Fluorine-free Slippery Liquid-infused Porous Surfaces Prepared Using Hierarchically Porous Aluminum

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Abstract

Slippery liquid infused porous surfaces (SLIPS) have been receiving increased attention. Fluorinated compounds have been used to prepare the SLIPS, but because of bioaccumulation and ecological impact of perfluoalkyl building blocks, fluorine-free SLIPS system is preferable for practical applications. In this study, SLIPS has been prepared from hierarchically porous aluminum substrate, tetradecylphosphonic acid monolayer coating and silicone lubricant oil. The several properties of the SLIPS, including durability against lubricant loss, ice adhesion and corrosion resistance, have been compared with those with a fluoroalkyl monolayer coating and a fluorinated lubricant oil. Findings in this study demonstrate that the fluorine-free SLIPS shows higher durability against the removal of lubricant in air and also in water. Both fluorinated and fluorine-free SLIPS samples exhibit extremely reduced ice adhesion compared with superhydrophobic surfaces obtained using the same hierarchically porous aluminum substrate with fluorinated or fluorine-free monolayer coating. The SLIPS samples have very high corrosion resistance in NaCl solution, but not in alkaline solution. The solution-dependent corrosion resistance is discussed in terms of the stability of monolayer coating.

1. Introduction

Inspired by the *Nepenthes* pitcher plant,[1] slippery liquid-infused porous surfaces (SLIPS) were first reported in 2011.[2] The SLIPS materials consist of low-surface-energy porous materials filled with a lubricant liquid and exhibit repellency to various liquids. In contrast to superhydrophobic surfaces that show water contact angles greater than 150º, relatively low contact angles less than 120º is revealed for various liquids on SLIPS. However, both SLIPS and superhydrophobic surfaces have low sliding angle of liquids of only a few degrees. Thus, SLIPS materials have attracted increased attention as self-cleaning, anti-fouling, anti-icing and corrosion-resistant surfaces. For instance, corrosion of metallic materials in marine industry environments, in which microbial fouling often accelerates corrosion, will be effectively suppressed by fabricating SLIPS surface.[3, 4] Icephobic surfaces that can repel ice or prevent ice formation are of importance in transportation and infrastructure, including aircraft wings, power transmission lines and air conditioning systems. It was demonstrated that the SLIPS is also icephobic.[5] Superhydrophobic surfaces with micro-/nano-textured surface morphology and low surface energy coatings possess several drawbacks including repellency only to liquids with high surface tensions (wetting to low-surface-tension-oils), low mechanical stability, irreversible Cassie-to-Wenzel transition.[6] These drawbacks can be overcome when SLIPS materials are used instead of superhydrophobic surfaces, although it is often pointed out that lubricant oil retention is a weakness of SLIPS.[7] Several methods to improve the lubricant liquid retention have also been reported recently.[8]

The most reported SLIPS use fluorinated materials or coatings for porous substrates and fluorinated lubricant liquids, such as DuPont Krytox oils and 3M Fluorinert FC-70.[9] These fluorinated compounds are expensive so that cheaper alternatives are awaited. In addition, bioaccumulation of perfluoroalkyl acids needs to be considered for practical application of fluorinated SLIPS.[10] Fluoroalkyl monolayers are often coated on porous substrates to form SLIPS and also superhydrophobic surfaces to reduce the surface free energy. However, because of economic and ecological drawbacks of fluorinated compounds, fluorine-free, const-effective and environmentally-friendly alternatives are demanded for industrial applications of SLIPS.

The formation of highly porous substrates by industrially acceptable processes is also important for practical applications of SLIPS. Porous aluminum surfaces have been prepared to fabricate SLIPS. [4, 8, 11, 12] Anodizing of aluminum and its alloys to form nanoporous alumina films with numbers of cylindrical nanopore channels, which is normal to the surface,

is one of the suitable ways to form porous aluminum substrates. Hierarchically porous aluminum surfaces that exhibit superhydrophobicity and also superoleophobicity were developed by a combination of chemical etching and subsequent anodizing of aluminum substrate, followed by organic monolayer coating.[13] The chemical etching introduced micropits of half-cubic shape on the entire aluminum surface and anodizing forms nanoporous anodic alumina film with the pore size and porosity being controlled by a post-pore-widening process, which is also a simple immersion process in phosphoric acid. The hierarchically porous surfaces with fluoroalkyl monolayer coating are superoleophobic even for low surface tension liquids. [13, 14] By utilizing the nanopores as reservoir of an organic healing agent, repeatable self-healing of superoleophobicity is also demonstrated.[15] Thus, the hierarchically porous aluminum substrate thus prepared will be suitable also for SLIPS.

Aluminum SLIPS materials have been prepared using porous anodic alumina [8, 16] and chemical etching and hydrothermal treatment. [12] In these SLIPS, fluorinated compounds were used for a thin organic coating and lubricant liquids. In the present study, SLIPS were prepared using a hierarchically porous aluminum substrate with a fluorine-free alkylphosphonic acid monolayer coating and silicone oil. The lubricant retention in air and in water, ice adhesion and corrosion resistance were examined and compared with fluorinated SLIPS prepared from the same porous aluminum substrate. The results obtained indicated superior properties of fluorine-free SLIPS (F-free SLIPS) comparable to or even better than those of Fluorinated SLUPS (F-SLIPS).

