

Mechanical and tribological properties of nanocomposites incorporated with two-dimensional materials

Zhengjia JI^{1,2}, Lin ZHANG¹, Guoxin XIE^{1,*}, Wenhui XU³, Dan Guo¹, Jianbin LUO^{1,*}, Braham PRAKASH¹

¹ State Key Laboratory of Tribology, Department of Mechanical Engineering, Tsinghua University, Beijing 100084, China

² College of Mechanical Engineering, Qinghai University, Xi'ning 810016, China

³ Key Laboratory of Tribology, School of Mechatronics Engineering, Nanchang University, Nanchang 330031, China

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Abstract: In recent years, attempts to improve the mechanical properties of composites have increased remarkably owing to the inadequate utilization of matrices in demanding technological systems where efficiency, durability, and environmental compatibility are the key requirements. The search for novel materials that can potentially have enhanced mechanical properties continues. Recent studies have demonstrated that two-dimensional (2D) nanomaterials can act as excellent reinforcements because they possess high modulus of elasticity, high strength, and ultralow friction. By incorporating 2D nanomaterials in a composite, 2D nanomaterial-based composites (2DNBCs) have been developed. In view of this, a critical review of recent mechanical and tribological studies based on 2DNBCs has been undertaken. Matrices such as polymers, ceramics, and metals, as well as most of the representative 2D nanomaterial reinforcements such as graphene, boron nitride (BN), molybdenum disulfide (MoS₂), and transition metal carbides and nitrides (MXenes) have been included in this review. Their preparation strategies, intrinsic mechanical properties, friction and lubrication performances, strengthening mechanisms, influencing factors, and potential applications have been comprehensively discussed. A brief summary and prospects are given in the final part, which would be useful in designing and fabricating advanced 2D nanocomposites in the future.

Keywords: two-dimensional (2D) nanomaterials; composites; mechanical property; friction; wear; lubrication

1 Introduction

Over the past decade, two-dimensional (2D) nanomaterials, which refer to ultrathin nanofilms having a thickness of only a single or few atoms, have received increasing attention [1–6]. A number of studies showed that 2D nanomaterials have highly improved properties and can be incorporated in composites to meet different application requirements. Accordingly, novel 2D nanomaterials are very important for a sustainable future.

Since Lee et al. [7] reported graphene's outstanding

mechanical properties, research on other 2D nanomaterials has also attracted attention worldwide. Studies on various 2D nanomaterials have significantly increased. Therefore, it is not surprising that 2D nanomaterials have been employed as promising reinforcements for polymers, metals, and ceramics to produce composites with enhanced mechanical properties. More importantly, it has been observed that even a very small amount of 2D nanomaterial can lead to a significant enhancement in the mechanical properties. As shown in Fig. 1, the numbers of publications on 2D materials and 2D

* Corresponding authors: Guoxin XIE, E-mail: xgx2014@tsinghua.edu.cn; Jianbin LUO, E-mail: luojb@tsinghua.edu.cn

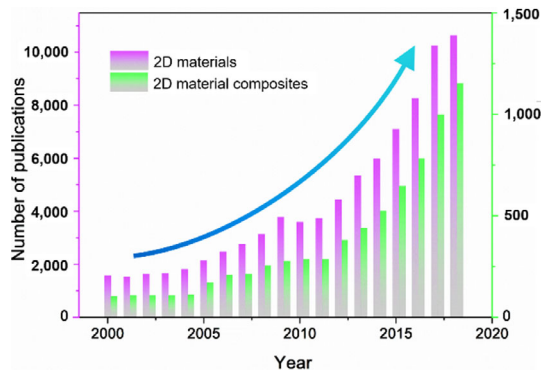


Fig. 1 Statistical data of articles on 2D materials and 2D material composites published during 2000–2018 based on the Web of Science database.

material composites have increased tremendously during the last decade. In view of the excellent mechanical properties and possibility for chemical modification, graphene has been applied in various fields ranging from aerospace to biomedicine. Inspired by the success of graphene-based nanocomposites, considerable effort has been devoted to incorporating other 2D materials in a number of composites as well as the exploration of their potential for various applications. Further, 2DNBCs have already attracted significant attention in many different fields. However, their mechanical and tribological applications have not yet been adequately explored. Recent studies have demonstrated that some novel ultrathin 2D nanomaterials possess excellent mechanical properties. For engineering applications, improving the mechanical properties is one of the most critical and challenging aspects in the technical advancement of these materials.

Many researchers are attempting to understand the reinforcing mechanism of composites with new 2D materials to improve their mechanical and tribological properties and achieve higher efficiency and longer durability for different engineering applications.

According to their chemical elements and atomic arrangements, all 2D nanomaterials can be typically classified into five categories: (1) Xenes, (2) transition metal carbides and nitrides (MXenes), (3) transition metal dichalcogenides (TMDs), (4) nitrides, and (5) organic frameworks. More specifically, (1) Xenes are composed of only one type of element, such as carbon, silicon, and phosphorous. A typical example

is graphene, which represents the carbon material of this type. Other examples are silicone, phosphorene [8], and borophene. (2) MXenes, where M is an early transition metal (e.g., Ti, V, and Mo) and X is carbon or nitrogen, are novel 2D nanomaterials discovered in 2011 [9–11]. (3) TMDs consist of hexagonal layers of metal atoms (M) sandwiched between two layers of chalcogen atoms (X) with a MX_2 stoichiometry, such as MoS_2 , MoSe_2 , and WS_2 . (4) A typical 2D nitride is hexagonal boron nitride (h-BN). (5) 2D organic frameworks include metal-organic frameworks (2DMOFs) and covalent-organic frameworks (2DCOFs) [12]. One of the most characteristic features of 2D organic frameworks is the presence of micropores or mesopores within the layer. All 2D nanomaterials have high in-plane strength, which is attributed to the in-plane covalent bonding [13]. The mechanical properties of some typical 2D nanomaterials are presented in Table 1. Notably, the modulus of phosphorene has been calculated along the different directions owing to its puckered structure.

Although a number of articles concerning 2DNBCs have been published, most of these papers focused on graphene-based nanocomposites [1, 21–24] and reviews regarding their mechanical properties have rarely been reported. This review intends to highlight the mechanical properties such as hardness, tensile strength, Young's modulus (Y), and tribological properties of composites reinforced with 2D nanomaterials from nanoscale to macroscale systems. As illustrated in Fig. 2, based on the elements of the matrices, the composites can be classified into three categories:

Table 1 Mechanical properties of some typical 2D nanomaterials.

2D nanomaterial	Thickness	Tensile strength (GPa)	Elastic modulus (GPa)
Graphene	—	125	~1,000 [14]
MoS_2	Monolayer	23 [15]	~270 [16]
h-BN	Monolayer	35 [17]	~865
Silicene	—	—	~82 [14]
Phosphorene	Monolayer	—	~106 (z) [18]
			~41 (a)
Ti_2C	Monolayer	—	~597 [19]
MOF	—	—	~5 [20]

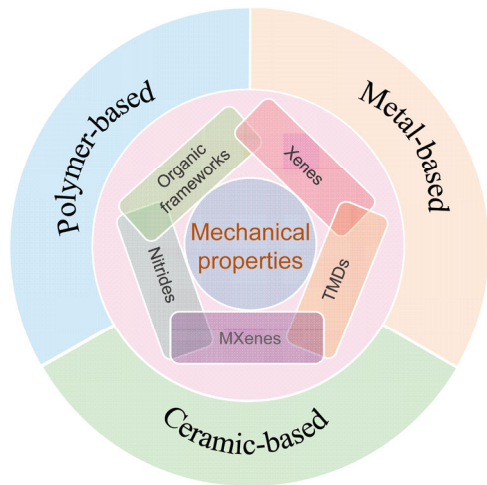


Fig. 2 Types of 2D nanomaterials and composites considered in this review.

polymer, ceramic, and metal composites. This review systematically describes the preparation and mechanical behaviors of various composites based on different matrices and 2D nanomaterials. More importantly, these 2DNBCs are promising for a variety of applications, such as aerospace, energy, and biomedicine, and they have potential to be utilized as structural materials and functional materials because of their excellent mechanical properties. Figure 3 depicts the basic framework of this review. In the following subsections, the preparation of 2DNBCs and the characterization methods of their mechanical properties are introduced.

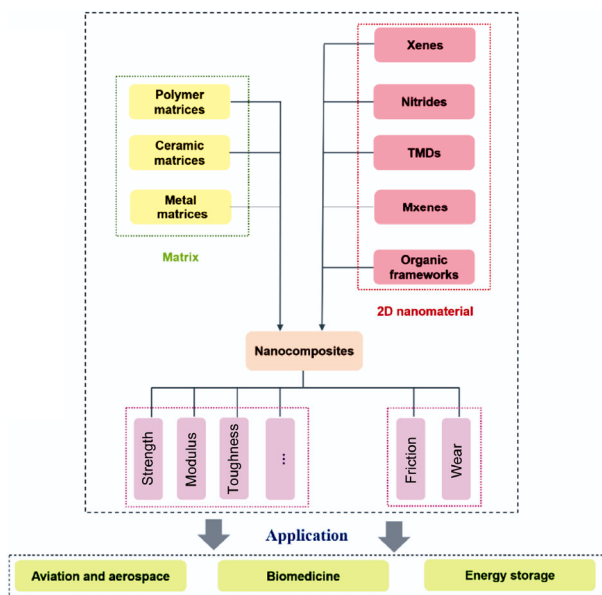


Fig. 3 Basic framework of this review.

2 Preparation methods

To achieve excellent mechanical properties, numerous methods, such as cold pressing, hot pressing, microwave sintering, powder metallurgy, and spark plasma sintering, have been developed for the preparation of 2DNBCs. Generally, all these methods can be classified into three categories:

1) For polymer composites, *in situ* polymerization, solution mixing, and melt blending [25, 26] are the most typical fabrication methods. *In situ* polymerization is a common approach for obtaining uniform dispersion of 2D nanomaterials in a polymer matrix. The 2D nanomaterials and the monomers or pre-polymers are swollen within the solvent and polymerization is then initiated by adjusting the conditions. Solution mixing is another method of incorporating 2D nanomaterials into a polymer matrix. The 2D nanomaterial and polymer can be dispersed easily in a solvent by ultrasonication or mechanical mixing and the solvent is then removed to obtain the composite. In this technique, the choice of a suitable solvent is a critical issue. There are many successful examples, such as epoxy (EP), polymethyl methacrylate (PMMA), polystyrene, and polytetrafluoroethylene (PTFE). Melt blending is performed by high temperature melting without a solvent. This method is generally used for fabricating thermoplastic composites. Recently, the resin transfer molding method has been presented to improve the dispersion of graphene in composites [27]. In this process, EP is incorporated into a three-dimensional (3D) graphene skeleton to prepare composites. Currently, research activities on 2DNBCs are mostly on composites reinforced with graphene or its derivatives. The preparation of other composites has also been achieved through similar strategies.

