lubricants

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Review

Graphene-Family Lubricant Additives: Recent Developments and Future Perspectives

Yanfei Liu [1,2], Shengtao Yu [1], Qiuyu Shi [3], Xiangyu Ge [1,*] and Wenzhong Wang [1]

[1] School of Mechanical Engineering, Beijing Institute of Technology, Beijing 100081, China
[2] State Key Laboratory of Tribology in Advanced Equipment, Tsinghua University, Beijing 100084, China
[3] State Grid Smart Grid Research Institute Co., Ltd., Beijing 102209, China
* Correspondence: gexy@bit.edu.cn

Abstract: Graphene-family materials have been investigated by researchers as promising additives for various lubrication systems due to their unique physical-chemical properties. It has been proven that graphene-family materials can lead to enhanced lubrication and wear-resistance performance, which have potential to reduce the energy losses and carbon emissions, and the wear of machines for industrial applications. Experimental, theoretical, and simulation studies have been performed to investigate the tribological behaviors of graphene-family materials as additives. The tribological properties of graphene-family materials, including graphene, reduced graphene oxide, functionalized graphene, and the combination of graphene-family materials and other materials as additives, and the fundamental mechanism are systematically reviewed and concluded. The authors also discuss the potential engineering applications of graphene-family materials as lubricating additives, and the unsolved issues and optimistic outlooks in the near future.

Keywords: graphene; friction; wear; additive

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1. Introduction

The friction and wear of machine parts usually cause huge carbon emissions and economic losses, restricting the sustainable development of human society [1]. Under such a background, advanced lubrication techniques have been proposed by researchers to reduce energy dissipation and enhance durability [2–10]. Additives are usually added to lubricants to enhance the lubrication performance, viscosity under high temperature, antioxidation, wear-reduction properties, and so on. Among all the lubricant additives, carbon nanomaterials have attracted a lot of attention due to the low cost, eco-friendliness, and significant improvement in lubrication and anti-wear performances [11–13].

Graphene, as an atomically thin carbon material, exhibits extremely high mechanical strength, high thermal and electronic conductivities, and other unique properties, which make it highly attractive for numerous applications [14,15]. Graphene also exhibits excellent tribological performance. Even single-layer graphene can lead to a significant reduction in coefficient of friction (COF) and wear between friction pairs [16,17]. At the nanoscale or microscale, superlubricity (COF < 0.01) can be achieved between graphene layers with incommensurate contact [18–21]. Although graphene shows great potential for various applications, it should also be noticed that graphene has some inherent properties including zero bandgaps and chemical inertness, restricting the application of graphene. That is one of the reasons for researchers investigating the functionalization of graphene through the reaction with inorganic or organic molecules, chemical modification of the surface of graphene, and the noncovalent interaction with graphene [22–27]. Functionalized graphene (Figure 1) also exhibits different dispersion properties and tribological behaviors [28–30]. The excellent tribological behaviors and the feasible functional design of graphene-family materials make them promising additives for different lubrication systems. Within this

work, the authors aim to review the recent achievements that have been realized with graphene-family materials as lubricating additives; and the functional mechanisms are also discussed in detail. Moreover, the unsolved issues and the optimistic outlooks for the graphene-family materials as additives are also discussed.

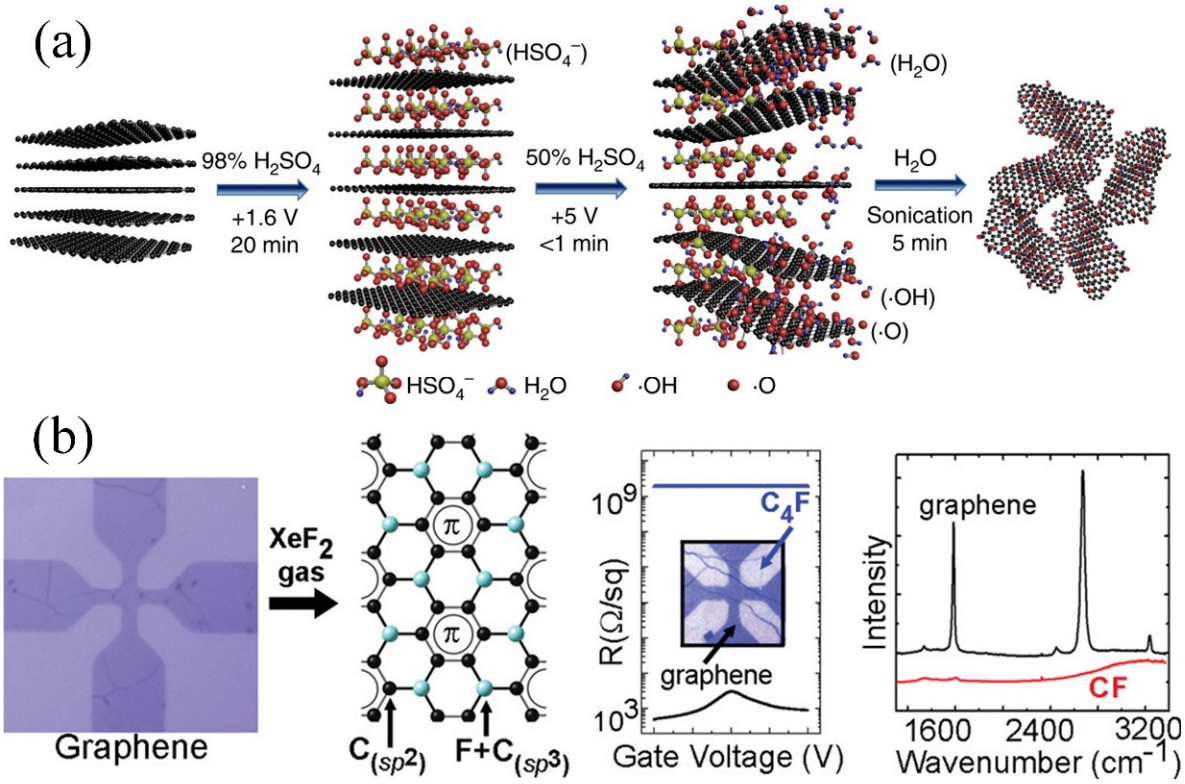

Figure 1. Functionalization of graphene-family materials including (**a**) oxidation [31] and (**b**) fluorination [32]. Reprinted with permission from Ref. [32]. American Chemical Society, 2010.

2. Tribological Behaviors of Graphene-Family Additives

Graphene-family materials have great potential as lubrication additives due to the atomic thin structure and high strength of graphene. Researchers have developed various lubricants containing graphene-family materials as additives, proving that graphene-family materials can effectively enhance lubrication and wear-resistance performances. However, different from the studies using graphene-family materials as solid lubricants, the detailed functional mechanisms of graphene-family materials as additives have not been elucidated due to the complexity of liquid-based lubricants. Hence, systematic studies are important for a better understanding of the tribological behaviors and the functional mechanisms of graphene-family materials as lubrication additives.

