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**REVIEW ARTICLE**

# Delivering quantum dots to lubricants: Current status and prospect

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**Abstract:** Very recently, two-dimensional quantum dots (2D QDs) have been pioneeringly investigated as lubricant additives, which exhibit superior friction-reducing and wear resistance. Compared with 2D nanoparticles, 2D QDs possess small size (~10 nm) and abundant active groups. These distinguished advantages enable them to quickly disperse into common lube mediums and maintain long-term storage stability. The good dispersion stability of 2D QDs not only effectively improves their embedding capacity, but also enables continuous supplements of lubricants during the sliding process. Therefore, 2D QDs are attracting increasing research interest as efficient lubricants with desirable service life. In this review, we focus on the latest studies of 2D QDs as liquid lubricant additives (both in polar and nonpolar mediums), self-lubricating solid coatings and gels, etc. Various advanced strategies for synthesis and modification of 2D QDs are summarized. A comprehensive insight into the tribological behavior of a variety of 2D QDs together with the associated mechanism is reviewed in detail. The superior lubricating performances of 2D QDs are attributed to various mechanisms, including rolling effect, self-mending performance, polishing effect, tribofilm formation, nanostructure transfer and synergistic effects, etc. Strategies for friction modulation of 2D QDs, including internal factors (surface modification, elemental doping) and extrinsic factors (counter surfaces, test conditions) are discussed, special attentions for achieving intelligent tribology toward superlubricity and bio-engineering, are also included. Finally, the future challenges and research directions regarding QDs as lubricants conforming to the concept of "green tribology" toward a sustainable society are discussed.

**Keywords:** quantum dots (QDs); design diversity; dispersibility; embedding stability; lubrication

## 1 Introduction

Tribology including friction and wear, has become a serious and challenging issue closely linked to energy crisis, carbon footprint, and equipment life [1–3]. Accordingly, tremendous efforts have been involved in developing highly efficient lubricants to reduce friction and wear. Lubricants mainly include solid lubricants and liquid lubricants. Thereinto, solid lubricants have been extensively adopted to minimize friction and wear for some specific conditions, such as heavy loads, high speeds, etc. [4, 5]. However, their tribological properties are sensitive to external environment, and the solid lubricant coatings will be worn out after serving for a long time. Historically, a variety of natural oils, such as castor oils, olive oils, coconut oils, lard oils, sperm oils, etc., are used to reduce friction and wear by creating a low-shear

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boundary film [6–8]. However, the disadvantages of natural oils, including inferior cold flow properties, poor oxidation stability and degradation, etc., severely restrict their prospective applications. Water-based lubricants are attractive as green lubricants owing to their exceptional features, such as good recyclability, excellent cooling capability, and eco-friendliness [9, 10]. Unfortunately, water itself is a poor lubricant owing to its relatively low viscosity, corrosion, poor lubricity, and poor film-forming/embedding ability. Therefore, it is essential to develop efficient liquid lubricant additives to improve their lubrication properties.

Since the first discovery of graphene in 2004, two-dimensional (2D) nanomaterials, including hexagonal boron nitride (*h*-BN), metal disulfide ($MoS_2$, $WS_2$), MXenes, and black phosphorous (BP), etc., have been extensively studied as attractive lubricants and/or additives due to their unique chemical, electrical, thermal, and mechanical properties [11–15]. In detail, the lamellar 2D nanosheets can easily slide against each other owing to the weak interlayer interaction, while the strong in-plane bonding endows them with remarkable mechanical strengths. In 2020, Ji et al. reported that the mechanical and tribological performances of nanocomposites would be obviously improved through the integrating with 2D nanomaterials [16]. However, 2D nanoparticles cannot be stably dispersed in common lube oils and tend to aggregate severely into clusters, which eventually precipitate out of the suspension according to the Derjaguin–Landau–Verwey–Overbeek (DLVO) theory.

Very recently, 2D quantum dots (QDs), naturally possessing fascinating advantages of 2D nanosheets, have attracted increasing attention in the field of tribology [17, 18]. Compared with traditional 2D nanoparticles, 2D QDs can be quickly dispersed into common lubricating mediums and maintain long-term storage stability benefiting from their high chemical activity and small particle size (~10 nm). The excellent dispersibility could avoid the complicated, time-consuming, and environmental pollution dispersion approaches as well as the influences of other dispersants. Particularly, the homogeneous dispersions endow 2D QDs improved film-forming and embedding stability through physical adsorptions (such as van der Waals) or interfacial tribo-chemical reaction [19]. Moreover, 2D QDs are small enough to *in-situ* fill the scratches of the rubbing surfaces, exhibiting self-healing/mending effects [20–22]. Consequently, the above-mentioned distinguished advantages of 2D QDs endow them to realize attractively efficient lubrication performances compared with traditional nanoparticles.

This review focuses on a comprehensive discussion of the latest research progress on 2D QDs in tribology. To begin, the advanced fabrication and versatile modified strategies of 2D QDs will be introduced. The particular advantages of 2D QDs are discussed in tribology fields, including good dispersion stability and diverse functionality. These distinguished advantages enable 2D QDs exceptional lubrication performances. Subsequently, the tribological behaviors of 2D QDs and corresponding mechanisms are discussed, which mainly include rolling effects, formation of protective films, mending effects, polishing effects, and synergistic effects. Then, recent advances for modulating the frictional performances are summarized. Finally, the prospects of extensively exploring QDs as lubricants toward sustainable future are discussed.

## 2 Why QDs?

QDs feature small size and abundant functional groups that have opened up new possibilities for effectively decreasing friction and wear. This review summarizes the latest advances of exploring QDs as attractive lubricant additives, solid coatings, and gels along with modulations strategies (mainly including surface functionalization, and elemental doping) as presented in Fig. 1.

### 2.1 Design diversity and modification

Since the first discovery of carbon QDs (CQDs) in 2004 [23], a growing number of literatures have emerged to exploiting fabrication methods of QDs, which can be mainly divided into two strategies: "top–down" and "bottom–up" approaches. The former one relies on breaking down larger bulks into smaller ones through dozens of chemical, electrochemical, or physical

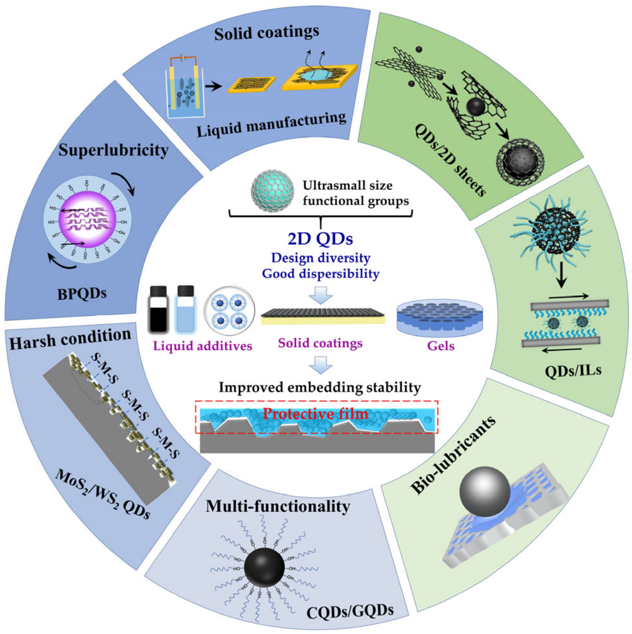



**Fig. 1** Overview of featured tribological performances, and modulation strategies of 2D QDs as lubricant additives, solid coatings, and gels.

approaches, such as acidic oxidation, hydrothermal method, laser ablation or electrochemical exfoliation, etc. [24, 25]. While the ''bottom-up'' methods synthesize QDs by carbonization of some inexpensive molecular precursors, such as citrate acid, glutamic acid, and carbohydrates, etc. [26–28]. Furthermore, modification strategies, e.g., surface functionalization and elemental doping, have been employed to elaborately design QDs to drive further task-specific multi-functionality [29, 30].

### 2.2 Promising physicochemical properties

#### *2.2.1 Good compatibility*

The polar groups (including hydroxyl groups, carboxyl groups, etc.) of QDs enable them with good affinity in most polar solvents, including water, polyethylene glycol (PEG), and ionic liquids (ILs), etc. Moreover, QDs with high chemical activity are conducive to be modified with target groups to improve colloidal stability and homogeneous distribution in nonpolar mediums, thus switching their affinity from hydrophilicity to hydrophobicity. Accordingly, QDs would exhibit switchable dispersibility and long-term stability in both polar and nonpolar mediums [31, 32]. For instance, Ye et al. modified CQDs with an organic agent via a chemically covalent grafting, and the modified CQDs in nonpolar polyalphaolefin (PAO) oil exhibited desired long-term stability and excellent lubrication performances [33].