2) For ceramic composites, many methods have been used and developed, such as powder metallurgy [28, 29], reaction bonding [30], chemical vapor deposition/infiltration [31], sol-gel [32], and polymer infiltration pyrolysis [33]. In the powder metallurgy technique, 2D nanomaterials and matrix powder particles are mixed using ball milling to form composites. Commonly, hot isostatic pressing, vacuum hot pressure sintering, and spark plasma sintering are employed in the later stage. Compared with

other methods, the powder metallurgy method is cost- and time-effective and is the most promising. Reaction bonding and chemical vapor deposition/infiltration belong to gas phase processes. With these techniques, good dispersion of 2D nanomaterials in the matrix can be achieved. The sol–gel method can provide uniform dispersion of 2D nanomaterials, which are dispersed early in a molecular precursor solution. The composite can then be formed through the sol–gel reaction. Polymer infiltration pyrolysis is an important method by which ceramic composites can be fabricated from preceramic polymers. The 2D nanomaterials can be dispersed in a liquid phase preceramic polymer beforehand.

3) For metal composites, the fabrication methods are mostly similar to those described above. For instance, both powder metallurgy [34] and melt blending are the most popular methods. However, there are some special methods for fabricating metal matrix composites. Electrochemical deposition and thermal spray are often reported in Ref. [35]. The electrochemical deposition method is an attractive approach for fabricating composite films. As reported, metal injection molding is one of the special ways [35]. Recently, additive manufacturing and molecular-level mixing techniques have also

been developed [36, 37]. In a previous review by Naseer et al. [38], these preparation methods have been discussed in detail.

Based on the summary (Table 2), the dispersion of 2D nanomaterials in a matrix is a crucial step and a key challenge in the production of composites. In recent years, a variety of processing routes for dispersing 2D nanomaterial fillers into matrices, such as liquid phase blending, melt mixing, and freezing-dried masterbatch strategy, have been reported [30, 35–37].

3 Characterization methods

Mechanical properties are important factors in the structural and functional applications of composites. Many studies have focused on the qualitative and quantitative characterization of the mechanical properties of various composites [47]. The characterization methods frequently used in studying the mechanical properties of 2DNBCs are briefly described as follows.

3.1 Universal testing machine approach

The universal testing machine is a conventional

Table 2 Summary of preparation methods for 2DNBCs.

Materials	Method	Advantages	Disadvantages	Ref.
Polymer matrix	<i>In situ</i> polymerization	Excellent adhesion and dispersion	Complicated to operate, high cost, and only used in lab	[39]
	Solution mixing	Flexibility in various materials	Hard to remove solution	[40–42]
	Melt blending	Simple process, eco-friendly	Unsuitable for thermosetting resin	[43]
Ceramic matrix	Powder metallurgy	Easy operation, saving in time	Environmental pollution, destructive to 2D materials	[28, 29]
	Reaction bonding	Lower processing time, high densification	High energy consumption	[30]
	Chemical vapor deposition/infiltration	Less energy required	Difficult control in shape	[31]
	Sol–gel	Excellent dispersion	Only used in lab	[32]
	Polymer infiltration pyrolysis	Steady quality	Less raw material sources	[33]
Metal matrix	Powder metallurgy	Easy operation, low cost	Environmental pollution	[44, 45]
	Melting blending	Excellent dispersion of 2D materials	High cost, dangerous, destructive to 2D materials	[46]
	Additive manufacturing	Ability to form various shapes, easy to industrialize, and environment-friendliness	Poor densification	[36]

equipment for measuring the tensile, compressive, and shear strengths. It is worth pointing out that most of the composites can be tested by this approach according to the relevant standard. Different standards have been developed by different countries and organizations. Several studies have applied this approach to characterize composites including graphene/EP [47], h-BN/poly [2,2'-(p-oxydiphenylene)-5,5'-bibenzimidazole] (OPBI) [48], and graphene/Cu [49].

3.2 Dynamic mechanical analysis (DMA)

DMA is another typical strategy that has been extensively used to study the elastic modulus and loss factor of composites [50, 51]. It is most widely used for determining typical stress–strain curves and interfacial adhesion of polymeric composite systems. For example, the storage modulus and loss angle tangent of MoS₂/chitosan as a function of temperature were shown in Ref. [52]. The forced frequency and free resonance are two typical approaches for calculating the modulus and viscosity values. It has been found that the DMA method is more suitable for polymer composites [50].

3.3 Finite element approach

Calculation methods are also effective for evaluating the mechanical properties of composites. The modulus of 2DNBCs can be calculated by the Mori Tanaka equation [53] and the Halpin–Tsai equation [54] or the modified Halpin–Tsai equation [55, 56]. The modified Halpin–Tsai equation is as follows:

$$E_C = \left\{ \frac{3}{8} \left[\frac{1 + (2L_G / 3T_G) \eta_L V_G}{1 - \eta_L V_G} \right] + \frac{5}{8} \left[\frac{1 + 2\eta_T V_G}{1 - \eta_T V_G} \right] \right\} E_p \quad (1)$$

$$\eta_L = \frac{(E_G / E_p) - 1}{E_G / E_p + 2L_G / 3T_G} \quad (2)$$

$$\eta_T = \frac{(E_G / E_p) - 1}{E_G / E_p + 2} \quad (3)$$

where E_C is the Y of the composite with randomly oriented graphene, E_G is the tensile modulus of graphene, E_p is the tensile modulus of polymer, L_G and T_G represent the length and thickness of graphene, respectively, and V_G is the volume fraction of graphene in the matrix. η_L and η_T are dimensionless parameters.

$$E_{\text{para}} = \left[\frac{1 + (2L_G / 3T_G) \eta_L V_G}{1 - \eta_L V_G} \right] E_p \quad (4)$$

where E_{para} is the Y of the composites with unidirectionally distributed graphene. Apparently, the calculation method is an effective way of deriving the theoretical value of Y .

Based on these relevant equations, simulation methods, such as finite element models [57, 58], molecular dynamics [59–62], and 3D computational models [63], are developed into effective routes for predicting the mechanical properties of 2DNBCs, as displayed in Fig. 4.

3.4 Nanoindentation method

The nanoindentation method has been used since the 1970s and has become one of the most effective methods for determining the mechanical properties of composites since the early 2000s. Nanoindentation is a microscopic method that allows characterization through small-volume deformations. Atomic force microscopy (AFM) nanoindentation is one of the most effective ways for investigating nanomechanical properties such as normal hardness and elastic modulus [64, 65]. Based on force curves and the approach by Du et al. [66], the elastic modulus (E) of composites can be extracted from the following equation:

$$\frac{1}{E_r} = \frac{(1 - \nu^2)}{E} + \frac{(1 - \nu_i^2)}{E_i} \quad (5)$$

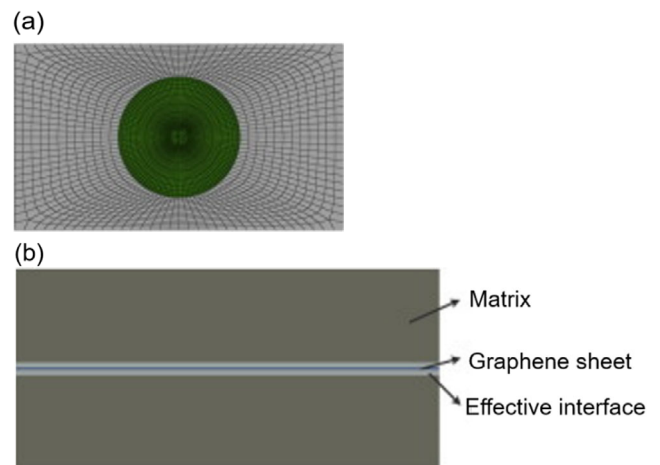


Fig. 4 (a, b) 3D computational model of the effective interface properties between a graphene sheet and polymer matrix. Reproduced with permission from Ref. [63], © Elsevier B.V. 2014.

where E_r and E_i are the reduced modulus and elastic modulus of the indenter, respectively, while ν and ν_i are the Poisson's ratios of the composite and indenter, respectively.

3.5 Tribological testing

As one important aspect of the mechanical properties of composites, the tribological properties have drawn much attention and need to be considered. To measure the coefficient of friction (COF) and wear rate, there are many different tribometers employing different test configurations such as pin (or ball) on disc, roller on plate, and block on ring [67, 68]. The COF is calculated from the frictional force and normal load. The wear rate is generally in the form of specific wear rate, which is calculated by the following equation:

$$W_s = \frac{\Delta m}{\rho L F_N} \quad (6)$$

where Δm and ρ are the mass loss and density of the composite, respectively, L is the sliding distance, and F_N is the normal load.

Since 2DNBCs consist of various matrices and 2D nanomaterials, their mechanical properties can be significantly altered by adjusting the chemical composition, microstructure, and content of 2D nanomaterials. The addition of very low amounts of 2D nanomaterials significantly improves the mechanical

properties of a matrix. Thus, the mechanical properties and behaviors of 2DNBCs are complex and challenging.

4 Mechanical and tribological properties

First, the main concepts should be introduced. In general, the interatomic bonds of composites mainly determine their mechanical properties [69]. Y , which is one of the most important mechanical properties, depends on the spring constant of the bond (k) and the interatomic distance (r_0). It can be expressed by the following formula: $Y = k/r_0$ [69]. It should be noted that the spring constant is a crucial term. For example, the value of the carbon–carbon bond is from 500 to 1,000 N/m and that of the metal or ionic bond is relatively lower [70]. If k of the material changes, the properties also change. Hence, the composite Y is greatly influenced by the matrix type and filler type as well as the interface bonding type between the matrix and filler (i.e., hydrogen bond).

In recent years, 2D nanomaterials such as graphene, BN, and MoS₂ [52] have been incorporated into a wide range of matrices for improving the mechanical performance. In the following sections, recent developments in the mechanical properties of 2DNBCs, from polymer composites to metal composites, are discussed. For comparison, the salient mechanical properties based on previous studies are summarized in Table 3.

Table 3 Summary of mechanical properties of 2DNBCs.

2D materials/the optimum content	Matrix	Tensile strength (MPa)/increase	Tensile modulus (GPa)/increase	Elongation at break (%)	Y(GPa)/increase	Bending strength (MPa)/increase	Ref.
Functionalized graphene (FGS) (0.75 wt%)	Polyimide (PI)	204.9/59%	2.93/62%	14.2	—	—	[71]
Graphene (2 wt%)	Polyvinyl chloride (PVC)	55/130%	—	40	2/58%	—	[72]
MoS ₂ (4 wt%)	Waterborne polyurethane (WPU)	28/140%	—	—	0.205/85%	—	[73]
Graphene (1 wt%)	Polyetheretherketone-carbon fiber (PEEK-CF)	160/60%	—	—	7/52%	—	[43]
Graphene (3 vol%)	PTFE	21.5/10%	—	—	2.62/223%	—	[74]
BN (1.0 wt%)	Al ₂ O ₃	—	—	—	—	432.8/58.6%	[75]
MoS ₂ (7 wt%)	Polyvinylidene fluoride (PVDF)	42/61.5%	—	470	—	—	[76]
Graphene (0.5%)	Cu	164/49.1%	—	—	—	—	[77]

(Continued)

