

Not All Sizes of Dust can be Removed by Jumping Condensates on Superhydrophobic Surfaces

Kangning Li,[§] Dandan Ma,[§] Chenxi Zhu, Jintao Yang,* Jing Zhang, and Jie Feng*Cite This: *ACS Omega* 2023, 8, 5731–5741

Read Online

ACCESS |



Metrics & More

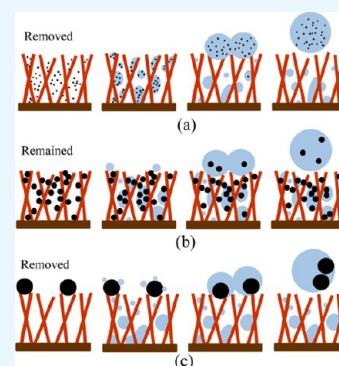


Article Recommendations



Supporting Information

ABSTRACT: It is well known that superhydrophobic surfaces (SHSs) possess self-cleaning ability, either by impacting or rolling water droplets or by self-propelled jumping condensate. However, contaminants that are present in the air are various. Is it possible that these contaminants can all be removed from SHSs by jumping condensate? In this study, hydrophilic SiO₂ micro- or nanoparticles with diameters larger than, comparable to, and smaller than the width of the nanogaps of the SHS were first filled in the nanogaps or suspended on the nanostructures with the help of ethanol, and the resulting SHS was exposed to condensing water vapor. Direct observation through microscopy showed that jumping condensation was still obvious on the SHS that were capped or filled with micro- or nanoparticles. Scanning electron microscopy (SEM) imaging demonstrated that following jumping condensation, particles that possessed diameters significantly smaller or larger than the width of the nanogaps were both removed from the SHS. However, most particles possessing diameters comparable to the width of the nanogaps remained on the SHS. This confirms for the first time that not all contaminants or dust can be removed from an SHS by self-propelled jumping condensate. Furthermore, the study also simply demonstrates that vapor condensation occurs within the nanogaps of the SHS. This study is helpful in further understanding the mechanism of the self-cleaning caused by jumping condensate and exploring the initial formation of condensate droplets on the SHS.



INTRODUCTION

It is well known that superhydrophobic surfaces (SHSs) possess self-cleaning,^{1–4} antifreezing,^{5,6} antideicing,⁷ microfluid nonloss transportation,⁸ water harvest,⁹ and biochemical separation¹⁰ abilities. The impacting or rolling water such as rain and washing water can bring away dust adhering on SHSs easily, although in a practical environment, due to the complexity of dust particles, the durability of such unique self-cleaning ability is suspectable,^{11–13} from a scientific viewpoint, this phenomenon and its associated mechanism have been widely accepted. In addition to extra water, in situ condensed water drops, here means those showing the jumping or Cassie state,^{14–18} can also bring away the dust deposited on the SHSs.^{19–21} This is another mechanism for the self-cleaning of SHSs, which was demonstrated by Chen et al. for the first time.¹⁹ Their findings offer new insights for the development of self-cleaning materials.

However, the diameters of dust particles used in the study of self-cleaning by jumping condensate are all much larger than the feature width and spacing (~200 nm) of the SHSs.^{19–21} Contaminants or dust particles in the atmosphere vary greatly in diameter, and the removal of smaller dust particles by jumping condensate has not been confirmed. Factually, so far, little study has been focused on the effect of dust size on the self-cleaning ability caused by jumping condensates even by rain or rinsing water.²² Can all sizes of dust particles be brought away by the jumping condensed water easily? Is it possible that some dust particles just are stuck between the micro- or nanostructures of

SHSs and thus could not be brought away by the jumping condensates from the SHSs easily?

In the past 14 years, more attention has been focused on the SHSs on which condensate droplets can spontaneously jump upon coalescence with each other.^{5,6,14–18,23} However, the mechanism of droplet nucleation, growth, and especially ascending in nanogaps before mobile coalescence, e.g., the location of droplet nucleation, have not yet been demonstrated simply due to the low definition and magnification in observing such phenomena using environmental scanning electron microscopy (E-SEM).^{24,25} Does the nucleation occur on the bottom of nanogaps, wall of the nanostructure, and tip of the nanostructure homogeneously? Or does it occur preferentially on the condensate droplets once the condensation occurs?

In this study, hydrophilic nanoparticles as dust models, with diameters larger, comparable to, and smaller than the width of nanogaps of the SHS, were first filled in the nanogaps of the SHSs or suspended on the nanostructures of the SHSs with the aid of ethanol. The resulting SHSs were then exposed to vapor condensation or water rinsing. Microscopic observations show

Received: November 15, 2022

Accepted: January 23, 2023

Published: February 6, 2023



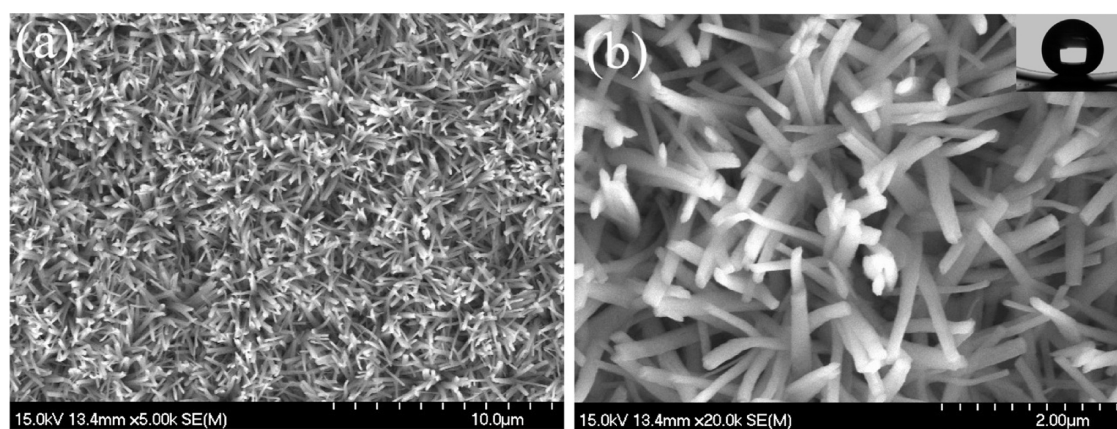


Figure 1. SEM images of copper surfaces after being immersed in 2.5 M NaOH and 0.1 M $(\text{NH}_4)_2\text{S}_2\text{O}_8$ at 4 °C for 1 h, followed by drying at 180 °C for 2 h and then treatment with FAS17 for 1 h. (b) Magnified view of (a). The inset shows the profiles of the 4 μL sessile water droplet with CA at 160°.

that jumping condensate is still present on the SHS. After condensation, particles possessing a diameter size significantly smaller or larger than the width of the nanogaps were both removed from the SHS. However, most particles with a diameter comparable to the width of the nanogaps remained on the SHS. This confirms for the first time that not all dust particles can be removed by self-propelled jumping condensates. Moreover, the study also simply demonstrates that vapor condensation occurs within the nanogaps of the SHS.

MATERIALS AND METHODS

Preparation of the Copper Superhydrophobic Surface. The surface of copper foils was superhydrophobilized according to procedures described in our previous studies.¹⁴ Briefly, copper foils with a size of 30 mm \times 60 mm \times 0.5 mm (purity 99.99%, Aldrich) were first washed by an ultrasonic bath with ethanol and deionized water for 5 min, separately, and then immersed in a 4 M HCl aqueous solution for 5 s to remove surface oxide. After that, the copper foils were incubated in an aqueous solution composed of 2.5 M NaOH and 0.1 M $(\text{NH}_4)_2\text{S}_2\text{O}_8$ at 4 °C for 1 h. Then, the blue copper foils were rinsed with deionized water and dried at 180 °C for 2 h. Subsequently, the copper foils were immersed at room temperature for 60 min in a 0.5 wt % hexane solution of 1H,1H,2H,2H-perfluorodecyltriethoxysilane (FAS17, Sicong New Material Ltd., Quanzhou, China) and then dried at 120 °C for 1 h.

