminum powder into a glass bottle, which was then rotated on a rotation rack for 30 min to promote the dispersion of GNPs in the aluminum powder. Then, they pressed the mixed powder in a die at 40 MPa at room temperature. HPT was performed at 298, 373, and 473 K, and composites made with different torsional turns (0, 1, 5, and 20) were studied to reveal the change in GNP distribution during this process. Fig. 7(a) shows the microstructure of composites processed by HPT at 298 K, which describes the GNP distribution in the Al matrix. As the number of turns increased, the distribution of GNPs gradually became uniform, and the agglomeration was considerably reduced. As shown in Fig. 7(b), the GNPs have a long, intact layered structure, and only slight bends were found. Fig. 8 shows the tensile results of samples processed at different temperatures. At a low processing temperature, the composite has high strength. On the basis of the experimental observations and simulation results, the researchers analyzed that the turbulent flow generated in the material during HPT caused the opening and uniform redistribution of GNP agglomeration, which is an important reason for the improvement of material properties. The application of HPT can fabricate composites at lower temperatures, thereby avoiding the problem of interface reaction. With this idea taken into consideration, the method has outstanding advantages over powder metallurgy or liquid methods.

Zhao *et al.* [39] obtained graphene/aluminum mixed powder by using ultrasonic and ball milling techniques. The powder was pressed into a tablet press machine for compaction, and a ϕ15 mm disc is cut off. A composite was prepared using an HPT device under 3 GPa for 10 revolutions at room temperature. The results of an electron probe microanalyzer show that the graphene in the prepared composite was uniformly distributed without agglomeration. The tensile strength of the composite reached 197 MPa when the content of graphene was only 0.5wt%, which is 25.5% higher than the matrix material, and also has certain advantages over the 0.5wt% MWCNT composite material prepared by the same method (its tensile strength is 181 MPa). The HPT can generate a strong shear force, which is beneficial for the further