#### *2.2.2 Improved embedding/film-forming ability and self-mending properties*

Owing to the small size and high surface area characters, 2D QDs are easy to enter into contact areas and form complete protection films via strong physical adsorption. Besides, the abundant active groups of 2D QDs enable them to react with freshly rubbing surfaces. This enables 2D QDs to exhibit improved embedding/film-forming abilities. Meanwhile, QDs can be deposited and/or adsorbed in bulges, cracks, and scratches of the rubbing surfaces, exhibiting "self-mending" effects. For instance, Huang et al. pointed out that the CQDs/$CuS_x$ nanocomposites in liquid paraffin oil exhibited excellent tribological properties and self-healing abilities on metal–metal surfaces by combining the advantages of graphitic CQDs and highly active $CuS_x$ nanoparticles [34].

#### *2.2.3 Multifunctionality: Anticorrosion and oxidation resistance*

For practical industrial applications, the corrosion resistance, especially for liquid lubricants, is a critical evaluation criterion. Recently, continuously encouraging progress has been reported that 2D QDs could form high-quality protective films on the tribo-pairs to prevent the penetration of oxygen and water molecules, thus are effectively impervious to corrosive gases and liquids [35]. For instance, Zhu et al. reported that introduction of carbon dots (CDs) could significantly improve the anticorrosion performances of polymer matrixes [36]. Cui et al. clarified that N-doped CDs (CDs-N) exhibited effective corrosion inhibition on carbon steels attributed to the chemical-/physical adsorptions as well as the special structures of CDs-N [37]. Besides corrosion, oxidative degradation of lubricants under oxygen atmosphere and metal catalysis, also threat their lubricating performances. Interestingly, Wang et al. demonstrated that chlorine-doped graphene QDs (Cl-GQDs) could exhibit ideal anti-oxidant activities via modification with functional dopants [38].

## 3 Essential tribological functions and corresponding mechanisms

### 3.1 Switchable compatibility with polar and nonpolar mediums

#### *3.1.1 CQDs and GQDs: Efficient aqueous lubricant additives*

Accordingly, the ideal liquid lubricant additives entail good compatibility with common polar and nonpolar mediums. In 2019, Hu et al. prepared water-soluble CDs by one-pot hydrolysis deriving from ammonium citrate (AC) (Fig. 2(a)) [39]. For comparison, graphene oxide (GO) was also prepared via prolonging the hydrolysis time of AC from 2 to 5 h. As shown in Fig. 2(b), the friction coefficient (COF) of CDs aqueous solution is 0.2671, which is much lower than that of GO (~0.3125) suspension under the identical test conditions (10 N, 0.1 mg/ml). Likewise, the wear rate (WR) of CDs is $1.5\times10^{-6}$ $mm^3/(N\cdot m)$, which is obviously lower than those of GO and bare AC solutions. The efficiency of CDs solution is contributed to the synergistic effects of rolling effects, formation of high-quality tribo-films, and self-repairing effects (Fig. 2(c)).

The load-carry capacity of lubricants is of great importance for practical engineering applications. Recently, Qiang et al. proved that load-bearing capacity of water-based lubricants can be obviously improved by introduction of an appropriate amount of GQDs [40]. Additionally, compared with GO nanosheets, GQDs dispersions exhibited better tribological performances in terms of a relatively a lower COF value of 0.23 and a longer service life (≥ 3,600 s) under a high load of 100 N. The superior tribological behaviors of GQDs can be described as follows: GQDs could effectively assemble on the steel–steel rubbing surfaces via physical adsorption or forming stable tribo-films,

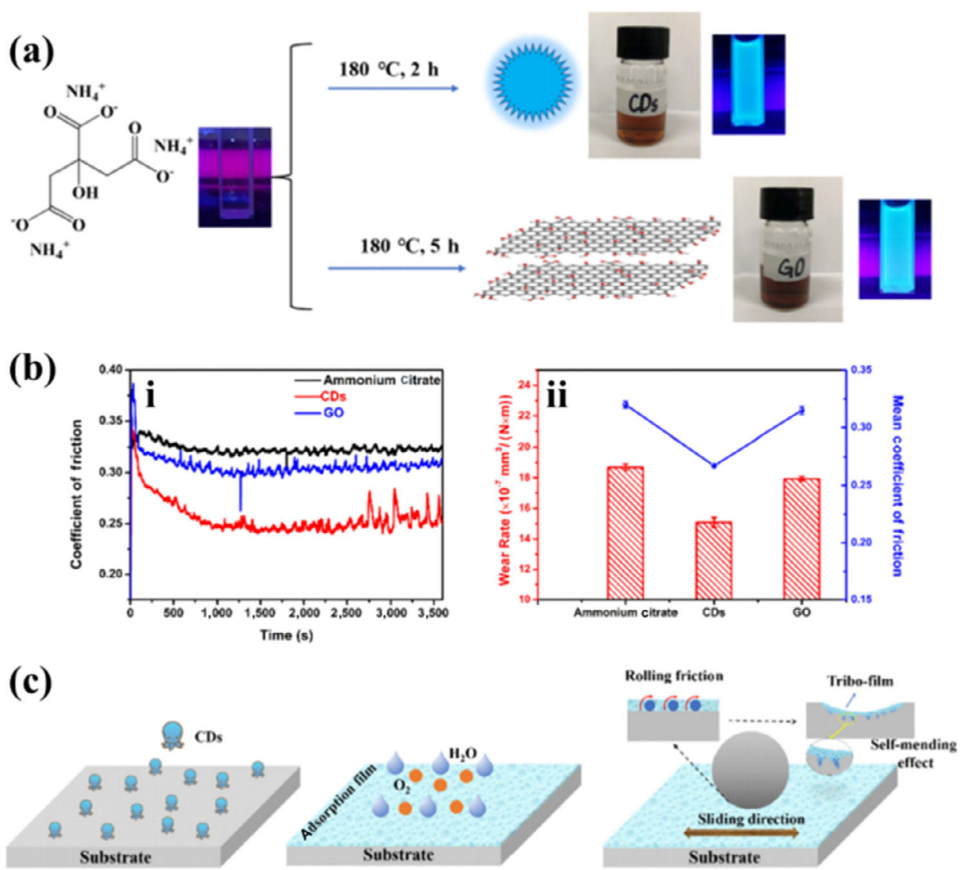



**Fig. 2** Fabrication and lubrication performances of CDs and GO: (a) schematic of preparation of CDs and GO, (b) (i) COF curves and (ii) mean COF and wear rate of AC, CDs, and GO aqueous solutions (0.1 mg/ml, 10 N), and (c) illustration of proposed lubrication mechanisms of CDs. Reproduced with permission from Ref. [39], © Elsevier Ltd. 2019.

and the excellent dispersion stability would ensure the continuous supply of GQDs in the rubbing interfaces during the sliding process. In contrast, GO nanosheets featured with microscale size would be easily pushed away from the contacting areas, resulting in a sharp increase of COF.

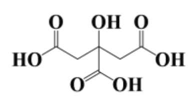
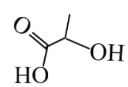
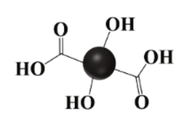
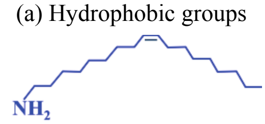
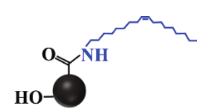
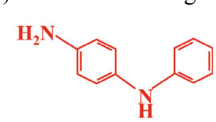
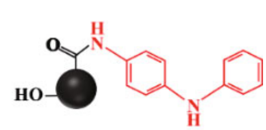
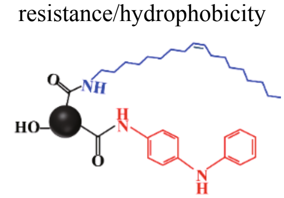
### *3.1.2 CQDs: Switchable polar and nonpolar lubricant additives*

The polar nature endows QDs excellent hydrophilicity, but indeed poses the question of their compatibility with nonpolar lubricating oils, such as PAO and mineral oils. For practical purposes, the hydrophobic nano-additives are largely needed because most of the vehicles and machines are lubricated by engine oils. Fortunately, QDs with abundant functional groups could easily switch from hydrophilicity to hydrophobicity by grafting target molecules or groups. From the viewpoint of the switchable compatibility required for lubricant additives, the modification strategies of QDs were listed in Table 1.

