Progress in experimental study of aqueous lubrication

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Aqueous lubrication is commonly regarded as friendly to the environment. We review current knowledge of the behaviors and mechanisms of aqueous lubrication. Major methods for investigating aqueous lubrication are addressed. We describe studies on both friction and film formation performance of various water-based lubricants. Additionally, we discuss possible underlying mechanisms of aqueous lubrication. Attention is also drawn to continuing topics of investigation and some unsolved problems.

lubrication, aqueous lubricant, friction, film formation

Lubrication theory and technology has been well developed in the past 10 years. Some new areas and concepts have been explored, e.g. superlubricity [1–5], nanoparticles or onion-like carbon nanoparticles in lubricant [6–9], nanolubrication under external electric voltage [10–15], bio-lubrication [16–19], lubrication with textured surfaces [20–22], and aqueous lubrication [23–25]. Aqueous solutions are drawing attention because of their ecological advantages, fire-resistance, high thermal conductivity, non-toxicity, accessibility, biodegradability, environmental friendliness, and good solvency. However, water-based solutions have several disadvantages as lubricants, such as corrosiveness, vaporization and non-viscosity [26,27]. In the past several decades, aqueous fluids have been extensively used as lubricants in metal working processes, food processing, biological systems etc. Since petroleum will be exhausted within 50 years, aqueous fluids will have wide application in many industrial fields. Thus, research on aqueous fluids has become an important branch of tribology.

Use of water-based lubrication in engineering can be traced back to about 2400 BC, when people used water as a lubricant in transporting statues in Egypt [28]. Meanwhile, water-based liquids have always been excellent lubricants in mammalian joints such as knees or hips, with friction coefficients $\mu < 0.002$ [29,30]. Till now this ultra-low friction has not been well emulated in human-made aqueous systems. In contrast to oils and organic solvents, water has many unique properties due to the polarity of its molecules, which makes aqueous lubrication much more complicated than traditional lubrication with oils.

Many studies have revealed both the characteristics and mechanisms of aqueous lubricants, with a wide variety of dimensions, conditions and application areas. In spite of the great efforts of both experimentalists and theorists, the study of aqueous lubrication still faces big challenges. As mentioned above, human-made systems seldom emulate natural lubrication well enough for practical use. Major efforts have been made to reduce the friction force of aqueous solutions between two solid surfaces and enhance the film formation ability at the same time. Nanotechnology has made much progress in revealing the underlying mechanisms and behaviors of materials at the nanometer-scale, especially the behaviors of atoms and molecules [31–33]. However, the mechanisms of aqueous lubrication are still not well known because of the diversity and complexity of aqueous solutions. This paper presents a general review of research on aqueous lubrication.

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1   Survey of investigation methods

Because of the progress of nanotechnology, several instruments have been developed to understand the atomic behaviors and mechanisms of materials, such as the atomic force microscope (AFM) [34,35], surface force apparatus (SFA) [36,37], interferometers, and so on. In tribology, the processes occurring between two materials when they are approaching or sliding past each other are very important [38–40]. This is especially true for experimental study of aqueous lubrication, where understanding the molecular mechanisms in thin-film surfaces is of considerable interest.

AFM and friction force microscopy (FFM) are used to investigate the surface roughness, adhesion, friction and normal forces and wear of surfaces, with high sensitivity by using a cantilever with a sharp probe on the order of nanometers [34,35].

Since the late 1960s, the SFA has been used to detect the interactions between mica (or coated) surfaces. The SFA has a major role in the study of aqueous liquid behaviors. Both film thickness and force (normal and frictional) can be precisely determined between two surfaces across various confined liquid media [41,42]. The unique sensitivity and resolution of the SFA has been achieved by taking advantage of both molecularly smooth mica surfaces and chromatic optical interferometry (which enables direct measurements of absolute separations with an accuracy of ± 0.1–0.3 nm) [43]. Compared to the AFM, which has very small contact areas and can measure only relative displacements, the SFA has been more extensively used in studies of aqueous lubrication.