2D materials/the optimum content	Matrix	Tensile strength (MPa)/increase	Tensile modulus (GPa)/increase	Elongation at break (%)	Y(GPa)/increase	Bending strength (MPa)/increase	Ref.
Graphene (0.5%)	Ti	1021/20.1%	125/14.6%	9.3	—	—	[44]
BN (2%)	Polyvinyl alcohol (PVA)	95/98%	—	—	—	—	[78]
BN (3%)	PMMA	—	—	—	5.11/130%	—	[79]
Graphene (0.05 wt%)	Cementitious	5/79%	—	—	—	—	[80]
BN (0.3 wt%)	PMMA	42/11%	—	—	2.13/22%	—	[17]
Graphene	Al	120/26%	—	—	—	—	[81]
BN (2.5 wt%)	Borosilicate glass	—	—	—	74	112	[82]
BN (0.2 wt%)	PVA	140/16%	—	—	3.4/36%	—	[83]
MoS ₂ (5 wt%)	PVA	105/24%	—	150	—	—	[84]
MoS ₂ with POSS (2 wt%)	PVA	58/57%	—	—	—	—	[85]
BN (0.5 wt%)	Cellulose acetate (CA)	115	7	—	—	—	[86]
BN (0.3 wt%)	PMMA	42/11%	—	—	2.3/22%	—	[87]
BN	PMMA	—	—	34.4	0.803/9%	—	[88]
Ni-GNPs (0.5 vol%)	Cu	281/24%	—	11.9	—	—	[89]
BN (1.5 wt%)	Styrene butadiene rubber (SBR)	48.7/150%	—	380	—	—	[90]
Reduced graphene oxide (rGo) (1.5 wt%)	Ni	948	—	12.1	—	—	[91]
Graphene (1.8 vol%)	PVA	42/150%	1.04/940%	98	—	—	[55]
Graphene (1 wt%)	Al5083 alloy	470/56%	—	3	—	—	[92]
Graphene	6063Al	277/23%	—	14.7	—	—	[93]
BN (2 wt%)	Polyarylene ether nitrile (PEN)	111/10%	—	2.75	—	—	[41]
rGO (1 wt%)	AlN	—	—	—	322/−10%	—	[94]
GO	Polyelectrolyte (PEC)	155/370%	—	9	4.41/270%	—	[95]
Graphene (5 wt%)	EP	62/−52%	4/48%	—	—	—	[96]
Graphene (2 wt%)	Polybutylene succinate (PBS)	37.2/21%	—	19	—	—	[97]
Silane-f-GO (0.1 wt%)	EP	80/45%	3.3/6%	—	—	—	[98]
Graphene (1.0 vol%)	Copper	32039%	—	—	132/61%	—	[99]
PBA–BN (0.3 wt %)	EP	71.9/54%	3.34/21%	—	—	—	[100]
Ti ₃ C ₂ T _x	NH ₂ –CF/EP	1210.9/40.8%	—	—	—	—	[101]
Graphene (0.1 wt %)	Ag/PVA	141.1/16.4%	—	—	0.701/126.9%	—	[102]
Graphene	Polypropylene (PP)	61.57/81%	—	19	2.314/100%	—	[26]

4.1 Polymer composites

To date, polymer composites enhanced with 2D nanomaterials such as graphene, MoS₂, BN, MXene, and MOF [103] have been widely reported and applied. In this section, the recent developments on

both strength and friction of 2DNBCs are described.

4.1.1 Strength

Most of the studies on polymer composites have been aimed at exploiting the high mechanical strength of fillers such as fibers [104, 105], nanoparticles

[106–108], whiskers [109, 110], and nanosheets [111–116]. Compared with other fillers, 2D nanomaterials have attracted more attention because of their unique planar structure and unconventional mechanical properties, as illustrated in Fig. 5. Herein, the critical strength of polymer composites incorporated with 2D nanomaterials is discussed in detail.

As presented in Fig. 6, the layered structure of typical 2D nanomaterials investigated by transmission electron microscopy (TEM) and AFM shows good dispersion in the composites. Four factors are responsible for enhancing the mechanical strength of polymer composites reinforced by 2D materials: (1) the large aspect ratio and high intrinsic mechanical properties of 2D materials [26], (2) the strong interfacial interaction between the 2D material and polymer matrix [26, 84, 118–120], (3) the homogeneous dispersion of 2D materials [78, 84, 118], and (4) the increased crystallinity [121].

As organic materials, polymers are highly flexible

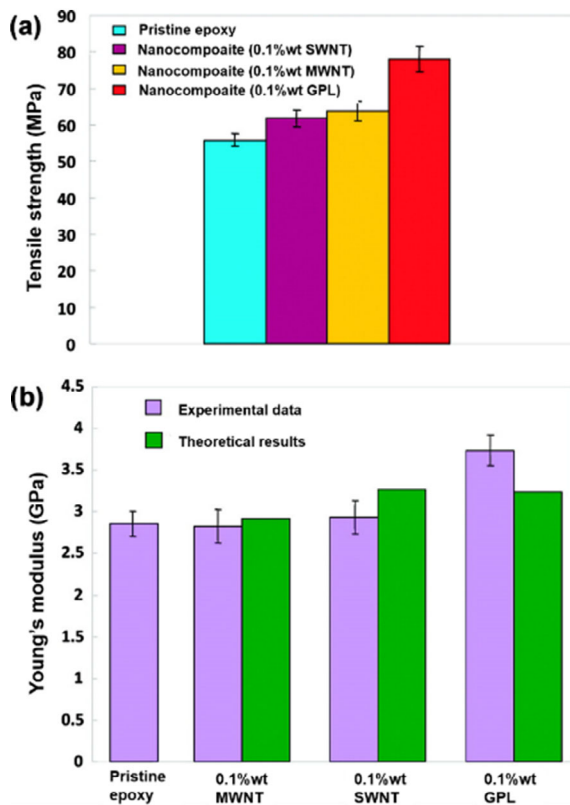


Fig. 5 (a) Tensile strength and (b) Y of pristine EP, single-walled carbon nanotube (SWNT)/EP, multi-walled carbon nanotube (MWNT)/EP, and graphene (GPL)/EP composites. Reproduced with permission from Ref. [117], © American Chemical Society 2009.

and have poor mechanical properties [122]. However, the addition of 2D nanomaterials significantly improves their characteristics. As a typical example, PVA was reinforced by various 2D nanomaterials including graphene [123], BN [78], MoS₂ [84], and Ti₃C₂T_x [124, 125]. The crystallinity of the PVA polymer was influenced by the addition of graphene or graphene oxide (GO) nanosheets, indicating that 2D nanomaterials act as nucleating agents in the matrix [26]. However, Wang et al. [97] reported that the crystallinity of polybutylene succinate (PBS) remained the same despite the presence of graphene. The addition of 2.0 wt% graphene in PBS increased the tensile strength and storage modulus by 21% and 24%, respectively. This is because, at room

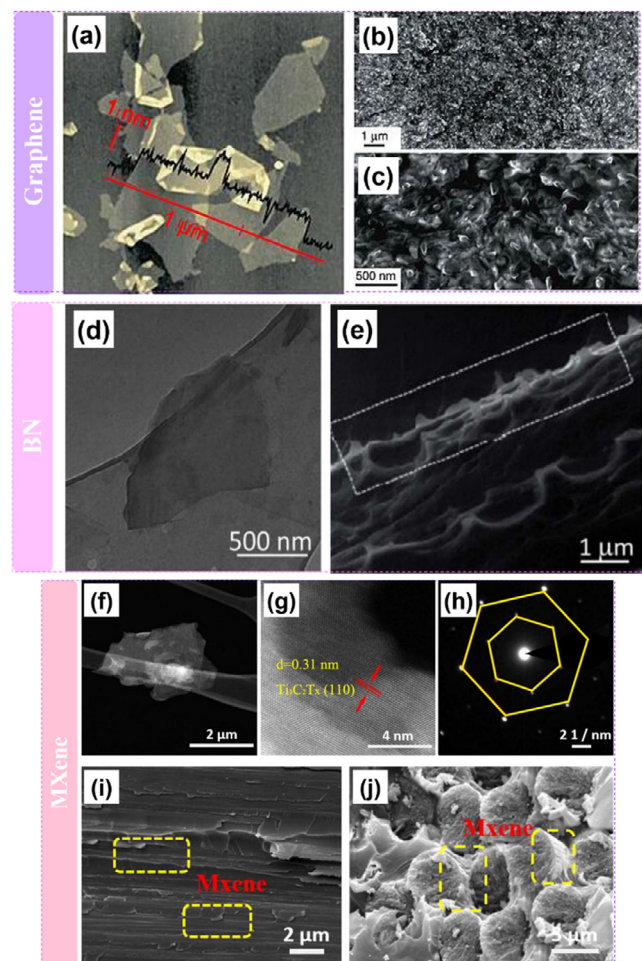


Fig. 6 Morphology images of (a) graphene and (b, c) its composite. Reproduced with permission from Ref. [1], © Springer Nature 2006. (d) BN and (e) its composite. Reproduced with permission from Ref. [83], © RSC 2009. (f–h) MXene and (i, j) its composite. Reproduced with permission from Ref. [101], © Springer Nature 2019.

temperature, the load transfer across the interface between the graphene and matrix is easily achieved owing to the large aspect ratio and uniform dispersion of graphene in the matrix. Since the crystallinity of PBS remained the same despite the presence of graphene, this could mean that the addition of 2D nanomaterials does not affect the crystallinity of composites and the improvement in mechanical properties depends mainly on the 2D material rather than on the matrix. The results are consistent with the shear lag theory, which describes the 2D nanomaterial behavior in a matrix [126].

The effect of graphene on the stiffness, strength, and toughness of OPBI was investigated by Wang et al. [51] through an *in situ* polymerization method. Later, significant progress of composites filled with other 2D nanomaterials has been reported. An appropriate concentration of the well-structured BN enables the enhancement of mechanical properties [48]. The addition of 4 wt% BN to OPBI has a similar effect on improving the tensile strength and modulus to that of filling with 0.4 wt% of graphene [48]. The difference in the amounts may be due to their intrinsic strengths ($\sigma_{\text{graphene}} \approx 130$ GPa [7, 127], $\sigma_{\text{BN}} \approx 85$ GPa [128]).

There are two main strategies for further enhancing the mechanical properties of polymer composites. First, the functionalization, using octa (aminophenyl) silsesquioxane, octadecylamine, fluoride, etc., is an efficient way [129–135], because both dispersion and stress transfer can significantly improve the strength. The functionalization of 2D nanomaterials is conducive to better dispersion [136]. The compatibility of the 2D nanomaterial and the matrix affects the interface energy transfer in composites [76]. Ramanathan et al. [2] added 0.05 wt% of single-layer functionalized graphene to PMMA and achieved an increase of 80% in elastic modulus and 20% in tensile strength. A similar result was obtained by Qiu et al. [137]. In addition, Xing et al. [138] also claimed that functionalized BN can enhance the mechanical properties of polymers. Another strategy is synergistic effect [102, 139–143]. Owing to the synergistic effect of the π - π interaction between the one-dimensional (1D) carbon nanotube (CNT) and 2D GO nanosheet as well as the strong interfacial interactions between the PEC matrix

and CNT-GO, the production of PEC/GO-CNT composites with higher tensile strength and Y is expected [95]. The incorporation of GO can improve the compressive strength of the PEC complex hydrogel [50]. The GO nanofiller bears most of the applied load in various states owing to its intrinsic high strength. Figure 5 displays a comparison between the tensile strengths and Y of pure EP and different nanocomposites. It is shown that the tensile strengths of the composites are higher than that of the pure matrix. On average, the strength increases by 20% to 140%. Note that even 0.5 wt% MoS₂ improved the tensile strength of chitosan to 207% [52]. Finally, it remains a challenge to understand the strengthening mechanisms of 2D nanomaterials in 2DNBCs owing to their diversity and complexity.