2.1. Pristine Graphene and Reduced Graphene Oxide (rGO)

Oil-based lubricants, including non-synthetic lubricating oils, synthetic lubricating oils, grease, and most recently, bio-based lubricating oil, are widely used as lubricants in both the market and the scientific research field. Typically, additives are needed for oil-based lubricants to improve the rheological, lubrication, anti-wear, and antioxidation behaviors. Different from traditional additives, nanomaterials have been used as additives due to the environmentally friendly, easy-processing properties and significantly improved lubrication and anti-wear performances [33]. Recently, graphene-family materials have attracted much attention from researchers as lubrication additives; and the performances of graphene-family materials as additives have been widely investigated. Among all the graphene-family materials, pristine graphene and reduced graphene oxide are ideal model materials for the understanding of the unique properties of graphene-family materials

as lubrication additives. Eswaraiah et al. [34] prepared ultra-thin graphene through the exfoliation of graphite oxide. Different from the graphite oxide as starting material, the as-prepared ultra-thin graphene has a low concentration of oxide. Ultra-thin graphene nanoflakes were added to engine oil to prepare nanofluids with graphene concentration of 0.0125–0.0625 mg/mL (Figure 2a). It was found that the optimized graphene concentration of nanofluids is 0.0250 mg/mL, which can significantly reduce the COF by 80% (Figure 2b) and wear scar diameter of friction pairs. The performance of graphene as additive was attributed to the nano-bearing effect of graphene and the ultrahigh mechanical strength of graphene. It was found that the concentration of graphene as an additive has an important influence on tribological behaviors. Guo et al. [35] prepared PAO2 oil with graphene concentration from 0.05–0.5 wt%, finding that the lowest COF can be achieved with the lowest graphene concentration of 0.05 wt%. Cai et al. [36] found that even a low graphene concentration of 0.01 wt% can effectively reduce the COF by 78% and wear rate by 90% with textured bronze plates as friction pairs. The function mechanism of graphene nanoflakes was attributed to the nano-bearing effect and the formation of protective film on the friction pairs. Sanes et al. [37] investigated the influence of graphene nanoflakes and the ionic liquid (1-octyl-3-methylimidazolium tetrafluoroborate) as additives on the tribological behaviors of additive-free isoparaffinic base oil and formulated motor oil. It was found that the addition of 0.005 wt% of graphene nanoflakes can lead to a 70% reduction of COF of motor oil at the temperature of 150 °C, while for the additive-free base oil, ionic liquid dispersed with graphene nanoflakes can significantly reduce the friction and wear at room temperature. It has also been found the tribological behaviors of PAO6 [38–40], hydraulic oil [41], 4010 AL base oil [42], vegetable oil [43], cashew nut shells liquid [44], wax extracted from *Codonopsis pilosula* [45], or lithium complex and polypropylene-thickened greases [46] can be also improved with graphene as additives. However, there is still room for the improvement of graphene as a lubrication additive. The structure of the sliding interface has significant influence on the lubrication performance of graphene or other 2D materials as solid lubricants, which has been widely investigated at the nanoscale [47–49] or macroscale [5,50,51]. For graphene as an additive, Jin et al. [52] regulated the structure of graphene through ball-milling treatment, suppressing the commonly observed wrinkles and curved edge-site (Figure 3). With the ultra-flat graphene as additive, the COF and wear depth can be reduced by 49% and 93% at high contact pressure of 2.5 GPa, respectively.

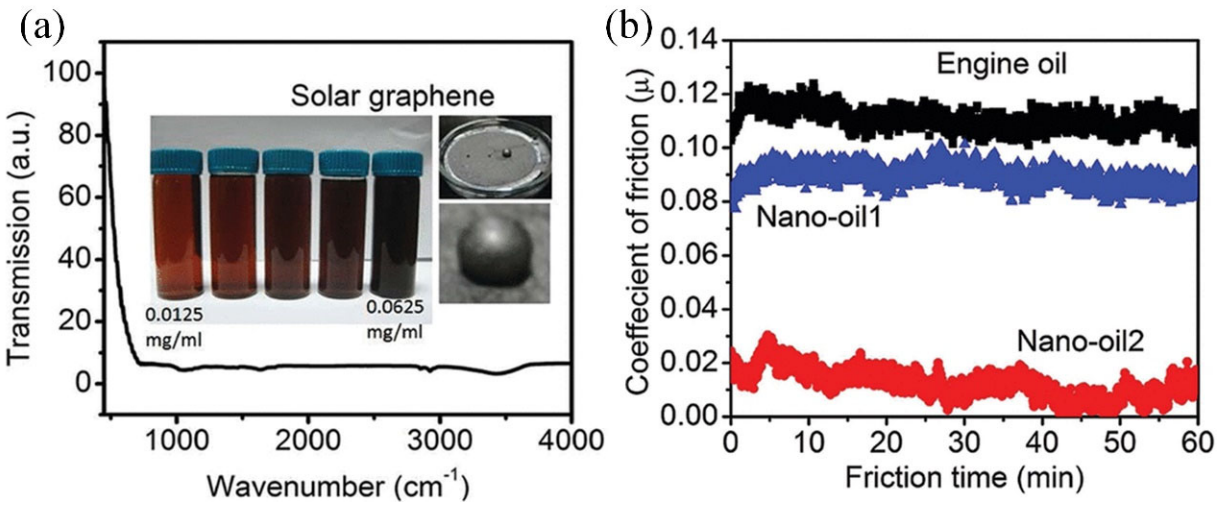

Figure 2. (**a**) Fourier transform infrared spectrum of as-prepared ultrathin graphene nanoflakes, where the insets are the photographs of the engine oil nanofluids with different concentrations of graphene, and the water droplet on graphene film; (**b**) COF versus time curves with engine oil and nanofluids. Reprinted with permission from Ref. [34]. American Chemical Society, 2011.