An interferometer based on calculated optical interference fringes was developed for measuring the elastohydrodynamic lubricant film thickness between a coated glass disc and a highly polished steel ball/roller [44,45]. As the traditional optical interference technique has been limited by weak resolution (visible thickness must be bigger than 1/4 wavelength) [45], great effort has been made to develop techniques with higher resolution and precision to study film formation mechanisms and characteristics at the nanometer scale during lubrication processes [46,47]. These include the space layer imaging technique [48–52], relative optical interference intensity (ROII) approach [53–57] and so on. The existing interference techniques can detect lubricant film thinner than 1 nm between two surfaces. Compared to both the SFA and AFM, which focus on static or quasistatic states and smooth atomic contacting surfaces, an interferometer can study lubrication in dynamic states, and can well simulate practical conditions.

Various other techniques have been used to study aqueous lubrication, such as different kinds of tribometers (which are used to study the wear and friction performance of solid surfaces across water-based solutions), quartz crystal microbalance (which can be used to detect the adsorption behaviors of solutions on solid surfaces [58]) and so on.

2   Investigations of aqueous lubrication

Because of the unique properties of water compared to oil and organic materials, studies on aqueous lubrication reveal big differences in various fields from bio-lubrication to colloid science. Some traditional lubrication theories are no longer applicable to aqueous lubrication because of the complex physical and chemical properties of water. Numerous topics and issues are involved. Most of them pertain to the following basic aspects.

2.1   Lubrication behaviors of aqueous liquids in confinement

Nowadays, most studies on lubrication aim to evaluate the superlubricity between two rubbing surfaces and to reveal the underlying mechanisms. Superlubricity reduces the conservation of friction energy and helps develop modern highly technical equipment and nano-techniques [59]. A kind of superlubricity results from the extremely high repulsive force between two surfaces, as shown in the studies below.

Liquid confined between surfaces within a nanogap is crucial for understanding lubrication, friction and adhesion behaviors, especially with water molecules [38]. With the development of nanotechnology, rubbing surfaces are extended to atomic roughness, providing an excellent opportunity to study the molecular mechanism of aqueous lubrication. Thus, water films at interfaces have been studied via experiments and computer simulation techniques, e.g. AFM [36], SFA [38,43], X-ray diffraction [60] and optical interferometry [23,61]. The term "effective viscosity" of water, which is significant to lubrication behavior, has a disputed history. Peschel et al. [62] first reported an anomalously high viscosity in 1970. Zhu and Granick [63] also reported an effective viscosity varying by orders of magnitude for water confined between mica crystals with a thickness of 1–2 water molecules. On the other hand, Horn et al. [64] found that water retains a fluidity characteristic of the bulk liquid, although the structure was modified. This was supported by the SFA results between bare mica surfaces from Klein et al. [65]. Klein et al. [65] studied the fluidity of highly purified deionized water confined in a gap less than 3.5 nm. The study suggested that the effective viscosity of water remains at the same order of its bulk even when confined in the range of 3.5±1 to 0.0±0.4 nm, which markedly differs from non-associative liquids [66]. As shown in Figure 1, confinement for water molecules seems primarily to suppress the formation of the highly directional hydrogen-bonded networks associated with freezing [65]. In the case of water with low ion concentration ($\sim 10^{-3}$ mol/L), the effective viscosity of a water film confined between two bare mica surfaces reportedly ranged from 10^3 to 10^7 Pa s within 0.2±0.2 nm confinement [63]. On the other hand, Raviv and Klein [66] showed that the effective viscosity of

confined water with low ion concentration is around 0.1 Pa s even when compressed down to 1.0 nm thick films. When the ion concentration was increased to 10^{-2} -10^{-1}mol/L, the water molecules bounded tenaciously to the surfaces as a result of the hydration ions [66] as shown in Figure 1(b). The rapid exchange of molecules ensures the hydration layer bounded to the surface remains very fluid under shearing [66]. As a result, the hydrated layers perform as highly efficient lubricants supporting a large normal load. Donose et al. [67] reported that the size of the counter ion in the water significantly influences the lubrication performance of surface-bounded hydration layers. They found that the bounded layers of more and smaller hydrated cations have a higher lubrication capacity than the layers of larger and less hydrated cations in electrolyte solutions. This attributes to the lateral mobility of the water molecule in the hydration shell of adsorbed cations [67], as shown in Figure 2.

