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Liquid-like Slippery Surfaces via Ultrasound Activation and Grafting of Polydimethylsiloxane

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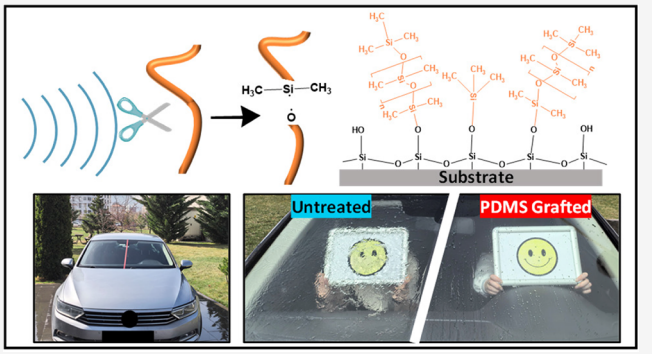
ABSTRACT: Slippery molecular coatings that dynamically repel polar and nonpolar liquids are in urgent need for a diverse range of applications. Nanoscopic films of grafted polydimethylsiloxane (PDMS) have particularly attracted significant attention for their liquid-like slipperiness, which arises from their flexible chains and low surface energy. Herein, we present ultrasound-assisted activation and grafting of methyl-terminated PDMS for generating fluorine-free omniphobic coatings on silicon oxide-terminated surfaces. Intense sound waves generated by an ultrasonic homogenizer induce the activation of unreactive and inert PDMS, allowing ambient grafting in less than an hour. The grafted PDMS films with a thickness of ∼4 nm exhibit sliding angles below 17° for liquids with surface tensions ranging from 20 to 73 mN/m. The sonochemical activation of PDMS under outdoor conditions and its subsequent deposition over square meters of surfaces demonstrate the practical potential of the proposed method. The ultrasound activation enables the versatile preparation of liquid-like slippery molecular coatings over large areas using inexpensive materials and ambient processes.



INTRODUCTION

There is strong demand for nonstick molecular coatings for a wide range of industrial and everyday applications. Liquid-like surfaces have received significant attention due to their slippery nature when in contact with a variety of liquids and substances. These surfaces are often referred to as omniphobic, because they exhibit low adhesion and easy sliding of polar and nonpolar liquids at low tilting angles. These slippery surfaces are typically composed of nanoscopic films prepared by grafting highly flexible polymers onto smooth solid substrates.[1−9] The flexibility of the chains ensures high mobility at room temperature and liquids, regardless of their surface tension, are dynamically repelled on such surfaces.[10] Overcoming disadvantages of fragile nanostructures and opacity of superhydrophobic surfaces and liquid depletion of liquid-infused surfaces, these liquid repellent molecular coatings are emerging for real-world applications with their self-cleaning,[11] antifouling,[12,13] anti-icing,[14] and antifogging characteristics.[15]

Omniphobic liquid-like surfaces rely on the free rotational ability of low surface energy chains, the smoothness and homogeneity of the surface to repel liquid droplets from the surface without the need for air pockets and lubricant infusion. Smoothness and flexibility are critical parameters affecting the behavior of omniphobic liquid-like surfaces. Smoothness facilitates low contact angle hysteresis by reducing friction between the liquid drop and the surface. Contact angle hysteresis is the difference between advancing and receding contact angles and characterizes the resistance to initiate liquid droplet to movement on surfaces.[16] The high mobility of the polymer chains reduces contact angle hysteresis for both polar and nonpolar liquids and diminishes the resistance to sliding off the surface. In this regard, grafted films of polydimethylsiloxane (PDMS), also called silicone oil, has particularly attracted significant attention due to their liquid-like slipperiness arising from their flexible chains and low surface energy. Furthermore, the strict regulations on fluorinated compounds due to their bioaccumulation and toxicity issues compel researchers to develop omniphobic coatings using fluorine-free and biocompatible molecules like PDMS.[17,18] The deposition of PDMS onto surfaces using various techniques has been widely reported in the literature for the fabrication of omniphobic coatings with high liquid repellency, low contact angle hysteresis, and diverse functional properties.[19−24]

The significant potential of omniphobic liquid-like surfaces based on grafted films of PDMS has led to the development of

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numerous preparation methods, which can be divided into two broad categories of grafting from and grafting to.[25,26] In the former approach, monomers of reactive alkylsilanes such as dichlorodimethylsilane[2,27−30] and dimethyldimethoxysilane[31] react with residual water and surface silanol groups, go through condensation/polymerization reaction, resulting in surface-grafted PDMS chains. In the "grafting to" method, presynthesized polymers either with functional groups or with unreactive methyl groups can be chemically grafted to substrate with surface oxide groups. In an interesting study, Teisala et al. demonstrated grafting of unreactive, methyl-terminated PDMS on a silicon wafer at room temperature, achieving a grafted PDMS layer with an average thickness of 3.1 nm. The surface exhibited omniphobic properties, exhibiting a sliding angle of 12° for water and 1° for toluene.[8] The grafting of methyl-terminated PDMS at room temperature typically requires long deposition times, which limit its practical applications.[32] To address this limitation, we previously introduced a mechanochemical grafting approach using ball milling, enabling the grafting of unreactive PDMS at room temperature within 1 h.[9] This study highlights the promise of exploring other means of mechanochemical processes to prepare omniphobic surfaces. Sonochemical approaches particularly stand out as effective and practical techniques.[33] At the heart of the sonochemical process is acoustic cavitation, which involves formation of a bubble (hot spot) in a liquid, growth of its maximum size and subsequent collapse.[34,35] The extreme temperature and pressure conditions that occur during the collapse of the bubble are recognized as the main mechanism that triggers the breaking of chemical bonds. While ball milling operates with random and spatially nonuniform force fields, ultrasonication provides directional and solution-based force transmission. This characteristic enables ultrasonication to facilitate precise, controlled, and reproducible mechanochemical activation. Moreover, sonication eliminates contamination risks arising from collisions between the milling chamber and balls during ball milling. The low-weight ultrasonic homogenizers mediate portability and in situ mechanochemical activation for outdoor applications. Owing to all these unique advantages, ultrasonication is a highly powerful and preferred technique for fundamental research and practical applications where selective and efficient polymer activation is critical.

In this study, we present ultrasound assisted activation and grafting of unreactive, methyl-terminated PDMS onto silicon wafer and glass substrates. The activation of PDMS is achieved solely by ultrasound activation in the absence of external heating and additional solvents and chemicals. The deposition of this activated PDMS followed by washing results in a slippery liquid-like surface with a sliding angle less than 17° for liquids with surface tensions that range between 20 and 73 mN/m. Conventional hydrophobic/superhydrophobic coatings primarily repel high-surface-tension liquids, such as water, and typically rely on long-chain hydrocarbon silanes. In contrast, the omniphobic coatings developed in this work exhibit superior functionality by repelling not only aqueous but also low-surface-tension liquids, including oils, thereby extending their applicability and enhancing performance under practical conditions. Key parameters regarding ultrasound activation and grafting are systematically studied as a function of wetting properties. The developed fluorine-free omniphobic liquid-like surface exhibits high mechanical, chemical, thermal and UV stability. The versatile and low-cost deposition of such nonstick molecular coatings over square meters areas, such as a windshield of a car demonstrate the practical potential of the proposed approach.

EXPERIMENTAL METHODS AND MATERIALS

Materials. Methyl-terminated PDMS with molecular weights of 1250 g/mol (10 cst, Lot: W02C007), 2000 g/mol (20 cst, Lot: Y26F047), 6000 g/mol (100 cst, Lot: Z14G033), 14,000 g/mol (350 cst, Lot: T12C031), and 28,000 g/mol (1000 cst, Lot: T08C011) were purchased from Alfa Aesar. Silicon wafers (⟨100⟩, n-doped) were obtained from Wafer World Inc. Glass slides (1 mm thick) and ethanol (99.9% purity, Lot: LR0090611APQ) were purchased from Isolab. Toluene (99.7% purity, Lot: STBK2371) and *N*,*N*-dimethylformamide (≥99.0% purity, Lot: SHBP1695) were purchased from Sigma-Aldrich. Formamide (≥99.0% purity, Lot: K43521808 306) and ethylene glycol (≥99.5% purity, Lot: K54175921 218) were purchased from Merck.

