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Review Article

Toward living lubrication: Designing hydrogels that learn, adapt, and respond

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Abstract

Biological lubrication rarely arises from a single, static low-friction state. Instead, living interfaces sustain efficient lubrication through the dynamic regulation of hydration, boundary renewal, fluid redistribution, and interfacial reconstruction in response to fluctuating environmental conditions. Hydrogels, as water-rich polymer networks with bioinspired adaptability, tunable chemistry, and structural diversity, provide an ideal platform for translating living tribological principles into synthetic materials. In this review, we summarize the evolution of hydrogel lubrication from conventional bioinspired designs to bioinspired adaptive lubricating hydrogels (BALHs). We then focus on BALHs, which integrate stimulus-responsive molecular motifs, reconfigurable interfaces, and stimulus-coupled transport or phase transitions. These systems enable lubrication states that are no longer immutable post-fabrication but can be renewed or reconfigured *in situ*. Adaptive lubrication is further discussed across mechano-, thermo-, photo-, pH-, electro-, magneto-, and multi-responsive systems, in which diverse external cues enable dynamic adjustment of interfacial states, boundary replenishment, and frictional performance. Emerging applications are highlighted in biomedical implants, soft robotics, and other dynamic tribological environments. Finally, we discuss the major challenges impeding practical translation, including the trade-off between responsiveness and durability, the stability of renewable boundary layers in complex media and multi-stimulus interference.

Keywords: Bioinspired hydrogel, Adaptive lubrication, Stimulus-responsive, Anti-friction

1. Introduction

Friction is ubiquitous at biological interfaces, including sliding, rotating, and rolling contacts, and poses persistent challenges to durability and functional efficiency [1–3]. To cope with these demands, living systems have evolved highly efficient lubrication strategies that minimize friction while preserving structural integrity under dynamic operating conditions [4–6]. However, biological systems rarely rely on a single, static lubrication state. Instead, nature often maintains low friction through adaptive lubrication, in which the interfacial state continuously adjusts in response to environmental changes [7,8]. For example, the peristome of *Nepenthes* remains grippable under dry conditions but becomes extremely slippery when wetted, thereby causing prey to slide into the trap [9]. Likewise, terrestrial gastropods such as snails and slugs can remain firmly attached under low deformation but generate a lubricious mucus layer during locomotion, thereby enabling smooth movement across solid surfaces [10]. Similarly, some intertidal fishes maintain a tough body surface under abrasive rocky conditions while secreting abundant mucus, which may help reduce friction and facilitate movement in confined spaces [11]. In the esophagus, mucosal tissues respond to transient pressure and shear by secreting saliva and mucins, instantly transforming the interface into a highly lubricious transport pathway [12]. The ocular surface likewise sustains efficient lubrication during repeated blinking through the continuous spreading

and renewal of the tear film, in which mucins contribute to hydration and interfacial lubrication [13,14]. These biological lubrication interfaces are governed by surface forces resulting from hydration layers, electrostatic interactions, and steric repulsion between hydrated macromolecules. These examples collectively demonstrate that effective lubrication in biological systems is often not an inherent property of the materials but a dynamic, environment-dependent, and interactive phenomenon with the environment [15–17].

Hydrogels, with their high-water content, tunable structure, biocompatibility, and bioinspired adaptive capabilities, have emerged as an ideal material platform for achieving efficient, low-consumption, and intelligent lubrication [18–24]. Current research efforts in this field have shifted toward transforming lubrication from a static, passive mechanism into a dynamically adjustable, active lubrication system, offering innovative solutions to the complex tribological challenges in biomedical and engineering applications [25–28]. The increasing number of publications on hydrogel lubrication also reflects the growing interest in this field (Figure 1), but the key challenge is no longer simply expanding material examples; rather, it is to clarify how different hydrogel designs regulate fundamental lubrication mechanisms under dynamic conditions.

Importantly, BALHs are not intended to replace conventional bioinspired hydrogels but rather to extend their functional boundaries, offering a paradigm shift in how lubrication is engineered at material interfaces. Conventional bioinspired hydrogels have successfully reproduced key structural and molecular features of natural lubrication

systems, whereas BALHs further incorporate stimulus-responsive or reconfigurable elements that enable the lubricating state to be dynamically regulated in situ rather than being fully fixed upon fabrication [29–32]. This regulation can be achieved through responsive molecular motifs, reconfigurable interfaces, and stimulus-coupled transport or phase transitions [33–37]. Such adaptability is of considerable importance for next-generation biomedical implants and coatings, where on-demand lubrication can reduce tissue damage and multifunctional switching may help control infection or inflammation [38–41]. Beyond biomedical applications, this capability is equally transformative for soft robotics, where reversible switching between high-friction (gripping) and low-friction (sliding) states can enhance locomotion efficiency [42–45]. Collectively, these attributes position BALHs as a versatile and intelligent material platform capable of addressing complex functional demands across diverse technological domains.

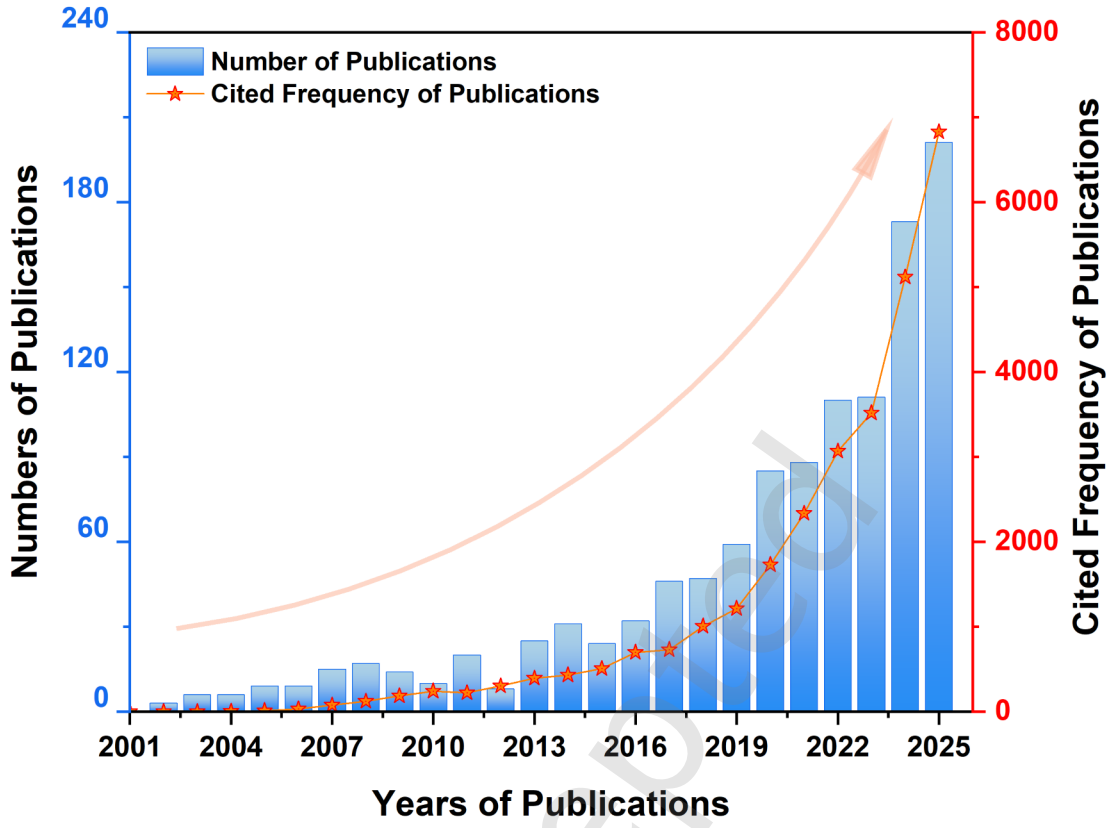

Figure 1. The annual publication and citation frequency data of hydrogels lubrication field from 2001 to 2025 (the insert shows the corresponding proportion of review articles to the annual publication of hydrogels in the lubrication field). This dataset was retrieved from the Web of Science database on January 5, 2026, using terms including "hydrogels" and "lubrication".

In this review, we first summarize representative conventional bioinspired lubrication strategies in hydrogels, focusing on how structural organization and interfacial hydration together regulate contact, load support, and boundary lubrication. We then focus on adaptive bioinspired lubrication, organized according to the external cues that govern interfacial regulation, including mechano-, thermo-, photo-, pH-, electro-, magneto-, and multi-responsive systems, in which external stimuli are converted into lubricant generation, interfacial renewal, friction switching, or coordinated

deformation–lubrication behavior (Figure 2).

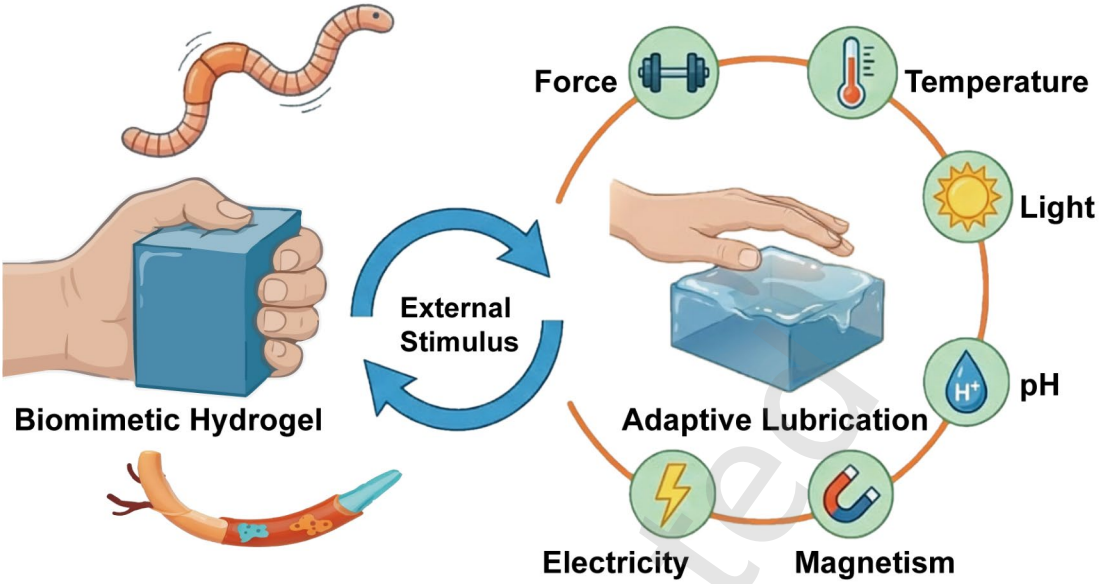

Figure 2. Schematic illustration of stimulus-responsive hydrogel lubrication. Representative stimuli and their typical lubrication-regulation mechanisms are summarized, including force-induced lubricant transport and boundary-layer renewal, temperature-regulated hydration/modulus transition, light-triggered interfacial switching or lubricant release, pH-mediated protonation/deprotonation and swelling changes, electrically driven ion/charge redistribution, magnetically induced deformation or interfacial transition, and multi-stimulus coupling for adaptive lubrication.

Beyond summarizing representative materials and mechanisms, we emphasize the broader significance of BALHs in biomedicine, soft robotics, and other dynamic tribological environments, where adaptive lubrication can minimize tissue damage, alleviate degeneration, integrate with regeneration, antifouling, or drug delivery, and enable reversible transitions between anchoring and sliding or between gripping and low-friction transport. We also discuss the major challenges that still limit translation, including the balance between responsiveness and durability, the stability of renewable boundary layers in complex media and stimulus interference in multi-responsive systems. Through this perspective, we aim to show that bioinspired hydrogels are

evolving from pre-designed lubricious materials into dynamic, self-adjusting, and multifunctional lubricating interfaces, and to provide a roadmap for the next generation of intelligent tribological soft matter.

2. Fundamental lubrication mechanisms of hydrogels

Friction is essentially an energy-dissipation process, whereas lubrication reduces this dissipation by altering how load, shear, and interfacial contact are accommodated. For hydrogels, this process is closely related to water retention, interstitial fluid transport, polymer-chain mobility, surface chemistry, and network deformation. Therefore, the lubrication behavior of hydrogels can be broadly understood through two basic modes: fluid lubrication and boundary lubrication.

Fluid lubrication occurs when a continuous or partially continuous liquid layer exists between the sliding surfaces. In this state, direct solid-like contact is largely avoided, and frictional energy is mainly dissipated through viscous shear of the interfacial fluid. For hydrogels, the lubricating fluid may originate from surface water, interstitial water within the polymer network, pressure-induced fluid exudation, or externally supplied lubricants. This allows the hydrogel network to temporarily support load through fluid pressurization and reduce direct contact between polymer-rich regions and the counterface. However, fluid lubrication is sensitive to long-term compression, dehydration, and lubricant depletion. Once the liquid layer is squeezed out, redistributed away from the contact region, or becomes too thin to separate the surfaces, the lubrication state gradually shifts toward boundary-dominated friction.

