lubricants

Review

Hydrogels for Lubrication: Synthesis, Properties, Mechanism, and Challenges

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Abstract: Hydrogels have received extensive attention as functional lubricants because of their excellent anti-friction and anti-wear properties, tunable tribological performances, and effectiveness in alleviating lubrication failures caused by the creeping or leakage of conventional liquid lubricants owing to their semi-solid nature. This review summarizes the current research advances in hydrogel lubricants fabricated with various organic and/or inorganic gelators, including organic polymeric or supramolecular hydrogels, inorganic particles-based hydrogels, and organic polymer-inorganic particle hybrid hydrogels. We illustrate not only the design strategies for constructing high-performance hydrogel lubricants but also the tribological behavior and mechanism of different types of hydrogel lubricants and their potential applications in industrial and biomimetic fields. Corresponding outlooks and suggestions for future studies have also been proposed.

Keywords: polymeric hydrogels; inorganic hydrogels; friction and wear; lubrication; mechanism; biomimetics

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Citation: Hu, L.; Yang, Y.; Yu, W.; Xu, L. Hydrogels for Lubrication: Synthesis, Properties, Mechanism, and Challenges. *Lubricants* **2024**, *12*, 186. https://doi.org/10.3390/lubricants12060186

Received: 22 March 2024
Revised: 13 May 2024
Accepted: 23 May 2024
Published: 24 May 2024

1. Introduction

Friction is the force that impedes the relative motion between two contacting interfaces. It can cause a list of adverse mechanical and tribological impacts such as severe damage to working apparatus, strong reductions in the service life of equipment and devices, and high energy consumption in industrial manufacturing. Statistical analyses show that friction and wear account for nearly 30% of the primary energy consumption, around 80% of total machinery damages, and approximately 50% of mechanical component failures worldwide every year [1]. Therefore, looking for innovative ways to minimize the adverse impacts of friction and wear down to negligible levels is necessary. Advanced lubricants can greatly improve mechanical efficiency to save energy and prolong the service life of the machine [2,3].

Currently, different types of lubricants have been developed, including liquid lubricants [4–6], gas lubricants [7,8], solid lubricating materials [9–11], and semi-solid lubricants (greases and gel) [12–14]. Mineral oil-based lubricants as conventional liquid lubricants have been widely utilized to minimize friction and wear by separating the contacting surfaces of the equipment in the field of industry [15]. However, the poor degradability of petroleum and additives used usually to improve the capability of lubricating oil have an irreversible impact on the environment. Consequently, the negative impact of mineral lubricants cannot be ignored. Despite mineral lubricants having been gradually replaced by synthetic lubricants based on plant and animal oil, the inherent challenges (e.g., leakage and creeping) of liquid lubricants during the operation have not been solved. The solid lubricants were developed to overcome the challenges of oil-based lubricants and applied in fields where liquid lubricants are inadequate such as aviation and space industry, which are perceived as stringent conditions (e.g., large temperature difference, high velocity, high vacuum, and intense radiation). Under these harsh conditions, solid lubricants have been extensively used due to their large operating temperature range, strong carrying capacity, resistance to high vacuum and radiation, etc. Moreover, solid lubricants have stable physical and chemical properties, as well as the function of protecting the friction interface due to their strong attachment to the friction surface. However, in comparison with lubricating oil, solid lubricants generally exhibit a poorer efficiency in reducing friction and wear. Moreover, very few solid lubricants could be used in industrial applications. Therefore, exploring high-performance lubricating materials that can not only integrate the advantages of liquid and solid lubricants but also make up for the lack of the two lubricants is important.

Semi-solid lubricants are a kind of materials that are in a semi-flowing state at normal temperature and pressure and can translate on demand to a liquid state upon exposure to strong shear forces. Compared to liquid lubricants, they have a better load-carrying capacity and damping and shock absorption ability, as well as the ability to solve the problem of creeping and leakage [16]. Greases, a kind of semi-solid lubricant mainly made of base oil and thickener, are more appropriate under long-term lubrication conditions because of the properties of non-leakage and anti-creeping [17,18]. Moreover, numerous organic additives inevitably are added to grease for the purpose of exhibiting desired characteristics when operating at high temperatures or extreme-pressure conditions [19]. Unfortunately, there are many disadvantages of grease. For instance, a large quantity of mineral oils, which have been extensively used as base oil (~88% market share) to synthesize grease for industry, are discarded in contact with water and soil, leading to irreparable damage to the environment [20]. The extensive use of thickeners and organic additives not only increases the cost but also reduces the lubrication efficiency. In addition, the complex preparation process and poor cooling performance unquestionably have become the challenges for grease. Gel lubricants, another semisolid lubricating material, usually show outstanding lubrication properties and have been explored in a variety of applications, especially in intelligent lubrication fields. There are two kinds of gel lubricants, oil-based gels and water-based gels. Over the past few years, oil-based gel lubricants have evolved from merely avoiding leakage to enabling domain-specific functionalization through modular molecular design [21–23]. However, the development of oil-based lubricating materials is also limited owing to the non-renewal of mineral resources [24]. Despite many researchers being committed to environmentally friendly additives [25,26], the impact of oil-based lubricating materials on the environment is, ultimately, irreversible. As a result, green lubricants, which are nontoxic and environmentally friendly, have gained increasing attention in recent years [27].

Recently, water-based lubricating materials have captured lots of interest and are widely used in water-based friction systems, e.g., cartilaginous joints [28], oral cavity [29], and subsea equipment [30]. As early as 2001, the role of the hydration layer to reduce friction in water-based lubrication was proposed by Jacob Klein [31]. Briefly, water molecules can form a hydration layer via the mutual attraction and restriction between the charges of water molecules and biological macromolecules. The hydration layer has high spatial stability and large carrying capacity, moreover, the water molecules in the hydration layer can exchange quickly with other free water molecules. Therefore, the hydration layer can maintain rapid relaxation efficiency, which shows a better hydrodynamic effect and a smaller friction coefficient in the face of shear forces. Various water-based lubricating materials have been developed based on this mechanism over the past 20 years. Ever since Jacob Klein et al. [32,33] systematically explained hydration lubrication of polymer brushes, numerous water-based polymer lubricating materials have been studied due to their excellent performance [34–37]. Remarkably, hydrogels, a class of three-dimensional (3D) networks formed by gelators embedded in a water-rich environment, have played a crucial role as water-based lubricants in recent years. Hydrogel lubricants not only have the advantage of eliminating lubrication failure compared to lubricating liquids due to their non-flowability but also show potential applications in the field of underwater lubrication and bio-lubrication [38,39]. Additionally, more and more researchers have been paying attention to the intelligent hydrogel lubricants, which are endowed with stimuli-responsive properties to achieve controllable interfacial friction through external stimulus (e.g., pH, temperature, light, magnetic field, electric field, and concentration) [40–43]. However, these intelligent and biomimetic hydrogel lubricants typically contain organic compounds or additives that may cause a series of environmental problems due to their flammability and toxicity. In this regard, creating environmentally friendly hydrogel lubricants is a meaningful study. In order to achieve this goal, a series of inorganic hydrogel lubricants solely constituted of inorganics and water were developed [44–46]. And many nanomaterials, such as SiO_2, graphene oxide (GO), carbon nanotube, MXene, and Fe_3O_4, have been adopted to construct inorganic hydrogel lubricants successfully [45–48].

Up to now, the studies on hydrogel lubricants have laid the foundation for their further development in biological and mechanical lubrication fields. In this review, we summarize the research on various hydrogel lubricants, including polymeric hydrogels, polymeric-inorganic hydrogels, and inorganic hydrogels. We not only introduce the design methods and performances of different hydrogel lubricants but also discuss their corresponding lubrication mechanisms. Particularly, we focus on the progress and the potential applications of biomimetic hydrogel lubricants. Finally, the current challenges and future prospects of hydrogel lubricants are underlined.

2. Polymeric Hydrogel Lubricant

2.1. Design Methods and Property of Polymeric Hydrogel Lubricants

Polymeric hydrogels are formed by cross-linking polymer chains dispersed in an aqueous media through a myriad of mechanisms. Based on the raw materials and gelation mechanism, the synthetic methods for polymeric hydrogels can be categorized into physically cross-linked methods, chemically cross-linked methods, and a combination of the above methods. Polymeric hydrogels have been investigated as ideal intelligent lubricants due to their tunable tribological performance by introducing functional components, which can be applied in the field of intelligent lubrication, such as smart actuators and controllable transmissions.