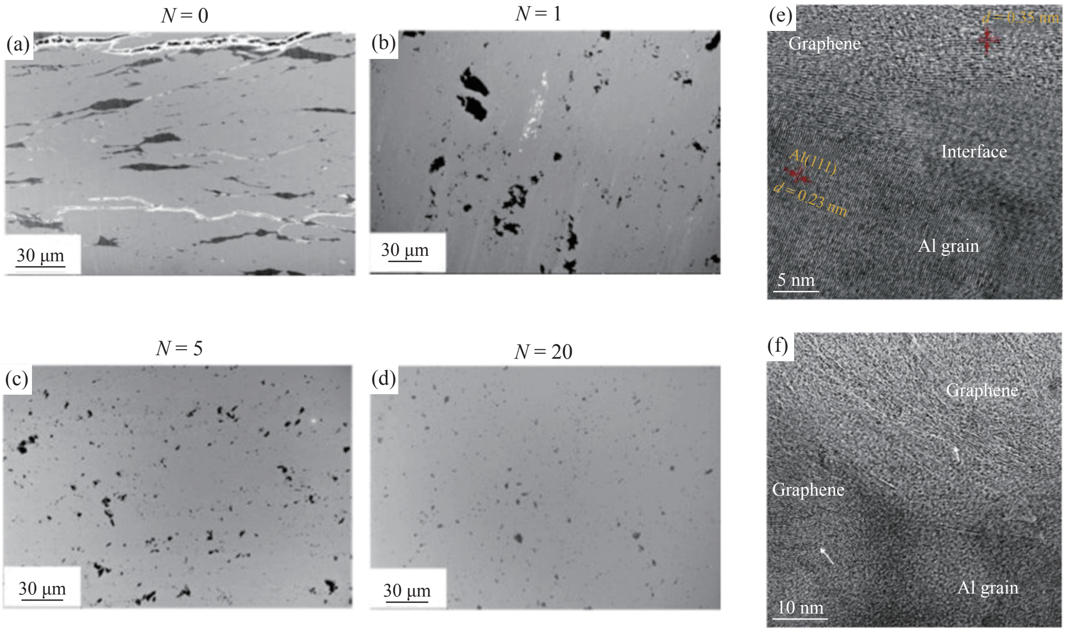

Fig. 7. Distribution of 5wt% GNPs in the Al matrix processed by HPT at 298 K with (a) *N* = 0, (b) *N* = 1, (c) *N* = 5, and (d) *N* = 20 (*N* is the number of turns). High-resolution scanning transmission electron microscopy (STEM) images of one turn sample processed by HPT at 298 K showing the interface between GNPs and the Al matrix under the condition of (e) graphene nanoplates and Al matrix having different orientations and (f) the existence of long, straight, and slightly curved GNPs in the Al matrix (*d* is the distance between graphene layers). Reprinted from *Acta Mater.*, 164, Y. Huang, P. Bazarnik, D.Q. Wan, D. Luo, P.H.R. Pereira, M. Lewandowska, J. Yao, B.E. Hayden, and T.G. Langdon, The fabrication of graphene-reinforced Al-based nanocomposites using high-pressure torsion, 499-511, Copyright 2019, with permission from Elsevier.

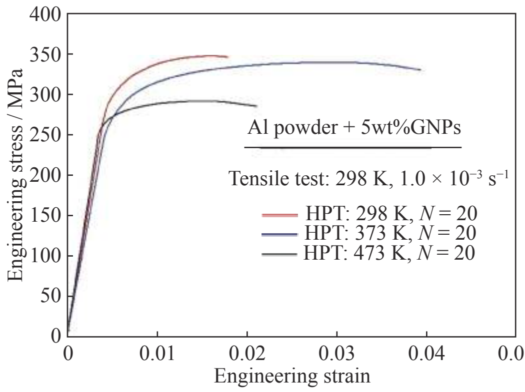

Fig. 8. Tensile results of 20 turn samples processed by HPT at different processing temperatures of 298, 373, and 473 K (the mechanical properties were tested at 298 K). Reprinted from *Acta Mater.*, 164, Y. Huang, P. Bazarnik, D.Q. Wan, D. Luo, P.H.R. Pereira, M. Lewandowska, J. Yao, B.E. Hayden, and T.G. Langdon, The fabrication of graphene-reinforced Al-based nanocomposites using high-pressure torsion, 499-511, Copyright 2019, with permission from Elsevier.

dispersion of graphene, but difficulties still arise in the large-scale preparation of the composite by using this method.

2.5. Drawing

To achieve improved distribution of reinforcement at nanoscale, researchers introduced multipass drawing into the deformation treatment of CNTs/Mg, CNTs/Cu, and CNTs/Al composites. Few reports have been published in the open literature. Li *et al.* [40] treated composites by multipass cold drawing to eliminate the GNP agglomeration. First, ball milling was used to prepare the mixed GNPs and aluminum powders. Then, the mixed powders were cold-compacted at ~200 MPa to fabricate green ingots, which were hot-extruded at 450°C with an extrusion ratio of 25:1. Finally, a sample with a diameter of 3 mm was cut for multiple passes (20% reduction in cross-sectional area per pass). Li *et al.* [40] prepared composites with a mass fraction of 0.4%, 2%, and monolithic Al by using the above process. The experimental results indicate the following: (1) with the increase in the cold drawing passes (a total of 31 passes), the holes in the material gradually decrease, the GNP agglomeration is gradually opened, and the deformation treatment can effectively improve the material performance, as shown in Fig. 9; (2) when the equivalent shear strain reaches 6.00, GNPs can be dispersed homogeneously in a matrix in 0.4wt% composite, while GNPs cannot be fully dispersed in 2.0wt% composite. With the reinforcing effect of graphene, the tensile strength of composites with 0.4wt% GNPs reaches 219 MPa, which improved by 52% compared with the matrix. The multipass process can introduce a large equivalent strain in the material, which is conducive to the improvement of the reinforcement distribution in the composite.

