Durable slippery lubricant-infused multiscale-textured surfaces for repelling highly adhesive liquids

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Abstract
Surfaces that can repel various types of liquid and retain surface properties over acceptably long periods of time are in great demand. Here, we presented a simple but effective technique to fabricate slippery, lubricant-infused surfaces with excellent liquid-repellent properties and resistance to hydrodynamic damage, evaporation, and high static pressure. Chemically-functionalized multiscale-textured surfaces were impregnated by highly-viscous and vacuum-grade lubricants that fully wetted the nanoscale roughness while conformed to the microscale textures. This generated slippery rough surfaces with improved liquid-resistant properties evaluated by water and highly-adhesive latex. The respective contact angles of water and latex droplets were above 130.1° ± 0.8° and 105.7° ± 1.1°, while water and latex sliding angles were less than 5.8° ± 0.7° and 8.7° ± 0.7°, respectively. More importantly, the slippery roughness reduced liquid-lubricant contact areas, and protected the lubricating layer from flow-induced erosion. The particular lubricant-infused surfaces can withstand an impact of a water jet speed up to 2.6 m s⁻¹ for at least 10 min. Furthermore, the viscous lubricant layer was unaffected by evaporation at 65 °C for at least 11 weeks, and stable under hydrostatic pressure of 150 kPa for 20 min.

1. Introduction

The past decade has witnessed rapid advancements in fundamental research and fabrication techniques to develop and produce surfaces that repel various types of liquids [1, 2]. Special surface functionalities offer promising applications across a broad range of technologies, including self-cleaning [3, 4], friction reduction [5–8], anti-icing [9], water harvesting [10], liquid separation [11] and transportation [12]. By exploiting the key fundamentals behind natural non-wetting structures, especially lotus leaves, researchers have successfully developed superhydrophobic surfaces (SHSs) that use surface roughness to create numerous air pockets between the substrate and the contacted liquid [13]. Water deposited on an SHS generally beads up and forms spherical droplets with apparent contact angles greater than 150° that roll off easily when the substrate is tilted at an angle typically smaller than 10° [14]. Liquid-repellent properties superior to those of bio-mimicking SHSs were developed by incorporating materials with ultra-low surface energy [15], or by utilizing complex surface textures generally associated with multiscale roughness or re-entrant structures [13, 14]. Such liquid-repellent surface engineering has extended the range of fluids that can be repelled to include liquid hydrocarbons with surface tensions less than 15 mN m⁻¹ [16], and complex liquids such as blood, ink and milk [17, 18]. Studies have shown that re-entrant surface structures play a central role in the resistance to liquid penetration of micro- and nano-sized surface cavities that uphold the liquid-air interfaces required by the Cassie-Baxter anti-wetting state [16, 19]. Furthermore, a surface that is hierarchically structured in multiple length scales was proved not only to increase the energy barrier inhibiting Cassie-Baxter to Wenzel wetting transition [20], but also to minimize the
direct liquid-solid contact area, greatly reducing surface adhesion [21, 22]. In a previous work, we demonstrated that a triple-scale surface roughness, obtained from the intrinsic structure of fluoroalkyl-functionalized silica aggregates, exhibited excellent anti-wetting properties that could repel even a highly sticky and viscoelastic liquid like concentrated natural latex [23]. In fact, considering its highly adhesive nature and the short attachment time of this polymeric material [24, 25], liquid latex would be one of the most challenging classes of liquids for surface repellent properties. Rubber latex is also among the most important industrial materials, used to manufacture products such as adhesives, paints, vehicle tires, gloves, and anti-vibration products. Therefore, a latex-repellent coating would improve processing efficiency and avoid material loss of about 10%–15% caused by adhesion during harvesting, handling, and transportation [26]. However, studies related to this application are hard to find in the literature.