2. Experimental Section

2.1. SLIPS preparation

An aluminum sheet (Nilaco, Japan) of 99.5% purity and 0.4 mm thickness was used in this study. After degreasing in acetone, the aluminum sheet was alkaline etched in 1 mol dm^{-3} NaOH solution at 60ºC for 120 s, followed by desmutting treatment in 1 mol dm^{-3} HNO_3 solution at 60ºC for 180 s. Chemical etching of the surface area of 30 x 15 mm^2 was performed in a mixed solution containing 0.2 mol dm^{-3} $CuCl_2$ and 1 mol dm^{-3} HCl at room temperature for 360 s. After chemical etching, deposited copper layer on aluminum was dissolved in concentrated HNO_3 solution. The subsequent anodizing was conducted at 25 V in 0.3 mol dm^{-3} H_2SO_4 electrolyte at 15ºC for 180 s to form a nanoporous anodic alumina film on entire etched aluminum surface. A two-electrode cell with aluminum counter electrode was used for anodizing. Then, the pore diameter was controlled to ~30 nm by immersing the anodized aluminum in 5 wt% H_3PO_4 solution at 30ºC for 1.2 ks.

Organic monolayer coatings were formed by immersing the porous aluminum substrate in ethanol solution containing either 1 mmol dm^{-3} tetradecylphosphonic acid (TDP) or 1 mmol dm^{-3} 1H, 1H, 2H, 2H perfluorooctanephosphonic acid (FOPA) for 2 days. Prior to the coatings, the porous aluminum substrate was treated with oxygen plasma (Harrik Plasma, PDC-32G plasma cleaner) for 320 s to remove a contaminant hydrocarbon surface layer and to introduce surface OH group. The lubricant liquids used for F-free SLIPS, in which TDP was utilized as an organic coating, were three types of Shin-Etsu Chemical silicone oils. For F-SLIPS with a FOPA coating, three types of fluorinated DuPont Krytox oils were used. The lubricant liquids of 100 μL were dropped onto the sample surface, and then spin-casted at 500 rpm for 60 s to get uniform lubricant liquid layer. The physical properties of the lubricant liquids used in this study were listed in **Table 1**.

Table 1. Viscosity, surface tension and pour point of the lubricant liquids used in this study.

Type	Lubricant liquid	Viscosity at 25ºC (cSt)	Surface tension at 25ºC (mN m^{-1})	Pour point (ºC)
F-free SLIPS	Si-20	20	20.8	-73
	Si-50	50	20.8	-50
	Si-100	100	20.0	-50
F-SLIPS	F-12	12	15.5	-70
	F-82	82	17.0	-60
	F-177	177	17.7	-51

2.2. Characterizations

2.2.1. Surface observations

Surfaces of aluminum sample after chemical etching, anodizing and pore widening were observed using a ZEISS, Sigma-500 field emission scanning electron microscope operated at a low accelerating voltage of 1.5 kV. Because of the use of low accelerating voltage, the as-formed sample was observed without any conductive coating.

2.2.2. Surface wettability

Static and dynamic contact angles for water, rapeseed oil and ethanol were measured using a Kyowa Interface Science, DM-C1 contact angle meter. The static contact angles were obtained with a droplet volume of 4 μL. The dynamic contact angles were measured by an extension/contraction method. The results obtained were analyzed using a FAMAS software. The sliding angles were measured by tilting a sample with a water droplet of 10 μL. The measurements were repeated at least 5 times to get reliable contact angles and sliding angles.

2.2.3. Ice adhesion

The ice adhesion was evaluated by measuring the shear stress between the SLIPS sample and ice, which was directly formed on the SLIPS samples. The SLIPS sample was placed on a Peltier cooling stage of -20ºC and ice block of 10 mm diameter was directly developed on the SLIPS sample by cooling. Then, the shear stress was measured using an Imada, ZTS-50N force gauge. For comparison, the ice adhesion of as-received aluminum sheet and superhydrophobic aluminum without lubricant liquid were also evaluated.

2.2.4. Corrosion resistance

Corrosion resistance of the SLIPS samples were examined by electrochemical potentiodynamic polarization measurements. A three-electrode electrochemical cell with a Pt counter electrode and an Ag/AgCl reference electrode was used. The potential was swept using an Ivium Technologies, Compactstat potentiostat/galvanostat system from approximately -200 mV from the corrosion potential toward anodic direction with a sweep rate of 1 mV s^{-1} after measuring the corrosion potential for 1.2 ks.