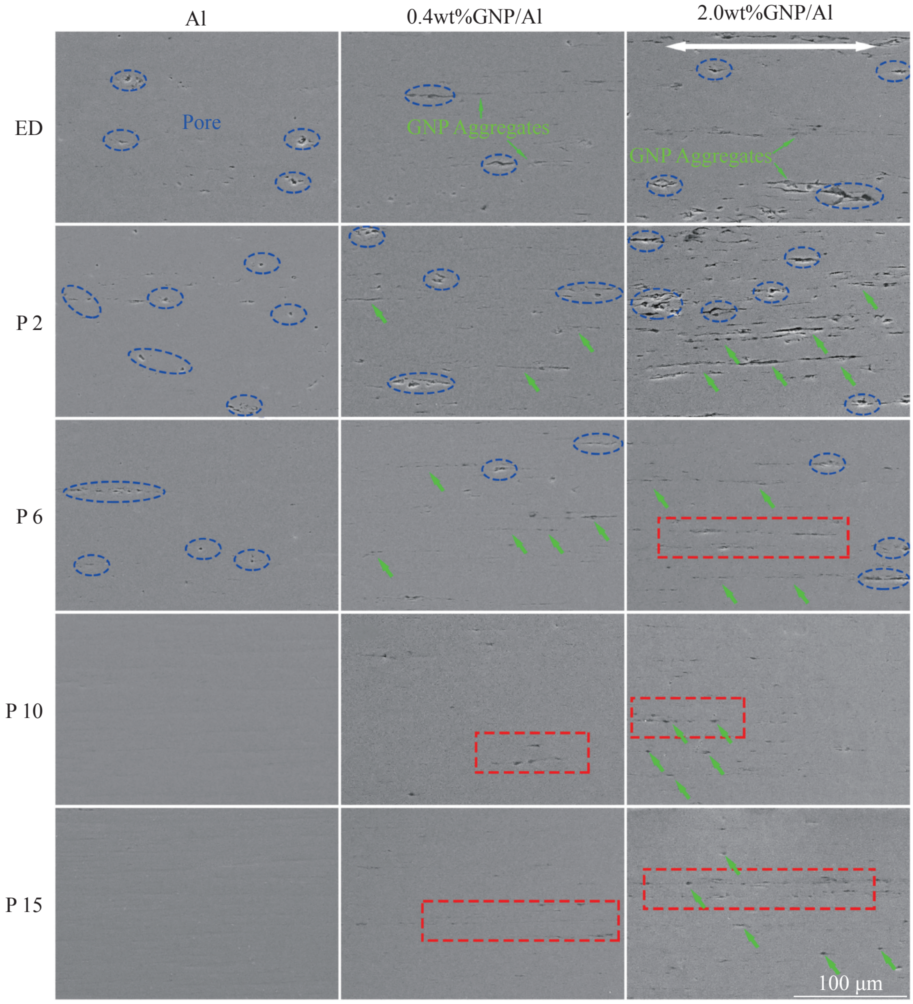

Fig. 9. Scanning electron microscope images showing the microstructure of the cold-drawn Al, 0.4wt%GNP/Al, and 2.0wt%GNP/Al composite wires at different passes (ED: extruded state; P: cold-drawn pass). Reprinted from *Mater. Des.*, 144, J.C. Li, X.X. Zhang, and L. Geng, Improving graphene distribution and mechanical properties of GNP/Al composites by cold drawing, 159-168, Copyright 2018, with permission from Elsevier.

2.6. Strengthening mechanism

At present, much research and discussion has been conducted on the strengthening mechanism. Corresponding calculation models (or modified models) are also proposed according to relevant knowledge. The shear lag model (including the modified model), Eqs. (2) and (3), is used to describe the behavior of reinforcements with large aspect ratios, where interface transfer is considered the main loading transfer method [41]. Expressions are established based on the mechanical equilibrium conditions and by taking the critical size l_c into consideration:

$$\sigma_C = \sigma_f V_f \left(\frac{l}{2l_C}\right) + \sigma_m (1 - V_f),\ l < l_C \tag{2}$$

$$\sigma_C = \sigma_f V_f \left(1 - \frac{l_C}{2l}\right) + \sigma_m (1 - V_f),\ l > l_C \tag{3}$$

where σ_C, σ_f, and σ_m mean the yield strength of composite, fiber reinforcement, and matrix respectively. V_f is the volume of fiber reinforcement.