Despite all research efforts, the practical applications of existing liquid-repellent surfaces are still restricted. The surfaces present long-term durability problems because their properties are susceptible to hydrostatic pressure [27, 28], physical wear [26, 29], and aging under certain conditions [30, 31]. At certain levels of external pressure, the essentially metastable non-wetting Cassie-Baxter state can collapse to a Wenzel wetting state. Air trapped in surface voids cannot withstand the pressure-induced intrusion of liquids, especially those with low surface tension. These liquids can then easily infiltrate and fully wet the surface. Furthermore, conventional SLIPSs are notoriously impaired by defect sites caused by physical damage or imperfect chemical passivation [32, 33], which are prone to induce contact line-pinning. The situation is even worse when liquids that contain elastic polymeric materials are involved. Irreversible polymer adsorptions at liquid-solid contacts rapidly aggravate any surface defect. In the case of liquid latex, the elasticity of accumulated rubber molecules substantially dissipated pulling forces [34] and more energy was required to separate an adhered droplet from the solid surface once attached, resulting in a substantial increase in contact angle hysteresis [35, 36]. When adsorbed polymer chains overlay the original surface composition, the surface will lose its repellent property.

Recently, a new strategy was devised to create a more durable liquid-repellent surface by replacing the Cassie-Baxter imperfect solid-liquid interface with a defect-free and molecularly smooth liquid-liquid interface [37]. This concept is exemplified by the slippery liquid-infused porous surface (SLIPS), which utilizes surface porosity to immobilize a layer of immiscible lubricant [34, 38]. Rather than relying on surface roughness to repel a liquid, a SLIPS presents a smooth and slippery outer layer of infused lubricant that allows droplets to slide effortlessly off the surface. The incompressible lubricant is infused between surface features, displacing the pockets of air. SLIPSs were reported intrinsically stable against external pressure and vapor condensation [39, 40]. Furthermore, lubricant-infused slippery surfaces were able to restore their liquid-repellent function when the porous substrate was damaged by abrasion or impact [41]. The fluidic nature of the lubricating layer allows the lubricant to wick into the damaged area by capillary action and refill the voids.

In general, fabrications of SLIPS surfaces utilize ultra-low surface tension lubricants to promote lubricant impregnation, and to render a smooth lubricating layer that enhances surface slipperiness. This strategy, however, poses durability problems, since low-molecular-weight lubricants vanish rapidly by evaporation [42]. Furthermore, a lubricating layer with low viscosity affords low shear strength that could be easily dragged away when contact to dynamic fluid. In fact, external shear flow and liquid impact were reported to induce failures of SLIPS properties, as the lubricating layer was drained over time [43–45]. This shear-driven damage problem would be worse for the case of adhesive liquid. To date, lubricant-infused surfaces that are robust enough to withstand high abrasive flow have not been realized. A solid lubricant incorporating 2D nanomaterials is one of the promising strategies to produce a firm slippery coating layer [46–48]. However, its liquid-repellent properties especially for highly-adhesive polymeric liquid need validations and more investigations.

In this research, we demonstrated fabrication technique to produce durable lubricant-impregnated surfaces that were stable against physical wear caused by high shear flow and liquid impact. Here, highly viscous lubricants with viscosity one to two orders of magnitude higher than normal were infused into a porous substrate to produce a slippery lubricant-infused multiscale-textured surface (SLIMS) with shear-resistant properties. Infusion of such viscous lubricants into micro/nanoscale pores was possible at elevated temperature where the viscosity was temporarily reduced, enabling capillary-driven permeation of the surface texture. Once cooled down, the lubricant regained its original viscosity and was firmly locked in placed by the multiscale textures with a slight decrease in its volume. The volume reduction exposed some degree of surface roughness, as the final lubricating layer conformed to the surface topography. This advantageous slippery rough feather reduced liquid-lubricant contact area, which further minimized lubricant loss by shear flow. Here, comparative liquid-repellent performance studies of surfaces impregnated by lubricants with different viscosities were carried out using water and adhesive liquid latex. Also, resistance to water-jet impact, evaporation loss, and static pressure, was evaluated.
2. Experimental