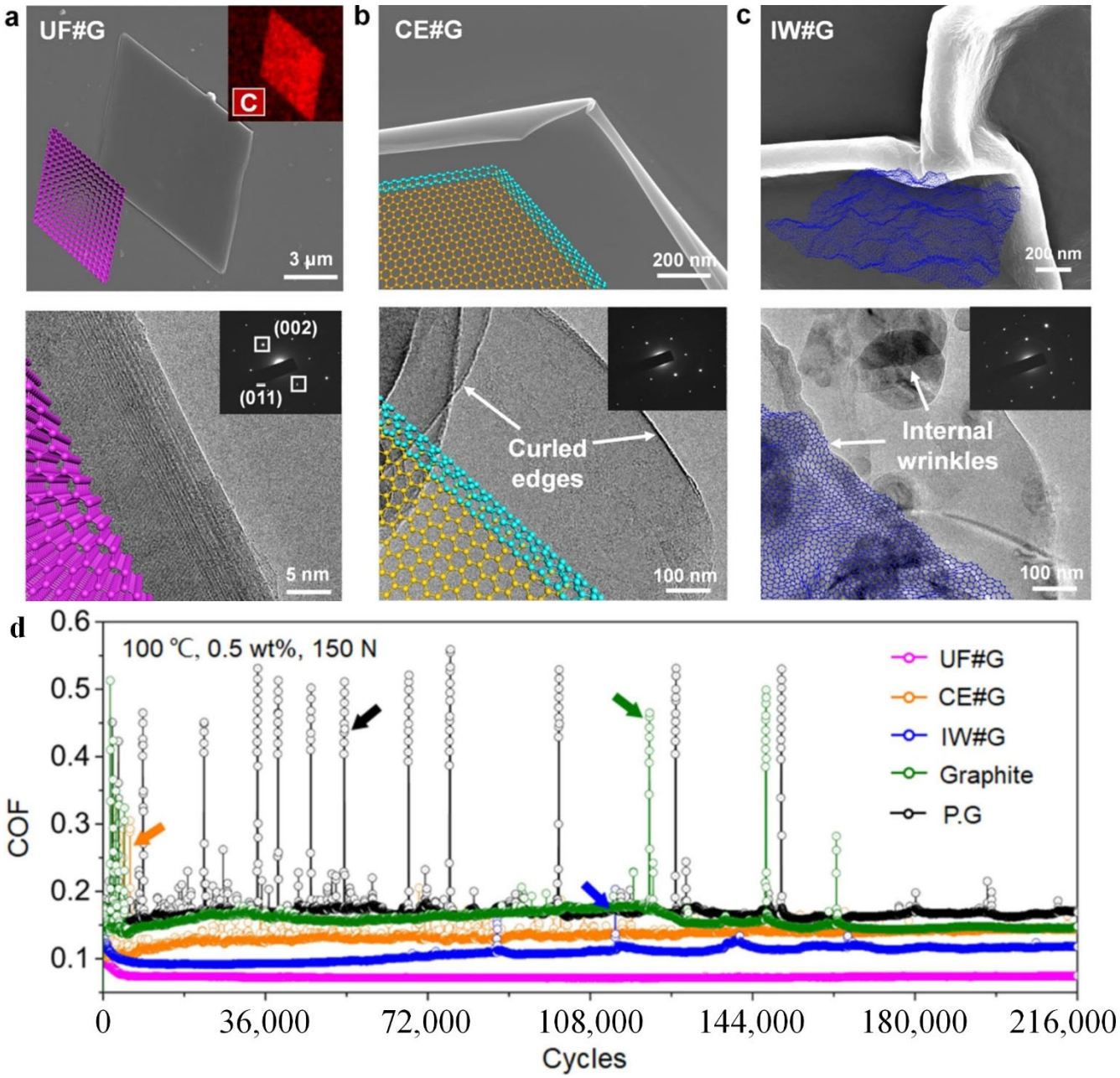

Figure 3. SEM and HRTEM images of (**a**) ultra-flat reduced graphene oxide, (**b**) curly edge reduced graphene oxide, and (**c**) internal wrinkle reduced graphene oxide; (**d**) COF versus time curves with different nanomaterials as additives. Reprinted with permission from Ref. [52]. Elsevier, 2022.

2.2. Functionalized Graphene

Pristine graphene or reduced graphene oxide exhibits excellent tribological behaviors as additives in the oil-based lubricant. However, those graphene-family materials are hard to be dispersed in water-based lubricants, restricting their application. In addition, the frictional and wear-resistant behaviors of graphene-family materials can be also regulated by functionalization. Hence, researchers have focused on the functionalization of graphene-family materials; and the tribological behaviors of functionalized graphene have also been systematically investigated. Wang et al. found that GO as an additive can effectively enhance the lubrication and wear-resistance performance of hexadecane-based oil [53]. Zhao et al. [38] compared the tribological behaviors of rGO and GO as additives in PAO6. It was found that PAO6 with GO exhibited much larger and unstable COF compared to

that with rGO, which was attributed to the hindered interlayer sliding between lattice layers by oxygen-containing groups. For the achievement of better tribological behaviors, Fan et al. [54] prepared fluorinated graphene through direct fluorination with F_2. Fluorinated graphene with different concentrations of F was added into liquid paraffin oil as additives. It was found that fluorination with C/F ratio of 1.0 can reduce friction and wear by 50.4% and 90.9%, respectively. The formation of tough tribofilm was believed the dominant mechanism for the friction and wear reduction properties. Paul et al. [55] prepared dodecylamine functionalized graphene nanosheets as an additive, which can reduce the COF by up to 40% compared to the base engine oil. Li et al. [56] prepared graphene oxide grafted with titanate coupling agent (T-GO) as an additive to hydraulic oil. With 0.08 wt% of T-GO, the COF and wear can be reduced by up to 50% and 20%, and better extreme pressure performance compared to the original hydraulic oil. Fu et al. [57] prepared ionic liquid-modified graphene. The excellent tribological behaviors of the ionic liquid-modified graphene as an additive can be attributed to the adsorption of modified graphene and tribochemical reaction of contact surfaces. The alkylated graphene [58] and graphene oxide [59,60], phosphonium-organophosphate-modified graphene [61], CuAAC-modified graphene oxide [62], 3,5-ditert-butyl-4-hydroxybenzaldehyde-grafted graphene [63], octadecylamine- and dicyclohexylcarbodiimide-modified GO [64], octadecylamine-functionalized rGO [65], rGO doped with N and B species and polyisobutylene succinimide-grafted graphene [66] also exhibit excellent dispersibility and tribological behaviors as additives of oil-based lubricants.