In 2018, Shang et al. synthesized hydrophilic nitrogen-doped CDs (N-CDs) with via a "bottom-up" method, and then modified the obtained N-CDs with hydrophobicity via a covalently grafting of alkyl chains from oleyl amine (OA) [41]. The hydrophilic and hydrophobic N-CDs disperse homogeneously in polar solvents, including PEG, $H_2O$, N,N-dimethylformamide (DMF), and nonpolar mediums (including PAO, toluene, and petroleum ether), respectively (Fig. 3(a)). Furthermore, the mean COF and wear volumes (WV) lubricated by hydrophilic N-CDs/PEG suspension account for 24.1% and 17.2% of those lubricated by PEG, demonstrating the improvement in the friction-reducing and anti-wear (AW) property with the addition of N-CDs. Likewise, the hydrophobic N-CDs in nonpolar PAO oil also exhibited improved tribological performances compared with those lubricated by pure PAO. As shown in Figs. 3(b) and 3(c), both the hydrophilic and hydrophobic N-CDs exhibited better tribological performances than traditional lubricant additives, such as bis(salicylato) borate ionic liquid (IL) and zinc dialkyldithiophosphates (ZDDP), respectively. In a follow-up study, He et al. prepared powdery GQDs by a one-pot gaseous detonation method, and further investigated their tribological performances in 150SN mineral oil. A low COF of 0.031 was achieved at an appropriate additive content of 0.8 wt% against a high load of 392 N, which is correspondingly improved by 65% of that lubricated by neat 150SN mineral oil. Additionally, GQDs play a vital role in reducing wear scar diameter (WSD) and depth [42]. Ye et al. designed and fabricated multi-functionalized CQDs (CQDs-N) through a one-pot hydrolysis method using organic 4-aminodiphenylamine (ADPA) as a multifunctional modifier [43]. The obtained CQDs-N could be suspended homogeneously in PEG solution with long-term storage stability (~5 months) owing to the strong interactions between oxygen groups deriving from CQDs surfaces and PEG chains. As expected, the CQDs-N in PEG exhibited lowest COF and WSD compared with CQDs and bare PEG, even under an extremely high load of 588 N.

Moreover, doping with metal ions offers another effective tool for improving the tribological behaviors of 2D QDs. The presence of metal ions endows 2D QDs

**Table 1** Summary of fabrication and modification of CQDs toward target-specific functions.

| Raw materials | Target-specific functions |
| --- | --- |
| Precursors  Citrate acid  Lactic acid |  Hydrophilic CQDs |
| Modifier agents (a) Hydrophobic groups  Oleyl amine (OA) |  Hydrophobic CQDs |
| (b) Multifunctional agents  4-Aminodiphenylamine (ADPA) | Anticorrosion/oxidation resistance  |
| | Anticorrosion/oxidation resistance/hydrophobicity  |

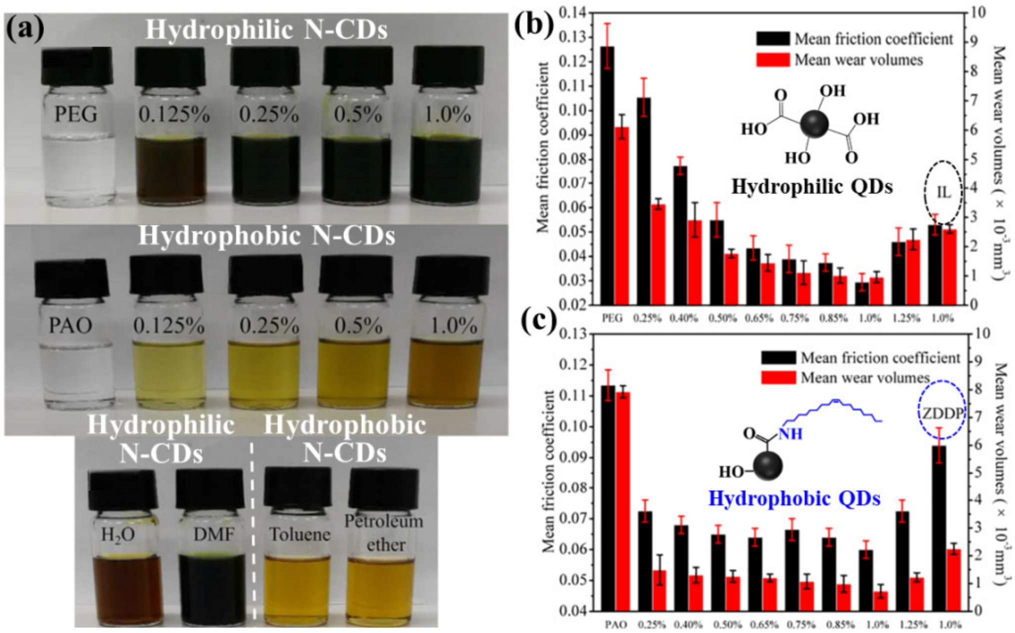



**Fig. 3** Dispersions and tribological characterizations of N-CDs: (a) digital images of N-CDs dispersions. Mean COF and WV of (b) hydrophilic N-CDs in PEG, and (c) hydrophobic N-CDs in PAO, respectively. Reproduced with permission from Ref. [41], © Elsevier B.V. 2018.

with improved chemical activity, which is beneficial to form a high quality tribochemical film. Recently, Tang et al. prepared a series of metal ions doped CDs ($Fe^{2+}$, $Cu^{2+}$, $Mg^{2+}$, and $Zn^{2+}$), and the synthesized metal-doped CDs exhibited excellent dispersion stability in water (> 3 months) [44].

Specifically, the load-carrying capacity was significantly enhanced from 120 N to no less than 500 N. Furthermore, the Zn-CDs exhibited good dispersion stability and lubricating performances in PEG via an anion replacing with $N(CF_3SO_2)^{2-}$. Table 2 summarizes the recent advances of CDs and GQDs as lubricant additives in both polar and nonpolar mediums.

### 3.1.3 *Solid coating*

Solid lubricant coatings can provide lubrication function for sliding tribopairs under boundary lubricating conditions, which is an efficient strategy to facilitate precise devices to run smoothly and safely [45, 46]. The good dispersion stability of 2D QDs is an important advantage for solution-based coatings. Recently, the solution-based coatings, including self-assembly approaches (e.g., solvent vaporization) and electrophoretic deposition (EPD), have been intensively used to realize large-scale and uniform solid coatings of 2D QDs. In 2020, Qiang et al. deposited GQDs coatings (GQDCs) on silicon (Si) substrate via an EPD approach, the thickness of GQDCs can be well-controlled via varying the deposition voltage [47]. Compared with GO coatings (GOCs) and bare Si substrate, the lowest COF and AW performance of GQDCs was immediately achieved owing to the synergistic effects of laminar slip effect and the formation of protective films. Moreover, the counterpart materials also influenced the interfacial nanostructures transformation and bonding state, thus affecting the friction properties. For instance, Yin et al. coated GQDs on different hydrogenated carbon films, involving graphite-like carbon (GLC), diamond-like carbon (DLC), and polymer-like carbon (PLC), and compared their tribological performances against self-mated counterparts or neat steels [48]. The lower COF values of self-mated counterparts are observed than those sliding against bare steel tribocouples. Specifically, the tribo-system containing GQDs coating on DLC surface is able to achieve superlubricity, which might attribute to the synergy of GQDs coating and the hydrogenated DLC film. However, the service life of this trio-system is very short (~115 s) due to their high sensitivity to environmental humidity.