Preparation of Silica Nano- and Microparticles. Monodisperse spherical silica particles were prepared using the classic Stöber method.²⁶ Briefly, for the preparation of silica particles with a diameter of 100 nm, in a 60 °C water bath, 9 mL of ammonia ($\text{NH}_3 \cdot \text{H}_2\text{O}$, 28 wt %) was first mixed with 100 mL of alcohol in a flask to form an alcoholic ammonia solution. Then, 9 mL of tetraethylorthosilicate (TEOS) was added to the flask with rapid stirring. After the reaction for 17 h, the resulting suspension was evaporated and centrifuged (4000 r/min) to decant most of the supernatant. The sediment was purified twice by resuspending and centrifugation using alcohol as a medium and finally stored in alcohol. To prepare 300 nm silica, a procedure identical to that described above was conducted, except the temperature of the water bath was 25 °C and the volume of ammonia was 15 mL. To prepare 1 μm silica, a seeding growth method was adopted. Briefly, 2 g of 300 nm silica particles as nucleation seeds were first filled in the above reaction solution for the preparation of 300 nm silica particles. After 17 h,

the resulting suspension was evaporated and centrifuged (2000 r/min) to give larger silica particles (with a diameter of 0.8–1.0 μm) and then purified and stored in alcohol.

Characterization of SHS and Silica Particles. The surface morphology of the resulting copper foils was scanned by field emission scanning electron microscopy (FE-SEM, S4700, Hitachi, Japan). A software named ImageJ was used to analyze the average diameters of nanoribbons and the width of the nanogaps. The water contact angles (CAs) and slide angles (SAs) of the surfaces were measured by a contact-angle system (Dataphysics OCA35, Germany). A 4 μL droplet was used. The CA and SA values are the averaged values of five different measuring points on each surface. The diameter and morphology of the synthesized silica particles were characterized by transmission electron microscopy (FEI, Tecnai G2 F30 S-Twin). The morphologies of the resulting copper SHSs filled with or covered by silica particles, before and after water vapor condensation, were characterized by the above FE-SEM.

Condensation Experiments. Obviously, using optical microscopy directly to observe vapor condensation under ambient conditions is more valuable than using environmental SEM. A closed room with a height of 3 m and an area of 25 m² was selected to perform the condensation experiments. Both the ambient temperature and the relative humidity (RH) were stable, respectively, at 26 ± 1 °C and $96 \pm 1\%$. An aluminum block was horizontally deposited and almost completely immersed in a mixture of ice and water (0–1 °C). Then, copper foils, with the size of 3 cm \times 3 cm \times 0.5 mm, were placed on the aluminum block. This ensured the stability of the copper foils, and thus, the following video could be focused on the same area. An optical 10 \times objective microscope (CVM-600E, Shanghai Changfang, China), together with a 25 fps charge-coupled device (CCD) camera, was used to observe and visualize the spontaneous motion of condensate droplets. All four videos of the condensation on the SHSs without or with dust particles were analyzed to quantify the spontaneous motion phenomenon. Five short periods of time (only 1 s), e.g., at 0, 15, 30, 45, and 60 s, were selected for statistics. The average number of discriminable droplet position changes (appearing or disappearing in continuous microscopic images) in the 1 s video was referred to here as “spontaneous motion frequency.”

To deposit silica particles on the SHSs, the silica particles/alcohol suspensions obtained from the above procedures were adjusted to a concentration of ~ 10 mg/mL and then dropped onto the copper foil SHSs (~ 0.05 mL/cm²). After drying at

room temperature and atmospheric pressure, the copper foils were exposed to water vapor condensation for 1 or 2.5 h at ambient RH and room temperature. As a control experiment, the copper foils filled with silica particles were also rinsed with flowing water at a speed of 750 mL/min and 7.5 kPa pressure for 1 h to compare the differences in removing silica particles using these two methods. To explore the removal mechanism of the silica particles by jumping condensates, two different condensation methods, one in which the SHS was placed horizontally and the other in which the SHS was placed vertically, were performed.

RESULTS AND DISCUSSION

Morphology and Wettability of the Prepared Surfaces. Figure 1 shows the top-view morphology of the prepared copper surface. We can see that dense nanoribbons with diameters of ~ 100 nm and lengths of ≥ 1 μm are present on the surface. The gap widths between these nanoribbons are less than 1 μm (0.3–1.0 μm). The superhydrophobicity of the prepared surfaces was confirmed by measuring water CAs and SAs. The results show that the CA and SA of a 4 μL water droplet on the copper surface are 159.0 ± 0.3 and 2° , respectively. This confirms that the prepared surfaces are typically superhydrophobic. In our previous studies,^{14,15,27} we confirmed that SHSs possessing such nanostructures generally lead to obvious jumping condensate when exposed to condensing water vapor.

Cassie Condensation before and after Silica Particle Deposition. Figure 2 and Movie S1 show the time-lapse top-

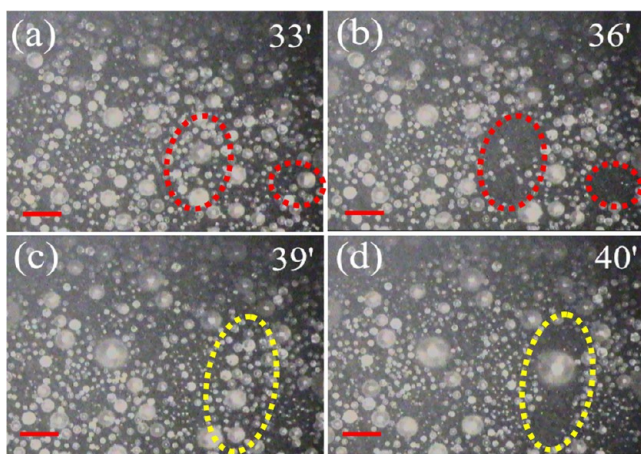


Figure 2. Time-lapse optical images (top-view) of vapor condensation on the horizontally placed copper SHS without silica particles at (a) 33 s, (b) 36 s, (c) 39 s, and (d) 40 s. The surface temperature is 0°C , and the ambient RH is $96 \pm 1\%$ ($26 \pm 1^\circ\text{C}$). The coalescence and spontaneous motion of condensate droplets are obvious. The scale bar is 100 μm .

view optical images of vapor condensation on the copper SHS without deposition of micro- and nanoparticles of silica, clearly showing that the Cassie condensation phenomenon occurs on the SHS, i.e., the rapid removal of condensate droplets was observed (see the dashed circles with the same color, the time scale is in seconds). Due to the substantial reduction of the total liquid–air interfacial area, the surface energy released upon drop mobile coalescence gives power to the spontaneous motion of condensate drops.^{19,28}

In our previous studies,^{14,15} we speculated and indirectly demonstrated the microscopic mechanism of the Cassie

condensation on the SHS, i.e., nucleation occurs at the top, side, and bottom of the nanostructures at the early beginning of vapor condensation. At a slightly later stage, the growth of the droplet is dominated by continuous vapor condensation and droplet coalescence until the gaps are filled. Subsequently, a Wenzel droplet, covering more than one gap with an external and relatively flat surface, forms. Finally, the Laplace pressure caused by the interconnected and hydrophobic narrow gaps forces the Wenzel droplets to ascend and rapidly become Cassie droplets. It is the coalescence between the Cassie droplets that initiates the jumping phenomenon. However, as the droplets formed during the Wenzel-to-Cassie transition are so small, it is extremely difficult to observe them directly.