2.1.1. Physically Cross-Linked Polymeric Hydrogel Lubricants

Physically cross-linked polymeric hydrogels, also known as supramolecular polymer hydrogels, are formed through non-covalent interactions such as hydrogen bonding, hydrophobic interaction, van der Waals force, electrostatic interaction, and coordination interactions. The interactions depend on the intrinsic properties of the polymers. The gelation of supramolecular polymer hydrogels is easy to achieve without the need for modifying the polymer chains [49–51].

Driven by the broad application prospects, many controlled friction surfaces regulated with reversible phase transition of self-lubrication hydrogel have been designed and fabricated. For example, Ha et al. [52] developed a novel composite coating composed of Y_2O_3 stabilized ZrO_2 ceramic reservoir and poly(ethylene glycol) (PEG)-modified silver nanoparticle (AgNP) hybrid hydrogel. The PEG-modified AgNPs and α-cyclodextrins (α-CDs) were trapped into the pores of a ceramic reservoir to in situ form a supramolecular hydrogel via the host–guest interaction between α-CDs and PEG chains on the surface of AgNPs through a vacuum infiltration method. The PEG-AgNPs hybrid supramolecular hydrogel showed excellent antibacterial properties, self-lubrication, and wear-resistance performance. During the frictional motion, the gel-phase hydrogel distributed onto the local contact pores would change to the sol-phase expand to the surface, and then form a lubricating layer. This research promotes the application of artificial joint lubrication. Furthermore, supramolecular polymeric hydrogels can be used as excellent lubricating materials because the friction protective film can be formed at the friction interface. Wang et al. [53] prepared a novel supramolecular hydrogel lubricant comprising 2-ethylhexylphosphoric acid mono-2-ethylhexyl ester and melamine, which could assemble into a fibrous structure through intermolecular interaction, exhibiting shear thinning and superior extreme-pressure and anti-wear properties even under harsh condition. As evidenced, such excellent lubrication performance mainly relied on an effective protection film formed between the steel-steel contact surface, that can prevent the direct contact of the tribopair, thus enhancing the friction and wear behaviors. It has a great potential to be applied in devices in underwater or harsh conditions.

Taking advantage of the dynamic properties of non-covalent interaction, the physical cross-linked hydrogels can achieve reversible phase transformation and then effectively alleviate friction and wear. However, these properties also reduce the mechanical strength of the hydrogels. The inherent contradiction between mechanical strength and dynamic characteristics poses a great challenge to the practical application of physically cross-linked hydrogels.

2.1.2. Chemically Cross-Linked Polymeric Hydrogel Lubricants

The chemically cross-linked polymer hydrogels are generally formed via a complex chain reaction of polymer connected by crosslinker and triggered by initiators. Different from physical cross-linked hydrogels, chemical cross-linking allows more controllable and precise management of the cross-linking procedure and is better at improving the stability and mechanical strength, potentially in the application of lubrication.

The hydrogel lubrication efficiency strongly depends on their physicochemical properties, such as cross-linking density, elasticity, and hydrophilicity [54–56]. Shoaib et al. [57] studied the frictional characteristics of hydrogels with acrylamide (AM) and N,N′-methylene bis(acrylamide) (MBAM) as monomer and chemical crosslinking agents, respectively. The crosslinking density is enhanced with an increase in polymer concentration. According to the results, hydrogels with lower cross-linked polymer chains can result in better lubrication because longer relaxation times are needed for polymer chains to re-adsorb on the counter-surface. Poly(N-isopropylacrylamide) (PNIPAM) has been widely employed to synthesize polymeric hydrogel lubricant due to the chains conformation transition at above or below the low critical solution temperature (LCST) about 32 °C [58], and thus switchable interfacial lubrication induced by swelling/shrinkage phase transition can be realized. For example, Chang et al. [59] prepared PNIPAM homopolymer hydrogels by free radical polymerization of NIPAM monomers with MBAM as the cross-linker, ammonium persulfate (AP) as the redox initiator, and N,N,N,N-tetramethyl ethylenediamine (TEMED) as a co-initiator. The hydrogels transferred from a swollen state to a hydrophobically collapsed state when the solvent changed from water to water/ methanol. In the collapsed state, the surface roughness and elastic modulus of the hydrogels raised leading to a higher coefficient of friction (CoF) than that of the swollen gel, and the changes of the CoF are reversible induced by the phase change triggered by the stimuli. Xu et al. [60] reported a PNIPAM microgel as a lubricant additive in bovine calf serum, which exhibited obvious temperature-sensitive tribological performances due to the phase transition property of PNIPAM. At LCST temperature, the microgel was in a swollen state and presented the lowest friction coefficient. While the temperature exceeded the LCST, the microgel dehydrated to shrink and the corresponding CoF gradually increased with the increase in temperatures.

2.1.3. Dual-Network (DN) Polymeric Hydrogel Lubricants

The structure of hydrogel lubricants has been thought to be a crucial influence on their tribological properties. Regulating the microstructure of hydrogel networks has been demonstrated to be one of the efficient routes to improve their mechanical and tribological properties [61,62]. Polymer hydrogels with both physical and chemical crosslinks, named dual-network polymeric hydrogels, incorporate the advantageous features of rigid and tightly cross-linked by covalent bonds as well as soft and loosely cross-linked through supramolecular interactions, making them promising material for lubrication. Ever since DN hydrogel was invented by Gong et al. [63], it has proved to be mechanically strong and highly wear-resistant, DN hydrogel has so far become one of the most extensively studied lubricants.

In recent years, researchers have been devoted to exploring dual- or even multi-crosslink polymer hydrogel lubricants. For instance, Zhang et al. [64] reported a shear-responsive lubricating artificial hydrogel prepared by combining non-covalent *N*-Fluorenyl-methoxycarbonyl-L-tryptophan (FT) supramolecular network with a covalently cross-linked polyacrylamide/polyvinyl alcohol (PAM/PVA) network. On one hand, once been sheared, the FT supramolecular network disassembled to transform into a sol-state and then a lubricating layer occurred on the hydrogel surface (Figure 1a), on the other hand, the stiff PAM/PVA network supplied physical support, that the both contribute to the shear force induced lubrication. With the constant shear applied on the surface, the CoF of the hydrogel decreased due to the lubricating layer triggered by shear force (Figure 1b). The mechanical strength enhanced with the increased shear cycles due to the decreased quantities of FT in the hydrogel also evidenced the presence of disassembly of the FT hydrogel (Figure 1c). This study is expected to bring about the possibility of developing force-induced lubricating materials, like cartilage substitutions. Different from shear-responsive hydrogel lubricant, Kang et al. [65] recently developed a mucosa-inspired electro-responsive hydrogel via the synergy between an electro-responsive silk fibroin supramolecular network and a covalent PAM/PVA polymer interpenetrating network. In this study, near the cathode, the gel-state silk fibroin hydrogel disassembled into a sol-state owing to electrostatic repulsion and the CoF decreased from ~0.115 to ~0.062. This strategy provided a promising idea for designing electric-field- or magnetic-field- or even other-field-controlled hydrogels. In fact, maintaining excellent lubrication and long-life wear resistance while adjusting and controlling frictional properties through the phase transition of the hydrogel is more challenging. Rong et al. [66] reported a bilayer hydrogel with an upper polyelectrolyte brushes and a lower Poly(acrylamide–acrylic acid–2-(2-bromoisobutyryloxy) ethyl methacrylate) (Poly(AM–AA–BrMA)) hydrogel substrate, named as high strength hydrogel grafted with polymer brushes (HHy-g-PBs) (Figure 2a). The lubricity of HHy-g-PBs could remain at a high and stable level under high contact pressure (Figure 2b,c) because the loose ionic polymer brush layer had a higher intensity of hydration, which played a role in lubricating and reducing wear, meanwhile, the tough hydrogel layer delivered the load-bearing capacity.