2.1. Materials

Precipitated silica particles (Ultrasil 7000 GR) with a specific surface area of 175 m²g⁻¹ were purchased from Evonik Industry. Xylene (dimethyl benzene), fluoroalkylsilane coupling agents 1H,1H,2H,2H-perfluorodecytriethoxysilane (FDTS), poly(vinylidene fluoride-co-hexafluoro propylene) (PVDF-HFP) and acetone (AR grade) were from Merck. Butyl-cellosolve (2-Butoxyethanol) was supplied by Hangzhou Tiankai Enterprise. Preserved natural latex (Hevea brasiliensis) concentrate (industrial standard, 60%w/w of rubber in water medium) was obtained from Thai Rubber Latex. Three types of perfluoropolyether (PFPE) lubricant with different kinematic viscosities were obtained: general-purpose Krytox 101 from Dupont, vacuum-pump grade Fomblin Y-LVAC 16/6, and diffusion-pump grade Fomblin Y-HVAC 140/13, both from Solvay. Their nominal viscosities were 16, 168, and 1508 cSt, respectively; their respective vapor pressures were listed as ∼10⁻⁴, 8 × 10⁻⁸ and 7 × 10⁻¹² torr at 20°C, and their lubricant surface tension values at 20°C, measured by the pendant drop method, were 28.6 ± 0.6, 33.1 ± 1.0, and 36.5 ± 0.3 mNm⁻¹.

2.2. Preparation of SHS and SLIMS samples

The coating formulation was based on functionalized silica nanoparticles prepared by transforming hydrophilic silanol groups (–SiOH) in their surface composition to hydrophobic fluoroalkyl groups (–CF₃). The modification was achieved by stirring 0.5 g of precipitated silica with 0.2 ml of FDTS molecules in 10 ml of xylene for 75 h at room temperature. The silica content in the solvent was approximately 5%w/v. A polymer binder solution was prepared separately by dissolving 10% w/v of PVDF-HFP in acetone under stirring at 45°C for 0.5 h. The functionalized silica stock and the polymer binder solution were then mixed at a PVDF-HFP:SiO₂ mass ratio of 1:2 and 1%v/v of butyl-cellosolve was added. The mixture was stirred for 0.5 h at 45°C to obtain a homogeneous formulation. The coating solution was applied on a glass slide substrate by a normal spray-casting technique and allowed to dry at ambient temperature for at least 24 h to obtain the SHS samples with multiscale surface roughness (figure 1). To produce the SLIMS samples, 2 ml of PFPE liquid were dropped onto SHS samples at 40°C. At this temperature, the infiltration mobility of the PFPE was improved and the liquid spread spontaneously. The temperature was maintained for 24 h to allow the complete infusion of the liquid before the temperature was lowered back to the ambient, immobilizing the fluoro-lubricants within the surface texture. Three different types of PFPE-based lubricants with viscosities of 16, 168, and 1508 cSt were successfully infused.

Figure 1. A schematic diagram illustrates (a) a brief fabrication process, (b) multiscale-textured structure of pre-infused SHS films, and (c) SLIMS structure with slippery roughness. This feature reduces effective contact area, minimizes shear-driven damage and prevents pinning points caused by polymeric adsorption.
into the multiscale-textured samples, which were denoted as SLIMS1, SLIMS2 and SLIMS3, respectively. The low-viscosity, low-surface-tension oil infused in SLIMS1 is the most commonly used lubricant for slippery liquid-infused surfaces [49].

2.3. Characterization of SHS and SLIMS

2.3.1. Surface morphology and topography

Surface microscopic topographies of SHS and SLIMS samples were characterized by atomic force microscopy (AFM, Nanosurf EasyScan 2) in non-contact mode with an ACL-A cantilever (AppNano) at room temperature. AFM images (50 × 50 μm²) were taken of random locations on the sample surfaces, and the root mean square (RMS) roughness was analyzed by Gwyddion software to examine the effects of the infused lubricant on surface roughness. Morphological structures of SHS and SLIMS were also studied using field-emission scanning electron microscopy (FESEM, FEI Quanta 400, 25 kV). Here, SLIMS3 was the only lubricant-infused sample that could undergo SEM operations as the PFPE lubricant was nonvolatile with an extremely low vapor pressure, on the order of 10⁻¹³ torr. Virtually no evaporation loss could be expected for a diffusion-pump-grade fluorofluid of this type under ordinary SEM operating vacuum conditions of ~10⁻⁴ torr at room temperature. SLIMS1 and SLIMS2, on the other hand, would have a strong tendency to release PFPE vapor into the SEM chamber.