**Table 2** Tribological results of CDs and GQDs as lubricant additives in polar and nonpolar mediums.

| Lubricant | Content (wt%) | Base liquid | Methodology (▼top-down ★bottom-top) | Friction mechanism | Tribo-pairs | Tribo-test | COF | Wear | Dis-persion stability |
|---|---|---|---|---|---|---|---|---|---|
| CDs | 0.01 | Water | ★Pyrolysis | Tribo-film; self-mending; rolling friction | Ball-on-disk ($Al_2O_3$/316 stainless steel) | 10 N $^a$, 5Hz $^b$, 5 mm $^c$, 60 min $^e$, RT $^f$ | 0.2671 | $1.5\times10^{-6}$ $mm^3$/(N·m) $^h$ | >6 months |
| GO | | | | | | | 0.3125 | $1.8\times10^{-6}$ $mm^3$/(N·m) $^h$ | >6 months |
| GQDs | 0.4 | Water | ▼Hydrothermal | Tribo-film | Ball-on-disk (GCr15 steel) | 100 N $^a$, 20 Hz $^b$, 1 mm $^c$, 60 min $^e$, RT $^f$ | 0.23 | $2.2\times10^{-12}$ $mm^3$ $^g$ | |
| GO | | | | | | | 0.305 | $1.76\times10^{-3}$ $mm^3$ $^g$ | |
| Hydrophilic N-CDs | 1.0 | PEG | ★Covalent grafting | Tribo-chemistry; boundary lubrication film | Four-ball (GCr15 steel) | 392 N $^a$, 1,200 rpm $^d$, 60 min $^e$, 75 ℃ $^f$ | 0.0295 | $0.966\times10^{-3}$ $mm^3$ $^g$ | |
| ILs | | | | | | | 0.0525 | $2.6\times10^{-3}$ $mm^3$ $^g$ | |
| Hydrophobic N-CDs | | PAO | ★Covalent grafting | Tribo-chemistry; boundary lubrication film | | | 0.0725 | $0.85\times10^{-3}$ $mm^3$ $^g$ | |
| ZDDP | | | | | | | 0.0935 | $2.5\times10^{-3}$ $mm^3$ $^g$ | |
| Zn-CDs | 1.0 | Water | ★Pyrolysis | Ball-bearing; tribo-film | Ball-on-plate (GCr15 steel) | 500 N $^a$, 5 Hz $^b$, 5 mm $^c$, 20 min $^e$, RT $^f$ | ~0.15 | ~$2.4\times10^{-6}$ $mm^3$ $^g$ | >3 months |
| Zn-CDs-$NTf_2$ | 0.15 | PEG | | | | 100 N $^a$, 5 Hz $^b$, 5 mm $^c$, 20 min $^e$, RT $^f$ | ~0.10 | ~$1.4\times10^{-7}$ $mm^3$ $^g$ | |

Tribo-test: $^a$ Load/N, $^b$ Frequency/Hz, $^c$ Amplitude, $^d$ Sliding speed/rpm, $^e$ Time/min, $^f$ Temp/℃.
Wear analysis: $^g$ Wear volume (WV)/$mm^3$, $^h$ Wear rate (WR)/$mm^3$/(N·m).

### 3.2 Extreme operating conditions & superlubricity

With the rapid development of manufacturing, most of the modern equipment is required to work availably at severe conditions (including elevated temperature, high speed, heavy loads, and oxidizing atmosphere, etc.). Thus, it is essential to develop special lubricant additives available at extreme operating conditions. Moreover, superlubricity is leading to a revolution in engineering technology, which is an ideal state that friction between two sliding surfaces is negligible [49]. It is still a challenge to achieve superlubricity at a macro-scale and reveal its underlying mechanisms.

#### *3.2.1 $MoS_2$ and $WS_2$ QDs: Elevated temperature and heavy load*

As emerging 2D materials, $MoS_2$ and $WS_2$ nanosheets have demonstrated outstanding tolerance to high-temperatures and heavy loads owing to their lamellar structures and unique physicochemical properties. However, the realization of dispersing $MoS_2$ and $WS_2$ nanosheets in common lube oils is still severely restricted by their solid characters. To address this problem, oil-soluble $MoS_2$ nanosheets have been reported via a surface modification [50]. The modified $MoS_2$ nanosheets exhibited superior extreme pressure properties (no less than 2,000 N) compared to other Mo-contained lubricant additives, e.g., molybdenum dialkyldithiophosphate (MoDDP) (600 N), and fullerene-like $MoS_2$ nanoparticles (IF $MoS_2$) (300 N), and micro $MoS_2$ (200 N).

Notably, the discovery of $MoS_2$ and $WS_2$ QDs opens up new possibility of lubricant additives at extreme operating conditions. In 2018, Wu et al. reported that $MoS_2$ and $WS_2$ QDs would exhibit better dispersion stability in polyalkylene glycol (PAG) than $MoS_2$ and $WS_2$ nanosheets (Fig. 4(a)), resulting in lower friction-reducing and AW behaviors under the identical conditions. XPS analysis proved that $MoS_2/WS_2$ QDs could bond with Fe atoms by Fe–S bonds, or iron oxide layer by M–O and S–O bonds, confirming the formation of tribo-films on the worn surfaces (Fig. 4(b)). The possible lubrication mechanism was described as follows: $MoS_2$ and $WS_2$ QDs are more conducive to enter into the contacting areas, creating high-quality boundary films and *in-situ* "filling" the asperity valleys (Fig. 4(c)). In contrast, $MoS_2$ and $WS_2$ nanosheets tend to be squeezed out of the contacting areas, resulting in gradually deteriorating lubrication performances [51]. Furthermore, Gong et al. revealed that $MoS_2/WS_2$ QDs could disperse stably in commercial IL, 1-butyl-3-methylimidazolium hexafluorophosphate ([BMIm]$PF_6$) (≥ 2 months).

Compared with neat ILs, the presence of $MoS_2/WS_2$ QDs would dramatically improve the friction-reducing and AW performances even under severe conditions (500 N, 150 °C), as shown in Fig. 4(d) [52]. It is quite attractive that $MoS_2$ QDs would also disperse stably in nonpolar paraffin oil and exhibit an extremely low COF of 0.061, which is much lower than that of bare paraffin oil (0.169) [53]. Table 3 summarizes the recent advances involving commonly used $MoS_2/WS_2$ QDs as lubricant additives.

### 3.2.2 BP QDs: Superlubricity

As emerging 2D materials, BP with armchair-zigzag orientation and attractive physicochemical properties

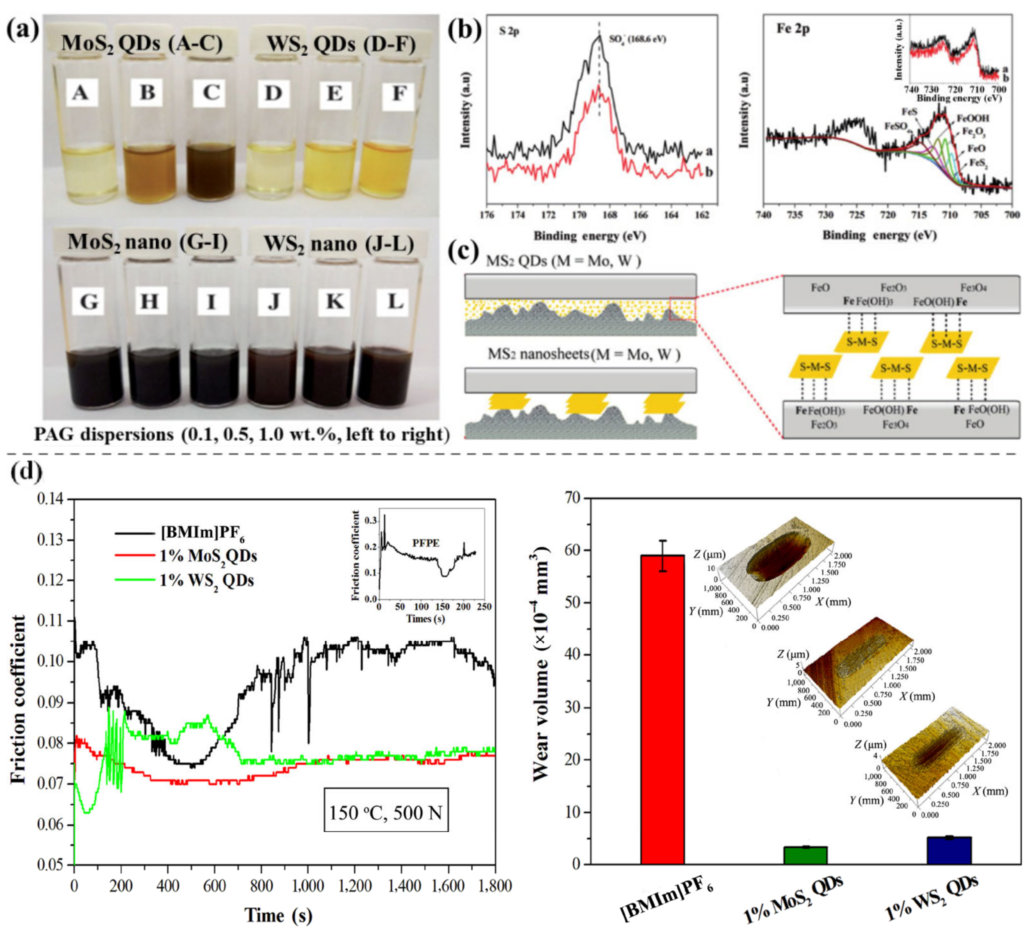



**Fig. 4** Dispersions and tribological performances of $MoS_2/WS_2$ QDs: (a) dispersions of PAG containing $MoS_2/WS_2$ QDs and $MoS_2/WS_2$ nanosheets, respectively, (b) XPS spectra of S 2p and Fe 2p of worn surfaces lubricated by 0.5 wt% $MoS_2/WS_2$ QDs in PAG, (c) scheme of possible lubrication mechanism of $MoS_2$ QDs and $MoS_2$ nanosheets. Reproduced with permission from Ref. [51], Wiley 2018. (d) COF and WV of ILs, and ILs with 1% $MoS_2/WS_2$ QDs (150 °C, 500 N), respectively. Reproduced with permission from Ref. [52], © The authors 2020.