The Halpin–Tsai model is a semiquantitative method for describing multi-walled carbon nanotubes (MWNTs)/ phenolic composites [42]. A nonlinear phenomenon occurs in the tensile strength–content curve as a result of the agglomeration problem when the CNT content is high. The modified Halpin–Tsai equation, Eq. (4), can be found using the experimental data within 3wt% of MWNTs and then used to evaluate the results for 4wt% and 5wt% of MWNTs:

$$\sigma_C = \frac{1 + \xi \eta V_f}{1 - \eta V_f} \sigma_m \tag{4}$$

where the ξ is an optimization parameter which need to take care of dispersion of the reinforcement $(2(l/d)e^{(-40V_f - 1.0)})$, d is the diameter of reinforcement, and η depends on σ_f/σ_m.

The Piggott model [43], represented by Eq. (5), modifies the discontinuous fiber model by considering two-dimensional geometric characteristics for platelet reinforcements:

$$\sigma_{\mathrm{C}} = \frac{\sigma_{\mathrm{m}} \dfrac{2l}{t} V_{\mathrm{p}}}{4} + V_{\mathrm{m}} \sigma_{\mathrm{m}} \tag{5}$$

where the V_{p} is the volume fraction of platelet, V_{m} is the volume fraction of matrix, and t is the thickness of platelet reinforcement. The strengthening mechanism of the Gr/Al composites has also been discussed in the literature according to the strengthening effect of the graphene: (1) strengthening by loading transfer ($\Delta\sigma_{\mathrm{L.T.}}$); (2) strengthening by the grain refinement ($\Delta\sigma_{\mathrm{G.R.}}$); (3) thermal mismatch enhancement ($\Delta\sigma_{\mathrm{T.M.}}$); (4) strengthening by Orowan looping system ($\Delta\sigma_{\mathrm{Oro.}}$) [44], which is shown as Eqs. (6) and (7):

$$\sigma_{\mathrm{C}} = \sigma_{\mathrm{m}} + \Delta\sigma_{\mathrm{R}} \tag{6}$$

$$\Delta\sigma_{\mathrm{R}} = \Delta\sigma_{\mathrm{L.T.}}(\mathrm{R}) + \Delta\sigma_{\mathrm{G.R.}}(\mathrm{M}) + \Delta\sigma_{\mathrm{T.M.}}(\mathrm{R}) + \Delta\sigma_{\mathrm{Oro.}}(\mathrm{R}) \tag{7}$$

where $\Delta\sigma_{\mathrm{R}}$ is strengthening brought by reinforcement. $\Delta\sigma_{\mathrm{G.R.}}(\mathrm{M})$, $\Delta\sigma_{\mathrm{L.T.}}(\mathrm{R})$, $\Delta\sigma_{\mathrm{T.M.}}(\mathrm{R})$, and $\Delta\sigma_{\mathrm{Oro.}}(\mathrm{R})$ are strengthening brought by matrix (M) and reinforcement (R), respectively. This method requires more sufficient characterization of the composites, but it may also introduce more evaluation errors. Therefore, the development of more reliable theoretical models is an important part of future research on Gr/Al composites.

3. Summary and future perspective

Graphene has high mechanical properties and excellent electrical and thermal properties, and it is an ideal reinforcement for metal matrix composites. It is expected to break through the performance limitations of traditional aluminum-based composites. How to disperse graphene uniformly in the Al matrix, maintain the integrity of the microstructure, and form a good interface with the Al matrix are key issues that need to be tackled in the future. Research on the synthesis methods is relatively complete, whereas research on deformation treatment is still insufficient.

Graphene has exhibited an extraordinary strengthening effect in composites. However, this effect has not yet achieved the application needs and the researchers' expectations for ultra-high performance. Many challenges still remain in the research process. Therefore, in the future, solutions can be sought from the following aspects.

(1) Through the subsequent deformation treatment, further dispersion of graphene can be achieved after consolidation of the composite and adjustment of a reasonable deformation parameter to exfoliate the graphene layers. Greater shear deformation needs to be introduced into the composites through severe plastic deformation process or other methods.

(2) Quantitative description of graphene dispersion. New characterization methods need to be developed to describe the dispersion of graphene in macroscale, which is not limited to microscale without further discussion.