2.3.2. Wetting properties of the coatings

The wetting properties of SHS and SLIMS samples were evaluated using two different types of test liquid: DI water and 40% w/w latex, which was obtained by diluting the 60% w/w latex concentrate with DI water to achieve the average concentration of freshly tapped natural latex. Contact angle (CA) measurements were performed using probe liquid droplets of ~2 μl placed at multiple locations on the sample surface. Droplet images were acquired with an optical contact angle measurement system (DataphysicsOCA-15EC), and analyzed by built-in drop shape analysis software. Sliding angle (SA) measurements were carried out by means of a tilting stage using droplets of ~10 μl on the sample surface. The droplet was set in motion by slowly adjusting the tilt angle. Multiple measurements were repeated for tilt angles in both clockwise and counterclockwise directions.

2.3.3. Durability tests

One of the most demanding requirements of slippery liquid-infused surfaces is the ability to maintain liquid-repellent properties over an acceptable time period under practical conditions. Here, the durability of the SLIMSs against water erosion, static pressure, and lubricant evaporation was evaluated by comparison with normal SHSs.

The mechanical robustness of the coating in the face of physical wear induced by hydrodynamic erosion is crucial to outdoor performance, where damage caused by rainfall would be unavoidable. Water jet erosion is an accelerated weathering test commonly used to examine the mechanical durability of coatings against fluid abrasion. The procedure of the water jet erosion test was detailed in [50]. Briefly, a continuous jet of water was directed at the sample at a 45° incident angle (figure 2(a)). The water jet had a diameter of approximately 0.5 cm and was released at pressures from 1–5 kPa, producing stream speeds from approximately 1.3 to 2.6 ms⁻¹. Samples were exposed to the water stream for 10 min before they were examined for changes in their wetting properties.

The resistance of the prepared SLIMSs to the loss of infused lubricant due to evaporation under raised temperatures was evaluated by placing samples in a temperature-controlled chamber with a slow rate of air exchange to prevent condensation of the evaporated lubricant molecules back onto the surface. The exposure temperature was set at a constant 65 °C. To measure wetting properties, samples were taken once a week over the 11 weeks of total exposure time.

To investigate the effects of external pressure on wetting behaviors, SHS and SLIMS samples were laid horizontally at the bottom of a high-pressure chamber containing test liquids of DI water or liquid latex to a depth of 1 cm (figure 2(b)). External pressure from 0 to 150 kPa was applied to the system using an air compression pump. The elevated pressure condition was maintained for 20 min and then the samples were taken out to be examined for resulting CA and SA values.

3. Results and discussion

3.1. Morphological and topographical observations

The surface topography of SHS and SLIMS samples were examined using AFM to produce color-mapped roughness profiles and cross sectional line scans. Imaging results confirmed several length scales of surface roughness, comprising high-frequency nanoscale roughness superimposed on underlying larger scales of height.
fluctuation (figure 3). SHS samples, in particular, clearly presented deep apertural structures of microscopic scale throughout the surface area, which resulted in a higher degree of overall roughness compared to SLIMS samples. Quantitative analysis of the RMS surface roughness of SHS samples gave a value of $1,225 \pm 420$ nm, which accounted for all the observed length scales of roughness. On the other hand, the RMS surface roughness values of SLIMS1, SLIMS2 and SLIMS3 were $773 \pm 146$, $646 \pm 176$ and $599 \pm 119$ nm, respectively. The reduced roughness was the result of lubricant impregnation. The RMS roughness of the SLIMS samples tended to reduce toward higher viscosity owing to the thickening of the lubricant layer. AFM results agreed well with SEM images that the microscale cavities of the SLIMS samples were filled with PFPE lubricating liquid but that the entire surface was not overlaid, since surface roughness features could still be observed. In contact with test liquid, these slippery rough surfaces could create air pockets underneath the liquid, combining Cassie-Baxter characteristics with pinning-free liquid-lubricant contact lines.