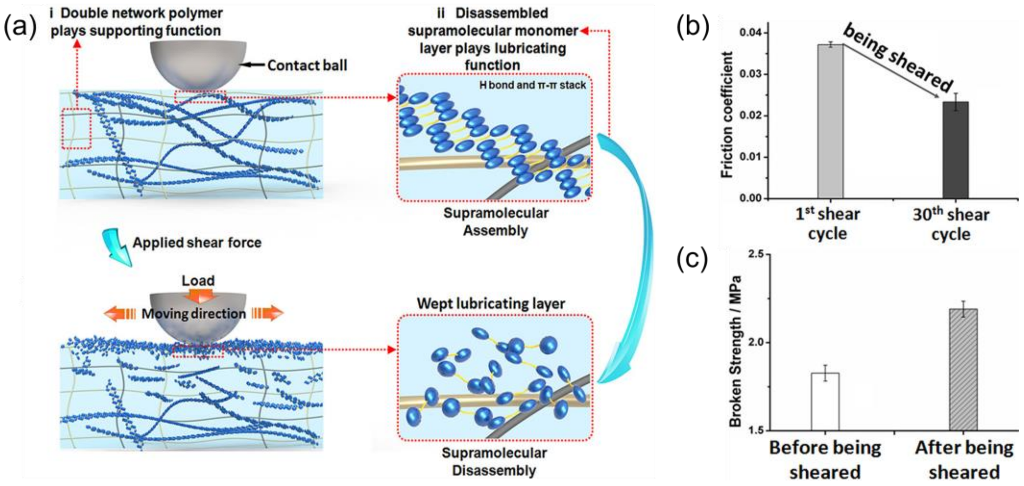

Figure 1. Before and after being sheared: the schematic diagram of internal structure (**a**), friction coefficients (**b**), and broken strength (**c**) of the hydrogel [64].

Hydrogel lubricants that are responsive to external stimuli have great potential for many applications such as soft robotics, intelligent devices, biosensors, and so on. For example, Wang et al. [41] designed a semi-convertible hydrogel composed of a stable PVA/PAAM covalent network, a α-CD/PEG supramolecular network and photo-responsive 1-[p-(phenylazo)benzyl]pyridinium bromide (AzoPB) used as competitive counterpart to tune the assembled/disassembled of α-CD/PEG supramolecular network under UV/Vis irradiation (Figure 3a). After Vis irradiation, α-CD/PEG hydrogel dissembled accompanied by gel-sol transition due to the competitive host-guest interaction of α-CD/*trans*-AzoPB and a lubricating sol layer appeared on the surface of the hydrogel, bringing on the improved lubricity. While the sol-state layer disappeared under UV irradiation causing the increase in CoF. The reversible responsive lubricating behaviors of the composite hydrogel were achieved by the adjustment of UV and Vis irradiation (Figure 3b).

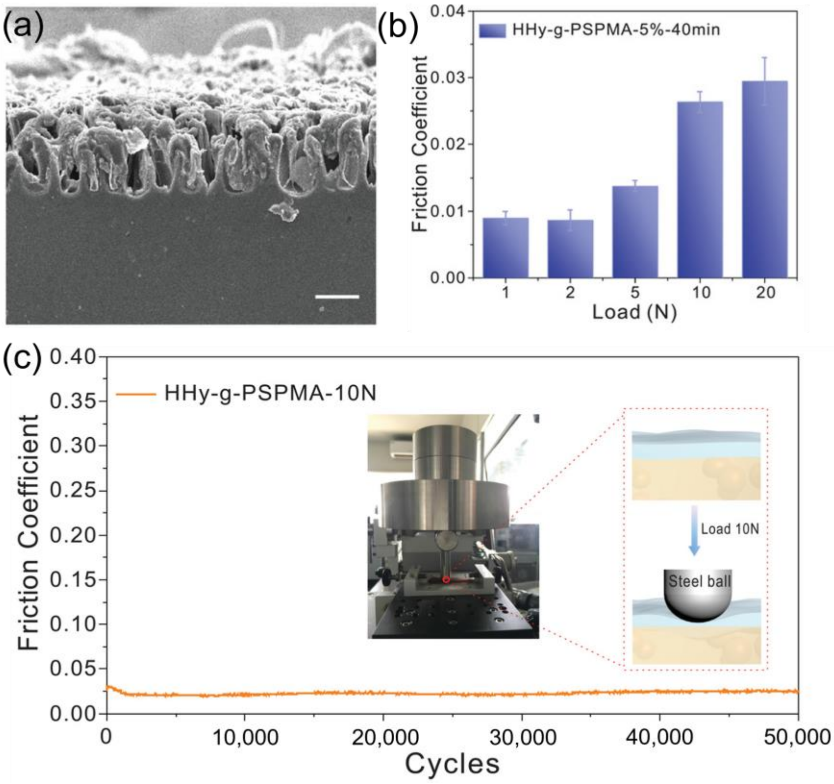

Figure 2. The cross-sectional morphologies (**a**), corresponding CoF under different loads (**b**), and long-life lubrication (**c**) of layered HHy-g-poly(3-sulfopropyl methacrylate potassium) (PSPMA) sample. Scale bar: 50 µm [66].

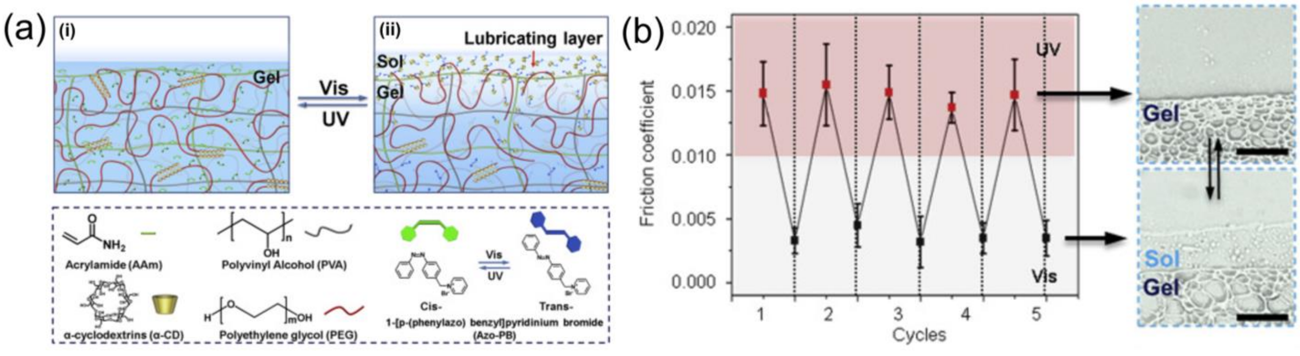

Figure 3. (**a**) The schematic diagram of the internal structure of the photoresponsive semiconvertible hydrogel under (i) UV and (ii) Vis irradiation. (**b**) Reversibility of photoresponsive lubrication under UV and Vis irradiation. Direct observations of the reversible sol layer after UV and Vis irradiation are shown in the photos on the right. Scale bar: 100 mm [41].

Overall, the structure of the DN design provides an efficient strategy to fabricate hydrogel lubricants with high strength and controllable friction properties, enabling their potential application in interfacial sensing, controlled actuation, and underwater lubrication devices. However, the complex and cumbersome two-step synthesis route of DN hydrogel lubricants limits their practical application, and establishing a simple and effective method to prepare DN hydrogel is still a great challenge.

2.2. Lubrication Mechanism and Bionic Design of Polymeric Hydrogel Lubricants

Owing to the water-rich soft structures hydrogels are similar to those of biological tribo-systems, tremendous research efforts have been conducted to illustrate the lubrication mechanism of hydrogel lubricants and explore their possible applications as replacement materials for biological tissue.

Numerous mechanisms have been proposed to explain the lubrication provided by hydrogels, such as hydrodynamic, viscous-adhesive model, and hydration lubrication [67]. Among them, hydration lubrication provided by the strong hydrated layer surrounding the polar group, as mentioned in the introduction, has been considered one of the most persuasive lubrication mechanisms for polymer hydrogels. Through the rational design and control of the hydration layer, different approaches have been taken to prepare polymer hydrogel lubricants with stimuli-responsive properties, a biomimetic design, and biomedical performance, etc. For example, PAM hydrogel particle/high-density polyethylene (HDPE) composites were designed to apply hydration lubrication in heavy-load engineering fields [68]. A hydration lubrication layer between the tribopairs could be formed effectively in the sliding process and improve the friction properties of hydrogel composites under water lubrication (Figure 4a). Thus, the obtained composites had the advantages of the excellent hydration lubrication of PAM hydrogel and the high mechanical properties of HDPE.