(3) Multiscale *in-situ* characterization is needed to study the behavior of graphene during deformation treatment, such as digital image correlation, synchrotron X-ray beam, and *in-situ* TEM. It can also explain the strengthening effect of grapheme.

(4) Multiscale simulation technology is needed to describe graphene dispersion under thermal deformation, including first-principles calculations, molecular dynamics, and crystal plastic finite element.

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References

[1] M.E. Smagorinski, P.G. Tsantrizos, S. Grenier, A. Cavasin, T. Brzezinski, and G. Kim, The properties and microstructure of Al-based composites reinforced with ceramic particles, *Mater. Sci. Eng. A*, 244(1998), No. 1, p. 86.

[2] B.Q. Han, J.Y. Huang, Y.T. Zhu, and E.J. Lavernia, Effect of strain rate on the ductility of a nanostructured aluminum alloy, *Scripta Mater.*, 54(2006), No. 6, p. 1175.

[3] E.S. Caballero, J. Cintas, F.G. Cuevas, J.M. Montes, and F. Ternero, Influence of milling atmosphere on the controlled formation of ultrafine dispersoids in Al-based MMCs, *Metals*, 6(2016), No. 9, p. 224.

[4] N. Chawla and K.K. Chawla, *Metal Matrix Composites*, 2nd Ed., Springer Science + Business Media, New York, 2013, p. 3.

[5] K.S. Novoselov, A.K. Geim, S.V. Morozov, D. Jiang, Y. Zhang, S.V. Dubonos, I.V. Grigorieva, and A.A. Firsov, Electric field effect in atomically thin carbon films, *Science*, 306(2004), No. 5696, p. 666.

[6] C.G. Lee, X.D. Wei, J.W. Kysar, and J. Hone, Measurement of the elastic properties and intrinsic strength of monolayer graphene, *Science*, 321(2008), No. 5887, p. 385.

[7] A.A. Balandin, S. Ghosh, W.Z. Bao, I. Calizo, D. Teweldebrhan, F. Miao, and C.N. Lau, Superior thermal conductivity of single-layer graphene, *Nano Lett.*, 8(2008), No. 3, p. 902.

[8] Y.B. Zhang, Y.W. Tan, H.L. Stormer, and P. Kim, Experimental observation of the quantum hall effect and Berry' s phase in graphene, *Nature*, 438(2005), No. 7065, p. 201.

[9] R.G. Wang, Z. Li, W.B. Liu, W.C. Jiao, L.F. Hao, and F. Yang, Attapulgite–graphene oxide hybrids as thermal and mechanical reinforcements for epoxy composites, *Compos. Sci. Technol.*, 87(2013), p. 29.

[10] Y.G. Han, T.Q. Wang, X.X. Gao, T.X. Li, and Q. Zhang, Preparation of thermally reduced graphene oxide and the influence of its reduction temperature on the thermal, mechanical, flame retardant performances of PS nanocomposites, *Composites Part*

A, 84(2016), p. 336.

[11] C. Ramirez, P. Miranzo, M. Belmonte, M.I. Osendi, P. Poza, S.M. Vega-Diaz, and M. Terrones, Extraordinary toughening enhancement and flexural strength in Si_3N_4 composites using graphene sheets, *J. Eur. Ceram. Soc.*, 34(2014), No. 2, p. 161.

[12] S.F. Bartoluccia, J. Paras, M.A. Rafiee, J. Rafiee, S.L. Lee, D. Kapoor, and N. Koratkar, Graphene–aluminum nanocomposites, *Mater. Sci. Eng. A*, 528(2011), No. 27, p. 7933.

[13] S.E. Shin, H.J. Choi, J.H. Shin, and D.H. Bae, Strengthening behavior of few-layered graphene/aluminum composites, *Carbon*, 82(2015), p. 143.

[14] Y.Y. Jiang, Z.Q. Tan, R. Xu, G.L. Fan, D.B. Xiong, Q. Guo, Y.S. Su, Z.Q. Li, and D. Zhang, Tailoring the structure and mechanical properties of graphene nanosheet/aluminum composites by flake powder metallurgy via shift-speed ball milling, *Composites Part A*, 111(2018), p. 73.

