Lubricant self-replenishing slippery surface with prolonged service life for fog harvesting

Yi CHEN^{1,2}, Weimin LIU^{1,2,*}, Jinxia HUANG^{1,2,*}, Zhiguang GUO^{1,3,*}

¹ State Key Laboratory of Solid Lubrication, Lanzhou Institute of Chemical Physics, Chinese Academy of Sciences, Lanzhou 730000, China

² University of Chinese Academy of Sciences, Beijing 100049, China

³ Ministry of Education Key Laboratory for the Green Preparation and Application of Functional Materials, Hubei University, Wuhan 430062, China

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Abstract: Slippery lubricant-infused surfaces exhibit excellent fog-harvesting capacities compared with superhydrophobic and superhydrophilic surfaces. However, lubricant depletion is typically unavoidable under dynamic conditions, and reinfused oil is generally needed to recover the fog-harvesting capacity. Herein, an effective strategy for delaying the depletion of lubricant to prolong the service life of fog harvesting is proposed. An ultrathin transparent lubricant self-replenishing slippery surface was fabricated via facile one-step solvent evaporation polymerization. The gel film of the lubricant self-replenishing slippery surface, which was embedded with oil microdroplets, was attached to glass slides via the phase separation and evaporation of tetrahydrofuran. The gel film GFs-150 (with oil content 150 wt% of aminopropyl-terminated polydimethyl siloxane (PDMS-NH₂)) exhibited superior slippery and fog-harvesting performance to other gel films. Furthermore, the slippery surfaces with the trait of oil secretion triggered by mechanical stress exhibited better fog-harvesting capabilities and longer service life than surfaces without the function of lubricant self-replenishment. The lubricant self-replenishing, ultrathin, and transparent slippery surfaces reported herein have considerable potential for applications involving narrow spaces, visualization, long service life, etc.

Keywords: prolonged service life; lubricant self-replenishing; slippery liquid-infused surfaces; fog harvesting

1 Introduction

Surfaces with special wettability are of considerable interest in various fields, including liquid transport [1–5], oil/water separation [5–7], self-cleaning [7–10], and water collection [11–15]. In these fields, harvesting water from fog is an effective solution for the shortage of fresh water, which has drawn wide attention [16–20]. Fog capture, water condensation, and water removal are generally recognized as the three stages of fog harvesting [21–24]. The wettability of artificial surfaces, including hydrophobic surfaces (HBSs), hydrophilic surfaces (HHSs), and slippery liquid-infused porous surfaces (SLIPs), can be controlled by adjusting the

surface morphology and surface chemical composition [25, 26]. Surfaces with differences in wettability differ significantly in the processes of fog capture, water condensation, and water removal, leading to differences in their fog-harvesting capabilities.

Generally, HBSs are endowed with a low surface energy, which is significantly different from that of water droplets with a high surface tension. This results in little droplet nucleation on the HBSs, as well as a low efficiency of fog capture [27]. With fewer droplets on the HBSs, coalescence of the droplets is more difficult. Meanwhile, microdroplets tend to form a wrapping layer at the water–air interfaces on HBSs, hindering droplet coalescence and further nucleation

* Corresponding authors: Weimin LIU, E-mail: wmliu@licp.cas.cn; Jinxia HUANG, E-mail: huangjx@licp.cas.cn; Zhiguang GUO, E-mail: zgao@licp.cas.cn

[28–30]. Furthermore, the condensed water droplets are easily pinned on the HBSs because the wetting changes from the Cassie–Baxter state to the Wenzel state [23, 31–33]. Because of these traits, HBSs have poor performance in all three stages of fog harvesting. Owing to their affinity to water droplets, HHSs generally have a higher droplet nucleation density than HBSs, and the tightly nucleated droplets are more likely to coalesce [34]. Despite the better performance of HHSs for fog capture and water condensation, it is difficult to remove water droplets from the surface, owing to the formation of water films. Lubricant-infused slippery surfaces have advantages for all three stages of fog harvesting [29, 31, 35, 36]. They have a high nucleation density of water droplets in the fog-capture process, which shortens the distance between water droplets and promotes the condensation of water droplets. The slippery property not only promotes the further condensation of water droplets but also allows the water droplets to slide off the surface easily [29, 30, 37]. Hence, SLIPs have a higher fog-harvesting efficiency than HBSs and HHSs. However, lubricant depletion is a common problem for SLIPs, which can be due to gravitational drainage [38, 39], evaporation [40–42], high-speed drop impact [43, 44], growth of frost or ice [44–46], formation of wetting ridges and cloaking layers [47], and shear flow impact [48, 49]. Although some of the causes of lubricant depletion can be circumvented, such as gravitational drainage (by using appropriate substrates with micro/nanostructures) [50, 51] and evaporation (by replacing lubricants with nonvolatile oils, such as ionic liquids) [41], lubricant depletion is difficult to avoid. Therefore, a surface with a continuous oil supply can effectively overcome this situation.

In nature, numerous living organisms have tissues that can secrete fluids to mediate defenses, wound healing, etc., for survival [52, 53]. For instance, earthworms continually secrete mucus under external stimuli to reduce body surface damage when they wriggle in the soil [53]. Inspired by earthworms, Zhao et al. [54] fabricated rough polymer coatings with self-replenishing lubrication, which had an excellent friction-reducing effect after 200 stimulation cycles. These films exhibit superior longevity under solid-based friction than films without a self-replenishing function.