Despite oil-based lubricants, functionalized graphene has been widely used as an additive to water-based lubricants due to its better dispersibility and stability [67–71]. Kinoshita et al. [72] found that with GO as an additive in pure water, the COF between tungsten carbide ball and stainless steel plate can be significantly reduced from around 0.4 to 0.05; and the wear of friction pairs can be also significantly suppressed. Adsorbed GO was found on the surface of friction pairs, which was believed to act as a protective coating for the lubrication and wear-resistance properties. He et al. [73] investigated the influence of pH value on the tribological behaviors of GO as an additive to water. It was found that the GO sheets can be broken down and chemically reduced under high pH value, leading to a higher COF. Wei et al. found that better lubrication behavior can be achieved with graphene oxide/polysaccharide copolymer nanohybrids [74]. With the nanohybrid as an additive in pure water, the COF can be reduced by 40% and 84% compared with pristine GO and individual copolymer as additives, respectively. Researchers have also designed various functionalized graphene to further enhance tribological behaviors. Fan et al. [75] prepared ionic liquids modified graphene oxide as an additive of multialkylated cyclopentanes, finding that the ionic liquids-modified GO can lead to a reduction of COF and wear by 27% and 74%, respectively. GO modified by imidazolyl dinitrile amine also exhibited improved tribological performance [76]. Liu et al. [77] prepared graphene grafted with polyethyleneimine and polyacrylic acid, which improves the multiple adsorption effects of graphene on the counterparts' surfaces, leading to the formation of tough and stable tribofilms. Researchers found that fluorinated graphene exhibits better tribological performance compared to pristine graphene at the macroscale [54,78], but the hydrophobic nature of fluorinated graphene makes it difficult to be used as an additive for water-based lubrication systems. Targeting this problem, Ye et al. [79] developed hydrophilic urea-modified fluorinated graphene. Using urea-modified fluorinated graphene as an additive to water with a concentration of 1 mg/mL, wear can be reduced by 64.4% compared to that lubricated by pure water. Min et al. [80] prepared fluorinated graphene oxide using hydrothermal reaction, obtaining excellent dispersibility in water and tremendous abrasion resistance performance. Fan et al. [81] prepared fluorinated graphene with relatively low F content under mild temperature conditions. The as-prepared fluorinated graphene oxide as an additive of water led to a 47% and 31% lower wear rate compared to that with the lubrication of pure water and water suspension of GO, respectively. Later, urea-modified fluorinated graphene oxide was also prepared as the additive for water lubrication [82].

With traditional lubricants, the COFs are usually in the range of 0.02–0.1 [81,83–86]. Under the background of energy saving and emission reduction, the concept of superlubricity (COF < 0.01) has attracted a lot of attention [1,87,88]. Ge et al. [89] found that macroscale superlubricity can be achieved by ethylene glycol water solution containing GO as an additive. The achievement of superlubricity was attributed to the synergy between hydrodynamic effect and boundary lubrication provided by GO tribofilm. Even better lubrication and wear-reduction performances were achieved with the combination of GO nanosheets and an ionic liquid as additives [90] (Figure 4). Superlubricity can be also achieved by GO and lithium salts as an additive for dihydric alcohol aqueous solutions [91]. The adsorbed GO and tribochemical reaction are both important for the achievement of superlubricity. The functional groups have a potential influence on the superlubricity behaviors with graphene-family as additives. Recently, GO-OH, GO–COOH, and GO–NH_2 were added to dihydric alcohols as additives [92]. It was found that robust superlubricity can be achieved using GO–NH_2 as an additive, which was believed attributed to the formation of the adsorption layer due to the high adhesion between GO–NH_2 and SiO_2 substrate. The achievement of superlubricity usually needs a running-in process with high COF and severe wear. Recently, Liu et al. [93] used GO quantum dots as nano-additive of ethylene glycol water solution. A superlubricity state with COF of 0.0068 can be achieved with an extremely short running-in process of 6 s. The tribofilms with adsorbed GOQDs were believed to be critical for the reduced running-in process, and the achievement of a superlubricity state at relatively high contact pressure, which was confirmed by the in-situ friction tests and surface characterization results. Graphene-family as additives can provide lubrication and wear-resistance performance. Superlubricity with high contact pressure and the extremely short running-in process can be achieved with water-based lubricants with graphene-family materials as additives. However, the choice of graphene-family materials for water-based superlubricity systems is largely restricted by dispersibility and stability. GO is commonly used for water-based lubrication systems, but the interlayer shearing strength of GO is higher than the pristine graphene and other functionalized graphene such as fluorinated graphene [94,95]; and the interlayer sliding can be further influenced by the adsorbed water molecules, leading to higher COF in the environments with the presence of water molecules [96,97]. Targeting the above-mentioned problems, Liu et al. [98] proposed a novel strategy to achieve superlubricity with water-based lubricant and hydrophobic graphene, where the coatings with different graphene-family coatings were firstly deposited on the SiO_2 substrate and the glycerol aqueous solution was then added on the coatings for lubrication. The performances of pristine graphene, GO, and fluorinated graphene was compared. All the deposited graphene coatings led to lower COF and wear of friction pairs. Among the graphene-family materials, pristine graphene coating exhibited the best lubrication performance at a sliding speed of 0.1 m/s, where macroscale superlubricity with COF of 0.004 can be achieved. The superlubricity behavior can be attributed to the hydrodynamic effect and the boundary lubrication provided by the tribofilm containing graphene nanoflakes (Figure 5). Combining graphene-family materials as additives and lubrication coatings is also a strategy to realize superlubricity. An extremely small COF of 0.002 was achieved on silicon-doped hydrogenated amorphous carbon film with the lubrication of ethylene glycol containing GO as an additive [99].

2.3. Synergy between Graphene-Family and Other Nanomaterials

Researchers also combined graphene-family materials and other nanomaterials including other kinks of 2D materials, carbon nanomaterials, metal or metal oxide nanoparticles, and silica nanoparticles as additives for further improved tribological behaviors. Xu et al. [100] investigated the synergetic effect between graphene and MoS_2 as additives for esterified bio-oil. The tribological behaviors of the bio-oil added with 0.5 wt% graphene, 0.5 wt% MoS_2, and 0.3 wt% graphene, and 0.2 wt% MoS_2 were compared, finding that the nanofluid with 0.3 wt% graphene and 0.2 wt% MoS_2 exhibited lower COF and wear rate comparing to other samples. The formation of tribofilm is critical for enhanced tribo-

logical behaviors. With the combination of graphene and MoS_2 nanoflakes, it was found that the oxidation and degradation of MoS_2 can be suppressed by graphene nanoflakes, and the structure of graphene and MoS_2 can be better maintained through the synergetic effect (Figure 6). Farsadi et al. [101] added functionalized GO and MoS_2 into petroleum-based oil as an additive, achieving improved tribological performances. MoS_2 can be also directly synthesized on graphene or GO through calcination [102] and hydrothermal reaction [103–105]. The obtained composited nanomaterials can effectively enhance the lubrication and wear resistance performance of polyalkylene glycol [102,104] and based oil [103,105]. The intrinsic incommensurate between graphene and MoS_2 was believed to be one of the reasons for the enhanced lubrication performance [105]. Graphene-family materials were also mixed with h-BN [106], or grafted with APTMS-modified h-BN to improve the tribological properties as additives [107].