Herein, the deposition of silica particles on the SHS provides a new insight for studying the microscopic mechanism of Cassie condensation. Figure 3 and Movies S2–S4 show the time-lapse

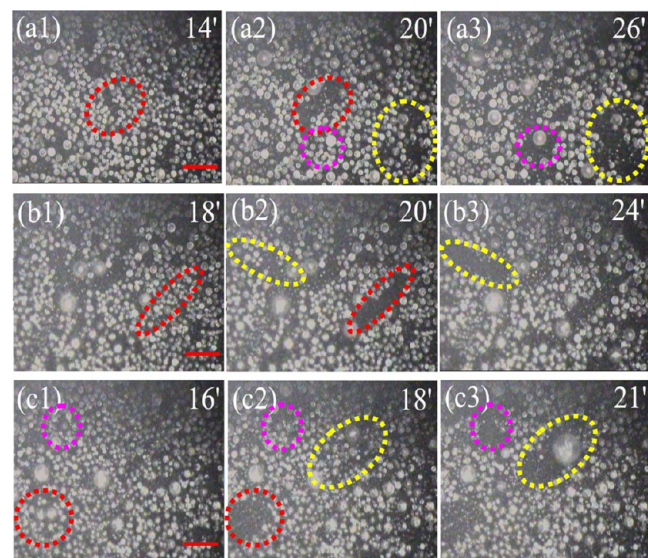


Figure 3. Time-lapse optical images (top-view) of vapor condensation on the horizontally placed copper SHS, followed by the deposition of silica particles with diameters of 100 nm (a), 300 nm (b), and 1 μm (c). The surface temperature is 0°C , and the ambient RH is $96 \pm 1\%$ ($T = 26 \pm 1^\circ\text{C}$). The coalescence and spontaneous motion of condensate droplets are obvious. The scale bar is 75 μm .

top-view optical images of vapor condensation on the copper SHSs being deposited with hydrophilic micro and nanoparticles of silica, clearly showing that in a 1 h experiment, the jumping condensate is still obvious on the SHSs being deposited with all three types of silica particles. The condensate droplets on the SHSs continuously depart from their original locations via coalescence with neighboring droplets (see the dashed circles with the same color, the time scale is in seconds). This departure exposes a fresh surface for the formation of new droplets. Compared with Figure 2, it appears that after the deposition of hydrophilic silica particles, the spontaneous motion frequency¹⁴ changes little, and the corresponding spontaneous motion frequencies of condensate drops were 72, 71, 69, and 70 drops/s (Table 1). In theory, hydrophilic dots on the SHS should accelerate vapor nucleation and result in higher spontaneous motion frequency;²⁹ the gravity of the particles may counteract such acceleration.

Self-Cleaning of SHS by Jumping Condensates. Using a cicada wing as a model surface, Chen et al. demonstrated that the self-propelled jumping condensate on the SHS is an effective

Table 1. Spontaneous Motion Frequencies of Condensate Drops on the SHSs with or without Hydrophilic Silica Particles

particle diameter (nm)	spontaneous motion frequencies of condensate drops (drops/s)
no particle	72
100 nm	71
300 nm	69
1000 nm	70

tool for the removal of a variety of contaminants, including both hydrophobic/hydrophilic and organic/inorganic particles.¹⁹ However, the contaminant models they used consist of hydrophilic glass particles (diameter $50\ \mu\text{m}$), silica particles ($20\ \mu\text{m}$), less hydrophilic poly(methyl methacrylate) (PMMA)

particles ($100\ \mu\text{m}$), pollen particles ($\sim 20\ \mu\text{m}$), and hydrophobic polystyrene particles ($8\ \mu\text{m}$). These particles are all much larger than the surface gap width of the cicada wing. This raises the question of how particles with diameters much smaller than or approximately comparable to the gap width can be autonomously removed by the jumping condensate.

Figure 4 shows the morphology of the copper SHS being filled with hydrophilic SiO_2 particles (diameter $100\ \text{nm}$) before (a, b) and after 1 h of Cassie condensation (c, d) or 1 h of water rinsing (e, f). As the silica nanoparticles are much smaller than the gap widths ($0.3\text{--}1.0\ \mu\text{m}$) of the SHS, all of the particles fall into the nanogaps of the SHS with the help of alcohol. However, after 1 h of Cassie condensation, no particles could be found on the SHS. The self-cleaning effect is very good. It is clear that these nanoparticles are removed from the SHS by the jumping

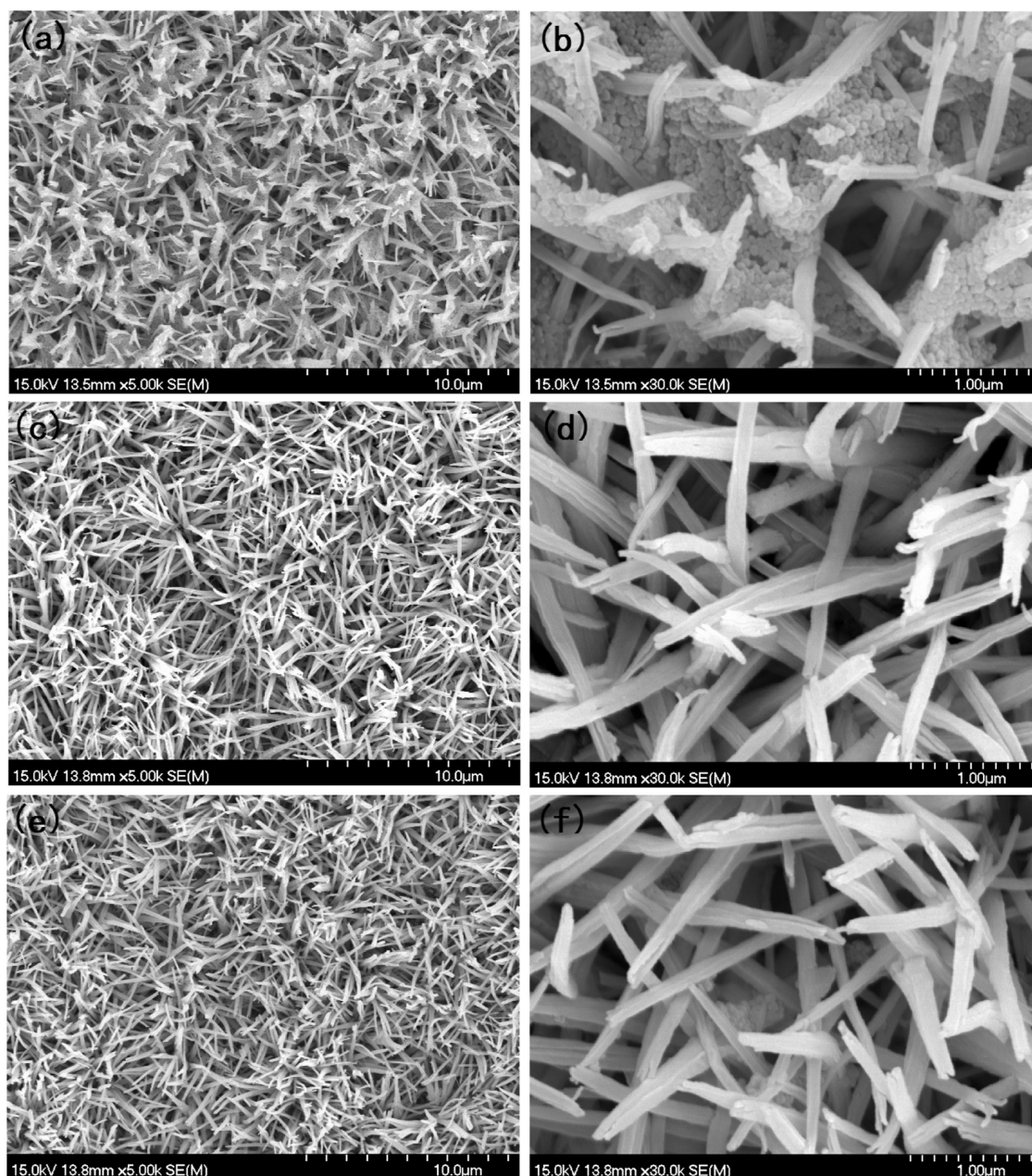


Figure 4. SEM of the copper SHS being filled with SiO_2 nanoparticles (diameter $100\ \text{nm}$) before (a, b) and after 1 h of Cassie condensation (c, d) or 1 h of water rinsing (e, f). The SHS temperature is $0\ ^\circ\text{C}$, and the ambient RH is $67 \pm 1\%$ ($T = 23 \pm 1\ ^\circ\text{C}$).

