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Advances in graphene reinforced metal matrix nanocomposites: Mechanisms, processing, modelling, properties and applications

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ABSTRACT

Graphene has been extensively explored to enhance functional and mechanical properties of metal matrix nanocomposites for wide-range applications due to their superior mechanical, electrical and thermal properties. This article discusses recent advances of key mechanisms, synthesis, manufacture, modelling and applications of graphene metal matrix nanocomposites. The main strengthening mechanisms include load transfer, Orowan cycle, thermal mismatch, and refinement strengthening. Synthesis technologies are discussed including some conventional methods (such as liquid metallurgy, powder metallurgy, thermal spraying and deposition technology) and some advanced processing methods (such as molecular-level mixing and friction stir processing). Analytical modelling (including phenomenological models, semi-empirical models, homogenization models, and self-consistent model) and numerical simulations (including finite elements method, finite difference method, and boundary element method) have been discussed for understanding the interface bonding and performance characteristics between graphene and different metal matrices (Al, Cu, Mg, Ni). Key challenges in applying graphene as a reinforcing component for the metal matrix composites and the potential solutions as well as prospectives of future development and opportunities are highlighted.

1. Introduction

To enhance specific strength of metal matrix composites (MMCs), novel reinforcements such as whiskers, ceramic particles, and fibers are often applied.[1] These MMCs generally not only enhance the strength of the matrix material, but also maintain its high toughness,[2] which have found wide applications in aerospace, marine, military and other fields including electronics and automotive industries.[3,4] In general, carbon material reinforced MMCs often show a better overall performance and excellent enhancement effect over ceramic reinforced MMCs owing to their high electrical and thermal conductivity, excellent vibration damping properties and self-lubricating properties.[5,6] Early studies showed that graphite and carbon fiber are effective reinforcement phases to MMCs, not only improving their strength, but also maintaining good electrical conductivity and small friction coefficient.[7–9] However, the density of graphite is signficantly less than that of the metal matrix, so graphite easily becomes agglomerated into graphite clusters, thereby reducing the mechanical properties of the metal matrix. Due to the anisotropy of carbon fibers, the shear strength of the carbon fiber based composite material along the fiber length direction is dramatically different from that perpendicular to the fiber direction. These severely limit the potential applications of graphite- and carbon fiber reinforced metal matrix composites.[10] One dimensional (1D) carbon materials, such as carbon nanotubes (CNTs) have also been reported difficult to be reinforced into metal matrix nanocomposites (MMNCs).[11] The main reasons are that small specific surface area of CNTs is not easy to form a strong bonding interface with the metal matrix, and the van der Waals force between CNTs is very large, which easily leads to

agglomeration,[12] thus affecting the mechanical properties and electrical properties of composite materials.

Graphene as a two dimensional (2D) carbon material with large surface areas shows higher intimate contact with the metal matrix,[13,14] and also shows ultra-high elastic modulus, yield strength, good electrical conductivity[15] and thermal conductivity.[16,17] Graphene reinforced metal matrix composites (GRMMCs) show excellent thermal and mechanical properties, which make it suitable for wide-range application prospects in the fields of catalysis, electronics, energy storage, conversion, sensing and biotechnology.[18–20] Currently the main challenges of GRMMCs include[21,22]: (1) avoiding the damage of the network structure of graphene; (2) improving the degree of uniform dispersion of graphene in the metal matrix; (3) strengthening the interface adhesion and wettability between graphene and the metal matrix.

Nowadays, considerable efforts have been made for preparing high-quality graphene in large quantities for research and potential applications. Since its discovery in 2004 using the micromechanical cleavage method,[15] various approaches have been utilised to prepare graphene and graphene derivatives including graphene oxide (GO), reduced graphene oxide (rGO) and functionalized graphene nanosheets. For example, liquid-phase exfoliation (top-down) method can be used to synthesize large quantities of single or multilayer graphene sheets with small width, and involves the isolation of graphene layers from graphite or graphite derivatives. Chemical vapor deposition (CVD), one of the bottom-up methods builds up the graphene sheet atom by atom to produce large size and high-quality nanosheets.[23–25] There are other methods such as epitaxial growth method,[25,26] molecular beam epitaxy,[26] and most recently flash graphene.[27] Graphene derivatives are usually considered as an economical alternative than graphene for large-scale production for fabricating nanocomposites. They have also proven to be effective as fillers in nanocomposite materials.[28,29]

In literature, there are a few review papers about the research progress of the graphene reinforced metal matrix nanocomposites (GRMMNCs). For example, Khorshid et al.[30] reviewed the process of preparing GRMMNCs using the powder metallurgy and various characterization methods for studying GRMMNCs. Xavior et al.[31] introduced two types of methods for making composites of graphene and metal matrix, including mechanical methods (ball milling, ultrasonic dispersion) and chemical method (electrodeposition), and highlighted problems of graphene agglomeration and weak composite interface in the preparation of GRMMCs. Yang et al.[32] reported influences of the interfacial bonding between graphene and different metals on the mechanical properties, electrical properties and thermal properties, magnetic properties and friction properties of composite materials, for applications in electronic equipment, aerospace, nuclear industry. However, most of these reviews have neither covered in-depth discussions of strengthening mechanisms of GRMMNCs, nor comprehensively reviewed various technologies for preparing the GRMMNCs. More importantly, the theoretical and analytical models used to predict the enhancement effect of graphene have seldom been mentioned.

Herein, we will thoroughly discuss and compare the preparation technologies of GRMMNCs, and focus on the key strengthening mechanisms and principles. The dispersibility and interfacial wettability between graphene and different metal matrices (Al, Cu, Mg, Ni) are evaluated and the cost effective performance is evaluated. Theoretical and analytical modelling methodologies for predicting the performance of GRMMNCs are then discussed. Finally, the key issues of GRMMNCs are summarized and their potential application prospects are highlighted.

2. Design and enhancing mechanisms

Exploring the enhancement mechanisms of graphene in the metal matrix is beneficial to the fundamental designs of MMCs, and can provide guidance on improving synthesis technology for achieving the best reinforcement effect. Mathematical models can be developed to represent the enhancement values brought by different enhancement mechanisms due to the addition of the graphene. The micromechanics model can be established to predict the local stress and strain fields at different geometry, phases, and to predict the responses of non-uniformity composite mechanical properties to the geometry and properties of the different phases.[33]

Several mechanisms have been proposed to explain the strengthening effects of graphene in MMCs: (1) Grain refinement in the MMCs via pinning effect; (2) Solution strengthening of the interstitial carbon, oxygen and nitrogen; (3) Dispersion strengthening of MMCs by the uniformly dispersed graphene; (4) Dislocation strengthening of in-situ formed carbides within the matrix; and (5) Effective load transferring function from the matrix to graphene. However, the exact strengthening mechanisms for a given composite is dependent on the nature of its structures. Therefore, the key enhancement mechanisms of graphene in the most commonly used metal substrates are load transfer,[34] Orowan cycle,[35] thermal mismatch mechanism[36] and grain refining strengthening,[34] which will be discussed in detail in the following sections.

2.1. Load transfer

In order to explain the strengthening effects of graphene in MMNCs, three types of distribution models are proposed (Fig. 1). The distribution of graphene in the metal matrix has a great influence on the load transfer efficiency. Fig. 1 shows the distribution type of graphene in the metal matrix. When the loading is along the direction as shown in Fig. 1, the graphene in Fig. 1(a) is along the interface, and its contribution to the load transfer is insignificant and even weakens the strength of the composite material. This is because the surface energy between the graphene planes is quite large, and more graphenes will be agglomerated. They are prone to separation when subjected to tensile stress, thus easily resulting in direct separation of the interface and no load transfer. On the other hand, the interface bonding between graphene and the metal matrix is inherently poor, and the composite interface will be separated easily when subjected to a load. In Fig. 1(b), the graphene has a certain inclination angle in the metal matrix, so when the load is transferred to the composite interface, the graphene will be rotated and stretched. This process will provide an enhanced load transfer efficiency. When graphene is vertically distributed in the metal matrix (Fig. 1(c)), the graphene will be stretched under the load. For composites, the contribution of load transfer is the largest under this distribution condition.

Fig. 2 shows a schematic diagram of the load transfer mechanism. The distribution type of graphene in the metal matrix is shown in Figs. 2(b) and (c). The bond between graphene and the matrix is strong. Fig. 2(a) shows that the interface is perfect and there are no cracks or defects produced. When a load is applied along the 2D length direction of reinforced graphene, the stress has been effectively transferred to the matrix as shown in Fig. 2(b). The matrix metal is initially elongated and deformed. Due to the strong interfacial adhesion between the graphene
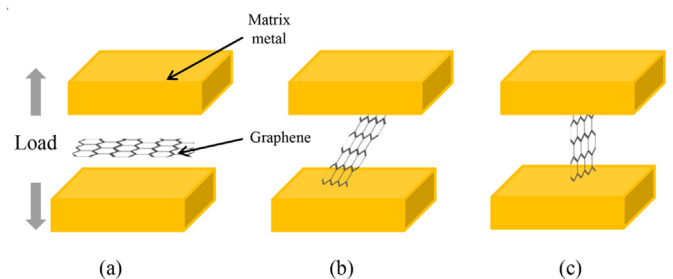
Fig. 1. Distribution model of graphene in metal matrix: (a) horizontally; (b) obliquely; (c) vertically.
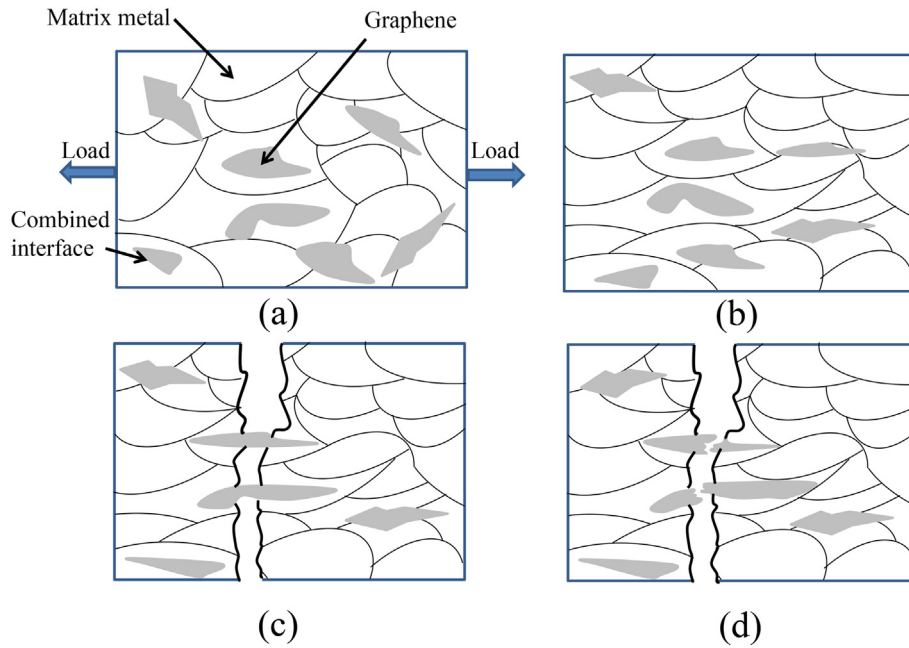
Fig. 2. Schematic diagram of load transfer: (a) the initial composite material with randomly distributed graphene; (b) the matrix deforms and the graphene rotates after being stressed; (c) the graphene is deformed and elongated; and (d) the graphene finally breaks.

and metal matrix, the load has been effectively transferred, causing the graphene to be elongated and deformed (Fig. 2c). Finally, a further increase in the load will lead to the signficant deformation of graphene structure or even break (Fig. 2d).

