Fibers reinforced composite hydrogels with improved lubrication and load-bearing capacity

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Abstract: Hydrogels as one kind of soft materials with a typical three-dimensional (3D) hydrophilic network have been getting great attention in the field of biolubrication. However, traditional hydrogels commonly show poor tribology performance under high-load conditions because of their poor mechanical strength and toughness. Herein, pure chemical-crosslinking hydrogels mixed with different types of the micron-scale fibers can meet the requirements of strength and toughness for biolubrication materials, meanwhile the corresponding tribology performance improves significantly. In a typical case, three kinds of reinforcement matrix including needle-punched fibers, alginate fibers, and cottons are separately combined with Poly(*n*-vinyl pyrrolidone)-poly(2-hydroxyethyl methacrylate) (PVP-PHEMA) hydrogels to prepare fibers reinforced composite hydrogels. The experimental results show that the mechanical properties of fibers reinforced composite hydrogels improve greatly comparable with pure PVP-PHEMA hydrogels. Among three kinds of fibers reinforced composite hydrogel, the as-prepared composite hydrogels reinforced with needle-punched fibers possess the best strength, modulus, and anti-tearing properties. Friction tests indicate that the fibers reinforced composite hydrogels demonstrate stable water-lubrication performance comparable with pure PVP-PHEMA hydrogels. Besides, the hydrogel-spunlace fiber samples show the best load-bearing and anti-wear capacities. The improved tribology performance of the composite hydrogels is highly related to mechanical property and the interaction between the fibers and hydrogel network. Finally, spunlace fibers reinforced hydrogel materials with high load-bearing and low friction properties are expected to be used as novel biomimetic lubrication materials.

Keywords: cartilage-inspired; fiber-enhanced; composite hydrogel; high strength; good toughness; lubrication; load-bearing; wear-resistance

1 Introduction

As one kind of wet/soft polymer-based material with a typical three-dimensional (3D) network, hydrogels possess unique physical and chemical

properties, which have got great attention in many fields [1–3]. Meanwhile, the feature of low friction for hydrogels endows hydrogel-based materials with good application potential in the field of biolubrication [4–9]. As reported by Gong et al.[10],

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synthetic hydrogels showed a much lower friction coefficient as low as 10^{-3} than hard solid materials. Especially, it is found that the polymer network structure of hydrogels is similar to the extracellular matrix, which plays a crucial role in the friction behavior of cartilage [11–14]. Therefore, hydrogels become ideal materials for artificial cartilage and tissue substitutes. Over the past 20 years, the studies focusing on friction of hydrogels-based materials have achieved great progress. However, the most serious problem of traditional hydrogels is its poor mechanical behavior for high load-bearing. As a result, the development of strong and tough hydrogel materials is the focus of current research [15–18]. For example, researchers have developed a series of hydrogels with high strength and good toughness, such as dual-network (DN) hydrogels [19, 20], nano-composite hydrogels [21–23], and double-crosslinking hydrogels [24, 25]. Their excellent mechanical properties can be attributed to the effective dissipation of energy by sacrificial bonds introduced in the network during fracture.

Even though many strategies have been developed for improving the mechanical strength of hydrogels, the inherent increase of the strength will shrink the mesh pore size and reduce the degree of surface hydration, and thus leading to the deterioration of surface lubrication property. So, balancing the mechanical properties and hydration capability of the hydrogels becomes a key challenge. Our cartilage simultaneously shows low friction, high strength, and excellent toughness. As been extensively studied, cartilage is considered as a biphasic structure composed of a solid matrix phase and an interstitial fluid phase, in which the interstitial fluid can support most of the contact force to reduce the interface friction, whereas the solid matrix can withstand dynamic loads under shearing. Inspired by this, numerous composite hydrogels with excellent mechanical properties are prepared through combining the fibers and pure hydrogels together, in which the hydrogels can provide a considerable hydration state to reduce the interface friction while the fibers can improve the mechanical property. For example, Huang et al. [26, 27] prepared a series of fibers composite hydrogels. King et al. [28] put forward a new material design strategy, that the hydrogels of the woven fibers were mixed to prepare a new

kind of reinforced fibers composite with five times of the strength of steel. However, up to now, enormous efforts devoted to the composite hydrogels mainly focus on the mechanical property, and few cases are focusing on biolubrication. So, developing strong and resilient hydrogels with considerable tribology performance remains a challenge.