Li et al. [21] reported a caterpillar-inspired fiber trichome with a self-replenishing lubricating layer. The lubricant can be replenished by mechanical stress stimulation after consumption.

Herein, a lubricant self-replenishing slippery surface is proposed for extending the service life of slippery surfaces and improving the efficiency of fog harvesting. The lubricant self-replenishing function is realized via a facile solvent evaporation polymerization method, in which oil droplets are embedded in a gel film after polymerization. The gel film with an oil content of 150% exhibited superior slippery and fog-harvesting performance to other gel films. Furthermore, slippery surfaces with the trait of oil secretion triggered by mechanical stress exhibited better fog-harvesting capabilities and longer service life than the surfaces without the function of self-replenishment. Thus, lubricant self-replenishing slippery surfaces are useful for applications involving narrow spaces, visualization, long service lives, etc.

2 Experimental

2.1 Materials

Glass slides were purchased from TaiZhou KangWei Medical Technology Co., Ltd., China. Bis aminopropyl-terminated polydimethyl siloxane (PDMS-NH₂, Mn 1000) was purchased from Macklin Biochemistry Co., Ltd., Shanghai, China. and Toluene 2,4-diisocyanate (TDI, 98%) was supplied by Sanyou International (Shanghai) Chemical Co., Ltd., China. Tetrahydrofuran (THF, ≥ 99.0%) was obtained from ChengDu Chron Chemicals Co., Ltd., China. Dimethyl silicone oil (viscosity of 100 cSt at 25 °C) was obtained from Sinopharm Chemical Reagent Co., Ltd., China. All the reagents employed in the present study were of analytical grade and used as received. Deionized (DI) water purified using a ModuPure system was used throughout the study.

2.2 Fabrication of oil-embedded gel films

The lubricant self-replenishing slippery surfaces were fabricated via facile one-step polymerization based on a previous method [55]. The glass slides were cut into 2.5 cm × 2.5 cm pieces, which were ultrasonically

cleaned in 0.1 M HCl and absolute ethanol (for 1 h each). The cut glass slides were dried in an oven at 60 °C for 10 min for further use. The prepolymers were prepared via a facile method; TDI (0.0871 g, 0.5 mmol) was added to PDMS-NH₂ (0.5 g, 0.5 mmol) in THF (2 g), followed by magnetic stirring for 5 min to obtain a uniform dispersion. Subsequently, different amounts of silicone oil (0, 80, 100, 120, 150, 180, and 200 wt% PDMS-NH₂) were added to the prepolymers with another 5 min of magnetic stirring to obtain a homogeneous prepolymer solution. The prepolymer solution (300 µL) was coated onto the glass slide using a spin-coating instrument at a shear rate of 500 rpm for 10 s to evenly spread the prepolymer solution, followed by storage in an ambient environment (temperature of 20 °C and relative humidity of 10%) out of direct sunlight for 24 h to obtain gel films adhered to the glass slide (GFs-0, GFs-80, GFs-100, GFs-120, GFs-150, GFs-180, and GFs-200). The gel films were formed by the evaporation of THF. Owing to the phase separation in the polymerization process, the oil microdroplets were embedded in the gel films, and then the silicone oil in the upper region of the gel films overlapped on the surface. The PDMS-NH₂ gel matrix approached the saturation point when the embedded silicone oil content was 150 wt% (Fig. S1 in the Electronic Supplementary Material (ESM)). Thus, samples GFs-80, GFs-100, GFs-120, and GFs-150 were used for the following characterization analysis.

2.3 Characterizations

After the oil layer was removed from the different gel films (20 mL of DI water was dripped onto the inclined sample surfaces dropwise, so that the water droplets removed the lubricating oil while sliding off the surface), the micromorphology and embedded oil droplets were examined using an optical microscope (Olympus BX43, USA). Three-dimensional (3D) profile images of the gel films (after the oil layer was removed and the gel films were pre-treated by Au sputtering) were obtained using a 3D noncontact profiler (MicroXAM-800, KLA-Tencor, USA). The statistical roughness values of different samples were obtained by analyzing a series of the west-east line of the 3D surface profile images using the software Apex 3D

Basic, USA. The thicknesses of the gel films were determined using the 3D noncontact profiler, and the gel films were scratched by the tips of a pen or a knife. The relative height was analyzed using the software Apex 3D Basic. The transparency of the glass and gel films was determined using an Agilent Cary 60 UV-Vis spectrophotometer within a light wavelength range of 200–800 nm. The wettability of the gel films was investigated by recording the static water contact angle (CA) and sliding angle (SA) with 10 µL of DI water using a JC2000D system (Zhong Chen Digital Equipment Co., Ltd., Shanghai, China) at room temperature. The lubricant self-replenishment was triggered by mechanical stress was performed by applying a pressure of 50 kPa and a weight of 31.25 N to a surface of $6.25 \times 10^{-4} \text{ m}^2$. The lubricant self-replenishing performance was tested by the cycle of removing the oil layer of the gel films with 20 mL of DI water dropwise and applying a pressure of 50 kPa for 10 min, which was terminated when the difference in SAs between the oil layer after being removed and replenished was less than 1°. The fog-harvesting capabilities of the gel films were evaluated using a laboratory-assembled system (composed of a humidifier, a specimen holder, and foam board packaging) in a relatively sealed environment at a temperature of 15 °C and humidity of 99%. The distance between the mist vent and the samples was 20 cm, and the flow rate and speed of the commercial humidifier were $0.5 \text{ g}\cdot\text{s}^{-1}$ and approximately $2.4 \text{ m}\cdot\text{s}^{-1}$, respectively. The organic content of the collected water was determined by measuring the chemical oxygen demand (COD) using the U.S. Environmental Protection Agency Method 8000 (HACH, DRB 200, USA). The wear resistances of the gel films were tested using a steel ball with a diameter 6 mm on an Anton Paar ball mill. The test conditions were as follows: temperature of 28 °C, load of 1 N, frequency of 3 Hz, and stroke of 5 mm. The test duration was 1,000 cycles. The wear volume was determined using a 3D non-contact profiler (MicroXAM-800, KLA-Tencor, USA). The wear rate was calculated as $w = v(PL)^{-1}$, where w , v , P , and L represent the wear rate ($\text{m}^3\cdot(\text{N}\cdot\text{m})^{-1}$), wear volume (m^3), external load (N), and scratch length (m), respectively. The temperature stability of the gel films was tested by keeping the lubricant