As mentioned above, owing to the resemblance between the hydrogel lubricants and biological tissues, hydrogels are identified as promising substitutes for artificial organs. Recent research on hydrogel components and structure according to bionics has made it possible to obtain hydrogels with excellent lubricating and mechanical properties through a rational design, which makes the application of hydrogel lubricants in regenerative medicine more realistic. Professor Nicholas D Spencer of ETH Zurich, a well-known expert in biomimetic tribology research, also pointed out that biomimetic polymer-based lubricants played a key role in water-based lubrication. Biological lubricants, such as hyaluronic acid, glycoprotein, proteoglycan, and other major components, are biomacromolecules with a "bottle-brush" structure in joint synovial fluid [69]. Glycoprotein and proteoglycan can be assembled into the molecular chain of hyaluronic acid via hydrophobic interaction and form a multi-level brush assembly with hyaluronic acid as the main chain and glycoprotein or proteoglycan as the side chain. These brush-like polymers can be dispersed in joint synovial fluid to play a part in excellent water lubrication. Inspired by the bottle brush-like bio-macromolecular lubricants, the research on the preparation and lubrication properties of structure-biomimetic polymer hydrogel lubricants has attracted extensive attention. For instance, a hydrogel lubricant with low concentrations of lipids (PC) was designed by Lin et al. [70]. These hydrogels could form self-lubricating layers by constantly oozing lipids to the friction interface and continuously self-lubricate during the friction process via the hydration layer attributed to PC-exposing boundary layers, which made friction and wear reduce by 80–99.3% compared to lipid-free hydrogels and achieved imitation or even improvement for the lubrication mechanism of human articular cartilage (Figure 4b). This research provided a strategy for manufacturing self-lubricating hydrogels that require low friction and wear. Moreover, inspired by articular cartilage, hydrogel lubricant with a bilayer structure can exhibit not only effective aqueous lubrication but also high load-bearing capacity [66]. Mu et al. [71] fabricated a polymer-filled macroporous hydrogel by freezing, thawing, and soaking the macroporous hydrogel in a polymer solution to mimic the hydration lubrication mechanism of the synovial joint. The resulting polymeric hydrogel can maintain low friction after long-term sliding as a result of a liquid-like hydration layer stabilized by the mobile chains (Figure 4c). Zhang et al. [72] synthesized gelatin-based hydrogels by photopolymerizing methacrylic anhydride grafted gelatin (GelMA), AM, and 2-methacryloyloxyethyl phosphorylcholine (MPC). The composite hydrogels exhibited remarkably enhanced lubricity due to the tenacious hydration shell formed around the zwitterionic phosphocholine headgroups (Figure 4d). In addition, the multi-functional biomimetic hydrogels have also attracted a lot of attention. For instance, inspired by the properties of self-adhesive mussel and lubricating cartilage, Han et al. [73] fabricated injectable hydrogel lubricating microspheres with a combination of biomimetic lubrication coating and drug delivery vehicle via dip-coating a self-adhesive polymer named dopamine methacrylamide (DMA)-MPC on GelMA hydrogel microspheres. The lubrication performance was improved due to the hydration lubrication of the zwitterionic phosphocholine in the copolymer and the rolling properties of the microspheres. The hydrogel microspheres not only greatly improved the lubricity but also achieved sustained local drug release, owning enormous potential for the treatment of osteoarthritis.

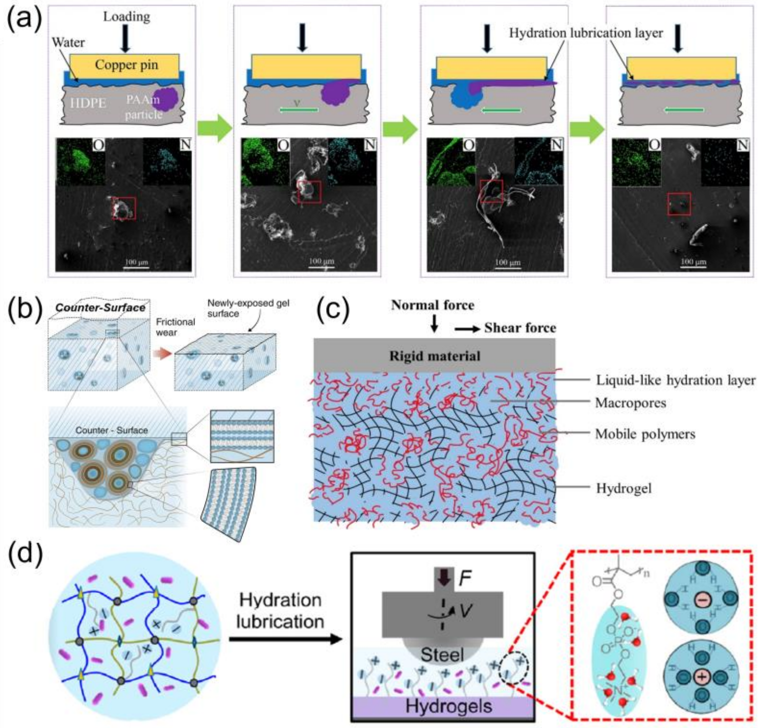

Figure 4. (**a**) The formation of hydration lubrication layer between the friction pairs in the rubbing process [68]. (**b**) The schematic representing the self-lubrication of lipid-incorporating hydrogels [70]. (**c**) Liquid-like hydration layer of polymer-filled macroporous hydrogels [71]. (**d**) Hydration lubrication mechanism of gelatin-based composite hydrogels [72].

Nevertheless, despite the promising potential in the field of biomimetic and intelligent materials, these polymeric hydrogel lubricants usually exhibit poor mechanical properties, which are prone to deformation under high interfacial contact stress, limiting their application. And the inherent toxicity of organic polymers also limited the development of polymeric hydrogels. Therefore, it is a critical challenge to study hydrogel lubricants with high mechanical properties, excellent lubrication, and hypotoxicity.

3. Polymeric-Inorganic Composite Hydrogel Lubricant

The polymeric-inorganic composite hydrogels, combing polymer network and inorganic nanomaterials, have been considered the strongest possible candidate for lubrication. The inorganic nanomaterials can not only help to improve the mechanical properties of polymer hydrogel through interacting with polymer chains, overcoming the poor capacity to resist external forces due to high water content but also enhance the tribological properties crediting to their rolling effect, load-carrying capacity, repairing ability [74–77]. Recently, many 0D quantum dots (e.g., carbon dots (CDs)), 1D nanowires, such as carbon nanotube (CNT) and fiber materials, 2D nanosheets (e.g., GO, MoS_2, and MXene), and 3D

NPs (e.g., TiO_2, SiO_2, and Fe_3O_4) have been employed to construct polymeric-inorganic composite hydrogel lubricants successfully [78–84].

3.1. Synthesis Methods and Property of Polymeric-Inorganic Hydrogel Lubricants

Generally, the use of inorganic nanomaterials as cross-linkers could reduce the restrictions exerted by dense cross-links existing in pure polymer networks. Taking advantage of their weak interlayer interaction, tunable surface properties, highly specific surface, and strong mechanics, 2D nanosheets could slide and form tribofilms between the friction pairs. Currently, GO has been widely used as a multifunctional crosslinker or reinforcement agent in composite hydrogels [85]. For example, Meng et al. [86] prepared a PVA/GO-PEG nanocomposite hydrogel via a freezing/thawing method. When frozen, PVA molecules were promoted to form the first network structure comprising molecular entanglements and crystallites through the van der Waals and hydrogen bonding force. In addition, the hydrogen bonding interaction between PVA and GO nanosheets led to the formation of the second network with GO cross-linked and the number of PVA molecules grafted on the GO surface increased with the help of PEG (Figure 5a). By introducing GO, the increased crystalline regions of PVA and crosslink density brought about the enhancement of the mechanical strength and toughness of hydrogel (Figure 5b). Moreover, due to the synergistic lubrication effect of GO-PEG, the composite hydrogel presented a remarkably lower CoF and a much smoother worn surface (Figure 5c). Wang et al. [87] synthesized a GO/zwitterionic poly(sulfobetaine methacrylate) (PSBMA) hydrogel through physical and chemical cross-linking, in which the polymer chains were effectively absorbed onto the surface of GO through the electrostatic interactions (Figure 6a). The hydrogel exhibited a significant enhancement in the compressive stress (from 0.08 MPa to 0.36 Mpa) (Figure 6b) and a remarkable reduction in the CoF even reaching the superlubricity state (from 0.025 to 0.008) (Figure 6c) with the increased GO content. These GO-based nanocomposite hydrogels exhibited appropriate combinations of excellent lubrication and mechanical capacity that provide potential requirements for cartilage replacement applications. In terms of the biocompatibility of lubricants, Wang et al. [88] synthesized a multifunctional PVA/polyacrylic acid (PAA)/GO/polydopamine (PDA) hydrogel coating by combining chemical assembly and freezing-thawing method. The hydrogel coating exhibited an 82.0% wear rate reduction of the bone tissue and possessed good corrosion resistance and bioinductive activity. Moreover, MXene and carbon nitride nanosheets have also been popularly employed in polymeric hydrogel lubricants. Recently, Miao et al. [89] constructed an MXene/PVA hydrogel via a simple salting out strategy, and the network structure of the hydrogel was reinforced by virtue of the multiple cross-linking and salting out effects of the introduced Zn^{2+}. The hydrogel exhibited long-term lubrication performance and prominent load-bearing capacity owing to an in-situ lubricating water film formed during the friction process. Kumru et al. [90] successfully prepared a composite hydrogel lubricant with the method of graphitic carbon nitrides grafted polydimethylsiloxane, the addition of graphitic carbon nitride allowed the participation of charged monomers, yielding tough and lubricant hydrogels via a photoinitiated reaction.