The extent of porosity and roughness is a crucial morphological component that enables a high affinity between a substrate film and an infused lubricant. SEM was employed to observe the complex morphology of the coated samples before and after infusion with the PFPE lubricant. SEM imaging revealed stochastic surface structures of SHS films in planar and cross-sectional views (figures 4(a)–(b)). Overall, the coated surfaces were crowded with multiscale micro- and nanostructures connected by a network of polymer binder. Surface roughness was created by hierarchical structures of primary and secondary silica particles (indicated by arrows and solid circles) which presented length scales of $\sim 10–15$ nm and $\sim 100–300$ nm, respectively. These dual-size particle aggregates that imparted nanoscale roughness to the film are essentially the natural form of precipitated silica nanoparticles. Larger structures (marked by dashed circles) of microscopic aggregates of tertiary particles, were also commonplace. These structures were mainly composites of secondary silica particles and polymer binder. The sub-level of the film comprised porous PVDF-HFP networks that provided robust mechanical support to the surface texture and added comparatively large re-entrant cavities that accommodated impregnation even by the more viscous liquids. There was no evidence of excessive binder build-up either on the surface or in the interstitial regions; porous features were therefore preserved across all length scales. This characteristic was attributed to the self-stratification of particulate film under a convection-dominated transport regime during quick film drying [31]. Here, acetone was used at the initial stage of film deposition to induce a rapid evaporation rate that produced a skin layer of high-density particles [32].

Rounder and smoother surface features were clearly observed on the SLIMS3 sample (figures 4(c)–(d)). The lubricant penetrated the polymer pores, wetting all the length scales of surface roughness. However, different lubricant wetting behaviors were found at different roughness length scales. Silica aggregate nanostructures with features much smaller than the capillary lengths of PFPE lubricants, were smoothly overlaid with a lubricant layer stabilized locally by surface tension with no bare substrate exposed. On the other hand, the lubricant tended to conform to the microscale topography as it sagged into the re-entrant cavities. Thus, the SLIMS samples could be regarded as slippery rough surfaces rather than conventional flat and smooth SLIPS, which are usually obtained from surfaces that present only nanoscale textures [43, 53].

3.2. Wetting properties
SHSs in general show highly liquid-repellent characteristics, which involve large liquid-solid contact angles and small droplet roll-off angles. When a water droplet was deposited on SHS samples, it appeared round with a
water contact angle (WCA) of 161.1° ± 0.6° and a water sliding angle (WSA) of 2.2° ± 0.2° (figure 5). The multiscale-textured SHS in the present study also exhibited decent anti-wetting behavior in contact with a sticky latex droplet. Owing to surface asperities that reduced the latex-solid contact area, the latex contact angle (LCA) was as large as 153.8° ± 0.2° and the latex sliding angle (LSA) was as small as 4.2° ± 0.3°. Theses WCA and WSA are in good agreement with the previous report [23]. Smaller contact angles for liquid latex can be attributed to its lower surface tension compared to water. Apparent contact angles were smaller on all SLIMSs than on the SHS as the liquid-solid-air interfaces were replaced by liquid-lubricant-air interfaces. Characteristic annular wetting ridges were also formed at droplet bases without detectable cloaking layers, which were the result of the moderately low surface tensions of the infused lubricants. SLIMS1 samples repelled liquid the best among all lubricant-infused samples, exhibiting a WCA of 155.4° ± 0.6°, an LCA of 115.3° ± 0.4°, a WSA of 3.5° ± 0.4° and an LSA of 6.3° ± 0.6°. Because of roughness effect, the SLIMS1 samples with commonly used low-surface-tension lubricant showed much higher water contact angles when compared with flat and smooth SLIPS cases [40]. However, liquid-repellent properties reduced progressively toward SLIMS2 and SLIMS3 with slight decreases in contact angle accompanied by increments of sliding angle (table 1). This behavior can be explained by the increasing surface tension values of the infused lubricants and the reduced roughness of the surfaces infused with more viscous lubricant.

Figure 3. AFM color-mapped images and the corresponding line scans were produced by scanning 50 × 50 μm² areas of surface coating structures on (a) SHS, (b) SLIMS1, (c) SLIMS2 and (d) SLIMS3 samples.
Figure 4. SEM images (a)–(b) reveal the multiscale roughness of an SHS sample from top and cross-section views. The arrow and solid circles indicate primary and secondary silica particles respectively, while the dashed circles surround the larger scale roughness produced by composites of secondary particles and porous PVDF-HFP structures. A SLIMS surface (SLIMS3 sample) shows the conformal coating of fluoro-lubricant, locked in place within the open structures (c)–(d).