self-replenishing slippery surfaces at 0, 20, 60, 80, and 100 °C for 1 h, and the CAs and SAs of the gel films were measured in a timely manner. The pH stability of the gel films was tested by sliding 10 μL of water with different pH values on the gel films. The long-term stability of the gel films was tested by sliding 10 μL of water on the lubricant self-replenishing slippery surfaces with a storage time of 1–10 days in an ambient environment.

3 Results and discussion

3.1 Fabrication and characterizations of lubricant self-replenishing slippery surfaces

The gel films attached to the glass slides were fabricated via facile one-step solvent evaporation polymerization of PDMS–NH₂ and TDI. Equal molar ratios of PDMS and TDI, as well as different contents of silicone oil, were evenly dispersed in the THF solvent to obtain homogenous prepolymer solutions, which were then spin-coated onto the glass slides. With the evaporation of THF in an ambient environment, the gel films with embedded oil droplets were formed through a highly coordinated polymerization process, in which the amino groups (–NH₂) of PDMS–NH₂ and the isocyanate group (–N=C=O) of TDI could generate a copolymer

of urea and PDMS at a low temperature (room temperature). Figure 1 shows the formation process of the oil droplet-embedded lubricant self-replenishing slippery surfaces. During the formation of the gel films, phase separation between the gel matrix and silicone oil occurred, causing the oil droplets to be embedded in the gel films, and then the oil droplets in the upper region of the gel films overlapped on the surface with the evaporation of THF [55].

The micromorphology and discrete embedded oil droplets were examined for the gel films with different contents of silicone oil using an optical microscope and a 3D noncontact profiler, as shown in Fig. 2. The embedded oil droplets were microscale, and the number and volume of oil droplets increased as the amount of added oil increased (Fig. 2). Thus, a higher oil content corresponded to a larger volume and number of oil droplets produced during phase separation. Interestingly, the surfaces of the gel films without silicone oil were highly flat (Figs. 2(a)–2(e)), whereas the surfaces of gel films with embedded silicone oil were rough, with concave–convex structures (Figs. 2(f), 2(k), 2(p), and 2(u)). The statistical roughness values of different gel films were obtained by analyzing the 3D profile images of the gel films (Fig. S2 in the ESM). The statistical roughness of GFs-0 was the lowest among all the samples ($0.119 \pm 0.110 \mu\text{m}$), and

Fig. 1 Formation process of the oil droplet-embedded gel films of the lubricant self-replenishing slippery surfaces. (a) The prepolymer solution was coated onto the glass slide, and (b) the phase separation between the gel matrix and silicone oil occurred with the evaporation of THF, causing the oil droplets to be embedded in the gel films.

Fig. 2 Micromorphologies of (a–d) GFs-0, (f–i) GFs-80, (k–n) GFs-100, (p–s) GFs-120, and (u–x) GFs-150 observed using an optical microscope with different enlargement factors and 3D surface profile images of (e) GFs-0, (j) GFs-80, (o) GFs-100, (t) GFs-120, and (y) GFs-150 with dimensions of 1 mm × 1 mm.

the statistical roughness increased with the addition of silicone oil, which agreed well with the optical microscopy images. Furthermore, most of the oil droplets were dispersed in the sunken region of the gel films, and few were distributed in the bulge region, which was mainly due to the changed surface tension between the THF evaporated regions and unevaporated regions as the THF evaporated randomly [56–58]. The surface tension of the THF ($27 \text{ mN}\cdot\text{m}^{-1}$) was higher than that of the silicone oil ($21 \text{ mN}\cdot\text{m}^{-1}$) and PDMS- NH_2 . The surface tension decreases in the sunken region owing to the rapid evaporation of THF, which drives the movement of the silicone oil [56]. Additionally, the thicknesses of all the gel films were measured, as shown in Fig. S3 in the ESM, revealing that all the gel films fabricated in this study were $< 10 \text{ }\mu\text{m}$ thick. The compositions of the PDMS- NH_2 , TDI, and gel films were investigated via Fourier transform infrared (FT-IR) spectroscopy, as shown in Figs. 3 and S4 in the ESM. The peak at $2,270 \text{ cm}^{-1}$ was associated with the antisymmetric stretching vibration absorption of the $-\text{N}=\text{C}=\text{O}$ group (Fig. 3(b)), which was observed in the FT-IR spectrum of TDI but not in the spectra of the gel films. In Fig. 3(a), the peak