As one kind of potential biological material, PHEMA hydrogels are studied extensively because of its good biocompatibility [29, 30]. However, the mechanical strength of pure PHEMA hydrogels is commonly poor and cannot meet the requirements of the cartilage-liked lubrication materials. As mentioned above, the combination of micro-scale fibers and hydrogels can meet both high strength and biocompatibility. Therefore, we prepared fiber reinforced composite hydrogels with both good strength and toughness. In the typical case, micro-scale fibers (spunlace fibers, alginate fibers, and cottons) are *in situ* integrated into pure PVP-PHEMA hydrogel network by physical mixing upon encountering radical polymerization. The fibers are robust confined within the hydrogel network through hydrogen bonds. When the normal loads and tangential stress are applied, the fibers can dissipate energy through elongated deformation, thereby improving the mechanical properties of the hydrogels. As a result, the composite hydrogel materials show both high load-bearing and low friction properties, which are expected to be used as novel biomimetic lubrication materials.

2 Material and methods

2-hydroxyethyl methacrylate (HEMA) was purchased from J&K Chemical Ltd. 1-vinyl-2-pyrrolidone was purchased from Shanghai Macklin Biochemical Co., Ltd. Trimethylolpropane trimethacrylate (TMPTA) was purchased from Guangyi Chemical Co., Ltd. 2,2-azobis (isobutyronitrile) (AIBN) was purchased from Tianjin Damao Chemical Reagent Co., Ltd. AIBN was purified by reflux in ethanol. Other reagents and solvents were purchased and used without any purification. Deionized water was applied for all polymerization and treatment processes.

2.1 Preparation of PVP-PHEMA hydrogels

The monomer solution was prepared from a mixture

of HEMA with linear reinforcing polymer-*n*-vinyl pyrrolidone (PVP, 1,300,000 Mw), AIBN (0.1%, w/w) as initiator, and TMPTA (0.5%, w/w) as crosslinker. Then the PVP-PHEMA hydrogels were prepared by thermal polymerization of monomer solution at 60 °C for 4 h. Subsequently, the hydrogel blends were immersed into distilled water for 2 days to remove the residues.

2.2 Preparation of fibers reinforced composite hydrogels

Three kinds of woven fabrics made by cotton fibers, spunlace fibers, and alginate fibers were separately used to prepare fibers reinforced composite hydrogels. Firstly, the woven fabrics were immersed in the mixture of monomer solution to allow the complete wettability and liquid saturation for the fiber network. Then the liquid-saturated fabrics were taken out to allow the thermal polymerization for 4 h at 60 °C to obtain the fibers reinforced PVP-PHEMA composite hydrogels, as exemplified in the schematic representation. Subsequently, the composite hydrogels were immersed into distilled water for 2 days to remove the residues.

2.3 Friction test

The friction test during aqueous-lubricated sliding was carried on the conventional ball-on-disk reciprocating tribometer (CSM Co., Ltd., Switzerland) by acquiring friction coefficient (μ) at different loads and frequencies. The elastomeric poly(dimethylsiloxane) (PDMS) hemispheres and 314 stainless steel balls with a diameter of 6 mm were used as pins. The PDMS hemispheres were prepared by a commercial silicone elastomer kit (SYLGARD 184 silicone elastomer, base and curing agents, Dow Corning, Midland, MI) and the weight ratio of the base and curing agents is 10:1. A polystyrene cell culture plate with round-shaped well (Siqu Biotechnology Co., Ltd., Beijing) was used to prepare PDMS hemispheres. After removing the bubbles by vacuum degassing, the mixtures were put into the mold and then incubated in a 70 °C oven for 4 h. Each sample was measured at least three times at different positions to obtain the average value.