3. Results and Discussion

3.1. Characterization and durability

Figure 1 shows the scanning electron micrographs of the hierarchically porous aluminum substrate, which was formed by chemical etching, anodizing and pore-widening processes. Chemical etching in HCl + $CuCl_2$ solution developed many micro-pits, as shown in Figure 1a. The micro-pits of aluminum are formed by faceted dissolution of (100) aluminum plane; in Figure 1a, the exposed crystallographic plane in the micro-pits should be (100). [17] The size of the micro-pits is roughly 1-2 μm under the present condition. The smaller pits are formed when the HCl concentration is increased in the etching solution. [13] The higher magnification scanning electron micrograph (Figure 1b) discloses the formation of nanoporous anodic alumina on the entire surface of etched aluminum. The average size of nanopores after pore widening is ~30 nm. Such porous surface morphology is uniformly formed on the entire aluminum surface and the size of pits and nanopores were highly reproducible under the present condition. The hierarchically micro-/nano-porous surface thus obtained is superhydrophobic and superoleophobic, as reported previously. [13, 15]

The surface wettability of the specimens was evaluated by dynamic water contact angle measurements (**Figure 2**). Figure 2a and 2b are advancing and receding water contact angles on the TDP-coated porous aluminum surface (F-free SHS), respectively. Both angles exceed

150º and the contact angle hysteresis, which is the difference between the advancing and receding angles, is only 4.3º. Thus, the surface is superhydrophobic. Slightly higher water contact angles are obtained on the FOPA-coated porous aluminum surface (F-free SHS), because of the lower surface energy of the fluoroalkyl surface (~6 mN m^{-1}) compared with the alkyl surface (~24 mN m^{-1}) (Figure 2c and 2d). Both the F-free and F-SHS have the low contact angle hysteresis, which is also one of the important criteria of superhydrophobicity. In fact, water droplet was readily rolled off when the specimen was slightly tilted.

The dynamic water contact angles on the lubricant-infused porous aluminum surfaces were also examined. The F-free SLIPS (TDP coating with silicone lubricant, Si-100) shows highly reduced dynamic water contact angles, but the contact angle hysteresis is still less than 5º (Figure 2e and 2f). Similar water contact angles were also obtained when other silicone oil lubricants listed in Table 1 were used. The dynamic water contact angles were increased to ~120º on the F-SLIPS (FOPA coating with fluorinated lubricant, F-12), but the contact angle hysteresis is also as low as ~3º (Figure 2g and 2h). In correlated with the low contact angle hysteresis, the water droplet on the SLIPS specimens was readily slipped on the slightly tilted surfaces, as demonstrated in the supporting information (Movie S1 to S3).

The lubricant retention properties of F-free and F-SLIPS were examined in air and in water. The SLIPS samples were rotated at several spin rates using spin casting facilities, and the change in the water sliding angle was measured (**Figure 3**). The water sliding angle at 1000 rpm is as low as ~2º regardless of lubricant liquid, indicating that all the SLIPS samples examined in this study present excellent slippery properties when enough lubricant liquid is remained on the surface. The sliding angle increases gradually as the spin rate increases and the rise is suppressed when the viscosity of lubricant is high for both F-SLIPS and F-free SLIPS (Figure 3a). However, the F-free SLIPS samples show lower sliding angles at high spin rates compared with the F-SLIPS, although the remaining amount of lubricant liquid is similar for both F-and F-free SLIPS at the same spin rate. Thus, F-free SLIPS samples appears to be better to keep low sliding angles at reduced lubricant amounts (Figure 3c). The reason for the better slippery properties of F-free SLIPS at low oil retention conditions is not clear and the subject of future study. From Figure 3c, it is obvious that the sliding angle is mainly controlled by the amount of remaining lubricant liquid regardless of viscosity of liquid when either F- or F-free lubricants are used.

Figure 4 shows the change in the water sliding angle measured after immersing the SLIPS samples in stirred water for various periods of time. Again, lower sliding angle is sustained for lubricant liquids with higher viscosities for both F- and F-free SLIPS samples. It is also

obvious that the F-free SLIPS samples sustain the lower sliding angles compared with the F-SLIPS. Since the water and silicone or fluorinated lubricant liquids used in this study are immiscible, one of the lubricant removal mechanisms from the porous surface is cloaking. [18] Whether cloaking of water droplets by a lubricant liquid occurs or not is determined by spreading coefficient, S_{ol}, of the lubricant liquid on water droplet and the value is given by the following equation:

$$S_{ol} = \gamma_{l} - \gamma_{o} - \gamma_{ol} \qquad (1)$$

where γ_{l}, γ_{o} and γ_{ol} are the water and lubricant surface tensions and the water-lubricant interfacial tension, respectively. Cloaking of lubricant occurs under the condition of the spreading coefficient $S_{ol} > 0$. Thus, the removal of lubricant liquids by cloaking can be suppressed by selecting the lubricant liquids showing $S_{ol} < 0$. However, both fluorinated lubricant liquids and silicone oils have positive spreading coefficients. [18] Thus, the better lubricant retention of the silicone oils compared with the fluorinated lubricant liquids may not be explained from the cloaking properties. The controlling factors of lubricant retention are not fully understood yet, and the similarity of the surface tension of lubricant liquid and surface energy of the porous solid surface, the amount of lubricant dissolution into water (even though small) and emulsion formation characteristics between water and lubricant may also influence the lubricant retention. Further detailed studies are awaited for a better understanding the durability of SLIPS. Hereafter, the most durable Si-100 and F-177 lubricant liquids were used for F-free and F-SLIPS, respectively, for further examination of SLIPS properties.