has been explored to achieve superlubricity through experiments and simulation [54, 55]. For instance, BP nanosheets attached with hydroxyl groups can deliver robust superlubricity [56]. Compared to nanosheets, BP QDs are promising to deliver superlubricity owing to the competitive advantages of QDs. Ren et al. investigated an initial lubricating performance of BP QDs-ethylene glycol aqueous ($EG_{aq}$) suspension [57]. As shown in Figs. 5(a)–5(c), a "robust" and "durable" macroscale superlubricity state of BP QDs-$EG_{aq}$ suspension was observed, which exhibited an extremely low COF value in the range of ~0.001–0.003 for more than 2 h. As depicted in Fig. 5(d), the superlubricity and excellent AW performances of BP QDs-$EG_{aq}$ suspension were attributed to the synergy of rolling effects, interlayer sliding, hydrogen bond layers

**Table 3** Literature summary of commonly used $MoS_2$/$WS_2$ QDs-based lubricant additives.

| Lubricants | Content (wt%) | Base liquid | Methodology (▼top-down ★bottom-top) | Friction mechanism | Tribo-test Ball-on-disk (GCr15 steel) | COF | Wear | Dispersion stability |
|---|---|---|---|---|---|---|---|---|
| $MoS_2$ nanosheets | 1 | Liquid paraffin | ★Wet chemistry | Protective film | Load climbing test: load range from 100 to 2,000 N, load increase at a rate of 100 N/2 min $^{a}$, 50 Hz $^{b}$, 2 mm $^{c}$, >50 min $^{d}$, 120 °C $^{e}$ | ~0.1–0.15 | 1.39±0.06 mm $^{g}$ | |
| $MoS_2$ QDs | 0.5 | PAG | ▼Sonication/ Mechanical stirring | Boundary lubrication film | 100 N $^{a}$, 25 Hz $^{b}$, 1 mm $^{c}$, 30 min $^{d}$, 150 °C $^{e}$ | ~0.092 | ~0.06×10$^{-3}$ mm$^{3\,h}$ | >2 weeks |
| $MoS_2$ nano | | | | | | ~0.126 | ~1.9×10$^{-3}$ mm$^{3\,h}$ | |
| $WS_2$ QDs | | | | | | ~0.097 | ~0.07×10$^{-3}$ mm$^{3\,h}$ | |
| $WS_2$ nano | | | | | | ~0.123 | ~2.0×10$^{-3}$ mm$^{3\,h}$ | |
| $MoS_2$ QDs | 1 | $[BMI_m]PF_6$ | ▼Sonication/ solvothermal | Boundary lubrication film | 500 N $^{a}$, 25 Hz $^{b}$, 1 mm $^{c}$, 30 min $^{d}$, 150 °C $^{e}$ | ~0.074 | ~0.4×10$^{-3}$ mm$^{3\,h}$ | ≥2 months |
| $WS_2$ QDs | | | | | | ~0.076 | ~0.5×10$^{-3}$ mm$^{3\,h}$ | |
| $[BMI_m]PF_6$ | | | | | | ~0.093 | ~6×10$^{-3}$ mm$^{3\,h}$ | |

Tribo-test: $^{a}$ Load/N, $^{b}$ Frequency/Hz, $^{c}$ Amplitude, $^{d}$ Time/min, $^{e}$ Temp/°C, $^{f}$ Tribo-pairs.
Wear analysis: $^{g}$ Width of the wear scar /mm, $^{h}$ Wear volume (WV)/mm$^3$.

**Table 4** Literature summary of commonly used BP QDs-based lubricant additives.

| Lubricants | Content (wt%) | Base liquid | Methodology (▼top-down; ★bottom-top) | Friction mechanism | Tribo-pairs | Tribo-test | | COF | Wear | Dispersion stability |
|---|---|---|---|---|---|---|---|---|---|---|
| BP-OH nanosheets | 7 | Water | ▼Ball milling | Lamellar slip; water layer | Ball-on-plate ($Si_3N_4$/$SiO_2$) | 3 N $^{a}$, 56 mm/s $^{d}$ 836 MPa $^{e}$, RT $^{g}$ | | 0.0006–0.006 | | |
| BPQDs-$EG_{aq}$ | 20 | Water | ▼Ball-milling | Hydrogen bond layers; rolling; shear sliding; protective film | Ball-on-disc ($Si_3N_4$/ sapphire) | 3 N $^{a}$, 100 mm/s $^{d}$, 336 MPa $^{e}$, RT $^{g}$ | ~120 min $^{f}$ | ~0.001–0.003 | 106 μm $^{h}$ | >2 weeks |
| BP-$EG_{aq}$ | | | | Rolling; shear sliding; protective film | | | ~20 min $^{f}$ | ~0.0109 | 205 μm $^{h}$ | |
| BP powder | 0.005 | Base liquid $^{j}$ | | | Ball-on plate (GCr15steel) | 40 N $^{a}$, 10 mm/s $^{d}$, 20 min $^{f}$, 30 ℃ $^{g}$ | | ~0.48 | 1.25× 10$^{-3}$ mm$^{3\,i}$ | |
| BP QDs | | | ▼Ultrasonic method | Ball-bearing effect; tribo-film | | | | ~0.32 | 2.5× 10$^{-4}$ mm$^{3\,i}$ | >2 weeks |

Tribo-test: $^{a}$ Load/N, $^{b}$ Frequency/Hz, $^{c}$ Amplitude, $^{d}$ Sliding speed/(mm/s), $^{e}$ Contact pressure/MPa, $^{f}$ Time/min, $^{g}$ Temp/°C.
Wear analysis: $^{h}$ Wear scar diameter (WSD)/μm, $^{i}$ Wear volume (WV)/mm$^3$, $^{j}$ 2.0 wt% TEA aqueous solution.

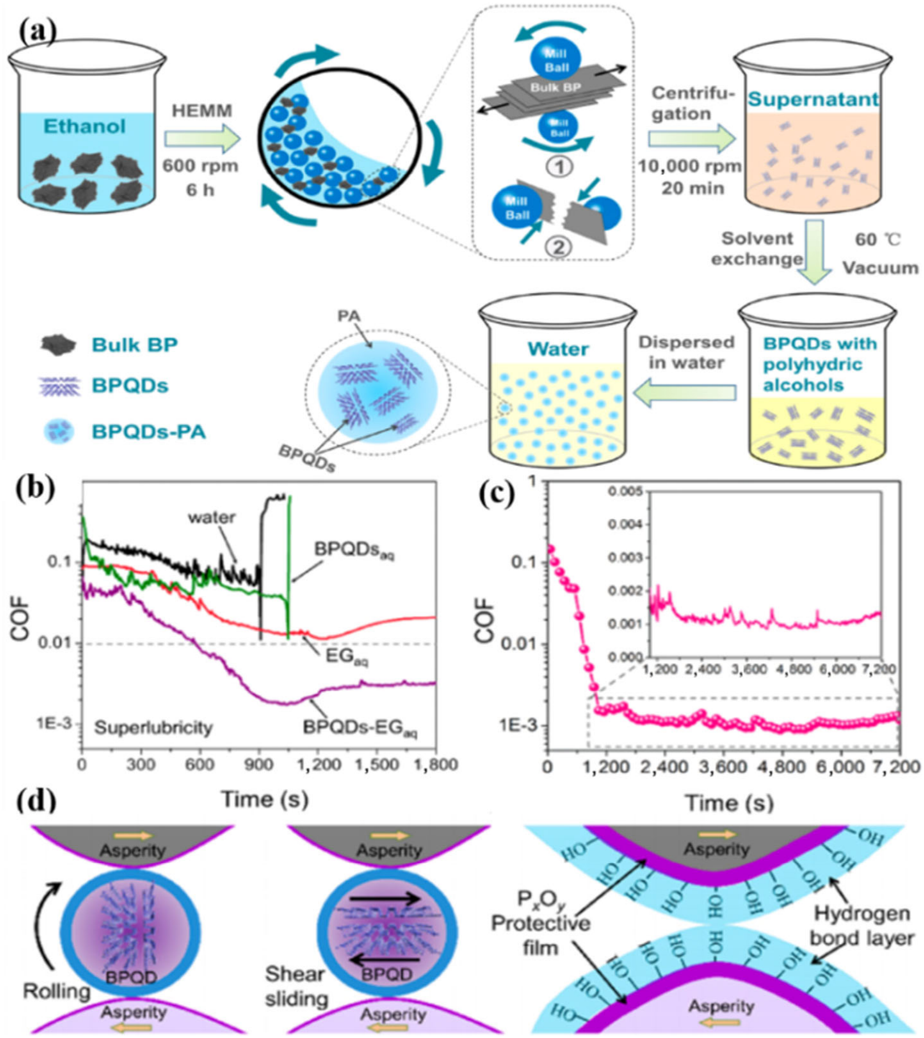



**Fig. 5** Fabrication and lubricating performances of BPQDs related lubricants: (a) schematic of preparation of BPQDs aqueous solution, (b) COF curves of water, $EG_{aq}$, $BPQDs_{aq}$, and BPQDs-$EG_{aq}$ solutions, (c) COF curves of BPQDs-$EG_{aq}$ solutions for 2 h, and (d) scheme of possible lubrication mechanism of BPQDs-$EG_{aq}$ solution. Reproduced with permission from Ref. [57], © American Chemical Society 2020.