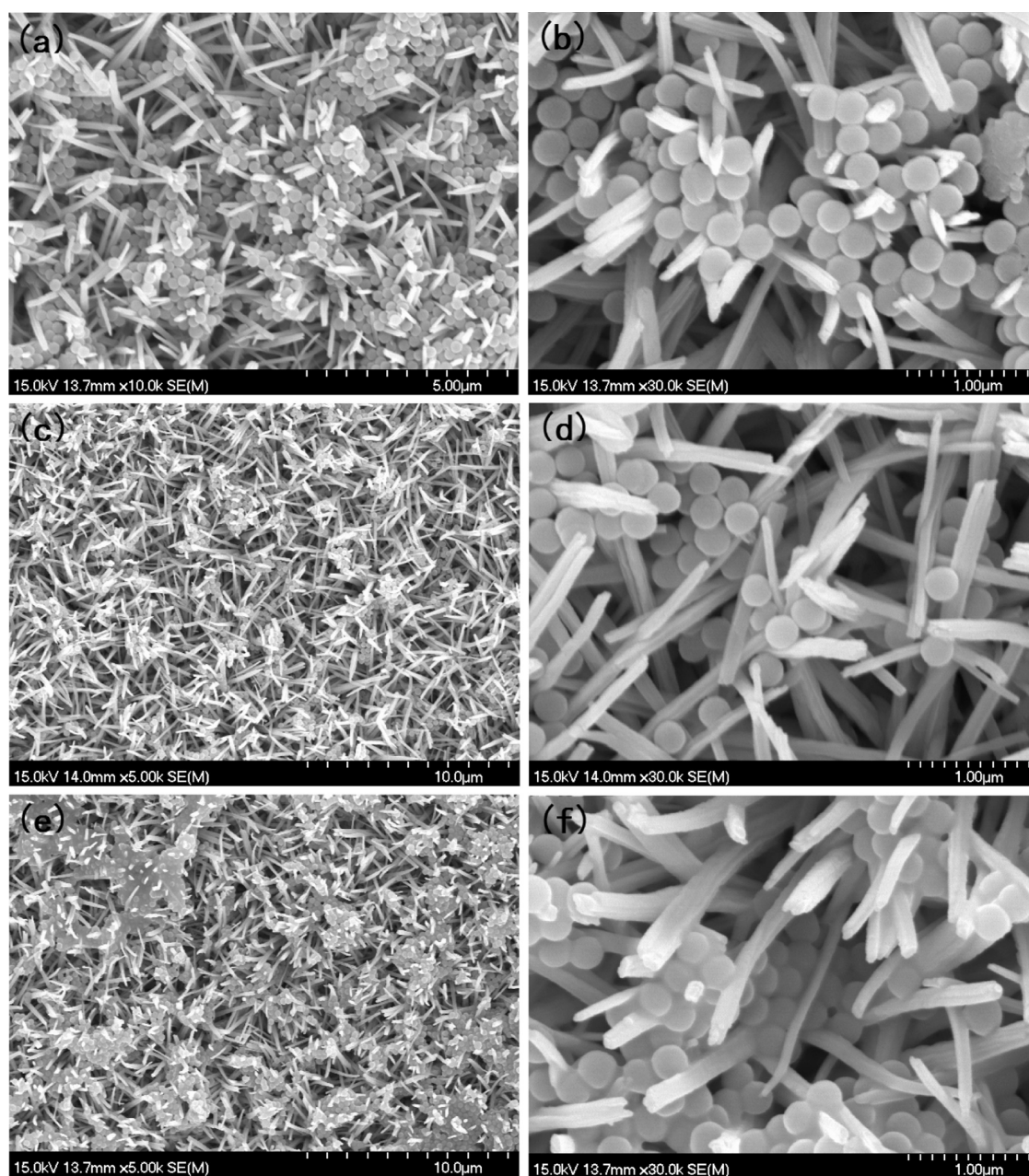


Figure 5. SEM of the copper SHS being filled with SiO₂ nanoparticles (diameter 300 nm) before (a, b) and after 2.5 h of Cassie condensation (c, d) or rinsing with water for 1 h (e, f). The SHS temperature is 0 °C, and the ambient RH is $67 \pm 1\%$ ($T = 23 \pm 1$ °C).

condensate. However, differing from the self-cleaning mechanism proposed by Chen et al. in which large microparticles are removed from the SHS by floating, lifting, and aggregating forces,¹⁹ the nanoparticles in our study are removed by an embedding force (Figure 7a). The vapor first condenses on the hydrophilic nanoparticles, and then the nanoparticles that are embedded in the condensate droplets ascend to the top of the nanoribbons, finally forming water/nanoparticle composite Cassie droplets. When these droplets coalesce with a neighboring droplet, the surface energy released upon coalescence triggers an out-of-plane jumping or sweeping condensate that carries away the embedded nanoparticles from the nanogaps of the SHS.

Recently, Gao et al.³⁰ fabricated a superhydrophobic surface with spatially heterogeneously patterned superhydrophilic microdots by a mask-assisted photodegradation method.

Based on such a novel surface, they realized confined growth, more efficient coalescence, and self-ejection of condensate droplets. In our study here, during vapor condensation nucleation, the hydrophilic silica nanoparticles deposited on the copper SHS play the same role as superhydrophilic microdots in the work of Gao et al.³⁰ The difference is that the hydrophilic particles were finally removed away from the SHS, while the superhydrophilic microdots could not be removed away.

A similar result was also obtained by rinsing the SHS filling with SiO₂ particles (100 nm) for 1 h (Figure 4). Moreover, the self-cleaning effect caused by the Cassie condensation appears to be a little better than rinsing with water. This demonstrates that particles smaller than the gap width of the SHS can be effectively removed from the SHS by jumping condensates. However, for particles with diameter sizes comparable to the width of the