Apart from 2D nanosheets, adopting 0D, 1D, and 3D nanomaterials as composition to fabricate composite hydrogel lubricant has also garnered numerous interests. A SiO_2 NPs combined bilayer nanocomposite hydrogel synthesized using AM, AA, and alginate via modulated free-radical polymerization [91]. The resultant hydrogel showed obvious improvement in tribological properties and wear resistance and mechanical performances due to the addition of SiO_2 NPs. The sustained release lubrication ability of hydrogels can also be achieved by incorporating inorganic NPs into polymeric hydrogels. To satisfy the application on artificial joints, Lu et al. [92] fabricated a high-strength and good slow-release lubricating CDs/PEG/chitosan/sodium glycerophosphate (CDs/PEG/CS/GP) composite hydrogel via hydrogen bond interaction. Figure 7a exhibited the intensified strength and anti-pressure capacities of hydrogels under high shear pressure and rate in the presence of CDs. The composite hydrogel could keep releasing along with low CoF for 10 days, suggesting a slow releasing process (Figure 7b). The better lubrication ability of CDs/PEG/CS/GP hydrogel originated from the hydrogen-bonded network structure of CDs, PEG, CS, and GP in the release solution (Figure 7c). Guo et al. [82] reported a thermosensitive poly(ε-caprolactone)-PEG-poly(ε-caprolactone) (PCEC) hydrogel embedded with CNT. The PCEC/CNT composite hydrogel exhibited release-lubrication behavior while maintaining its injectability, and it showed a promising prospect of providing a slow-release lubrication effect on artificial joints.

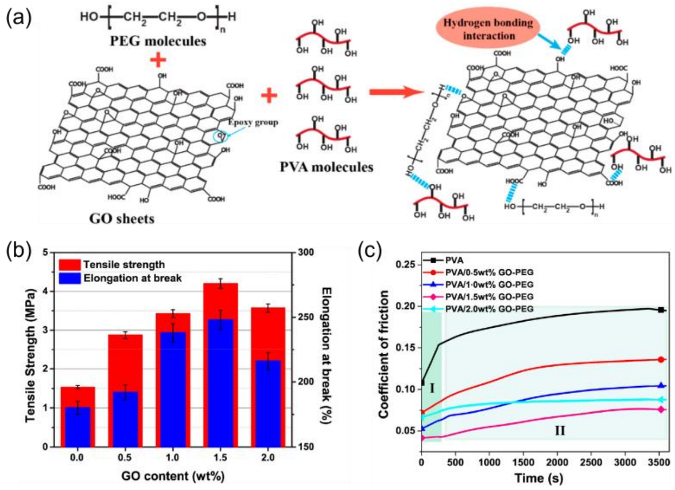

Figure 5. (**a**) Schematic diagram of the intermolecular interactions inside the PVA/GO-PEG hydrogels. (**b**) Mechanical properties and (**c**) CoF of PVA/GO-PEG hydrogels with different GO content [86].

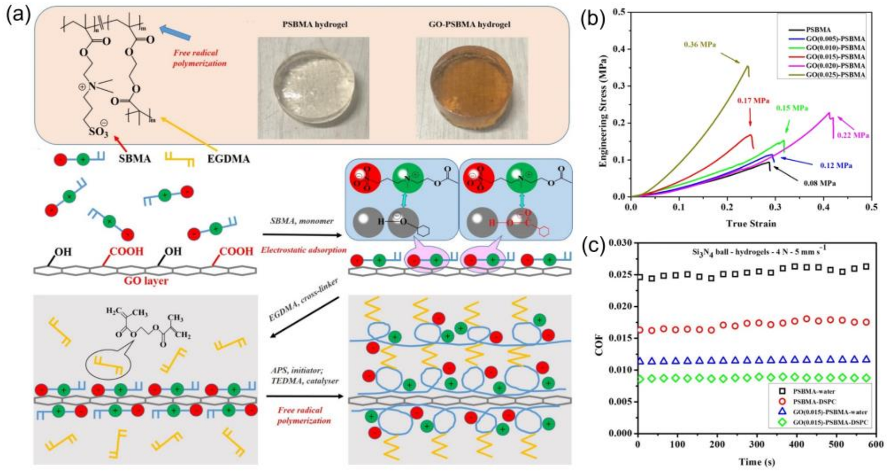

Figure 6. (**a**) Formation mechanism of the GO-PSBMA nanocomposite hydrogel. (**b**) Stress–strain and (**c**) Variations of the CoFs with time of the GO-PSBMA hydrogels with different GO concentrations [87].

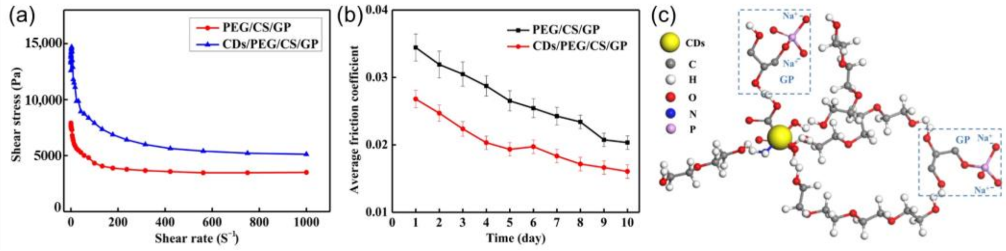

Figure 7. (**a**) The shear stress curve and (**b**) friction coefficient during sustained release of the hydrogels with and without CDs. (**c**) Schematic diagram of internal cross-linking of CDs, PEG, and GP [92].

3.2. Lubrication Mechanism and Intelligent Design of Polymeric-Inorganic Hydrogel Lubricants

The lubrication mechanisms of the polymeric-inorganic composite hydrogel can be divided into the following aspects: a hydration layer, a protective lubricating film including physically adsorbed film and transfer film and tribochemical film, sliding friction, and rolling friction, and repairing actions. Currently, the most common strategy to improve the lubricity of polymeric-inorganic composite hydrogel is the utilization of the synergistic effect of methods. For instance, Fu et al. [93] reported a GO-loaded supramolecular composite hydrogel with surprising self-lubrication performance, anti-wear, and antibacterial properties synthesized via an in situ host–guest interaction-induced self-assembly strategy. Due to the gel-sol transition behavior, the GO hybrid hydrogel could be converted into a liquid phase with the action of shear force and friction heat, forming a lubricating film to reduce friction and wear. Moreover, the pseudo-polyrotaxane superstructures in the film can also adsorb the water molecules through hydrogen bond interaction to form a hydration layer, which provides reduced shear resistance during the rubbing process (Figure 8a). In addition, ultrathin lamellar GO in the lubricating film was easily attached to the contact surface to further improve the friction-reducing and wear-resistant performance based on the layer slipping mechanism. Wang et al. [88] also verified that the content of inorganic nanosheets in hydrogels could directly affect the thickness of the formed interfacial lubrication film, which in turn affected the lubrication efficiency.