Ultrasound Activation and Grafting of PDMS. 20 mL of methyl-terminated PDMS was placed in a 40 mL beaker and an ultrasonic probe MS73 (3 mm) of the ultrasonic homogenizer (model HD 2070.2, SONOPULS) was immersed into the PDMS at a depth of 1 cm. The ultrasonic homogenizer was programmed to operate at a power level of 100% corresponding to 70 W at 20 kHz for durations from 1−120 min. Before grafting, the substrates were washed with ethanol for 10 min. Then they were cleaned in a UV-ozone chamber (BioForce Procleaner) for 30 min. PDMS activated with the ultrasonic homogenizer (200 μL) was dropped onto the surface of the cleaned substrates and left to graft at ambient atmospheric conditions and room temperature for desired durations of 15, 30, 60, and 120 min. Thereafter, the modified substrates were thoroughly washed in toluene, ethanol, and water for 10 min each. Ultrasonically treated PDMS is referred to as "activated PDMS" throughout the manuscript. For comparison, untreated PDMS was grafted by simply placing PDMS on surfaces for a certain time, followed by thorough washing in toluene, ethanol, and water for 10 min each. To demonstrate a practical application, ultrasound activated PDMS was applied using a brush onto the freshly cleaned windshield (70 × 70 cm^2) of a car and left for 1 h. The windshield was then sprayed with pressurized ethanol and water to remove the ungrafted PDMS.

Characterization. The contact angle and the sliding angle were measured using an optical tensiometer system (Attension, Theta Lite) at least at three different locations of the substrate, using 5 and 10 μL droplets, respectively. The advancing and receding contact angles were measured by moving 10 μL water drops downward on an inclined plate. The advancing and receding contact angles were determined using the tilting plate method. In this approach, a 10 μL water droplet was placed on the surface, and the plate was gradually tilted until the droplet initiated motion. The front and rear angles at the onset of motion were recorded as the advancing and receding contact angles, respectively. The morphology and elemental composition of the PDMS-grafted substrate surfaces was analyzed using a scanning electron microscope (SEM, Zeiss EVO LS10) and an energy-dispersive X-ray (EDX) detector (XFlash 6110, Bruker). Before SEM analysis, the samples were sputter-coated with a thin layer of gold to improve conductivity. The chemical composition of the coatings was analyzed through X-ray photoelectron spectroscopy (XPS) using a Thermo Scientific K-Alpha system. The survey spectra scans were performed with a transition energy of 80 eV, 10 sweeps, and an energy step of 1 eV. The temperature of the PDMS during ultrasonic activation was measured with a Fluke TIS75 thermal camera. The optical transparency of PDMS grafted glass substrates was obtained using an UV−vis spectrophotometer (PerkinElmer Lambda 25). The viscosity of PDMS was determined using a viscometer (Brookfield-model DV-E). The thickness of the grafted PDMS was measured using a Gaertner LSE Stokes ellipsometer on silicon substrates. The refractive index of PDMS was set to 1.4025 and a native oxide layer of 1.5 nm was taken into account. Measurements were conducted at a 70° incidence angle using a single-wavelength

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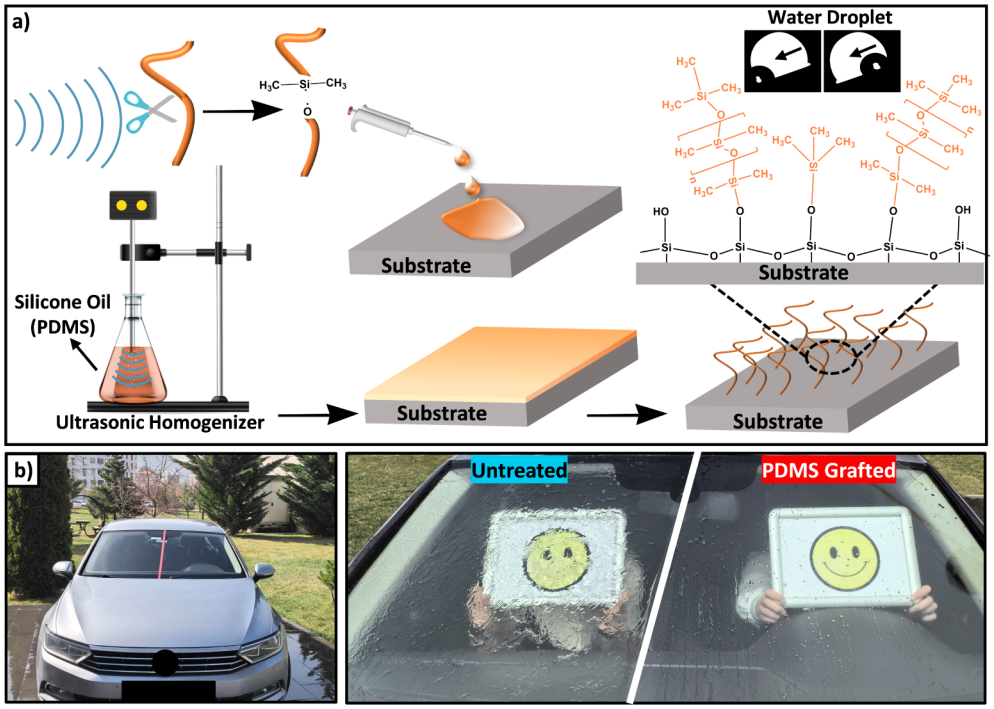

Figure 1. Ultrasound activation and grafting of PDMS. (a) A schematic representation of the process. The inset on the top right corner shows the images of a water droplet sliding down a tilted substrate grafted with PDMS. (b) A photograph demonstrating the brush-coating of a car windshield with ultrasound activated PDMS and the subsequent evaluation of its wetting behavior. Water repellency comparison of a car windshield, where one-half is PDMS-grafted, and the other remains untreated. A paper with a smiling emoji is held behind the windshield while water is sprayed onto the exterior surface, illustrating the high transparency and water-repellence properties of the coating.

He−Ne laser (632.8 nm). Three measurements near the center of each sample were averaged to obtain the reported thickness.

Durability Tests. The durability of the grafted PDMS films was studied by different tests. In the continuous water flow impact test, the samples were exposed to water flowing through a 5.8 mm diameter tube at a flow rate of 0.25 L/s for 1 h. In the water spray impact test, water droplets were sprayed to the surface from a distance of 2.5 cm using a spray gun with a nozzle diameter of 0.35 mm, producing an impact velocity of 3.0 m/s. To investigate the thermal stability, the substrate was heated at 200 °C for 24 h. To assess UV stability, the substrates were subjected to ultraviolet light irradiation for 24 h, with a distance of 5 cm maintained between the substrate and the UV light (245 nm, 60 W) source. In the tape peel test, an adhesive tape was adhered to the sample surface, and a 200 g weight was applied on top. After waiting for one min, the tape was removed, and this procedure was repeated 100 times. In the sonication test, the sample was placed in water and sonicated for 60 min.

■ RESULTS AND DISCUSSION

Figure 1a schematically depicts the ultrasound assisted activation and grafting of methyl-terminated PDMS, also known as silicone oil. Treating a PDMS melt for 1 h with an ultrasonic homogenizer in the absence of external heating and solvents or chemicals activates the unreactive PDMS. The activated PDMS is then drop casted or brushed onto the silicon oxide terminated silicon or glass substrates at ambient conditions. Finally, the excess material is removed by sequential washing in toluene, ethanol and water to ensure that only chemically grafted PDMS chains remain on the substrate. Toluene, particularly is a good solvent for PDMS with the Hildebrand solubility parameter of ∼0.4 $(cal/cm^3)^{1/2}$.[36] Therefore, washing with toluene effectively removes unbound PDMS chains. This approach is scalable to coat large areas with the ability to rapidly activate low-cost unreactive silicone with portable tools and adaptability to solution-based coating techniques such as brushing and drop-casting. Furthermore, the method is compatible with dip-coating, a widely reported approach in the literature, offering a practical and industrially relevant fabrication strategy.[37,38] The ambient grafting in short durations without need for any additional heating, closed containers, bulky equipment and light sources are key advantages. PDMS grafted onto the glass surface via ultrasonic activation exhibited a water contact angle of 108° and a sliding angle of 18°. Figure S1 comparatively presents the wetting behavior of the PDMS-grafted glass surface and the untreated glass surface. While water droplets on the untreated glass tend to spread across the surface, those on the PDMS-modified surface readily slide off in droplet form. This behavior suggests that applying PDMS coatings to surfaces such as commercial building glass facades or solar panels can impart self-cleaning functionality, thereby reducing or even eliminating the need for costly maintenance and cleaning processes. Notably, approximately 60 mL of silicone is sufficient to cover 1 m^2 of surface area, and based on local manufacturer prices, the estimated coating cost is less than 0.30 USD/m^2. To demonstrate a large-area application, a car windshield was coated with the ultrasound activated PDMS (Figure 1b). The deposition was achieved via brushing and ungrafted PDMS was removed by washing using ethanol and water. To discern the variation in wettability, half of the car windshield was grafted with PDMS, while the other half remained untreated. A spray of water droplets was applied onto the windshield, with two individuals positioned inside the vehicle, each holding an emoji. In the untreated half of the windshield, a film of water formed on the surface, obscuring the emoji and compromising visual perception. In contrast, in the PDMS-grafted half of the