3.2. Ice adhesion

The ice adhesion on the SLIPS samples and superhydrophobic surfaces was examined at -20ºC and the results are summarized in **Table 2**. Rather high adhesion strength of 2.1 kPa was obtained on the as-received aluminum substrate with relatively smooth surface. Under this condition, further increased ice adhesion was found on the superhydrophobic surfaces of both TDP-coated hierarchically porous aluminum (F-free SHS) and FOPA-coated one (F-SHS). Ice should be formed within the micro-pits and nanopores from vapor phase at -20ºC in addition to the outer surface of the samples. Because of the anchoring effect, the ice adhesion strength should be enhanced under the present condition. It is often reported that superhydrophobic surfaces are icephobic. [19-22] The present study discloses that the superhydrophobic surfaces are not always icephobic, being probably dependent upon the environmental conditions. In

contrast, the F-SLIPS and F-Free SLIPS exhibit significantly reduced ice adhesion. Both the F-and F-free SLIPS samples have the ice adhesion as low as 20-30 kPa, being two orders of magnitude lower than those of superhydrophobic surfaces.

Table 2. Ice adhesion strength on several aluminum samples at -20ºC.

Sample	As-received Al	F-free SHS	F-SHS	F-free SLIPS	F-SLIPS
Adhesion (kPa)	2.1×10^3	2.8×10^3	2.6×10^3	20	29

However, the ice adhesion increased gradually when the ice adhesion measurements were repeated (**Figure 5**). After each adhesion measurement, sliding angle of water droplet was also measured (Figure 5b). The sliding angle also increases with the measurement cycle. After the fifth cycle, lubricant liquid was impregnated again, and then, the ice adhesion and sliding angle were measured. As shown in Figure 5, both the ice adhesion and sliding angle are almost recovered to the initial values. The gradual increase in the ice adhesion and sliding angle may be associated with the depletion of lubricant, not the serious mechanical damage of the porous solid surface with organic coating. The introduction of self-replenishing lubrication methods will be able to extend the durability of icephobic SLIPS. [23, 24]

3.3. Corrosion resistance

The corrosion behavior of the SLIPS samples was examined by potentiodynamic polarization curves in NaCl, HCl and KOH aqueous solutions (**Figure 6**). The polarization curves of superhydrophobic aluminum samples were also measured for comparison. Figure 6a shows the polarization curves in 0.5 mol dm^{-3} NaCl solution. The anodic polarization of the as-received aluminum shows steep increase in anodic current to ~100 mA cm^{-3} because of pitting corrosion. The anodic current density and corrosion current density are highly suppressed for the F-free and F-SHS samples. Thus, superhydrophobicity is useful to suppress the corrosion in this corrosive environment. Further reduced corrosion current density and anodic current density are observed for the SLIPS samples. In particular, the anodic current density of the F-free SLIPS is as low as the order of 10^{-10} A cm^{-2}. The fluctuation of the anodic current of the F-SLIPS may be associated with the local film breakdown and repassivation. It is likely that fluorinated lubricant liquid on F-SLIPS is not completely filled within the pores, causing the partial breakdown.

Similar trend was found in 0.1 mol dm^{-3} HCl solution. The corrosion current density decreased in the following order: as-received > superhydrophobic > SLIPS. Thus, SLIPS

samples show better corrosion protection performance, compared with the superhydrophobic surfaces, although the anodic current density was fluctuated for both F-free and F-SLIPS in this more aggressive corrosion environment. Wu et al. compared the lubricant oil penetration by vacuum and air-pressure impregnation into porous anodic alumina nanochannels and the impregnation was evaluated using cryo-SEM technique.[25] They clarified better filling of nanopore channels with lubricant oil by the vacuum impregnation. In the present study, air-pressure impregnation was used to form a lubricant layer, resulting in the presence of insufficiently impregnated regions in porous layer. Thus, in aggressive HCl solution partial breakdown was induced during anodic polarization. The fluctuation of anodic current for the F-SLIPS sample in milder NaCl solution suggests the presence of higher degree of insufficient lubricant filling regions in the F-SLIPS compared with the F-free SLIPS.

Table 3 compares the corrosion current density of various SLIPS samples formed on aluminum substrate in NaCl and HCl solutions. The corrosion current density of the present F-free SLIPS in both solutions is comparable to the previous F-containing SLIPS and exhibits sufficiently low corrosion current density. Thus, the F-free SLIPS samples with alkyl monolayer coating and silicone lubricant are promising as corrosion-resistant layers.

Table 3. Comparison of the corrosion currents of various SLIPS samples formed on aluminum in NaCl and HCl solutions.

Coating/lubricant	Corrosive solution	Corrosion current (A cm^{-2})	Reference
TDP/silicone	0.5 mol dm^{-3} NaCl 1 mol dm^{-3} HCl	~1 x 10^{-10} ~1 x 10^{-9}	This study
FAS/Krytox100	3.5 wt% NaCl	~10^{-10}	[26]
FAS/silicone oil	3.5 wt% NaCl	~10^{-11}	[11]
FAS/Krytox100	3.5 wt% NaCl	~10^{-7}	[12]
Teflon/Krytox100	1 mol dm^{-3} HCl	6.25 x 10^{-9}	[16]
FAS/mineral oil	1 mol dm^{-3} NaCl	~3.0 x 10^{-10}	[25]