at $3,340 \text{ cm}^{-1}$ was ascribed to the stretching vibration absorption of the $-\text{N}-\text{H}-$ group, which appeared in the FT-IR spectra of the gel films but not that of PDMS- NH_2 . The peaks at $1,638$ and $1,080 \text{ cm}^{-1}$ correspond to the urea group and the $-\text{C}-\text{N}-$ group in PDMS- NH_2 , respectively (Fig. 3(c)). The peak of the urea group was observed in the spectra of the gel films but not those of PDMS- NH_2 and TDI. The intensity of the peak of the $-\text{C}-\text{N}-$ group for the gel films was lower than that for PDMS- NH_2 . These results indicated that the $-\text{NH}_2$ groups in PDMS- NH_2 , as well as the $\text{N}=\text{C}=\text{O}$ group in TDI, were successfully polymerized to the urea group. Moreover, the paper-thin gel films were almost as transparent as glass, ensuring that the gel films do not affect the transparency of the substrate so as to achieve the purpose of negligible in the process of use (Fig. S5 in the ESM).

3.2 Lubricant self-replenishing mechanism and performance of slippery surfaces

The mechanism of lubricant self-replenishment was analyzed to further understand the self-replenishing process. Figure 4 shows a schematic of the lubricant

Fig. 3 FT-IR spectra of (a) the -N-H- group, (b) -N=C=O group, and (c) -HN-CO-NH- and -C-N- groups.

Fig. 4 Schematic of the lubricant self-replenishing mechanism. (a) Homogeneous prepolymer solutions. (b) Gel films formed on the glass after the evaporation of THF and the polymerization of PDMS- NH_2 . Meanwhile, the oil microdroplets were embedded in the gel films, and part of the oil droplets moved to the surfaces as THF evaporated. (c) Gel films after the oil layer were removed via the dropwise addition of 20 mL of DI water. (d) Oil droplets were excreted from the gel films after a pressure of 50 kPa was applied. (e) Optical images of the weighing papers imprinted with the trace of the gel films after the films were rinsed with DI water (left) and loaded with a pressure of 50 kPa (right).

self-replenishing mechanism. There were three stages in the process of lubricant self-replenishment: solvent evaporation and gelation, oil removal, and self-replenishment. In the first stage, the amino groups (-NH_2) of PDMS- NH_2 and the isocyanate group (-N=C=O) of TDI were crosslinked with the continued evaporation of THF from the homogeneous prepolymer solutions (Fig. 4(a)). The silicone oil nucleated inside the prepolymer solution and grew by combining with vicinal nucleated oil droplets during the gelation process of PDMS- NH_2 . Meanwhile, phase separation

of the gel matrix and oil droplets occurred, and some of the oil droplets moved to the surfaces of the gel films simultaneously to form a thin oil layer as THF continuously evaporated. After the gelation of prepolymers and the completion of the THF evaporation, the newly nucleated and un-excreted oil droplets were trapped inside the gel films (Fig. 4(b)) [54–56]. The oil removal process was then executed to further investigate the self-replenishing properties. The oil layer on the surfaces of the gel films after gelation was removed via dropwise addition of 20 mL of DI

water (Fig. 4(c)). An optical image of the weighing papers imprinted with the trace of the gel films after being rinsed with DI water is presented in Fig. 4(e) left, which shows that the oil was only distributed at the edges of the gel films after the films were rinsed in DI water. The self-replenishing property was tested by loading a pressure of 50 kPa. The hydrogen bonds of the urea units broke under the pressure, which allowed the trapped oil droplets to combine with vicinal nucleated oil droplets again; these droplets were excreted from the gel film surfaces under pressure, forming a new thin oil layer (Fig. 4(d)) [26, 54–56]. An optical image of the weighing papers imprinted with the trace of the gel films after being loaded at a pressure of 50 kPa is presented in Fig. 4(e) right. As shown, the oil layer was replenished on the gel films after the pressure of 50 kPa was applied. In summary, the oil layer self-replenishment of the gel films was triggered by mechanical stress, and the service life of the gel films can be prolonged in this manner.

The wettability differences of the gel films were analyzed by investigating the CAs and SAs (Fig. 5(a)). The contents of the original oil (OOC), first surface oil (SOC), and residual oil (ROC), and the thickness of the first surface oil for the gel films were calculated. As shown in Fig. S1 in the ESM, the OOC, SOC, and ROC increased with the oil content in the gel films. The CA of GFs-0 was $106.6^\circ \pm 3.4^\circ$, and the CAs of gel films with different silicone oil contents did not change significantly (fluctuated around 88°). The SAs of the gel films decreased slightly with the increasing silicone oil content, mainly owing to the thicker oil layer of GFs-150 after 24 h of preparation compared with GFs-80, GFs-100, and GFs-120 (Fig. S1(c) in the ESM). The lubricant self-replenishing properties of the slippery surfaces were further tested, and the cycles of oil-layer removal and replenishment were used to represent the relative service life of different gel films. In each cycle, the oil layer was removed by 20 mL of DI water dropwise and replenished by loading a pressure of 50 kPa (Fig. 5(b)). Lubricant self-replenishing performance tests of the slippery property were performed, as shown in Figs. 5(c) and 5(d), in which the cycle tests of the self-replenishing performance were terminated when the difference in SAs between the oil layer after being removed and replenished was