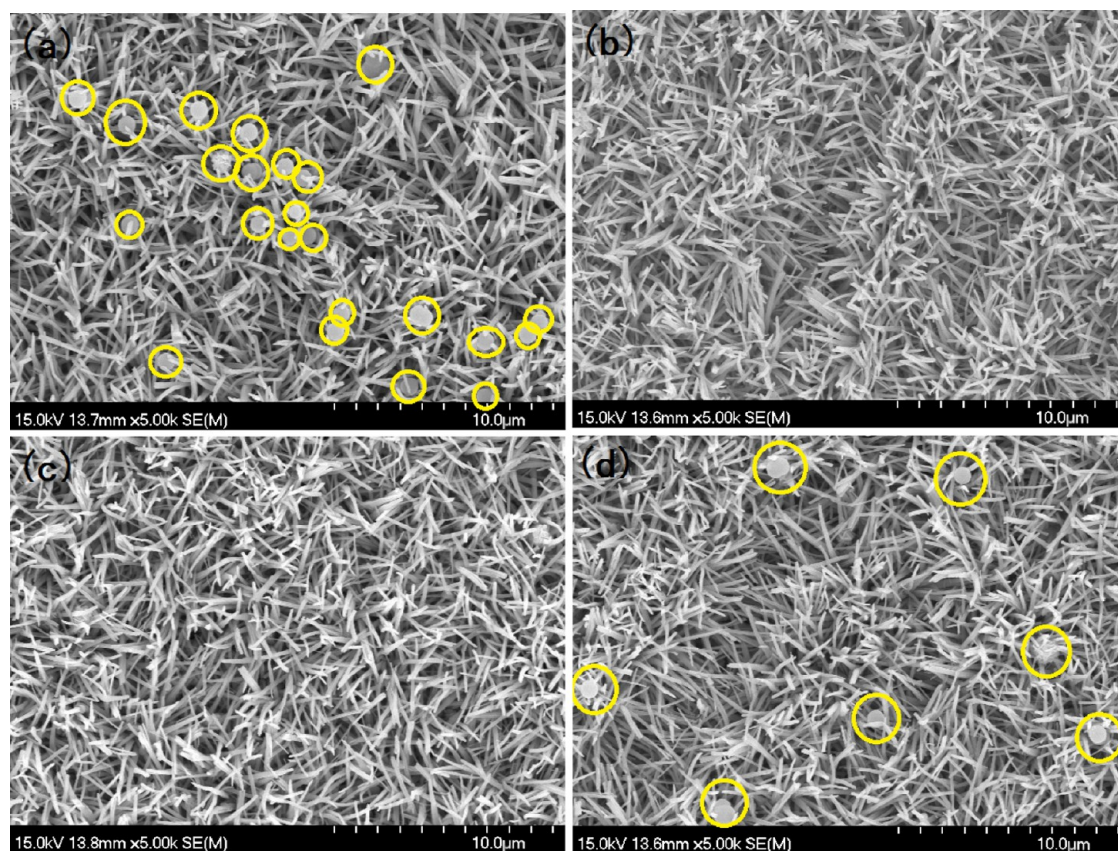


Figure 6. SEM of the copper SHS being filled with SiO₂ nanoparticles (diameter 1 μm) before (a) and after 1 h of Cassie condensation (b), 2.5 h of Cassie condensation (c), or 1 h of water rinsing (d). The SHS temperature is 0 °C, and the ambient RH is 67 ± 1% ($T = 23 \pm 1$ °C).

nanogaps, e.g., particles with a diameter of 300 nm, the self-cleaning result is quite different (Figure 5). First, most of the gaps on the SHS are filled with silica particles. Moreover, because the surface of silica particles is hydrophilic, agglomeration did occur during alcohol volatilization after the silica particles/alcohol suspensions were dropped onto the SHS. The agglomeration formed larger particles. Not all of the gaps have an open and wide entrance. Particles can be trapped by gaps with narrow tops and wide bottoms created by declining and overlapping nanoribbons. Following 1 h of Cassie condensation, the number of particles on the SHS decreases slightly. Only particles present on the SHS are removed. Following 2.5 h of Cassie condensation, many particles are still embedded in the gaps of the SHS. A similar result was obtained after rinsing with water for 1 h, i.e., many particles are still embedded in the gaps of the SHS.

Particles with diameters (~ 1 μm) significantly larger than the width of the nanogaps of the SHS are all capped on top of the nanoribbons (Figure 6). However, after rinsing with water for 1 h, many particles remain on the SHS. Contrastively, after only 1 h of Cassie condensation, these larger particles are all removed from the SHS. From the microscopic viewpoint, the vapor may condense simultaneously on the hydrophilic surface of these large particles and in the nanogaps of the SHS. Then, the condensate droplets around the particles and in the nanogaps grow, merge, and finally form Cassie droplets on the SHS. The surface energy released upon coalescence of these Cassie droplets results in the removal of the larger silica particles (Figure 7c). On the contrary, when the SHS was washed, due to the presence of water pressure, some particles with a diameter of

1 μm may be pressed into the gaps on the surface, e.g., cannot be removed effectively. Another reason is that these residual larger particles may be locked by the declining nanoribbons. It must be noted that since the concentration of silica particles is ~ 10 mg/mL, the number of 1 μm silica particles in Figure 6 is smaller than those in Figures 4 and 5.

The dimensions matching the particles' diameters and gap widths should be responsible for such firm locking or unlocking, i.e., the particles with diameters comparable to the width of the nanogaps are locked in the gaps by the nanoribbons of the SHS (Figure 7b), while particles with diameters significantly smaller or larger than the width of the nanogaps can be easily removed from the SHS (Figure 7a,c). The quantification of the above matching or mismatching is listed in Table 2.

Because the surface of the silica particles is hydrophilic, agglomeration did occur during alcohol volatilization after the silica particles/alcohol suspensions were dropped onto the SHS. The agglomeration may form larger particles. As a result, even though the gaps definitely look bigger (Figure 5), most silica particles with a 300 nm diameter still could not be removed away by jumping condensates. Silica particles with diameters of 100 nm or 1 μm also agglomerate; however, the jumping condensates can weaken such agglomeration and then remove the silica particles away from the SHS.

To further explore the mechanism of removal of the silica particles by Cassie condensation, the SHS was placed vertically. The results showed that for both 100 nm and 1 μm silica particles, vertical jumping condensation appears to be less effective than horizontal jumping condensation for particle removal (Figures 8 and 9). This is because in horizontal

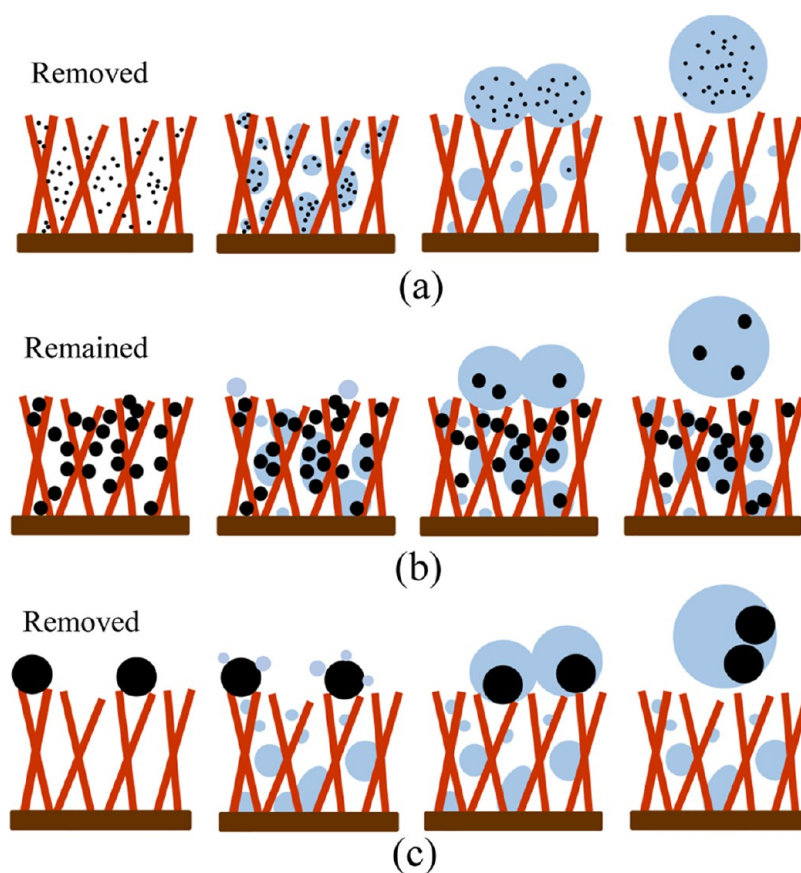


Figure 7. Self-cleaning mechanism of the silica nano- and microparticles with diameters significantly smaller (a), comparable to (b), and significantly larger (c) than the width of the nanogaps of the SHS. Only particles with diameters smaller or larger than the width of the nanogaps of the SHS could be removed by jumping condensates. The particles with diameters comparable to the width of the nanogaps of the SHS are mostly locked by the nanoribbons.

Table 2. Matching of the Particles' Diameters, Nanogap Widths, and Self-Cleaning or Not

widths of the nanogap (nm)	particle diameter (nm)	self-cleaned or not
300–1000	100	yes
300–1000	300	no
300–1000	1000	yes

condensation, in addition to jumping condensates, there is a sweeping condensation effect, i.e., some of the droplets roll off the surface upon coalescence and sweep up other droplets in the rolling path.³¹ In vertical condensation, once they emerged, the condensate droplets would self-ejected and fell down directly due to gravity. As a result, there is less sweeping condensation, and thus, the efficiency of silica particle removal is lower.