Yan's group reported that the value of the yield strength provided by the load transfer mechanism in graphene-reinforced aluminum composites follows the following formula[37]:

$$\sigma_s = \left(\sigma_0 + kd^{-\frac{1}{2}}\right)\left[\frac{V_f(s+4)}{4} + (1-V_f)\right] \tag{1}$$

where σ_0 and k are constants associated with the crystal type; d is the matrix size; V_f is the graphene's volume fraction; and s is the aspect ratio of the graphene. It can be seen that when the graphene's volume fraction V_f is fixed, with the increase of values of s, the yield strength (σ_s) increases. Using this model, the yield strength of graphene-Ni/Cu composites was theoretically predicted, which is similar to the experimental result.[38] The effective transfer of load is the main reason for

the enhanced strength. Addition of Ni nanoparticles not only improves the dispersibility of graphene, but also enhances the wettability at the interfaces and improves the interface bonding.[38] However, this mechanism does not take into account the negative effects of agglomeration when the graphene content is large. It is only applicable for the ideal situation where graphene is homogeneously dispersed. The reinforcement effect of large amount of graphene is much lower than the theoretical calculation values. Theoretical studies[39] showed that aspect ratio, dispersion, orientation direction of graphene and interfacial structures determine the efficiency of load transfer effects within the GRMMNCs.

2.2. Orowan cycle

Fig. 3 shows a schematic illustration of Orowan cycle strengthening mechanism, which is mainly due to the uniform dispersion of graphene in the MMCs. When the composite material is plastically deformed, the dislocations will occur (Fig. 3a). The movement of dislocations shown in
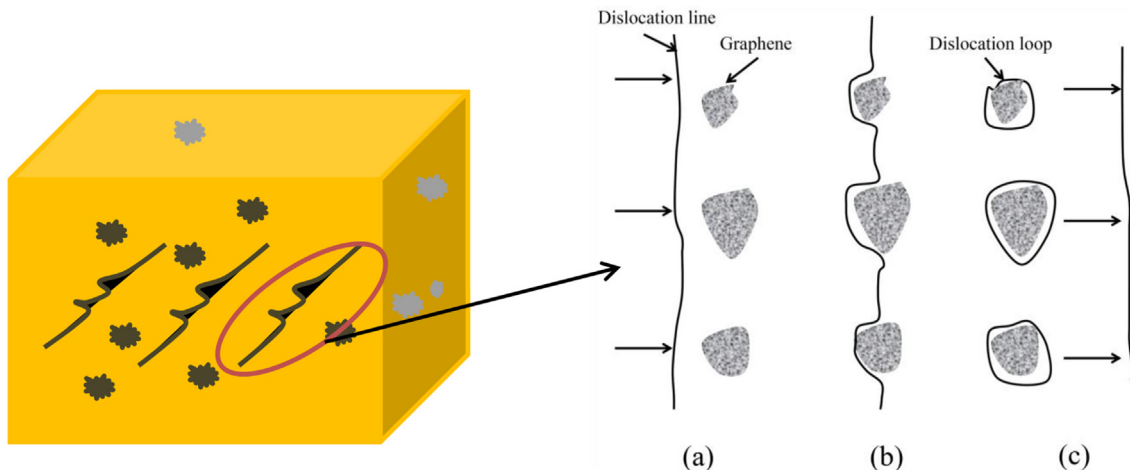
Fig. 3. Schematic diagram of Orowan cycle strengthening: (a) initial dislocation; (b) dislocation hindered deformation; (c) dislocation loop formation.

Fig. 3(b) is hindered when these dislocations are in contact with graphene. Therefore, it is necessary to further increase the applied stress, so that the dislocation will cut through or bypass the graphene and then continue to move. There will be dislocation rings generated surrounding the graphene as shown in Fig. 3(c). The dislocation hardening mechanism can be expressed using Eq. (2).[40]

$$\tau = \frac{0.84 MGb}{(L_\mathrm{m} - S)} \qquad (2)$$

where τ is the shear stress, M is the Taylor coefficient as a constant, G is the shear modulus of the matrix, b is the Burgers vector, L_m is the distance between the reinforcements, and S is the average size of graphene. Moreover, a smaller distance among the graphene will result in a larger shear strength of the composites.

2.3. Thermal mismatch mechanism

The coefficients of thermal expansion of graphene and metal substrates usually differ by about an order of magnitude, and increase in the thermal mismatch has been proposed as one of the enhancement mechanism for the GRMMNCs. Arsenault and Fisher[41] pointed out in SiC/Al composite, the mismatch of thermal expansion of the reinforcement and the substrate during thermal processing, such as sintering, annealing heat treatment, etc. results in significant differences in the volume expansion and contraction, thus increasing the defect levels.[36] Generally speaking, the larger the interface areas are, the larger the dislocation density is along the composite interface, and the more significant this enhancement effect is. The dislocation density formula and strength can be expressed using formula (3) and formula (4).[42]

$$\rho = \frac{10A\epsilon}{bt(1-A)} \qquad (3)$$

where ρ is the dislocation density, A is the volume fraction of the reinforcements, ϵ is the thermal strain, b is the Burgers vector, and t is the size of the reinforcement. The enhancement strength $\Delta\sigma$ can be written as:

$$\Delta\sigma = \alpha\mu\rho^{\frac{1}{2}}b \qquad (4)$$

where α is a constant, and μ is the stiffness modulus of the matrix. From formula (3), the more content of reinforcement, the larger the dislocation density and the higher the theoretical strength. However, when the size of graphene is too small and/or its content is too low, the thermal mismatch mechanism becomes insignificant.[43] The higher graphene content will lead to an increased chance of graphene agglomeration, which will significantly reduce its strengthening effect. An increase of graphene content will also increase the composites' defect levels, decrease the material density, and reduce the strengthening effect.

2.4. Grain refining strengthening

Large specific surface area of the graphene effectively prevents the growth of metal matrix grains during sintering, which plays a critical role in refining grains. Meanwhile, the interfaces between graphene and the metal matrix hinder dislocation motion and crack propagation. The effect of grain refinement can be quantified according to the Hall-Petch formula[34]:

$$\sigma_\mathrm{s} = \sigma_0 + kd^{\frac{1}{2}} \qquad (5)$$

Table 1
Key technologies used to process the GRMMCs.[45–59]

Preparation technology	Specific method	Advantages	Disadvantages	Refs.
Liquid metallurgy (LM)	Arc melting, laser cladding, stirring casting	(1) Simple, convenient and practical; (2) excellent compactness; (3) mass production capability.	(1) Graphene has a low density and is easy to float during the smelting process; (2) graphene has a poor wetting effect with the metal matrix, resulting in poor interface bonding.	45–47
Thermal spraying (TS)	Plasma spraying, high velocity oxy fuel spraying, flame spraying, electric arc spraying, detonation flame spraying	Graphene is evenly distributed in the feedstock.	(1) The sprayed layer is relatively thick, which is not suitable for thin film composite coating; (2) the process is complicated; (3) thickness uniformity is poor; (4) empty drum; (5) partially uncoated.	48–51
Powder metallurgy (PM)	Hot press sintering, spark plasma sintering (SPS), microwave sintering, laser sintering, secondary deformation treatment (hot extrusion, hot forging, equal channel angle processing)	(1) The process is simple and easy to operate; (2) graphene has a high degree of dispersion.	(1) Structural integrity of graphene is easily damaged, and degree of defects is large; (2) density of composite materials is poor.	52–55
Surface deposition	Electrochemical deposition (ED), chemical vapor deposition (CVD)	(1) Graphene has a high quality; (2) graphene has a good dispersibility; (3) composite interface is well bonded.	(1) The thickness of the composite coating prepared by ED technology is limited; (2) CVD technology will have carbon precursor residues; (3) coarse grains.	56, 57
Other methods	Molecular level mixing (MLM), friction stir processing (FSP)	(1) Graphene has a high quality; (2) composite interface is well bonded.	The process is complicated and not convenient for mass production.	58, 59

The above formula shows that as the grain size d decreases, the yield strength of the material increases. Previous work showed that addition of graphene to W70Cu30 composites can effectively refine the tungsten grains.[44] The tungsten grains (14.67 μm) with 1.0 wt% graphene were significantly smaller than those without adding it (45.50 μm).

3. Processing technologies and graphene dispersing techniques

Preparation technologies for the GRMMNCs have signficant influences on the dispersion uniformity of graphene, interfacial adhesion and the composites' structural integrity. The key technologies for preparing GRMMNCs can be divided into chemical and physical methods according to the presence or absence of chemical reactions in the preparation process. Among these methods, the physical means include liquid metallurgy (LM), thermal spraying (TS), powder metallurgy (PM), molecular-level mixing technology (MLM) and friction stir processing (FSP); whereas the chemical means mainly include chemical vapor deposition (CVD) and electrochemical deposition (ED). Table 1 lists the key technologies of preparing GRMMNCs, including their advantages and disadvantages. Table 2 summarizes performances of different types of GRMMNCs after different processes, as well as the comparison of physical properties with other reinforcement/metal matrix composites.

3.1. Liquid metallurgy (LM)

Fig. 4 shows two commonly used LM processes, namely stirring casting technology and arc melting technology. To solve the uneven distribution of graphene in the LM technology, the stirring casting technology has been developed. During the process of adding graphene into the molten metal, an additional stirring process is added so that graphene can be uniformly distributed into the matrix.[81] The process is simple and easy to operate, suitable for mass production. However, as shown in Fig. 4, because the density of graphene is much lower than that of metal, it often floats on the upper part of the molten metal, making an uneven distribution of graphene. The poor wettability between graphene and metal leads to a reduction in the contact surface of the molten metal and graphene. At the same time, a very high heating temperature will increase the fabrication cost, and lead to the reaction between the graphene and the matrix, as well as the severe oxidation of matrix metal.

In addition, during the laser cladding process, excessively high temperatures cause the formation of interface products such as carbide and increase in thermal stress, which is the problem of limiting laser cladding based LM technology at this stage.

3.2. Thermal spray (TS)

TS technology was initially used to prepare surface coatings on the industrial components, for examples, antirust coatings on the surface of automobile shells, and thermal softening prevention coatings in the military industry, thermal barrier coatings and biological coatings for aerospace and medical industries.[82,83] The TS process mainly includes sequenial depositions of coating materials through droplet formation process[84,85], droplet spraying process[86], and atomizing droplet deposition and cooling process on the substrate.[87] However, as shown in Fig. 5, the nozzle moving speed and the size of the discharge port have significant influences on the coating quality and thickness uniformity.[50,51,88]

Fig. 5 also shows the process of obtaining GRMMNCs using the TS method. The graphene composite powders were melted to a molten state in the combustion chamber, and then they were accelerated to a high speed and deposited on the substrate plate. For example, Derelizade et al.[89] used a suspended high-speed oxygen fuel spraying technology to deposit graphene nanoplates on stainless steel plates. Compared with the uncoated substrate, the friction coefficient was reduced by 7 times.

3.3. Powder metallurgy (PM)

PM technology uses graphene powder and metal matrix powder as raw materials for mixing, and then uses these mixtures for consolidation treatment in order to obtain composite materials.[90,91]

The preparation process and problems of PM technology are shown in Fig. 6. The PM process generally includes two main procedures: powder mixing and consolidation. These procedures are convenient and