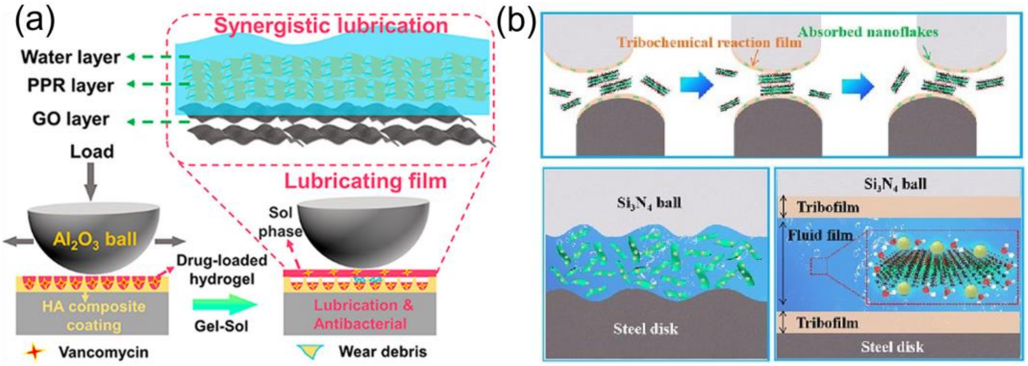

Figure 8. (**a**) Graphic illustration of the possible friction self-responsive lubrication and wear resistance mechanism of GO-loaded supramolecular composite hydrogel [93]. (**b**) Lubrication mechanism of Mxene/Fe_3O_4/PNIPAM as a water-based lubricant additive [80].

Forming a chemical protective film through a tribological reaction is also an important means to reduce friction and wear. Cui et al. [80] fabricated an MXene/Fe_3O_4/PNIPAM microgel as a water-based lubricant additive. The microgel can effectively reduce friction and wear attributed to the synergistic lubrication effect of the chemical protective film, physical adsorption film, and hydration layer acting as boundary lubrication film (Figure 8b). During the friction process, interlayer shear of lamellar Mxene nanosheets and spherical Fe_3O_4/PNIPAM played a key role in sliding friction and rolling friction mechanism, respectively, and the tribochemical reaction between Ti, Fe, and oxygen took place to form a protection film composed of metallic oxides. Moreover, a hydration layer was formed through the hydrophilic groups of the microgel and acted as a boundary lubrication film to reduce friction.

In addition to achieving ultralow friction, it is interesting to regulate friction by external triggers, such as pH [94], temperature [95], light [96], electricity [97], and magnetism [98]. Wu et al. [99] synthesized a thermosensitive GO/PNIPAM/AM composite hydrogel through the free-radical copolymerization of AM and PNIPAM. GO was added as an additive to enhance the mechanical and load-carrying capacities. The CoF of hydrogels was demonstrated to be associated with the gel state; moreover, it could reversibly switch for several cycles between ultralow friction (CoF $\approx$ 0.03) when the gel was swollen below the LCST, and it had very high friction (CoF $\approx$ 0.49) when it was shrunken above the

LCST, which had a significant impact on the design of special coatings for micromechanical devices. Wu et al. [42] then reported dual- and even multi-responsive hybrid hydrogels prepared polymer monomer (NIPAM, sodium methacrylate, or 2-(dimethylamino)ethyl methacrylate), BIS, and GO to mimic fish skin. Ultra-low and high CoF occurred when the gel was in a swelling and shrinking state adjusted by the change of temperature and/or pH.

Although most stimuli-responsive hydrogels are mainly based on responsive polymers [100] and phase-change materials [101], the participation of nanomaterials could diversify the adjustment modes of the properties of hydrogel lubricant. Fe_3O_4 NPs are favored by researchers as intelligent components to construct polymeric-inorganic hydrogel lubricants because of their low price, excellent magnetic and photothermal conversion properties, and recycling possibility. Liu et al. [102]. Synthesized a Fe_3O_4-loaded PNIPAM thermosensitive nanogel via emulsifier-free emulsion polymerization. The lubricity of the hydrogel can be multiply-regulated by magnetism, temperature, and NIR light (Figure 9a). The hydration or dehydration phase transformation of PNIPAM triggered by heating irradiation was attributed to the photothermal effect of Fe_3O_4 NPs. The presence/absence led to thermally regulated lubrication of the hydrogel. Regulation of friction under NIR light of an external magnetic field could cause aggregation/redispersing of nanogels at the friction interface, thus realizing the friction reversibly switch. The multiply regulated tribological property provided new development strategies for intelligent lubrication materials for engineering and biomedical applications. Similarly, a photothermal Fe_3O_4@poly(NIPAM-co-PAA)(Fe_3O_4@PNA) microgels lubricating coating was prepared and its surficial friction was controllably adjusted by the illumination of NIR light (Figure 9b) [103]. Without NIR light irradiation, the gel coating was hydrophilic and achieved relatively low friction due to the outstanding hydration lubrication. However, the temperature of the microgel rapidly increased because of the photothermal effect of Fe_3O_4 under the irradiation of NIR light, and the thermosensitive PNA shell was transformed to hydrophobicity, leading to the failure of hydration lubrication and a remarkable increase in friction. Liu et al. [104] reported a thermo-responsive Fe_3O_4 NPs-integrated layered hydrogel lubricants by modifying an initiator-embedded, high-strength [poly(NIPAM-co-AA-co-initiator/Fe^{3+})] hydrogel with a poly(3-sulfopropyl methacrylate potassium salt) polyelectrolyte brush. The hydrogel not only possessed considerable load-bearing capacity but also could realize in situ lubrication improvement via applying an NIR laser (Figure 9c). They [105] also created an intelligent Fe_3O_4-based hydrogel actuator inspired by the typical feeding mechanism of the esophagus system which combined a lubricating mucosa surface with the peristaltic contraction deformation of the cavity synergistically, realizing the remote operation of the cavity channel of thermosensitive gel tube device by NIR light. Mxene has also been widely used in the preparation of programmable stimuli-responsive composite hydrogels due to its good electrical conductivity and photothermal conversion properties [106,107]. Wu et al. [43] reported an Mxene / PNIPAM double network hydrogel that could achieve switchable lubrication with/without the NIR light irradiated. The prepared hydrogel showed much lower CoF (~0.021) at 25 °C because the hydration layer formed on the hydrogel surface. After the NIR light irradiation, the temperature of the hydrogel with good photothermal effect rapidly rose above LCST due to the photothermal property of Mxene, leading to an obvious increase in CoF (~0.135) because of the destruction of the hydration layer. In addition, the hydrogel showed remarkable recyclability and controllability by tuning the "on-off" of the NIR light.

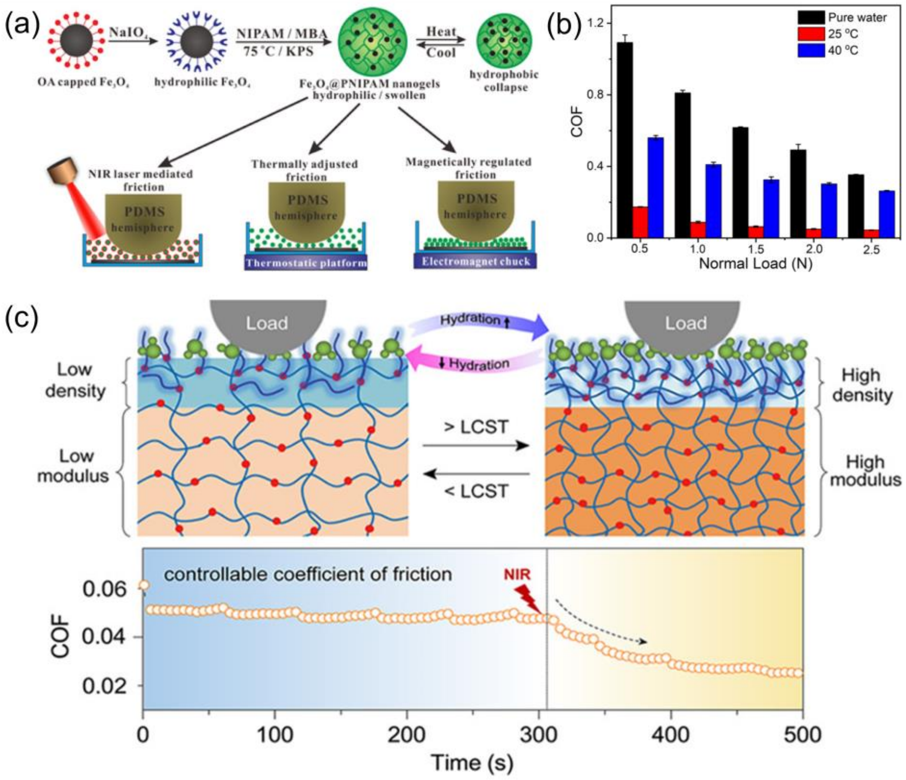

Figure 9. (**a**) Schematic diagram of the synthetic process and friction regulation with temperature, magnetic field, and NIR laser of Fe_3O_4-loaded composite hydrogel [102]. (**b**) The photothermal effect of the Fe_3O_4-based hydrogel [103]. (**c**) Schematic diagram of change in the interface hydration degree and polymer brush chain density in response to temperature. CoF change curve for an Fe_3O_4-based hydrogel by applying NIR laser irradiation [104].