Different behavior was observed in 0.1 mol dm^{-3} KOH solution, in which all the samples show rather high corrosion current density and anodic current density. No remarkable suppression of corrosion was found for both superhydrophobic and SLIPS samples. Poor corrosion protection ability in KOH solution may be associated with the stability of organic

monolayer coating. It was reported that alkyl phosphonic acid monolayer coating exhibited long-term stability in acid and neutral aqueous solutions, but the degradation occurred in alkaline solution. [27] In fact, when a droplet of NaCl or HCl solution is placed on SLIPS samples, the sliding angles were low, being less than 3º, but the sliding angles of KOH solution droplet were more than 20º. The alkylphosphonic acid or fluoroalkylphosphonic acid monolayer coating may not be stable in KOH solution even under the presence of lubricant liquid. Thus, the aggressive solution penetrates through the lubricant layer, attacking the aluminum substrate. To improve the corrosion resistance of aluminum by SLIPS, more stable organic coatings in alkaline solution needs to be used.

4. Conclusions

Environmentally friendly fluorine-free SLIPS (F-free SLIPS) samples are prepared using hierarchically porous aluminum substrate, alkylphosphonic acid monolayer coating and silicone oils, and their lubricant retention properties, ice adhesion and corrosion protection properties are compared with fluorinated SLIPS (F-SLIPS) samples. Lower sliding angles are maintained for F-free SLIPS even if the remaining lubricant liquid on the porous substrate is reduced. Better stability of F-free SLIPS is also found in water. The ice adhesion and corrosion resistance of the F-free and F-SLIPS samples are superior compared with the superhydrophobic samples. Thus, the fluorine-free SLIPS are promising for many practical applications for anti-fouling, self-cleaning, icephobic and corrosion protection.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

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References

[1] H. F. Bohn, W. Federle *Proc. Natl. Acad. Sci. USA.* **2004**, *101*, 14138-14143.

[2] T. S. Wong, S. H. Kang, S. K. Y. Tang, E. J. Smythe, B. D. Hatton, A. Grinthal, J. Aizenberg *Nature*. **2011**, *477*, 443-447.

[3] Z. Shi, Y. Xiao, R. Qiu, S. Niu, P. Wang *Surf. Coat. Technol.* **2017**, *330*, 102-112.

[4] P. Wang, Z. Lu, D. Zhang *Corros. Sci.* **2015**, *93*, 159-166.

[5] P. Kim, T.-S. Wong, J. Alvarenga, M. J. Kreder, W. E. Adorno-Martinez, J. Aizenberg *ACS Nano*. **2012**, *6*, 6569-6577.

[6] J. S. Li, E. Ueda, D. Paulssen, P. A. Levkin *Adv. Func. Mater.* **2019**, *29*, 1802317.

[7] M. Tonelli, S. Peppou-Chapman, F. Ridi, C. Neto *J. Phys. Chem. C*. **2019**, *123*, 2987-2995.

[8] J. Lee, Y. Jiang, F. Hizal, G. H. Ban, S. Jun, C. H. Choi *J. Colloid Interface Sci.* **2019**, *553*, 734-745.

[9] M. Villegas, Y. X. Zhang, N. Abu Jarad, L. Soleymani, T. F. Didar *ACS Nano*. **2019**, *13*, 8517-8536.

[10] F. M. Hekster, R. W. P. M. Laane, P. de Voogt in *Environmental and Toxicity Effects of Perfluoroalkylated Substances, Vol.*, Springer New York, New York, NY, **2003**, pp.99-121.

[11] J. Sun, C. Wang, J. L. Song, L. Huang, Y. K. Sun, Z. A. Liu, C. L. Zhao, Y. X. Li *J. Mater. Sci.* **2018**, *53*, 16099-16109.

[12] Y. J. Tuo, H. F. Zhang, W. P. Chen, X. W. Liu *Appl. Surf. Sci.* **2017**, *423*, 365-374.

[13] K. Nakayama, E. Tsuji, Y. Aoki, H. Habazaki *RSC Adv.* **2014**, *4*, 30927-30933.

[14] K. Nakayama, E. Tsuji, Y. Aoki, S.-G. Park, H. Habazaki *J. Phys. Chem. C*. **2016**, *120*, 15684-15690.

[15] K. Nakayama, A. Koyama, C. Zhu, Y. Aoki, H. Habazaki *Adv. Mater. Interfaces*. **2018**, *5*, 1800566.

[16] J. Lee, S. Shin, Y. H. Jiang, C. Jeong, H. A. Stone, C. H. Choi *Adv. Func. Mater.* **2017**, *27*.

[17] S. Ono, T. Makino, R. S. Alwitt *J. Electrochem. Soc.* **2005**, *152*, B39-B44.

[18] S. Sett, X. Yan, G. Barac, L. W. Bolton, N. Miljkovic *ACS Appl. Mater. Interfaces*. **2017**, *9*, 36400-36408.

[19] K. Golovin, S. P. R. Kobaku, D. H. Lee, E. T. DiLoreto, J. M. Mabry, A. Tuteja *Sci. Adv.* **2016**, *2*, 12.

[20] J. Y. Lv, Y. L. Song, L. Jiang, J. J. Wang *ACS Nano*. **2014**, *8*, 3152-3169.