[15] S.E. Shin and D.H. Bae, Deformation behavior of aluminum alloy matrix composites reinforced with few-layer graphene, *Composites Part A*, 78(2015), p. 42.

[16] P.Z. Shao, W.S. Yang, Q. Zhang, Q.Y. Meng, X. Tan, Z.Y. Xiu, J. Qiao, Z.H. Yu, and G.H. Wu, Microstructure and tensile properties of 5083 Al matrix composites reinforced with graphene oxide and graphene nanoplates prepared by pressure infiltration method, *Composites Part A*, 109(2018), p. 151.

[17] M. Tabandeh-Khorshid, A. Kumar, E. Omrani, C. Kim, and P. Rohatgi, Synthesis, characterization, and properties of graphene reinforced metal-matrix nanocomposites, *Composites Part B*, 183(2020), art. No. 107664.

[18] A. Nieto, A. Bisht, D. Lahiri, C. Zhang, and A. Agarwal, Graphene reinforced metal and ceramic matrix composites: A review, *Int. Mater. Rev.*, 62(2017), No. 5, p. 241.

[19] F. Chen, N. Gupta, R.K. Behera, and P.K. Rohatgi, Graphene-reinforced aluminum matrix composites: A review of synthesis methods and properties, *JOM*, 70(2018), No. 6, p. 837.

[20] M.X. Li, Y.H. Zhao, L.W. Chen, J.Q. Liang, T. Zhang, and H. Hou, Research progress on preparation technology of graphene-reinforced aluminum matrix composites, *Mater. Res. Express*, 6(2018), No. 3, art. No. 032002.

[21] S.C. Tjong, Recent progress in the development and properties of novel metal matrix nanocomposites reinforced with carbon nanotubes and graphene nanosheets, *Mater. Sci. Eng. R*, 74(2013), No. 10, p. 281.

[22] P.Z. Shao, G.Q. Chen, B.Y. Ju, W.S. Yang, Q. Zhang, Z.J. Wang, X. Tan, Y.Y. Pei, S.J. Zhong, M. Hussain, and G. Wu, Effect of hot extrusion temperature on graphene nanoplatelets reinforced Al6061 composite fabricated by pressure infiltration method, *Carbon*, 162(2020), p. 455.

[23] M. Alipour and R. Eslami-Farsani, Synthesis and characterization of graphene nanoplatelets reinforced AA7068 matrix nanocomposites produced by liquid metallurgy route, *Mater. Sci. Eng. A*, 706(2017), p. 71.

[24] W.W. Zhou, Y.C. Fan, X.P. Feng, K. Kikuchi, N. Nomura, and A. Kawasaki, Creation of individual few-layer graphene incorporated in an aluminum matrix, *Composites Part A*, 112(2018), p. 168.

[25] W.S. Yang, Q.Q. Zhao, L. Xin, J. Qiao, J.Y. Zou, P.Z. Shao, Z.H. Yu, Q. Zhang, and G.H. Wu, Microstructure and mechanical properties of graphene nanoplates reinforced pure Al matrix composites prepared by pressure infiltration method, *J. Alloys Compd.*, 732(2018), p. 748.

[26] R. Xu, Z.Q. Tan, G.L. Fan, G. Ji, D.B. Xiong, Q. Guo, Y.S. Su, Z.Q. Li, and D. Zhang, High-strength CNT/Al–Zn–Mg–Cu composites with improved ductility achieved by flake powder metallurgy via elemental alloying, *Composites Part A*, 111(2018), p. 1.

[27] H. Kurita, M. Estili, H. Kwon, T. Miyazaki, W.W. Zhou, J.F. Silvain, and A. Kawasaki, Load-bearing contribution of multi-walled carbon nanotubes on tensile response of aluminum, *Composites Part A*, 68(2015), p. 133.

[28] Z.Y. Liu, B.L. Xiao, W.G. Wang, and Z.Y. Ma, Modelling of carbon nanotube dispersion and strengthening mechanisms in Al matrix composites prepared by high energy ball milling-powder metallurgy method, *Composites Part A*, 94(2017), p. 189.

[29] S.E. Shin, H.J. Choi, J.Y. Hwang, and D.H. Bae, Strengthening behavior of carbon/metal nanocomposites, *Sci. Rep.*, 5(2015), art. No. 16114.