less than 1° . The CAs of all the gel films remained close to 98° regardless of the removal and replenishment of the oil layer, which was slightly larger than the previous value (88°), because of the removal of the first thicker oil layer (Fig. S1(c) in the ESM). The slight changes in the wettability of the oil layer on the surfaces were caused by the replenishment and removal of the thin oil layer. The SAs are distinguished in different states of the gel films in Fig. 5; the pink rhombus represents the SAs of the gel films after the oil layer was removed, and the gray circles represent the SAs after the gel films were stimulated by mechanical stress. In the gel film tests after the removal of the first oil layer, the SAs of GFs-80 and GFs-100 were $> 5^\circ$, but those of GFs-120 and GFs-150 were $< 5^\circ$ and increased as the cycles progressed, which indicated that the slippery property after the removal of the oil layer of GFs-120 and GFs-150 was superior to that of GFs-80 and GFs-100. Thereafter, the SAs of the four gel films that underwent the first pressure stimulation were $< 5^\circ$, and the SAs decreased as the oil content increased, indicating that GFs-150 had a better slippery property than the other films. The SAs of the gel films increased slightly after the removal of the oil layer and recovered to a lower value after the replenishment of the oil layer. The SA of each gel film increased after each cycle test but remained less than 10° after the termination of the self-replenishing cycle tests, indicating the excellent slippery property of the gel films before or after the removal of the oil layer. The lubricant self-replenishing cycle tests of the gel films were terminated after 2, 3, 5, and 7 cycles for GFs-80, GFs-100, GFs-120, and GFs-150, respectively, indicating that a higher oil content corresponded to a larger ROC of the gel film, resulting in a longer service life. Owing to the restrictions of the thickness and ROC of the gel films, the number of cycles was < 10 . Thus, service life was sacrificed for a thinner gel film, which made it easier to use in practical applications. Therefore, the slippery properties of the gel films can be maintained for a prolonged time simply via pressure stimulation.

3.3 Fog-harvesting properties of lubricant self-replenishing slippery surfaces

Fog capture, water condensation, and water removal

Fig. 5 Wettability and lubricant self-replenishing performance of the slippery surfaces. (a) Wettability and slippery properties of the slippery surfaces before the lubricant self-replenishing cycle tests. (b) State transition of the slippery surfaces during the lubricant self-replenishing cycle tests. The wettability and slippery properties of (c) GFs-80, (d) GFs-100, (e) GFs-120, and (f) GFs-150 with different numbers of cycles of oil-layer removal (rhombus) and replenishment (circles).

are generally recognized as the three stages of fog harvesting [22, 59]. First, the fog-capture process based on a humidifier is captured by small droplets from a mist flow by a surface, in which the oil-infused slippery surfaces have a higher droplet nucleation density than HBSs [37]. Water condensation is the process whereby the volumes of the droplets increase gradually. Furthermore, the higher droplet density on oil-infused slippery surfaces allows faster water condensation by accelerating the congealment of the droplets with adjacent droplets. Finally, when the droplets grow sufficiently, they slide off the surface because of gravity. Owing to the excellent slippery performance of oil-infused slippery surfaces, the droplets slide off easily. In this section, the effect of the oil layer on fog harvesting is investigated. Figure 6 shows the three stages of the fog-harvesting process for all the gel films. The small droplets from the mist flow were captured by the gel films, and the fog capture was faster on the silicone oil-containing gel films than on GFs-0 (the droplet nucleation to a similar density only took approximately 2 s on the oil-containing gel films (GFs-80, GFs-100, GFs-120, and GFs-150) but took

approximately 5 s on the gel film without oil (GFs-0)), indicating that the oil layer on the oil-containing gel films underwent more rapid droplet nucleation than the surfaces without oil. For all the gel films, in the process of water condensation, the small droplets grew and congealed to the critical volume, allowing them to slide off the surfaces. The droplets grew as more small droplets were captured on the surface; moreover, the droplets congealed with adjacent droplets, increasing their volume. Owing to the poor slippery property (Fig. 5(a)), GFs-0 required a larger area of the droplets to be congealed to the extent of sliding off the surfaces than the oil-containing gel films. Moreover, owing to the excellent slippery property, GFs-80, GFs-100, GFs-120, and GFs-150 required a smaller area and less time for the droplets to slide off than GFs-0 in the water-condensation process. GFs-150 exhibited the best fog-harvesting properties, with the smallest volume and shortest time for the droplets to slide off. These results indicate that the oil layer on the gel films was helpful for the water-condensation process. Finally, the droplets grew to the critical volume and slid off the surface owing

Fig. 6 Fog-harvesting procedures for (a) GFs-0, (b) GFs-80, (c) GFs-100, (d) GFs-120, and (e) GFs-150, including the stages of fog capture, water condensation, and water removal.

to gravity during the process of water removal. The volume, as well as the required time for the complete removal of droplets from the surfaces, decreased as the oil content of the gel films increased. The final removed droplets needed the most surrounding droplets and most time on the surfaces of GFs-0. The results indicate that the oil layer on the gel films played an active role in the three stages of the fog-harvesting process and that a thicker oil layer corresponded to a better fog-harvesting effect.