Figure 5. Wetting characteristics of (a)–(b) water droplets on SHS and SLIMS3 samples, in comparison with (c–d) latex droplets. The arrows indicate characteristic wetting ridges formed at the liquid-lubricant-solid interface.
\[ \cos \theta_Y = \frac{\gamma_{OL} - \gamma_{LG}}{\gamma_{LG}}, \]

with the subscripts \( O \), \( L \), and \( G \) represent oil (lubricant), liquid, and gas phases, respectively. When accounting for surface roughness, a substrate can be modeled as a composite between oil and air, and the apparent contact angles can be determined from the weighted averages of liquid-lubricant and liquid-air components according to the Cassie- Baxter model,

\[ \cos \theta_{CB} = f \cos \theta_Y - (1 - f), \]

where \( f \) is the fractional area associated with a liquid-lubricant interface. Therefore, with a higher degree of roughness, the apparent contact angle becomes larger.

### 3.3. Durability tests

To perform successfully in applications outside laboratories, liquid-infused surfaces must be robust enough to conserve their slippery functions against various external conditions. In term of mechanical robustness, SHS and SLIMS samples were assessed for the degradation of their anti-wetting properties due to hydrodynamic damage. Samples were exposed for 10 min to a water jet at various pressures at an incident angle of 45° that generated both shear and normal impact damage. The properties of the conventional SHS were easily diminished even at low jet pressure, observed from the significant drop of WCA together with the increase of WSA (figure 6). The damaged surfaces produced similar behaviors from both water and latex test liquids. The dynamic pressure exerted by the water jet physically damaged the particulate film, reducing its roughness, and enabling water to infuse the surface texture. On the contrary, all SLIMS samples exhibited good resistance to flow-induced wear, and both WCA and LCA were almost unaffected. A small increase in WSA and LSA was observed but the liquid infuse the surface texture. On the contrary, all SLIMS samples exhibited good resistance to hydrodynamic damage. Samples were exposed for 10 min to a water jet at various pressures at an incident angle of 45°. As shown in figure 6, the fabrication of SLIMSs withstood jet speeds up to 2.6 ms\(^{-1}\) for 10 min, retaining their pinning-free liquid-repellent properties even when tested with sticky liquid latex. According to the cited calculation \([4, 44]\), water jet impact momentum at a speed \( V_{\text{jet}} \) for a duration \( t_{\text{jet}} \) could produce erosion damage equivalent to a rainfall duration, \( t_{\text{rain}} \), given by the following equation,

\[ t_{\text{rain}} = \frac{V_{\text{jet}}^2 t_{\text{jet}}}{f_{\text{rain}} V_{\text{drop}}}, \]

where, \( V_{\text{drop}} \) is raindrop volume, \( V_{\text{rain}} \) is rain final speed, and \( f_{\text{rain}} \) is rain impact frequency. Since in this study, \( V_{\text{jet}} = 2.6 \) ms\(^{-1}\), \( t_{\text{jet}} = 10 \) min, \( V_{\text{rain}} = 9 \) ms\(^{-1}\), \( f_{\text{rain}} = 108 \) drops s\(^{-1}\) m\(^{-2}\) and the average raindrop diameter of 5 mm, this water jet imparted the same damage as a rainfall of ~2 years.

Lubricant evaporation is among the critical factors that gradually remove lubricating material from SLIPs, causing more defect sites and increasing adhesion. Here, the liquid-repellent behaviors of the coated samples were examined for the effect of evaporation at a temperature of 65 °C at ambient pressure. Wetting parameters, including WCA, WSA, LCA and LSA, were traced for an 11-week period. All SHS and SLIMS samples displayed insignificant changes in water-repellent properties after aging at this temperature (figure 7). Their WCA and WSA values were nearly constant throughout the studied period. However, different outcomes were encountered when the surfaces were tested with natural latex liquid. After a few weeks of testing, the SHS sample
Figure 6. Traces of the changing SHS and SLIMS wetting properties show values for WCA (a), WSA (b), LCA (c) and LSA (d), after 10 min exposure to a water jet of various speeds.

