Supporting Information

A triple-scale superhydrophobic surface with excellent anti-icing and icephobic performance via ultrafast laser hybrid fabrication

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Figure S1. The XRD result of the prepared triple-scale micro/nanostructure surfaces



Figure S2. The water sessile drops illustrating contact angle and sliding angle of the resultant MNGF superhydrophobic surface (the uncertainties of the correlation evaluations can be found in Experimental Section)

Surfaces -	Different Critical Laplace Pressure (Pa)			Wenzel state
	P_{CCA}	P_{CTD}	P_{CC}	
MNGF	1549	1446	1446	No
MNSF	1460	1450	1450	No

Table S1. P_{CCA} , P_{CTD} and P_{CC} of different hierarchical surfaces and whether Wenzel state occurs or not (All the measured/calculated numerical values are averages. The uncertainties can be found in *Methods Section*.)

Note: In Table S1, P_{CCA} denotes the critical Laplace pressure for CA when the transformation from superhydrophobic to hydrophobic state happens, i.e. the CA starts to be lower than 150°. P_{CTD} is the critical Laplace pressure for TPCL diameter (P_{CTD}) at the onset of its asymptotic behavior, i.e. when TPCL diameter starts to be unchanged with the further increase of Laplace pressure within a certain degree.

The dependence of the structural parameters on the Cassie state stability

Figure S3 shows the LSCM (laser scanning confocal microscope) images of the microcone arrays with different heights of the MNGF surfaces. The microcone heights were 4µm, 25µm, 50µm and 60µm, respectively. The morphology of MNGF surfaces with different microcone heights after chemical oxidation are shown in Figure S4. All the surfaces are composed of the microcone arrays covered with dense nanograsses and dispersively distributed microflowers. These are typical morphologies of the triplescale micro/nanostructures considered in the present study. As shown in Fig. S3, with the increase of the microcone height, the size of nanograsses stay almost the same. However, the diameters of the microflowers increase from 1µm to 5µm when the microcone height increases from 4µm to 50µm, respectively. Further increase of the microcone height up to 60µm shows no significant impact on the diameters of microflowers. This is because, with the increase of the microcone height (ablated by the ultrafast laser), the reaction area and the population of the active sites increases, which in turn promotes the reaction between the textured surface and the reaction solution. However, when the microcone height reaches to a certain magnitude, further increase of the reaction area and active sites on the surface show no significant effect on the chemical reaction.

The dependences of CA and the TPCL diameter on the Laplace pressure during droplet evaporation processes on MNGF superhydrophobic surfaces are presented in Fig. S5. The critical Laplace pressures of the tested MNGF superhydrophobic surfaces with microcone heights of 4 μ m, 25 μ m, 50 μ m and 60 μ m are 593 Pa, 788 Pa, 1446 Pa and 1431 Pa, respectively. This indicates that an increase of the microcone height can efficiently enhance the Cassie state stability of MNGF superhydrophobic surfaces to a certain degree. This is because the increased microcone height increases the surface roughness and correspondingly the volume of the air pockets, which is beneficial for enhancing the superhydrophobicity of a surface based on the Cassie and Wenzel equations. However, after reaching a certain height value, a further increase of the microcone height may not impact significantly the Cassie state stability of MNGF superhydrophobic surfaces.

Similarly, Fig. S6 shows the morphology of MNGF superhydrophobic surfaces with different microcone periodicities, which are 60µm, 80µm and 100µm, respectively. The

effect of microcone periodicity on Cassie state stability of the MNGF superhydrophobic surfaces is shown in Fig. S7. It can be seen that the critical Laplace pressure of the tested MNGF with the superhydrophobic surfaces with microcone periodicity of 40 μ m, 60 μ m, 80 μ m and 100 μ m are 1446 Pa, 927 Pa, 761 Pa and 450 Pa, respectively, indicating that the increase of microcone periodicity decreases the Cassie state stability of the MNGF superhydrophobic surfaces. The reason for that outcome may be attributed to the reduced surface roughness and the increased solid-liquid contact area, which would have an adverse impact on enhancing the superhydrophobicity and Cassie state stability (based on the Cassie and Wenzel equations).

Thus, from the above discussion, one may conclude that the microcone structural parameters of the MNGF superhydrophobic surface are optimized to be with the parameter values of 50 µm height and 40 µm periodicity.



Figure S3 The LSCM (laser scanning confocal microscope) images of the microcone arrays with different heights of the MNGF surfaces: (a)-(a1) 4μm; (b)-(b1) 25μm; (c)-(c1) 50μm; (d)-(d1) 60μm. (a1)-(d1) are the corresponding side profiles of the microcone arrays in (a)-(d).



Figure S4 The morphology of MNGF surfaces with different microcone heights: (a)-(a2) 4μm; (b)-(b2) 25μm; (c)-(c2) 50μm; (d)-(d2) 60μm.