Boundary lubrication becomes dominant when the interfacial fluid layer is extremely

thin, discontinuous, or insufficient to fully separate the two sliding surfaces. In this regime, friction is mainly controlled by the physicochemical properties of the interfacial boundary layers, rather than solely by the viscosity of the fluid. These boundary layers may consist of hydrated polymer chains, polymer brushes, phospholipid assemblies, zwitterionic groups, adsorbed macromolecules, or sol-like supramolecular layers. Energy dissipation may arise from intermolecular adhesion, van der Waals interactions, electrostatic interactions, polymer-chain deformation or disentanglement, and rearrangement of interfacial molecules at the slip plane. Therefore, boundary lubrication is particularly important under high contact pressure, low sliding speed, long-term loading, or lubricant-starved conditions. Hydration lubrication is an efficient form of boundary lubrication in aqueous hydrogel systems. Charged, polar, or zwitterionic groups can strongly bind water molecules to form stable hydration layers. Under shear, hydration water can rapidly exchange with surrounding water molecules, allowing the hydrated layer to relax quickly and behave in a fluid-like manner. Therefore, hydrated boundary layers can simultaneously provide load-bearing capacity and reduce shear dissipation.

In practical hydrogel contacts, fluid lubrication and boundary lubrication are often coupled rather than completely separated. At the beginning of sliding, surface water or exuded interstitial water may reduce friction through fluid lubrication. During repeated loading, water redistribution, dehydration, surface wear, or boundary-layer depletion may increase molecular contact and shift the interface toward boundary lubrication. Therefore, well-designed hydrogels can enhance lubrication performance by retaining

water, replenishing boundary components, forming hydrated surface layers, or reconstructing interfacial lubricating layers during sliding. In the following sections, representative hydrogel systems are discussed in terms of how they establish, maintain, switch, or reconstruct these lubrication states under different structural designs and external stimuli.

3. Conventional bioinspired lubrication

In nature, biological tissues maintain lubricated interfaces and long-term resistance to deformation and wear through the coupling of bulk mechanical support, interfacial compliance, and fluid-mediated stress accommodation [6, 46-48]. Conventional bioinspired lubricating hydrogels achieve stable, durable, and low-friction interfaces under predefined conditions by mimicking the key principles underlying natural lubricating interfaces [49-52]. Therefore, in hydrogel tribology, bioinspired design remains a core and highly valuable strategy, because before lubrication can become adaptive, it must first be made robust, long-lasting, and mechanically compatible with the intended tribological environment.

In nature, low friction is often supported not only by hydration itself, but also by anisotropic architectures [53-57]. Mechanical anisotropy refers to direction-dependent bulk stiffness, strength, and deformation resistance, which mainly contributes to load-bearing support, contact-state stabilization, and reduced excessive deformation under load. In contrast, surface topographical anisotropy refers to directionally organized surface features, such as aligned fibers, microgrooves, micro-ridges, or oriented swelling patterns, which more directly affect sliding direction, interfacial contact area,

lubricant or water distribution, and therefore the direction-dependent coefficient of friction (COF).

A representative example is the scallion leaf, whose slippery behavior arises from the synergy of oriented surface micro-topography, directional bulk stiffness, and a hydrated mucus layer (Figure 3(a)). Inspired by this architecture, Liu et al. [58] developed an anisotropic layered lubricating hydrogel (ALLH) by chemically embedding thick hydrophilic polyelectrolyte brush chains into the subsurface of a high-strength anisotropic hydrogel bulk. In this system, mechanical anisotropy originates from the pre-stretched double-crosslinked polymer network and is reflected in direction-dependent tensile strength and elastic modulus. Its main role is to enhance load-bearing capacity and suppress excessive bulk deformation during sliding. By contrast, the anisotropic surface micro-morphology, including unidirectional swollen micro-ridges and directional infiltration behavior, more directly affects sliding-direction-dependent friction by regulating interfacial contact and hydration. Under the reported testing conditions, ALLH maintained COF values below 0.01 over a contact-stress range of about 0.2–2.4 MPa, with particularly low friction observed when sliding along the surface micro-strip direction.

Along a related but distinct route, Xu et al. [59] constructed sugarcane-inspired anisotropic composite hydrogels by filling hydrogel monomer precursors into aligned sugarcane nanofiber scaffolds, followed by free-radical polymerization (Figure 3(b)). The aligned nanofiber scaffold and hydrogen-bonding interactions give these hydrogels clear mechanical anisotropy, including direction-dependent tensile and tearing

properties. However, the direction-dependent COF is more directly associated with surface topographical anisotropy and hydration state along different sliding directions. For pAM and p(AM-co-AA) sugarcane composite hydrogels, the COF was lower along the parallel nanofiber direction than along the vertical direction, whereas pHEMA showed the opposite trend, which was attributed to its different surface hydration and wear behavior. Therefore, these examples show that mechanical anisotropy and surface topographical anisotropy contribute differently to hydrogel lubrication: the former mainly improves mechanical support and contact stability, whereas the latter more directly governs directional friction and COF anisotropy.

While anisotropic architectures regulate lubrication through directional mechanical support, biological interfaces can also maintain low friction through layered and compliant surface structures. Another representative example is the mucosa, which facilitates organ movement through a lubricative fluid layer while simultaneously serving as a protective biological interface and possesses a bilayer organization consisting of an epithelial layer and an underlying stromal layer. Inspired by this feature, Bai et al. [60] developed a mucosa-like conformal hydrogel coating, in which a thin hydrogel layer mimicking the epithelial surface was firmly anchored to the substrate through robust interfacial linkage (Figure 3. (c)). The resulting coating combined mucosa-matched compliance with a low COF of 0.018 ± 0.003, showing that contact-state regulation can also be achieved through a soft and conformal hydrated surface that minimizes interfacial damage during repeated motion.

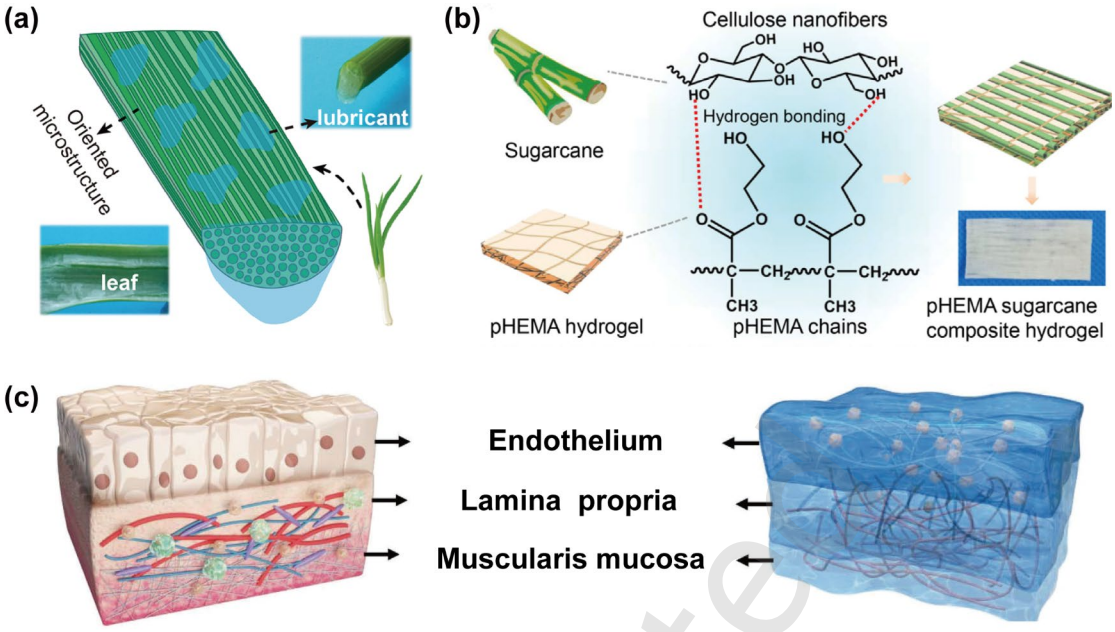

Figure 3. Bioinspired structural strategies for hydrogel lubrication. (a) Scallion-leaf-inspired layered anisotropic hydrogel. Reprinted with permission from Liu et al. [58] Copyright© 2024 Wiley-VCH GmbH. (b) Anisotropic composite hydrogels inspired by sugarcane. Reprinted with permission from Xu et al. [59] Copyright© 2021 Wiley-VCH GmbH. (c) Schematic diagram of the similarity between the hydrogel coating and the mucosa. Reprinted with permission from Bai et al. [60] Copyright© 2022 Wiley-VCH GmbH.

Besides structural organization for contact and mechanical support, the maintenance of low friction at biological lubrication interfaces also depends strongly on highly hydrated boundary structures at the slip plane [61-63]. In synovial joints, boundary lubrication is closely associated with phospholipid-containing surface complexes, in which phosphocholine-rich motifs provide strongly hydrated interfacial groups, while living tissues can also replenish such boundary components during use [64-67]. Accordingly, bioinspired hydrogels in this category mainly aim to engineer the sliding interface itself by stabilizing highly hydrated slip planes and constructing boundary-lubricating layers that remain effective during friction [68-72].

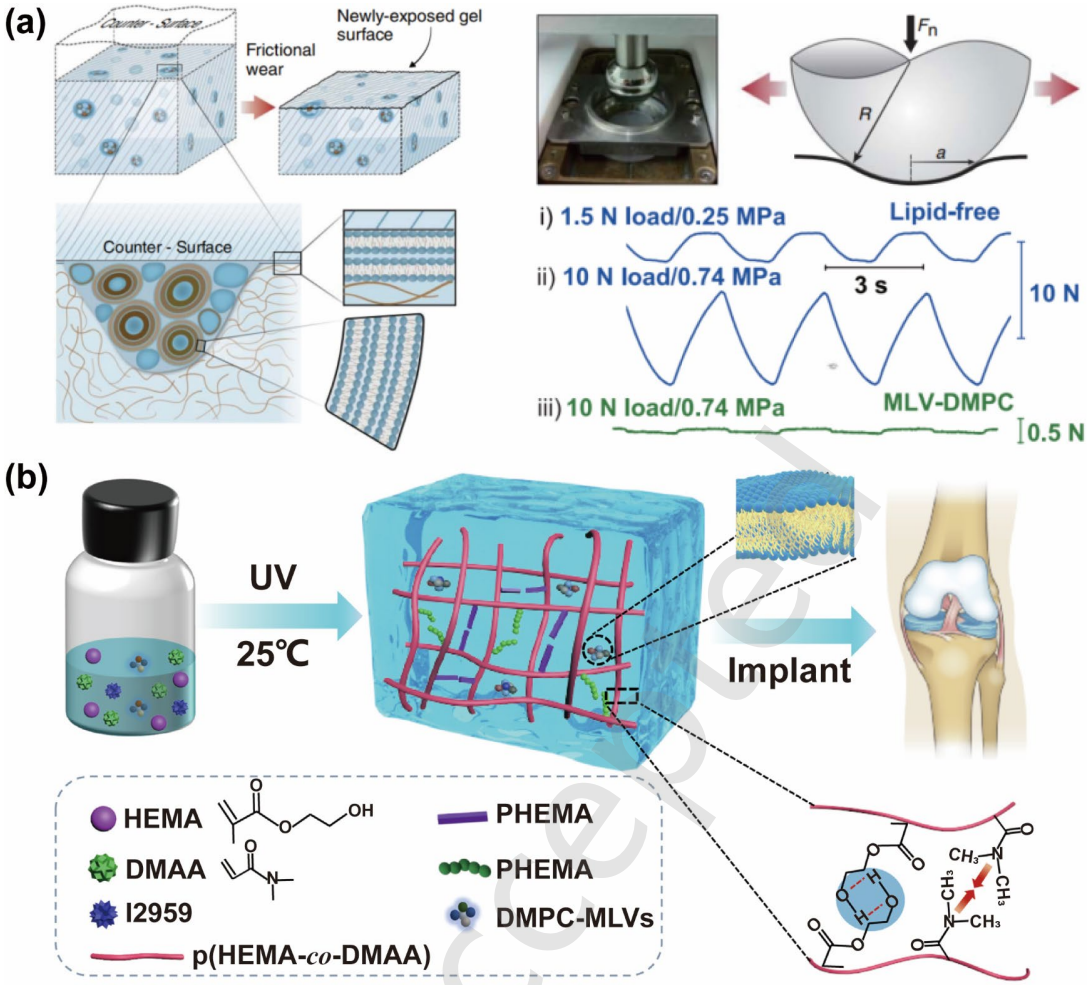

Figure 4. Design of chemical and molecular bioinspired lubricating hydrogels. (a) Inspired by cartilage, integrating PC liposomes into the hydrogel matrix can significantly reduce the friction of the material. Reprinted with permission from Lin et al. [73] Copyright© 2020 The American Association for the Advancement of Science. (b) Inspired by the lipid-assisted boundary lubrication in joints, DMPC was used as a lipid reservoir to synthesize lipid-based lubricating hydrogels. Reprinted with permission from Xiao et al. [81] Copyright© 2023 Elsevier B.V.