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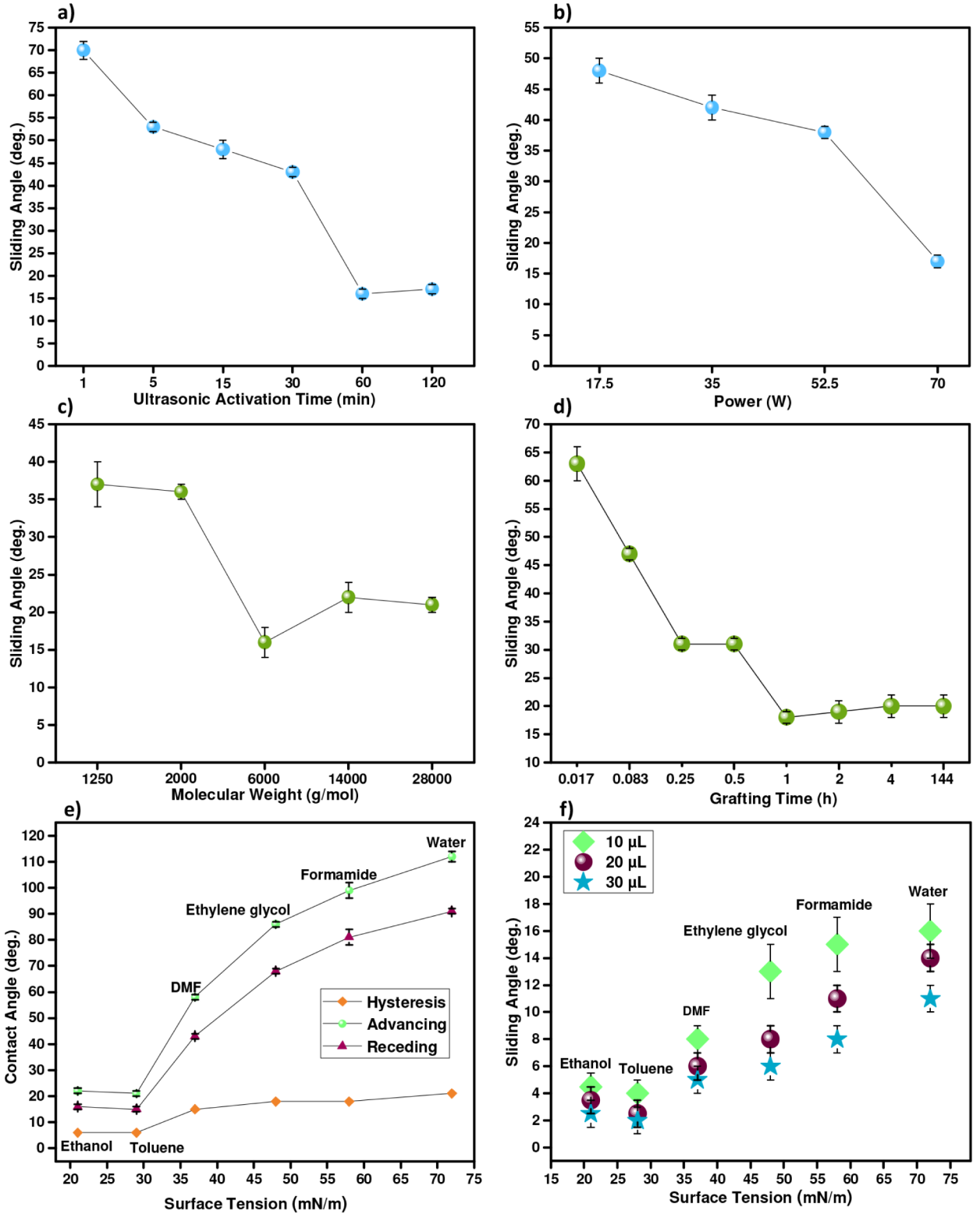

Figure 2. Effects of ultrasound activation, molecular weight, and grafting time. (a) Effect of the ultrasonic activation time on the water sliding angle. Grafting time was kept constant at 60 min and the molecular weight of PDMS was 6000 g/mol. (b) Effects of ultrasonic power on the wetting properties. Ultrasonic homogenizer time and grafting time were kept at 60 min. The molecular weight of PDMS was 6000 g/mol. (c) Effect of the molecular weight of PDMS on the water sliding angle. The ultrasonic activation and grafting time were kept constant at 60 min. (d) Wetting properties of surfaces prepared by different grafting time. The ultrasonic activation was kept constant at 60 min and the molecular weight of PDMS was 6000 g/mol. (e) Advancing contact angle, receding contact angle, and contact angle hysteresis of various liquids on PDMS-grafted surfaces. (f) Sliding angle for different volume drops and different surface tension liquids. Ultrasonic homogenizer time and grafting time were kept at 60 min. The molecular weight of PDMS was 6000 g/mol.

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windshield, water forms droplets that slide off the surface, making the emoji clearly visible (Figure 1b and Video S1). It should be noted that the droplets generated by the spray are larger in volume than the 10 μL droplets used for sliding angle measurements. This difference in the volume of droplets plays a role in the observed water repellency. The grafting mechanism relies on covalent siloxane bonds with surface silanol groups preventing direct application on substrates lacking such chemistry. The current methodology is effective in generating slippery surfaces on silicon wafers, glass and metal oxide terminated substrates. However, a set of important industrial applications require modification of metallic substrates lacking the necessary surface chemical groups. One possible solution is deposition of a thin interlayer such as silicon dioxide, so that the ultrasound activated PDMS can be grafted. Although deposition of such an interlayer was effectively demonstrated on aluminum, stainless steel, polyester fabric, and paper substrates, this additional step may limit the scalability of the process for applications, which require coating of substantially large areas.[8]

To determine optimal ultrasound activation and deposition conditions, we first studied effects of activation time, homogenizer power, grafting time and molecular weight of PDMS. This study was performed based on the sliding angle, which practically shows the dynamic water repellency of the surface at the minimum tilting angle. The first parameter affecting the sonochemical activation and grafting of PDMS is the duration of ultrasound activation. PDMS was exposed to the ultrasonic homogenizer for periods ranging between 1 and 120 min (Figure 2a). PDMS exhibits a sliding angle of 16° when grafted onto the surface following 60 min of exposure to ultrasonic homogenization. No significant changes were observed when PDMS was subjected to ultrasonication for durations exceeding 60 min. At shorter durations, the sliding angle of PDMS-grafted substrates was higher, which indicates insufficient activation. For example, when PDMS is grafted onto the surface after only 1 min, the water sliding angle of the silicon wafer, initially at >90°, decreased to 70°. The likely reason for such a high sliding angle is that the 1 min activation period was insufficient for complete PDMS activation. While some activation may have occurred, PDMS is likely not homogeneously grafted onto the surface. These inhomogeneities can cause localized variations in the interaction between the liquid and the surface, which in turn increases the friction between the water droplet and the surface, thereby elevating the sliding angle. Similarly, a direct relationship was observed between the ultrasonic activation time and the contact angle, and the contact angle value increased as the activation time increased. However, this increase is not as pronounced as the change in the sliding angle (Figure S2). Consequently, a duration of 60 min was selected as the ultrasonic homogenization time and will be utilized in subsequent sections.