Superlubricity can be also caused by the formation of an effective lubricant film between the two surfaces. A coefficient of 0.0028 has been achieved by Ma et al. [4] using a mixed aqueous solution of glycerol and boric acid under a high contact pressure. Superlubricity is reported for the strongly adsorbed diglycerin borate layer on the sliding surface which holds the water molecules acting as good lubricant for the contact. Luo et al. [5] observed the superlubricity of phosphoric acid (H_3PO_4) under ambient conditions, which is due to the water-containing nanofilm with a hydrogen-bonded network structure induced by the pressure and shear effect.
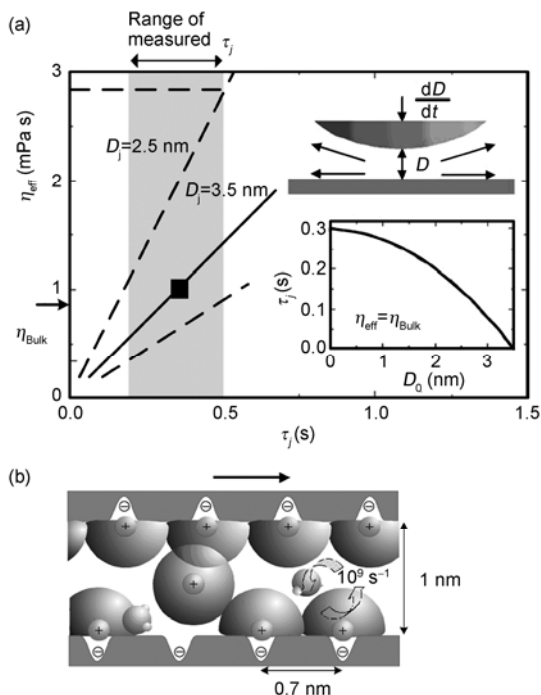

Figure 1   (a) Evaluation of the effective viscosity of confined water [43], Copyright 2011, Nature Publishing Group; (b) schematic of the interactions across high-concentration aqueous salt solutions [65], Copyright 2004, IOP Publishing.
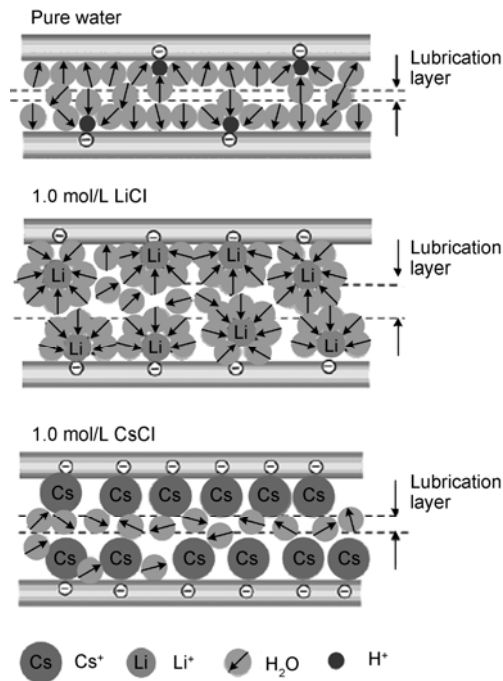

Figure 2   Schematic of hypothetical frictional mechanisms of different cation size and quantity [67], Copyright 2005, American Chemical Society.

Film thickness of a lubricant confined by two surfaces is considered an important factor for evaluating the lubricating effectiveness. The SFA studies above focused mainly on static or qusistatic states. In practical cases, the formation of elastohydrodynamic lubrication (EHL) films between two surfaces is crucial to the lubrication efficiency. A technique combining optical interference with the disc-ball/roller rolling set-up has been developed for investigating lubricant films confined between two solid surfaces under heavy load and high speed conditions. A big distance between two contacted surfaces will efficiently reduce the friction and wear [26]. Numerous results have been reported on aqueous lubricants at the nanometer scale using this approach under both static and dynamic contacts [68,69]. Till now, it has been commonly acknowledged that the film formation ability of pure water is quite poor because of its low viscosity. Rubbing surfaces under a practical load cannot be effectively lubricated by pure water, because the water film under such conditions is quite difficult to detect experimentally [69,70]. This is also confirmed by elastic hydrodynamic lubrication theory, as shown in Figure 3 (when the rolling speed is even bigger than 5 m/s, the water film is still less than 10 nm) [71]. Since water itself does not efficiently separate two surfaces sliding with respect to each other, various additives have been used to improve the film formation ability of water, such as surfactants, polymers, oil and so on. Both the lubrication mechanisms and behaviors are different for different types of aqueous liquids, which makes it quite difficult to investigate aqueous lubrication.