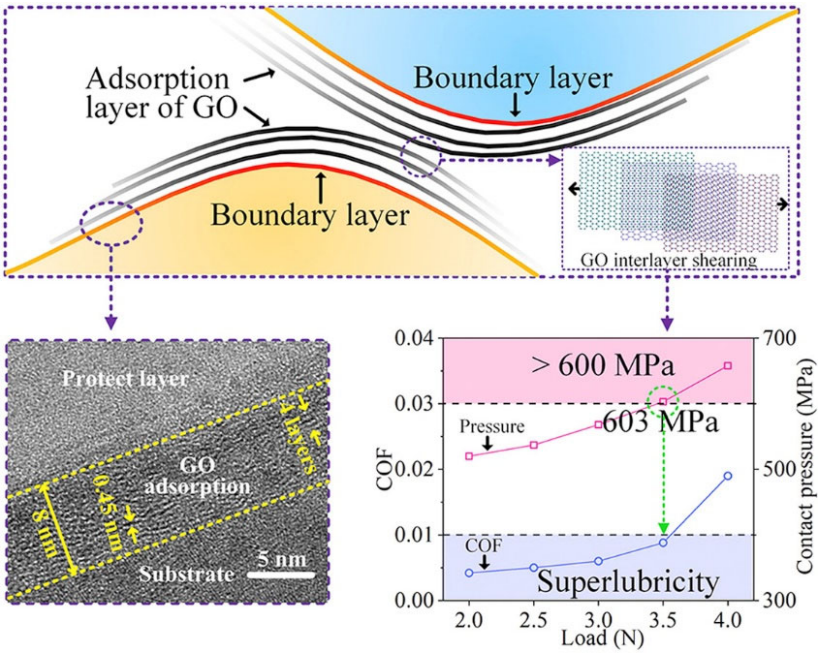

Figure 4. Superlubricity is achieved with the combination of GO nanosheets and an ionic liquid as additives in ethylene glycol water solution. Adsorbed GO can be observed onto the wear surface, which is critical for the achievement of superlubricity. Reprinted with permission from Ref. [90]. Elsevier, 2019.

The synergetic lubrication performance between graphene-family materials and nanoparticle or nanotubes was also investigated. A lubricant with GO and nanodiamonds as additives in pure water was developed by Wu et al. [108] to improve tribological performance. The low friction of approximately 0.03 can be obtained with 0.1 wt.% and 0.5 wt.% for GO and ND, respectively. Nanostructured tribofilm was found on the surface of friction pairs (Figure 7). The low shearing strength between graphene layers and the possible nano-bearing effect of ND was believed to be the mechanism for the remarkable lubrication performance. In addition ND, functionalized carbon spheres were also used combined with graphene, which led to an 18% reduction of COF compared to the CASTROL-20 W 40 engine oil [109]. Graphene-family materials have been combined with WS_2 [110], onion-like carbon [111], metal [112–120] or metal oxide [121–129] nanoparticles, silica nanoparticles [130–135], or carbon nanotubes [136–138] as additives for oil-based or water-based lubricants to provide better lubrication and wear-resistance performance.

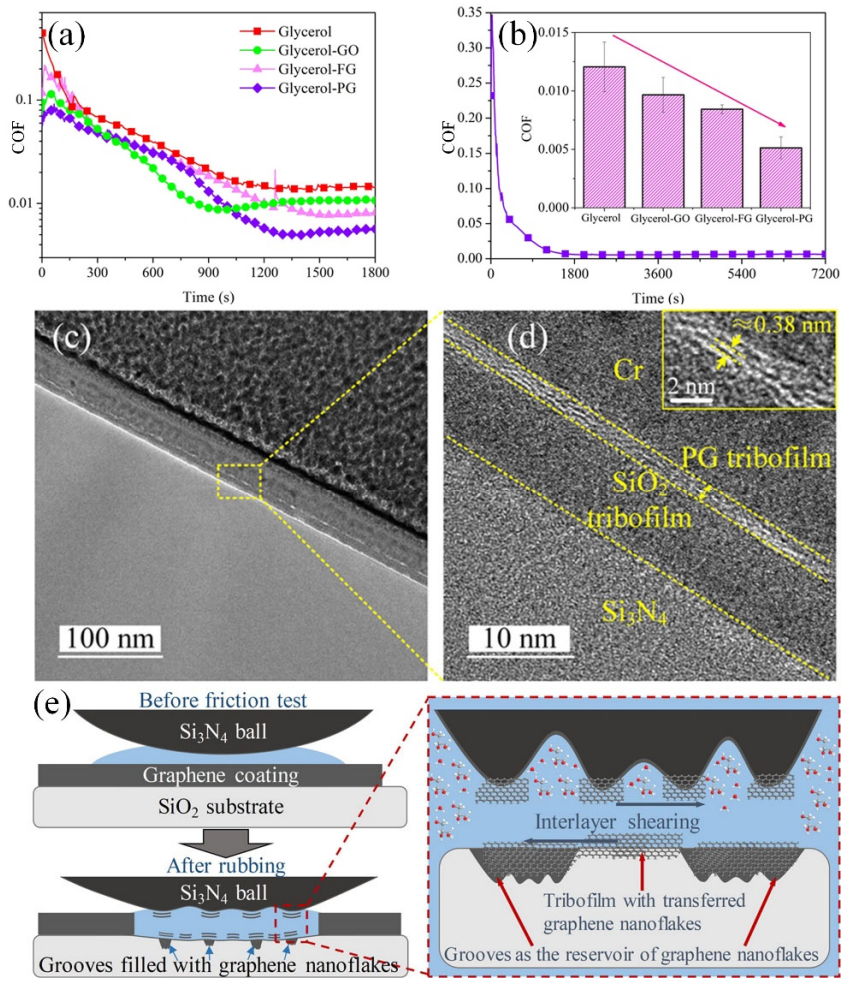

Figure 5. Superlubricity is achieved with the hydrophobic graphene coating and the glycerol aqueous solution. (**a**) COF versus time curves with different lubricants and the (**b**) long duration result with pristine graphene coating; (**c**) TEM and (**d**) HRTEM images of the tribofilm after the friction test with deposited pristine graphene coating; (**e**) schematic diagram of the superlubricity mechanism with hydrophobic graphene and water-based lubricant. Reprinted with permission from Ref. [98]. American Chemical Society, 2020.

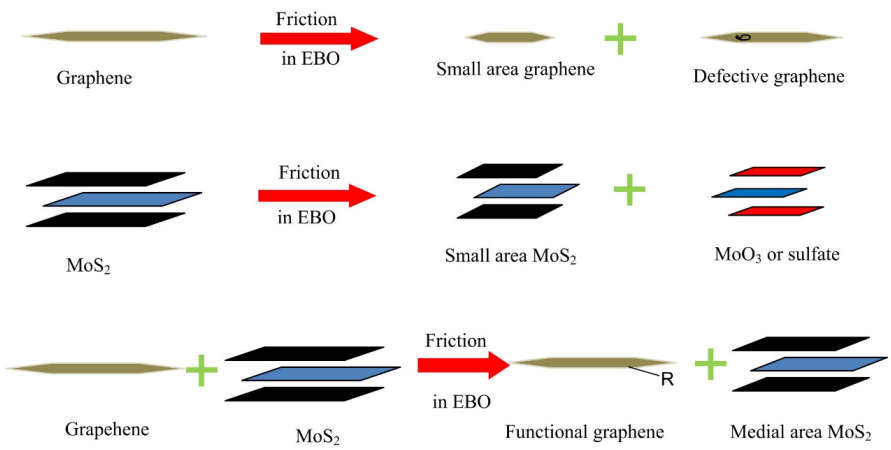

Figure 6. A proposed synergetic effect between graphene and MoS_2 nanoflakes as an additive in bio-oil. Schematic diagram of the structural evolution and chemical changes with graphene, MoS_2, and graphene combined with MoS_2 additives during friction process. Reprinted with permission from Ref. [100]. Elsevier, 2015.