Table 2
Comparisons of physical and mechanical properties of GRMMCs with those from literature.[10,60–80]

| Materials	Optimized amount	Preparation methods	Mechanical properties				
			Hardness/HV	Tensile strength/MPa	Yield Strength/MPa	Elongation/%	Refs.
Al-graphene	1060Al-1.5 wt% graphene	Deformation-driven metallurgy (DDM)	–	497	–	–	60
	Al-1.5 wt% graphene/Ni	PM and hot extrusion	65.3	–	204.5	–	61
	Al-0.5 wt% graphene	Field activated and pressure assisted synthesis	49.5	131	–	42	62
	AL-0.34 wt% CNPs/0.16 wt% Cu	PM	–	180	–	22.5	63
Al-CNTs	Al-Cu-4 vol% CNTs	PM	193.18	601	450	–	64
	Al-5 vol% CNTs	PM	137.1	–	–	2.45	65
Cu-graphene	Cu-0.3 wt% graphene	PM	51	208	–	17.5	10
	Cu-0.3 wt% graphene	PM	–	187	172	–	66
	Cu-0.2 wt% GNPs	SPS	108.6	233	171	23	67
	Cu-CNPs-Cu	Electrostatic self-assembly and electroless Cu plating	–	274	195	–	68
Cu-SiC	Cu-20 vol% SiC	High-pressure torsion	119	–	142	7.9	69
	Cu-40 vol% SiC	Sintering and forging	175	–	–	–	70
Cu-CNTs	Cu-Ti-0.2 wt% CNTs	PM	–	407	–	9.7	71
	Cu-2.0 wt% CNTs	Microwave sintering and rolling process	372	344	234	8.2	72
Mg-graphene	Mg-1Al-1Sn-0.18 wt% GNPs	PM	–	269	208	18	73
	Mg-0.25 wt% GNPs	DMD		202	122	14	74
Mg-CNTs	Mg-4.0 vol% CNTs	PM	70.3	265.5	–	6.3	75
	AZ31-0.1 vol% CNTs	Two-step dispersion	88	322	270	16	76
Mg-SiC	Mg-1Al-2.4 wt% SiC	SPS	56	262	147	–	77
	Mg-1 vol% SiC	Microwave sintering	43.2	203	157	7.6	78
WCu-graphene	W80Cu20–0.45 wt% Cu @ graphene	SPS	238	–	–	–	79
Ti6AL4V-graphene	Ti6AL4V-0.5 wt% graphene	SPS	472	–	–	–	80
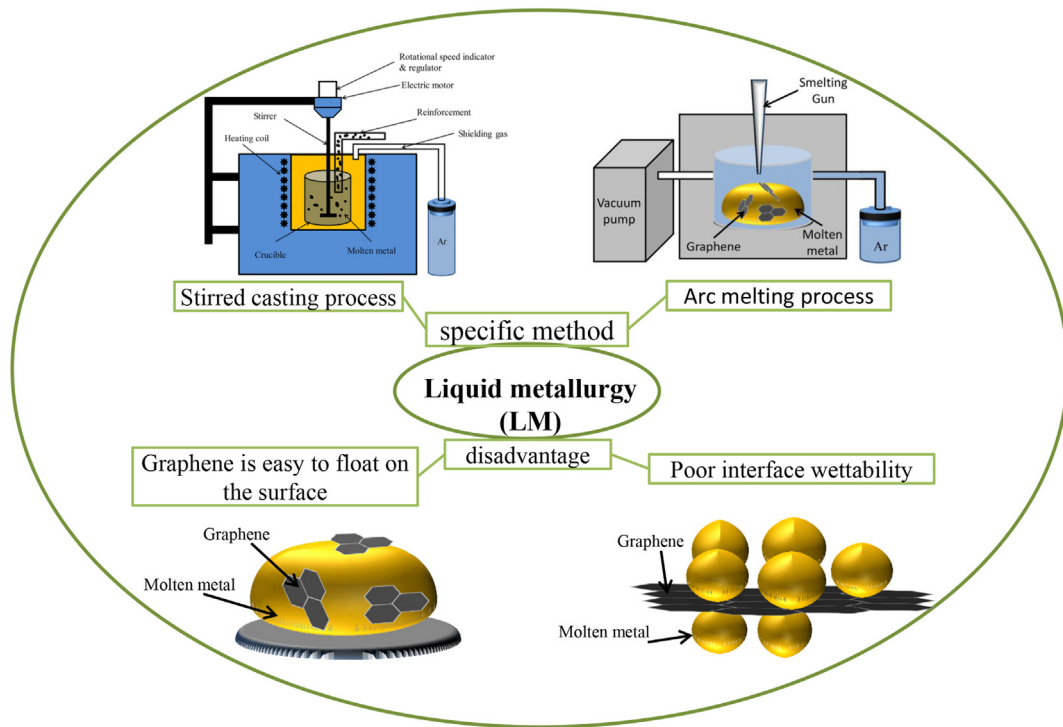
Fig. 4. Schematic diagram and disadvantages of LM technology.
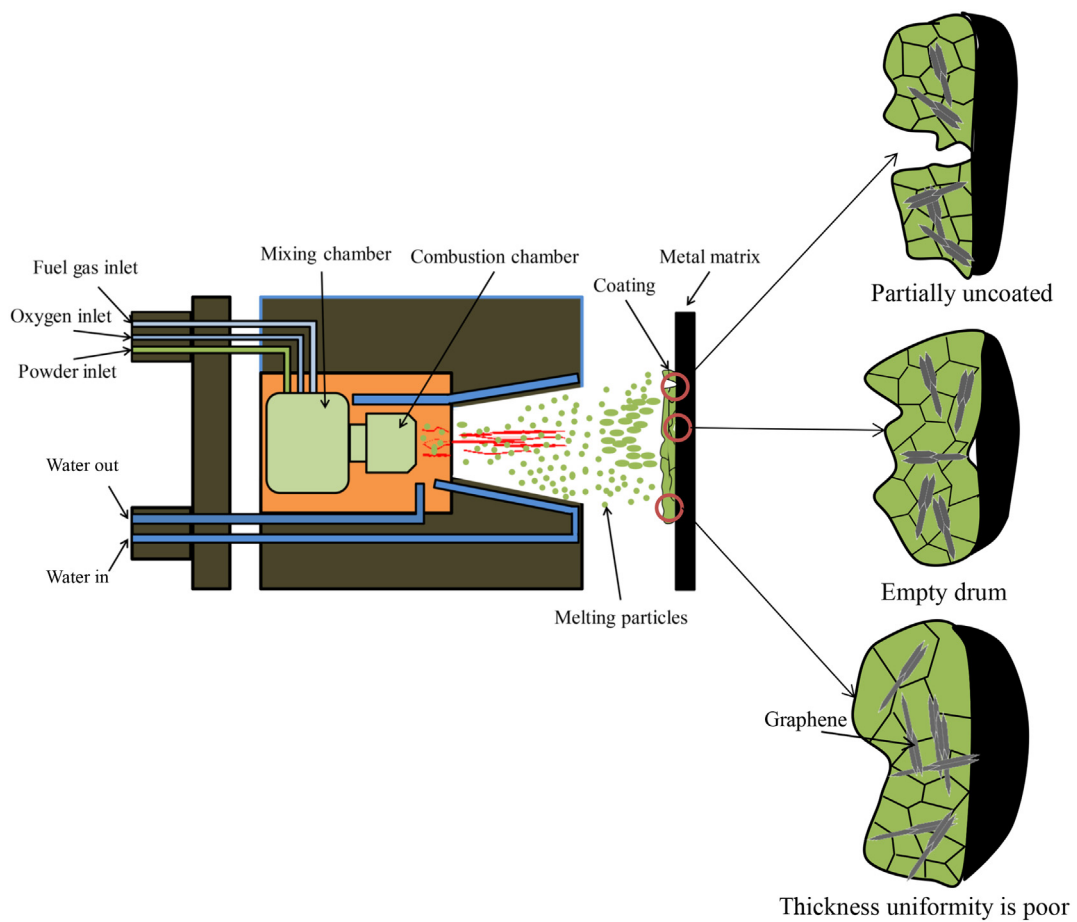

Fig. 5. Schematic diagram of TS process and existing problems.

simple to operate, so they are widely used in the preparation of composite materials. The powder mixing stage of PM technology has a significant influence on the dispersion of the graphene. The powder is often mixed by the ball milling method, in which a large deformation occurs during the powder mixing process, and repeated welding and fracture phenomena between the graphene and the metal matrix happen, thereby improving the dispersibility of graphene. However, this process will cause the fracture of graphene structures, thereby increasing the degree of defects and reducing the strengthening effect. Whereas a solvent mixing method uses organic solvents or deionized water to uniformly disperse the graphene inside the solvent under an ultrasonic agitation. The composite powder could be obtained using a freeze-drying method, and then the composite material is made using the sintering. However, this method has the problems of poor bonding strength between graphene and metal structures, and graphene is easily agglomerated during solvent removal process. At the same time, the composite materials prepared using PM technology inevitably have macro-defects such as pores, which reduce the density of the composite materials, thereby affecting the strength, electrical conductivity, and thermal conductivity of the GRMMNCs.

The sintering process can be divided into liquid phase sintering and solid phase sintering. The microstructure obtained by liquid phase sintering is denser and the carbon nanomaterials are easily dispersed. However, the high sintering temperature will result in coarsening of the matrix microstructure and the decrease of mechanical properties. Although the solid phase sintering can avoid the coarsening of the microstructure, the defect degree of the sintered body is quite high.

In recent years, many advanced sintering processes have been developed, which have the characteristics of low sintering temperature and short time, such as spark plasma sintering (SPS), laser sintering, and microwave sintering.[92–95] These newly developed sintering processes can effectively avoid the shortcomings caused by high temperature and long sintering durations in the traditional PM method. In some cases, sintering is not the final process, and further deformation processing is applied to improve mechanical properties of the composites (Fig. 6). These include hot processing and cold processing techniques such as hot/cold extrusion,[52] hot/cold forging,[53] hot/cold rolling[96] and equal channel angle processing.[97]

For example, Kady et al.[98] prepared Cu/WC-TiC-Co/graphene nanoplates (GNPs) composite using the PM method. When the content of GNPs was 0.25 wt%, the relative density of the composite reached its maximum value. Increasing the content of GNPs will lead to the agglomeration of graphene and generate a lot of pores. Sengupta et al.[30] used polyvinyl alcohol (PVA) as a dispersant to disperse graphene oxide (GO) and Fe powders uniformly before using the laser sintering technology.[99] The schematic diagram of this process is shown in Fig. 7(a). The PVA is evaporated from the base metal through the liquid phase sintering process, and the
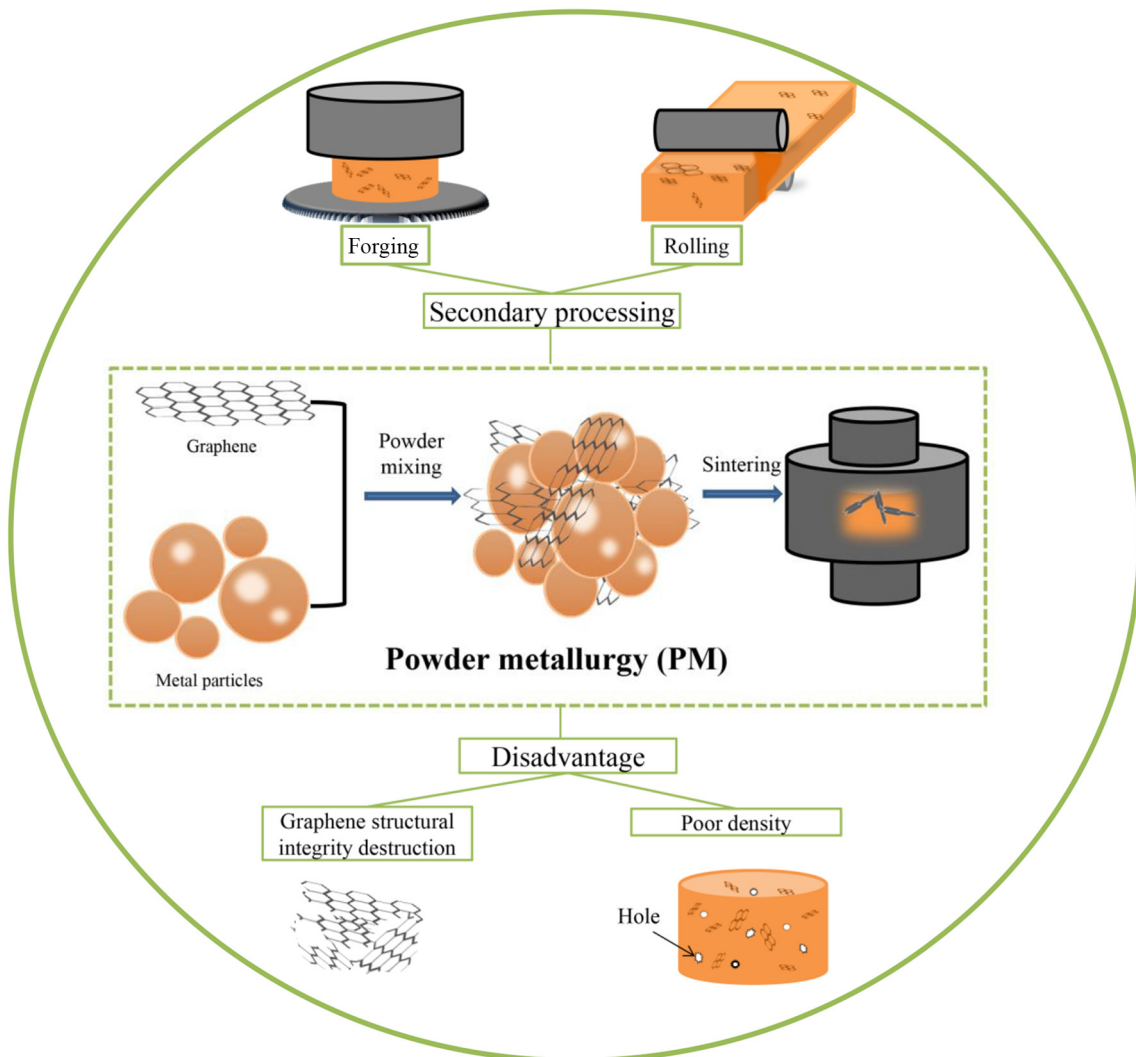

Fig. 6. Schematic diagram of PM process and existing problems.

alignment of GO along the vertical direction of the cross-section is shown in Fig. 7(b).