[21] L. B. Boinovich, A. M. Emelyanenko, V. K. Ivanov, A. S. Pashinin *ACS Appl. Mater. Interfaces*. **2013**, *5*, 2549-2554.

[22] D. K. Sarkar, M. Farzaneh *J. Adhesion Sci. Technol.* **2009**, *23*, 1215-1237.

[23] H. X. Zhao, Q. Q. Sun, X. Deng, J. X. Cui *Adv. Mater.* **2018**, *30*, 1802141.

[24] C. Howell, T. L. Vu, J. J. Lin, S. Kolle, N. Juthani, E. Watson, J. C. Weaver, J. Alvarenga, J. Aizenberg *ACS Appl. Mater. Interfaces*. **2014**, *6*, 13299-13307.

[25] D. Wu, D. Zhang, Y. Ye, L. Ma, B. Minhas, B. Liu, H. A. Terryn, J. M. C. Mol, X. Li *Chemical Engineering Journal*. **2019**, *368*, 138-147.

[26] M. L. Zhang, R. R. Chen, Q. Liu, J. Y. Liu, J. Yu, D. L. Song, P. L. Liu, L. T. Gao, J. Wang *ChemElectroChem*. **2019**, *6*, 3911-3919.

[27] M. Kosian, M. M. J. Smulders, H. Zuilhof *Langmuir*. **2016**, *32*, 1047-1057.

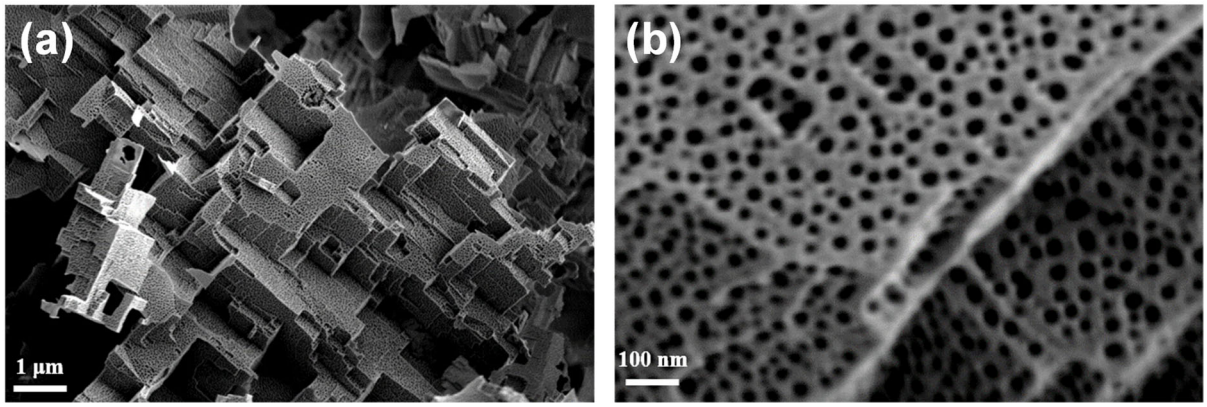

Figure 1. Scanning electron micrographs of aluminum sheet after chemical etching, anodizing and pore widening: (a) low and (b) high magnifications.

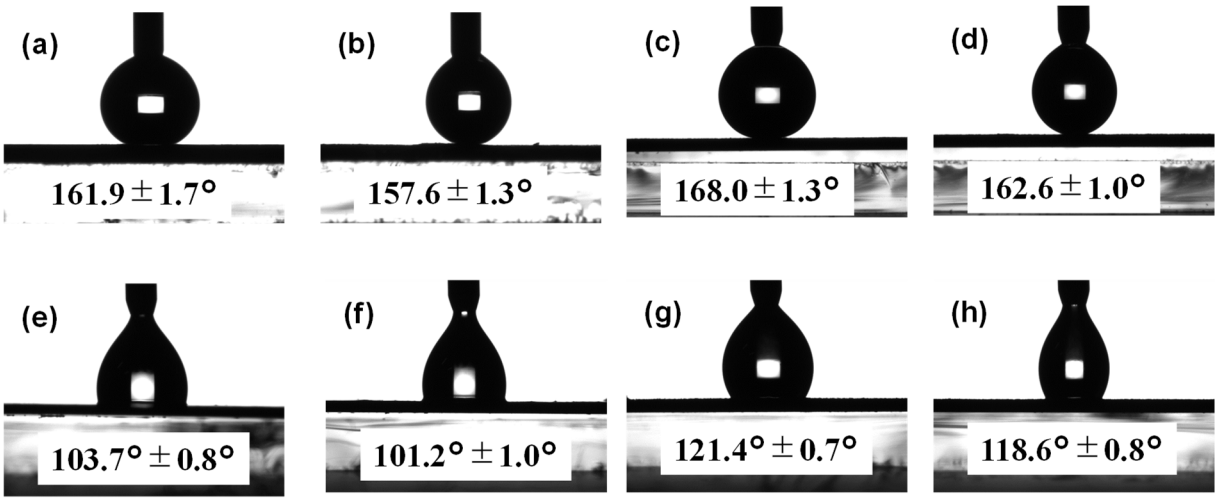

Figure 2. Dynamic water contact angles of (a, b) F-free SHS, (c, d) F-SHS, (e, f) F-free SLIPS and (g, h) F-SLIPS: (a, c, e, g) advancing and (b, d, f, h) receding contact angles.