4.1.2 Friction and wear characteristics

Besides the mechanical strength, 2D nanomaterials can improve the tribological performance of polymers as well, because these 2D nanomaterials exhibit in-plane mechanical isotropy and a weak interlayer interaction [144]. The good tribological performance, which can be achieved by incorporating 2D nanomaterials in the matrix, comprises two aspects: (1) reduction in COF and (2) enhancement in wear resistance. This section discusses the effect of using 2D nanomaterials as fillers in a polymer matrix on the frictional properties. All the results mentioned for the COF and wear rates are summarized in Table 4. For comparison, various 2D nanomaterials added in the polymer matrix are presented [145–147].

The COF of PTFE is significantly reduced when the added graphene in the matrix is 4.0 wt% [148]. However, a higher content of graphene may compromise the lubrication property. The addition of graphene to a PTFE matrix decreases the wear rate by three or four orders of magnitude [157]. The friction reduction and improved wear resistance of the composite are attributed to the good dispersion and self-lubrication of 2D nanofillers [144]. Ultrahigh molecular weight polyethylene (UHMWPE) had been reinforced by graphene [158] or Ti₃C₂ [156]. The results showed that the 2D nanomaterial reduced the effective lateral force and increased the wear resistance. Further, 2D nanomaterials can

Table 4 Tribological properties of polymer composites reinforced with 2D materials.

Matrix	2D material	Method	Operating condition	COF	Wear rate ($\text{mm}^3/(\text{N}\cdot\text{m})^{-1}$)	Ref.
PTFE	4.0 wt% graphene	Cold compression and sintering	20 N, 0.1 m/s	0.18	7.5×10^{-6}	[148]
PTFE	10 wt% MoS ₂	Cold press and sintering	5 N, 0.12 m/s	0.15	2.5×10^{-5}	[149]
PTFE	10 wt% g-C ₃ N ₄	Cold press and sintering	5 N, 0.12 m/s	0.18	1.0×10^{-5}	[149]
PTFE	5 wt% phosphorene	Ball milling and SPS	3 N, 20 mm/s	0.041	6.9×10^{-6}	[150]
EP	0.5 wt% BN	Mechanical mixing	5 N, 20 mm/s	0.5	16×10^{-5}	[151]
PI	2 wt% BN	Mechanical mixing and spin-coated	3 N, 20 mm/s	0.10	2.79×10^{-6}	[152]
PEEK	10 vol% g-C ₃ N ₄	Mechanical mixing and hot pressing	50 N, 1 m/s	0.6	4.0×10^{-7}	[68]
Phenol formaldehyde (PF)	0.3 wt% Graphene oxide	Solution mixing	320 N, 2.24 m/s	0.12	—	[153]
Polyurethane	3 wt% MoS ₂	Solution mixing	3 N, 60 mm/s	0.10	9×10^{-5}	[145]
EP	1.0 wt% MoS ₂ @PPN	Mechanical mixing	80 N, 0.05 m/s	0.58	22.3×10^{-5}	[154]
Bismaleimide (BMI)	0.6 wt% PHbP@rGO/WS ₂	Solution mixing	196 N, 0.42 m/s	0.13	1.22×10^{-6}	[155]
UHMWPE	2.0 wt% Ti ₃ C ₂	Hot compression mold	200 N, 0.4 m/s	0.128	—	[156]
EP	2.0 wt% Ti ₂ CT _x	Mechanical mixing	98 N, 0.3 m/s	0.228	—	[39]

significantly contribute to the excellent mechanical properties, lubricity, and thermal conductivity [151]. These 2D nanomaterials also have potential to solve the frictional heat problems of polymer composites. Qiu et al. [154] employed an infrared thermal imager to investigate the melting behavior of composites. The results showed that the addition of MoS₂ reduced the frictional heat and retarded the melting wear. In Ref. [159], a 2D nanomaterial was also added in a base oil owing to its good self-lubricating property. Thus, 2D nanomaterials are promising fillers and can significantly enhance the tribological performance of polymer composites because of their good lubricating properties under harsh conditions [160].

To understand the wear mechanism of polymer composites incorporating 2D nanomaterials, many studies have focused on the analysis of the worn surface of composites (Fig. 7). The mechanism reported includes two different categories: (i) slightly abrasive wear [155] and (ii) melting wear [154]. Based on the observation of friction surfaces, many studies have demonstrated that the formation of a load-carrying transfer layer is the key factor for improving

the tribological properties of polymer composites (Fig. 8) [149, 150].

In summary, the friction performance of polymer matrices reinforced with 2D nanomaterials mainly depends on the type and content of the 2D nanomaterial as well as the interfacial bonding. As discussed above, the functionalization of 2D nanomaterials is an important method for strengthening the interfacial bonding. To further enhance the tribological performance, several challenges related to the interfacial bonding must be tackled urgently [40].

4.2 Ceramic composites

Similar to polymer composites reinforced with 2D nanomaterials, ceramic composites require a homogeneous distribution of 2D nanomaterials for improving their mechanical performance. However, designing advanced ceramic composites is still a challenging task owing to interdiffusion and chemical reactions at high temperatures. To address this challenge, novel preparation methods have been developed as described in Section 2. Thus, their mechanical and tribological properties would be

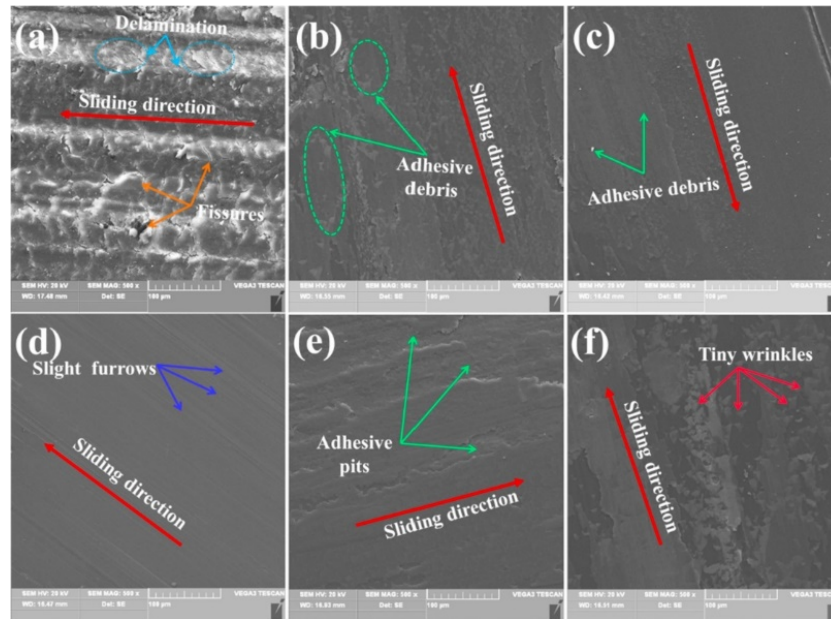


Fig. 7 Scanning electron microscopy images of worn surfaces of (a) bismaleimide resin and its composites incorporating: (b) 0.2 wt%, (c) 0.4 wt%, (d) 0.6 wt%, (e) 0.8 wt%, and (f) 1.0 wt% PHbP@rGO/WS₂. Reproduced with permission from Ref. [155], © Elsevier Ltd. 2019.

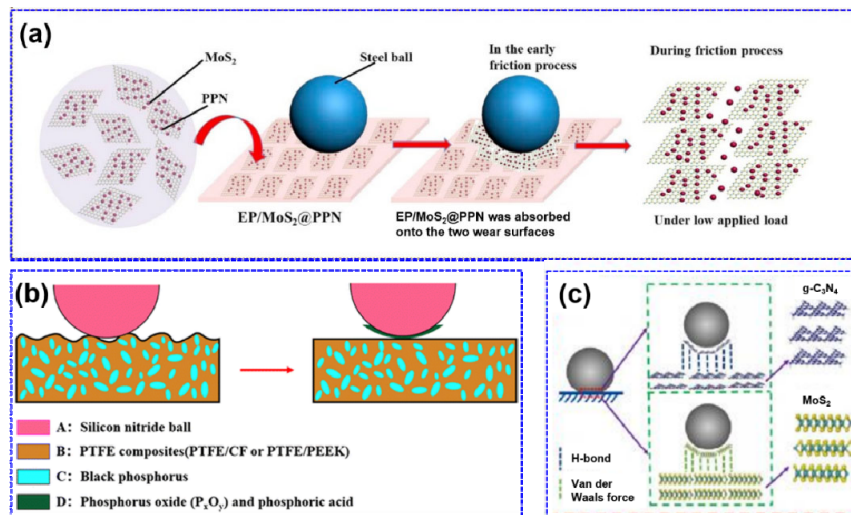


Fig. 8 Schematic of friction mechanism for (a) EP/MoS₂@ PPN composites. Reproduced with permission from Ref. [154], © Elsevier Ltd. 2018. (b) PTFE/phosphorene. Reproduced with permission from Ref. [150], © Springer Nature 2018. (c) PTFE/MoS₂ and PTFE/g-C₃N₄. Reproduced with permission from Ref. [149], © Springer Nature 2019.

discussed in the following sections.

4.2.1 Strength

Graphene-reinforced ceramic composites, which possess the strongest 2D nanomaterial, have been studied extensively [161–170]. Two reviews have already examined critically the effect of graphene [171, 172]. Even a very small content of graphene can significantly improve the tensile strength and

fracture toughness of ceramic composites. For example, the fracture toughness of zirconia ceramics can improve with 0.01 wt% graphene [161]. Krystek et al. [80] reported that the addition of 0.05 wt% graphene to ordinary cement results in a significant increase of up to 79% in the tensile strength. In another example, with increasing graphene content, the flexural strength and fracture toughness of AlN composites initially increased and then decreased

[166]. Graphene nanosheet (GNS) bridging the crack propagation path indicated the crack bridging effect of GNSs during crack propagation, which in turn, increased the fracture toughness. Furthermore, the microstructure of composites was investigated to explain the reinforcing mechanism of 2D nanomaterials [166]. It was found that the addition of graphene promotes the hydration reaction of both alite and belite, and thus leading to the formation of a large fraction of $3\text{CaO}\cdot 2\text{SiO}_2\cdot 3\text{H}_2\text{O}$ (C–S–H) phase. In fact, the 2D nanomaterials proved to be superior to the 1D and 3D nanomaterials, such as nanotubes and nanoparticles, in improving the mechanical properties of ceramic matrices [173]. A detailed review by Papageorgiou et al. [24] summarizes the research on improving the mechanical properties of graphene nanocomposites.

Owing to the poor toughness of ceramic matrices, BN can be used to improve their mechanical properties [174]. Lee et al. [28] explored the mechanical properties of a BN/Si₃N₄ nanocomposite by the single-edge notched beam method and microstructural analyses. The strengthening mechanism is summarized in Fig. 9. Moreover, it was shown that the introduction of BN results in the strengthening and toughening of ceramic composites [28, 82, 175].

MoS₂ nanoplatelets were also incorporated into bioceramic scaffolds fabricated by selective laser sintering (Fig. 10) by Shuai et al. [15]. At 0.5 wt% MoS₂ nanoplatelets, the compressive strength and fracture toughness of the bioceramic scaffolds improved by 46% and 24%, respectively. The MoS₂ crack bridge/deflection enhances energy dissipation, leading to the improvement.