3.4. Surface coating deposition

(1) Electrochemical deposition (ED)

In order to improve the wear resistance and decrease the friction co-efficient of many components involved in contacts, friction and wear processes,[100,101] graphene is added to the surfaces of these components, which can increase the hardness and at the same time can effectively reduce wear due to its self-lubricating performance, thus increasing the service life of the parts. This could be more effective than many conventional methods such as surface lubricants. However, much research is needed to effectively apply graphene as the effective reinforcement phase of the metal matrix.[102,103] The ED technology can deposit a graphene-based composite film on the surface of the substrate to improve the wear resistance of the material.[104,105] During the process, graphene is combined uniformly with the metal ions in the electrolytes by mechanical stirring or addition of surfactant, and then the composite coating is deposited on the surface of the cathode substrate by pulse current or direct current methods.[106–108] Finally, the composite material is obtained by sintering, and the preparation process is shown in Fig. 8 (a). Recently, pulsed reverse current has been used as an excitation source, which can improve the dispersibility of graphene and improve interfacial bonding of graphene with metal matrix (Fig. 8b and c).[109] In Ref. [110], the GNPs/Cu composites prepared by ED and atmospheric protection sintering technology have shown significantly improved dispersibility of graphene and enhanced interfacial bonding.

(2) Chemical vapor deposition (CVD)

CVD is a commonly used method for large-scale production of graphene on metal substrates.[111,112]. Since graphene is a two-dimensional material, during CVD process, the spherical metal matrix powder is firstly subjected to treatments such as ball milling to make it into a flake powder. Then with the help of different carbon precursors, graphene can be formed on the substrate surface under either thermal heating or adding the metal catalysts (Fig. 9). Microwave,[113,114] magnetocaloric effect[115] and arc discharge[116] are often applied to generate plasma sources in the deposition chamber in order to promote the formation of high quality graphene in the CVD (or plasma enhanced chemical vapor deposition (PECVD)) processes. GRMMCs[117] can be obtained by in-situ preparation of graphene on metal powder or nano porous metal by CVD. Graphene has a good binding ability with metal matrix,[118] and can achieve uniform dispersion.[119] However, as shown

in Fig. 9, the surface defects of graphene are often increased due to the residual of carbon precursor in the CVD technology.[111] At the same time, the CVD process is generally carried out at a high temperature, which will cause the metal matrix crystal grains to increase significantly, and thus deteriorate the performance of the composite material.

Fig. 10 shows that Ni nanoparticles are in-situ combined with graphene using CVD method in order to prevent agglomeration of the graphene. In the composites, Ni nanoparticles are uniformly distributed on the surface of graphene.[120] In Ref.[121], the radio frequency plasma was used to enhance the CVD process, and the graphene layers are vertically produced on a 3D foam copper substrate,[121] which can be effectively used as a battery electrode material. Chen et al.[122] applied the CVD technology to in-situ produce graphene on the surface of copper powders, thus forming copper-based composite materials.

3.5. Other methods

In recent years, molecular-level mixing (MLM) technology has been widely used in CNTs reinforced MMCs.[123] Using the MLM process, the defects of the reinforced phase are signficantly reduced, which is helpful to synthesize high quality composite materials, such as GRMMNCs. Fig. 11 schematically illustrate the MLM process,[57] which include: (1) uniform dispersion of GO and metal ions in an aqueous solvent through chemical reactions to obtain composite powders; (2) reduction treatment to obtain rGO/metal composite powders; (3) consolidation treatment to obtain the final composites. For example, using the MLM technology, the rGO can be uniformly dispersed and form a strong inter-face bond through the oxygen-assisted bonds at the composite inter-faces of rGO and Cu, thus achieving a signficant enhancement effect.[124]

Friction stir processing (FSP) uses the heat generated by friction to soften metal and increase mechanical agitation to disperse the graphene into the metal matrix.[125,126] Fig. 12(a) shows a schematic diagram of the friction agitation process.[126] It is reported that the GNPs/Al6061 composite prepared by a multi-channel FSP was superior to the surface morphology of a single-channel FSP,[127] as shown in Fig. 12(b) and (c).

4. Different types of graphene metal matrix composites

In recent years, researchers have synthesized GRMMNCs based on the different metal matrices including Al, Cu, Mg, Ni, etc.[128–131] Different metal substrates will be combined with graphene with two variations: (1) the interactions between the above-mentioned interfaces are different; and (2) the interface lattice mis-matching is different. According to different interactions between graphene and the metal matrix (Table 3), there are two types of short-distance chemical adsorption and long-distance physical adsorption interactions. Table 3 lists the equilibrium
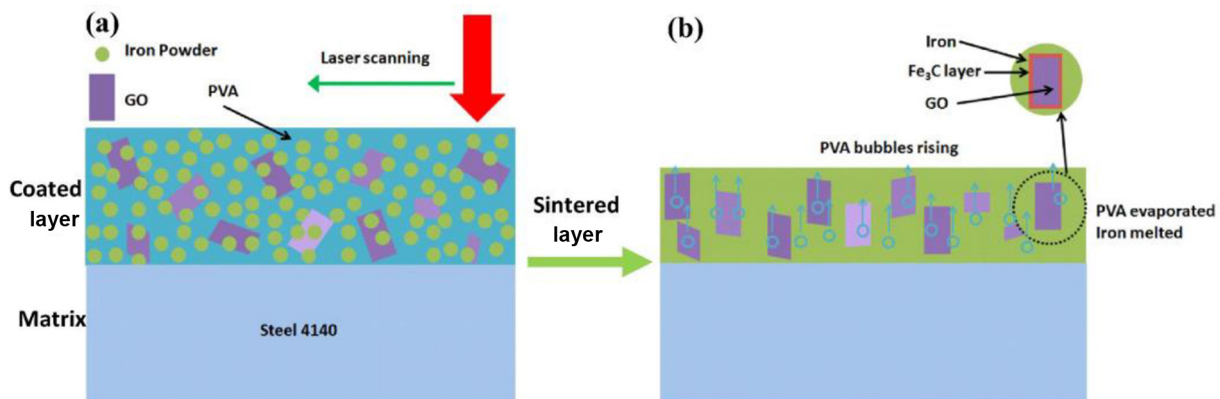
Fig. 7. (a) Schematic diagram of the GO and Fe powder mixed coating on the surface of the steel plate; (b) schematic diagram of the regular arrangement of GO during laser sintering. Reproduced from Ref.[99]
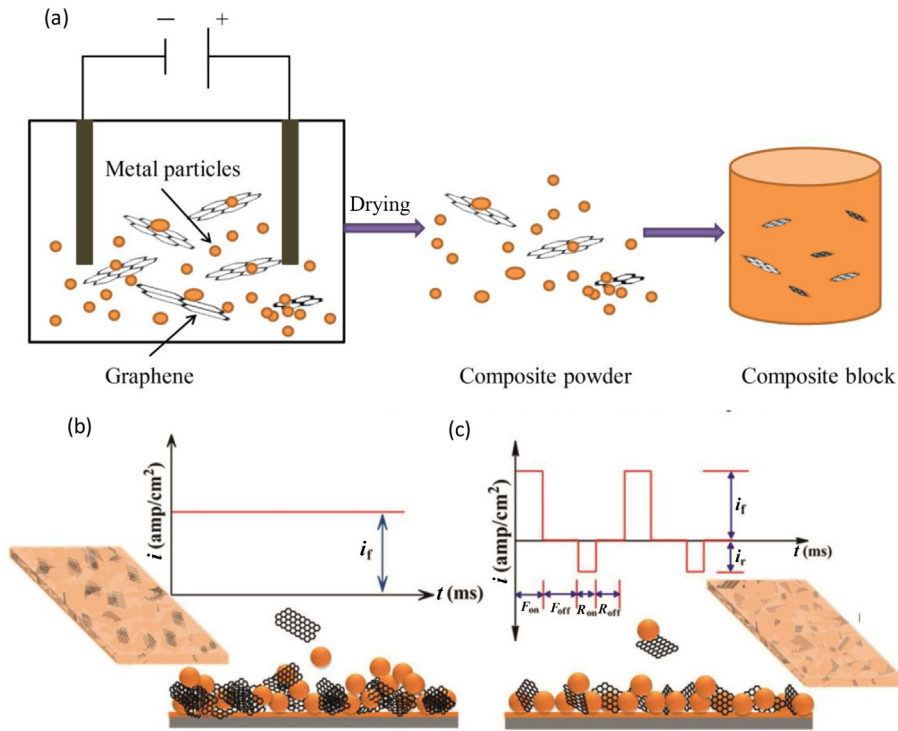
Fig. 8. (a) Schematic diagram of the preparation of GRMMNCs by ED and sintering; (b) schematic diagram of DC deposition (Reproduced from Ref.[109]); (c) schematic diagram of pulse reverse deposition (Reproduced from Ref.[109]).

atomic distances between the graphene and different metal substrates. For metals such as Cu, Al, and Ag, their equilibrium atomic distances with graphene are between 0.33 and 0.35 nm,[132] which are basically the same as the graphene layer spacing of 0.34 nm.[133] Therefore, the bonds between these metals and graphene are van der Waals forces which are very weak. However, as listed in Table 3, the equilibrium atomic distances between metals such as Ni or Ti and graphene are between 0.20 and 0.21 nm,[132,134,135] and the bonding is chemical bond which is very strong.

In the following sections, we will discuss the interactions and lattice matching degree of these metal-graphene interfaces with Al, Cu, Mg, Ni and other metals as the matrix.