In 2018, Zhang et al.²⁰ fabricated SHSs with nanopore structures on glass and then investigated particle removal behavior from the SHSs via jumping condensate for different particle numbers and sizes. They find that smaller particles, which could not be removed by the action of vibration, gravity, and wind, can be removed away via the self-jumping of water droplets. However, the particles they adopted are much larger than the width of nanogaps on the SHSs. Ding et al.²¹ fabricated hydrophilic, hydrophobic, and SH coatings on copper and studied the effect of the surface wettability on dust removal by condensate water. They found that the remnant dust weight on the SH surface is 69.9% lower than that on the uncoated surface, 61.1% lower than that on the hydrophilic surface, and only

3.59% lower than that on the hydrophobic surface. This means that the SH has no obvious advantage in antisoiling by condensate water. Moreover, they seem to omit studying the relationship between the diameter of dust particles and the SH characteristic morphology.

Lyons et al.³² think that condensation exacerbates soiling rates and leads to stronger adhesion, which makes cleaning more expensive. They found that hydrophobic and SH-coated glass exhibited 42% lower soiling rates than hydrophilic and bare glass in the presence of condensed water. The hydrophobic surfaces were also easier to clean using only water. Huang et al.³³ prepared an SHS and deposited dust on it. They studied the dust removal process on a condensation visualization platform and found the removal included three consecutive processes, e.g., dust agglomerating, condensate sliding, and rolling with jumping condensate. Over 95% of particles can be removed by condensation within 90 min condensation. However, the dust they used is far larger than the width of nanogaps on the SHS.

Some literature studies^{34–36} show that the sizes of dust particles in the daily environment from different countries are generally micro-sized and submicro-sized. Figgis et al.³⁴ reported that the dominant size range of particles accumulating on outdoor surfaces is 10–30 μm in the U.S.A. (Chicago) and 8–16 μm in Kuwait. In our previous work,³⁵ we analyzed the size of dust particles accumulating in Hangzhou, China, showing that the minimum particle size was about 500 nm. When the width of nanogaps is far below 500 nm, the dust in the daily environment of Hangzhou can be cleaned by jumping condensation.

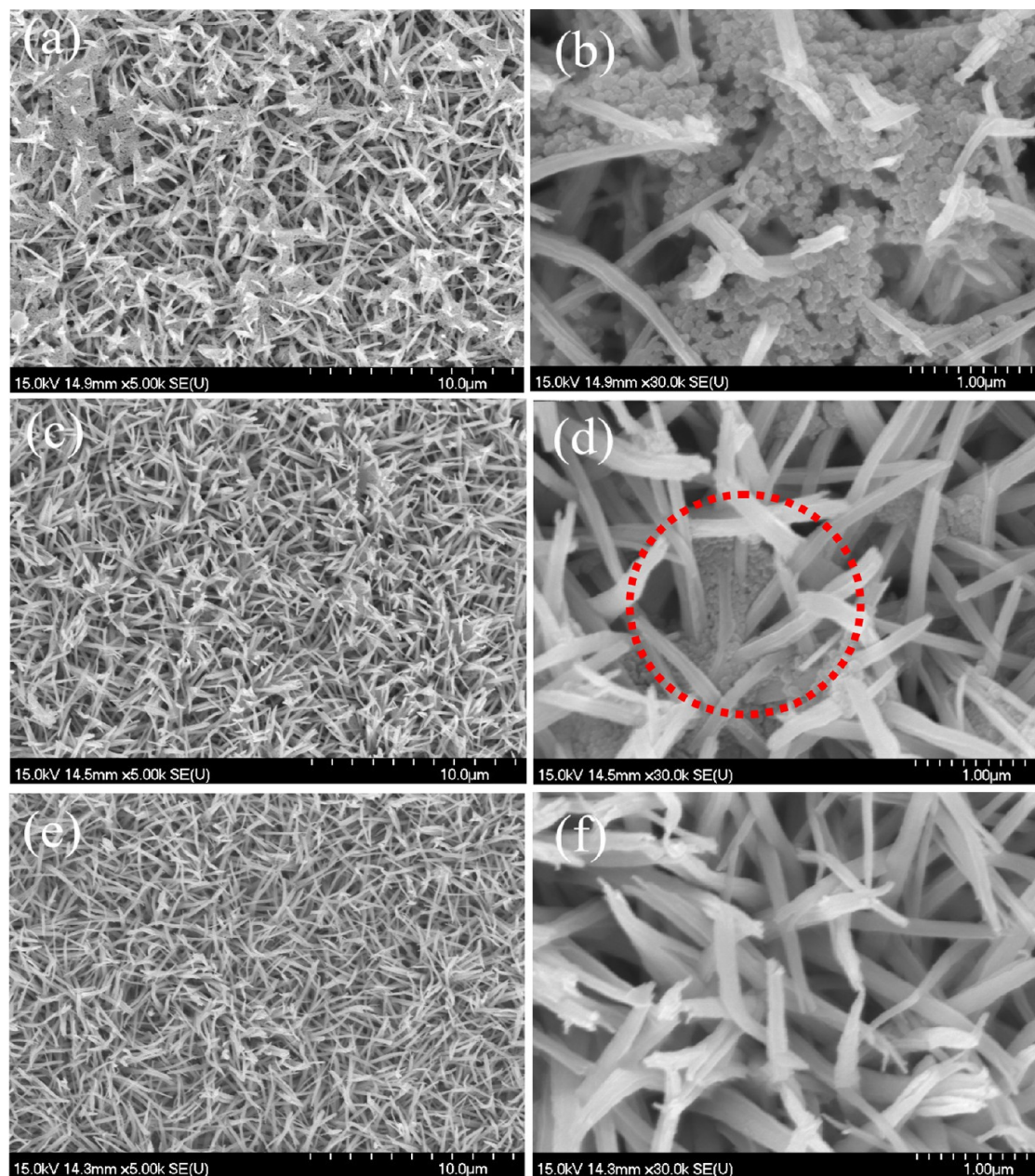


Figure 8. SEM of the copper SHS being filled with SiO₂ nanoparticles (diameter 100 nm) before (a, b) and after 1 h of Cassie condensation vertically (c, d) and horizontally (e, f). The SHS temperature is 0 °C, and the ambient RH is 65 ± 1% ($T = 26 \pm 1$ °C).

CONCLUSIONS

In summary, condensation-induced self-cleaning of dust on the SH surface has attracted worldwide interest. However, so far, almost all of the relative conclusions are positive, e.g. jumping condensates can bring away all sizes of dust from the SH surface. Since contaminants or dust particles in the atmosphere vary greatly in diameter, it is a pity that the effect of dust size on the self-cleaning ability caused by jumping condensates even by rain or rinsing water had been ignored. To our knowledge, our study here is the first one focusing on the relationship between the diameter of the dust deposited and the removal efficiency. Through systematic studies concerning the removal behavior of silica particles with different diameters by jumping condensates from an SHS, we not only confirm that the nucleation location of water vapor condensation on the SHS occurs within the

nanogaps but also show for the first time that not all sizes of dust can be removed by self-propelled jumping condensates.