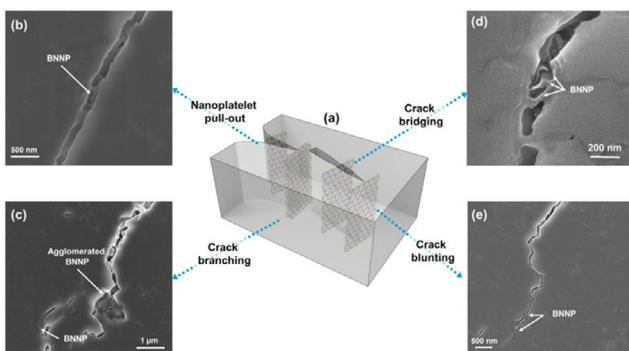


Fig. 9 Microstructure of a ceramic composite reinforced with BN. Reproduced with permission from Ref. [28], © Springer Nature 2016.

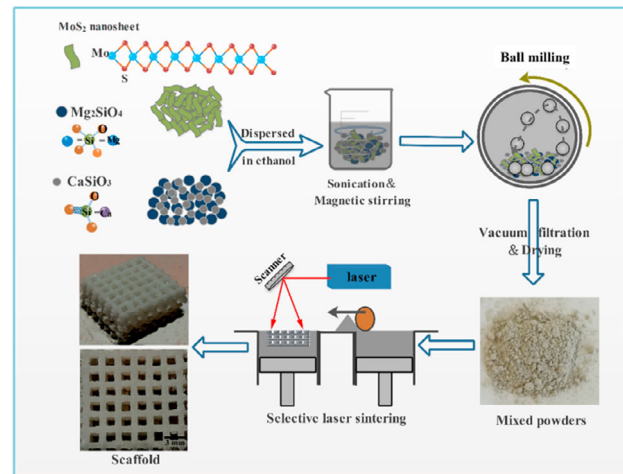


Fig. 10 Fabrication process of a bioceramic scaffold incorporating MoS₂. Reproduced with permission from Ref. [15], © Elsevier Ltd. 2017.

MXenes for high-strength composites have attracted much attention and were incorporated into ceramic composites. Incorporating 2 wt% Ti₃C₂T_x into an alumina ceramic composite can improve the fracture toughness, bending strength, and hardness to ~300%, ~150%, and ~300%, respectively [176]. The hardness and elastic modulus of ZnO ceramic were increased dramatically with 0.5 wt% Ti₃C₂T_x [177]. The microstructure of the nanocomposites and the outstanding intrinsic mechanical properties of Ti₃C₂T_x were the reasons for such improvements.

In summary, the toughness of ceramics can be improved by 2D nanomaterials, which can solve the brittleness problem. The strengthening of ceramic composites by 2D nanomaterials is mainly attributed to two reasons. The first reason is the microstructure of nanocomposites. The 2D nanomaterials are mainly dispersed at the grain boundaries of the ceramic matrices, resulting in a remarkable increase in the grain boundary strength and energy dissipation. Second, the outstanding intrinsic mechanical property and large surface area of 2D nanomaterials are equally important.

4.2.2 Friction and wear characteristics

As discussed in Section 4.1.2, very significant improvements have been achieved in ceramic matrices by using various types of 2D nanomaterials. Further research on the tribological performance of ceramics reinforced with 2D nanomaterials is necessary [161,

163–165, 178, 179].

In a previous study by Belmonte et al. [178], graphene was added into a silicon nitride ceramic and the tribological properties of the graphene/ceramic composite were reported. One of the most interesting properties of the graphene/Si₃N₄ composite is its excellent wear resistance, which can be attributed to the formation of a protective tribofilm by graphene. In addition, the tribological performance of zirconia composites reinforced with an *in situ*-reduced GO was reported recently [180]. The wear rate was decreased from 2.33×10^{-5} to 4.66×10^{-6} mm³·N⁻¹·m⁻¹ with 0.5 wt% GO (Fig. 11(a)). Moreover, various amounts of graphene were added into a ceramic to evaluate the friction and wear [181]. The results are interesting because all composites exhibited outstanding tribological performance, even

those composites containing large amounts of graphene.

Besides graphene, other 2D nanomaterials including BN [28], MoS₂ [182], and WS₂ [183] have been incorporated into ceramic matrices by various fabrication methods. The results revealed that the tribological performance of ceramic composites is better than that of pure ceramics (Figs. 11(b) and 11(c)). These results verified the advantage of incorporating 2D nanomaterials in ceramic composites.

From the above discussion, it can be concluded that to obtain the lowest COF and best wear resistance, 2D nanomaterials can be considered as one of the most effective reinforcements (as listed in Table 5). The resulting microstructures of ceramic composites reinforced with 2D nanomaterials lead to significantly lower friction and wear [184]. The main wear mechanisms of ceramic composites reinforced with

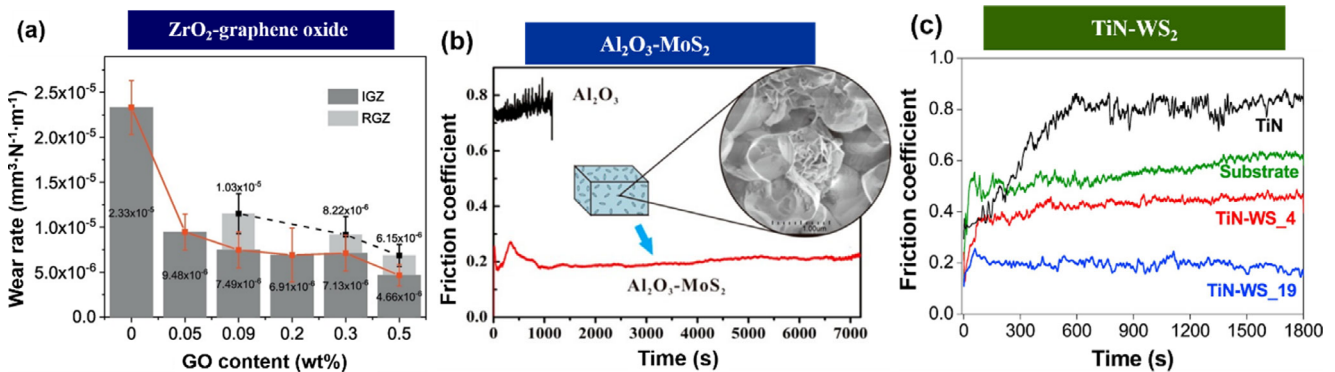


Fig. 11 (a) Specific wear rates of ZrO₂ and ZrO₂-GO (IGZ: *in situ* reduced graphene oxide reinforced 3 mol% yttria stabilized zirconia; RGZ: Pre-reduced graphene oxide reinforced 3 mol% yttria stabilized zirconia). Reproduced with permission from Ref. [180], © Elsevier Ltd. 2018. COFs of (b) Al₂O₃ and Al₂O₃-MoS₂ composites. Reproduced with permission from Ref. [182], © American Chemical Society 2017. (c) TiN and TiN-WS₂ composites. Reproduced with permission from Ref. [183], © Elsevier Ltd and Techna Group S.r.l. 2019.

Table 5 Tribological properties of ceramic composites reinforced with 2D materials.

Matrix	2D material	Method	Operating condition	COF	Wear rate (mm ³ /(N·m) ⁻¹)	Ref.
Al ₂ O ₃	1.0 vol% graphene	Ball milling and cold isostatic press	25 N, 0.1 m/s	0.35	2.0×10^{-5}	[185]
Si ₃ N ₄	20.6 vol% graphene	Solution mixing and SPS	180 N, 0.1 m/s, lubrication with iso-octane	0.10	5.0×10^{-8}	[181]
SiC	20 vol% graphene	Solution mixing and SPS	180 N, 0.1 m/s, lubrication with iso-octane	0.12	1.0×10^{-6}	[186]
ZrO ₂	0.5 wt% Graphene oxide	Ultrasonic and SPS	30 N, 0.12 m/s	0.76	4.66×10^{-6}	[180]
SiO ₂	5.0 vol% graphene	Ball milling and SPS	5 N, 0.1 m/s	0.56	7.7×10^{-6}	[179]
Si ₃ N ₄	2.0 vol% BN	Hot pressing	39.2 N, 7.85 mm/s	0.30	—	[28]
TiN	19 wt% WS ₂	Reactive magnetron sputtering	2 N, 14 mm/s	0.19	—	[183]
Al ₂ O ₃	10 vol% MoS ₂	Ultrasonic and vacuum infusing	5 N, 5 cm/s, high vacuum	0.20	1.4×10^{-6}	[182]

2D nanomaterials are plastic deformation [180, 183] and microcracking [180]. These findings suggest that 2D nanomaterial-based ceramic composites have the potential for various tribological applications.

4.3 Metal composites

Metal composites are widely used in aerospace, automotive, precision instruments, and large infrastructure owing to their high strength and stiffness. Besides the well-designed 2D nanomaterial-based polymer and ceramic composites, 2D nanomaterials have also been used as reinforcements in metal matrices including Cu [99, 187–191], Al [192–194], Mg [195], Ni [91, 196], and alloys [197]. The mechanical properties of metal composites incorporating 2D nanomaterials are discussed in the following subsections.

4.3.1 Strength

Recent studies have shown that metal composites filled with 2D nanomaterials exhibit superior hardness, γ , compressive strength, and tensile strength at room temperature compared to their corresponding unreinforced matrices. For example, Cu/graphene composite is typical, as described in Refs. [198–200]. Several recent studies reported the outstanding mechanical properties of composites with 2D nanomaterial contents from 0.05 wt% to 2 wt% [201]. However, the properties are highly dependent on the type and content of the 2D nanomaterial, as shown in Table 2.

The reinforcement of metal matrices using graphene has become one of the hottest topics in metal composite research [44, 45, 77, 81, 91, 92, 99, 187–189, 192, 193, 195, 196, 202–207]. Early studies on the mechanical properties of composites have demonstrated the advantages of incorporating graphene and its derivatives in metal composites [81, 194, 208–212]. The strengthening efficiency shown in Fig. 12 indicates that functionalized graphene can reinforce a copper matrix more effectively than other nanomaterials including carbon fiber, CNT, and pristine graphene, leading to the best strengthening efficiency for metal matrices. To ensure uniform dispersion of graphene in the copper matrix, Shao et al. [49] fabricated graphene-

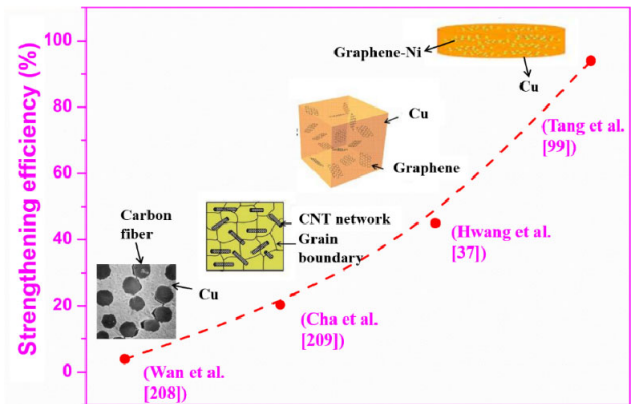


Fig. 12 Summary of progress in the strengthening efficiency of Cu matrix composites. Graphene–Ni. Reproduced with permission from Ref. [99], © Elsevier B.V. 2014. Carbon fiber. Reproduced with permission from Ref. [208], © Elsevier Science S.A. 2000. CNT network. Reproduced with permission from Ref. [209], © John Wiley and Sons 2005. Graphene. Reproduced with permission from Ref. [37], © John Wiley and Sons 2013.

nanoplatelets/copper (GNPs/Cu) composites by electrostatic self-assembly and spark plasma sintering. The resulting GNPs/Cu composite exhibited outstanding tensile strength and hardness. In another study, it was observed that the Cu–O–C bonding can be formed in graphene/Cu composites fabricated by electrochemical deposition [213]. Oxygen-mediated bonding was reported by Zhao [91], as depicted in Fig. 13. This suggests that interface bonding is important for achieving excellent mechanical properties in metal composites. Recently, the effects of ball milling time [93], content [214], and functional group [140] on the mechanical properties of metal composites have been studied in detail. Researchers are continuously extending the research scope of graphene/metal composites.