4.1. Al-matrix composites

The advances of aluminum-based composite materials such as low density and good wear resistance make them having suitable applications in the automotive and aerospace fields.[136] However, there are
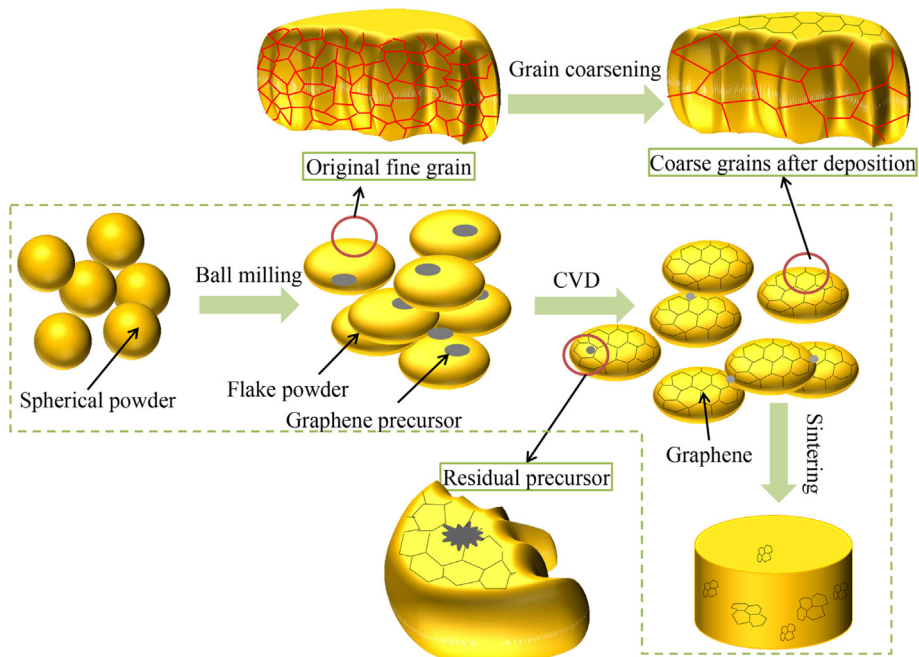
Fig. 9. Schematic diagram and disadvantages of CVD technology.
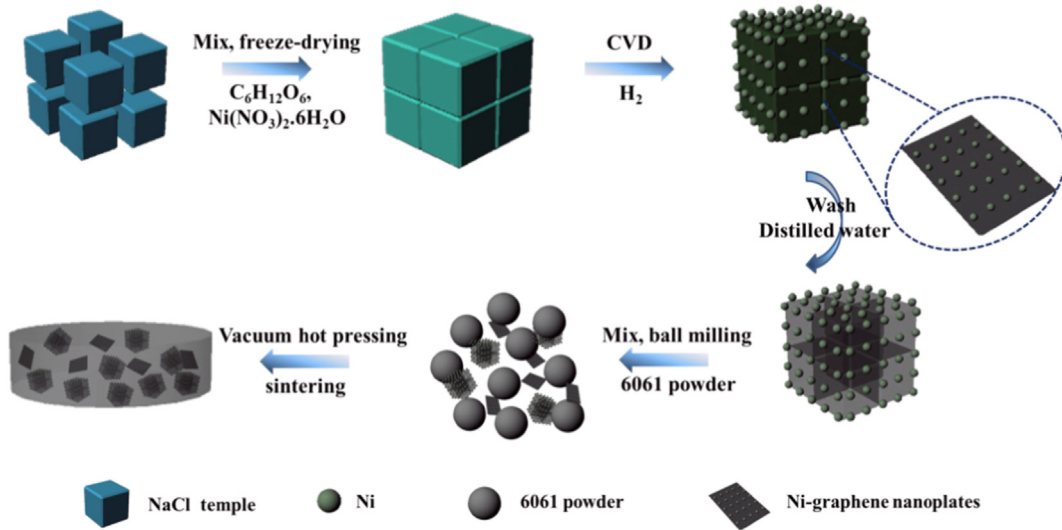
Fig. 10. Schematic diagram of preparation process for in-situ CVD, ball milling and hot pressing sintering of Ni@GNPs/Al composite block. Reproduced from Ref.[120]

requirements for these automotive and aerospace materials to be lightweight with good mechanical properties. The enhancement of traditional ceramic particles makes the aluminum matrix composites with much increased weight.[60,137] Graphene has small density, large surface area, excellent thermal conductivity, electrical conductivity and wear resistance, so it is regarded as an excellent reinforcement of aluminum-based composite materials.[138] It not only reduces the weight of the aluminum-based composite material, but also improves the mechanical properties of the aluminum-based composite materials. At the same time, its self-lubricating properties greatly improve the wear resistance of aluminum-based composite materials, which is unmatched by other reinforcing materials.[139]

As shown in Table 3, the equilibrium atomic distance between the graphene and Al matrix is 0.34 nm, so the interaction is quite weak at the physical adsorption interface. There are high degrees of lattice mismatch between the lattice of graphene and the lattice of the Al matrix, so a periodic lattice matching will occur and cause the generation of Moiré pattern, which leads to buckling of graphene.[132]

Affected by the van der Waals forces, the graphene layers are often agglomerated and form thick sheets, which generate a large number of defects at their bonding interfaces, thus the strength of composite is reduced.[140,141] Therefore, an effective increase in graphene dispersibility inside a metal matrix such as aluminum will surely enhance the strengthening effect of graphene in the GRMMNCs.[10,142] The composite interface without a strong interfacial reaction is only combined by physical bonds (for examples, van der Waals force and hydrogen bond) rather than chemical bonds (e.g., metal bond, ionic bond, and covalent bond). Fig. 13(a) shows different types of bonding between graphene and aluminum substrates. Among them, the composite interface is only bonded by van der Waals force, the Al ions are bonded by metal bonds, and the C atoms are bonded by covalent bonds. There are only weak van der Waals force between the graphene and Al matrix. On the other hand, wettability is also important for the adhesion of the composite interface. Increasing the wettability can increase the bonding surface between the graphene and Al, thereby improving the interface bonding.

Fig. 11. Schematic diagram of MLM process for preparing rGO/Cu composite materials. Reproduced from Ref.[58]

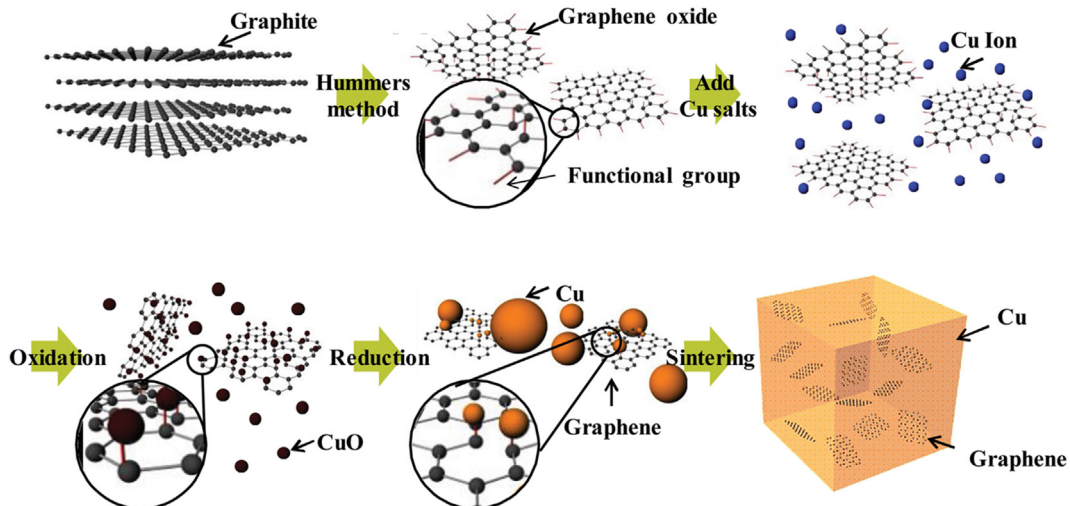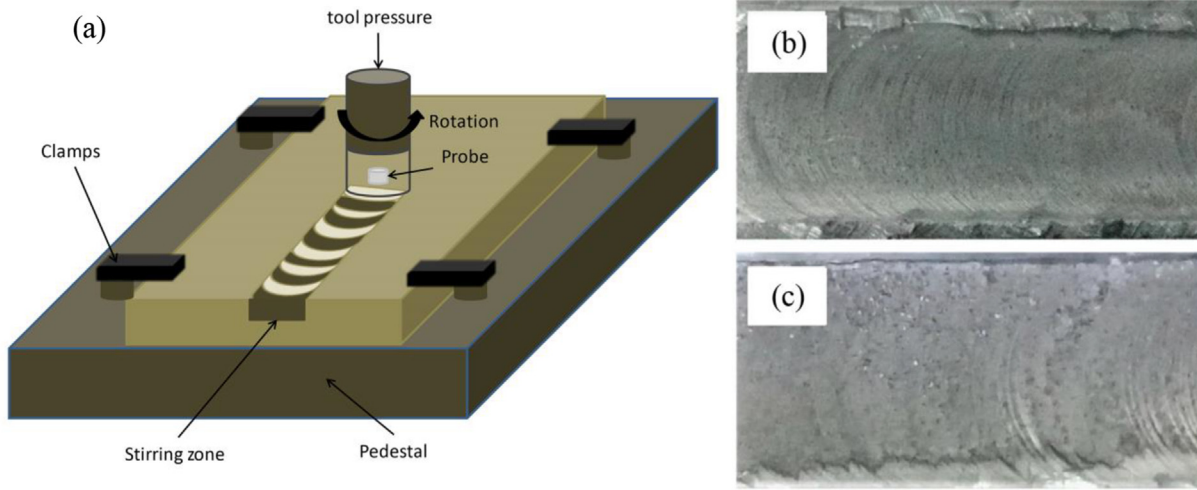**Fig. 12.** (a) FSP process diagram; (b) multi-channel surface (Reproduced from ref. [127]); (c) single channel surface (Reproduced from ref.[127]).

Table 3
The equilibrium atomic distances between graphene and different metal substrates.

Types	Substrate	Equilibrium atomic distances (nm)
Physisorbed metals	Cu	0.32–0.33
	Al	0.34
	Ag	0.33
	Au	0.33
Chemisorbed metals	Ni	0.20–0.32
	Ti	0.21
	Re	0.21
	Co	0.20–0.32

As explained in the above section, the ball milling process is one of the most commonly used methods for uniform mixing of powder, but the large deformation of the materials during the ball milling process will damage the structure of graphene and thus reduce the enhancement effect.[72]

In order to achieve a uniform dispersion of graphene in the Al matrix and improve the wettability between the graphene and Al, the existing solution is to modify the graphene surface, reduce the surface energy of
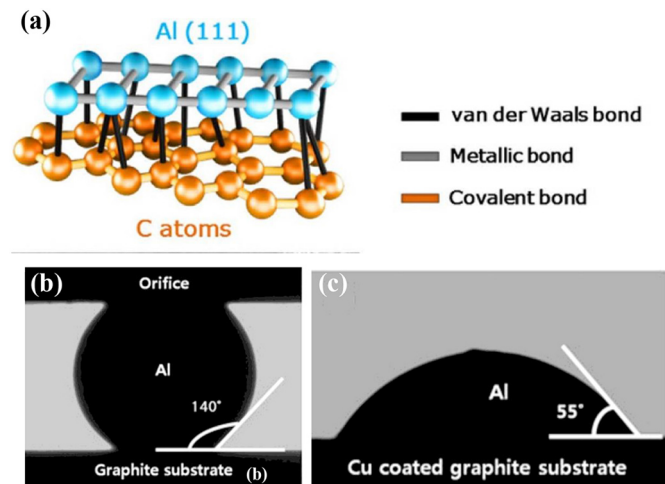

Fig. 13. (a) Schematic diagram of the graphene/Al bonding method (Reproduced from Ref.[39]); (b) graphite/Al is a wetting interface (Reproduced from Ref.[144]); (c) Cu-modified graphite/Al wetting the surface (Reproduced from Ref.[144]).

graphene, and improve the wetting between graphene and Al. For example, Ju et al.[143] used a water suspension mixing method to load Mg^{2+} ions on the surface of graphene, and then ball-milled the mixed powders. This is followed by an SPS process to form graphene/Al composites. The Mg^{2+} ions can reduce the surface energy of the graphene, and form a bridging effect connecting graphene and Al matrix, thus improving the composite's yield strength and tensile strength. Studies have shown that covering the surface of graphene with Cu can effectively improve the wettability between graphene and Al.[144] As shown in Fig. 13(b), the wetting angle between the unmodified graphite plate and Al is 140 °C, while the wetting angle at the interface between the modified graphite platelet and Al is decreased significantly to 55 °C (Fig. 13(c)).[39] This can improve the bonding strength of the Al matrix and the graphene interface.

Sun et al.[145] introduced Cu^{2+} ions into the Al-GO aqueous suspension, and then the positively charged Cu^{2+} ions and negatively charged GO were combined by electrostatic adsorption. The Cu^{2+} ions help to reduce the GO into rGOs. The composite materials were finally obtained by vacuum hot pressing sintering of Cu-doped rGO/Al composites. Doping with 3 wt% Cu^{2+} resulted in the best reduction effect of the GOs, and the microhardness of the obtained composites are 121% higher than that of Al substrate, and the compressive strength are 121% higher than that of Al. However, with a further increase of the doping contents of Cu^{2+}, the dispersibility, hardness and compressive strength are all decreased due to the significant agglomeration effects.