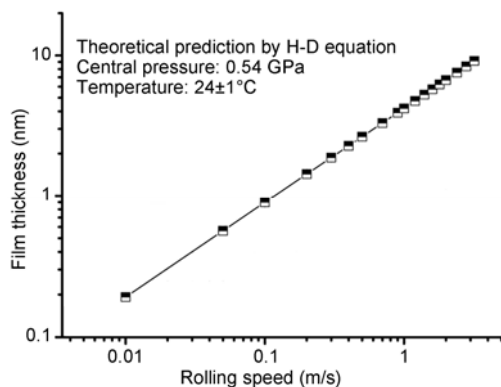A surfactant has a unique amphiphilic molecular structure

Figure 3   Film formation of pure water under point contact predicted by Hamrock-Dowson (H-D) EHL theory [71].

with both hydrophobic tails and hydrophilic heads. By migrating to the water surface, the alignment and aggregation of surfactant molecules will alter the surface properties, by which the surfactant influences the performance of aqueous lubrication. In the past several decades, the lubrication behaviors of surfactant solutions in confinement have been studied by many researchers. Ratoi and Spikes [26] studied the film-forming characteristics of several types of surfactant solutions using an interferometer. The results showed that film formation is dependent upon both the type and concentration of the surfactant and also on the pH value of the surfactant solutions. The film formations shown in Figure 4 show that the boundary film formation is due to the adsorption on the solid surfaces of mono- or bi-layers of surfactant at low speeds, and the film is enhanced by the hydrodynamic entrainment of water solutions at high speeds [26]. Boschkova et al. [72] studied the cation, amphiphile and gemini surfactants systematically by using the Quartz Crystal Microbalance (QCM), AFM and SFA. The results indicated that the surfactants will adsorb at surfaces forming either monolayers or multi-(liquid crystalline) layers. Packing of surfactant molecules at the surfaces resulted in good film-forming capability and load-carrying capability, which will be changed by the length of the spacer group [72,58]. Figure 5 shows the liquid crystalline mesophase structure presented by Boschkova et al. [73]. Liu [74] investigated
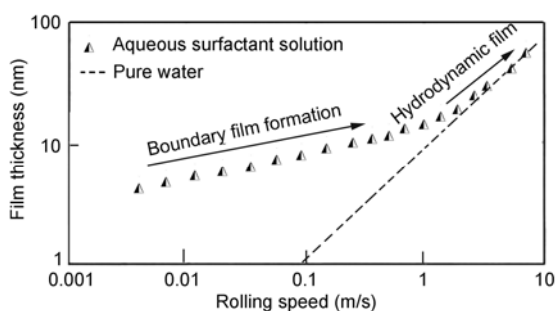

Figure 4   Film-forming properties of different types of aqueous surfactant solutions.
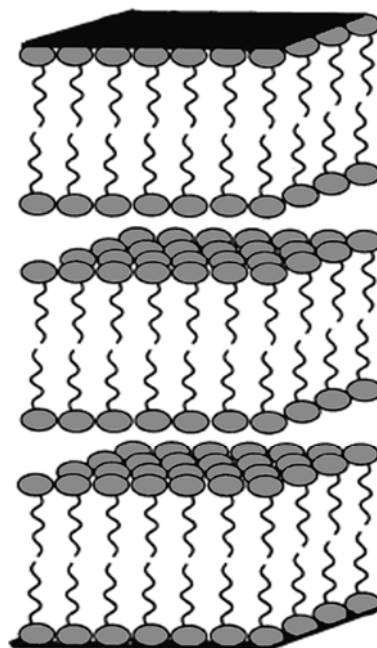

Figure 5   Schematic of surfactant molecules arranged in bilayers of a lamellar liquid crystalline mesophase between two surfaces [73], Copyright 2000, Elsevier.

the film formation properties of ionic surfactants (taking sodium dodecyl sulfate as an example) under the point contact condition by using a relative optical interference intensity (ROII) interferometer [56]. They reported that the film formation of an aqueous solution is obviously improved by the adsorption of the surfactants on the solid surfaces [74].