Figure 7. The comparative evolution of SHS and SLIMS wetting parameters over 11 weeks of exposure to an elevated temperature of 65 °C is illustrated by plots of WCA (a), WSA (b), LCA (c) and LSA (d).
completely lost its ability to repel latex droplets due to the proliferation of defect sites during the test. SLIMS1 also exhibited similarly reduced wetting behavior for latex, where the LCA dropped substantially, accompanied by a sharp rise of LSA to 90°: characteristics of sticky surfaces. The low-vapor-pressure lubricant infused in the SLIMS1 sample had become more volatile in the warmer condition and the sample was unable to retain a stable lubricating layer. The newly generated defect sites could directly interact with the liquid latex. The resulting polymer adhesion and altered surface chemical composition quickly degraded the anti-wetting properties of this sample. On the other hand, being infused with lubricants with much lower vapor pressure, the SLIMS2 and SLIMS3 samples, showed excellent stability in the elevated temperature condition, although SLIMS2 showed a slight reduction in LCA after eight weeks. Generally associated with high viscosity, resistance to mass loss due to evaporation is a necessary property of a lubricant layer that requires a prolonged slippery and pinning-free lifetime.

The ability of liquid-repellent surfaces to withstand hydrostatic pressure is also a vital requirement for real applications such as anti-adhesive coatings for liquid storage containers. Here, the wetting-resistant properties of the samples were evaluated after immersion for 20 min in water under an applied static pressure. Distinguishable behaviors between the SHS and SLIMS samples were observed (figure 8). Large numbers of air bubbles were released from the SHS, but only a few were released from the SLIMS3. The conventional SHS failed to sustain the Cassie-Baxter anti-wetting state against static pressure as the water intruded into the surface cavities, displacing entrapped pockets of air. This degassing phenomenon did not occur in any SLIMS sample, since the surface cavities were already filled with incompressible lubricant.

Further detailed investigations related to the effects of static pressure on slippery behaviors were carried out. Again, due to the hierarchically rough textures, SHS could still resist external pressure to some extent. WCA was maintained above 150° and WSA below 10° on an SHS subjected to applied pressure up to ~80 kPa (figure 9). When the SHS was subjected to greater pressure, the wetting property was permanently fixed in the Wenzel state. However, the repellent properties of the SHS became less durable when tested with liquid latex. The LCA and LSA values fell more rapidly even with small increments of applied pressure and the liquid latex adhered to the SHS after the application of almost every level of pressure. SLIMS samples, on the other hand, showed more stable wetting behaviors for both water and latex, even subjected to a pressure of 150 kPa, which is equivalent to the hydrostatic pressure at a depth of ~15 m. This level of resistance is sufficient for general industrial applications. Although the SLIMS1 sample displayed better liquid-repellent properties at the initial time, it became less stable than SLIMS2 and SLIMS3 against liquid compression at pressures above ~60 kPa. Therefore, lubricants with higher viscosity are more able to withstand high static pressure.

Figure 10 illustrates the dynamics of latex-repellent behavior on a single substrate coated with SLIMS3 formulation and an SHS layer on different regions. The sample was submerged in static latex at a pressure of 60 kPa for 20 min and then tested for droplet motion. As displayed by sequential movie frames, a latex droplet slid easily across the SLIMS3 coating without leaving any trace behind. In contrast, some residual latex adhered to the substrate on the non-functionalized SHS coating.
4. Conclusions

We demonstrated fabrication strategies to produce slippery liquid-infused multiscale-textured surfaces that were highly robust against shear-induced damage, evaporation, and high static pressure. High-viscosity lubricants with extremely low evaporation rates were infused into particulate triple-scaled rough surfaces at warm temperature and allowed to cool, yielding a firm lubricant film wrapping over the surface topography. The
final films possessed some degree of slippery roughness, enhancing liquid-repellent properties that repelled even highly adhesive liquid latex. For the SLIPS3 samples infused with the most viscous lubricant, WCA of 130.1 ± 0.8° and LCA of 105.7 ± 1.1° as well as WSA of 5.8 ± 0.7° and LSA of 8.7 ± 0.7° were achieved. These samples also exhibited good resistance to abrasive flow as the wetting-resistant properties were unaffected by an impact of a water jet speed up to 2.6 ms⁻¹ for 10 min duration. A viscous lubricant layer was also nonvolatile, preventing lubricant loss from evaporation at 65 °C for at least 11 weeks of study. Furthermore, the SLIPS3 samples can maintain their wetting-resistance properties against external pressure of 150 kPa for 20 min. In general, SLIMS3 showed superior long-term repellent properties to SLIM2S, SLIM2 and SHS respectively.

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