2.4 Mechanical property measurement

The mechanical property of the samples was measured by an electrical universal material testing machine with a 500 N load cell (EZ-Test, SHIMADZU). The test samples were cut into rectangular shape. All mechanical tests were carried out after the sample had reached the swelling equilibrium in water. In the tensile test, the samples were cut into 20 mm in length, 5 mm in width, and 2 mm in thickness. The crosshead velocity was kept at 50 mm/min in the tensile measurement. The elastic modulus in the tensile test was calculated from the slope 5%–15% of the strain ratio of the stress–strain curve. In the compression test, the samples were cut into 10 mm in length and width, 3 mm in thickness, respectively. The compression test was performed at a velocity of 5 mm/min with a strain limitation of 70%. The elastic modulus in compression test was calculated from the slope 5%–15% of the strain ratio of the stress–strain curve.

The trouser tearing test was performed in the same machine with a tensile velocity of 50 mm/min. The samples were firstly cut into a rectangle of 40 mm in length and 10 mm in width, respectively. Then the samples were cut into pants in the length direction, and the length of the forked and undivided portions both were 20 mm.

$$G_c = \frac{1}{t \times L_{\text{bulk}}} \int F d\Delta \quad (1)$$

where the G_c represents the energy required to tear. The thickness of the specimen and the length of the tear are indicated by t and L_{bulk} , respectively. F and Δ stand for load and displacement, respectively (The geometry and dimensions are shown in Ref. [31]).

2.5 Morphology characterization

The sample morphologies were characterized by field-emission scanning electron microscope (FESEM) (JSM-6701F, JEOL Inc., Japan) at an accelerating voltage of 5 kV. The test samples were prepared according to the standardized process. In a typical case, the test samples were frozen with liquid nitrogen or by an automatic refrigeration system and then were dried in the freeze-drying machine

for 24 h at a vacuum degree of 5 Pa (SCIEN TZ-10N, Ningbo Scienzt Biotechnology Co., Ltd.).

3 Results and discussion

3.1 Synthesis process of composite hydrogels

The fibers play an important role in the wear resistance of composite gels. According to the previous work, PVP-PHEMA gel was capable of achieving a low friction coefficient (~ 0.03) [32]. Upon integrating fibers into hydrogels, the composite hydrogels can both realize high load-bearing and low friction. Figure 1 shows the schematic illustration of the fabrication process for the composite hydrogels. First, the mixture containing monomers of HEMA and NVP, initiator of AIBN, and crosslinker of TMPTA was poured into the mold, and then the fiber sheet was added to allow it completely wetted. Finally, the samples were put into 60 °C oven for 4 h to perform radical polymerization. After polymerization, the samples were placed into the water to remove the residues. Correspondingly, the fibers penetrate and distribute throughout the entire hydrogel network, and the hydrogen bonds can be formed between

fiber surface and polymer chains of hydrogels. So, it can be speculated that the fibers can effectively improve the mechanical strength of hydrogels.

3.2 Morphology of bare fibers and fiber-enhanced composite hydrogels

In the hydrogel network, two different kinds of polymer chains (PHEMA and PVP) entangled each other to form a hydrophilic composite network with a large number of hydrogen bonds. As observed from the SEM images, the pristine spunlace fibers exhibit an irregular arrangement with a single fiber diameter of 10–15 μm (Fig. 2(a)), and the alginate fibers show a little regular arrangement with a single fiber diameter of 5–8 μm (Fig. 2(b)), while the cotton fibers show scattered arrangement with single fiber diameter of 15–20 μm (Fig. 2(c)). The formation of a highly porous 3D fiber network ensures the composite hydrogels show high strength and toughness in a different direction. The entire mechanical property of the composite hydrogels must be closely related to the microstructures of the hydrogel network. After freezing-drying cycles, the morphology of hydrogel samples was observed

Fig. 1 Schematic illustration showing the preparation process of the fiber-enhanced PVP-PHEMA composite hydrogels along with corresponding molecular structure and network structure, and existing hydrogen interaction between fiber surface and hydrogel network.