4. Inorganic Hydrogel Lubricant

Composed of 3D inorganic skeletons and as high as 99% water, inorganic hydrogels serviced as versatile green lubricants have attracted much attention in the past few years. They not only exhibit excellent lubricity comparable to that of organic hydrogel lubricants but also overcome some defects such as toxicity, flammability, non-biocompatibility, and degradability of organic constituents. Recently, many inorganic nanomaterials, such as SiO_2 NPs, GO, CNT, and Mxene, have been employed as construction blocks of hydrogel lubricants successfully. The framework of inorganic hydrogels is typically formed via covalent or noncovalent bonds, such as hydrogen bonding, van der Waals force, π–π stacking. For instance, Yu et al. [47] created a SiO_2-skeleton organohydrogel containing SiO_2 NPs, water, and glycerol. As verified through experiments, the reduction in the pH of silica solution induced an attenuation of the electrostatic repulsion between particles, which accelerated the aggregation and condensation reaction of the adjacent NPs and then formed hydrogels through covalent -Si-O-Si bonds (Figure 10a). The presence of glycerol endowed the hydrogel ultralow friction, outstanding low-temperature lubrication (as low as −80 °C), anti-dehydration, and corrosive inhibition, which could find extensive potential applications in extreme conditions. The lubrication mechanism of the gel was demonstrated to be a synergistic effect between the physical protective film formed by the glycerol/water mixture that prevented direct contact among the friction pairs and the rolling and friction surface repairing actions of silica particles (Figure 10b). They also constructed high-water-content Mxene gels using brief and moderate centrifugation [48]. Based on the changes in the surface terminations of Mxene nanosheets and consequently topological reconfiguration in their internal structures triggered by acid/alkaline, the gels expressed better lubrication effect and low abrasion at relatively low pH values due to the weak interlayer associations, however, they presented relatively high friction at high pH values because of the formation of the randomly arrayed structure of Mxene nanosheets. Yang et al. [44] fabricated mechanically strong and self-healable inorganic hydrogel lubricants using aluminum hydroxide nanosheets (AHNSs) as gelators and water as dispersants. The gained hydrogel not only showed an excellent elasticity (>10 Mpa) due to the formation of a long-range ordered lamellar structure consisting of self-assembled side-to-side in repulsion-dominant gel (Gel_{Rep}) or interlaced-stacking nanosheets superstructures in attraction-dominant gel (Gel_{Att}), but also exhibited tunable lubrication by tuning the internal nanosheets' configuration and salt content (Figure 10c). The lubrication mechanism of the hydrogel was considered the synergistic effect of hydrodynamic lubrication and the physical lubricating film of AHNSs.

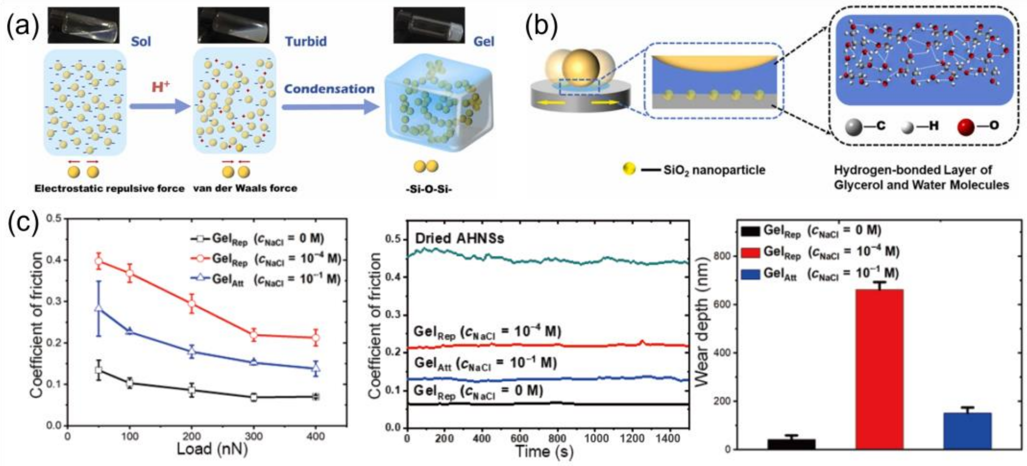

Figure 10. Schematic illustration of (**a**) the gelation process and (**b**) the ultralow friction of the silica hydrogel [47]. (**c**) The dependence of the CoFs of the AHNS hydrogel on the applied load and NaCl contents. The wear depths with different hydrogel lubricants under 400 nN applied load [44].

Although researchers have made some achievements in the preparation of biomimetic hydrogel lubricants using polymers or low-molecular-weight organics as building blocks [40,100,108–111], the adoption of rigid inorganic components remains greatly challenging due to their poor flexibility and processibility. Recently, Hu et al. reported a series of intelligent carbon-based inorganic hydrogel lubricants, which demonstrated multiple responsiveness to external stimuli like pH, temperature, and light. For example, they constructed a GO hydrogel lubricant with mechanical and tribological adaptivity based on its pH and temperature sensitivity (Figure 11a) [45]. The mechanical and tribological performances of hydrogel were capable of reversibly switching originating from the internal structure varied with the change of the pH and/or temperature (Figure 11b,c). The layer-by-layer structure of hydrogel positively influences its mechanical and tribological properties due to the weak association between the carboxyl groups favoring the fracture separations to occur in the edge-to-edge regions of GO sheets, resulting in stronger adhesion and transverse stretching, thus improving the structural stability to allow more load transfer and energy dissipation between nanosheets. In addition, the hydrogel could carry out long-term continuous lubrication and was demonstrated to be both biocompatible and biodegradable. In a follow-up study, the researcher explored the gelation behavior of carbon-based 0D CDs, 1D CNT, and 3D fullerenes particles [46]. According to the research, 0D CDs could not form hydrogels due to the super-dispersibility of quantum dots, 1D CNT could act similarly to the aforementioned 2D GO nanosheets owing to their high structural anisotropy, 3D fullerene hydrogel was not suitable as biomimetic materials because of their poor mechanically and tribologically adaptive properties although it could undergo topological reconfigurations in its internal structure upon pH varied. Therefore, in order to satisfy the need for smart surfaces and controllable transportation systems that require remote manipulation, both pH- and NIR light-responsive CNT hydrogel lubricant was fabricated (Figure 11d). The lubrication mechanism of CNT hydrogel was also considered the hydrodynamic lubrication as well as the rolling and repairing actions of CNT. These studies provided a new strategy for fabricating smart adaptive hydrogels free of organic molecules and new insights into materials science, colloidal chemistry, and nanotechnology.