Ball milling process is also used to optimize the powder mixing process, a homogeneous mixing of graphene and Al powders can be achieved using a low-energy ball milling while ensuring the structural integrity of graphene.[146] It should be noted that the interface between graphene and Al is prone to interface reactions and formation of Al_4C_3 intermetallic compounds,[147] which are unfavorable for Al metal matrix composites.

$$4AL + 3C \leftrightharpoons AL_4C_3 \tag{6}$$

In addition to the mechanical properties enhancement of the Al matrix, the addition of graphene can also improve the friction properties. For example, Zeng et al.[148] reported that composite materials (rGO/SiC/Al—Si) were prepared using hybrid reinforcement materials through PM technology. The friction coefficient of the composite material is significantly lower than that of the Al—Si matrix. This shows that the self-lubricity of graphene can improve the wear resistance and reduce the friction.[149]

4.2. Cu-matrix composites

The development of copper-based composites is mainly driven by the requirement for improving low strength of pure copper metals. Additions of various reinforcements nanomaterials will increase the strength but often their conductivity values are decreased, which is unacceptable for many practical applications.[150,151] Graphene is regarded as the best material to simultaneously improve these material properties.[152,153] However, the interface wettability between Cu and graphene is generally poor, and their interface bonds are weak.[58,154] It is generally difficult to disperse graphene uniformly inside the Cu matrix.[155] Similar to that in the aluminum-based composite materials, the interaction between graphene and Cu is still weak and the lattice mismatch is large. The difference is that the Cu matrix does not chemically react with graphene to form metal carbides. It was reported that the graphene/Cu composite interface relies on oxygen atoms as

intermediate atoms to connect graphene and Cu matrix,[156] as illustrated in Fig. 14.

The problems for the graphene/Cu composites are basically the same as those for the graphene/Al composites. The only difference is that graphene and Cu matrix are not prone to severe chemical reactions, e.g., without formation of a lot of metal carbides. Generally speaking, the presence of metal carbides will affect the enhancement effect of graphene on the strength of the Cu matrix, while limiting the electron transfer rate and lowering the electrical conductivity and thermal conductivity. The methods for solving the problems including poor graphene dispersibility and weak interface bonding of graphene in the Cu matrix, and poor interface wettability are basically similar to those for the graphene/Al composites. These include graphene modified by loading metal particles, improving the preparation processes and using new technological methods such as molecular level mixing (MLM). For example, Zhang et al.[129] prepared GNPs/Cu composite
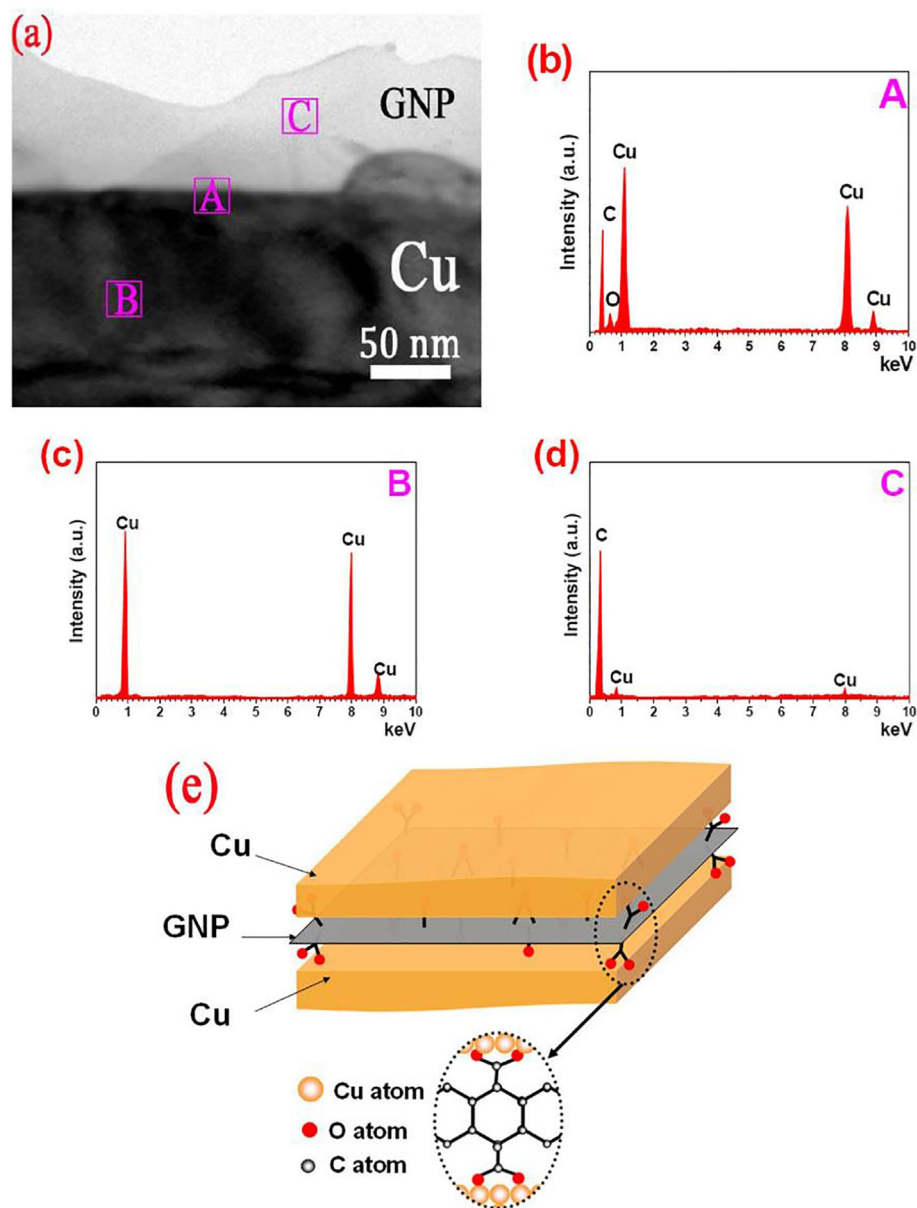
Fig. 14. (a) Transmission electron microscope (TEM) image of graphene/Cu composite interface; (b) energy dispersive spectrometer (EDS) diagram of area A in(a); (c) EDS diagram of area B in(a); (d) EDS diagram of area C in (a); (e) schematic diagram of GNP/Cu interface bonding. Reproduced from Ref.[156]. The TEM and EDS diagrams proved that the C—O—Cu bond is generated.

powders using MLM technology with NaOH solution, and then synthesized composites using the SPS method.

Molecular dynamics simulation has been applied to study the graphene/Cu composites,[157] and the results show that graphene can improve both the elastic modulus and yield strength of the composite. Simulations results show that the addition of graphene can significantly inhibit the expansion of cracks. Additionally, the plastic deformation of the composites was mainly shown as sliding along the surface of graphene, indicating that the interfacial properties of graphene and Cu have a dramatic influences on the overall performance of the composite. For example, our study shows that the use of rare earth element of cerium and metallic element of Ag to modify rGO and the use of SPS technology to obtain rGO/Cu composites result in a lower tensile strength than that of pure copper.[158] The reason for this result is largely due to the low interface connectivity between the graphene and the matrix, which is prone to cracking and failure during the stretching process.

4.3. Mg-matrix composites

Magnesium alloys have the advantages of low density and good damping performance,[159] but they have some disadvantages such as low strength and hardness, which seriously restrict further applications.[160] Current key research is to apply reinforcing materials to improve the mechanical properties of magnesium alloys.[161] The Mg matrix has a large lattice mismatch with graphene. The chemical nature of Mg is quite active, therefore, MgO will be formed during the preparation process, which can improve the degree of lattice matching. For example, Chen et al.[162] synthesized 1.2 vol% graphene nanosheets (GNS) reinforced Mg matrix composites using liquid ultrasonic treatment and solid-state stirring methods, and fabricated nanomaterials (CNTs, SiC, SiO$_2$ and Al$_2$O$_3$) reinforced Mg matrix composite materials using the same method. Among them, the GNS/Mg composite (66 kg/mm^2) has shown the highest microhardness, and the graphene and Mg substrates can be properly adjusted by optimizing the MgO at the interfaces, as shown in Fig. 15.

Adding graphene into the Mg matrix has shown a hardness enhancement effect. However, serious agglomeration of graphene and the poor interface structures is often the problem for the poor enhancement effect.[163,164] The current solution is mainly focused on optimizing existing processes or adopting new preparation processes. For example, after the conventional composite process (e.g., melting, PM, vacuum hot pressing, etc.), additional secondary processes (e.g., hot extrusion, hot rolling, etc.) are often used, and the yield strength and tensile strength of the obtained graphene/Mg composite material have been improved.[165,166] The wear rates of graphene/Mg composites are also found to be significantly lower than those of magnesium alloys without graphene, mainly due to the good self-lubrication effect of the graphene.[167]

4.4. Ni-matrix composites

Due to their high strength and hardness and good wear resistance, Ni-based composite materials are suitable for solid lubricating materials.[169] Due to its self-lubricating properties, graphene has been added into Ni matrix in order to improve mechanical properties such
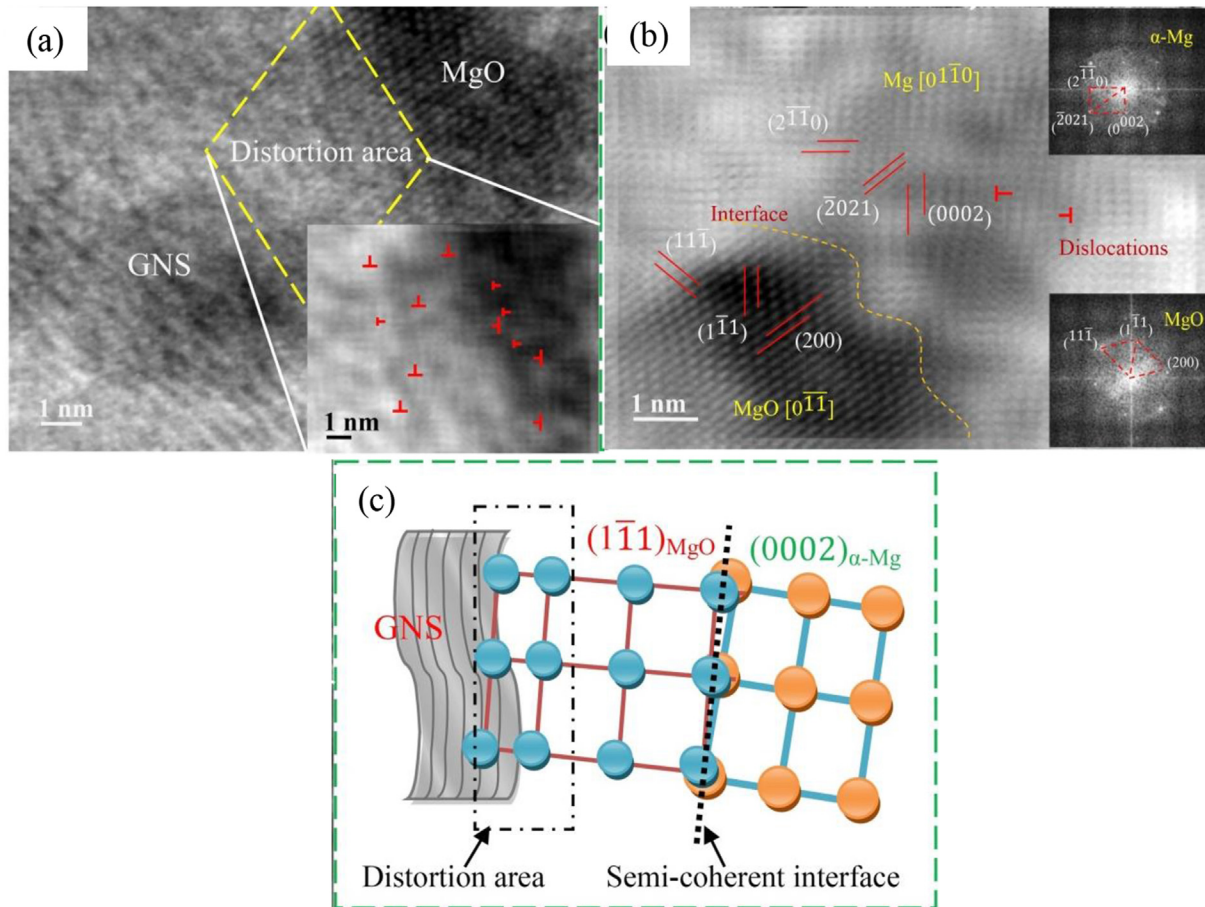
Fig. 15. (a) Inverse Fourier image of the high-resolution transmission electron microscope (HRTEM) of GNS/MgO interface, which shows that there is a high density of dislocations at the GNS/MgO interface. (b) Inverse Fourier image of the HRTEM of GO/α-Mg interface. It can be seen that the (1$\bar{1}$1) crystal plane of MgO and the (0002) crystal plane of α-Mg are arranged in parallel. Therefore, it has a high degree of lattic matching, and MgO and α-Mg are tightly bound. (c) Schematic diagram of the combined GNS/MgO/α-Mg interface. Reproduced from Ref.[168]

as strength and hardness, and corrosion resistance and wear resistance.[170] For example, Yasin et al.[171] studied the enhancement of mechanical properties and corrosion resistance performance of nanocomposite coatings of Ni/graphene nanocomposite coating. Hassannejad et al.[172] proposed an approach for deposition of nickel–graphene nanocomposite coating on copper substrates and reported that with the presence of graphene in the nickel coating, the wear rate was dramatically decreased.