A representative example was reported by Lin et al. [73], who developed a cartilage-inspired hydrogel containing phosphatidylcholine lipid microreservoirs throughout the bulk network (Figure 4. (a)). In this design, friction progressively exposes new reservoirs, allowing the continuous reconstruction of a molecularly thin lipid-based boundary layer at the hydrogel surface. The low friction is therefore sustained by hydration lubrication at the slip plane between highly hydrated phosphocholine

headgroups. These zwitterionic groups bind water molecules strongly to form stable hydration shells, which generate short-range hydration repulsion and prevent direct surface contact. At the same time, rapid exchange of hydration water enables fluid-like shear, allowing the boundary layer to sustain load while maintaining low friction, while the internal microreservoirs compensate for boundary-layer depletion during wear [74-80]. As a result, the lipid-incorporating hydrogel exhibited sustained low friction, with a COF of approximately 0.005–0.02, corresponding to a friction and wear were reduced by 80% to 99.3% relative to the lipid-free gel. This study clearly illustrates how cartilage-like boundary lubrication can be translated into a renewable hydrogel interface through the coupling of interfacial hydration and internal lipid supply. Along a related line, Xiao et al. [81] embedded DMPC multilamellar vesicles into a hydrogel as lipid reservoirs. This reduced the average COF from 0.437 ± 0.021 for the blank gel to 0.069 ± 0.004 for the lipid-containing gel (Figure 4. (b)).

4. Adaptive Bioinspired Lubrication

Based on the structural and interfacial design principles of conventional bioinspired hydrogels, BALHs further endow these systems with the ability to regulate lubricating performance during use. The key advance lies in transforming the predesigned lubricating interface into a dynamic and adjustable interface that can respond to environmental changes. In this sense, BALHs are closer to the operating mode of living lubricating interfaces, where lubrication performance can be updated, reconstructed, or rebalanced in situ. This regulation can be achieved through stimulus-coupled lubricant transport, hydration renewal, boundary-layer regeneration, interfacial phase transitions,

modulus/contact-state switching, or other forms of interfacial reconstruction in response to external signals [82-85].

The following subsections are organized according to how different stimuli couple with hydrogel lubrication mechanisms. Mechanical cues are discussed first because they arise directly from the sliding contact itself and can trigger lubricant transport, boundary-layer renewal, or shear-induced gel–sol transition. Thermal, pH, and photoresponsive systems are then discussed as examples in which environmental or externally applied stimuli regulate hydration, swelling, phase transition, molecular conformation, or interfacial lubricant release. Electroresponsive and magnetoresponsive systems further extend this logic to field-regulated interfaces, where electrical or magnetic inputs can remotely modulate charge distribution, interfacial structure, deformation, or contact state. Finally, multi-responsive systems are discussed as integrated designs that combine more than one of these mechanisms to adapt to complex or fluctuating tribological environments.

4.1 Bioinspired Mechano-Adaptive Lubrication

In real sliding contacts, the lubricious state of conventional hydrogels cannot automatically adjust to changing forces. Under prolonged compression and repeated shear, boundary lubricating layers can be gradually depleted, while biphasic/poroelastic load support weakens as interstitial fluid redistributes [48, 86–88]. These limitations motivate the development of mechano-responsive adaptive hydrogels, in which mechanical inputs such as compression and shear are not merely external challenges,

but are instead converted into signals for lubricant transport, interfacial reconstruction, or hydration renewal.

In nature, earthworms move efficiently through soil under continuous compression and shear because their cuticle maintains a persistently slippery interface through the cooperation of wrinkled surface microstructures and gland-assisted lubricant replenishment. Inspired by this surface-texture-assisted retention and gland-assisted supply strategy, Ma et al. [82] developed a multilevel structural super-lubrication hydrogel (MS-SLH), in which a wrinkled, rapidly hydrating lubrication layer and gland-like textured pores together transform a static lubricant reservoir into a force-triggered self-pumping unit (Figure 5. (a)). Under relatively low contact pressure, lubrication is mainly governed by the hydrated surface layer; under higher pressure and repeated shear, however, lubricants are continuously transported from the gland-like reservoirs to the contact interface, thereby compensating for depletion and maintaining lubrication under starvation. Correspondingly, under fully hydrated conditions, the optimized SLH layer already exhibited excellent high-pressure superlubrication, showing a COF of about 0.0079 at 11.32 MPa and a stable ultralow-friction lifespan of 100,000 cycles at 8.48 MPa (COF ≈ 0.0028). After further introducing gland-like textured pores to construct the MS-SLH system, the material additionally gained sustained lubricity under lubricant-starved, quasi-wet conditions, maintaining lubrication in air for about 3700 cycles with only 5 μL of lubricant.

Natural mechano-responsive lubrication, however, is not limited to structural storage and release. In articular cartilage, loading and shearing do not simply consume

lubrication; they can also promote the dynamic exudation and redistribution of interstitial fluid and boundary components at the surface, a phenomenon often described as weeping lubrication [89-91]. This dynamic behavior is conceptually different from the conventional cartilage-inspired designs. Here, the bioinspired target is instead how cartilage behaves during motion. Inspired by this shear-coupled lubrication behavior, Zhang et al. [36] developed a bioinspired supramolecular hydrogel by integrating a thixotropic *N*-fluorenylmethoxycarbonyl-L-tryptophan (FT) supramolecular network with a PAAm/PVA double-network framework (Figure 5. (b)). In this design, the FT supramolecular network acts as the force-responsive component. Once shear is applied, the FT network undergoes a local gel–sol transition. This transition is governed by the reversible disruption of weak noncovalent interactions, such as hydrogen bonding and π–π stacking, which reorganize the interfacial structure and modify the surface forces within the confined aqueous layer. As a result, a sol-like lubricating layer appears at the surface; when shear is relaxed, the supramolecular structure can recover. The PAAm/PVA double network, meanwhile, provides the supporting skeleton. In this way, force is no longer simply a destructive factor, but becomes the trigger for interfacial lubrication generation. Correspondingly, the COF decreased from 0.0372 ± 0.0007 to 0.0233 ± 0.0021 during repeated shearing, confirming that shear can directly generate lubrication through interfacial molecular reorganization.

Following the same natural logic, later work moved one step further toward the way living tissues maintain function under continuous motion, where lubrication must not

only be activated under shear, but also recovered after damage in a dynamic environment [92–94]. Zhang et al. [95] further extended shear-induced sol-layer lubrication to a self-healing semi-convertible hydrogel (SHSCH) by integrating an FT supramolecular network, a self-healing poly(hydroxyethyl acrylamide) (PHEAA) network, and a rigid PVA covalent network (Figure 5. (c)). Under dynamic shear, the FT network disassembles to form a lubricating sol layer; after surface damage, the disassembled FT species can refill the scratched region, while hydrogen-bond reformation in PHEAA and π–π reassembly of FT restore both interfacial structure and lubricating function. As a result, the COF decreased from 0.62 ± 0.03 to 0.39 ± 0.03 in one formulation, and in an artificial worn cartilage model, the COF recovered from 0.68 ± 0.03 after wear to 0.09 ± 0.02 after dynamic self-healing.

Mechano-responsive adaptive lubrication is not only valuable for sustaining low friction under dynamic loading, but is also particularly relevant to diseased joints, where mechanical stress, inflammation, matrix degradation, and lubrication failure are tightly coupled. In such settings, force no longer acts merely as a tribological variable; instead, it becomes part of the pathological microenvironment that drives progressive cartilage damage. Accordingly, an important translational direction is to convert shear or loading from a source of degeneration into a trigger for boundary-layer renewal, therapeutic delivery, or regeneration-supportive interfacial regulation [96-98].

A representative example was reported by Lei et al. [33], whose design was inspired by the boundary-lubrication strategy of synovial joints (Figure 5. (d)). In native joints, phospholipid-containing boundary layers and other synovial components can cooperate

to maintain low friction, whereas under pathological conditions shear damage, inflammation, and lubrication failure often occur simultaneously. To mimic the renewable character of this biological boundary lubrication, they developed a shear-responsive boundary-lubricated drug-loaded hydrogel by incorporating celecoxib-loaded HSPC liposomes into a dynamic Schiff-base HA network. Unlike permanently crosslinked hydrogels, in which liposome microreservoirs remain immobilized within the bulk, the dynamic HA network can rearrange under shear and drive internal liposome reservoirs toward the sliding surface. In this way, shear is converted from a damaging factor into a cue for bioinspired boundary-layer renewal, while the hydrated HA matrix also helps sustain lubrication. Correspondingly, the average COF decreased from 0.062 ± 0.007 for PBS to 0.041 ± 0.004 for HA-gel and further to 0.031 ± 0.003 for Lipo@HA-gel. More importantly, the worn Lipo@HA-gel surface showed significantly more liposome signal than the pre-friction state (1.52 ± 0.09-fold versus 1.00 ± 0.09-fold), directly supporting shear-induced accumulation of boundary-lubricating reservoirs at the interface. This bioinspired force-responsive design was further coupled with celecoxib delivery, enabling the material to alleviate cartilage wear and attenuate osteoarthritis progression.

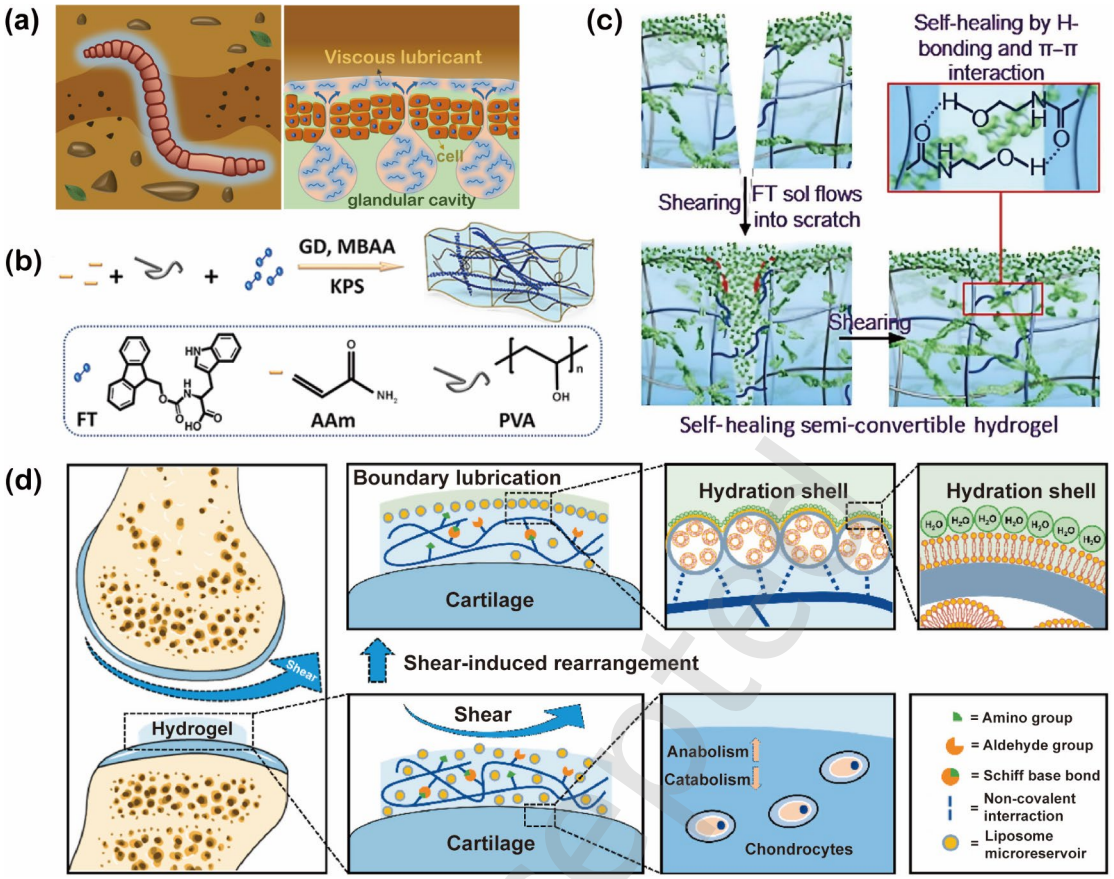

Figure 5. Schematic diagram of the mechanism of bioinspired force-responsive lubricating hydrogels. (a) The body surface of earthworms is covered with a lubricating layer (left), which is composed of hydrated viscous protein molecules secreted by cells or glands (right). Reprinted with permission from Ma et al. [82] Copyright© 2025 Springer Nature Limited. (b) The preparation of FT-PAAm/PVA supramolecular hydrogel. Reprinted with permission from Zhang et al. [36] Copyright© 2018 American Chemical Society. (c) Through the synergy of π-π stacking and hydrogen bonding, reversible recovery of active lubrication and mechanical integrity is achieved under dynamic shear. Reprinted with permission from Zhang et al. [95] Copyright© 2021 The Authors. Published by Chinese Chemical Society. (d) The hydrogels can expose internal liposome microreservoirs on the outer surfaces to form boundary layers via shear-induced structural rearrangements, and deliver celecoxib to improve the anabolic-catabolic balance of the extracellular matrix. Reprinted with permission from Lei et al. [33] Copyright© 2022 The Authors, under exclusive licence to KeAi Communications Co., Ltd.

Beyond therapeutic lubrication, mechano-responsive bioinspired design can also be extended to materials involved in cartilage regeneration. This is because in an environment of high-frequency movement, lubrication, load adaptation, and matrix

remodeling must coexist. Based on this concept, Qiu et al. [99] developed a shear-responsive and lubricating hyaluronic acid–chondroitin sulfate–decellularized matrix hydrogel for articular cartilage repair. Here, the bioinspired character lies not only in the use of HA, chondroitin sulfate, and cartilage-derived extracellular matrix components, but also in the attempt to reproduce the dynamic mechanical behavior of cartilage itself. The hydrogel can undergo a low-strain gel-to-sol transition under shear, while dECMMA fibers become oriented and aligned along the shearing direction, allowing the material to adapt to joint motion without losing lubricity. Correspondingly, the hydrogel achieves a stable coefficient of friction of about 0.082 under a 10 N load. In parallel, the HA/Chs-rich composition supports lubrication and hydration, whereas the dECM-containing dual-crosslinked structure provides a regenerative microenvironment and prolongs structural support during repair. Thus, this work extends force-responsive adaptive lubrication from friction reduction alone to a broader bioinspired paradigm of lubrication–regeneration coupling under realistic joint mechanics.