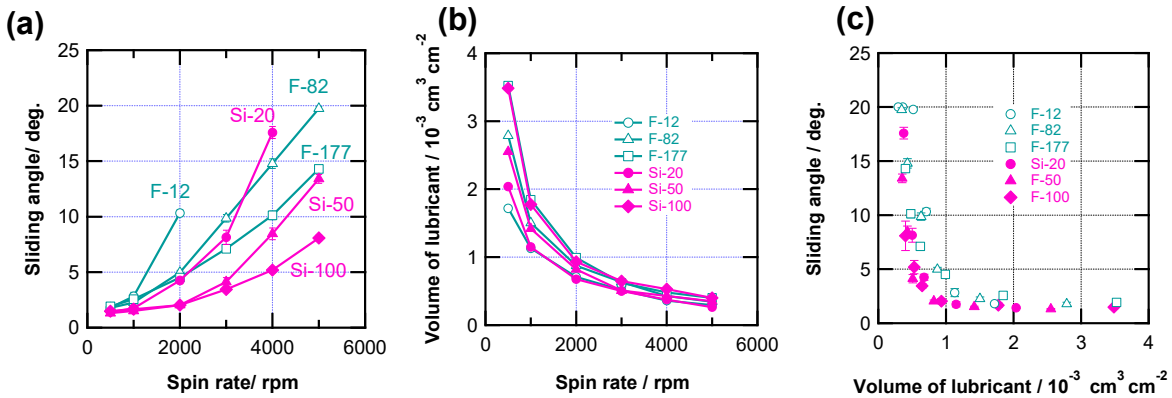

Figure 3. Changes in (a) water sliding angle and (b) lubricant volume with spin rate of SLIPS samples and (c) correlation between the sliding angle and lubricant volume.

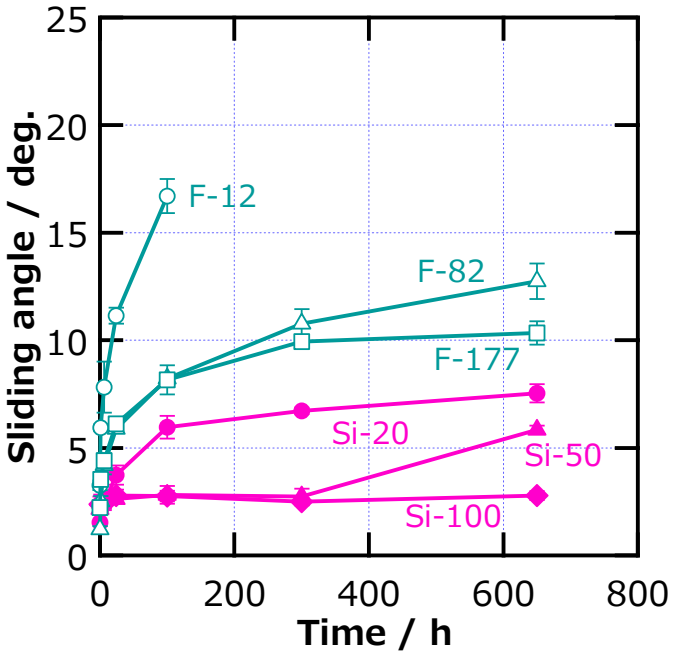

Figure 4. Change in the water sliding angle with immersion time of SLIPS samples in stirred.

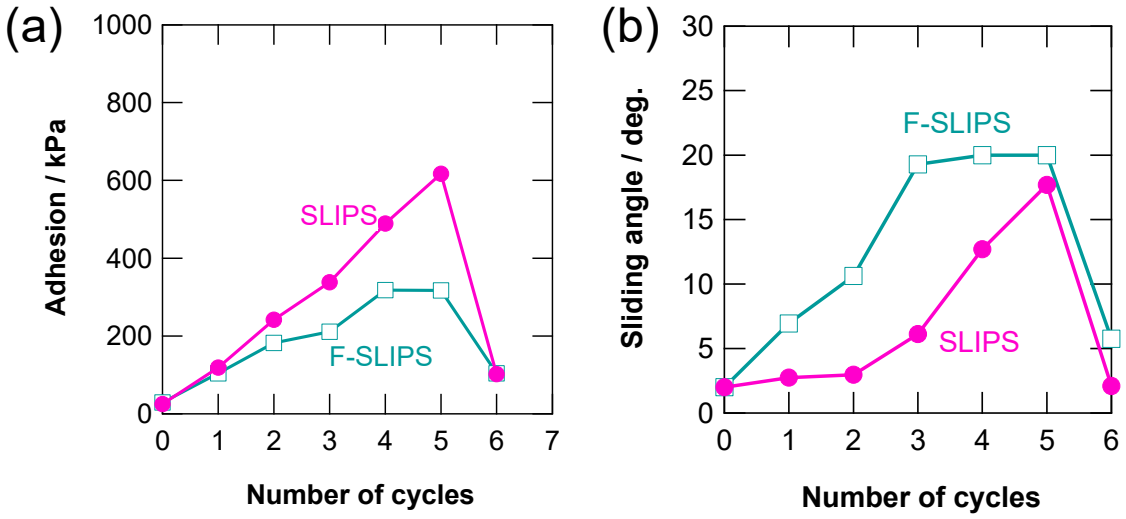

Figure 5. Changes in (a) ice adhesion strength and (b) water sliding angle with the measurement cycle. After the fifth measurements, lubricant liquid was impregnated again.