between BP QDs, and formation of protective films consisted of oxidized BPQDs ($P_xO_y$). The tribological performances of BP QDs as water-dispersible lubricant additives were also assessed by Tang et al. [58]. Compared with bare base liquid (2.0 wt% TEA aqueous solution), the load-supporting capacity of 0.005 wt% BP QDs dispersion was dramatically improved from 120 N to over 300 N. The obtained BP QDs could maintain long-term period (~120 min) of stable lubrication effect at a relatively high load of 80 N. Additionally, the anti-friction and AW performances of small BP QDs are considerably better than those of the large-sized BP powder. These results can be explained that the BP QDs can easily to react with the freshly rubbing surfaces to form a robust boundary tribo-film, while BP powder might be squeezed from rubbing surfaces during the friction process.

## 4 Strategies for friction modulation

### 4.1 Impacts of substrates on interfacial friction: Tribo-pair effects

In general, friction behaviors at contact surfaces were largely governed by physical conditions of contact surfaces (roughness, hardness, etc.) and chemical interactions between the sliding interfaces. Though QDs exhibited promising lubricating behaviors, their lubrication effects and mechanisms vary with different tribopairs. The non-uniform protective film formed by QDs on the worn surface is usually not tough enough due to the weak interactions between QDs and sliding surfaces, and will eventually lose lubrication under harsh conditions (e.g., high load, high temperature, etc.). Therefore, further studies are necessary particularly focusing on the tribopairs effects.

Physically, amorphous carbon (a-C) coating is considered as an effective approach to achieve ultralow COF and excellent wear-resistance owing to their chemical inertness, low adhesiveness, and high smoothness. Tang et al. employed CDs aqueous solutions to enhance the lubricity for a-C/a-C contacts [59], and found that the introduction of 0.1 wt% CDs added to water exhibited a much low COF of 0.03 owing to the synergistic effect of CDs and the smooth a-C surfaces. Another effective approach for achieving super low friction is choosing appropriate tribological counterparts. The commonly used bearing steels are prone to react with most nanolubricants, leading to unexpected corrosive attack. Recently, polymers with suitable elastic modulus have been gradually employed as tribopairs. Among diverse polymers, polytetrafluoroethylene (PTFE) with self-lubrication and suitable Young modulus is considered as applicable tribopairs to conduct promising lubrication performances. Yin et al. suggested that the unprecedented lubrication of 2D $Ti_3C_2$/GQDs coating against PTFE surfaces should be assigned to the synergy of shielding effect, self-lubrication, rolling effect, slipping effect, and intercalation effect [45].

## 4.2 Influences of surface functionalization of QDs

### *4.2.1 QDs/2D nanomaterials: Synergistic effects*

Despite various successful reports in the literatures, commercialization of QDs as lubricants is still a great challenge. One of the major challenges is QDs will lose their ''nanorolling effect" under high-load conditions due to their ultrasmall size. Different from the high load bearing capacity of 2D nanosheets [60], the 2D QDs could be completely extruded out of the contacting surfaces under ultrahigh loads and thus lost the high load lubricating performances. Facing the challenge, our group proposed a universal strategy to improve the load-capacity of CQDs in synergy with 2D nanosheets [61]. The aqueous dispersions of CQDs decorated 2D nanosheets (*h*-BN, $MoS_2$, $MoSe_2$, $WS_2$, and graphene) were prepared using CQDs as efficient stabilizers and exfoliation agents through π–π interactivity (Fig. 6(a)). These CQDs decorated 2D nanosheets suspensions exhibited stable and desirable tribological performances compared with those of bare CQDs suspensions (Fig. 6(b)). Not only that, these CQDs/2D nanosheets suspensions also exhibited improved load-carrying capacity from 100 to 300 N, which could be attributed to the synergetic effects of 2D nanosheets and CQDs as depicted in Fig. 6(c).

Recently, chemical modified graphene (GO), is considered one of the main strategies to achieve stable graphene relevant aqueous solutions [62]. However, their tribological properties are severely limited due to the strong van der Waals force and π–π stacking interactions of GO sheets, leading to the severe concerns of dispersibility and storage stability. Considering the conjugated π structure, CQDs are expected to form strong noncovalent π–π interactions with GOs, preventing the stacking of GO nanosheets. In 2018, Shang et al. reported that CQDs/GO hybrid could dissolve in PEG with ultrahigh dispersion stability after 6 months [63]. The COF and wear volume of CQDs/GO hybrid in PEG were 0.039, $3.11\times10^{-3}$ $mm^3$, which accounted for 32.5% and 16.5% of those lubricated by neat PEG under a high load of 588N. The better lubricating properties of QDs/GO hybrid in PEG are contributed to synergistic effect of sphere-like CQDs and lamellar GOs layers.

### *4.2.2 GQDs/2D nanocomposites: Microstructure transformation*

As discussed above, the sliding-induced microstructure transformation of carbon-based nanomaterials is an important way for modulation of their lubricating performances, especially under high loads and fast sliding speeds. For instance, Wang et al. addressed that fullerene-like carbon would convert to graphene during the sliding process, thus exhibiting super-low COF values [64]. However, 2D graphene would be severely ripped under fast sliding speed, and eventually lost its lubrication effects. Therefore, it is important to deep understanding the structural evolution, interfacial interactions, and lubrication mechanisms of nanolubricants towards practical applications. Very

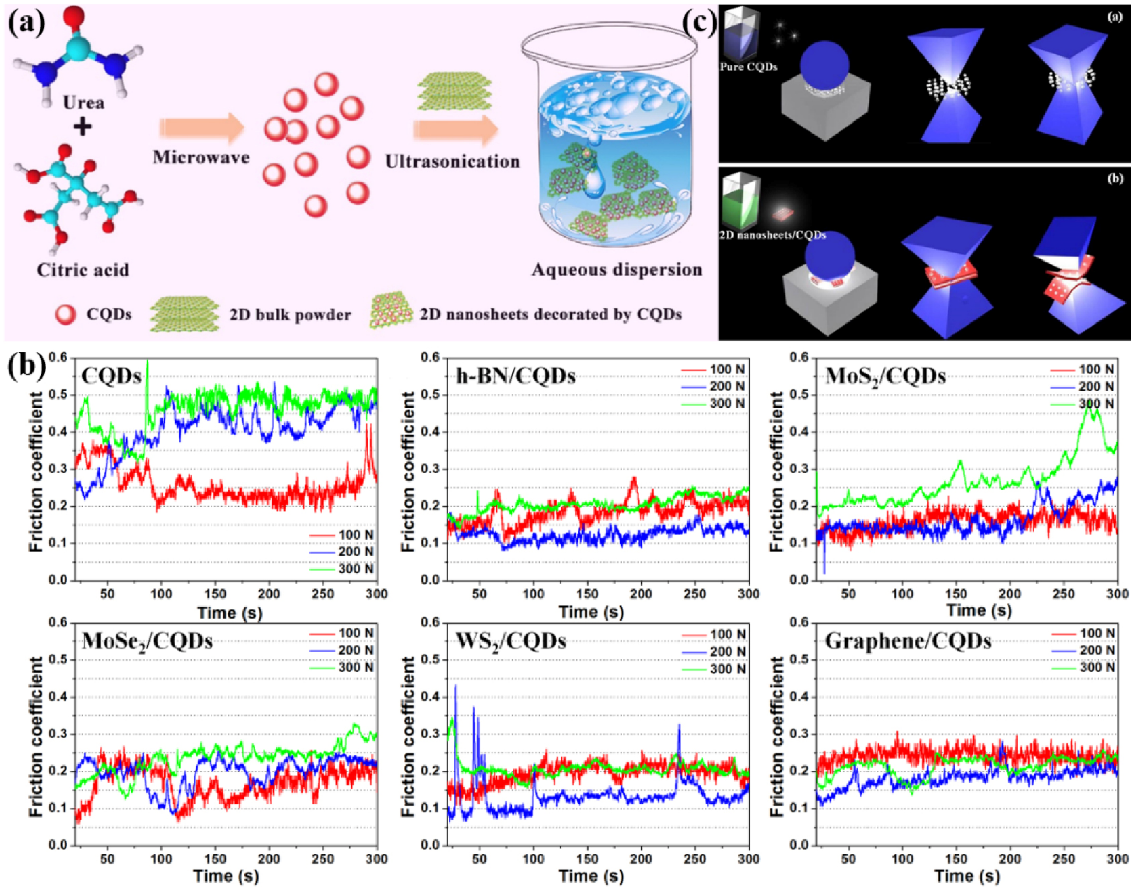



**Fig. 6** Fabrication and tribological performances of CQDs and 2D nanosheets/CQDs: (a) illustration of preparation, (b) COF curves of CQDs and 2D nanosheets/CQDs dispersions, and (c) their proposed lubrication mechanisms. Reproduced with permission from Ref. [61], © American Chemical Society 2016.