Taken together, these studies highlight lubrication as the central organizing theme of mechano-responsive hydrogel design. In natural systems, force is not only a source of frictional challenge, but also a cue for maintaining lubrication through lubricant secretion, fluid redistribution, and boundary-layer renewal. Inspired by these biological strategies, adaptive hydrogels increasingly use compression and shear not merely to resist mechanical damage, but to regulate lubrication itself—through lubricant generation, transport, renewal, therapeutic delivery, and lubrication-assisted tissue

repair. In this sense, the importance of force-responsive design lies in expanding lubrication from a passive low-friction state into an actively maintained and functionally integrated interfacial process. After these contact-driven mechanoresponsive systems, thermoresponsive designs provide another route in which lubrication is regulated by temperature-dependent hydration, modulus change, or lubricant release.

4.2 Bioinspired Thermoresponsive Adaptive Lubrication

Temperature is a pervasive and biologically relevant stimulus in living systems, not only because local warming accompanies exercise, inflammation, or external irradiation, but also because many biological interfaces change their lubrication state when temperature alters hydration, chain conformation, or tissue stiffness [100]. A simple but instructive example is fish skin, where a highly hydrated mucus layer gives rise to ultralow friction. Inspired by the hydrated mucus-mediated lubrication mechanism of fish skin, Wu et al. [101] developed thermoresponsive hydrogels that mimicked fish-skin lubrication and demonstrated that thermal stimuli could reversibly tune friction through hydration/dehydration-induced chain conformational changes. In their system, swollen chains at low temperature maintained an ultralow-friction state, whereas thermal collapse of the responsive network increased friction, thereby illustrating one of the earliest bioinspired demonstrations that temperature can act as a switch for hydrogel lubrication.

Natural thermal regulation of lubrication, however, is more sophisticated than simply changing hydration. In biological systems, frictional state can also be altered by

coupling a slippery surface with temperature-dependent changes in subsurface stiffness or body mechanics. A representative hydrogel example was provided by Liu et al. [102], who developed a layered thermoresponsive hydrogel by grafting a poly(3-sulfopropyl methacrylate potassium salt) (PSPMA) polyelectrolyte brush onto a poly(N-isopropylacrylamide-co-acrylic acid-co-initiator/Fe^{3+}) [P(NIPAAm-AA-iBr/Fe^{3+})] substrate (Figure 6. (a)). In this system, the top soft hydrogel/brush composite layer preserved aqueous lubrication, while the bottom thermoresponsive hydrogel layer adaptively stiffened above the lower critical solution temperature (LCST, 32.5 °C), thereby reducing friction through enhanced load-bearing capacity without sacrificing the hydrated surface state. More importantly, after integrating Fe_3O_4 nanoparticles, the hydrogel exhibited a typical in situ thermoresponsive lubrication behavior under near-infrared (NIR) irradiation, with the COF decreasing from 0.047 to 0.025. These results show that thermal cues can regulate lubrication through the cooperative tuning of surface hydration and subsurface mechanics, rather than by simple hydration/dehydration switching alone.

A more vivid natural analogy for this temperature–modulus–lubrication coupling is found in the longsnout catfish. Its skin is covered by a mucus layer that provides a slippery surface, yet when the fish struggles, internal muscle stiffening rapidly increases the skin modulus and changes the interfacial frictional state, making the fish much harder to catch. Inspired by this mucus-like lubricating surface and muscle-like thermally stiffening support strategy, Zhang et al. [83] developed a modulus-adaptive lubricating hydrogel (MALH) composed of a top hydrophilic lubricating layer and a

bottom thermo-stiffening phase-separation hydrogel. Upon heating, the bottom layer underwent rapid phase separation and stiffened dramatically, while the hydrated top layer remained lubricious; as a result, the material switched almost instantaneously from a soft/high-friction state (~0.3 MPa, COF ~0.37) to a stiff/lubricious state (~120 MPa, COF ~0.027) in water (Figure 6. (b)). Importantly, this work made clear that thermal adaptivity in lubrication need not rely solely on changing hydration; it can also operate through temperature-triggered contact-state regulation via modulus switching, which more closely resembles how biological tissues adapt their friction during active motion.

Thermoresponsive adaptive lubrication can be taken one step further when the biological inspiration shifts from external slipping surfaces to muscle itself, whose performance improves when warming promotes actin–myosin cross-bridge formation and greater isometric force generation with minimal length change. Ji et al. [103] introduced a muscle-mimetic thermomechanical route by intertwining linear poly(N-isopropylacrylamide) (L-PNIPAM) chains within a robust PVA network. Heating induced aggregation of L-PNIPAM within the PVA matrix, producing reversible isometric strengthening with limited macroscopic volume change. This internal polymer reorganization also switched the surface frictional state, with the COF decreasing from approximately 0.17 at 20 °C to about 0.02 at 35 °C under a 5 N model tissue contact. This example highlights the thermal adaptive lubrication achieved through internal network reconfiguration, thereby expanding thermal adaptive lubrication from a simple interface switching to a combination of internal polymer

reorganization and friction control, similar to the function of muscles (Figure 6. (c)), rather than relying solely on changes in surface hydration or the hardening of the supporting layer.

Thermally adaptive lubrication can also be achieved through a different route, namely using heat as a trigger for in situ lubricant generation rather than merely for friction or stiffness regulation. In the human body, the tendon sheath is a representative dynamically lubricated structure that combines reversible contraction with synovial-fluid secretion to maintain near-frictionless tendon sliding. Inspired by this sheath behavior, Yang et al. [104] designed a sheath-inspired durable semi-convertible hydrogel (SheathSCH) that integrates a contractile PNIPAM/PVA covalent framework with a supramolecular gelatin/polydopamine lubricant-generating component. In this design, near-infrared irradiation induces photothermal heating, which simultaneously contracts the PNIPAM/PVA framework and triggers gelatin gel–sol transition, thereby generating a lubricating sol phase in situ. This system thus moves toward on-demand thermally triggered lubricant secretion, much closer to the living lubrication behavior of tendon sheaths. Notably, after introducing hydrophosphatidylcholine, the hydrogel reached a superlubricity level (COF ≈ 0.007) and maintained extremely low wear even after 100,000 shearing cycles, illustrating how thermal adaptivity can be coupled with durability and controllable actuation.

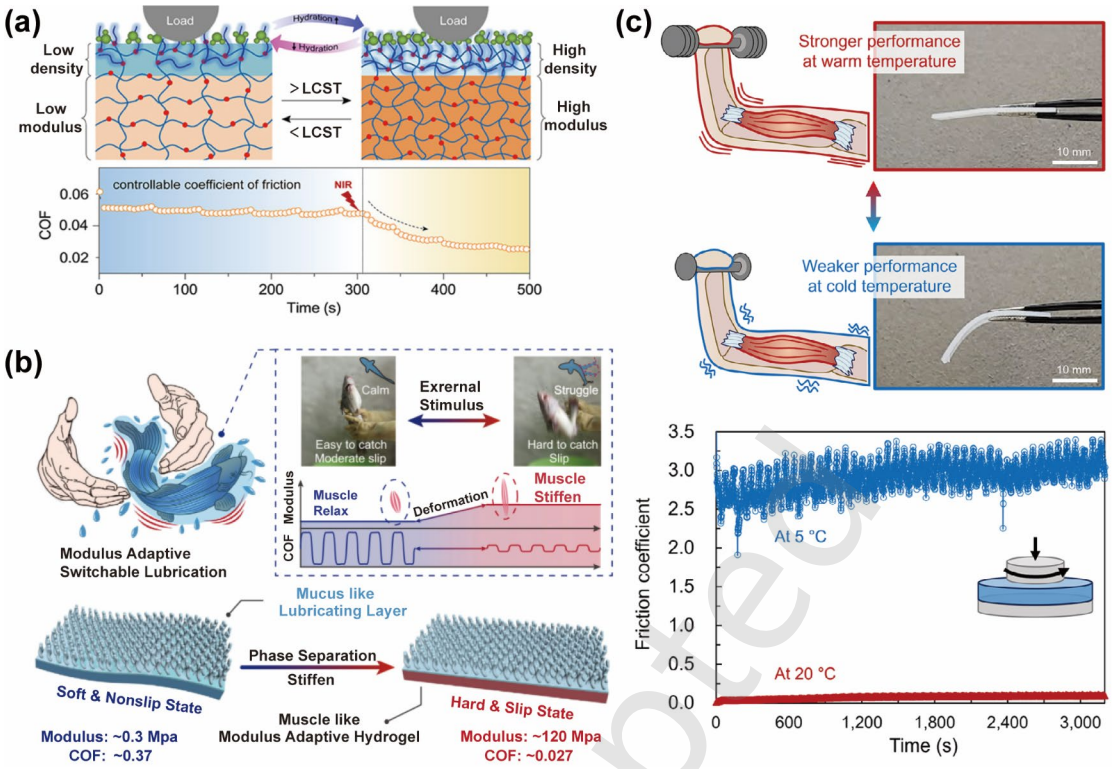

Figure 6. Schematic diagram of the mechanism of bioinspired thermoresponsive adaptive lubricating hydrogels. (a) Cartilage mimics adaptive lubrication. Reprinted with permission from Liu et al. [102] Copyright© 2020 American Chemical Society. (b) Schematic diagram of the MALH concept structure inspired by the modulus adaptive switchable lubrication and instantaneous muscle hardening mechanism of fish during fishing. Reprinted with permission from Zhang et al. [83] Copyright© 2022 Springer Nature Limited. (c) Thermomechanical double-network hydrogels for mimicking isometric muscle motion and their COF at 20 and 35 °C. Reprinted with permission from Ji et al. [103] Copyright© 2025 Wiley-VCH GmbH.

Thermoresponsive lubrication has also been extended to on-demand release systems. Lv et al. [132] designed heterogeneous organohydrogels with water-driven and thermally driven oil-release capabilities, where environmental changes induce dispersion-medium exchange and deswelling, enabling controllable oil secretion for self-lubrication and antifouling applications. This route differs from modulus-switching hydrogels because the thermal cue mainly regulates lubricant release rather than only contact stiffness. Thermoresponsive hydrogel microspheres have also been explored for

localized therapy. Xu et al. [133] developed biocompatible HPC-based hydrogel microspheres with temperature-controllable drug release and lubricating properties. Their design uses thermally induced hydrogel shrinkage near physiological temperature to promote on-demand release while retaining lubricity and biocompatibility. These examples broaden thermoresponsive lubrication from contact-state regulation to controlled lubricant or drug release.

On the whole, thermoresponsive adaptive lubrication can be realized through several distinct but related bioinspired routes. It may begin with fish-skin-inspired hydration switching, extend to the cooperative regulation of surface hydration and subsurface mechanics, develop further into catfish-inspired modulus-switchable lubrication, and then advance to muscle-mimetic internal polymer reorganization and tendon-sheath-inspired in situ lubricant generation. In this sense, thermal adaptive lubrication is not merely about heating a hydrogel to change friction; rather, it is about using temperature to regulate interfacial hydration, contact-state support, bulk modulus, internal polymer conformation, and even on-demand lubricant secretion, thereby bringing synthetic hydrogel interfaces closer to the dynamic and multifunctional lubrication strategies of living tissues. Compared with temperature, light provides a more spatially and temporally controllable external stimulus, enabling remote regulation of hydrogel lubrication through photothermal, photochemical, or supramolecular pathways.

4.3 Bioinspired Photoresponsive Adaptive Lubrication

Light is a distinctive biological cue because living systems frequently use it to regulate function in a non-contact, spatially precise, and temporally controllable manner, as

exemplified by processes such as photosynthesis, phototaxis, and phototropism [105,106]. Inspired by this broader natural principle, photoresponsive hydrogels offer an appealing route for adaptive lubrication, because light can modulate interfacial states remotely without direct mechanical intervention or chemical dosing.

A traditional bioinspired route to photoresponsive lubrication was to use light not as a direct chemical switch, but as a remote regulator of modulus and contact state. In this context, Zhao et al. [107] developed an MXene-based hydrogel system that combined NIR photothermal conversion, a thermally stiffening hydrogel substrate, and a poly(3-sulfopropyl methacrylate potassium) (PSPMA) polyelectrolyte brush (Figure 7. (a)). The biological inspiration behind this design was twofold, adaptive cartilage lubrication, where a hyperhydrated surface cooperates with a load-bearing subsurface, and catfish-inspired friction control, where increased surface modulus alters interfacial tribology. Under ambient conditions, the hydrogel remained soft and exhibited a relatively high COF of about 0.24; under NIR irradiation, MXene raised the local temperature, strengthened the substrate through phase separation, and reduced the COF to about 0.10 while the hydrated PSPMA brush preserved aqueous lubrication. In this way, light served as a remote trigger for photothermally controlled friction reduction, extending the logic of thermoresponsive adaptive lubrication into an optically addressable format.