[30] Z. Li, Q. Guo, Z.Q. Li, G.L. Fan, D.B. Xiong, Y.S. Su, J. Zhang, and D. Zhang, Enhanced mechanical properties of graphene (reduced graphene oxide)/aluminum composites with a bioinspired nanolaminated structure, *Nano Lett.*, 15(2015), No. 12, p. 8077.

[31] X.D. Li, Z.H. Xu, and R.Z. Wang, *In situ* observation of nanograin rotation and deformation in nacre, *Nano Lett.*, 6(2006), No. 10, p. 2301.

[32] U.G.K. Wegst, H. Bai, E. Saiz, A.P. Tomsia, and R.O. Ritchie, Bioinspired structural materials, *Nat. Mater.*, 14(2015), No. 1, p. 23.

[33] Z.Y. Liu, B.L. Xiao, W.G. Wang, and Z.Y. Ma, Analysis of carbon nanotube shortening and composite strengthening in carbon nanotube/aluminum composites fabricated by multi-pass friction stir processing, *Carbon*, 69(2014), p. 264.

[34] Z.W. Zhang, Z.Y. Liu, B.L. Xiao, D.R. Ni, and Z.Y. Ma, High efficiency dispersal and strengthening of graphene reinforced aluminum alloy composites fabricated by powder metallurgy combined with friction stir processing, *Carbon*, 135(2018), p. 215.

[35] F. Khodabakhshi, S.M. Arab, P. Švec, and A.P. Gerlich, Fabrication of a new Al–Mg/graphene nanocomposite by multi-pass friction-stir processing: Dispersion, microstructure, stability, and strengthening, *Mater. Charact.*, 132(2017), p. 92.

[36] W.T. Sun, X.G. Qiao, M.Y. Zheng, C. Xu, S. Kamado, X.J. Zhao, H.W. Chen, N. Gao, and M.J. Starink, Altered ageing behaviour of a nanostructured Mg–8.2Gd–3.8Y–1.0Zn–0.4Zr alloy processed by high pressure torsion, *Acta Mater.*, 151(2018), p. 260.

[37] P. Jenei, J. Gubicza, E.Y. Yoon, H.S. Kim, and J.L. Lábár, High temperature thermal stability of pure copper and copper–carbon nanotube composites consolidated by High Pressure Torsion, *Composites Part A*, 51(2013), p. 71.

[38] Y. Huang, P. Bazarnik, D.Q. Wan, D. Luo, P.H.R. Pereira, M. Lewandowska, J. Yao, B.E. Hayden, and T.G. Langdon, The fabrication of graphene-reinforced Al-based nanocomposites using high-pressure torsion, *Acta Mater.*, 164(2019), p. 499.

[39] L.Y. Zhao, H.M. Lu, and Z.J. Gao, Microstructure and mechanical properties of Al/graphene composite produced by high-pressure torsion, *Adv. Eng. Mater.*, 17(2015), No. 7, p. 976.

[40] J.C. Li, X.X. Zhang, and L. Geng, Improving graphene distribution and mechanical properties of GNP/Al composites by cold drawing, *Mater. Des.*, 144(2018), p. 159.

[41] J.M. Hedgepeth, *Stress Concentrations in Filamentary Structures*, NASA Technical Note D-882, NASA Langley Research Center, Langley Field, VA, 1961.

[42] M.K. Yeh, N.H. Tai, and J.H. Liu, Mechanical behavior of phenolic-based composites reinforced with multi-walled carbon nanotubes, *Carbon*, 44(2006), No. 1, p. 1.

[43] M.R. Piggott, *Load Bearing Fibre Composites*, 1st ed., Pergamon, Oxford, 1980, p. 59.

[44] X. Zhang, S.F. Li, B. Pan, D. Pan, S.Y. Zhou, S.H. Yang, L. Jia, and K. Kondoh, A novel strengthening effect of *in-situ* nano Al_2O_3w on CNTs reinforced aluminum matrix nanocomposites and the matched strengthening mechanisms, *J. Alloys Compd.*, 764(2018), p. 279.