The water sliding angle of PDMS-grafted substrates inversely depended on the power of ultrasonic homogenizer used to active PDMS chains. As the power of the ultrasonic homogenizer increased, the sliding angle decreased (Figure 2b). For instance, PDMS exhibits a sliding angle of 48° after being exposed to ultrasound waves at a power of 17.5 W and grafted onto the surface. When PDMS was treated at an ultrasound power of 70 W prior to grafting, the surface exhibited a sliding angle of 16°. Thus, higher power levels led to lower adhesion of water droplets to the PDMS grafted substrate. The findings can be explained by the enhanced formation of free radicals and chain scission, similar to the effect of ball milling rotation speed on the activation and grafting of PDMS.[9,32]

To analyze the effect of molecular weight, activated PDMS samples with molecular weights ranging from 1250 to 28,000 g/mol were drop-cast on the surface (Figures 2c and S3). As the molecular weight of PDMS increased, the sliding angle of the surfaces also decreased. Similarly, a significant correlation was observed between molecular weight and contact angle (Figure S3). This trend is consistent with previous reports in the literature[19,31] The thickness of the PDMS layer grafted onto the surface increased with molecular weight from 3 nm for 1250 g/mol to 7 nm at 28,000 g/mol as measured by ellipsometry (Figure S4). The measured thicknesses are lower than those expected for the fully extended PDMS chains due to their flexible, coiled structure and parallel alignment to the surface, which alters chain packing and significantly impacts wetting behavior.[8,39] The surface prepared with PDMS of 1250 g/mol molecular weight likely exhibited a lower ability to resist water-surface interactions compared to surfaces prepared with higher molecular weight PDMS, resulting in a higher water sliding angle. The grafted film prepared with high molecular weight PDMS (28,000 g/mol) exhibits a higher thickness (~6.9 nm) compared to the ~4 nm thickness of PDMS with 6000 g/mol. However, it shows higher water sliding angle. This is due to the high molecular weight PDMS chains being highly coiled, which restricts chain mobility and consequently reduces the surface's water repellency. It should be noted that these molecular-weight-dependent effects are not limited to film thickness but also influenced by several other parameters, including grafting density, surface roughness, and chain conformation. Conventional AFM imaging was insufficient to resolve structural details and differences among liquid-like surfaces prepared by using different molecular weights of PDMS. Thus, based on the findings of this experimental study, the optimal molecular weight was determined to be 6000 g/mol, which will be utilized in the following sections unless otherwise specified. This finding confirms previous studies that this range of molecular weight PDMS is best suited for surfaces exhibiting low adhesion to liquids.[8,9,39]

Another critical parameter that determines the wetting behavior of the surface is the grafting time. Various grafting durations were investigated to assess the effect of grafting time on surfaces produced via the sonochemical approach (Figure 2d). When PDMS is grafted onto the surface after only 1 min, the water sliding angle of the silicon wafer, initially at >90°, decreased to 63°. As the grafting density of PDMS chains on the surface increases with time, the sliding angle gradually decreases until it reaches a certain threshold. After 60 min of grafting, the developed surface exhibited sliding angle of 16°. Since there was no significant change in the wetting behavior of the surface after 60 min of grafting, the optimum grafting time was determined as 60 min. In contrast, a previous study reported that methyl-terminated PDMS could be grafted onto a silicon wafer at room temperature for 24 h without any additional treatment, resulting in a surface with a sliding angle of 12° for water.[8] This comparison underscores that, while prolonged grafting in the literature enabled omniphobicity, the ultrasound-assisted approach developed in the present work achieves comparable surface wetting property within a substantially shorter processing time. Regardless of the extent of ultrasound treatment of PDMS, the reactive groups

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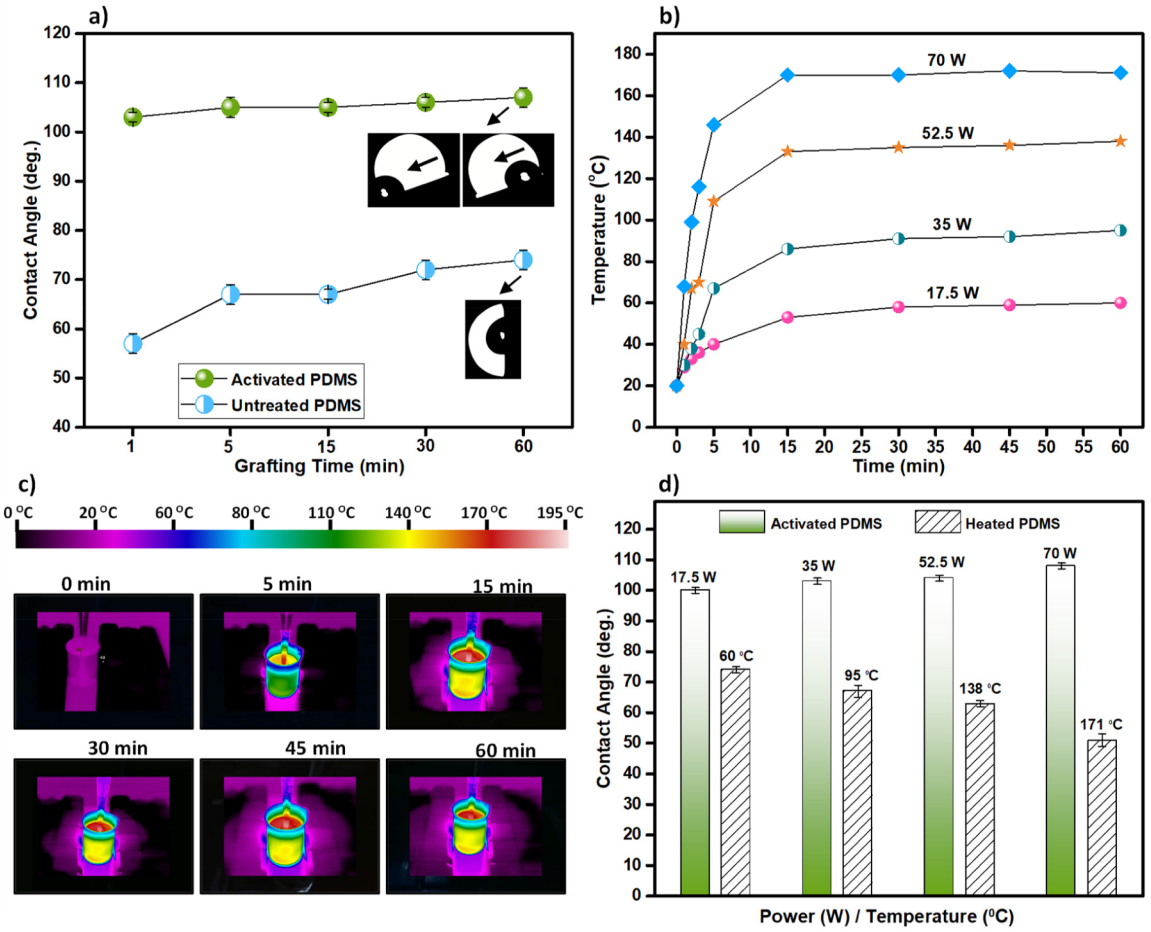

Figure 3. Control experiments. (a) Effect of grafting time on wetting properties for surfaces prepared by depositing ultrasound activated and untreated PDMS. Images of water droplets sliding down a tilted a substrate grafted with PDMS are presented. Ultrasonic activation time for activated PDMS was kept constant at 60 min and the molecular weight of PDMS was 6000 g/mol. (b) Effects of ultrasonic power on the temperature of PDMS. The molecular weight of PDMS was 6000 g/mol. (c) Infrared camera images of PDMS with a molecular weight of 6000 g/mol during ultrasonic processing. (d) Effects of ultrasonic power on the wetting properties and temperature of PDMS on the wetting properties following grafting of untreated PDMS heated at the specified temperatures. Ultrasonic homogenizer time and grafting time were kept at 60 min. The molecular weight of PDMS was 6000 g/mol.

generated in the environment tend to recombine over time, losing the ability to graft to the substrate. To investigate this effect, PDMS activated by ultrasonic homogenization was stored for varying durations before being deposited onto the surface (Figure S5). As storage time increased, the contact angle decreased, while the sliding angle increased. This suggests that the free radicals in the environment gradually rebond with each other over time, leading to a reduction in reactive species. These findings agree with our previous study, in which PDMS was converted into a reactive form via ball milling and was shown to exhibit a similar tendency toward recombination.[9]

The grafting of ultrasound activated PDMS leads to surfaces that exhibit low adhesion to liquids with a broad range of surface tension. The contact angle hysteresis plays a critical role in the lateral adhesion force between a liquid droplet and solid surface. Figure 2e presents advancing and receding contact angles and contact angle hysteresis for liquids with different surface tensions. A drop starts sliding if the lateral component of the gravitational force exceeds the adhesion force as given with eq 1.

$$\mathbf{F}_G = mg\sin\alpha > F_{adh} \approx w\gamma(\cos\theta_r - \cos\theta_a) \quad (1)$$

In this equation, m represents the mass of the liquid droplet, g is the gravitational acceleration (9.81 m/s^2), α is the inclination of the substrate, w is the lateral width of the droplet, γ is the surface tension of the liquid, θ_r is the receding contact angle and θ_a is the advancing contact angle. As indicated by the equation, with the reduction of the contact angle hysteresis ($\theta_a - \theta_r$), the gravitational force becomes larger than the lateral force at reduced inclination angles and results in low sliding angle of liquids. Figure 2f presents sliding angle for different liquids on PDMS grafted surfaces prepared by the ultrasound activation. As consistent with the equation, the gravitational force increases with the volume of drops and larger drops typically exhibit lower sliding angles. Liquids with lower surface tension show lower contact angle hysteresis and lower sliding angles in close agreement with the literature.[22,31,40] In the case of water, the contact angle hysteresis achieved in our study, while sufficient to demonstrate omniphobic behavior, is higher than some examples reported in the literature.[6] Further understanding of the mechanism of ultrasound-mediated activation and grafting of PDMS is needed to improve dynamic water repellence of these molecular coatings.