Yet natural light-regulated behavior is not limited to remote heating. Another photoresponsive route is based on light-triggered interfacial state conversion, in which optical stimulation induces the formation or removal of a lubricating surface layer. This logic was realized by Wang et al. [85] in their semi-convertible hydrogel enabled

photoresponsive lubrication. Their design integrated a responsive supramolecular α-cyclodextrin/polyethylene glycol (α-CD/PEG) network and a competitive photoresponsive guest 1-[p-(phenylazo)benzyl]pyridinium bromide (AzoPB) within a tough PVA/PAAm framework (Figure 7. (b)). Under visible light, trans-AzoPB competitively disassembled the α-CD/PEG supramolecular network, driving a partial gel–sol transition and producing a lubricating sol layer on the hydrogel surface, while the PVA/PAAm framework preserved structural integrity; under UV light, the system reassembled and the sol layer disappeared. This reversible appearance/disappearance of a surface sol layer allowed lubrication to be directly controlled by light. Correspondingly, with a thin water film on the surface, the COF of PSCH50 was about 0.0144 ± 0.0035 after UV irradiation but decreased further to 0.0045 ± 0.0022 after visible-light irradiation, and the photoresponsive lubricating behavior remained reversible over multiple cycles. Moreover, histological and force–distance observations showed that the lubricating layer appeared rapidly and could reach a thickness of roughly 90 μm after 2 h of visible-light treatment, confirming that light here does not merely tune bulk mechanics but actively reconstructs the interfacial lubricating state.

Additional photoresponsive systems further broaden the optical regulation routes of hydrogel lubrication. Wang et al. [134] recently developed a photoresponsive double-network hydrogel for ocular lubrication, in which a PVP hydrogel framework was combined with a β-CD/PEG host–guest system and a competitive azobenzene derivative guest (Azo-C_2). Under visible-light irradiation, Azo-C_2 competitively binds to β-CD and releases PEG chains, which migrate toward the hydrogel surface and

artificial tear medium to form a hydrated lubricating layer. This design achieved a 25% reduction in COF after 6 h of irradiation and enabled superlubricity in the soaking solution. Unlike photothermal systems or light-induced gel-sol transition hydrogels, this example represents a molecular-lubricant-release route for photoresponsive lubrication.

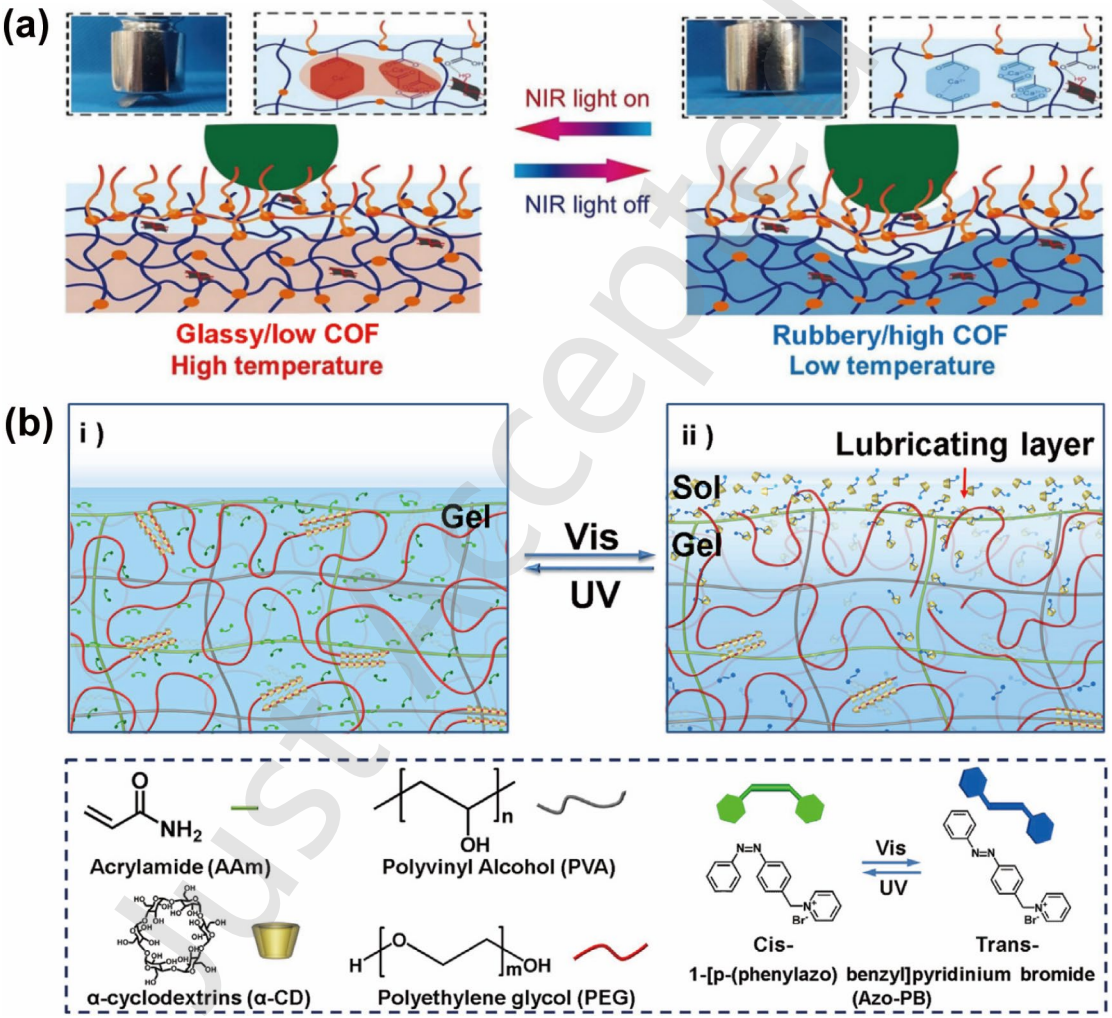

Figure 7. Schematic diagram of the mechanism of bioinspired photoresponsive lubricating hydrogels. (a) Controlling interface lubrication with near-infrared light. Reprinted with permission from Zhao et al. [107] Copyright© 2024 SciOpen. (b) The possible photoresponsive lubricating mechanism of the prepared photoresponsive semi convertible supramolecular hydrogel under (i) UV and (ii) Vis irradiation. Reprinted with permission from Wang et al. [85] Copyright© 2020 The Authors. Published by Elsevier Ltd.

The next step in this story is to move from light-triggered sol-layer generation to direct photochemical control of hydrogel hydrophilicity and friction. In this direction, Chau et al. [108] provided a complementary photochemical strategy by incorporating methoxy-spiropyran motifs into a PNIPAM-based hydrogel. Under 470 nm light, the hydrophilic merocyanine form converted to the more hydrophobic spiropyran form, causing deswelling, increased modulus, and higher friction. The COF increased from about 0.022 ± 0.002 in the dark to about 0.085 ± 0.039 after irradiation, showing that molecular polarity switching can directly regulate hydrogel friction.

Overall, photoresponsive adaptive lubrication originated from the natural inspiration that light is an extremely precise biological control signal, and then evolved from remote modulus regulation to photothermal friction control, further advanced to light-induced gel-sol interface transformation, forming a lubricating layer on demand, and finally developed into direct control of hydrophilicity and friction at the molecular level through photochemical switches. They are bioinspired adaptive interfaces that utilize light to regulate surface hydration, interfacial phase, overall stiffness, and the formation of lubricating layers, with spatial and temporal precision that is difficult to achieve solely through force or heat.

4.4 Bioinspired pH-Responsive Adaptive Lubrication

In living systems, pH is not a static background parameter but a highly organized environmental cue. Biological fluids span a broad pH range, blood and interstitial fluids are nearly neutral, gastric environments are strongly acidic, and bile or pancreatic fluids are mildly alkaline [109,110]. Correspondingly, tissue functions such as lubrication,

nutrient transport, and interfacial signaling are all influenced by local pH [111,112]. This natural pH heterogeneity provides an important bioinspired example for adaptive lubricating materials to sense the changes in protonated environments and convert them into alterations in hydration states, interfacial structures, and frictional states.

The most direct translation of this idea is to exploit the pH-dependent ionization of weak polyelectrolytes, because protonation and deprotonation can reorganize electrostatic interactions, swelling, and hydration lubrication at the hydrogel surface. A representative early example is the [P(AAm-co-AA)] system studied by Chau et al. [113], in this design, AA serves as the pH-responsive unit, when the surrounding environment changes, the protonation state of the acrylic acid groups changes accordingly, altering electrostatic interactions at the interface and thereby tuning friction. Rather than mimicking a single anatomical structure, this strategy more broadly emulates how biological aqueous interfaces adjust function across different pH microenvironments. The tribological response was striking, because by varying pH and acrylic acid content, the COF could be tuned over nearly two orders of magnitude, and hydrogels with the highest AA content achieved superlubricity, with COF = 0.005 ± 0.001 in deionized water and COF = 0.005 ± 0.002 in 0.5 M NaOH. Friction generally decreased with increasing AA content and increasing pH, supporting the view that pH-regulated protonation and electrostatic interactions can directly control hydrogel lubricity.

Natural pH adaptation, however, is often more than a gradual change in charge density; in many biological contexts, local acid–base environments can trigger more discrete changes in interfacial state. This inspires a second strategy of pH-responsive lubrication,

pH-triggered partial gel–sol transition, in which the interface transforms into a lubricating sol layer under specific pH conditions. Kang et al. [114] implemented this idea by embedding a dynamic PVA–borax network within a covalent PAAm framework and further introducing carbon dots (CDs) to enhance both mechanics and lubrication (Figure 8. (a)). Here, the biological inspiration again comes from pH-diverse body environments, but the synthetic translation is more explicit in that when the pH drops below the effective borate-ester stability window, the dynamic PVA–borax network partially disassembles, causing a partial gel–sol transition while the PAAm framework preserves the bulk gel state. As a result, a pH-triggered lubricating sol layer appears on the hydrogel surface. Specifically, at pH 6.0, a surface sol layer was observed, whereas no such layer appeared at pH 8.5 or after treatment with water alone. The friction response tracked this transition closely. For example, in a representative carbon-dot-enhanced pH-responsive lubricating hydrogel (CPLH), the COF decreased from 0.069 ± 0.011 to 0.041 ± 0.003 after treatment with pH 6.0 solution for 10 min, and the response was reversible when the system was switched back to alkaline conditions. Thus, pH here does not merely tune the degree of hydration; it triggers a surface-state conversion that actively generates a lubricating layer.

A further step toward practical pH-responsive adaptive lubrication is to combine pH-triggered state regulation with durability, self-healing, and injectability, because real lubricating environments are not only chemically variable but also mechanically demanding. Huang et al. [115] addressed this by developing an injectable and self-healing MXene-reinforced pH-responsive hydrogel based on polyvinylpyrrolidone (PVP),

phytic acid (PA), MXene, and glycerol (Figure 8. (b)). In this system, pH responsiveness originates from the pH-dependent noncovalent interactions within the gel network, so that under acidic conditions the system tends toward a sol state, whereas near neutral pH it forms a stable gel with favorable lubrication performance; when the pH becomes overly alkaline, the network again partially dissociates and friction rises. This design therefore mirrors a biologically meaningful requirement in that the lubricant should not only respond to pH, but should also maintain a useful low-friction window under the pH conditions most relevant to practical application. The resulting hydrogel displayed durable lubrication, maintaining a low-friction state for 3 h and reaching a COF of 0.01, while the addition of MXene and glycerol produced a synergistic friction reduction of 69% and wear-resistance improvement of up to 96%. Importantly, the pH-responsive gel/sol regulation remained compatible with self-healing, injectability, and biocompatibility, pushing pH-responsive lubrication beyond proof-of-concept switching toward more deployable intelligent lubricants.

Additional pH-responsive systems further demonstrate that pH regulation can be coupled with lubrication, antibacterial protection, and disease-microenvironment remodeling. Cheng et al. [135] developed a pH-triggered antibacterial and lubricating hydrogel coating for urinary catheters, in which an outer pH-responsive MgO@AAm/SA/TA hydrogel layer provided hydration lubrication and accelerated MgO release under alkaline infection-like conditions. In another biomedical direction, Zhang et al. [136] reported a lubricating and dual-responsive injectable hydrogel based on ZIF-8@Que for osteoarthritis treatment, combining thermosensitive gelation, HA-

assisted lubricity, and pH/ROS-responsive drug release. These examples indicate that pH-responsive lubricating hydrogels are not limited to friction reduction alone, but can also be integrated with antibacterial or therapeutic functions.