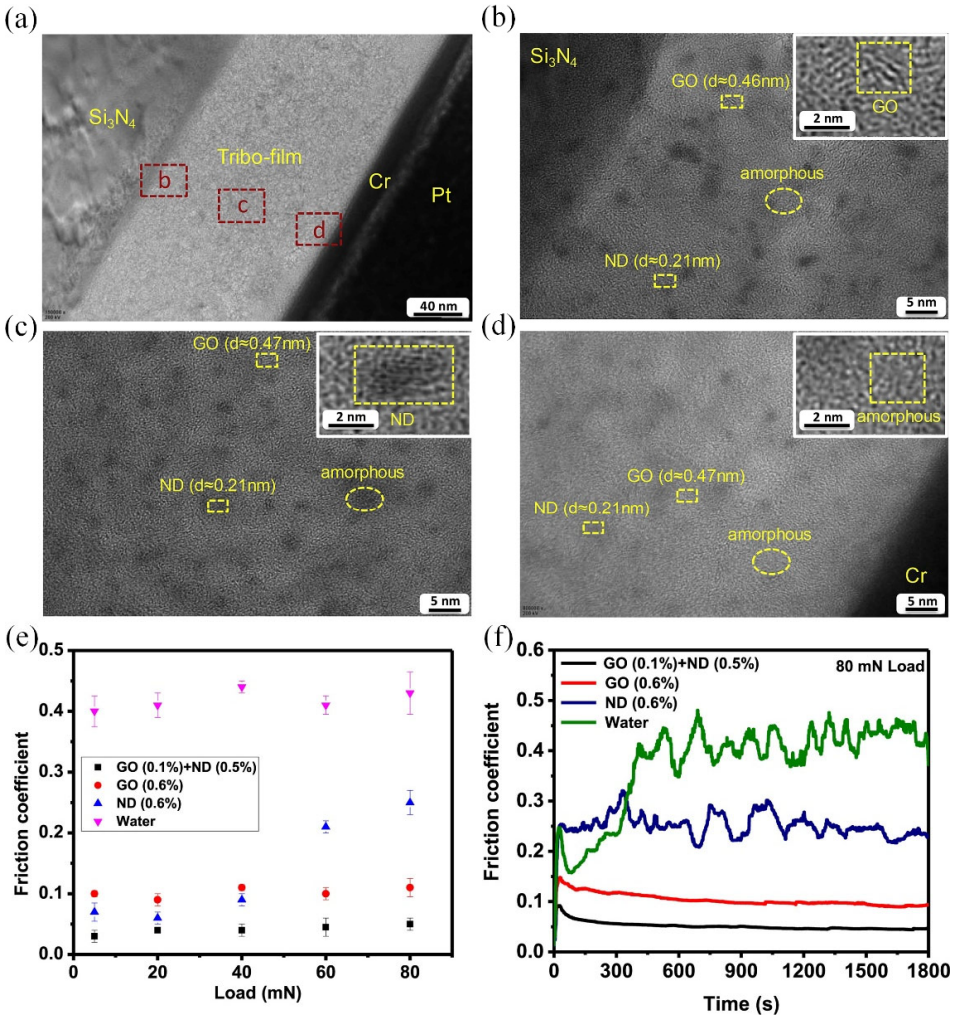

Figure 7. (**a**–**d**) Structure of the tribofilm containing GO and ND; (**e**,**f**) COFs with the lubrication of pure water, GO, ND, and the GO-ND suspensions. Reprinted with permission from Ref. [108]. Elsevier, 2019.

2.4. Lubrication Mechanisms of Graphene-Family Materials as Additives

The friction behavior with graphene-family materials as additives is a complex physical-chemical process, especially at the macroscale, leading to different tribological behaviors reported in the literature (Table 1). Various lubrication mechanisms have been proposed.

Table 1. Statistical data of studies related to graphene-family additives in liquid lubrication. Reproduced with permission. [15,139].

Additive (Fraction), in Base Liquid	Test Mode	Specimen Details	Test Parameters	Results		Lubrication Mechanisms
				COF Reduction	Wear Reduction	
GO (0.1 wt%), in 150 SN	Upper disk on the lower ball, Rotary	Ball: X45Cr13 steel, 52−54 HRC, Φ8 mm. Disk: X155CrVMo12-1 steel, 60 HRC, Ra 0.5 µm, Φ105 mm,	0.05−2.1 m/s; 30, 60, 90 N; 25, 50, 80 °C	30%	27%	Deposited GO protective layer [140]

Table 1. *Cont.*

Additive (Fraction), in Base Liquid	Test Mode	Specimen Details	Test Parameters	Results		Lubrication Mechanisms
				COF Reduction	Wear Reduction	
Zinc borate/GO composite (2 wt%), in 500 SN	Four-ball	—	1200 rpm; 147 N; Test 2 h	48.2%	40%	Protective layer [141]
Cu nanoparticles decorated on polydopamine functionalized GO (0.1 wt%), in soybean oil	Ball on disk	Ball: GCr15 steel, Φ9.525 mm, 62 HRC. Disk: 45#steel	100−500 rpm; 1−12 N; Test 0.5 h	57%	27%	Tribolayer [113]
Modified graphene (0.075 wt%), in 350 SN	Four-ball	GCr15A steel, Φ12.7 mm, 61 HRC, Test standard: ASTM D4172-82	1200 rpm; 147 N; 75 ± 2 °C; Test 1 h	37%	—	Protective layer [142]
Single layer GO (0.06%), in water	Ball on three disks	Ball: Cr alloy steel, Ra 11.1 ± 0.4 nm, 64 HRC. Disk: AISI304 stainless steel, Ra 37.0 ± 6.2 nm, 92 HRB	50 mm/s; 20 N; sliding 7.5 m	44.4%	17.1%	Tribolayer [73]
Multilayer graphene (0.1 wt%), in benton grease	Ball on disk, reciprocation	Ball: AISI52100 steel, Φ10 mm, 710 HV. Disk: AISI52100, 664 HV	100−500 N; 10−50 Hz; Test 0.5 h	10.4%	25−50%	Tribolayer [143]
Graphene (0.01 wt%), in PAO4	Ball on disk	Ball: GCr15 steel, Φ9.525 mm. Disk: Bronze contained elliptical dimple (area ratios: 0, 5%, 10%, 20%)	5 mm/s; 5 N; Sliding 8 mm; 25, 60, 100, 150 °C Test 1.67 h	78%	90%	Tribolayer and texturing [36]
CeO_2-decorated graphene (0.06 wt%), in paraffin oil	Reciprocation, test method: ASTM D6425-05	Ball: GCr15 steel, Φ10 mm, Ra 20 nm, 62 HRC. Disk: GCr15 steel, 792 HV, Ra: 50 nm	6 cm/s; 15 Hz; 50 N; 17% RH; Test 0.5 h	52%	1.5%	Transfer layer and nanoparticle spacer between graphene sheets [129]
ZrO_2/rGO composite (0.06 wt%), in paraffin oil	Reciprocation, test method: ASTM D6425-05,	Ball: GCr15 steel, Φ10 mm, Ra 20 nm, 62 HRC. Disk: GCr15 steel, 790−820 HV, Ra 50 nm	6 cm/s; 50−450 N; 25 ± 5% RH; Test 0.5 h	56%	6.4%	Protective layer and ball bearing [123]
Graphene (23.8−110 μg/mL), in water	Ball on disk, rotary	Ball: GCr15 steel, Φ9.53 mm. Disk: GCr15 steel	62.8−251.2 mm/s; 2−15 N; 25 ± 5% RH; Test 0.5 h	81.3%	61.8%	Fluid adhesive layer and graphene protective layer [144]