Additionally, the water-collection performance was tested to investigate the effects of the oil layer on fog harvesting. Figure 7(a) shows a schematic of the fog-harvesting system used in the present study. Figure 7(b) shows the time required for the first water droplet to be removed from the surface. The time required for the first water droplet to be removed from GFs-0 was 24.7 s, and those for GFs-80, GFs-100, GFs-120, and GFs-150 were approximately 2.7, 3.1, 4.9, and 5.1 s, respectively. Moreover, the time required for GFs-0 from water condensation to water removal from the surfaces was longer than those for the oil-containing gel films. The droplets on the surfaces of the oil-containing gel films coalesced with adjacent

droplets and slid off the surfaces quickly. However, a layer of water film remained after the droplets were removed from the surfaces of GFs-0 (Fig. 6(a)), which was beneficial for droplet nucleation but was an obstacle to the movement of droplets, resulting in poor performance in the water-condensation and water-removal stages. The water collection rate (WCR) of the gel films was determined by harvesting the mist flow for 30 min (Fig. 7(c)). Owing to their excellent slippery properties, the oil-containing gel films exhibited better fog-harvesting behavior than GFs-0. The WCR generally increased with the oil content, which is consistent with the analysis results for the fog-harvesting process. The WCR was 0.029 ± 0.004 g·min⁻¹·cm⁻² for GFs-150, which was approximately 395%, 134%, 119%, and 103% higher than those of GFs-0, GFs-80, GFs-100, and GFs-120, respectively. The excellent fog-harvesting performance of GFs-150 benefited from the superior slippery property and thicker oil layer compared with the other gel films. In contrast, the formation of a water film on GFs-0 after water removal led to the poor fog-harvesting performance of GFs-0. Additionally, the mass loss of lubricant from the gel films, as well as the COD of the collected

Fig. 7 Fog-harvesting properties of the lubricant self-replenishing slippery surfaces. (a) Schematic of the fog-harvesting system. (b) Time needed for the first water droplet to be removed from the surfaces. (c) Water collection rate for different films within 30 min. (d) Mass of water collected in 120 min for GFs-0 and GFs-150. WCRs of GFs-150 in different cycles (e) with and (f) without pressure stimulation.

water generated during the fog-harvesting process, was tested to assess the quality of the collected water (Fig. S6 in the ESM). Because there was no oil layer on the surface of GFs-0, the mass loss of the lubricant of GFs-0 was 0. The mass loss of the lubricant of GFs-150 after 30 min of fog harvesting was 4.87 ± 1.63 mg, which was approximately 256%, 203%, and 168% larger than those of GFs-80, GFs-100, and GFs-120, respectively. The COD is the required oxygen equivalent in the process where reductive substances in the collected water are oxidized by a strong oxidant. A higher COD indicates that more reductive substances were present in the collected water. In the present study, the reductive substances were mainly the evaporative residue of THF and lost silicone oil of the gel films, which were removed by droplets from the gel films during the droplet-removal stage in the fog-harvesting process. The COD of GFs-0 was 105 ± 1 $\text{mg}\cdot\text{L}^{-1}$, which was mainly generated by the evaporative residue of THF. The CODs of GFs-80, GFs-100, GFs-120, and GFs-150 were 163 ± 2 , 179 ± 2 , 183 ± 1 , and 181 ± 1 $\text{mg}\cdot\text{L}^{-1}$, respectively. Owing to the constant mass of prepolymers carried on the glass at a shear rate of 500 rpm, there were differences in the rates of PDMS-NH₂, silicone oil, and THF in the gel films. Table S2 in the ESM presents the mass ratios of

silicone oil, THF, and both of them, which accounted for the total mass (denoted as R_2 , R_3 , and R_4 , respectively). As the content of silicone oil increased, R_2 and R_4 increased, but R_3 decreased, as shown in Table S2 in the ESM. The total mass of silicone oil and THF reserved on the glass increased with the increasing silicone oil content, which was consistent with the COD tendency. The COD was influenced by the synergistic effect of R_3 and R_2 , and the COD of THF was higher than that of silicone oil by consuming the same quality of THF and silicone oil. The COD of GFs-150 was slightly reduced, mainly owing to the decrease in R_3 . The COD obtained in the present study was slightly higher than the standard for daily water, which can be satisfied by simple sewage treatment. Moreover, the mass of the water collected in 120 min was tested for GFs-0 and GFs-150 (Fig. 7(d)). The mass of collected water in 120 min for GFs-150 was 2.38 ± 0.27 $\text{g}\cdot\text{cm}^{-2}$, which was 251% larger than that for GFs-0. These results indicate the superiority of gel films with an oil layer for fog harvesting.

Additionally, fog-harvesting tests of GFs-150 with and without pressure stimulation were performed (Figs. 7(e) and 7(f)), in which the surface of GFs-150 without pressure stimulation represented a slippery surface without the self-replenishing function. Because

of the mechanical stress trigger, the fog-harvesting capability, as well as the service life, of the GFs-150 surface was superior to those of the surfaces without pressure stimulation. This suggests that the lubricant self-replenishing slippery surfaces had a better fog-harvesting capability and service life than the surfaces without the function of self-replenishment. The WCR after the first pressure stimulation was $0.029 \pm 0.001 \text{ g} \cdot \text{min}^{-1} \cdot \text{cm}^{-2}$ (Fig. 7(e)), which was similar to the WCR of the gel films before the first oil layer was removed. The WCR decreased as the cycle progressed, but the WCR of GFs-150 after seven cycles was only slightly lower than that of GFs-80 before the removal of the oil layer and was still higher than the WCR of GFs-0 owing to the self-replenishment function of GFs-150. The COD of the water collected after each pressure stimulation is shown in Fig. S9 in the ESM. It decreased as the cycle progressed, resulting from the reduced content of excretive oil. The WCR of GFs-150 without pressure stimulation decreased sharply (Fig. 7(f)), and the WCR of the fourth cycle was similar to that of GFs-0 because the oil layer faded away with the fog-harvesting tests. These results indicate that the service life of lubricant self-replenishing slippery surfaces for fog harvesting can be extended only by pressure stimulation. Table 1 presents recent studies on liquid-infused slippery surfaces for fog harvesting. The WCR achieved in this study was better than those of some previous studies; additionally, the lubricant self-replenishing slippery surfaces had an extended service life, which was not achieved in previous studies.