The addition of BN to a Ni₃Al matrix has enhanced the elastic modulus and thereby shows its significant strengthening effect [36]. In another study, the enhancement in the mechanical properties of composites was attributed to the higher strength of the 2D nanomaterials [75]. Metal composites incorporating MXene or 2DMOF have not yet been explored owing to their high cost and expected defects in structure.

Five strengthening mechanisms are summarized here. First, dislocation strengthening occurs when 2D nanomaterials become effective barriers to dislocation migration [215, 216]. Molecular dynamics

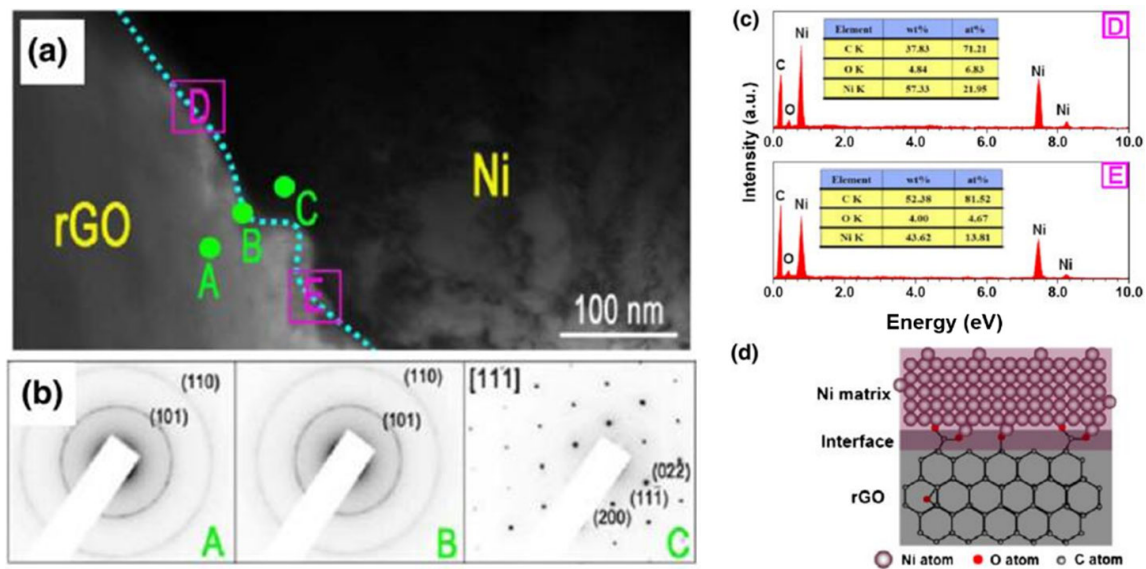


Fig. 13 (a) Transmission electron microscopy (TEM) image of the interface between rGO and Ni matrix. (b) Selected area electron diffraction patterns and (c) electron energy-dispersive X-ray spectrometry results at the regions marked in (a). (d) Schematic of interface bonding. Reproduced with permission from Ref. [91], © Springer Nature 2014.

simulation results also corroborate this mechanism [217]. Second, it is generally accepted that load transfer can be achieved owing to the large surface area and high strength of 2D materials [204, 218]. Third, grain refinement in metal matrices also occurs as revealed by TEM [219]. In addition, thermal mismatch strengthening [211] and Orowan strengthening [36, 193] have been mentioned in some cases.

4.3.2 Friction and wear characteristics

The use of 2D nanomaterials to reinforce metals usually leads to excellent improvement in the self-lubrication and anti-wear properties of the composites. The COF of 0.3 wt% graphene/Cu composite decreases by 65% compared to that of pure copper [220]. Graphene addition causes a plastic zone and a decrease in grain size [45]. It is noteworthy that as a 2D-layered nanomaterial, graphene has a lubricating effect [221], leading to a fairly stable frictional behavior in metal composites [197]. Moreover, the excellent antifriction performance of graphene has been revealed by both simulation and experimental studies [13]. BN-reinforced Ni_3Al composites also showed outstanding lubrication and anti-wear resistance [36]. Compared to metal composites reinforced with graphene, those reinforced with BN exhibit high COF and poor wear resistance. This is because graphene possesses better self-lubrication characteristics [220].

Mai et al. [222] first demonstrated that MXenes can significantly improve the tribological properties of metal matrix composites. However, different from other 2D nanomaterials such as graphene, it is difficult to observe transfer layers on the contact surface of the $\text{Ti}_3\text{C}_2/\text{Cu}$ composite. Thus, the lack of transfer layers is one of the major reasons for its poor tribological properties. Therefore, it is worthwhile to carry out further research on how to improve this aspect in the future.

The above discussions show that the COF and wear rate of metal composites decrease with the addition of 2D nanomaterials. This improved tribological performance is mainly attributed to the formation of a tribolayer on the contacting surfaces [223–227]. Table 6 summarizes typical data on the tribological properties of metal composites reinforced with 2D materials.

5 Salient factors influencing the performance of composites

The enhancement in the mechanical and tribological properties of composites by 2D nanomaterial reinforcement also depends on numerous factors such as aspect ratio, volume fraction of nanomaterials, orientation of nanosheet, and interface bonding between the filler and matrix [230]. Naturally, the

Table 6 Tribological properties of metal composites reinforced with 2D materials.

Matrix	2D material	Method	Operating condition	COF	Wear rate ($\text{mm}^3/(\text{N}\cdot\text{m})^{-1}$)	Ref.
TiAl	3.5 wt% graphene	SPS	10 N, 0.2 m/s	0.33	0.33×10^{-4}	[197]
Ni ₃ Al	1.0 wt% graphene	Ball milling and SPS	11.65 N, 1.0 m/s	0.20	9.0×10^{-6}	[228]
Ni ₃ Al	BN	Selective laser melting	9 N	0.22	2.1×10^{-5}	[36]
Cu	10 vol% graphene	Hot pressing	2 N, 1 m/s	0.17	1.8×10^{-4}	[229]
Cu	4.0 vol% graphene	Molecular level mixing and SPS	5 N, 0.01 m/s	0.25	—	[45]
Cu	2 wt% Ni@graphene	Ball-milling and hot-pressing	6 N, 0.105 m/s	0.2	0.1×10^{-3}	[224]
Cu	Graphene	Ambient-pressure CVD	1 N, 2.74 cm/s	0.2	2.8×10^{-5}	[223]
Cu	Ti ₃ C ₂	Electrodeposition Technique	1 N, 5.2 cm/s	0.27	3.7×10^{-5}	[222]
Ni	0.68 wt% graphene	Powder metallurgy	5 N, 9 mm/s	0.39	5×10^{-4}	[225]
Al	2 wt% WS ₂	Cold spray	1 N, 2 mm/s	0.6	88×10^{-3}	[227]
Al	10 vol% graphene	Powder metallurgy	10 N, 0.1 m/s	0.2	9×10^{-3}	[226]
Al	15 vol% MoS ₂	Powder metallurgy	10 N, 0.1 m/s	0.2	20×10^{-3}	[226]
Al	15 vol% BN	Powder metallurgy	10 N, 0.1 m/s	0.4	100×10^{-3}	[226]

improvement in mechanical properties depends on various factors including the matrix, reinforcement, and process. These influencing factors in the context of the mechanical properties of 2DNBCs are discussed as follows.

5.1 Matrix type

As is well known, there are many types of matrix materials that can be selected. It is also essential to emphasize that the mechanical properties of composites fundamentally depend on the matrix [231]. Commonly, the order of the magnitude of tensile strength is ceramic matrix > metal matrix > polymer matrix. After being reinforced by 2D nanomaterials, the tensile strength of composites maintains a similar trend, as displayed in Fig. 14. In addition, the order of the magnitude of elastic modulus is similar (ceramics: 30–1,000 GPa; metals: 50–300 GPa; polymers: 0.4–4.0 GPa). Further, different varieties of the same type of material also exhibit different mechanical performances owing to differences in the chemical composition and structure.

5.2 Reinforcement state

In the previously highlighted key points, the majority

of the effects on the mechanical properties were attributed primarily to the physical properties of 2D nanomaterials (Fig. 15). The synergistic effect of graphene with other 2D nanomaterials was also reported by many researchers. Previous AFM-based indentation experiments have also demonstrated its significance [65].

5.2.1 Geometry

The geometry of 2D nanomaterials, such as lateral dimension, thickness, and aspect ratio, determines its mechanical properties. A key factor that affects many properties including mechanical properties is the lateral dimension [29]. 2D nanomaterials have a layered structure with a large lateral dimension and atomic thickness. Khan et al. [240, 241] found that the mean sizes of graphene and MoS₂ nanosheets have evident effects on the mechanical properties of composites. The high aspect ratio of BN sheets is a critical factor for improving the mechanical property of a composite [86]. The macromolecular chains of the matrix are confined by 2D nanomaterials with a high aspect ratio. The effect has also been confirmed in graphene/PEEK [43].

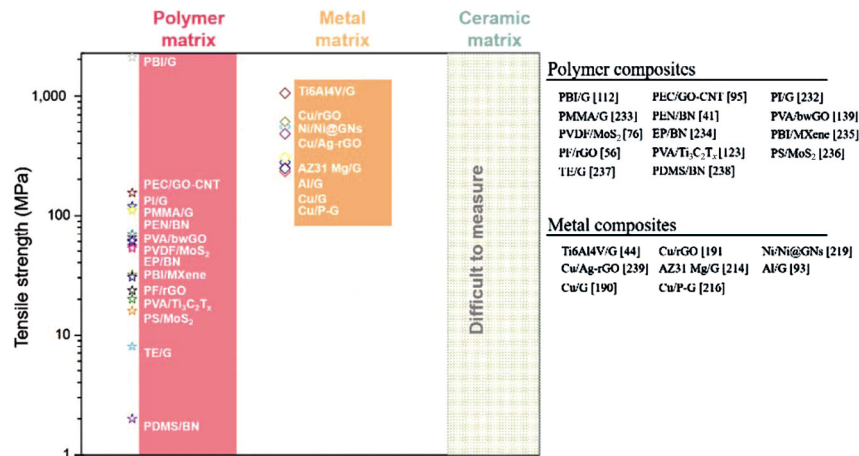


Fig. 14 Comparison of various matrices incorporating 2D materials (PBI: polybenzimidazole; PS: polystyrene; TE: thermoplastic elastomers; PDMS: polydimethylsiloxane; bwGO: base-washed graphene oxide; P-G: plasma treated graphene).