recently, molecular dynamic (MD) simulation has been employed to reveal and predict the frictional mechanisms of diverse nanoparticles during the frictional process [65]. Ma et al. verified that the a-C films underwent shear-induced $sp^3$-to-$sp^2$ structure transformation by using the MD simulations [66]. To achieve desirable lubrication performances, Yin et al. confirmed that the QDs with attractive interfacial activities can deliver promising lubrication behaviors via the strong intermolecular interactions with the rubbing surfaces and formation of lubricative tribofilms on the rubbing surfaces, which was supported by the MD simulation [48]. From the experimental aspect, Zhang et al. evaluated the tribological revolutions of graphene/GQDs suspensions in YG8 hard alloy contacts under a high sliding speed of 1,450 rpm [67]. The sulfurized isobutene (SIB) was employed as an extreme pressure (EP). Surprisingly, the graphene/GQDs/SIB performed exceptional effectiveness in reducing friction and wear. They attributed the attractive lubricating performances to the synergistic effect of SIB-assisting microstructures transformation from GQDs to fullerene QDs and the tribo-films formation during the severely sliding process (Fig. 7).

#### 4.2.3 *QDs/ILs: Improved durability and embedded stability*

Currently, ILs have received increasing attention owing to their good dispersibility, inherent polarity, and strong interfacial bonding [68]. Additionally, the structures and constituent ions of ILs are tailorable, which enable them either to adapt to various frictional contacts, or to meet some task-specific functions. However, the cost of ILs is relatively high, and some ILs containing halogen groups usually cause metal corrosion. Considering the excellent lubricating properties of QDs, integrating QDs with ILs, especially with halogen-free ILs [69], has becoming increasingly popular. Some strategies to synthesize ILs with QDs are summarized in Table 5, including one-pot pyrolysis, covalent grafting, and assembly, etc. Their lubricating behaviors will be discussed in the following section.

*i) QDs/imidazolium-based ILs*

Among diverse ILs, imidazolium-based ILs have attracted growing attention due to their relatively high thermal stability, and flexibility of molecular design. Tang et al. evaluated the lubricating properties of 1-aminopropyl-3-methyl-imidazolium bromide capped CDs (CDs/[AMIm][$Br^-$]) [70]. The results revealed that the obtained CDs/[AMIm][$Br^-$] exhibited lowest COF and wear volumes compared with those lubricated by ILs and CDs under parallel test

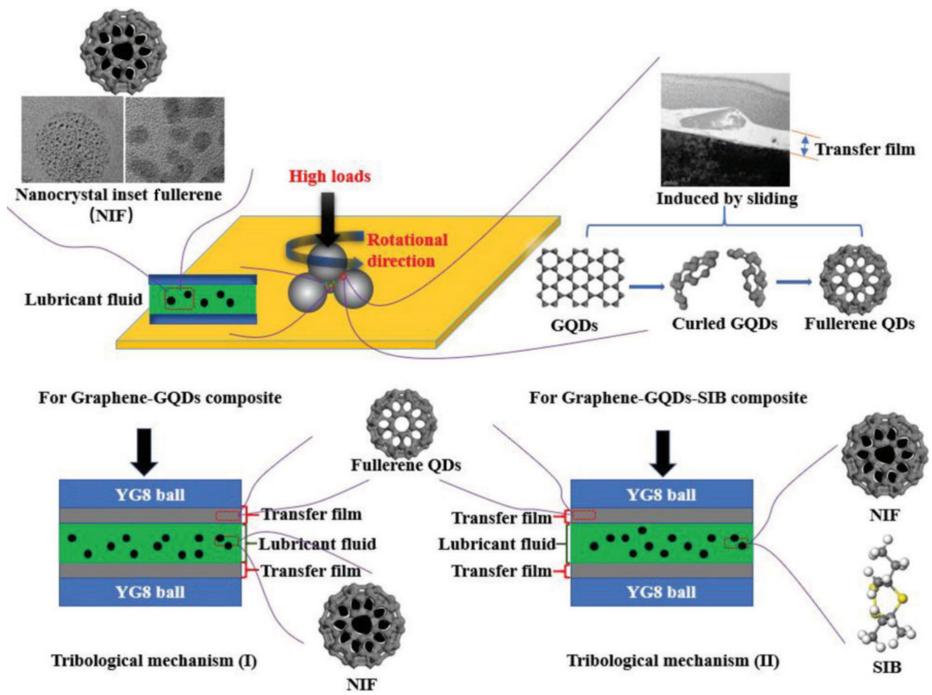
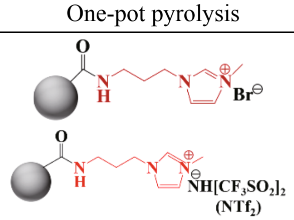
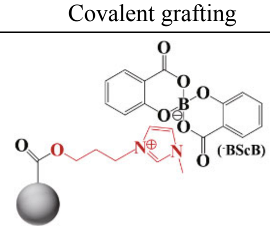
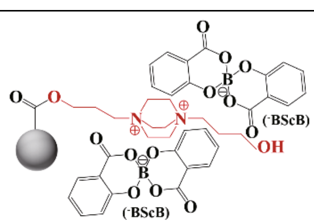
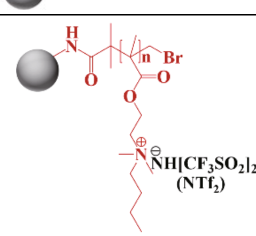
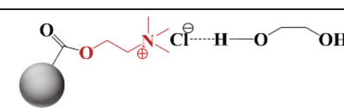



**Fig. 7** Schematic of sliding-induced microstructure transformation of graphene/GQDs. Reproduced with permission from Ref. [67], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2019.

**Table 5** Some strategies for synthesis of QDs/ILs.

| | One-pot pyrolysis | Covalent grafting | Assembly |
|---|---|---|---|
| a |  |  | |
| b |  |  | |
| c | | |  |

[a] Imidazolium based ILs, [b] Quaternary ammonia based ILs, [c] Deep eutectic solvents (DES)

conditions. Additionally, compared with ILs modified other conventional carbon nanomaterials, such as [AMIm][$Br^-$]/MWCNTs [71], the CDs/[AMIm][$Br^-$] exhibited better friction-reducing performances even with a smaller dosage. Moreover, to reduce the corrosion, the $Br^-$ anion group can be replaced with less corrosive $NTf_2^-$ anion by an anionic exchange reaction [72]. Additionally, ILs with some active elements (such as B, N, P, S) are also beneficial to form stable protective films via strong tribo-chemistry [73, 74]. For instance, Shang et al. reported that CQDs with chelated orthoborates IL (CQDs/[OHMim][BScB]) delivered an ultralow COF value of 0.037 under a high load of 588 N [75].

*ii) QDs/Quaternary ammonium-based ILs*

Recently, quaternary ammonium based ILs has attracted increasing attention owing to their exceptional advantages, such as low cost, easy synthesis, and good oil solubility. As pointed out previously, ILs or polyelectrolytes with abundant groups exhibit stable and extreme low COF due to the strong interactions/interplay between ions pairs of ILs and charged rubbing surfaces [76, 77]. Therefore, developing polymerized ILs or di-ionic ILs will provide more sufficient anchors on the rubbing surfaces, thus deliver more desirable lubricating behaviors. Mou et al. reported the polymerized quaternary ammonium/CDs hybrids (CDs-PILs) [78], and the obtained CDs-PILs in PEG200 performed high-performance lubricating performances. As shown in Fig. 8, based on the transmission scanning electron microscopy (TEM) and energy dispersive X-ray spectroscopy (EDS) analysis of the worn surfaces, we can conclude that the efficiency of CDs-PILs is mainly attributed to the continuous formation of tribofilms and nanolubrication of CDs. Interestingly, Zhang et al. addressed that dicationic ILs modified CDs (CD/IL) can exhibit an extreme low and stable COF of 0.045, which accounted for 38% for the neat PEG 200 under 588 N [79].