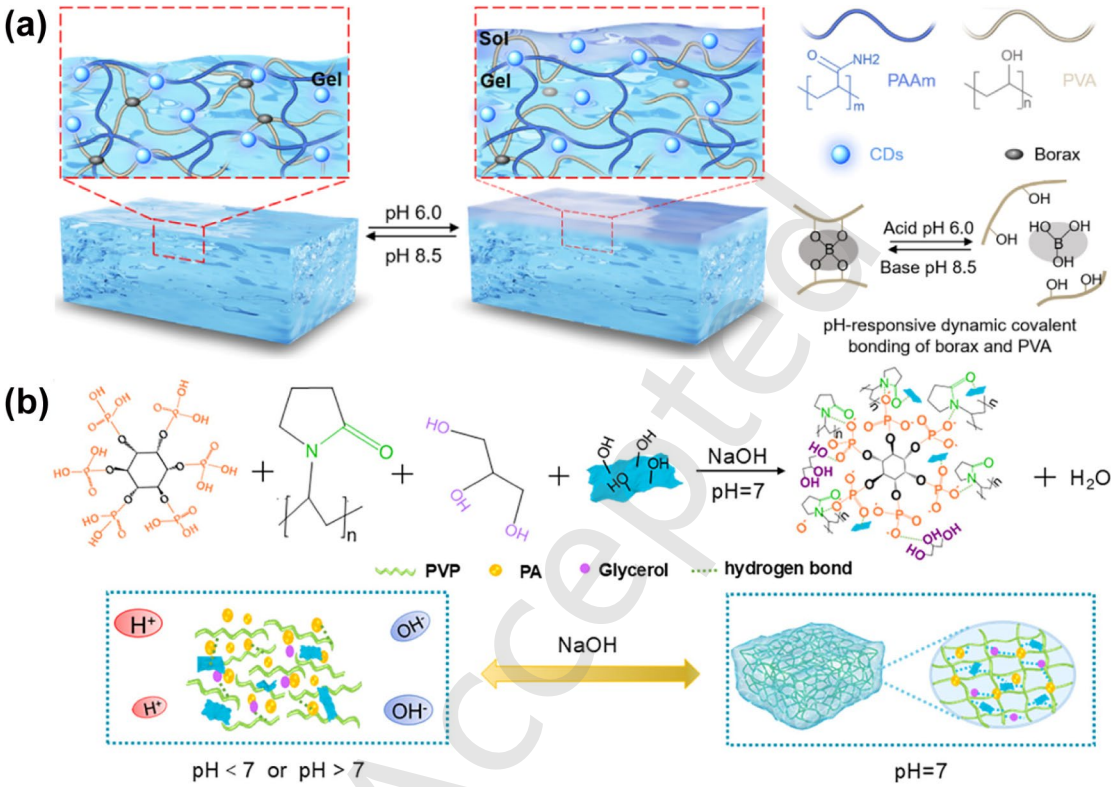

Figure 8. Schematic diagram of the mechanism of bioinspired pH-responsive lubricating hydrogel. (a) The lubricating mechanism of the CDs-enhanced pH-responsive lubricating hydrogel, and composition of the prepared hydrogel. Reprinted with permission from Kang et al. [114] Copyright© 2024 The Authors. Published by Elsevier Ltd. (b) Formation and mechanism of a pH-responsive PMGP hydrogel. Reprinted with permission from Huang et al. [115] Copyright© 2024 American Chemical Society.

In conclusion, pH-responsive adaptive lubrication originated from the natural observation of living tissues operating in highly heterogeneous pH environments, where interfacial functions, including lubrication, must remain effective. It first developed into pH-controlled electrostatic and hydration lubrication in weak polyelectrolyte hydrogels, then advanced to pH-triggered partial gel-sol interface

transitions, thereby generating lubricating sol layers on demand, and finally expanded to durable, injectable, and self-healing pH-responsive lubricants suitable for more realistic application conditions. In this sense, pH-responsive lubricating hydrogels are bioinspired adaptive interfaces where pH is used to regulate ionization, hydration, interfacial phase states, and lubricating layer formation, thus bringing synthetic lubricants closer to the environmental adaptability behavior of living tissues. While pH-responsive systems rely on chemical microenvironment changes, electroresponsive systems introduce a more directly programmable field input to regulate charged interfaces, swelling, and friction.

4.5 Bioinspired Electroresponsive Adaptive Lubrication

Electric signals are a distinctive regulatory cue in living systems because many biological interfaces do not merely maintain a static wet surface, but instead dynamically adjust interfacial states under bioelectrical and biochemical signals [116–118]. A representative example is mucosa, where secreted mucus continuously protects and lubricates ocular, gastrointestinal, respiratory, and cervical interfaces while also supporting signal communication. Inspired by this mucus-secreting capability of mucosa, Kang et al. [84] developed a mucosa-inspired electro-responsive lubricating supramolecular–covalent hydrogel (Figure 9. (a)). In this design, an electro-responsive silk fibroin supramolecular network was embedded into a tough PAAm/PVA covalent framework. The key bioinspired idea is that the supramolecular silk fibroin network mimics the dynamically secreted mucus phase, whereas the PAAm/PVA network plays the role of the mechanically stable tissue framework. When an electric field is applied,

the silk fibroin supramolecular network near the cathode disassembles because of electrostatic repulsion, causing a partial gel–sol transition and generating a surface sol layer that functions as a lubricating film; when the polarity is reversed, the supramolecular network reassembles and the lubrication state correspondingly recovers. In other words, this hydrogel does not simply change friction under electricity. Rather, it mimics electrically triggered mucus secretion by converting an electric cue into interfacial sol-layer generation. Correspondingly, the COF of a typical ESCH sample decreased from 0.115 ± 0.005 to 0.062 ± 0.003 under a 30 V field, and the diffusion of the sol layer could already be observed on a very short timescale after the electric field was turned on. This example most directly illustrates the bioinspired logic of electroresponsive lubrication, in which a bioelectric cue induces a secretion-like interfacial transformation and, in turn, renews lubrication at the interface.

This bioinspired concept is further strengthened by a recent cell-inspired study on friction regulation across lipid bilayers under transmembrane electric fields. Zhang et al. [119] showed that electric fields resembling those across cell membranes can induce a massive and fully reversible modulation of friction, reaching a 200-fold variation between phosphatidylcholine-bearing interfaces. Mechanistically, atomistic simulations revealed that the transverse field triggers reversible electroporation of the confined bilayers together with the formation of inter-bilayer bridges analogous to fusion stalks. These structural changes reduce hydration at the slip plane and partially shift sliding from the low-dissipation hydrated phosphocholine-headgroup interface to the much more dissipative acyl tail–tail interface, thereby dramatically increasing

interfacial dissipation. The process is reversible when the electric field is removed. It provides an important mechanistic basis for electroresponsive lubricating hydrogels. Bioelectric-style fields can directly reconfigure lipid-like boundary layers and strongly regulate interfacial dissipation through hydration loss and topological reconstruction.

Once this principle is established, electrical control can be extended beyond secretion-mimetic systems to a broader class of adaptive lubricating interfaces. In this broader sense, an electric field can also serve as a highly precise external signal for actively switching frictional states, particularly in conductive or ionic hydrogels where interfacial charge distribution, ion transport, or boundary-layer composition can be reorganized in real time. A representative example is the electrotunable ionic hydrogel reported by Liu et al. [120], in which a PVA/LiTFSI ionic hydrogel was used as an electroresponsive frictional material (Figure 9. (b)). It extends the same adaptive concept toward a more general tribotronic interface, electrical input is converted into real-time changes in interfacial lubrication. Mechanistically, the applied electric field drives ion migration and electroosmotic redistribution at the contact interface; when the hydrogel is positively charged and the metal counterpart is negatively charged, a removable salt-rich interfacial boundary film forms and dramatically lowers friction, whereas opposite polarity promotes oxidation of the metal surface and increases friction. As a result, the coefficient of friction can be reversibly modulated by more than fifty-fold, decreasing to ≈0.03 at −30 V while remaining at 1–2 at 0 V or +30 V, without the need for external liquid lubricants. Notably, this electrotunable friction was further

translated into applications such as a crawling robot and robotic gripping, demonstrating how electrically regulated lubrication can be coupled to adaptive motion.

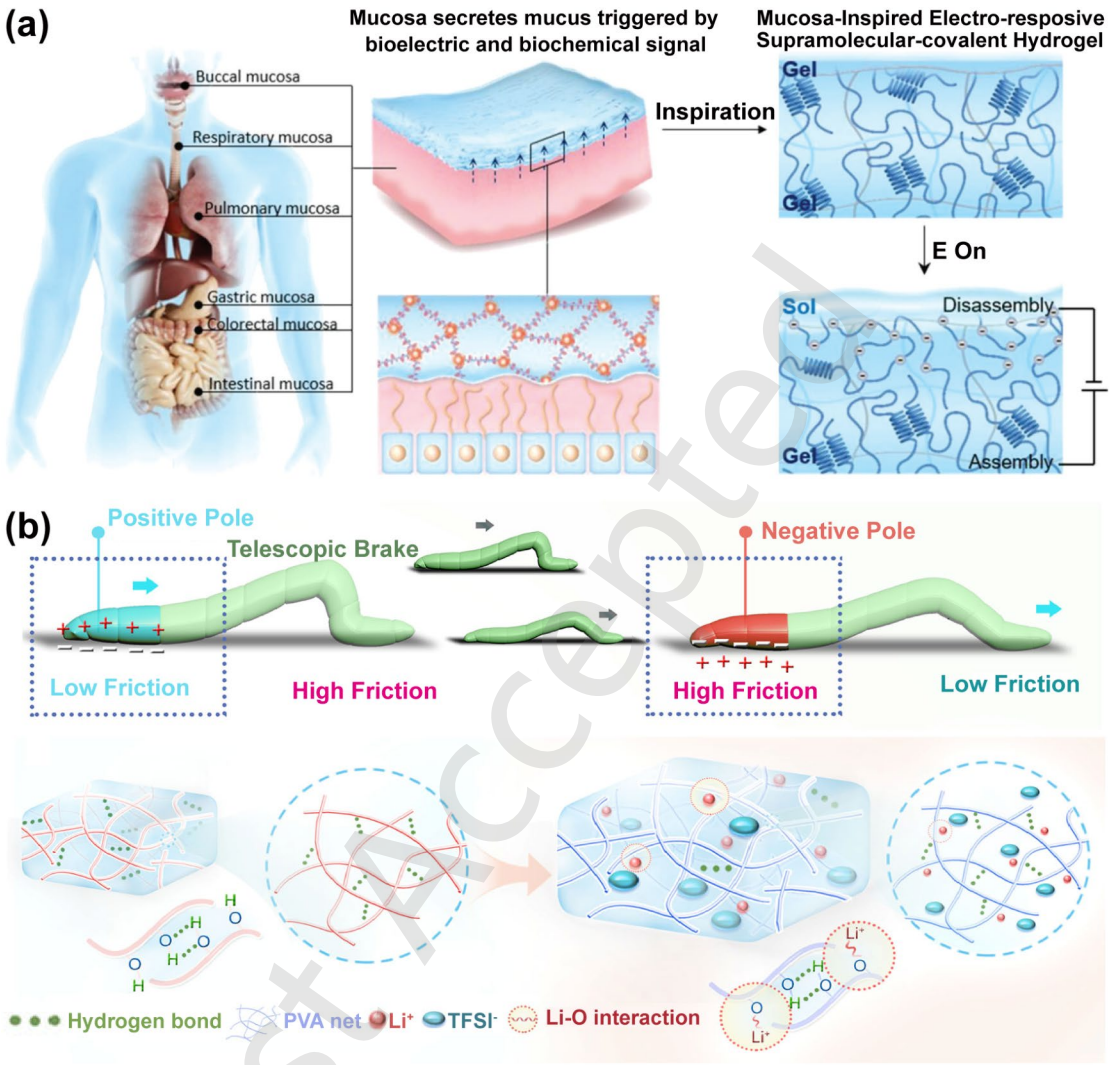

Figure 9. Schematic diagram of the mechanism of bioinspired electroresponsive lubricating hydrogel. (a) The electroresponsive mechanism of the mucosa-inspired supramolecular-covalent hydrogel under an electric field. Reprinted with permission from Kang et al. [84] Copyright© 2023 Wiley-VCH GmbH. (b) Schematic illustration of design principles of electrotunable friction. Reprinted with permission from Liu et al. [120] Copyright© 2025 Wiley-VCH GmbH.

Mechanistic studies on charged double-network hydrogels also provide useful evidence for electrically tunable hydrogel friction. Lee and Espinosa-Marzal [118] investigated agarose/poly (acrylamide-co-acrylic acid) double-network hydrogels and showed that intrinsic charge, salt concentration, swelling, and heterogeneous surface microstructure

jointly determine friction and adhesion. In a proof-of-concept experiment, electric bias dynamically altered surface topography, stiffness, adhesion, and friction, indicating that charged hydrogel interfaces can be regulated by electrical input. This work provides important mechanistic support for electroresponsive friction control.

From a bioinspired perspective, electroresponsive adaptive lubrication represents a progression from biologically inspired electrically triggered secretion to artificially programmed electrotunable frictional interfaces. It begins with inspiration drawn from mucosal tissues that dynamically regulate mucus secretion and lubrication under bioelectrical cues, advances to hydrogels that convert electric inputs into partial gel–sol transition and lubricating-layer generation, and is further supported by cell-inspired mechanistic evidence showing that electric fields can directly reorganize lipid-like hydrated boundary layers through electroporation and bridge formation. Electroresponsive lubricating hydrogels should therefore be understood not merely as electrically driven materials, but as bioinspired adaptive interfaces in which electrical cues are harnessed to regulate interfacial phase state, lubricant generation, boundary-layer dissipation, and frictional function with high precision and reversibility. Magnetic fields provide another remote actuation mode, particularly useful when deeper penetration or contactless deformation control is desired.