4.5. Other types of metal-matrix composites

Ti matrix has a significant interaction with graphene, and the lattice mismatch between Ti and graphene is much larger than that between Ni matrix and graphene.[39] There is not only van der Waals force bonding but also ionic bonding between the graphene and Ti. This is because the outer d-electron orbit of Ti is not fully filled and thus ionic bonds are formed between Ti and the graphene's dangling carbon atoms, resulting in a relatively strong composite interface.[39] In previous studies, Dong et al.[173] confirmed that TiC is formed between rGO and Ti matrix at 1000 °C, which will increase the tensile strength, yield strength and

hardness of the composite. Recently, we observed three effective interfacial microstructures in GNPs/Ti-6Al-4 V composites and GNPs/CT20 composites, i.e. GNPs-Ti matrix, TiC-GNPs and TiC-Ti matrix, as shown in Fig. 16, leading to synergetic enhancement of strength and ductility of Ti matrix composites.[174,175]

Studies also show that the addition of graphene reduces the friction coefficient and wear rate of TiAl substrates,[176,177] which can be attributed to the self-lubricity of the graphene and the improvement of the hardness of Ti-based composite materials due to the addition of graphene.

The addition of graphene to metal substrates such as tungsten alloys and stainless steels can also enhance the strengthening effect of graphene. For example, we have added graphene into W70Cu30 composites and found that because graphene refines the grains of the matrix and hinders the movement of dislocations, the hardness and yield strength of the matrix have been significantly improved.[44] Mandal et al.[178] added graphene to 316L stainless steel and the hardness of the composite material was significantly improved, which is attributed to the good load transfer and thermal mismatch mechanism between the graphene and stainless steel.[179]
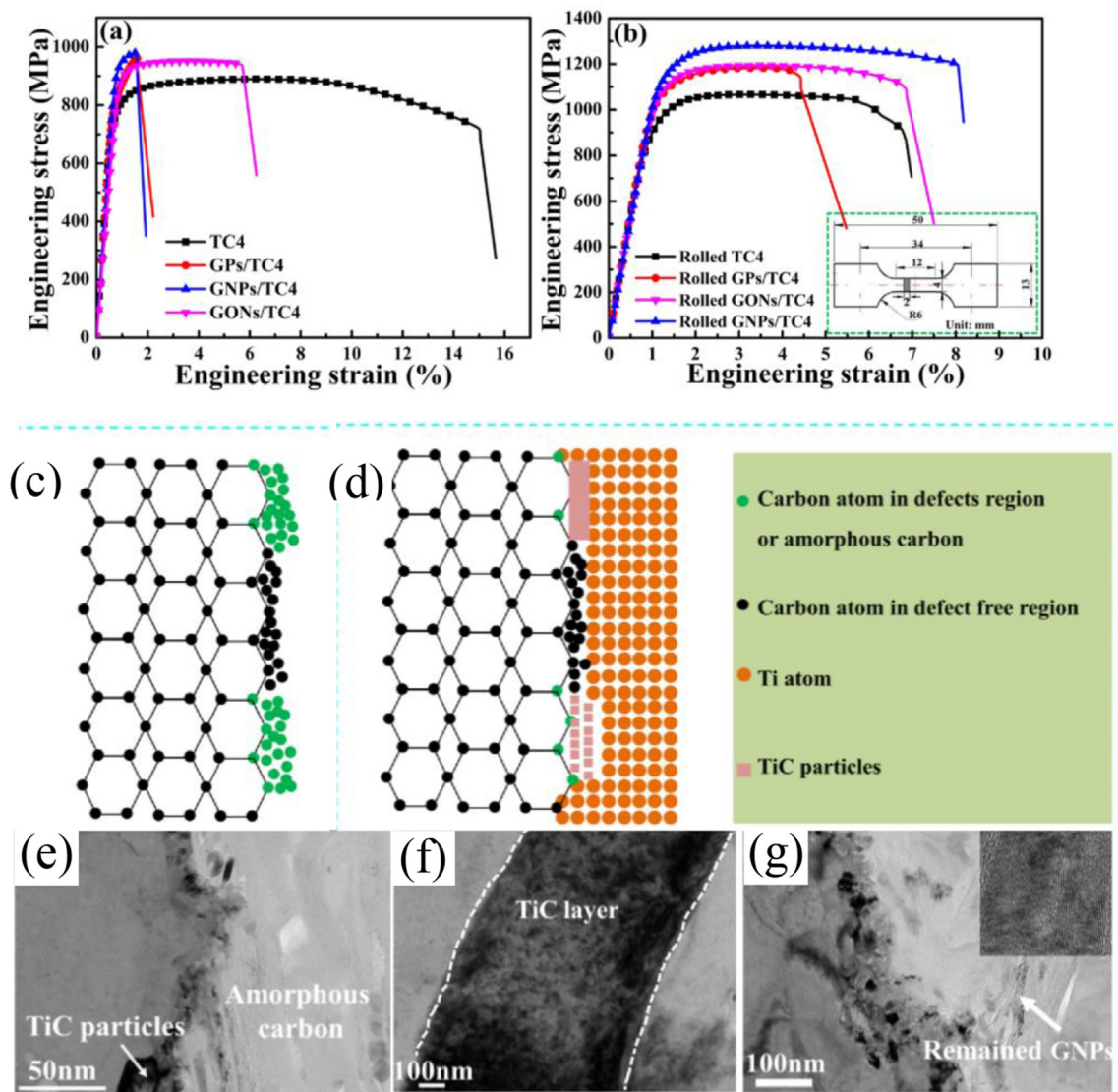
Fig. 16. (a) Engineering stress-strain curve of the composite material obtained after sintering. (b) The stress-strain curve of the composite material obtained after sintering and rolling. The stress-strain curve shows that the yield strength and tensile strength of GNPs/Ti-6Al-4V are higher. (c), (d) Schematic diagrams of the interface bonding mechanism between GNPs and Ti-6Al-4V. (e), (f) and (g) GNPs and Ti-6Al-4V interface bonding morphology. The morphology results show the formation of TiC. Reproduced from Ref.[174]

5. Modelling and computational mechanics of graphene composites

Modelling is popularly used to explore the enhancing mechanisms and understand the behaviour of materials, which then facilitates the design of structures or constructions. The model is able to predict the material or structure performance against external stimulants even before their manufacturing and applications.[180,181]

Over the years, there have been a number of analytical models to produce appropriate predictions of the properties of reinforced composites. Analytical techniques could be divided into four categories: phenomenological models (e.g. rule of mixture (ROM)[182]; Reuss model[183]), semi-empirical models (e.g. modified rule of mixture (MROM)[180]; Halpin–Tsai model[184]; Chamis model[185]), homogenization models (e.g. Mori–Tanaka model[186]), and self-consistent model[187].

In practice, to determine upper and lower bounds of properties, two distinct situations where the nanocomposites are considered to have uniform strain and uniform stress are investigated. In the case of uniform strain, it is found that Young's modulus E_c of the composites is given by the rule of mixtures.[188]

$$E_c = v_g E_{eff} + v_m E_m \tag{7}$$

Whereas in the case of uniform stress, the Young's modulus of the composite is given by

$$E_c = \frac{E_{eff} E_m}{v_m E_{eff} + v_g E_m} \tag{8}$$

where E_{eff} is the Young's modulus of the multilayer graphene filler that depends only upon its structure;[26,188] E_m is the Young's modulus of the matrix, and v_g and v_m are the volume fraction of graphene reinforcements and matrix, respectively.

Taking into account the orientation of the reinforcing particles and their finite length, we can obtain:

$$E_c = K\eta v_f E_{eff} + v_m E_m \tag{9}$$

where η is length efficiency factor; K is Krenchel factor[189], and for randomly-oriented nanoplatelets, $K = 8/15$.[190]

Halpin and Tsai proposed a semi-empirical formula based upon the self-consistent micromechanics method of Hill that enabled them to predict the elastic behaviour of a composite for a variety of reinforcement geometries.[184,191] For the elastic modulus of random short fiber reinforced composites,[192,193] by taking into account the distribution of graphene and gravity ratio, we can obtain:

$$E_c = [3/8 \times (1 + 2/3\eta_L pf)/(1 - \eta_L f) + 5/8 \times (1 + 2\eta_T f)/(1 - \eta_L f)]E_m \tag{10}$$

$$E_{\parallel} = (1 + 2/3\eta_L pf)/(1 - \eta_L f)E_m \tag{11}$$

in which E_c is the elastic modulus; $\eta_L = (E_g/E_m - 1)/(E_g/E_m + 2/3p)$; $\eta_T = (E_g/E_m - 1)/(E_g/E_m + 2)$; subscripts m, g, c and II represent the matrix, graphene, random graphene distribution and uniaxial orientation of the composite material, respectively; p and f are graphene diameter ratio and volume fraction, respectively.

The behaviour of composites could be modelled by using a macromechanical approach based on introducing the experimentally determined properties of composite material into an analytical tool, e.g. finite element analysis (FEA). Or this can be realized by using a micromechanical approach which considers the properties of each individual constituent, thus allowing determination of the composite properties for any volume fraction or any orientation prior to manufacturing of the composite itself and without the cost of experimental testing of the materials. It can, therefore, assist in designing the composite materials as well as the structures comprising of them.[194,195]

Multiscale modelling of composites refers to a bottom-up modelling strategy of simulating their behaviour through multiple time and/or length scale. The strategy begins with the in situ measurement of the constituent mechanical properties at the microscale level to build up a ladder of simulations, which take into account the relevant deformation and failure mechanisms at different length scales all the way to global/structural scale.[196,197] Multiscale modelling approaches could be further classified into a number of micromechanics models, ranging from simple fully analytical equations to a numerical FE-based approach to provide the global FE model with the effective behaviour of composites.[198]

Numerical modelling is a reliable tool, but unlike analytical models, it is time-consuming due to the definition of the geometrical dimensions.[198,199] There are various numerical techniques for solving practical engineering problems with a reasonable degree of accuracy such as the finite elements method (FEM), the finite difference method (FDM), and the boundary element method (BEM).[180] However, numerical FEM modelling is the most widely used for investigating and predicting the effective properties of composites.[198,200,201]

For example, Song et al.[202] used FEM to establish three-dimensional models of two different lateral scales (0.23 μm and 1.1 μm) when exploring graphene aluminum matrix composites. The results show that the engineering stress-strain curve predicted by the numerical simulation is basically consistent with the experimental measurement, as shown in Fig. 17(a) and (b). The morphology of the tensile fracture predicted by observation is in a good agreement with the experimental one, as shown in Fig. 17(c) and (d).

6. Conclusion and perspective

6.1. Current research trends and problems with graphene enhanced MMNCs

6.1.1. Applications and industrial relevance of GRMMNCs

Graphene market size is growing steadily in recent years. It is expected that among other types of graphene, graphene nanoplatelets will dominate the market share of graphene derivatives in the next 5–10 years due to their immense potentials as reinforcement materials for the development of lightweight and cost-effective composites with superior mechanical, thermal, and electrical properties.[203] Even though no commercial applications or commercial products based on graphene nanocomposites have been reported,[204] there are several potential applications including engineering, electronics, medicine, energy, industrial, household design etc.[205] For example, Cu-graphene composite films could be used for electro-friction applications such as brushes in generators and motors or heat sink structures for electronic devices.[204] Graphene–metal composites (e.g. those composites based on Fe—Ni, Ag, Au, Pt Pb, and Cd) are often used as catalysts, photo-catalysts, energy storage and transformation materials and biosensors.[206]

Until now a significant focus has been devoted to commercially producing thermoplastic and thermosetting systems, however, GRMMCs represent a promising materials sector.