Table 1. *Cont.*

Additive (Fraction), in Base Liquid	Test Mode	Specimen Details	Test Parameters	Results		Lubrication Mechanisms
				COF Reduction	Wear Reduction	
Single layer graphene (1 mg/mL), in ethanol	Ball on disk	Ball: 100Cr6 steel, Φ4 mm. Disk: Iron (99.98% pure) and bronze (98% Cu and 2% Sn), Ra 30 nm	100 mm/s; 1 N; 50% RH	48%	—	Chemical passivation of iron by graphene [145]
Silica/GO composite (0.125 wt%), in EG	Pin on disk, rotary	Ball: AISI420 steel, Φ12.7 mm. Disk: AISI52100 steel, Φ40 mm, Ra < 0.1 μm	0.008 m/s; 68.9 N; 50% RH; Test 1 h	38%	31%	Tribolayer and ball bearing [135]
Modified GO, in oil/water emulsion	Ball on ring	Ball: bearing steel, Φ25.4 mm, 65 HRC, Ra 30 ± 10 nm. Ring: AISI52100 steel, 65 HRC, Ra 100 ± 50 nm	20−200 mm/s; 0.5 N; 25 ± 5% RH; sliding 100 m	18%	48%	Transfer layer, adsorption layer, and tribolayer; and the lubricity of emulsion droplets [146]
Graphene (0.01 wt%), in Span-80/PAO4	Ball on disk, reciprocation	Ball: GCr15 steel, Φ4 mm, 766 HV. Disk: RTCr2 alloy cast iron, 220 HV, Ra ≤ 0.03 μm	—	—	50% on normal surface, 90% on textured surface	Polishing, self-repairing, and tribolayer [147]
GO (2 mg/mL) in ethylene glycol aqueous solution	Ball on disk, rotary	Ball: Si_3N_4, Φ4 mm, Disk: SiO_2	0.1 m/s; 3 N; 10−25% RH	Superlubricity with COF ≈ 0.004	99.5% compared to ethylene glycol aqueous solution	Adsorbed GO; low shear stress between GO layers and the interface between GO and ethylene glycol aqueous solution, and the formation of hydrated networks [89]
GO quantum dots (1 mg/mL) in ethylene glycol aqueous solution	Ball on disk, rotary	Ball: Si_3N_4, Φ20 mm, Disk: sapphire	0.1 m/s; 15 N; 10−25% RH	Superlubricity with COF of 0.0068	90%	Formation of tribofilm containing GO quantum dots [93]

For the graphene-family additives, well-accepted lubrication and wear-resistance mechanism is the formation of graphene-containing tribofilm, which has been observed in both oil-based [38–43] and water-based lubrication systems [72,89]. However, the investigation of the formation and function of adsorbed graphene-family during friction is still at an early stage. There are still many opening questions to be solved. It was proven that the graphene-containing tribofilms can effectively reduce friction and wear, but the studies related to the formation of graphene-containing tribofilms are still insufficient. Ge et al. [92] found that the GO-NH_2 exhibited better lubrication performance compared to GO with -OH and -COOH groups, which was attributed to the better robustness of GO-NH_2 tribofilm arising from the larger adhesive force between functional groups of GO-NH_2 and contact surfaces. In addition to the adhesion force, tribochemical reactions between graphene-family materials also have potential influence on the tribofilm formation, thus the tribological behaviors. For fluorinated graphene, the formation of metal-F bonds during

friction is believed to be an important mechanism for the formation of robust tribofilm and better macroscale lubrication performance [28]. In addition to the functional groups, it was found that the size of graphene-family materials also influences the formation of tribofilm and the lubrication behavior. Li et al. [148] investigated the influence of flake size and concentration of graphene as additive of linear alpha olefin on the lubrication behaviors using molecular dynamics simulation. It was found that when the size of graphene flakes is larger than 40 carbon atoms, the graphene flakes can be anchored on the surface of friction pairs to form a protective tribofilm, leading to a passivated and smoothened sliding interface and ultralow COF (Figure 8).

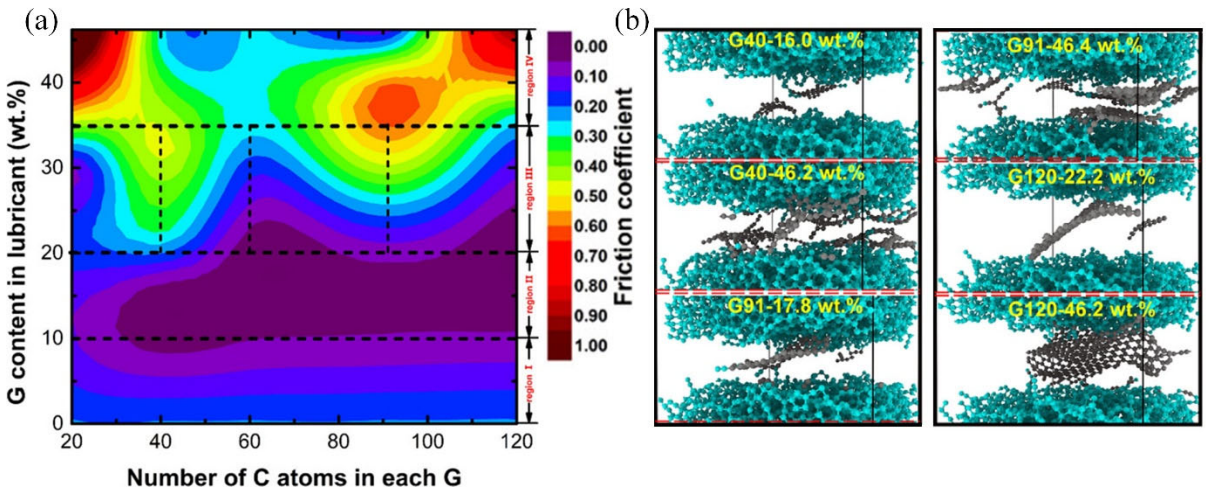

Figure 8. Effect of size and concentration of the graphene as additive on the COFs. (**a**) Change of COF with size and concentration of graphene; (**b**) interfacial structures containing graphene with different sizes and concentrations (the molecules of base oil are neglected for better observation of the structure of graphene). Reprinted with permission from Ref. [148]. American Chemical Society, 2020.