Control experiments in the absence of ultrasound homogenizer treatment confirm the necessity of such activation step

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to successfully generate liquid-like slippery surfaces. We first comparatively study deposition of ultrasound activated and untreated PDMS on a silicon substrate. Both liquid PDMS were deposited for varied times followed by sequential washing in toluene, ethanol, and water for 10 min each. There is a strong contrast in static contact angles and dynamic wetting behavior of these two surfaces (Figure 3a). The contact angle of surfaces prepared by using untreated PDMS was consistently lower than the ones that were ultrasound activated. For example, upon 60 min of grafting, the measured contact angle reached 74° for untreated PDMS, whereas the contact angle was greater than 100° for the ultrasound activated PDMS. More importantly, water drops pinned to the surfaces prepared by deposition of untreated PDMS for 1 h, whereas effective sliding was observed on the ultrasound activated PDMS. Previous literature has shown that, over long periods of time (typically >24 h), methyl-terminated PDMS can bond to silicon-oxide-terminated surfaces and form a slippery coating.[8] This study demonstrated that linear PDMS chains can be grafted onto silicon oxide surfaces at room temperature via hydrolysis and condensation reactions, without any additional processing steps. However, the grafting times for ultrasound-activated PDMS are substantially shorter, confirming the critical role of this activation strategy in the effective grafting of PDMS onto surfaces. During ultrasonic treatment, PDMS chains may partially cleave, possibly generating highly reactive species through the scission of siloxane bonds, thereby leading to the formation of siloxy mechanoradicals and silicon-centered radicals. In addition, the presence of residual water in the system could result in the formation of hydroxyl-terminated PDMS chains. These reactive species (radicals or hydroxyl-terminated PDMS) are likely to undergo condensation reactions with surface silanol groups, which may enhance the grafting efficiency. To verify this mechanism, a control experiment was performed using hydroxyl-terminated PDMS (OH-PDMS, Mn = 4200 g/mol). Without any pretreatment, OH-PDMS was grafted onto the surface following the previously established procedure. The resulting surface exhibited a static water contact angle of 95° and a sliding angle of 56°. In comparison, methyl-terminated PDMS, grafted under the same conditions without ultrasonic activation, yielded a surface with a static contact angle of 74° but did not exhibit any sliding behavior. However, surfaces prepared with ultrasound-activated methyl-terminated PDMS showed superior liquid repellence. Overall, these results clearly demonstrate the effectiveness of the ultrasonic activation methodology.

The second set of control experiments targeted concerns regarding the rise of temperature during the ultrasound activation. A significant increase in the temperature of liquid PDMS was observed during the ultrasonic treatment (Figure 3b). PDMS was exposed to ultrasonic sound waves at different powers (17.5 W, 35 W, 52.5 W, and 70 W) for 1 h and the temperature changes were monitored with a thermal camera, which was double-checked with a thermocouple. The temperature of PDMS rises significantly during the ultrasonic treatment, reaching to 146 °C in only 5 min at 70 W. The temperature of PDMS increased with increasing of the ultrasonic power. For example, when PDMS was subjected to 1 h of ultrasonic treatment at 17.5 W, the temperature reached 60 °C, whereas at 70 W this value increases to 171 °C (Figure 3c). These observations raise the question: Is the heating during ultrasonication responsible for the PDMS grafting? To address this question, PDMS (6000 g/mol) samples were heated at temperatures that were observed at different ultrasonic powers and these liquid PDMS were then drop-cast onto the silicon wafer for 1 h. In all cases, only heated samples did not show dynamic repellence, and water drops pinned on the surfaces. Therefore, only static contact angle measurements are presented in Figure 3d. We attribute the decreasing static contact angle (Figure 3d) of surfaces prepared by using only heated PDMS samples with the increasing temperature to reduction in the viscosity of samples. A similar experiment was conducted using OH-PDMS. The OH-PDMS was heated to 171 °C and then drop-cast onto the substrate. This temperature was selected because it corresponds to the approximate temperature reached during the ultrasonication process under optimal conditions. After 1 h of grafting, the samples were rinsed according to the procedure described in the experimental section. The resulting surface exhibited a water contact angle of 77° and a sliding angle of 88°. These results further confirm that ultrasonication is the key factor enabling effective PDMS grafting onto the substrate. The contact angles of surfaces prepared by deposition of only heated samples exhibited significantly lower values in comparison with their ultrasonically activated counterparts. We highlight that only liquid PDMS was heated in these experiments and there was no additional heating after the deposition of PDMS and grafting. Otherwise, heating the substrate after deposition of methyl-terminated PDMS is an effective approach to fabricate low contact angle hysteresis omniphobic surfaces.[41]

The ultrasound activation results in a PDMS molecular weight-dependent temperature rise and a viscosity reduction. PDMS samples with different molecular weights show significant temperature differences after 1 h of ultrasonic treatment (Figure S6). The rise of temperature during ultrasound treatment scales with the molecular weight of PDMS. After 1 h of ultrasonic treatment, the temperature of PDMS with a molecular weight of 1250 g/mol reached 74 °C and that of PDMS with a molecular weight of 28,000 g/mol reached 226 °C. This observation may be related to the entanglement of PDMS chains at high molecular weights, which may cause enhanced friction and temperature rise. An additional observation is significant decrease in viscosity of PDMS after ultrasound activation (Figure S7). Viscosity is of interest due to its indication of the molecular weight of PDMS. For example, the viscosity of PDMS with a molecular weight of 1250 g/mol decreased from 10 cSt to 5 cSt after ultrasonic treatment, while the viscosity of PDMS with a molecular weight of 28,000 g/mol decreased from 1000 cSt to 385 cSt. These results show a viscosity reduction of about 2 and 3 times in low and high molecular weight PDMS samples, respectively. However, the viscosity of 6000 g/mol PDMS, which was determined as the optimum molecular weight, decreased from 100 cSt before ultrasonic treatment to 18 cSt after treatment, a decrease of more than 5 times. However, since temperature increases during the ultrasonic activation process, viscosity changes depending on temperature should also be taken into account. The viscosity of ultrasound activated PDMS was typically lower than the only heated PDMS samples. The reduction in viscosity is likely due to a combination of increased temperature, chain scission, and shear thinning. Upon ultrasonic activation, the number of chain entanglements decreases, resulting in reduced resistance to flow and, consequently, lower viscosity.

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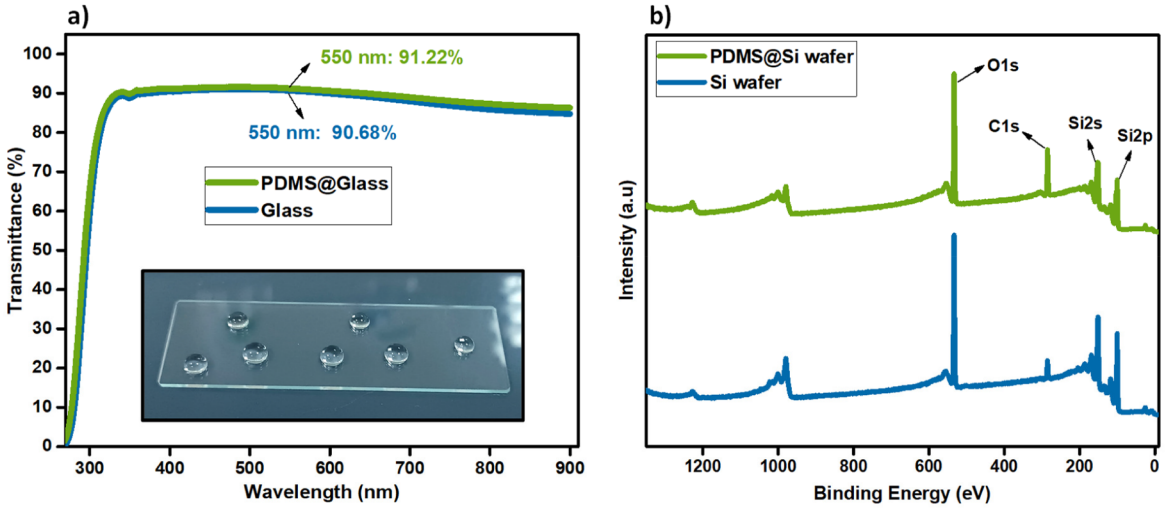

Figure 4. (a) Optical transmittance of PDMS-grafted and bare glass substrate. (b) XPS survey spectra of Si wafer and PDMS grafted Si wafer. Ultrasonic homogenizer time and grafting time were kept at 60 min. The molecular weight of PDMS was 6000 g/mol.