Only particles with diameters smaller or larger than the widths of the nanogaps of the SHS could be removed by jumping condensation. Nanoparticles with diameters comparable to the width of the nanogaps of the SHS could not be removed easily by jumping condensates or even by impacting water drops, due to the nanoparticles being locked in place by the nanoribbons. This implies that for SHSs to be used outdoors, the nanogap width of the SHS should be designed to avoid the dust particle size. For example, if most of the dust in the air is 0.5 μm in diameter, the nanogap width of the SHS should be designed to avoid such size thinking of self-cleaning by jumping condensates. Another question in self-cleaning by jumping condensates is where was the dust brought away from the SH surface? Is it the jumping condensates that re-evaporate into the air near the cool SHS,

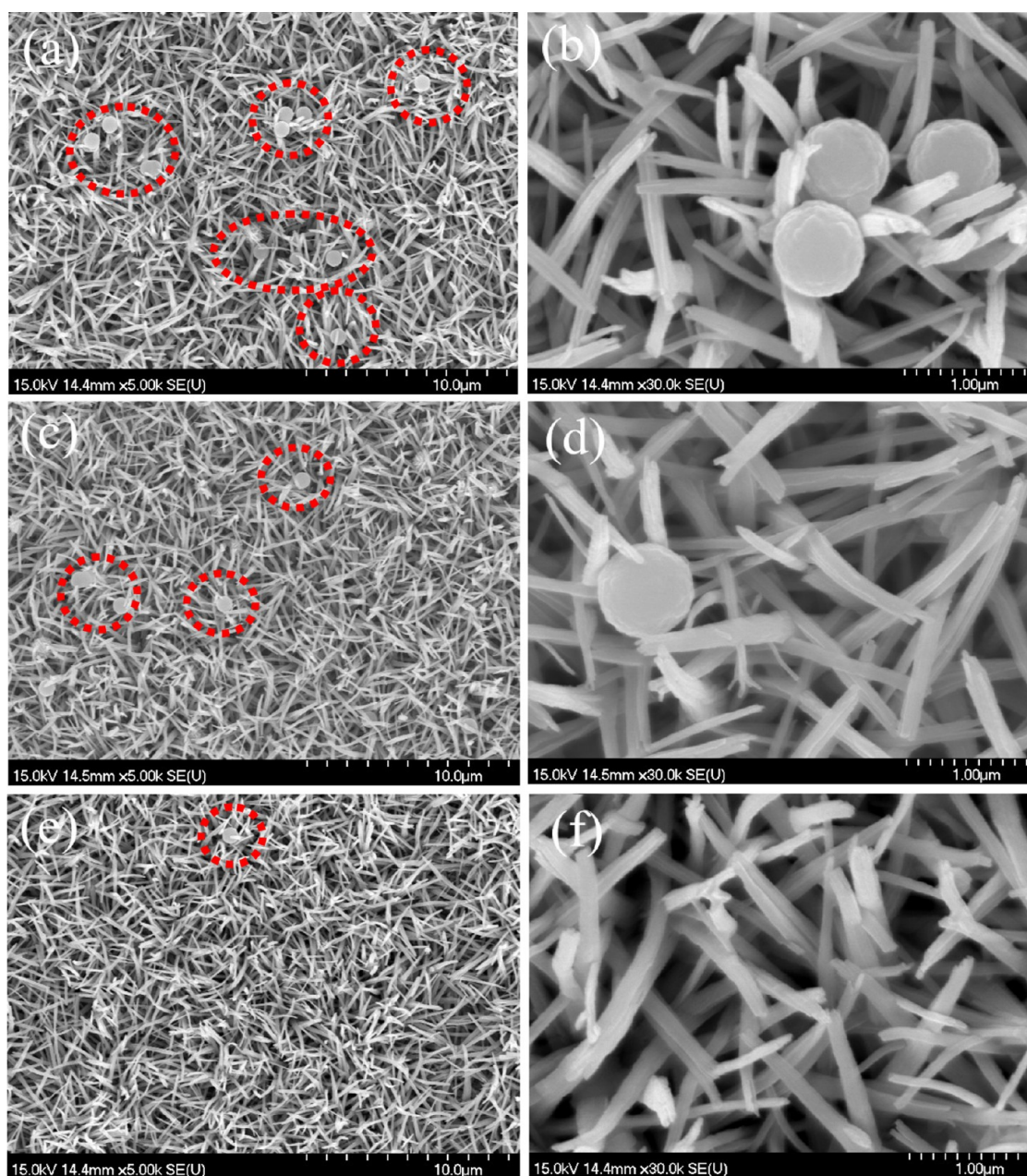


Figure 9. SEM of the copper SHS being filled with SiO₂ nanoparticles (diameter 1 μm) before (a, b) and after 1 h of Cassie condensation vertically (c, d) or horizontally (e, f). The SHS temperature is 0 °C, and the ambient RH is 65 ± 1% ($T = 26 \pm 1$ °C).

conveniently bringing the particles into the air and then making them being brought away from the SHS by the wind? This is the study we will carry out in the future.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsomega.2c07328>.

Water vapor condensation on the copper SHS (Movie S1) (MP4)

Vapor condensation on the SHS being deposited with 100 nm particles (Movie S2) (MP4)

Vapor condensation on the SHS being deposited with 300 nm particles (Movie S3) (MP4)

Vapor condensation on the SHS being deposited with 1 μm particles (Movie S4) (MP4)

■ AUTHOR INFORMATION

Corresponding Authors

Jintao Yang – College of Materials Science and Engineering, Zhejiang University of Technology, Hangzhou 310014, China; orcid.org/0000-0002-3133-1246; Email: yangjt@zjut.edu.cn

Jie Feng – College of Materials Science and Engineering, Zhejiang University of Technology, Hangzhou 310014, China; orcid.org/0000-0001-7228-117X; Email: fengjie@zjut.edu.cn

Authors

Kangning Li – College of Materials Science and Engineering, Zhejiang University of Technology, Hangzhou 310014, China; Jinhua Polytechnic, Jinhua 321007, China

Dandan Ma – College of Materials Science and Engineering, Zhejiang University of Technology, Hangzhou 310014, China
Chenxi Zhu – College of Materials Science and Engineering, Zhejiang University of Technology, Hangzhou 310014, China
Jing Zhang – College of Materials Science and Engineering, Zhejiang University of Technology, Hangzhou 310014, China; orcid.org/0000-0002-0245-7149

Complete contact information is available at:
<https://pubs.acs.org/10.1021/acsomega.2c07328>

Author Contributions

[§]K.L. and D.M. contributed equally to this work. K.L.: investigation and writing—original draft. D.M. and C.Z.: data curation and investigation. J.Y. and J.Z.: supervision. J.F.: conceptualization, supervision, writing—review & editing, and project administration.

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

The authors gratefully acknowledge financial support from the National Natural Science Foundation of China (Nos. 51172206 and 52173085) and the Zhejiang Provincial Natural Science Foundation of China (No. LQ18E010005).

ABBREVIATIONS

CAs, contact angles
CCD, charge-coupled device
E-SEM, environmental scanning electron microscopy
FAS17, 1H,1H,2H,2H-perfluorodecyltriethoxysilane
FE-SEM, field emission scanning electron microscopy
PMMA, poly(methyl methacrylate)
RH, relative humidity
SAs, slide angles
SEM, scanning electron microscopy
SHS, superhydrophobic surface
SHSs, superhydrophobic surfaces
TEOS, tetraethylorthosilicate