Fig. 2 (a) FESEM images of bare spunlace fibers, (b) alginate fibers, and (c) cotton fibers; network morphologies of (d) spunlace fiber composite hydrogels, (e) alginate fiber composite hydrogels, and (f) cotton fiber composite hydrogels.

by FESEM. The results show that all kinds of fibers were completely encapsulated within hydrogels. In the typical case, after integrating spunlace fibers, the as-prepared composite hydrogels can show a highly porous structure, and the fibers are obvious and irregularly interspersed across the entire samples (Fig. 2(d)). The alginate fiber composite hydrogels (Fig. 2(e)) and the cotton fiber composite gel (Fig. 2(f)) also exhibit similar network distribution structure, but the distribution of cotton fibers within the composite hydrogels is more dispersed than that of the other two fibers.

3.3 Mechanical property tests for bare fibers and fiber-enhanced composite hydrogels

Figure 3(a) shows the typical stress–strain curves of pure PVP-PHEMA hydrogels, three kinds of fibers, and fiber-enhanced composite hydrogels. It can be seen that the tensile stress of pure PVP-PHEMA hydrogels is only about 300 KPa at the strain of 4%, which indicates the pure hydrogels is so soft and brittle. The maximum tensile stresses of alginate fibers, cotton fibers, and spunlace fibers are separately 100 KPa, 1.5 MPa, and 3 MPa, and their corresponding strains are 49%, 55%, and 38%, respectively. Surprisingly, the stress and strain of the hydrogels reinforced

with three different kinds of fibers increase sharply. Taking spunlace fiber-enhanced composite hydrogels as an example, the tensile stress of the composite hydrogels can achieve to as high as 9.6 MPa, which is ~30 times higher than pure PVP-PHEMA hydrogels, and the corresponding fracture strain increases to 17%. This indicates that existing fibers can significantly enhance the network density of the hydrogels to endow its good mechanical property. The corresponding elastic modulus shows the same change trend as stress (Fig. 3(b)). The elastic modulus of the pure hydrogels and spunlace fibers are ~1.3 and ~3.95 MPa, respectively. The composite hydrogels reinforced with spunlace fibers can sharply increases to ~28 MPa. These data intuitively demonstrate the significant role of fibers within PVP-PHEMA hydrogels. As speculated, the formed compact network of composite hydrogels would effectively shackle the hydrogels and fibers, whereas the fibers break down to dissipate energy. It can be shown that the tensile stress of the spunlace fiber composite hydrogels is the most significant, followed by cotton fibers, and finally the alginate fibers (Fig. 3(a)).

Besides, the excellent mechanical properties of the composite hydrogels were further confirmed by

Fig. 3 (a) Tensile stress–strain curves of pure PVP-PHEMA hydrogels, three kinds of bare fibers, and corresponding fiber-enhanced composite hydrogels and (b) the calculated elastic modulus; (c) compression stress–strain curves of pure PVP-PHEMA hydrogels, three kinds of bare fiber and corresponding fibers-enhanced composite hydrogels, and (d) the calculated compressive modulus.

performing compression tests. In terms of spunlace fiber composite hydrogels, it can be compressed to 75% without fracture and its compression stress can reach ~12 MPa (Fig. 3(c)) and corresponding compressed modulus can reach 20 MPa (Fig. 3(d)). As expected, the excellent anti-compression capacity of composite hydrogels would provide a basic condition to realize high load-bearing under dynamic loading and shearing process. The compression property of the other two kinds of composite hydrogels shows the same trend as that of the spunlace fibers, but relying on the specific feature of each fiber.