4.6 Bioinspired Magnetoresponsive Adaptive Lubrication

Magnetic fields can regulate material behavior remotely, noninvasively, and directionally, which makes them especially attractive for adaptive soft materials operating in confined or inaccessible environments [121–123]. However, a magnetic field

by itself does not directly provide a biological lubrication mechanism. The more meaningful bioinspired inspiration comes from living systems in which tissues preserve an integrated body while locally secreting or presenting a liquid lubricating phase at the surface. Examples include synovial fluid on cartilage surfaces, mucus on mucosa, and the lubricating secretions used by organisms such as snails, fish, and earthworms during locomotion [10, 124, 125]. These natural interfaces suggest a key design principle for magnetoresponsive lubrication, an ideal bioinspired system should retain its structural integrity while generating a localized lubricating sol layer on demand.

Inspired by this living liquid lubrication mechanism of biological systems, Gao et al. [126] developed a magnetoresponsive semi-convertible hydrogel (MSCH) by embedding a gelatin/Fe_3O_4-NH_2 supramolecular network within a PVA covalent framework (Figure 10. (a)). Under a 300 mT magnetic field, magnetic forces disrupted the gelatin-based supramolecular network near the surface, inducing a localized gel–sol transition and generating a lubricating sol layer while the covalent framework preserved the bulk gel state. The COF decreased from 0.071 ± 0.004 to 0.022 ± 0.006, and the sol layer grew to about 45 μm after 60 min, demonstrating magnetic-field-triggered boundary-layer generation with limited residue release.

This bioinspired design translated directly into adaptive lubrication performance. Under a magnetic field of 300 mT, the COF of the optimized MSCH decreased from 0.071 ± 0.004 to 0.022 ± 0.006, accompanied by progressive growth of the surface sol layer, which reached about 45 μm after 60 min. The friction reduction remained evident across different humidity, load, and sliding-velocity conditions, and the hydrogel exhibited

reversible On/Off lubricating behavior during multiple magnetic cycles. Importantly, because lubrication was generated through surface-localized disassembly rather than whole-body liquefaction, the hydrogel could move through narrow channels, overcome obstacles, and even escape from tweezers with much lower residue than a conventional sol-state magnetofluid. Thus, this work provides a representative example of magnetoresponsive lubrication, in which a magnetic signal is converted into partial interfacial gel–sol transition and on-demand lubricating-layer generation while the material retains body integrity.

Natural locomotion, however, often relies on more than lubrication alone. In earthworms, for example, successful movement through soil depends on the synergy of body contraction and mucus-assisted lubrication, which allows the animal to reduce friction while simultaneously changing its shape to pass through confined pores [127–129]. Inspired by this richer biological strategy, Gao et al. [130] later extended magnetoresponsive lubrication into a magneto-thermal responsive semi-convertible hydrogel (MTSCH). Here, the biological inspiration is not merely earthworms are slippery, but that earthworms coordinate muscular contraction with living lubrication to traverse narrow and underground environments (Figure 10. (b)). To translate this into a synthetic system, the authors introduced a magneto-thermal gelatin/Fe_3O_4-NH_2 supramolecular network into a thermoresponsive PNIPAm covalent network and further constructed a macroporous architecture by ice templating. Under an alternating magnetic field, Fe_3O_4-NH_2 rapidly generated heat, which simultaneously triggered gelatin gel–sol transition to produce a lubricating sol layer and PNIPAm contraction to

reduce volume. In this way, magnetic stimulation no longer served only to create lubrication, but also to coordinate lubrication with active contraction, much closer to living locomotion.

The resulting adaptive behavior was significantly more integrated than in the earlier static-field system. Under AMF-On, the optimized MTSCH showed reversible volume contraction of about 65%, while its COF decreased from 0.449 ± 0.054 to 0.176 ± 0.062. The lubricating sol layer appeared rapidly, becoming visible within 40 s and reaching a thickness of about 2.03 ± 0.30 mm after 120 s, while the hydrogel simultaneously contracted. Because lubrication and contraction were triggered together, the material could slide down inclined surfaces, escape confined cages in water, and even move out of a simulated underground environment, whereas the nonresponsive comparison systems could not. This design provides a bioinspired example in which lubrication and contraction are coupled to support motion in confined environments.

Magnetic actuation has also been used to regulate hydrogel deformation and frictional locomotion in confined environments. Shen et al. [137] developed PNIPAAm/Fe_3O_4 hydrogel mag-bots that reversibly contracted and elongated under an alternating magnetic field and moved through confined spaces by interacting with asymmetric micro-patterned surfaces. It highlights an alternative role of magnetic fields in regulating hydrogel deformation, contact, and frictional interaction. Such studies may inspire future magnetoresponsive lubricating hydrogels that combine remote actuation with controlled interfacial contact.

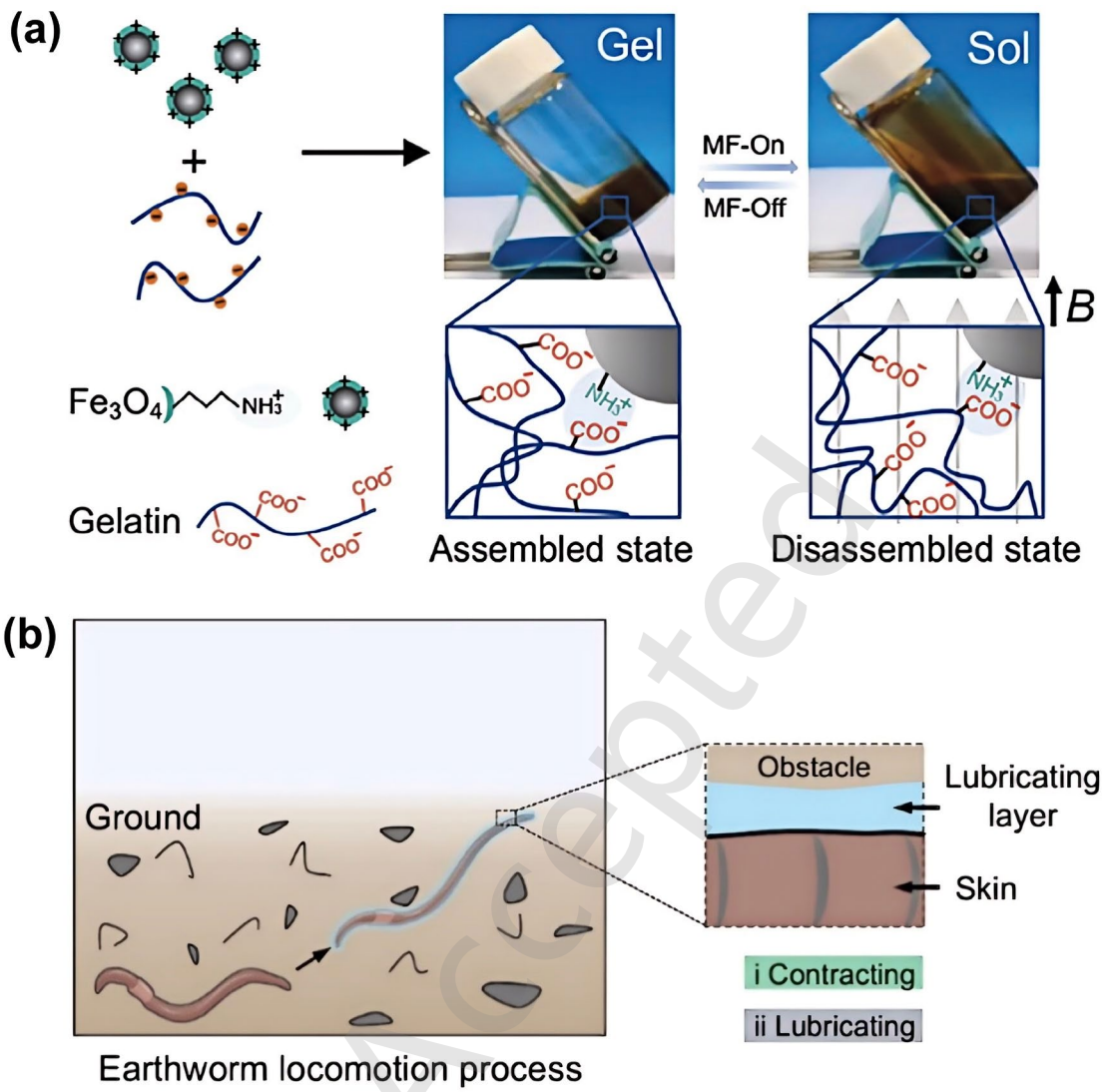

Figure 10. Schematic diagram of the mechanism of bioinspired magnetoresponsive lubricating hydrogel. (a) Scheme of the synthesis of magnetic supramolecular hydrogel composed of positive Fe_3O_4-NH_2 and negative gelatin, photographs of the gel–sol transition process under magnetic field (300 mT). Reprinted with permission from Gao et al. [126] Copyright© 2024 Wiley-VCH GmbH. (b) Inspired by the movement process of earthworms in soil, the response mechanism of bioinspired magnetothermal lubricating hydrogels under alternating magnetic fields. Reprinted with permission from Gao et al. [130] Copyright© 2026 American Chemical Society.

Viewed in this way, bioinspired magnetoresponsive lubrication evolves from the natural principle that biological interfaces can remain structurally integrated while locally presenting a liquid lubricating phase. Current magnetoresponsive lubricating hydrogels mainly follow two related routes. One route uses magnetic-field-induced interfacial

gel–sol transition to generate a lubricating surface layer, while another couples magnetothermal actuation with lubrication and reversible deformation for motion in confined environments.

4.7 Bioinspired Multi-Responsive Adaptive Lubrication

In living systems, lubrication is often regulated not by a single cue, but by the cooperation of multiple environmental signals. Multiple external stimuli may act together to produce the same biological effect, such as mucus secretion, interfacial phase transition, or lubricant renewal. This provides an important bioinspired principle for adaptive hydrogels, namely that rather than responding to isolated stimuli independently, multi-responsive systems can be designed so that different cues act on a shared dynamic lubricating phase, thereby enabling broader environmental adaptability and more robust interfacial regulation.

A representative example is the hagfish-inspired dual-responsive hydrogel reported by Deng et al. [25], in nature, hagfish rapidly secrete mucus when exposed to external disturbance, thereby reducing friction and facilitating escape (Figure 11. (a)). Inspired by this secretion-based adaptive lubrication, the authors developed a thermal/shear-responsive hydrogel (PPRA) by integrating a stimuli-responsive RPAD supramolecular network into a stable P(AAm-co-AAc)/PVA framework (Figure 11. (b)). In this design, the covalent polymer network preserves the structural integrity of the gel, whereas the RPAD supramolecular component acts as the adaptive lubricating phase. Upon heating or shearing, weak noncovalent interactions within RPAD are disrupted, inducing a partial gel–sol transition and causing a sol-state supramolecular layer to appear on the

hydrogel surface. This surface layer then serves as a lubricating film. Correspondingly, for PPRA-3, the COF decreased from 0.087 ± 0.003 to 0.029 ± 0.002 after repeated sliding at about 33 °C, while at 50 °C it dropped more rapidly and stabilized at 0.009 ± 0.001. Importantly, the same responsive transition also increased the fracture strength of the hydrogel from 1.23 ± 0.07 MPa in the original state to 1.92 ± 0.13 MPa after shearing and 2.1 ± 0.14 MPa after heating, indicating that the dual-responsive process did not simply lower friction, but reorganized the internal supramolecular network in a way that simultaneously benefited lubrication and mechanical performance.

A second example extends this multi-responsive logic toward more extreme operating conditions. Wang et al. [131] developed a self-secreting organohydrogel (OHG) inspired by the aquatic organisms, where lubrication is maintained even in challenging xeric or cryogenic environments. In their design, supramolecular organogel microspheres were dispersed within a PDMA hydrogel matrix, so that the hydrogel body provided flexibility and load support while the internal organogel served as a secretable lubricating phase. Because the organogel exhibits both thermoreversible sol–gel transition and shear-thixotropic behavior, temperature and shear can both trigger oil-phase secretion at the hydrogel surface. As a result, the system no longer relies solely on conventional hydration lubrication. Under dry conditions, the coefficient of friction decreased from about 0.4 at 25 °C to about 0.14 at 75 °C, and under repeated shearing at room temperature it gradually fell to a similar low level as secretion accumulated at the interface. More importantly, the organohydrogel retained flexibility and maintained a stable coefficient of friction of about 0.1 even at −10 °C, and in aqueous environments

the combined effects of hydration lubrication and oil-phase secretion further reduced the COF to about 0.07. This work therefore shows that thermo-/shear-coupled adaptive lubrication can be used not only for switchable friction control, but also for extending hydrogel lubrication toward dry, wet, and subzero environments where purely hydration-dominated mechanisms may be less effective.