*iii) QDs/ILs analogue (deep eutectic solvent, DES)*

Very recently, deep eutectic solvents (DES), a new family of synthesis-free and biocompatible ILs

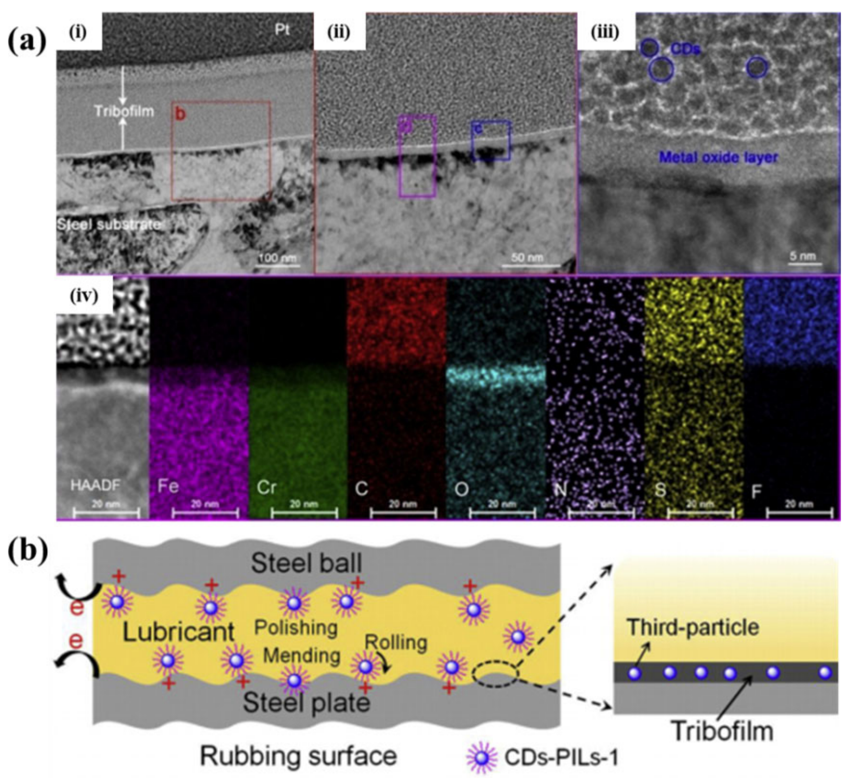



**Fig. 8** Surface analysis and possible lubrication mechanism of CDs-PILs/PEG200 suspension: (a) cross-section TEM images (i–iii), and EDS elemental mappings (iv) of worn surface lubricated by CDs-PILs/PEG200 suspension, and (b) schematic diagram of the corresponding lubrication mechanism. Reproduced with permission from Ref. [78], © Elsevier Ltd. 2019.

analogues, have drawn extensively research interest. For instance, Ma et al. proposed an initial assessment of lubricating properties of DES modified CQDs (DES/CQDs) [80]. The DES/CQDs with sphere-in-shell structure exhibited superlubricity with an extremely low COF of 0.006 for $Al_2O_3$ contacts owing to the advantages of synergistic effects of interlayer shearing, sliding, and rolling effects.

### 4.3 Intelligent lubrication

With the rapid development of industrial technology, newly developed tribo-systems are needed to meet the continuously rising stringent requirements for mechanical systems [81, 82]. In pursuit of high lubricating performances, core–shell nanoparticles have been employed which possess excellent lubricity, superior mechanical properties and flexibility owing to the advantage of synergy [83]. In 2019, He et al. reported core–shell structured PEG modified CDs (PEG/CDs) through a facile and productive ultrasonic treatment [84]. The PEG/CDs exhibited stable and super-low COF of 0.02 assigning to the surface passivation of PEG and rolling effects of CDs.

For some specific industrial applications, it is essential to develop intelligent lubrication systems with real-time self-repair, self-storage, and self-diagnosis [85]. For instance, a real-time and self-powered system was developed to monitor the lubricating oil condition aiming to online evaluate the reliability of working machines [86]. Moreover, developing new materials and approaches towards the biological lubrication systems is of major importance for human's well-being, including artificial joints, artificial teeth, artificial heart valves and other important parts that are easily damaged [87, 88].

Inspired by the artificial joints, microencapsulation has been implemented to achieve biological lubricity with controllable release of lubricants and long service life. Additionally, lubricants in gelation formation can prevent the oil creeping and evaporating loss, and thus are beneficial for maintenance and operation. Recently, eco-friendly CDs/PEG/chitosan (CDs/PEG/CS) gels were prepared and stored in bio-inspired grooved surfaces based on cartilage-embedded structures [89]. The CDs/PEG/CS gel solution exhibits superior lubricity for UHMWPE contacts and long service lifetime owing to the synergy of bio-inspired structure and strong interactions between CDs and PEG.

## 5 "Green nanotribology": Summary and prospects

In future, tribology will gradually play an increasingly important role in manufacturing, power output equipment, transportation, biological and other fields [2, 90, 91]. With regard to this situation, a concept of "green tribology" is proposed by Zhang towards a sustainable society in 2008 [92]. Guided by the viewpoint of green tribology, one of the most important approaches is to develop bio-/eco-lubricants with exceptional friction-reducing and wear resistance performances [93].

As we summarized in this review, the discovery of 2D QDs has become a burning research topic for green tribology, especially since the possibility of their large-scale and cost-effective manufacturing [94]. Recent studies have verified that 2D QDs-based lubricants could provide efficient lubrication functions in forms of liquid nanoadditives, solid coatings and gels, etc. The corresponding lubrication mechanisms are mainly categorized into nanolubrications (ball-bearing effects, self-mending effects, polishing effects, and formation of high-quality protective films) and synergistic effects.

Additionally, the rapid realization of superlubricity of 2D QDs, especially at macro-scale and in harsh working conditions, enables them to spark more broad application fields, including aerospace industry, and marine engineering, etc. Moving forward, there are still many exciting opportunities and challenges for further breakthroughs to discover QDs in the tribology field. As depicted in Fig. 9, we list some frontiers as follows:

i) From chemistry and physics perspectives, in-depth theoretical models performed by artificial intelligence (AI) (e.g. machine learning) [95, 96], and empirical experiments [97] are needed to delicately predict, design, and fabricate QD-based lubricants with desirable multi-functionality. Besides 2D QDs, more efforts should be concentrated to exploit other QDs with potential lubrication performances, such as ceramic QDs, metal oxide QDs, etc.

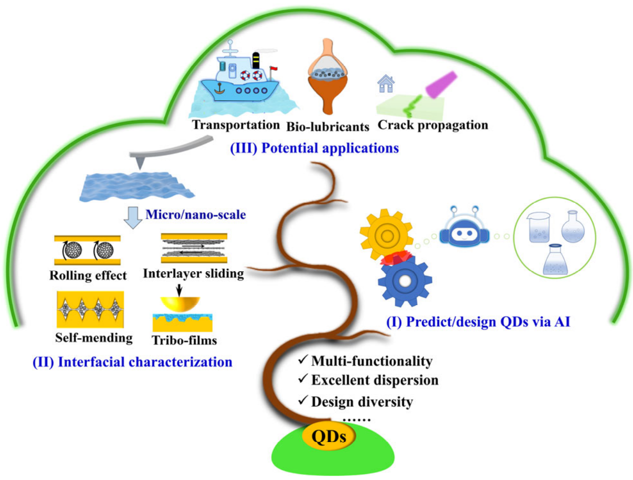



**Fig. 9** Schematic of exciting opportunities and challenges of QDs relevant lubricants towards a sustainable society.

ii) The *in-situ* characterization of interfacial phenomenon occurring at the microscale, even at the atomic and molecular scales, is needed to disclose the underlying lubricating mechanisms of nanolubricants, combined with advanced characterization technology such as atomic force microscope (AFM), surface force apparatus (SFA), scanning tunneling microscope (STM), and confocal microscopy, etc. [98].

iii) Moving forward, from an engineering perspective, the widely tunable physicochemical, optical and electrical properties of QDs will drive further exciting opportunities of tribology-oriented applications. For instance, owing to the fluorescence properties, the large area QDs coatings on metals or ceramics will intuitively real-time monitor the crack propagation for specific engineering fields.

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