3.4 Tearing test

The trouser-shape tearing test was used to further verify the toughening mechanism of the composite hydrogels [27]. As shown in Fig. 4(a), the upper end of the hydrogels was clamped to the platform and the other end was fixed to the fixture, and deformed under stress until it was completely torn. Figure 4(b) shows the real-time tear strength–

distance curves that are recorded in the tearing test. When the gel is torn along the stretching direction, it first reaches a critical force value than the gel tears following a “zig-zag” tensile stress and exhibits a very long tear distance (Fig. 4(c)). The tear resistance of composite hydrogels is significantly increased compared to pure hydrogel samples. When the spunlace fiber composite hydrogels are torn along the stretching direction, the tear force experiences a rapid increase and then ruptures very rapidly along with short tear distance. This means that the energy required to tear the composite hydrogels is much greater than the energy required for stretching. The maximum tear strength of the spunlace fiber composite hydrogels is 10.1 N/mm, which is about 25 times higher than pure PVP-PHEMA hydrogels (0.4 N/mm) and about 10 times higher than bare spunlace fibers (1 N/mm). The other two fiber composite hydrogels have lower tear strength than spunlace fiber composite hydrogels (cotton composite hydrogels: 6.8 N/mm; alginate

Fig. 4 (a) Photographs of the tearing test for fiber-enhanced composite hydrogels before (left) and (right) after undergoing a large strain deformation; (b) real-time tensile strength-distance curves in tearing tests for pure PVP-PHEMA hydrogels, three kinds of bare fibers and corresponding fibers composite hydrogels; (c) schematic of the tear test and (d) corresponding calculated fracture energy.

fiber composite hydrogels: 2.6 N/mm), but both improved obviously compared to bare fiber samples. What's more, the corresponding fracture energy is shown in Fig. 4(d), which is calculated from the force–distance curves. The fracture energy of the spunlace fiber composite hydrogels can be achieved as 17,000 J/m², which is much higher than the 500 J/m² of the pure hydrogels. These results indicate that the interaction of the hydrogels matrix with the fiber-reinforced phase promotes synergistic enhancement and toughness to endow the composite hydrogels with excellent mechanical properties. Meanwhile, similar to the tensile strength, the fracture energy of the composite hydrogels also increases as the fiber density of the fiber-reinforced phase increases.

Moreover, to understand the toughening mechanism

of the composite hydrogels, the interface morphology of the cracked sample was observed by SEM (Fig. 5(b)). It can be found that during the crack extension process, the fibers inside the hydrogels are gradually stretched, and then occur delayed fracture, which effectively dissipates the energy, so that the hydrogels exhibit excellent tear toughness. Furthermore, at the fracture tip, there is a large amount of intact fibers that can continue to function under a further stretching process until the samples are completely torn (Fig. 5(c)). It should be noted that all three kinds of fibers can form hydrogen bonds with the hydrogel network, but the interaction degree may be slightly different. Therefore, the hydrogen bonds between the fibers and hydrogels play a critical role in the enhancement and toughening of the hydrogels (Fig. 5(a)).

Fig. 5 (a) Schematic diagram of interaction mechanism based on hydrogen bonds between fibers and hydrogel phases; (b) SEM image of the locally interface amplification in tearing tests of spunlace fiber-enhanced PVP-PHEMA hydrogels; (c) the model of delayed-fracture induced energy dissipation mechanism under further stretching tear process.