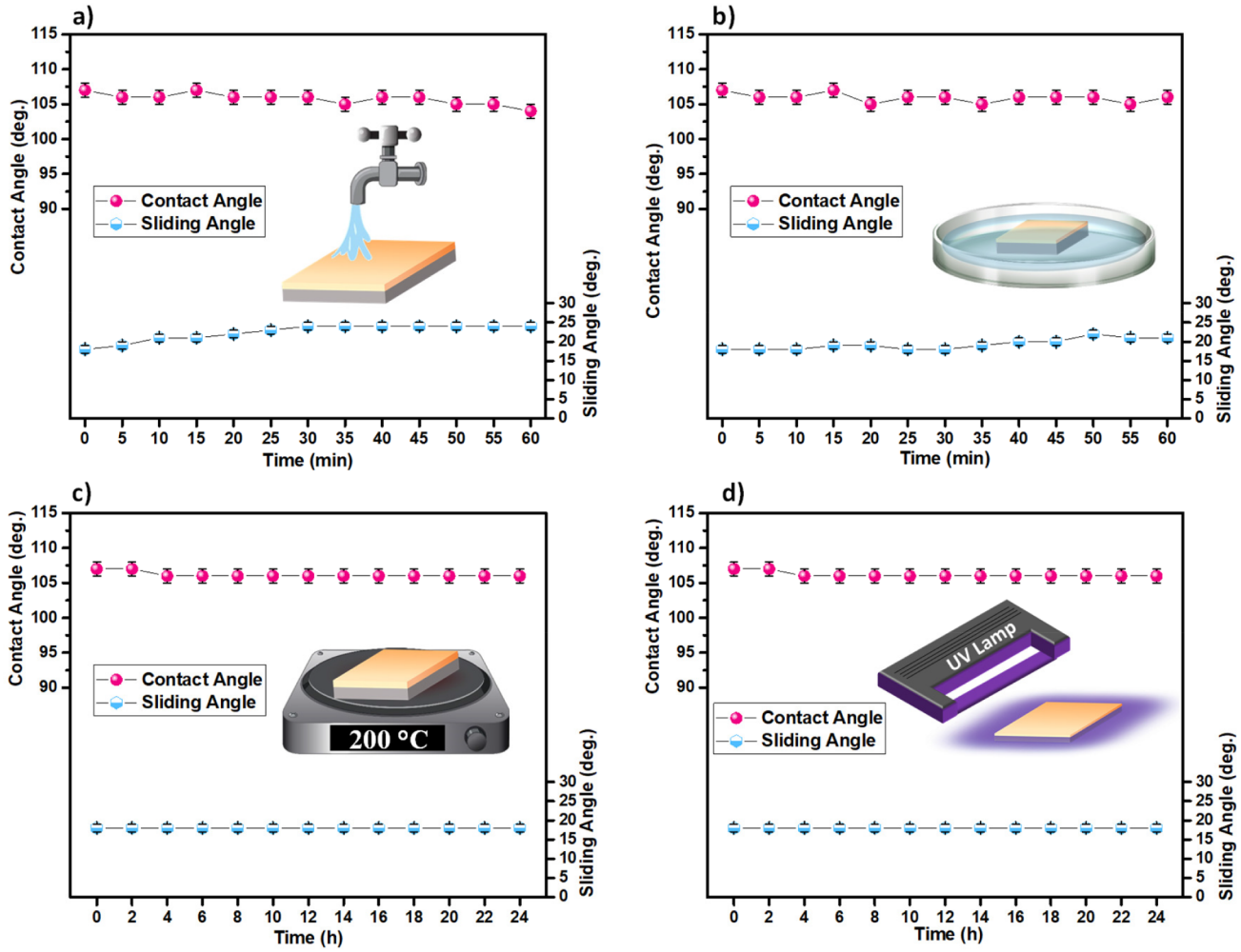

Figure 5. Durability of PDMS grafted surface, evaluated using (a) water jet impact test, (b) sonication-washing time test, (c) thermal stability test, (d) UV-irradiation test. Ultrasonic homogenizer time and grafting time were kept at 60 min. The molecular weight of PDMS was 6000 g/mol.

A key characteristic of the grafted PDMS surface, which is only a few nanometers thick, is its minimal light scattering, thereby maintaining the transparency and appearance of the coated surfaces (Figure 4a). In the visible light spectrum, no significant difference in transmittance was observed between the PDMS-grafted glass and the bare glass substrate. At a wavelength of 550 nm, 91.22% of light passed through the PDMS-grafted surface, compared to 90.68% for the bare glass

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substrate. XPS and EDX analyses provide evidence supporting the grafting of activated PDMS onto the surface (Figures 4b and S8). The presence of carbon from the methyl side groups of PDMS led to a significant increase in carbon content, with the C 1s peak at 248.78 eV showing a 104% increase compared to the untreated silicon wafer (Table S1). The increase in the carbon (C) element ratio observed in XPS analysis after the grafting process confirms the presence of PDMS on the surface, and this finding is highly consistent with the results reported in the literature.[8]

The long-term stability issues of liquid repellent coatings are a major limiting factor in many potential applications such as self-cleaning surfaces,[42,43] food packaging,[44,45] anti-ice surfaces,[46,47] anticorrosion applications,[48] water treatment systems,[49,50] and enhancing detection limits by concentrating molecules in biological assays.[51] The stability of liquid repellent coatings under different environmental and application conditions is of great importance for the effectiveness and sustainability of these technologies in practical applications. The soft interface formed by the grafting of ultrasound activated PDMS exhibits significant stability (Figure 5). Mechanical durability is crucial for the practical application of liquid-like surfaces, so various techniques were employed to assess the robustness of PDMS-grafted surfaces. First, the PDMS-grafted substrate was exposed to pressurized water using a water jet for 30 min. Although no change was observed in the contact angle after this exposure, the sliding angle increased slightly by 6° (Figure 5a). Second, water was sprayed onto the surface. After 1000 cycles of the water spray test, the surface retained its high repellency (Figure S9). In another water stability test, the PDMS grafted surface was sonicated in water for 60 min. After this treatment, the sliding angle of the surface increased by 3°, while the contact angle did not change significantly (Figure 5b). Despite the high polarity of water, the PDMS remained stable in aqueous environments due to the strong grafting. The thermal stability of the PDMS-grafted substrate was investigated by subjecting the samples to heating at 200 °C for 24 h (Figure 5c). After heating, no alteration was detected in the water-repellence of the PDMS-grafted surface, consistent with previous reports in the literature.[52] To assess UV stability, the substrates were exposed to ultraviolet light for 24 h (Figure 5d). Similarly, no alteration in the wetting behavior of the PDMS-grafted surface was observed following UV exposure. A tape peel test was conducted to simulate the mechanical forces the PDMS-grafted surface might encounter in practical applications. The tape was uniformly applied to the surface and peeled off repeatedly for 100 cycles (Figure S10). After 100 cycles, the sliding angle of the PDMS-grafted surface increased from 18° to 37°, indicating some loss of performance but still maintaining functional repellency. A notable observation was the stability of PDMS-grafted windshields (Figure 1b) after driving 2000 km in rainy conditions and continuously operating the windshield wipers for approximately 200 km. Water droplets on the coated car windshield were observed to runoff the surface with relative ease after 2000 km drive (Video S2). Although these results show promise, systematic studies are required to comprehensively assess the long-term performance of these molecular coatings. For broader industrial applications, it would be more appropriate to conduct stability assessments in accordance with relevant ASTM standards.[53]

CONCLUSION

This study has presented a versatile method for fabricating slippery liquid-like surfaces composed of grafted PDMS chains. The method is based on ultrasound activation of presynthesized and unfunctional methyl-terminated PDMS followed by grafting to silicon oxide terminated substrates. The resulting nanoscopic films exhibit low adhesion to liquids with a broad range of surface tensions and droplets can slide at low angles. The simple activation by ultrasonic homogenizer and direct solvent-free deposition by brushing and drop-casting ensure scalable and low-cost fabrication on various substrates, including glass and metal oxide-terminated surfaces. The demonstrated ability to generate liquid-like nonstick molecular coatings over square meters of areas show the scalable nature of the proposed method. The ambient grafting in short durations without need for any additional heating, closed containers, bulky equipment and light sources are key advantages of the proposed method. The penetration of ultrasound waves could offer an intriguing way to control the grafting of PDMS chains from a distance.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.langmuir.5c03707.