Polymers are good at separating two rubbing surfaces by forming a protective layer on the surfaces and improving both the load-carrying and lubrication capability of water [59]. Lee et al. [75] showed that poly(*L*-lysine)-g-poly (ethylene glycol) (PLL-g-PEG) improves the lubrication capability of water for metal-oxide-based tribo-systems. A relatively low friction coefficient (about 0.0006–0.001) even at low sliding speeds and at pressures of up to several atmospheres was reported by Raviv et al. [65] for the polyelectrolytes (PMMA-b-PSGMA) using the SFA. The results show that brushes of charged polymers attached to surfaces result in superior lubrication ability. Figure 6 shows the results of Raviv and his co-workers [65]. The good lubrication performance is attributed to the strong resistance to interpenetration of compressed brushes, combined with the fluidity of the hydration layers surrounding the charged polymer segments. Plaza et al. [76] explored the tribological properties of polyethoxyglycol esters of dithiphosphate acid derivative aqueous solutions using four-ball and ball-on-disk machines. Such solutions showed excellent load-carrying performances and strongly reduced friction coefficients in contrast to pure water. Wang et al. [77] reported a good wear resistance of carbon fiber reinforced polytetrafluoroethylene (CF/PTFE) in both pure and sea
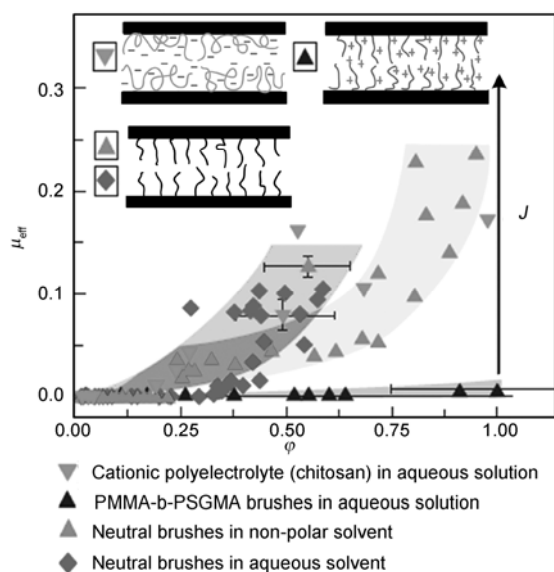
Figure 6 Friction coefficient μ_{eff} for different polymer lubricants as a function of mean surfactant layer volume fraction φ under compression [1], Copyright 2003, Nature Publishing Group.

water by investigating the tribological behaviors of some polymeric materials. An octadecyltrichlorosilane (OTS) monolayer reportedly has considerable anti-wear ability in a very humid environment [78].

An oil-in-water emulsion, a kind of water-based liquid different from aqueous solutions, has been widely used in metal working where a non-flammable lubricant is required. The lubrication performances of emulsions are significantly enhanced by their oil. The development of testing techniques has enabled many researchers to explore film-forming and lubrication behaviors. The emulsion films have been found to collapse to a relatively low thickness at a critical speed after a rise in the film thickness [69]. Kimura and Okada [79,80] studied the influence of the emulsifier type on the film formation of emulsions under a line contact. Film formation is significantly influenced by the oil concentration, emulsifier concentration and type [81]. A different emulsifier was reported to possess a different capability to remove the oil from the emulsion when contacting the solid surfaces. Although the emulsion film formation curve has been recognized, the film formation mechanism is still not well known. Consequently, several theories have been put forward [43]. Film formation at a low speed is commonly explained by the adsorption and plate-out of oil droplets on the surfaces [82]. The starvation theory was used to explain the fall of the film thickness at higher speed [83], which was initially developed for single-phase lubricants. The dynamic concentration theory was developed based on the concept that the oil droplets will concentrate before coming into the contact area and form a continuous oily pool [84], as shown in Figure 7. Re-emulsification was put forward to explain the film formation of O/W emulsions by exploring extremely low oil concentration(0.005 vol%),
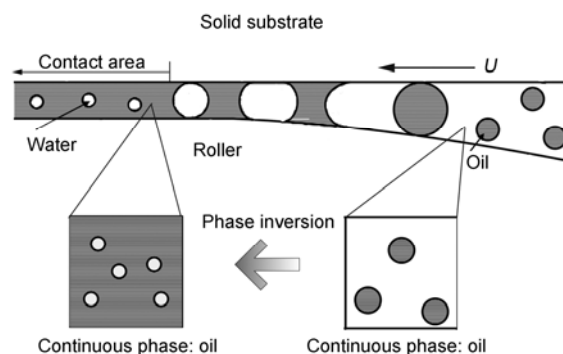