Figure S5 The dependence of CA (a) and TPCL diameter (b) on the Laplace pressure during the droplet evaporation processes on MNGF superhydrophobic surfaces with different microcone heights



Figure S6 The LSCM images and the morphology of MNGF surfaces with different microcone periodicity: (a)-(a3) 60µm; (b)-(b3) 80µm; (c)-(c3) 100µm



Figure S7 The dependence of CA (a) and TPCL diameter (b) on the Laplace pressure during the droplet evaporation processes on MNGF superhydrophobic surfaces with different microcone periodicity

The typical dropwise condensation on the MNGF surface

In Fig. S8(a), the marked small droplets (with visible diameters ranging from $5\mu m$ to $50\mu m$ in the zone A) merge into one bigger droplet with a diameter of around 60 μm in the next frame, as shown in zone A' of Fig. S8(b). Similar phenomenon can be seen in zones C and C' as marked in Fig. S8(d) and (e). In Fig. S8(b), the droplets with visible dimeters ranging from $10\mu m$ to $70\mu m$ in the marked domain B coalesce with each other and jump out of the surface due to the released surface energy, as marked in Fig. S8(c). Thus, refreshing the surface and restarting a new cycle of droplet condensation takes place (nucleation, growth, coalescence and coalescence-induced jumping). Similar phenomenon can be seen in Figs. S8(f)~(i) as well.



Figure S8. The dropwise condensation on the MNGF surface horizontally-placed on the cooling stage with temperature constant at 0°C and humidity at 50%. Scale bar 80 μ m. (a)~(b) and (d)~(e) the coalescence of the condensed droplets; (b)~(c), (f)~(g) (g)~(h) and (h)~(i) the self-propelled jumping of the droplets induced by the coalescence of the surrounding droplets.



Figure S9. Schematic diagram of hierarchical condensation cycles sustaining stable Cassie state of PCDs, rendering inspired by Refence 1. During condensation process, small SCDs near the hilltop domains nucleate and grow beneath PCDs in Cassie state (a); When the SCDs touch the liquid-air interface at the base of the PCDs or coalescence-induced jumping happens, they are absorbed into the PCDs (b); For the SCDs at the valley bottom, they become increasingly large by coalesensing the adjacent smaller SCDs and spontaneously move upwards due to the capillary forces and the released surface energy via coalescence ((c)~(e)), until they touch the above PCDs or coalescence-induced jumping happens and get absorbed into the PCDs. As a result, the microcone surface is refreshed and a new cycle of secondary condensation starts. The PCDs can either be removed by gravity or by the self-propelled jumping by coalescence with the adjacent PCDs. Then the cycle of hierarchical condensation initiates again. Thus, the sustainable hierarchical condensation keeps our designed triple-scale supehydrophobic surface exhibiting stable Cassie state.



Figure S10. The morphology of MNGF (a) and MNSF (b) surfaces after 10 icingdeicing cycles

Mechanical durability of the MNGF superhydrophobic surface

To further illustrate the mechanical robustness of the MNGF superhydrophobic surfaces, 20 more icing-deicing cycles and additional mechanical durability tests (including tape peeling test, linear abrasion test and water flow impact test, respectively) were performed for the MNGF superhydrophobic surfaces (with the optimized structural papramter for Cassie state stability). The detailed mechanical durability testing method and procedures can be found in refrences ²⁻³. The corresponding results are presented in below.

Figure S11 shows the effect of icing-deicing cycles on the ice adhesion strengths of a MNGF superhydrophobic surface. Overall, the ice adhesion strength of the MNGF surface increases gradually with the increase of the icing-deicing number of cycles. However, the ice adhesion strength remains below 10 kPa within 15 icing-deicing cycles. In addition, although the average value of the ice adhesion strength increases to 20 kPa when the icing-deicing cycle is in the domain from 15 to 30, the formed ice can still be readily removed, since the tested ice adhesion strength is around the defined "benchmark value" (e.g. 20 kPa) of the icephobic surface that allows the ice to be readily removed.



Figure S11 The effect of icing-deicing cycles on the ice adhesion strengths of the MNGF superhydrophobic surface

Figure S12 shows the effect of tape peeling cycles on the CAs and SAs of the MNGF surface. With the increase of the number of tape peeling cycles, the CA decreases and the SA increases gradually. But even the surface being exposed to 1000 tape peeling

cycles, the CA is still above 150°. The SA remains lower than 10° until after 800 tape peeling cycles, indicating that the MNGF superhydrophobic surface could withstand 800 tape peeling cycles before losing its superhydrophobicity. Figure S13 presents the effect of linear abrasion cycles on the CAs and SAs of the MNGF surface. Figure S13 shows that after being linear abraded for 4 cycles, the CA decreases to around 150° and SA increases up to 10°, indicating that the MNGF superhydrophobic surface could withstand 4 cycles of linear abrasion before losing its superhydrophobic surface could withstand 4 cycles of linear abrasion before losing its superhydrophobic surface from 4 to 10 leads to a fast increase of SA to 44°, while the CA still stays around 150°. This indicate that the droplet exhibits Wenzel state on the tested MNGF superhydrophobic surface. Figure 14 presents the effect of water impacting time on the CAs and the SAs of the MNGF superhydrophobic surface. After being water impacted for 60 min, the CA is still high, up to 153°, and the SA is still less than 5°. This indicates that the MNGF superhydrophobic surface possesses a good water impact durability.

Thus, it can be concluded that the prepared MNGF superhydrophobic surface could withstand up to 800 tape peeling cycles, 4 linear abrasion cycles and at least 60 min of water impact exposure before losing its superhydrophobicity, indicating a relatively good mechanical durability.



Figure S12 The effect of tape peeling cycles on contact angles and sliding angles of the MNGF surface



Figure S13 The effect of linear abrasion cycles on contact angles and sliding angles



of the MNGF surface

Figure S14 The effect of water impacting time on contact angles and sliding angles of the MNGF surface



Figure S15. The fixture of ice adhesion strength tests for superhydrophobic surfaces

References

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