REFERENCES

- (1) Latthe, S. S.; Sutar, R. S.; Kodag, V. S.; Bhosale, A. K.; Kumar, A. M.; Sadasivuni, K. K.; Xing, R.; Liu, S. Self-cleaning superhydrophobic coatings: Potential industrial applications. *Prog. Org. Coat.* **2019**, *128*, 52–58.
- (2) Sun, Y.; Guo, Z. A scalable, self-healing and hot liquid repelling superamphiphobic spray coating with remarkable mechanochemical robustness for real-life applications. *Nanoscale* **2019**, *11*, 13853–13862.
- (3) Wen, G.; Guo, Z.; Liu, W. Biomimetic polymeric superhydrophobic surfaces and nano-structures: from fabrication to applications. *Nanoscale* **2017**, *9*, 3338–3366.
- (4) Kong, X.; Zhang, J.; Xuan, Q.; Lu, J.; Feng, J. Superhydrophobic Coating for Antifouling of Chinese Paintings. *Langmuir* **2018**, *34*, 8294–8301.
- (5) Tian, J.; Zhu, J.; Guo, H.; Li, J.; Feng, X.; Gao, X. Efficient self-propelling of small-scale condensed microdrops by closely packed ZnO nanoneedles. *J. Phys. Chem. Lett.* **2014**, *5*, 2084–2088.
- (6) Zhao, Y.; Luo, Y.; Li, J.; Yin, F.; Zhu, J.; Gao, X. Condensate microdrop self-propelling Aluminum surfaces based on controllable fabrication of Alumina rod-capped nanopores. *ACS Appl. Mater. Interfaces* **2015**, *7*, 11079–11082.
- (7) Mouterde, T.; Lehoucq, G.; Xavier, S.; Checco, A.; Black, C. T.; Rahman, A.; Midavaine, T.; Clanet, C.; Quéré, D. Antifogging abilities of model nanotextures. *Nat. Mater.* **2017**, *16*, 658–663.
- (8) Vilian, A. T. E.; Hwang, S.-K.; Lee, M. J.; Bagavathi, M.; Huh, Y. S.; Han, Y.-K. Facile synthesis of petal-like VS2 anchored onto graphene nanosheets for the rapid sensing of toxic pesticide in polluted water. *Ecotoxicol. Environ. Saf.* **2021**, *228*, No. 113021.
- (9) Zhu, H.; Guo, Z.; Liu, W. Biomimetic water-collecting materials inspired by nature. *Chem. Commun.* **2016**, *52*, 3863–3879.
- (10) Leng, X.; Sun, L.; Long, Y.; Lu, Y. Bioinspired superwetting materials for water manipulation. *Droplet* **2022**, *1*, 139–169.
- (11) Zhang, J.; Zhang, W.; Lu, J.; Zhu, C.; Lin, W.; Feng, J. Aqueous epoxy-based superhydrophobic coatings: Fabrication and stability in water. *Prog. Org. Coat.* **2018**, *121*, 201–208.
- (12) Mortazavi, V.; Khonsari, M. M. On the degradation of superhydrophobic surfaces: A review. *Wear* **2017**, *372–373*, 145–157.
- (13) Dalawai, S. P.; Aly, M. A. S.; Latthe, S. S.; Xing, R.; Sutar, R. S.; Nagappan, S.; Ha, C. S.; Sadasivuni, K. K.; Liu, S. Recent advances in durability of superhydrophobic self-cleaning technology: a critical review. *Prog. Org. Coat.* **2020**, *138*, No. 105381.
- (14) Feng, J.; Qin, Z.; Yao, S. Factors affecting the spontaneous motion of condensate drops on superhydrophobic copper surfaces. *Langmuir* **2012**, *28*, 6067–6075.
- (15) Feng, J.; Pang, Y.; Qin, Z.; Ma, R.; Yao, S. Why condensate drops can spontaneously move away on some superhydrophobic surfaces but not on others. *ACS Appl. Mater. Interfaces* **2012**, *4*, 6618–6625.
- (16) Yan, X.; Zhang, L.; Sett, S.; Feng, L.; Zhao, C.; Huang, Z.; Vahabi, H.; Kota, A. K.; Chen, F.; Miljkovic, N. Droplet jumping: effects of droplet size, surface structure, pinning, and liquid properties. *ACS Nano* **2019**, *13*, 1309–1323.
- (17) Chen, X.; Wang, P.; Zhang, D. Designing a superhydrophobic surface for enhanced atmospheric corrosion resistance based on coalescence-induced droplet jumping behavior. *ACS Appl. Mater. Interfaces* **2019**, *11*, 38276–38284.
- (18) Zhu, Y.; Tso, C. Y.; Ho, T. C.; Leung, M. K. H.; Yao, S. Coalescence-induced jumping droplets on nanostructured biphilic surfaces with contact electrification effects. *ACS Appl. Mater. Interfaces* **2021**, *13*, 11470–11479.
- (19) Wisdom, K. M.; Watson, J. A.; Qu, X.; Liu, F.; Watson, G. S.; Chen, C. Self-cleaning of superhydrophobic surfaces by self-propelled jumping condensate. *Prog. Natl. Acad. Sci. U.S.A.* **2013**, *110*, 7992–7997.
- (20) Zhan, W.; Wang, W.; Xiao, Z.; Yu, X.; Zhang, Y. Water-free dedusting on antireflective glass with durable superhydrophobicity. *Surf. Coat. Technol.* **2018**, *356*, 123–131.
- (21) Yang, Y.; Zhuang, D.; Ding, G. Effect of surface wettability of fins on dust removal by condensate water. *Int. J. Heat Mass Transfer* **2019**, *130*, 1260–1271.
- (22) Farokhirad, S.; Lee, T. Computational study of microparticle effect on self-propelled jumping of droplets from superhydrophobic substrates. *Int. J. Multiphase Flow* **2017**, *95*, 220–234.
- (23) Chen, S.; Wang, R.; Wu, F.; Zhang, H.; Gao, X.; Jiang, L. Copper-based high-efficiency condensation heat transfer interface consisting of superhydrophobic hierarchical microgroove and nancone structure. *Mater. Today Phys.* **2021**, *19*, No. 100407.
- (24) Meng, K.; Fan, W.; Wang, H. Dynamic scenario simulation of dropwise condensation on a superhydrophobic surface with droplet jumping. *Appl. Therm. Eng.* **2019**, *148*, 316–323.
- (25) Wen, R.; Lan, Z.; Peng, B.; Xu, W.; Yang, R.; Ma, X. Wetting transition of condensed droplets on nanostructured superhydrophobic surfaces: coordination of surface properties and condensing conditions. *ACS Appl. Mater. Interfaces* **2017**, *9*, 13770–13777.
- (26) Stöber, W.; Fink, A.; Bohn, E. Controlled growth of monodisperse silica spheres in the micron size range. *J. Colloid Interface Sci.* **1968**, *26*, 62–69.
- (27) Hao, Q.; Pang, Y.; Zhao, Y.; Zhang, J.; Feng, J.; Yao, S. Mechanism of delayed frost growth on superhydrophobic surfaces with jumping condensates: More than interdrop freezing. *Langmuir* **2014**, *30*, 15416–15422.
- (28) Boreyko, J. B.; Chen, C. Self-propelled dropwise condensate on superhydrophobic surfaces. *Phys. Rev. Lett.* **2009**, *103*, No. 184501.

(29) Chen, X.; Wu, J.; Ma, R.; Hua, M.; Koratkar, N.; Yao, S.; Wang, Z. Nanograssed micropyrarnidal architectures for continuous dropwise condensation. *Adv. Funct. Mater.* **2011**, *21*, 4617–4623.

(30) Xing, D.; Wang, R.; Wu, F.; Gao, X. Confined growth and controlled coalescence/ self-removal of condensate microdrops on a spatially heterogeneously patterned superhydrophilic- superhydrophobic surface. *ACS Appl. Mater. Interfaces* **2020**, *12*, 29946–29952.

(31) Mohammadian, B.; Annavarapu, R.; Raiyan, A.; Nemani, S. K.; Kim, S.; Wang, M.; Sojoudi, H. Delayed frost growth on nanoporous microstructured surfaces utilizing jumping and sweeping condensates. *Langmuir* **2020**, *36*, 6635–6650.

(32) Nayshevsky, I.; Xu, Q.; Barahman, G.; Lyons, A. M. Fluoropolymer coatings for solar cover glass: Anti-soiling mechanisms in the presence of dew. *Sol. Energy Mater. Sol. Cells* **2020**, *206*, No. 110281.

(33) Yuan, W.; Liao, Z.; He, K.; Liu, Q.; Huang, S. An experimental investigation on condensation-induced self-cleaning of dust on superhydrophobic surface. *Appl. Surf. Sci.* **2021**, *566*, No. 150702.

(34) Figgis, B.; Ennaoui, A.; Ahzi, S.; Rémond, Y. Review of PV soiling particle mechanics in desert environments. *Renewable Sustainable Energy Rev.* **2017**, *76*, 872–881.

(35) Zhu, C.; Yu, X.; Lv, J.; Zhang, J.; Yang, J.; Hao, N.; Feng, J. Anti-soiling performance of lotus leaf and other leaves after prolonged outdoor exposure. *ACS Appl. Mater. Interfaces* **2020**, *12*, 53394–53402.

(36) Ilse, K. K.; Figgis, B. W.; Naumann, V.; Hagendorf, C.; Bagdahn, J. Fundamentals of soiling processes on photovoltaic modules. *Renewable Sustainable Energy Rev.* **2018**, *98*, 239–254.