3.5 Friction test of fiber-enhanced composite hydrogels

HEMA and NVP are widely used in the field of contact lenses due to their excellent biocompatibility and good lubricity [33–35]. Figure 6 shows the typical contact image obtained from the sliding process and the corresponding friction curves for pure PVP-PHEMA hydrogels. Friction tests were performed under a ball-on-disk contact style by employing a soft PDMS ball (Young's modulus: ~ 2.0 MPa), hard steel ball as sliding pairs, and the deionized water was used as a lubricant. The average friction coefficient was chosen for analysis, which is summarized in Fig. 6(a). It can be seen that the friction coefficient of pure PVA-PHEMA hydrogels against both sliding pairs increased with the normal loads, and the higher friction coefficient was observed when the steel ball was used as a contact counterpart. In the typical case, the friction coefficient is ~ 0.018 at the load of 0.2 N and it increased to ~ 0.03 at the load of 2 N against PDMS pair, and it is ~ 0.026 at the load of 0.2 N and increased to ~ 0.039 at the load of 2 N against steel ball pair, indicating the poor load-bearing property of the pure hydrogels. Correspondingly, the local interface contact process was observed. As shown

in Fig. 6(b), the PDMS ball completely penetrated the hydrogel sheet at the applied load of 3 N, resulting from poor mechanical property of the pure PVP-PHEMA hydrogels. The real-time friction coefficient curves were shown under different normal loads against PDMS (Fig. 6(c)) and steel ball (Fig. 6(d)) as sliding pairs. At the initial stage of friction, the sliding interface showed a stable low friction coefficient, but it appeared a rapid increase of the coefficient due to complete broken of hydrogels. This indicates that low mechanical strength of pure PVP-PHEMA hydrogels is not enough for high load-bearing.

The friction coefficients of the spunlace fiber composite hydrogels under different normal loads upon employing PDMS ball and steel ball as sliding pairs are shown in Fig. 7(a). The composite hydrogels exhibit low friction coefficients (< 0.06) under a wide range of load from 0.2 to 5 N both against two kinds of sliding pairs. Compared with pure hydrogels (Figs. 6(c) and 6(d)), the load-bearing capacity of the spunlace fiber composite hydrogels improves highly. In the typical case, the friction coefficient of the composite hydrogels can always maintain a low value of ~ 0.05 under a high load of 5 N in the whole 10,000 test cycles (Fig. 7(b)). Correspondingly, the real-time friction coefficient

Fig. 6 (a) Change of friction coefficient values with normal loads upon employing PDMS ball and steel ball for pure PVP-PHEMA hydrogels; (b) the photograph of the local contact of pure PVP-PHEMA hydrogels against PDMS ball under normal load of 3 N; the real-time friction coefficient curves of pure PVP-PHEMA hydrogels against (c) PDMS ball and (d) steel ball with sliding time under normal loads of 3 and 5 N (velocity: 0.01 m/s).

curves of spunlace fiber composite hydrogels against (Fig. 7(c)) PDMS ball and (Fig. 7(d)) steel ball were also provided. Compared with soft PDMS ball, the friction coefficient curves of the steel ball case generated slight fluctuations with the typical stick-slip phenomenon. The reason responsible for this can be attributed to that hard steel pair would generate a higher contact stress to induce larger elastic deformation of hydrogel network under the same normal load, causing a much larger friction coefficient than that of using PDMS pair. Besides, to evaluate the wear-resistance capability of the composite hydrogels, the surface wear morphology of spunlace fiber composite hydrogels was also observed after encountering 10,000 reciprocating sliding cycles. As shown in Fig. 7(e), the surface of spunlace fiber composite hydrogels only appeared slight wear marks without generating obvious peeling, indicating its excellent load-bearing and anti-resistance capacities. As speculated, the spunlace fibers are tightly distributed within the hydrogels network based on the hydrogen bonding interaction, which

would significantly improve the inherent robustness of the hydrogel network. Combining with the analysis for friction performance, these above results indicate that the as-prepared spunlace fiber composite hydrogels possess good lubrication, load-bearing, and anti-resistance properties.