6.1.2. GRMMNCs in aeronautical and automotive structures

Aviation and aerospace industry is the foremost adopters of advanced composite materials. With the exploitation of GRMMCs and their potential uses, future integrated approaches could be realised for structural aircraft components, e.g. thermoelectrical, lightning strike protection, fire and water barrier, and de-icing systems to keep aircraft parts ice-free, without affecting aerodynamic properties. Many projects are already in development[207,208] to increase the technology readiness level (TRL) of graphene-based systems and to reach highly integrated components. With the increasing number of automotive-grade graphene suppliers, many projects and companies in the automotive sector are also seeking to enhance their TRLs and increase their adoption of graphene-based components in new applications including electronics, thermal management and structural uses.
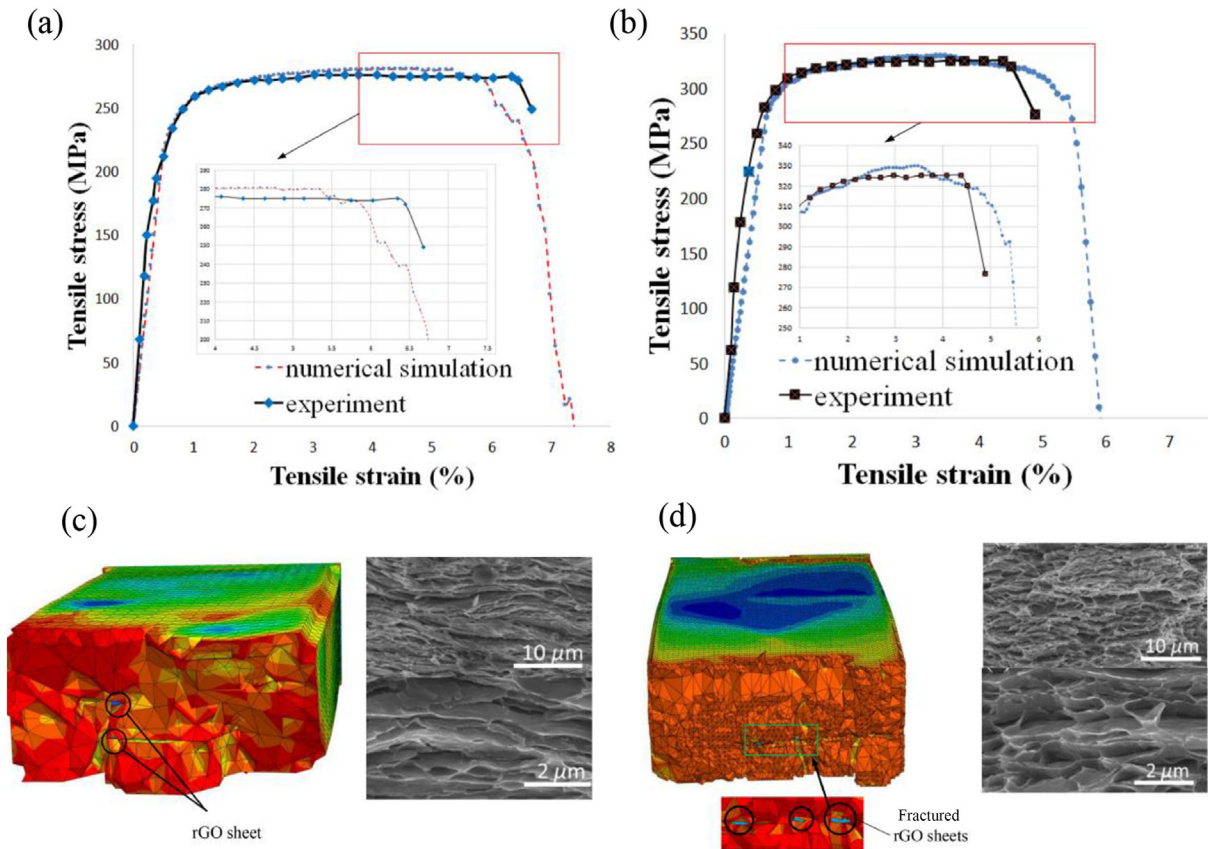
Fig. 17. (a) 0.23 μm rGO/Al simulation and experimental stress-strain curve; (b) 1.1 μm rGO/Al simulation and experimental stress-strain curve; (c) 0.23 μm rGO/Al simulation and experimental fracture morphology; (d) 1.1 μm rGO/Al simulation and experimental fracture morphology. Reproduced from Ref.[202]

6.1.3. GRMMNCs applications in construction

In the construction domain, graphene is generally viewed as next-generation materials. Graphene could be used in construction as a coating for steel or could be incorporated into traditional concrete production to substantially improve its characteristics. Although many constructional and functional benefits could be achieved through the addition of graphene to the suitable composite systems, e.g. enhanced fracture toughness, increased strength and stiffness, superior erosion resistance, the incorporation and processing techniques for these materials have not yet reached the same level of maturity. However, the near future looks for their promising applications.

6.2. The problem of graphene-enhanced MMNCs

Current research shows that graphene has the following major problems when enhancing MMNCs: (1) severe agglomeration of graphene, (2) weak interfacial bonding and (3) poor structural integrity of graphene.

A large number of studies have shown that during the recombination of graphene and metal matrix,[34,66,209] the phenomenon of its agglomeration is quite serious. The reason for this is that the surface energy of graphene is very large, causing the graphene sheets to spontaneously agglomerate by van der Waals forces to lose its inherent properties, which in turn deteriorates the mechanical properties and electrical properties of the metal matrix.

Wettability is a critical issue when the molten liquid is generated during the compounding process, which determines the strength of the composite interface. The wetting effect has a crucial influence on the performance of the composite material during the fabrication process. For a liquid on the surface of an ideal solid, the relationship between the wetting angle and the surface tension can be expressed by Young's equation.[210]

$$\gamma_{SV} = \gamma_{SL} + \gamma_{LV} \cos \theta \qquad (12)$$

$$\cos \theta = (\gamma_{SV} - \gamma_{SL})/\gamma_{LV} \qquad (13)$$

where θ is the wetting angle; γ_{SV}, γ_{SL} and γ_{LV} are the surface tensions of solid–vapor, solid–liquid and liquid–vapor, respectively. The smaller the wetting angle, the better the wettability between the droplet and the solid. When $\gamma_{SV} > \gamma_{SL}$, it indicates that the droplet can be wetted. Specifically, when $\cos \theta = 1$, the droplets have the best wetting effect and can be completely wetted. When $\cos \theta > 0$, the wetting partially occurs. When $\cos \theta < 0$, the surface is not wetted. The research shows that the wettability of graphite and molten metal is very poor.[211] For example, the wetting angle of graphite and Al is 140°−160°, that is, $\cos \theta < 0$ shows no wetting.[144]

High temperature and high pressure are generally the critical conditions for the preparation of composite materials, however, during these processes, the inherent structure of graphene could be seriously damaged, thus reducing their reinforcement effect.[212]

6.3. General methodologies to solve the problems of graphene enhanced MMNCs

For the above problems, researchers have developed a large number of solutions. It can be divided into process optimization method and graphene surface modification. These methods have improved the problems of GRMMNCs to a large extent and improved the

enhancement effect of graphene, but the conventional methods still cannot completely solve the existing problems. The following sections will introduce the solutions and their pros and cons.

6.3.1. Process optimization

The process optimization includes applying some modfied methods in dispersion methods such as ball milling dispersion and ultrasonic dispersion.[213,214] Although the ball milling method can achieve the dispersion effect in the process, due to the large deformation, graphene will be destroyed, and the metal matrix is often oxidized, thus affecting the strengthening effect.[215] Therefore, in the process of ball milling, the precise control of ball milling time is helpful to control the reinforcing phase and avoid a lot of defects.[209] Alcohol or protective gas can be added to prevent oxidation during ball milling. Dispersion can be done in anhydrous ethanol under the action of ultrasonic, and the subsequent mixing and drying with metal matrix powder can obtain graphene/metal matrix composite powders.[216]

6.3.2. Surface modification of graphene

The surface modification of graphene includes metal modified graphene, rare earth modified graphene and other modifications (for example, surface functional groups can be obtained by using the derivative GO)[120].

(1) Metal elements can be added to graphene to form (metal@graphene) powder, which is then mixed with the base metal powder. Because the addition of metal elements can effectively reduce the surface activity of graphene and inhibit the agglomeration of graphene, the metal elements can effectively combine with the base metal. The interface between graphene and the base metal can be well realised by using the intermediate metal elements as the bridge.[120,217] For example, Our previous research[218] showed that coating a layer of Cu on the surface of graphene can be used to reinforce WCu-based composites. Results showed that when the graphene content obtained by SPS infiltration sintering was 0.8 wt%, the material showed high hardness and high strength while maintaining good electrical and thermal conductivity. Reza et al.[219] used Ag nanoparticles to modify the rGO, and the Ag/rGO powder and Cu powder were mixed by ball milling and SPS compaction. Their results showed that when the content of graphene was 1.6 vol%, the microhardness of Ag/rGO-Cu composite block reached the maximum value of 81 HV. The flexural strength of the Ag/0.8 vol% rGO-Cu composite reached 472 MPa. The modification of rGO by Ag improved the poor interfacial bonding between graphene and Cu matrix, and formed Ag—Cu bond to increase the enhancement effect of the reinforcing phase.

(2) Modification with rare earth elements. Because rare earth elements are effective auxiliary additives in other materials, researchers used the rare earth elements to modify graphene, thereby improving the dispersibility of graphene and the weak bonding interface with the matrix.[220] Rare earth elements have a high coordination number with graphene to form a coordination bond. This will reduce the interface energy and surface energy of graphene, and improve the dispersion of graphene. At the same time, the rare earth element has low electronegativity and large activity, and can reduce the defects at the composite interfaces and improve the interface bonding strength.[221]

(3) GO surfaces often have a large number of hydrophilic functional groups containing oxygen, which will make graphene oxide evenly dispersed in water or solution. The metal matrix powders can then be added to the active agent to obtain the solvent containing metal matrix ions.[222] In this way, the negative graphene oxides and the positive metal matrix ions have electrostatic adsorption, which can inhibit the agglomeration of GOs. For example, Gao et al.[10] prepared the negatively charged GOs using the improved Hummers method, then treated the copper powder surface with cetyltrimethylammonium bromide to make it positively charged. They prepared the GO/Cu powder by electrostatic self-assembly, and then formed the composites using the PM technology. It was found that the graphene in the Cu based composite prepared by this method had a good dispersion and no obvious agglomeration. The tensile strength and hardness reached the maximum value, and the thermal conductivity was as high as 396 W·m^{-1}·K^{-1}.

6.4. Prospects for the future development of GRMMNCs

In future research, the following directions can be explored and researched.

(a) Using modern rapid sintering methods such as microwave sintering and SPS to reduce the defects of composite materials and increase the density, as well as in-situ growth CVD and electroless plating technology.

(b) Improving the dispersion and distribution of graphene nanofillers. It is well known that when the material reaches the nanometer scale, its surface energy is very high. Therefore, the graphene nanofillers are difficult to disperse into the matrix. For example, functionalization or modification of graphene can be done before it is introduced into metal matrix.

(c) Improving interactions between graphene and the metal matrix. Improving the interaction between graphene and metal matrix can effectively improve the interface bonding strength, and thus improve the mechanical, physical and chemical properties of GRMMNCs. This is also one of the current research hot topics. For example, it is very important to search for new intermediate materials to optimize the bond between the matrix and graphene.

(d) Controlling reduction of graphene oxide in nanocomposites. GO has a simple preparation method and is low cost, so it is currently the most widely used graphene derivative, and the addition of GO to the matrix requires reduction treatment to obtain rGO. At present, the most commonly used reduction methods include thermal reduction and chemical reduction, but the reduction degree of these methods is not high. After the reduction, there are still oxygen-containing groups on the surface of GO, which leads to an increase in the degree of composite interface defects. Therefore, it is the current research trend of GRMMNCs to develop methods with high reduction degree and controllable reduction.

(e) Since graphene-reinforced metal matrix composites are still in the theoretical development stage, the use of numerical simulation to broaden theoretical research has broad research prospects.

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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