Figure 7 Sketch of the dynamic concentration of oil droplets under a rolling speed *U*.

which is concerned more with the interfacial behavior of both oil and emulsifier under dynamic conditions [24]. The oil adsorbed on the surfaces is confirmed to re-emulsify and return to the aqueous bulk as droplets under the effect of the emulsifier combined with the mechanical shearing effect [24] (see Figure 8 for sketch).

2.2 Friction behaviors of different rubbing materials across water-based lubricants

For the oxidation of water to metals, the material of the surfaces contacting with the aqueous lubricant should be selected. Numerous studies have searched for suitable materials for the rubbing solid surfaces.

High-polymer materials have been widely used in practical aqueous lubrication because of their self-lubrication properties, such as bakelite, vinyl fluoride and polytetrafluoroethylene (PTFE). These materials have been used as solid lubricants to reduce friction, wear, and energy consumption of machinery. de Vicente et al. [85,86] explored the friction and lubrication behaviors of polymer solutions and hydrocolloids between the silicone elastomer and steel surfaces. The results show that the polymer solution and microgels can form mixed efficient fluid/boundary lubricating films, causing a low friction coefficient [85,86].

Polymer solids have several disadvantages as rubbing materials. They have poor thermal stabilities and are easily aged. In contrast, ceramic materials perform better when they are used in aqueous solutions with small density and
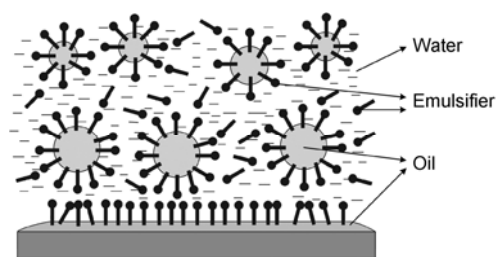

Figure 8 Illustration of the re-emulsification effect when the oil is partly removed [71].

good abrasive resistance. Silicon nitride and silicon carbide ceramics have been widely used in aqueous lubrication fields such as water bearings and space engines. Many investigations have revealed both the characteristics and mechanism of the friction of ceramic materials across water-based media. Silicon nitride was reported to efficiently reduce the friction coefficient to less than 0.002 when sliding in water after a suitable running-in condition [87–91]. The low friction coefficient obtained after the running-in process was attributed to mixed lubrication, i.e. partially hydrodynamic lubrication due to the ultra-smooth surface and partial boundary lubrication due to the colloidal silica layer caused by the reaction between silicon nitride and water. Zhang et al. [92] studied the friction and wear behaviors of a (Ca, Mg)-sialon sialon/SAE 52100 steel pair under the lubrication of water and polyol aqueous solutions. They found that the friction coefficient decreased and the wear volume loss of sialon ceramic increased for the (Ca, Mg)-sialonin/SAE 52100 steel pair [92]. Low friction forces and small wear volume loss have been obtained in water by using both alumina [93] and chromium oxide [94]. Although ceramic materials have good friction performances in aqueous environments, they are difficult to process and have high rigidity, which easily causes rupture. Film coatings (such as diamond-like carbon [95] and amorphous carbon nitride [96] films) on metal surfaces have also been found to efficiently reduce the friction coefficient of sliding in aqueous solutions.

3   Conclusions

These studies show that an aqueous lubrication system is very complex. In spite of the complexity, there are still several advantages. Both the boundary and elastohydrodynamic films prevent the sliding surfaces from contacting directly. The hydration effect of the ions in the water solution keeps the water molecules resistant against high pressure and maintains low friction. The surfactants and polymers serve as the protecting brushes on the surfaces, which partially reduce the friction coefficient. In contrast to normal lubricants such as oil and organic liquids, it is difficult to obtain a general model for water-based lubrication because of the diversity of aqueous lubricants and rubbing materials. Both the behaviors and mechanism of confined aqueous solutions and colloids are complicated and still not well known, which calls for deeper studies.

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