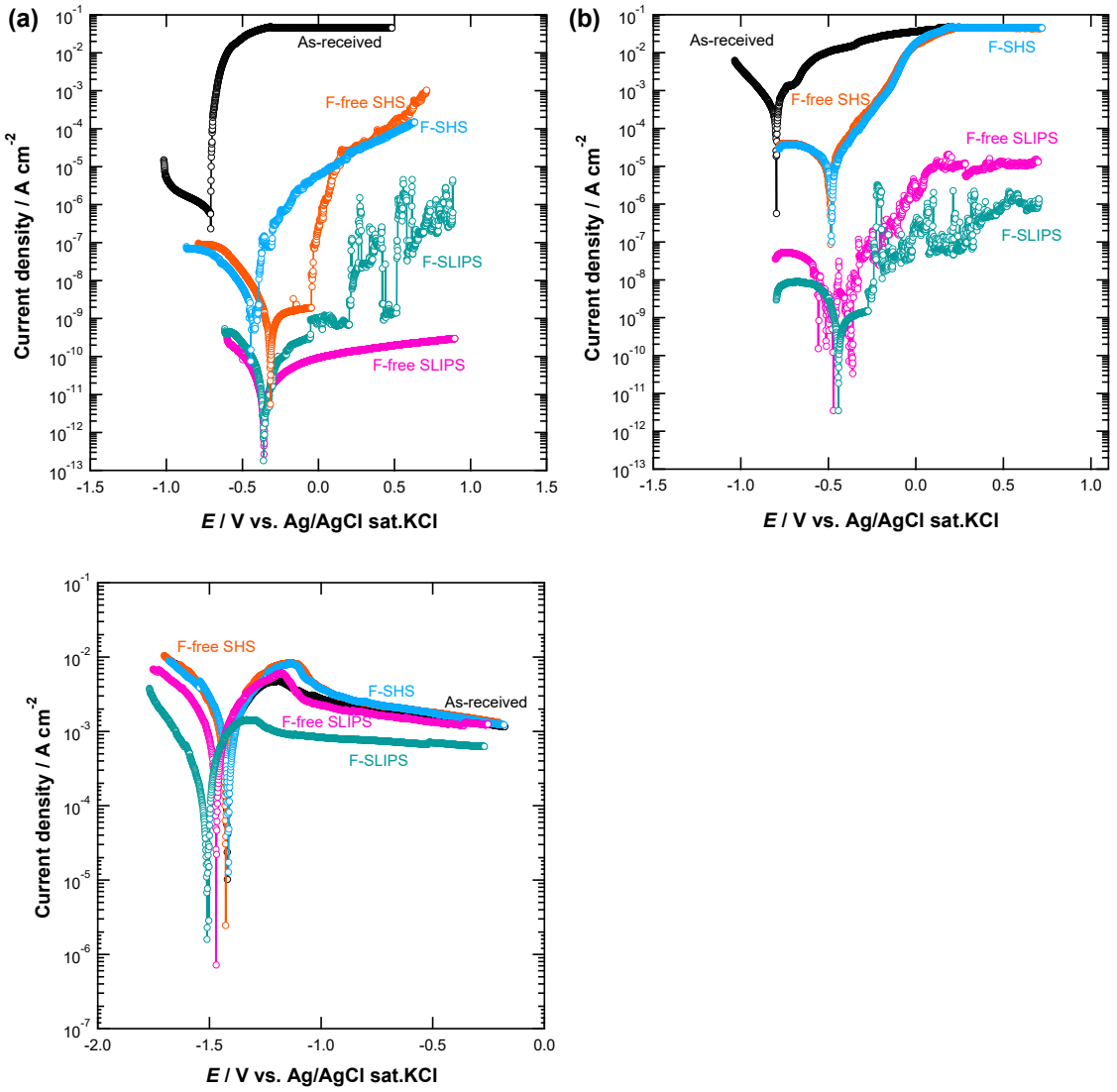

Figure 6. Potentiodynamic polarization curves of the as-received, F-free and F-SHS and F-free and F-SLIPS aluminum samples in (a) 0.5 mol dm^{-3} NaCl, (b) 0.1 mol dm^{-3} HCl and (c) 0.1 mol dm^{-3} KOH solutions.

Supporting information

Movie S1. Water droplet on the as-received aluminum.

Movie S2. Water droplet on the F-free SHS.

Movie S3. Water droplet on the F-free SLIPS.

Table of Contents

Fluorine-free slippery liquid infused porous aluminum surfaces with silicone oil lubricants are prepared and the lubricant retention, ice adhesion and corrosion behavior are compared with those of the fluorinated counterparts. The low ice adhesion strength and high corrosion resistance in HCl and NaCl aqueous solutions of the fluorine-free aluminum samples are revealed in this study. The better lubricant retention of the fluorine-free samples compared with the fluorinated ones is also found in this study.