EDX mapping analyses, XPS elemental composition, stability tests, and experimental parameters affecting omniphobicity, including additional experimental characterizations (PDF)

Water repellency of a car windshield, where one half is PDMS-grafted, and the other remains untreated (MP4)

Water repellency of the PDMS grafted car windshield after 2000 km drive (MP4)

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Notes

The authors declare no competing financial interest.

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■ REFERENCES

(1) Fadeev, A. Y.; McCarthy, T. J. A New Route to Covalently Attached Monolayers: Reaction of Hydridosilanes with Titanium and Other Metal Surfaces. *J. Am. Chem. Soc.* **1999**, *121* (51), 12184−12185.

(2) Fadeev, A. Y.; McCarthy, T. J. Self-Assembly Is Not the Only Reaction Possible between Alkyltrichlorosilanes and Surfaces: Monomolecular and Oligomeric Covalently Attached Layers of Dichloro- and Trichloroalkylsilanes on Silicon. *Langmuir* **2000**, *16* (18), 7268−7274.

(3) Krumpfer, J. W.; McCarthy, T. J. Rediscovering Silicones: "Unreactive" Silicones React with Inorganic Surfaces. *Langmuir* **2011**, *27* (18), 11514−11519.

(4) Cheng, D. F.; Urata, C.; Masheder, B.; Hozumi, A. A Physical Approach To Specifically Improve the Mobility of Alkane Liquid Drops. *J. Am. Chem. Soc.* **2012**, *134* (24), 10191−10199.

(5) Liu, P.; Zhang, H.; He, W.; Li, H.; Jiang, J.; Liu, M.; Sun, H.; He, M.; Cui, J.; Jiang, L.; Yao, X. Development of "Liquid-like" Copolymer Nanocoatings for Reactive Oil-Repellent Surface. *ACS Nano* **2017**, *11* (2), 2248−2256.

(6) Wooh, S.; Vollmer, D. Silicone Brushes: Omniphobic Surfaces with Low Sliding Angles. *Angew. Chem. Int. Ed.* **2016**, *55* (24), 6822−6824.

(7) Singh, N.; Kakiuchida, H.; Sato, T.; Hönes, R.; Yagihashi, M.; Urata, C.; Hozumi, A. Omniphobic Metal Surfaces with Low Contact Angle Hysteresis and Tilt Angles. *Langmuir* **2018**, *34* (38), 11405−11413.

(8) Teisala, H.; Baumli, P.; Weber, S. A. L.; Vollmer, D.; Butt, H.-J. Grafting Silicone at Room Temperature—a Transparent, Scratch-Resistant Nonstick Molecular Coating. *Langmuir* **2020**, *36* (16), 4416−4431.

(9) Celik, N.; Sahin, F.; Ruzi, M.; Ceylan, A.; Butt, H.-J.; Onses, M. S. Mechanochemical Activation of Silicone for Large-Scale Fabrication of Anti-Biofouling Liquid-like Surfaces. *ACS Appl. Mater. Interfaces* **2023**, *15* (46), 54060−54072.

(10) Mayoussi, F.; Chen, Y.; Mössner, D.; Franco Corredor, S.; Goralczyk, A.; Mader, M.; Warmbold, A.; Kotz-Helmer, F.; Helmer, D.; Rapp, B. E. Smooth Transparent Omniphobic Coatings with Remarkable Liquid Repellence. *Adv. Mater. Interfaces* **2022**, *9* (28), 2201080.

(11) Peng, J.; Ye, P.; Xu, F.; Geng, H.; Wang, R.; Jiang, Y.; Zhu, Y.; Wang, H. Highly Transparent, Durable, and Omniphobic Liquid-like Coatings for Efficient Dynamic de-Wetting and Self-Cleaning Applications. *Chem. Eng. J.* **2023**, *477*, 147090.

(12) Wu, Q.; Yang, C.; Su, C.; Zhong, L.; Zhou, L.; Hang, T.; Lin, H.; Chen, W.; Li, L.; Xie, X. Slippery Liquid-Attached Surface for Robust Biofouling Resistance. *ACS Biomater. Sci. Eng.* **2020**, *6* (1), 358−366.

(13) Li, S.; Lam, C. W. E.; Donati, M.; Regulagadda, K.; Yavuz, E.; Pfeiffer, T.; Sarkiris, P.; Gogolides, E.; Milionis, A.; Poulikakos, D.; Butt, H.-J.; Kappl, M. D. Ultrathin, and Antifouling Polymer Brush Coating for Efficient Condensation Heat Transfer. *ACS Appl. Mater. Interfaces* **2024**, *16* (1), 1941−1949.

(14) Fenero, M.; Knez, M.; Saric, I.; Petravic, M.; Grande, H.; Palenzuela, J. Omniphobic Etched Aluminum Surfaces with Anti-Icing Ability. *Langmuir* **2020**, *36* (37), 10916−10922.

(15) Yu, M.; Liu, M.; Hou, Y.; Fu, S.; Zhang, L.; Li, M.; Wang, D. Facile Fabrication of Biomimetic Slippery Lubricant-Infused Transparent and Multifunctional Omniphobic Surfaces. *J. Mater. Sci.* **2020**, *55* (10), 4225−4237.

(16) McHale, G.; Gao, N.; Wells, G. G.; Barrio-Zhang, H.; Ledesma-Aguilar, R. Friction Coefficients for Droplets on Solids: The Liquid−Solid Amontons' Laws. *Langmuir* **2022**, *38* (14), 4425−4433.

(17) Kwon, J.; Jung, H.; Jung, H.; Lee, J. Micro/Nanostructured Coating for Cotton Textiles That Repel Oil, Water, and Chemical Warfare Agents. *Polymers* **2020**, *12*, 1826.

(18) Alipanahrostami, M.; Coolidge, C.; Wang, Y.; Wang, W.; Tong, T. Minimizing the Use of Per- and Polyfluoroalkyl Substances for Textured Wetting-Resistant Surfaces. *Environ. Sci. Technol.* **2025**, *59* (7), 3355−3365.

(19) Khatir, B.; Lin, A.; Vuong, T. V.; Serles, P.; Shayesteh, A.; Hsu, N. S. Y.; Sinton, D.; Tran, H.; Master, E. R.; Filleter, T.; Golovin, K. Molecular Structure of Omniphobic, Surface-Grafted Polydimethylsiloxane Chains. *Small* **2025**, *21* (8), 2406089.

(20) Lai, Z.; Wang, J.; Liu, G. Poly(Dimethyl Siloxane) Bimodal Brush: Simple Method of Preparation and Performance Enhancement of Omniphobic Coatings. *Chem. Eng. J.* **2025**, *503*, 158417.

(21) Khatir, B.; Serles, P.; Wen, T.; Vuong, T. V.; Shayesteh, A.; Au, S.; Sinton, D.; Master, E. R.; Filleter, T.; Golovin, K. Autophobic Polydimethylsiloxane Nanodroplets Enable Abrasion-Tolerant Omniphobic Surfaces. *Chem. Eng. J.* **2024**, *502*, 157718.

(22) Kartsev, D. D.; Prilepskii, A. Y.; Lukyanov, I. M.; Sharapenkov, E. G.; Klaving, A. V.; Goltaev, A.; Mozharov, A.; Dvoretckaia, L.; Mukhin, I.; Levkin, P. A. Fabrication of Omniphobic-Omniphilic Micropatterns Using GPOSS-PDMS Coating. *Adv. Mater. Interfaces* **2023**, *10* (16), 2300156.

(23) Wang, X.; Wang, Z.; Heng, L.; Jiang, L. Stable Omniphobic Anisotropic Covalently Grafted Slippery Surfaces for Directional Transportation of Drops and Bubbles. *Adv. Funct. Mater.* **2020**, *30* (1), 1902686.