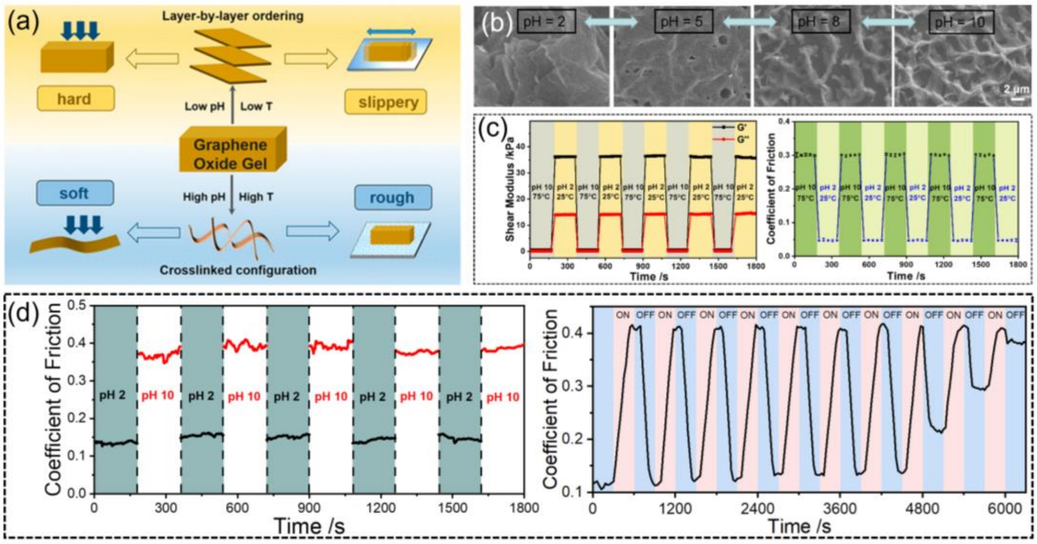

Figure 11. (**a**) Schematic diagram of the frictional and mechanical properties of GO hydrogel varying in pH and temperature [45]. (**b**) Cryogenic scanning electron microscopy (cryo-SEM) images of GO hydrogel at different pH values [45]. (**c**) Shear modulus and CoF of GO hydrogel upon synergistically switching pH and temperature [45]. (**d**) CoF upon alternately changing pH and illuminating the NIR laser [46].

To sum up, the structures and compositions determine the key properties of hydrogels like lubricity, mechanical strength, and stimuli-responsiveness. The properties of hydrogel lubricants with different structures and components have been summarized in Table 1.

Table 1. The properties of hydrogels with different structures and compositions.

Hydrogels	Compositions	Properties	Refs.
Physically cross-linked polymeric hydrogel	PEG, AgNPs, α-CD	Antibacterial, self-lubrication and wear-resistance	[52]
	2-ethylhexylphosphoric acid mono-2-ethylhexyl ester, melamine	Shear thinning, extreme-pressure, and anti-wear	[53]
Chemically cross-linked polymeric hydrogel	NIPAM, MBAM, AP, TEMED	temperature-responsive lubrication	[59]
Dual-network polymeric hydrogel	FT, PAM/PVA	Outstanding mechanical capacity, shear-responsive lubrication	[64]
	Silk fibroin, PAM/PVA	Electro-responsive tribological behavior	[65]
	Poly(AM–AA–BrMA), polymer brush	Load-bearing capacity, stable low-friction	[66]
	PVA/PAM, α-CD/PEG, AzoPB	Photo-responsive lubricating and mechanical properties	[41]

Table 1. *Cont.*

Hydrogels	Compositions	Properties	Refs.
Polymeric-inorganic hydrogel	PVA, GO/PEG	Splendid mechanical strength and toughness, good lubricity	[86]
	GO, PSBMA	High mechanical strength and excellent lubrication	[87]
	GO, PNIPAM, AM	thermal-responsive lubrication	[99]
	NIPAM, NaMA, DMAEMA, BIS, GO	pH- and thermal-responsive lubricity	[42]
	PVA, PAA, GO, PDA	Lubrication and corrosion resistance	[88]
	MXene, PVA	Long-term lubrication, load-bearing capacity	[89]
	Graphitic carbon nitrides, polydimethylsiloxane	High mechanical strength and lubricity	[90]
	SiO_2 NPs, AM, AA, alginate	Anti-friction and anti-wear, high mechanical strength	[91]
	CDs, PEG, CS, GP	Slow-release lubrication capability, good rheological and mechanical properties, biocompatibility	[92]
	PCEC, CNT	Release-lubrication effect, shear and strain resistance, thermo-sensitivity	[82]
	Fe_3O_4 NPs, PNIPAM	Photothermal-responsive lubrication	[102]
	Fe_3O_4 NPs, PNA	Photothermal-responsive lubrication	[103]
	Fe_3O_4 NPs, polyelectrolyte brush, poly(NIPAM-co-AA-co-initiator/Fe^{3+})	Load-bearing capacity, in situ lubrication-improvement	[104]
	MXene, PNIPAM	Photothermal-responsive lubrication	[43]
Inorganic hydrogel	SiO_2, glycerol	Low-temperature lubrication, anti-dehydration, and corrosive inhibition	[47]
	MXene	Tunable rheological and tribological performance	[48]
	AHNSs	Mechanical robustness, self-healing, controlled lubrication and anti-corrosion	[44]
	GO	pH-responsive friction and mechanical properties, long-term lubrication, biocompatibility and biodegradability	[45]
	CNT	pH- and light-responsive friction and mechanical properties	[46]

5. Conclusions and Outlooks

In the past few decades, semi-solid hydrogels, which are usually prepared by encapsulating water into a cross-linked polymeric or inorganic network, have drawn particular attention as functioned lubricants that benefit from their advantages in their non-flowability, anti-creeping and anti-irradiation properties, high surface adhesion, and on-demand reversible transitions between liquid and solid states upon exposure to strong shear forces or external stimuli. In this review, we provide a detailed overview of the hydrogel lubricants composed of polymer, inorganic particles, and the composites of both, and mainly discuss their synthetic methods, lubrication properties and mechanisms, and potential applications in bionic fields. Thus far, polymeric and polymeric-inorganic hydrogel lubricants have been intensively investigated due to the advantages of their tunable internal self-assembled structures, susceptibility to external stimuli, and resemblance to biological tissues [64,112]. Moreover, combining excellent tribological performances, simple synthesis process, and eco-friendliness, these features highlight inorganic hydrogels as potentially emerging materials for lubrication in the engineering, industrial, and biomimetic fields.

Although significant breakthroughs have been made in the development of hydrogel lubricants, there are still several critical issues that should be addressed:

(1) Regarding the preparation of hydrogel lubricants, it is still challenging to satisfy the requirements of practical applications owing to the limited production, as current methods typically generate products on a diminutive scale. It is essential to devote more effort to simplifying and optimizing the current synthetic strategies of hydrogel lubricants without degenerating their friction and wear properties.

(2) The lubrication mechanism of hydrogel lubricants with different microstructures needs to be further studied. It is necessary to explore the atomic-level interaction of hydrogels with the surface of friction substrates, the dynamic mechanisms of friction processes, the synergistic lubrication effect of components, and so on.

(3) At present, the evaporation and freezing of water make the most hydrogel lubricants that can only be used in humid environments and temperatures above zero, limiting their application, especially in extremely dry or cold weather. Moreover, the movement occurring between the hydrogel and the equipment during the process of running can also induce lubrication failure. Therefore, developing strong base-adhesion and environmentally robust hydrogel lubricants with effective anti-sliding, anti-dehydration, and anti-freezing properties to ensure the stable operation of mechanical devices is important.

(4) Although the lubricity of inorganic hydrogel can be comparable to polymer hydrogels, the formation of inorganic hydrogel networks primarily depends on weak hydrogen bonding or van der Waals interactions, leading to their inherent mechanical limitations in tensile strength and deformation resistance. To expand the applications of inorganic hydrogel lubricants, it is important to break through the limit.

In conclusion, to address these limitations, hydrogels capable of excellent lubrication, good mechanical properties, and macroscopic preparation are expected to be realized. The development of novel green hydrogel lubricants with desired performances is also expected to create a more energy-efficient and low-carbon society.

Author Contributions: Conceptualization, L.H. and L.X.; writing—original draft preparation, L.H.; writing—review and editing, W.Y. and L.X.; visualization, Y.Y.; supervision, L.X. All authors have read and agreed to the published version of the manuscript.

Funding: This work is financially supported by the National Natural Science Foundation of China (22202220 & 22302215), the Talents Program of Chinese Academy of Sciences (E30247YB), the Talents Program of Lanzhou Institute of Chemical Physics (E0SX0282), the National Natural Science Foundation of Shandong Province (ZR2022QB190), and the Innovative Research Funds of Shandong Laboratory of Advanced Materials and Green Manufacturing at Yantai (E1R06SXM07 and E1R06SXM09).

Data Availability Statement: Not applicable.

Conflicts of Interest: The authors declare no conflicts of interest.

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