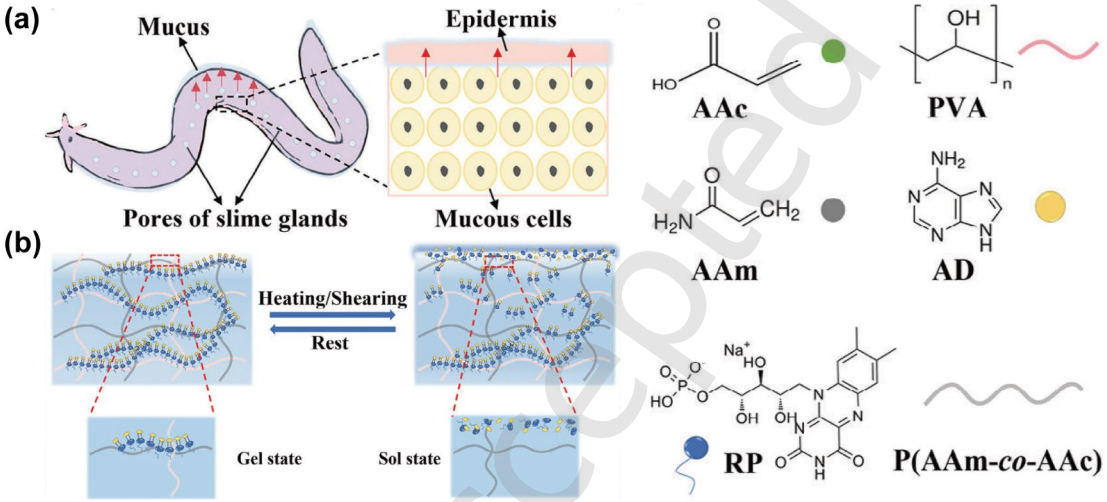

Figure 11. Schematic diagram of the mechanism of bioinspired multi-responsive lubricating hydrogel. (a) Schematic diagram illustrating the secretion of mucus of the hagfish. (b) The possible lubricating mechanism of the prepared thermal/shear-responsive lubricating hydrogel. Reprinted with permission from Deng et al. [25] Copyright© 2025 Wiley-VCH GmbH.

Overall, these studies show that multi-responsive adaptive lubrication can emerge when different external cues converge on the same dynamically reconfigurable lubricating phase. In the current thermo-/shear-coupled examples, temperature and shear do not act as separate regulators of friction; instead, they cooperatively trigger interfacial secretion, partial gel–sol transition, or lubricant-layer generation. In this sense, bioinspired multi-responsive hydrogels move adaptive lubrication beyond single-stimulus switching toward a more integrated mode of interfacial regulation, one that is closer to the way living systems coordinate multiple environmental signals to preserve

lubrication under complex and changing conditions. At last, Table 1 summarizes the representative stimulus-responsive hydrogels discussed in this section from the perspectives of external stimulus, bioinspired types, COFs, and response time scales. This table highlights how different external stimuli are transformed into various lubrication regulation modes, which are reflected in the changes of the COFs.

Table 1. Comparative summary of representative bioinspired adaptive lubricating hydrogels. NM = Not mentioned.

Stimulus type	Bioinspired types	Dominant lubrication mechanism	COFs/Reduction	Response time	Operating conditions	Durability / reversibility	Ref.
Mechanical	Cartilage	Shear-induced gel-sol transition plus self-healing H-bonding and π-πreassembly	~0.68 (~87% reduction)	~6 h	Parallel-plate rheometer (R = 25 mm); load 10 mN; shear velocity 0.1 mm/s. Shearing/wear tests: 50 kPa/80 mm/s and 5 N/80 mm/s.	Lubricating function can recover under dynamic shear	[95]
Mechanical	Synovium	Shear-induced HA network rearrangement exposes liposome reservoirs and renews lipid boundary layers	~0.031 (~50% reduction)	NM	PE sphere (d = 8 mm) vs stainless-steel disk; linear reciprocating; load 1 N; frequency 1 Hz; amplitude 4 mm; max contact pressure 25.68 MPa.	Shear-responsive boundary-layer renewal	[33]
Mechanical	Earthworm	Gland-like pores store lubricant; pore opening/closing pumps lubricant during sliding	~0.0079 at 11.32 MPa; ~0.0028 at 8.48 MPa; NM	NM	Load 10-80 N; frequency 1 Hz; contact pressure up to 8.48/11.32 MPa.	~100,000 cycles at 8.48 MPa	[82]
Thermal	Catfish	Thermal phase separation stiffens bottom layer and reduces contact deformation	~0.37 to ~0.027 (~93% reduction)	~1 s	Sphere-to-disk: glass ball (d = 6 mm) vs hydrogel, load 1 N. Disk-to-disk: glass plate (10 x 10 x 1 mm) vs hydrogel, load 1 N. Load-dependence tests: 0.2-5 N.	Reversible	[83]
Thermal	Muscle	Temperature-induced aggregation/untangling chains changes mechanics and friction	~0.17 to ~0.02 (~88% reduction)	NM	Rheometer friction test; aluminum plate (D = 20 mm) vs hydrogel, Fn ≈ 5 N.	Reversible	[103]
Thermal	Tendon sheath	NIR photothermal-triggered gelatin disassembly generates sol layer	~0.007; NM	~60 s	Rotational tribometer; load/contact pressure 51 kPa; shear velocity 0.1 mm/s. Load range 25.5-126.5 kPa; velocity range 0.10-1.00 mm/s.	15,000 compression cycles; 100,000 shearing cycles	[104]
Thermal	Plant secretory tissue	Water-/thermal-driven medium exchange and deswelling trigger oil secretion	NM	NM	PDMS sphere and stainless-steel sphere (r = 3 mm) as contact pairs; load 0.5-2.0 N; sliding velocity 0.1-2.0 mm/s; stroke 30 mm.	NM	[132]
Photo / UV-visible	Dynamic liquid	UV/visible-light-controlled α-CD/PEG/AzoPB assembly/disassembly generates/removes sol layer	~0.0144 to ~0.0045 (~69% reduction)	NM	Ball-on-disk test	Reversible	[85]
Photo / Visible light	Ocular-lubrication	Visible-light-triggered Azo-C_2/β-CD competition releases PEG to form hydrated layer	~0.0098; ~25% reduction	~6 h	NM	NM	[134].

Stimulus type	Bioinspired types	Dominant lubrication mechanism	COFs/Reduction	Response time	Operating conditions	Durability / reversibility	Ref.
Photo / molecular switch	Light-controlled molecular	Light-induced polarity/hydrophilicity switching regulates friction	~0.085; ~74% reduction	NM	NM	Repeatable over three light cycles	[108]
pH	Biological environment	pH-controlled dynamic covalent PVA-borax disassembly forms lubricating sol layer	~0.041; ~41% reduction	~ 10 min	Load 0.20 N; frequency 0.1 s^{-1}. Load range 0.10-0.30 N; frequency range 0.025-0.400 s^{-1}.	Reversible	[114]
pH	NM	pH-regulated sol-gel state	~0.01; ~0.13 to ~0.04 under varied load (~69% reduction)	NM	Reciprocating friction test; load 10 N; frequency 10 Hz; stroke 2 mm. Load range 1-15 N; frequency range 5-20 Hz.	Stable low friction for 3 h	[115]
Electrical	Mucosal mucus	Electric-field-induced partial gel-sol transition near cathode generates liquid lubricating layer	0.062 (~46% reduction)	~ 30 s	NM	Reversible	[84]
Electrical	NM	Electric bias drives electroosmotic extraction of salt-rich interfacial layer	~1-2 to ~0.03 under electric bias (>97% reduction; >50-fold modulation)	NM	Metal ball (d = 2 mm) vs hydrogel; normal load 20 mN; sliding velocity 2 mm/s.	Reversible	[120]
Electrical	Cell membrane	Transmembrane electric field induces reversible electroporation/bridging and changes hydrated slip plane	NM; ~200-fold reduction	NM	NM	Reversible	[119]
Magnetic	Maggot	Magnetic-field-induced contraction/elongation	NM	NM	NM	Reversible	[137]
Magnetic	Mucus/ synovia-like interfaces	Magnetic-field-induced gelatin/Fe_3O_4 network disassembly forms sol layer	~0.022; ~69% reduction	~15 min	Rheometer/friction test; pressure 10.0 kPa; velocity 0.1 mm/s.	Reversible	[126]
Magneto-thermal	Earthworm locomotion	Alternating magnetic field produces heating; gelatin gel-sol transition forms sol layer	~0.176; ~61% reduction	~2 min	Rheometer friction test; pressure 4.0 kPa; shearing velocity 0.1 mm/s.	Reversible	[130]
Thermal / shear	Hagfish	Thermal or shear stimulus disrupts π-π/H-bonded supramolecular network and forms surface sol layer	~0.029; ~67% reduction	~10 min	Ball-on-disk reciprocating tribometer; steel ball (d = 10 mm) as friction pair	Reversible	[25]
Thermal / shear	Aquatic self-secreting	Thermal or shear-induced secretion of oil-rich lubricating phase	~0.4 to ~0.1 in dry state (~75% reduction); ~0.07 in water (~65% reduction)	NM	PDMS sphere and stainless-steel sphere (r = 3 mm) as contact pairs; load 0.5-2.0 N; sliding velocity 0.1-2.0 mm/s; stroke 30 mm.	NM	[131]
Thermal / pH	Fish skin	pH- and temperature-controlled swelling/deswelling and chain conformation change	~0.5; ~88.1% reduction	NM	Face-to-face hydrogel/hydrogel contact; load/stress 1 N; sliding speed 200 mm/min.	Reversible	[101]

5. Conclusions

This review has outlined the evolution of bioinspired hydrogel lubrication from conventional bioinspired to adaptive bioinspired lubrication. Conventional bioinspired hydrogels have successfully reproduced many key structural and molecular principles of natural lubricating systems, including structural organization for contact regulation and hydrated boundary-layer design for maintaining low-friction interfaces. These designs have firmly established hydrogels as effective platforms for achieving low-

friction and wear-resistant interfaces under predefined conditions. At the same time, however, their lubricating states are in most cases largely preconfigured upon fabrication and may become less effective when the tribological environment changes. Against this background, BALHs represent not a replacement for conventional bioinspired hydrogels, but an important functional extension of them. Their key significance lies in moving hydrogel lubrication closer to the operating mode of living interfaces, where lubrication is not a fixed low-friction state, but a renewable, reconfigurable, and stimulus-coupled interfacial process. Across the systems discussed in this review, a common design principle emerges, namely that adaptive lubrication becomes possible when dynamic molecular motifs, reconfigurable interfaces, or stimulus-coupled transport and phase transitions are integrated into biomimetic hydrogel systems, so that external or environmental cues can be translated into lubricant generation, boundary-layer renewal, modulus/contact-state adaptation, interfacial phase reconstruction, or coordinated deformation–lubrication behavior. In this sense, mechano-, thermo-, photo-, pH-, electro-, magneto-, and multi-responsive systems should not be viewed as isolated categories, but as different routes toward the same goal of maintaining or reconstructing a favorable lubricating interface under changing conditions.

This shift is important both scientifically and technologically. Scientifically, it redefines lubrication as a dynamic interfacial function rather than a passive material property. Technologically, it opens the way toward hydrogel systems capable of maintaining performance in fluctuating and resource-limited environments. In biomedical settings,

adaptive lubrication enables low-friction interfaces to be integrated with tissue protection, regeneration, anti-inflammatory therapy, antifouling, or drug delivery. In soft robotics and intelligent devices, it enables reversible switching between anchoring and sliding, transport and gripping, or deformation and lubrication. BALHs should thus be regarded not simply as responsive materials, but as a broader bioinspired framework for constructing self-adjusting, multifunctional, and environmentally adaptive tribological interfaces.

6. Challenges and Future Perspectives

Despite the rapid development of bioinspired adaptive lubricating hydrogels, several key challenges still need to be addressed before their practical translation. First, long-term durability remains a central issue, because these hydrogels must maintain low friction under repeated compression, shear, swelling, dehydration, and wear. Promising strategies include decoupling load bearing from lubrication through hierarchical architectures, in which a mechanically robust bulk network supports the load while a soft hydrated surface layer reduces shear, as well as introducing dynamic sacrificial bonds, reversible crosslinks, or self-healing motifs to dissipate energy and recover damage during cyclic sliding. Second, the stability of lubricating boundary layers is equally critical. Hydrated polymer chains, zwitterionic groups, polymer brushes, lipid layers, and sol-like interfacial layers can effectively reduce friction, but they may be depleted, desorbed, diluted, or worn away during long-term sliding. Therefore, future designs should either strengthen boundary-layer anchoring through covalent grafting, interpenetrated surface layers, or strongly hydrated charged groups, or construct

renewable boundary layers using internal lubricant reservoirs, liposome-containing hydrogels, lubricant-release systems, semi-convertible hydrogels, or self-secreting organohydrogels. In this context, balancing boundary-layer regeneration with lubricant retention, leakage control, and long-term biocompatibility will be essential. Third, multi-stimulus interference becomes increasingly important for adaptive systems operating in complex environments. Temperature, pH, light, electric fields, magnetic fields, and shear may simultaneously affect swelling, modulus, ion transport, lubricant release, and interfacial reconstruction, leading to synergistic, antagonistic, or independent responses. To improve predictability, orthogonal response pathways should be designed by assigning different stimuli to distinct molecular motifs, network domains, or interfacial layers, while response maps under combined stimuli, such as temperature–pH, shear–pH, light–temperature, and electric field–ion concentration, should be established. Addressing these issues will be crucial for developing adaptive lubricating hydrogels with durable, stable, and controllable lubrication performance under application-relevant conditions.

Overall, the future of BALHs lies not simply in making hydrogels more lubricious, but in making them more intelligent, more context-aware, and more functionally integrated. The most transformative systems will likely be those that combine durable load-bearing frameworks, renewable lubricating interfaces, multi-stimulus responsiveness, and biologically or mechanically relevant feedback regulation. Such materials could ultimately define a new generation of tribological soft matter that no longer passively

tolerates environmental change, but actively adapts to it, much like the living lubricating systems that originally inspired this field.

Declaration of competing interest

The authors have no competing interests to declare that are relevant to the content of this article.

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