3.4 Stability of lubricant self-replenishing slippery surfaces

Furthermore, the physical stability and chemical stability of the lubricant self-replenishing slippery surfaces were evaluated (Figs. 8(b)–8(f) and S10–S12 in the ESM). First, the sliding process of 10- μL water droplets on GFs-150 with an SA of 0.75° was examined, as shown in Fig. 8(a). The distance slid by the 10 μL of water droplets in 60 s was 2.21 mm, and the sliding speed of 10- μL water droplets on the surface of GFs-150 with an SA of 0.75° was $0.037 \text{ mm} \cdot \text{s}^{-1}$. In the physical-stability tests, the wear resistance, temperature stability, and long-term stability of the lubricant self-replenishing slippery surfaces were investigated. Figures 8(b) and 8(c) show the friction coefficients and wear rates of the lubricant self-replenishing slippery surfaces. The friction coefficient of GFs-0 increased before 200 cycles and stabilized at approximately 0.74, which is close to the friction coefficient of glass [63]. Therefore, it can be considered that the gel film of GFs-0 was worn through after 200 cycles, and the wear volume of GFs-0 can be considered as the volume of gel films worn through. The friction coefficients of GFs-80, GFs-100, GFs-120, and GFs-150 stabilized at approximately 0.062 after a running-in period of approximately 150 cycles. The wear rate of the gel films decreased with an increasing oil content, indicating that the lubricant self-replenishing slippery surfaces reduced friction more effectively than GFs-0 and had a superior wear resistance. Regarding the temperature stability, the CAs of the gel films were smaller at 20°C

Table 1 Previously reported liquid-infused slippery surfaces for water harvesting.

Surface type	<i>T</i> ($^\circ\text{C}$)	<i>RH</i> (%)	Fog speed ($\text{cm} \cdot \text{s}^{-1}$)	Flow rate ($\text{L} \cdot \text{h}^{-1}$)	Distance between sample and fog outlet (cm)	WCR ($\text{g} \cdot \text{min}^{-1} \cdot \text{cm}^{-2}$)	Ref.
Hierarchical micro/nanostructure slippery surface	20	80	40–50	Not reported	6	0.014	[30]
WO ₃ -based slippery surface	18	85	Not reported	0.25	5	0.008	[60]
PDMS grafted ZnO slippery surfaces	18	90	50	0.25	5	0.011	[61]
Patterned slippery-superhydrophilic surface	20	80	40–50	Not reported	6	0.036	[62]
Perfluoropolyether (PFPE)-infused CuO-coated slippery surface	18	90	50	0.25	2	0.011	[23]
Lubricant self-replenishing slippery surface	15	99	240	1.8	20	0.029 ± 0.004	This work

T represents the temperature and *RH* represents the relative humidity.

than at 0, 60, 80, and 100 °C, which mainly resulted from the variations in the surface tension of water at different temperatures [41]. The SAs of GFs-150 at temperatures of 0, 20, 60, 80, and 100 °C were $1.37^\circ \pm 0.05^\circ$, $1.49^\circ \pm 0.08^\circ$, $1.42^\circ \pm 0.07^\circ$, $1.38^\circ \pm 0.16^\circ$, and $1.39^\circ \pm 0.10^\circ$, respectively, which were generally stable with the variations of temperature (Fig. 8(d)). The SAs of GFs-80, GFs-100, and GFs-120 changed slightly within varies of temperature (Fig. S10 in the ESM), indicating that the lubricant self-replenishing slippery surfaces were stable below 100 °C. The long-term stability of the gel films was tested by assessing the CAs and SAs of the lubricant self-replenishing slippery surfaces with a storage time of 1–10 days in an ambient environment (Figs. 8(e) and S11 in the ESM). There were no significant changes in the CAs or SAs of water on the surfaces of the gel films over time, indicating the long-term stability of the gel films. The antifouling properties of slippery surfaces are important for practical applications. To evaluate the chemical stability, the pH stability of the lubricant self-replenishing slippery surfaces was tested (Figs. 8(f) and S12 in the ESM). The CAs of acidic droplets on the surfaces of GFs-80, GFs-100, GFs-120, and GFs-150

fluctuated around 90° . The CAs of the alkaline droplets fluctuated around 90° on the surfaces of GFs-80 and GFs-100 but decreased slightly with the increasing pH on the surfaces of GFs-120 and GFs-150. The CAs of strong alkaline droplets (pH = 14) on the surfaces of GFs-80, GFs-100, GFs-120, and GFs-150 decreased significantly, to $31.8^\circ \pm 2.8^\circ$, $37.1^\circ \pm 4.4^\circ$, $21.9^\circ \pm 3.5^\circ$, and $16.5^\circ \pm 2.6^\circ$, respectively. Furthermore, the SAs of acidic and alkaline droplets on the surfaces of the gel films fluctuated around 3° , which indicated the excellent slippery properties of the lubricant self-replenishing slippery surfaces. Generally, the gel films were stable for acidic and alkaline droplets, with strong alkaline droplets spreading rapidly over their surfaces, and the wettability of the gel films became superhydrophilic upon exposure to strong alkaline droplets. This indicates that the lubricant self-replenishing slippery surfaces prepared in this study were stable against acids but susceptible to alkalis [27]. Nevertheless, the slippery property of the gel films was highly stable for droplets with different pH values. Additionally, as shown in Fig. 9, the antifouling properties of the lubricant self-replenishing slippery surfaces were tested by sliding liquid droplets available