(24) Yu, M.; Liu, M.; Hou, Y.; Fu, S.; Zhang, L.; Li, M.; Wang, D. Covalently Grafted Liquids for Transparent and Omniphobic Surfaces via Thiol-Ene Click Chemistry. *J. Mater. Sci.* **2020**, *55* (27), 12811−12825.

(25) Gresham, I. J.; Neto, C. Advances and Challenges in Slippery Covalently-Attached Liquid Surfaces. *Adv. Colloid Interface Sci.* **2023**, *315*, 102906.

(26) Foadi, F.; Celik, N.; Esidir, A.; Onses, M. S. Roughness-Dependent Hydrophobicity of Polydimethylsiloxane Grafted Titanium Thin Films. *Surf. Coat. Technol.* **2024**, *483*, 130749.

(27) Herzberg, W. J.; Marian, J. E.; Vermeulen, T. The Receding Contact Angle. *J. Colloid Interface Sci.* **1970**, *33* (1), 164−171.

(28) Chen, W.; Fadeev, A. Y.; Hsieh, M. C.; Öner, D.; Youngblood, J.; McCarthy, T. J. Ultrahydrophobic and Ultralyophobic Surfaces: Some Comments and Examples. *Langmuir* **1999**, *15* (10), 3395−3399.

(29) Khatir, B.; Shabanian, S.; Golovin, K. Design and High-Resolution Characterization of Silicon Wafer-like Omniphobic Liquid Layers Applicable to Any Substrate. *ACS Appl. Mater. Interfaces* **2020**, *12* (28), 31933−31939.

(30) Liu, J.; Sun, Y.; Zhou, X.; Li, X.; Kappl, M.; Steffen, W.; Butt, H.-J. One-Step Synthesis of a Durable and Liquid-Repellent Poly(Dimethylsiloxane) Coating. *Adv. Mater.* **2021**, *33* (23), 2100237.
(31) Wang, L.; McCarthy, T. J. Covalently Attached Liquids: Instant Omniphobic Surfaces with Unprecedented Repellency. *Angew. Chem. Int. Ed.* **2016**, *55* (1), 244−248.
(32) Celik, N.; Akay, S.; Sahin, F.; Sezer, G.; Dagasan Bulucu, E.; Ruzi, M.; Butt, H.-J.; Onses, M. S. Sustainable and Practical Superhydrophobic Surfaces via Mechanochemical Grafting. *Adv. Mater. Interfaces* **2023**, *10* (15), 2300069.
(33) Rosales Pérez, A.; Esquivel Escalante, K. The Evolution of Sonochemistry: From the Beginnings to Novel Applications. *ChemPluschem* **2024**, *89* (6), No. e202300660.
(34) Gedanken, A. Using Sonochemistry for the Fabrication of Nanomaterials. *Ultrason. Sonochem* **2004**, *11* (2), 47−55.
(35) Ashokkumar, M. The Characterization of Acoustic Cavitation Bubbles − An Overview. *Ultrason. Sonochem.* **2011**, *18* (4), 864−872.
(36) Lee, J. N.; Park, C.; Whitesides, G. M. Solvent Compatibility of Poly(Dimethylsiloxane)-Based Microfluidic Devices. *Anal. Chem.* **2003**, *75* (23), 6544−6554.
(37) Zhao, H.; Khodakarami, S.; Deshpande, C. A.; Ma, J.; Wu, Q.; Sett, S.; Miljkovic, N. Scalable Slippery Omniphobic Covalently Attached Liquid Coatings for Flow Fouling Reduction. *ACS Appl. Mater. Interfaces* **2021**, *13* (32), 38666−38679.
(38) Li, Z.; Liu, Y.; Liu, Y.; Feng, K.; Li, J.; Wu, Y.; Zhou, F. Robust Transparent Photothermal Omniphobic Coating for Efficient Anti/Deicing and Antifogging. *ACS Appl. Mater. Interfaces* **2024**, *16* (27), 35805−35814.
(39) Chen, L.; Huang, S.; Ras, R. H. A.; Tian, X. Omniphobic Liquid-like Surfaces. *Nat. Rev. Chem.* **2023**, *7* (2), 123−137.
(40) Zhang, K.; Huang, S.; Wang, J.; Liu, G. Transparent Omniphobic Coating with Glass-Like Wear Resistance and Polymer-Like Bendability. *Angew. Chem., Int. Ed.* **2019**, *131* (35), 12132−12137.
(41) Abbas, A.; Wells, G. G.; McHale, G.; Sefiane, K.; Orejon, D. Silicone Oil-Grafted Low-Hysteresis Water-Repellent Surfaces. *ACS Appl. Mater. Interfaces* **2023**, *15* (8), 11281−11295.
(42) Celik, N.; Sezen, B.; Sahin, F.; Ceylan, A.; Ruzi, M.; Onses, M. S. Mechanochemical Coupling of Alkylsilanes to Nanoparticles for Solvent-Free and Rapid Fabrication of Superhydrophobic Materials. *ACS Appl. Nano Mater.* **2023**, *6* (16), 14921−14930.
(43) Wang, H.; Wang, K.; Lu, H.; Parkin, I. P.; Zhang, X. Flexible and Strong Robust Superhydrophobic Monoliths with Antibacterial Property. *ACS Appl. Polym. Mater.* **2020**, *2* (11), 4856−4863.
(44) Zhang, S.; Li, M.; Wang, R.; Chang, L.; Ju, H.; Lin, W.; Zhao, W.; Tang, Y.; Lin, S. Superhydrophobic and Antioxidative Film Based on Edible Materials for Food Packaging. *Langmuir* **2021**, *37* (16), 5066−5072.
(45) Ruzi, M.; Celik, N.; Onses, M. S. Superhydrophobic Coatings for Food Packaging Applications: A Review. *Food Packag. Shelf Life* **2022**, *32*, 100823.
(46) Torun, I.; Celik, N.; Kiremitler, N. B.; Huang, X.; Onses, M. S. Fully Transparent and Superhydrophobic Electrodes Enabled by Soft Interfaces. *Surf. Interfaces* **2023**, *36*, 102576.
(47) He, Q.; Ma, Y.; Wang, X.; Jia, Y.; Li, K.; Li, A. Superhydrophobic Flexible Silicone Rubber with Stable Performance, Anti-Icing, and Multilevel Rough Structure. *ACS Appl. Polym. Mater.* **2023**, *5* (7), 4729−4737.
(48) Huang, J.; Yang, M.; Zhang, H.; Zhu, J. Solvent-Free Fabrication of Robust Superhydrophobic Powder Coatings. *ACS Appl. Mater. Interfaces* **2021**, *13* (1), 1323−1332.
(49) Celik, N.; Karagoz, S.; Sakir, M.; Yilmaz, E.; Genc, A.; Slater, T. J. A.; Onses, M. S. Solvent-Free Preparation of Superhydrophobic and Photocatalytic Monoliths for Water Treatment Applications. *Colloids Surf., A* **2025**, *721*, 137227.
(50) Wang, X.; Yu, P.; Zhang, K.; Wu, M.; Wu, Q.; Liu, J.; Yang, J.; Zhang, J. Superhydrophobic/Superoleophilic Cotton for Efficient Oil−Water Separation Based on the Combined Octadecanoyl Chain Bonding and Polymer Grafting via Surface-Initiated ATRP. *ACS Appl. Polym. Mater.* **2019**, *1* (11), 2875−2882.
(51) Ruzi, M.; Celik, N.; Sahin, F.; Sakir, M.; Onses, M. S. Nanostructured Surfaces with Plasmonic Activity and Superhydrophobicity: Review of Fabrication Strategies and Applications. *Small* **2025**, *21* (6), 2408189.
(52) Ma, J.; Zhang, C.; Wong, W. S. Y.; Song, J. Facile, scalable and Substrate-Independent omniphobic surface. *Appl. Surf. Sci.* **2025**, *682*, 161726.
(53) Thukral, T. S.; Rabbi, K. F.; Khodakarami, S.; Yang, W.; Sudarshan, A.; Pitschman, M. A.; Fourspring, P. M.; Miljkovic, N. Scalable and Durable Polydimethylsiloxane Coating for Anti-Corrosion, Anti-Fouling, and Condensation Applications. *Cell Rep. Phys. Sci.* **2024**, *5* (10), 102243.

https://doi.org/10.1021/acs.langmuir.5c03707
Langmuir 2025, 41, 31816−31826