Furthermore, the tribological properties of alginate fibers and cotton composite hydrogels were also investigated under different normal loads based on the same test condition. As shown in Fig. 8(a), the friction coefficients of the alginate fiber composite hydrogels are both significantly higher (~ 0.135) than that of the spunlace fiber composite hydrogels when the PDMS ball and steel ball were used as the friction pairs (Fig. 7(a)). This indicates that the alginate fiber composite hydrogels would have poor wear-resistance property under high load condition, and its surface would rapidly reach the heavy-wear stage. This can be attributed to the fact that alginate fibers are too thin to perform a good load-bearing function (Fig. 2(b)). As shown in Fig. 8(b), the friction coefficient of the cotton

Fig. 7 (a) Comparison of friction coefficient obtained in both contact counterpart on spunlace fiber composite hydrogels at different normal loads; (b) the long-time friction coefficient curve with sliding time under the load of 5 N at the sliding velocity of 0.01 m/s upon employing PDMS as friction pair; corresponding friction coefficient curves with sliding time under different loads when (c) PDMS and (d) steel ball were used as friction pairs; (e, f) the optical microscope images of the spunlace fiber composite hydrogels surface after encountering 10,000 friction tests cycles.

composite hydrogels is ~ 0.115 at the load of 0.2 N against PDMS pair and it increased to ~ 0.15 against the steel ball pair. This can be attributed to the loose fiber structure distribution inside the cotton (Fig. 2(c)), resulting in relatively rough surface after composition with the hydrogels. So, among these three composite hydrogels, the spunlace fiber composite hydrogels exhibit better tribological property than that of the other two composite hydrogels, which is highly related to its

toughness and strength of fibers. However, for cotton fiber composite hydrogels, the fiber network is so dispersed along with poor toughness and strength to endure larger loads. As a result, the cotton composite hydrogels show relatively poor load-bearing and wear-resistance properties. By contrast, alginate fiber composite hydrogel shows moderate fiber distribution density, so its tribological property is between the spunlace fiber composite hydrogels and cotton fiber composite

Fig. 8 Change of friction coefficient values with normal loads upon employing PDMS ball and steel ball as friction pairs for (a) alginate fiber composite hydrogels and (b) cotton composite hydrogels; typical friction coefficient curves with sliding time under different loads for alginate fiber composite hydrogels when (c) PDMS and (d) steel ball were used as friction pairs; typical friction coefficient curves with sliding time under different loads for cotton composite hydrogels when (e) PDMS and (f) steel ball were used as friction pairs.

hydrogels. In short, the combination of fibers and PVP-PHEMA hydrogels can satisfy the fundamental requirements of cartilage-like high strength and load-bearing.

4 Conclusions

Inspired by the structural feature of the articular cartilage network, several kinds of composite hydrogel materials are prepared by integrating fibers into

hydrogel networks based on the hydrogen bond interactions. Compared with pure hydrogels samples, the fiber-enhanced composite hydrogels exhibit excellent mechanical properties. When the fiber-enhanced composite hydrogels are performed with mechanical shearing and tearing, the fibers confined into the hydrogel network can dissipate energy by elongating the fracture, and thus highly enhancing the strength and toughness of the hydrogels. As a result, the wear-resistance and load-bearing properties

of the composite hydrogels improve obviously. Taking the spunlace fiber-enhanced composite hydrogels as an example, it still shows stable low-friction, excellent load-bearing, and wear-resistance after 10,000 cycles of sliding test using PDMS ball as friction pair. Meanwhile, it is found that the wear resistance of the as-prepared composite hydrogels is highly related to the properties of the fibers themselves. Overall, the combination of fibers and PVP-PHEMA hydrogels can successfully prepare the novel fiber-enhanced composite hydrogel materials with considerable mechanical strength and toughness. The composite hydrogels exhibit improved lubrication, load-bearing, and anti-wear properties in comparison with the traditional hydrogel system, and show considerable potential in the field of biomimetic materials.

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