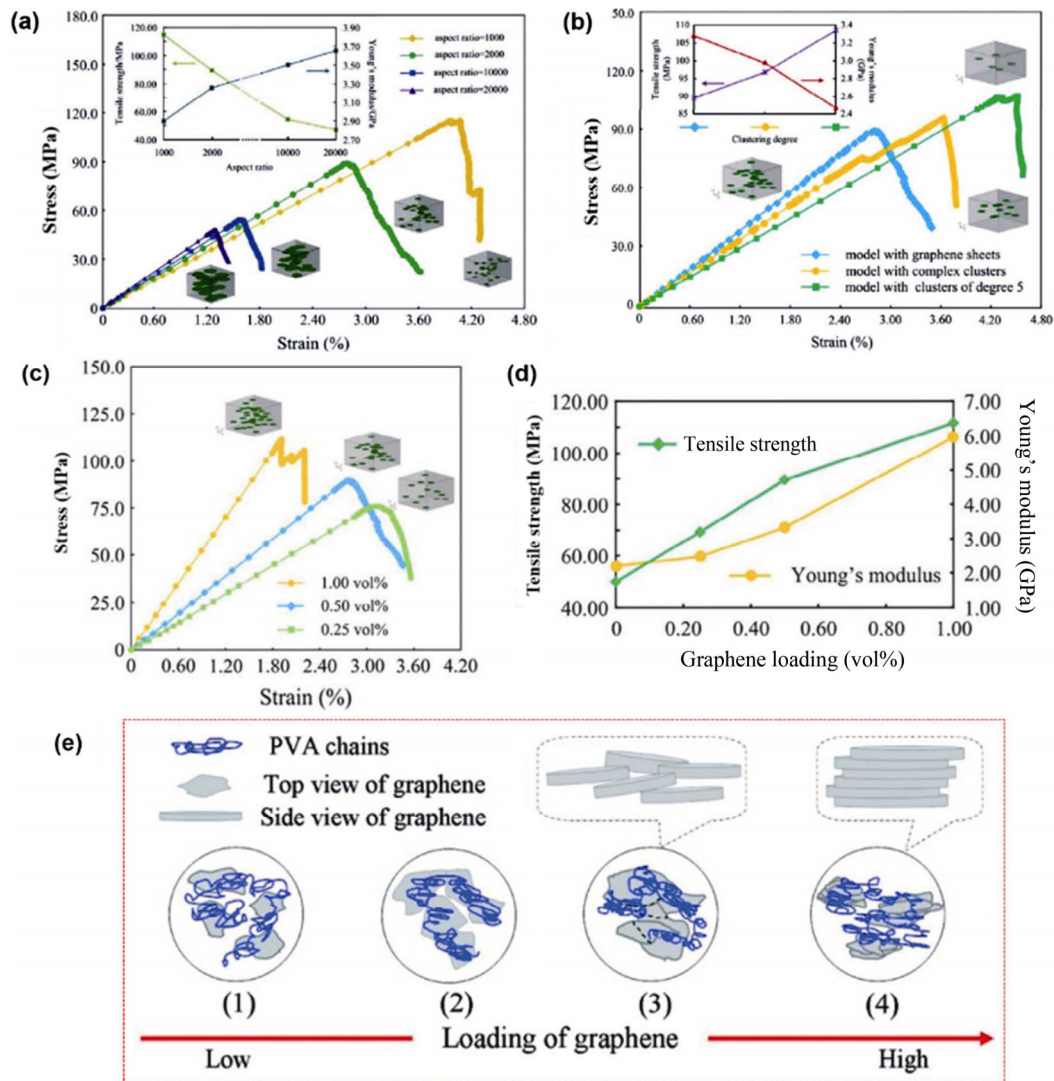


Fig. 15 Effect of (a) aspect ratio, (b) dispersion, and (c, d) volume content of graphene on the strain–stress curves of nanocomposites. Reproduced with permission from Ref. [63], © Elsevier B.V. 2014. (e) Schematic of the effect of volume content on the microstructure. Reproduced with permission from Ref. [55], © American Chemical Society 2010.

Moreover, thickness plays an important role in the mechanical properties of composites. Kiran et al. [79] demonstrated that the mechanical properties decrease linearly with increasing number of BN layers. Furthermore, several researchers reported the effect of thickness on wear of graphene/PTFE composites [148]. The results revealed the relation between wear and thickness.

When the 2D nanomaterials are small and thin, the composites have high tensile strength and ductility [242, 243]. These results demonstrate that the mechanical properties of polymer composites containing 2D nanomaterials depend primarily on the number of layers, with the properties decreasing linearly with increasing number of layers.

Although the aspect ratio plays a crucial role in determining the composite properties, little attention has been given to its influence on the mechanical and tribological properties of composites. A large aspect ratio can promote the load transfer ability in composite systems [236].

5.2.2 Volume content

The volume content of 2D nanomaterials is a key factor in controlling the mechanical strength, COF, and wear rate [45, 244, 245]. An extremely small amount of 2D nanomaterial, usually no more than 5 wt%, can significantly improve the mechanical properties of composites. As indicated in Fig. 15(e), the microstructure of a polymer composite has changed remarkably as the volume content of graphene increases. With the high volume content of graphene, the original advantage of a layered structure disappears, resulting in the deterioration of the mechanical properties.

The tensile strength of a composite can be calculated as [49]

$$\sigma_c = \sigma_g V_g + (1 - V_g) \sigma_m \quad (7)$$

where σ_g , σ_m , and σ_c are the tensile strength of graphene, the matrix, and the composite, respectively; whereas V_g represents the volume content of graphene. It may be noted that the volume content is crucial to the tensile strength of composites. As mentioned above, there is a critical point of the volume content [56]. At the critical point, the composite exhibits the best mechanical property

[246]. The strength and modulus of composites both increase with the increase in the 2D nanomaterial content [63]. At a higher volume content, 2D nanomaterials are prone to self-aggregation, resulting in the deterioration of the mechanical properties of the composite [41].

5.2.3 Dispersion

The effect of the dispersion of 2D nanomaterials on the mechanical properties of 2DNBCs has been extensively studied [6, 47, 121, 160, 218, 247–249]. To summarize, the degree of dispersion, including that of the 2D nanomaterials and other fillers, has a significant impact on the mechanical performance of composites. Figure 15(b) illustrates the effect of dispersion of graphene on the strain–stress curves of nanocomposites. Poor dispersion decreases the effective aspect ratio of 2D nanomaterials, leading to the deterioration in mechanical properties. Thus, dispersion is a major challenge in the fabrication of various composites [250]. Luo et al. [239] found that homogeneous dispersion of graphene can be achieved by the modification of silver nanoparticles. All the aspects mentioned above suggest that dispersion is one of the key factors for improving the mechanical properties. Achieving effective dispersion of 2D materials in the matrix should therefore be explored further.

5.2.4 Orientation

Recent studies have examined the role of the orientation of 2D nanomaterials within a material in its mechanical and tribological properties [234, 251]. A well-ordered orientation of 2D nanomaterials in composites maximizes the interactions between the 2D nanomaterials and the matrices, which lead to efficient stress transfer [252]. Conversely, a random distribution of 2D nanomaterials causes low load transfer ability [253]. Li et al. [251] reported that the Krenchel orientation factor can predict the effect of spatial orientation on the mechanical properties of 2DNBCs. Jan et al. [254] found that improving the alignment of BN nanosheets by uniaxial drawing can enhance the mechanical properties of composites. To control the orientation of graphene and its derivatives, a previous review

summarized a series of methods and evaluated the resulting properties [255].

5.2.5 Interface bonding

Interface bonding comprises hydrogen bonding, covalent bonding, ionic interactions, electrostatic

interactions, and so on. Most studies have mentioned that interface bonding is an important factor to be considered [232, 256–258], as displayed in Fig. 16. Under axial stress, the parallel lines become distorted owing to the low γ of the matrix (Fig. 16(g)). This means that 2D nanomaterials carry most of the

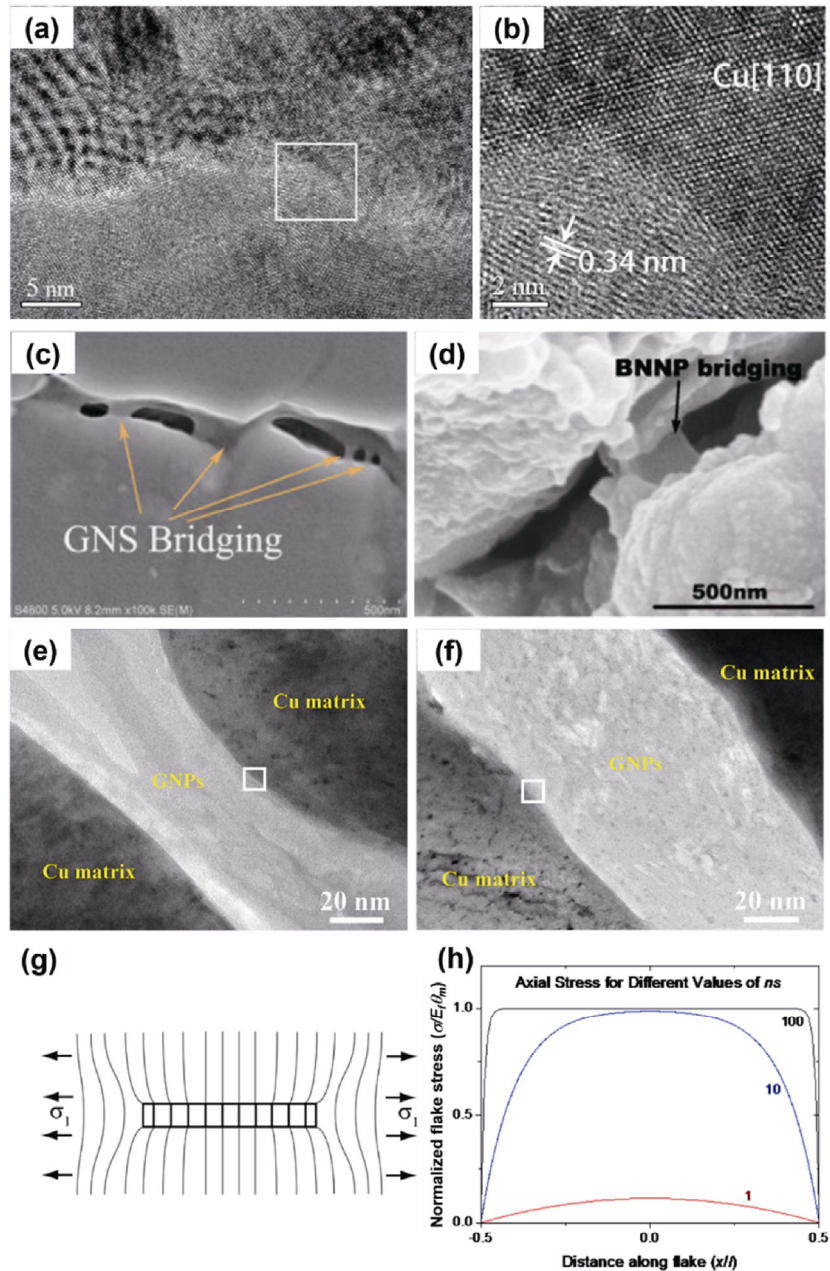


Fig. 16 Typical interface between graphene and copper matrix: (a, b) the interplanar distance of graphene is 0.34 nm. Reproduced with permission from Ref. [204], © American Chemical Society 2015. Bridging of 2D materials in composite systems: (c) graphene bridging. Reproduced with permission from Ref. [166], © Elsevier Ltd and Techna Group S.r.l. 2015. (d) BN bridging. Reproduced with permission from Ref. [36], © The Authors 2019. (e, f) Graphene bridging. Reproduced with permission from Ref. [49], © Elsevier B.V. 2018. (g, h) Deformation patterns of 2D material in a matrix under axial stress σ_1 . Reproduced with permission from Ref. [126], © The Authors 2017.

load in the composite. According to the elastic contact theory [259], the Y of the matrix will be affected by the interfacial contact between the 2D nanomaterial and matrix. To ensure adequate stress transfer, strong interface bonding between the 2D nanomaterial and matrix is essential. In view of the importance of interface bonding, the interface microstructure was investigated by TEM in Ref. [49]. The 2D nanomaterial was incorporated into composites, resulting in the presence of 2D bridging.