In addition to the formation of graphene-containing tribofilm, understanding how the graphene-containing tribofilms perform lubrication performance is also important. With the protection of graphene-containing tribofilm, the direct contact between asperities under boundary or mixed lubrication conditions can be prevented by the tribofilms, providing a low shear strength, and thus enhancing lubrication and wear-resistance performance. In addition, the wear interaction between graphene-containing tribofilm and lubricant molecules is also believed to be an important mechanism for the lubrication performance of graphene-family additives. A new strategy was proposed by Li et al. [149] to investigate the interaction between water molecules and graphene surfaces. A highly hydrophobic surface of self-assembled fluoroalkyl monolayers was prepared on the SiO_2 tip for the friction tests conducted using AFM. A superlubricity state with an extremely low COF of 0.0003 was obtained between the modified SiO_2 tip and graphene surface in a water environment, where the weak interaction between graphene surface and water molecules contributed to the achievement of superlubricity (Figure 9). The influence of weak interaction between graphene and liquid molecules on the lubrication performance was also verified by molecular dynamics simulations. It was found that the graphene tribofilm can promote the mobility of water molecules [150] or oil molecules [151,152], leading to a lower COF. In addition to those mechanisms, the nano-ball bearing [153], self-repairing [154], the micro-polishing effect [155], tribochemical reaction [57], and the incommensurate contact between graphene-family materials and other nanomaterials were also proposed as lubrication mechanisms for the graphene-family materials as additives.

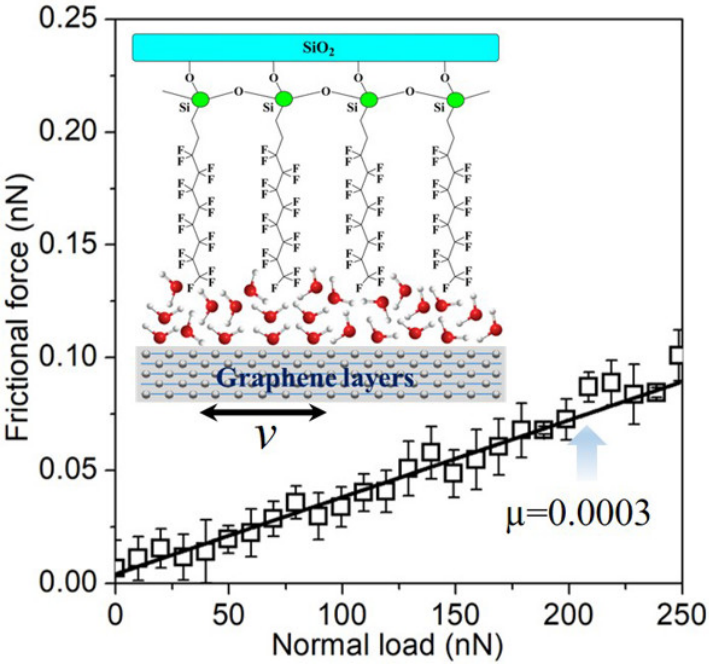

Figure 9. Superlubricity achieved by self-assembled fluoroalkyl monolayers and graphene surfaces in a water environment. Reprinted with permission from Ref. [149]. American Chemical Society, 2019.

2.5. Engineering Applications of Graphene-Family Materials as Additives

With decades of development, researchers have also focused on the commercialization and engineering applications of graphene-family materials [156,157]. The function of graphene-family materials as lubrication additives for engineering applications was also investigated. Rasheed et al. [158] investigated the heat transfer and tribological performance of engine oil with graphene as nano-additive using an internal combustion engine. It was found that with 0.01 wt.% of graphene, COF can be reduced by 21%, and the thermal conductivity of lubricant can be enhanced by 23%. In addition, heat transfer rate of the engine can be also enhanced by 70%. The wear of engine can be also suppressed with graphene nanoflakes as additives. The potential applications of graphene-family additives for machining process were also investigated. Baldin et al. [159] studied the effect of graphene addition in cutting fluids applied by minimum quantity lubrication (MQL) for end milling of AISI 1045 steel. It was found that the addition of graphene nanoflakes can enhance the lubrication performance of cutting fluids. For some cutting fluid, the addition of graphene nanoflakes leads to increased tool life, while for some cutting fluid, the addition of graphene nanoflakes can leads to reduced tool life. Li et al. [160] investigated the performance of vegetable oil-based cutting fluid dispersed with graphene nanoparticles for the MQL milling of Ti6Al4V. It was found that the graphene additive could enhance the cooling and lubrication performances of the oil film formed at the milling zone, leading to reduced milling temperature, milling force, tool wear, and enhanced tool life. The influence of graphene-family additive for drilling [161] and hard-turning [162] was also investigated, finding that graphene-family materials as additives could provide excellent lubrication, and improved surface roughness. Graphene-family additives have also been added into drilling fluid for the petroleum industry [157] to enhance the fluid loss control, rheology properties, and lubrication and wear-resistance performances. Those studies indicate that graphene-family materials are promising for various industrial applications.

3. Conclusions and Perspectives

Graphene-family materials have been investigated by researchers as promising additives for various lubrication systems due to the unique physical-chemical properties. It has been proven that graphene-family materials can lead to enhanced lubrication and

wear-resistance performance. Experimental, theoretical, and simulation studies have been performed to investigate the tribological behaviors of graphene-family materials as additives, and various function mechanisms have been proposed. Over the years, many breakthroughs have been achieved, but there are still problems for both scientific research and engineering applications.

- For graphene-family additives, dispersion stability is an important issue related to the instability in oil or water-based lubrication systems. However, the high temperature introduced wear debris, and the material degradation all have potential influence on the long-term stability of graphene-family additives, which still need further investigation.
- The cost of graphene-family materials is still high for industrial applications. Developing a large-scale, low-cost preparation process is important for the practical application of graphene-family materials as additives.
- There is still a lack of widely accepted criteria for the designing of graphene-family materials as additives. For example, the optimized parameters such as particle size, layer numbers, types, and concentration of functional groups for specified application condition are still unclear. An in-depth investigation of the fundamental mechanisms and advanced techniques [163–165] for the guidance of designing and application of graphene-family materials as additives is needed in the future.

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