Fig. 8 Physical and chemical stability of the lubricant self-replenishing slippery surfaces. (a) Sliding procedure for 10 μL of water droplets sliding on GFs-150 tilted at 0.75° ; the scale bar represents 1 mm. (b) Friction coefficient (the inset shows a magnification of the friction coefficient of gel films with silicone oil added) and (c) wear rate of the lubricant self-replenishing slippery surfaces. (d) CAs and SAs of 10-μL water droplets sliding on GFs-150 at different temperatures. (e) CAs and SAs of 10 μL of water droplets sliding on GFs-150 after a storage time of 1–10 days. (f) CAs and SAs of 10-μL water droplets with different pH values sliding on GFs-150.

Fig. 9 Antifouling tests: 10- μ L water, tea, energy drink, cola, milk, and coffee sliding on (a) GFs-150 and (b) GFs-0. The droplets slid on GFs-150 but were pinned on GFs-0.

in daily life: water, tea, energy drinks, cola, milk, and coffee, which had different surface tensions. All six types of liquid droplets slid smoothly on GFs-150, whereas they were pinned immediately for GFs-0. The excellent durability and antifouling properties of the lubricant self-replenishing slippery surfaces prepared in the present study suggest their practical applicability.

4 Conclusions

An ultrathin transparent lubricant self-replenishing slippery surface was fabricated via facile one-step solvent evaporate polymerization of PDMS-NH₂ and TDI, in which a slippery gel film was attached to glass slides and oil microdroplets were embedded in it via phase separation and evaporation of THF. The FT-IR spectra confirmed that the amino groups of PDMS-NH₂ and the isocyanate group of TDI were successfully polymerized to the urea group. Lubricant self-replenishment cycle tests, in which the oil layer was removed and then replenished, were conducted to evaluate the lubricant self-replenishing performance of the slippery surfaces. GFs-150 exhibited the longest service life among the gel films. Additionally, the fog-harvesting performance of the lubricant self-replenishing slippery surfaces was investigated. GFs-150 exhibited superior performance to the other gel films, as well as a longer service life, owing to the higher content of silicone oil. Only 4.8 s was needed for

the first water droplet to be removed from GFs-150 (five times faster than the case of GFs-0 and faster than previous studies), and the WCR of GFs-150 was 395% higher than that of GFs-0. After the first cycle test, the slippery and fog-harvesting performance of GFs-150 was recovered nearly relative to that before the oil layer was removed. The WCR of GFs-150 after seven cycles was only slightly lower than that of GFs-80 before the removal of the oil layer and was higher than that of GFs-0, owing to the self-replenishment function of GFs-150. Owing to the trait of oil secretion triggered by mechanical stress, the lubricant self-replenishing slippery surfaces had a better fog-harvesting capability and longer service life than surfaces without the function of self-replenishment. The ultrathin, transparent, and lubricant self-replenishing slippery surface is suitable for applications involving narrow spaces, visualization, long service lives, etc.

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Yi CHEN. She is currently a Ph.D. candidate at the Lanzhou Institute of Chemical Physics (LICP), Chinese Academy of Sciences (CAS), under the supervision of Prof. Zhiguang

GUO. She received her B.S. degree (2016) and M.S. degree (2019) in North University of China (NUC) under the supervision of Prof. Chenyang XUE. Her current scientific interests are focused on liquid-infused surfaces and its application.

Jinxia HUANG. She received her Ph.D. degree from the Lanzhou Institute of Chemical Physics (LICP), Chinese Academy of Sciences (CAS), in 2015. After that, she joined Prof.

Guo's Biomimetic Materials of Tribology (BMT) group in LICP. Her current scientific interests are focused on the biomimetic materials of tribology and its application.

Zhiguang GUO. He received his Ph.D. degree from the Lanzhou Institute of Chemical Physics (LICP), Chinese Academy of Sciences (CAS), in 2007. After that, he joined Hubei University. From Oct. 2007 to Aug. 2008, he worked in University of

Namur (FUNDP), Belgium, as a post-doctor. From Sept. 2008 to Mar. 2011, he worked in the Funds of

National Research Science (FNRS), Belgium, as a “Charge de Researcher”. During Feb. 2009–Feb. 2010, he worked in the Department of Physics, University of Oxford, UK, as a visiting scholar. Now he is a full professor and the dean of School of Materials Science and Engineering, Hubei University. His current scientific interests are focused on the biomimetic materials of tribology and its application.

Weimin LIU. He received his Ph.D. degree in lubricating materials and tribology from the Lanzhou Institute of Chemical Physics (LICP) of the Chinese Academy of Sciences (CAS), in 1990. After that, he joined the State Key Laboratory of Solid Lubrication

(LSL) of the LICP. From June 1993 to June 1994, he worked as a visiting scholar at Pennsylvania State

University, USA. In 2013 and 2016, he was elected the member of the Chinese Academy of Sciences and the fellow of The World Academy of Sciences (TWAS). Currently, he is a professor of the LICP and the head of the State Key Laboratory of Solid Lubrication. His research interests mainly focus on space and aviation lubrication, high performance lubricating materials and tribochemistry.