Interface bonding can be controlled by modification [2, 130] and ball milling [93]. As the interface bonding is improved, the final Y of the composite becomes almost three times higher than that of the pure matrix [2]. Thus, the design of the interface is essential owing to its importance in suppressing shear bands of the composites. It is also called interface-dominated/enabled mechanical behavior [260]. In addition to its use as enhancement fillers in composites, 2D nanomaterials have proved to be an efficient potent coupling agent [261]. Ultrathin GO was introduced in the interface between silica and EP resin to optimize the mechanical properties [262]. The results showed that the mechanical reinforcement is superior compared to those reported by others. $\text{Ti}_3\text{C}_2\text{T}_x$ MXene nanosheets are already used for enhancing the interfacial connection between carbon fibers and EP resins as well as for reducing the stress concentration of the interface [101]. The design might be complicated, but it has the great advantage of improving the tensile, flexural, shear, and impact strengths.

5.3 Processing parameters

Composites fabricated by different methods may possess different mechanical properties. In fact, there are many processing parameters in every method, such as pressure, temperature, and time. As described in Section 2, the main methods used for fabricating 2DNBCs are *in situ* polymerization, solution mixing, melt blending, powder metallurgy, reaction bonding, chemical vapor deposition/infiltration, and molecular-level mixing. Additionally, novel approaches of fabricating advanced composites [263] have been developed. Each processing parameter directly affects the quality of the composites and

even creates defects. Thus, the mechanical properties of composites are affected significantly by the processing parameters.

6 Potential applications

In Section 4, the mechanical properties of 2DNBCs are reviewed. Excellent mechanical properties can lead to more applications and reduced costs of composites. In this section, we will focus on the discussion about the potential of the aforementioned prepared composites for a number of applications based on their outstanding mechanical properties (Fig. 17).

6.1 Aviation and aerospace

Composites have been used for a long time and

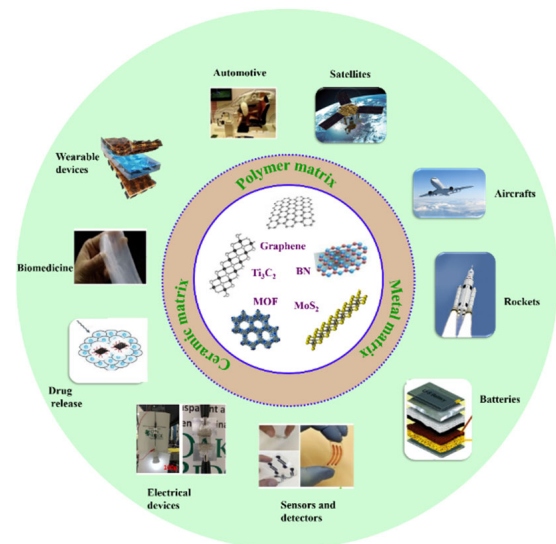


Fig. 17 Advanced technological applications of 2DNBCs. Ti_3C_2 . Reproduced with permission from Ref. [264], © John Wiley and Sons 2014. BN. Reproduced with permission from Ref. [265], © John Wiley and Sons 2018. MoS_2 . Reproduced with permission from Ref. [266], © American Chemical Society 2017. MOF. Reproduced with permission from Ref. [267], © John Wiley and Sons 2015. Drug release. Reproduced with permission from Ref. [268], © Springer Nature 2017. Wearable devices. Reproduced with permission from Ref. [269], © American Chemical Society 2018. Electrical devices. Reproduced with permission from Ref. [270], © American Chemical Society 2015. Biomedicine, automotive, sensors, and detectors. Reproduced with permission from Ref. [271], © Elsevier B.V. 2016. Batteries. Reproduced with permission from Ref. [272], © Science China Press 2017.

have become important materials in the field of aviation and aerospace. To further improve their performance, studies on reinforcing them with 2DNBCs have been reported widely in the literature. Among the family of 2DNBCs, graphene-based nanocomposites are one of the most promising materials in view of their excellent specific strength and good wear resistance. Besides graphene-based nanocomposites, composites incorporating MoS₂ and BN also exhibit excellent tribological properties. The different types of applications in aviation and aerospace can be categorized into structural components, e.g., fuselages, wings, and moving components, such as bearings, gears, and hatch seals [34]. It is well known that the aviation and aerospace industry depends on many structural and moving machine components; thus, 2DNBCs will play significant roles in this industry.

6.2 Biomedicine

The biomedical applications of 2DNBCs mainly include tissue engineering scaffolds [15, 273], drug delivery [274], thermotherapy [269, 275], and biosensors [276, 277]. Effective drug loading can be achieved by 2DNBCs owing to the 2D structure and delocalized surface π electrons [274]. Thus, composites have potential applications in drug delivery systems. Commonly, polymer composites used in tissue engineering scaffolds usually require structural strength and support [273]. To enhance their mechanical properties, 2D nanomaterials such as graphene and GO are incorporated into these matrices. For instance, Das et al. [65] demonstrated that incorporating functionalized few-layer graphene into PVA and PMMA can significantly increase their hardness and elastic modulus.

6.3 Energy storage

Energy storage including supercapacitors and batteries is a hot research area [278]. Based on the ultrathin flexible 2D structure, 2DNBCs applied to supercapacitors can endure different mechanical deformations [272]. For example, the graphene/polymer has led to successful implementation in supercapacitor [279]. In this case, graphene provided the most effective flexibility.

MXene/PVA composite has been used as electrode for supercapacitors and exhibits impressive performance [280]. It is noted that MXene provides an excellent flexibility and then the composite can sustain 5,000 times its own weight. In addition, 2DNBCs have potential to be used as protection materials and barrier materials such as body armor, ambient armor due to the in-plane mechanical isotropy of the 2D nanomaterial. Its critical penetration velocity gives 2DNBC a unique advantage over traditional woven fabric [281].

Overall, the potential application of 2DNBCs will be further expanded with the development of study of 2D nanomaterial [282]. It will have to be recognized that more work is needed in order to realize the true potential of 2DNBCs and to explore new application in the future.

7 Conclusions

In recent years, there has been a rapid development in the field of composites, especially in the design and characterization of novel 2DNBCs. Here, various characterization methods, including the use of universal testing machine, tribological testing, DMA, nanoindentation, and AFM, are introduced. All of these methods are proposed for the measurement of the mechanical and tribological properties of composites. Compared with conventional methods, microscopic methods, such as AFM, nanoindentation, and theoretical calculations, are becoming more important. This review reports the recent progress pertaining to the mechanical and tribological properties of composites reinforced with different 2D nanomaterials. Such nanomaterials including graphene, GO, rGO, phosphorene, BN, g-C₃N₄, MoS₂, WS₂, Ti₃C₂T_x, and MOF can effectively enhance the properties of composites because of their distinctive characteristics that can result in uniform dispersion, optimized orientation, and strong interface bonding. Of these 2D nanomaterials, graphene has been studied the most because of its ultrahigh strength, relatively low cost, and mature technology. In addition, owing to the weak interlayer van der Waals interaction of 2D nanomaterials, composites possessing outstanding tribological properties such

as ultralow friction, self-lubricating action, and ultralow wear rate can be realized. For these reasons, 2D nanomaterials are considered as the best reinforcements for various composites, such as polymers, ceramics, and metals. Particularly, the intrinsic mechanical properties of graphene have been proven to be excellent and it has been widely used for reinforcing various matrices. Further, the functionalization of 2D nanomaterials will be an effective approach for strengthening the interface bonding between the matrix and reinforcement.

Despite the enormous advances, several problems of 2DNBCs have not been fully resolved yet. These challenges include the followings:

1) Preparation of tailored 2D nanomaterials. Though much progress has been made in the preparation of 2D nanomaterials, high-quality and single-layer 2D nanomaterials except for graphene are still lacking. This is one of the main challenges in this field.

2) Uniform distribution of 2D nanomaterials in matrices. This determines the strength of the interface between the matrix and the 2D nanomaterial. Some easy and effective approaches are still expected. In addition, optimal orientation of the 2D nanomaterial needs to be maintained as much as possible by an appropriate technology such as external electric fields and magnetic fields.

3) Fundamental mechanism of strengthening and tribology for various composite systems. Much research has been attempted to elucidate the fundamental mechanism. However, some new systems such as phosphorene/polymer and MXene/polymer have not been investigated.

4) Low-cost applications of 2DNBCs. An increasing number of composite materials are being developed; thus, it is necessary to consider their particular applications. The cost and scalability should also be taken into consideration for real applications.

Considering the importance of composites for human activities, effectively designing and fabricating 2DNBCs need to be addressed in the future. This review opens up a new path toward the design of advanced composites having potential applications in various technological systems.

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Zhengjia JI. He received his Ph.D. degree in materials science and engineering from China University of Geosciences (Wuhan) in 2017. Afterward, he worked as a postdoctoral research fellow working with Prof. Jianbin LUO and Prof. Guoxin XIE at State Key Laboratory

of Tribology, Tsinghua University, Beijing, China. He is currently a lecturer at Qinghai University, China. He has published more than 10 papers in international journals. His major research areas include friction materials, self-lubrication materials, and 2D materials.



Guoxin XIE. He received his Ph.D. degree at Tsinghua University, China, in 2010, majoring in mechanical engineering. After that, he spent two years at State Key Laboratory of Tribology, Tsinghua University, China, for postdoctoral research.

From 2012 to 2014, he worked at Royal Institute of Technology, Sweden, for another two-year post-

doctoral research. Since 2014, he has worked at Tsinghua University as an associate professor. His research interests include intelligent self-lubrication, electric contact lubrication, etc. He has published more than 50 referred papers in the international journals. He won several important academic awards, such as Chinese Thousands of Young Talents, the Excellent Doctoral Dissertation Award of China, and Ragnar Holm Plaque from KTH, Sweden.



Jianbin LUO. He received his B.Eng. degree from Northeastern University in 1982, and got his M.Eng. degree from Xi'an University of Architecture and Technology in 1988. In 1994, he received his Ph.D. degree from Tsinghua University and then joined

the faculty of Tsinghua University. Prof. Jianbin LUO is an academican of the Chinese Academy of Sciences and a Yangtze River Scholar Distinguished

Professor of Tsinghua University, Beijing, China. He was awarded the STLE International Award (2013), the Chinese National Technology Progress Prize (2008), the Chinese National Natural Science Prize (2018, 2001), and the Chinese National Invention Prize (1996). Prof. LUO has been engaged in the research of thin film lubrication and tribology in nanomanufacturing. He has been invited as a keynote or plenary speaker for 20 times on the international conferences.