Supplementary Information

Ultrasonic Plasticizing and Pressing of High-Aspect Ratio Micropillar Arrays with Superhydrophobic and Superoleophilic Properties

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Supplementary Figure S1 and Notes

In order to compare the deformation and damage of the template microstructures after being used in UPP and the ultrasonic molding method of placing polymer pellets directly on the template microstructures and applying ultrasonic vibration to make the polymer pellets plasticize and fill the template micropores, two templates with different microstructures were designed and processed. One is a rectangular bar array with a height of 500 μm, a width of 150 μm and a pitch of 650 μm (see Supplementary Figure S1a), which was obtained by low speed wire electrical discharge machining (WEDM-LS) 304 stainless steel plate. The other is a micropore (through-hole) array with a pore margin of about 30 μm on the upper surface and a pore margin of about 75 μm on the lower surface (see Supplementary Figure S1d), which was obtained by laser cutting 304 stainless steel sheet with a thickness of 0.5 mm. Supplementary Figures 1b and 1e show the above two templates after being used a certain number of times in the method of placing polymer pellets directly on the template microstructures, respectively, while Supplementary Figures 1c and 1f show the above two templates after being used the same number of times in UPP with the same process parameters, respectively. Obviously, deformation is present at multiple positions of the microstructure in Supplementary Figure S1b, while only individual positions in Supplementary Figure S1c. The microstructure in Supplementary Figure S1e is fractured, while in Supplementary Figure S1f is intact.

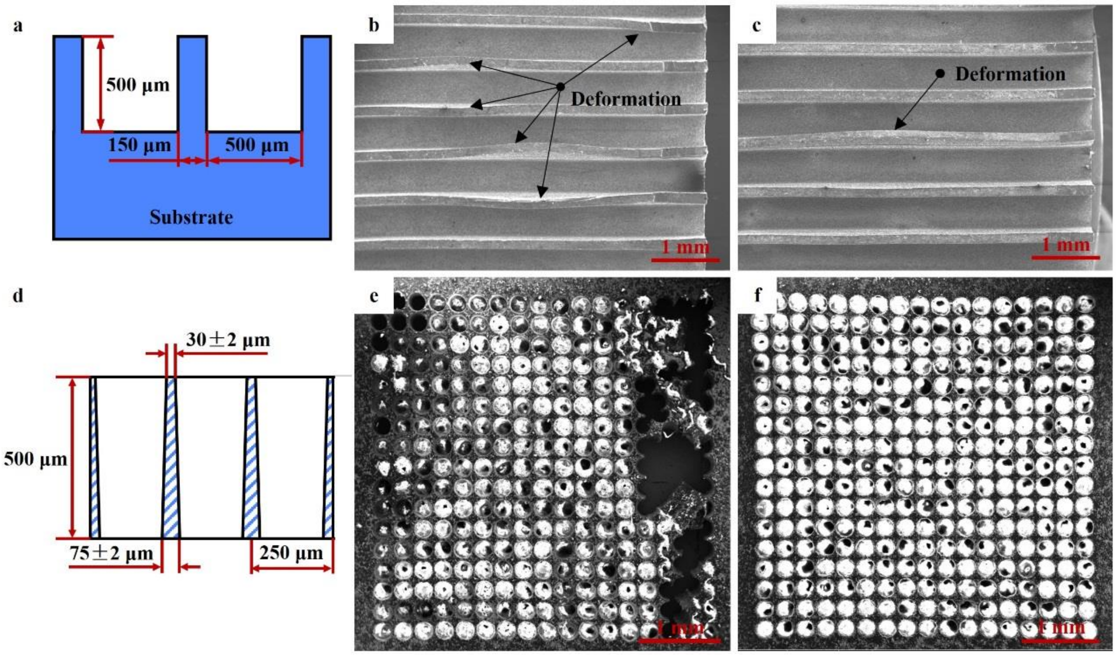

Figure S1. The deformation and damage of the template microstructures after being used. (**a**) The schematic dimensions of the rectangular bar array. (**b**) and (**c**) Rectangular bar array templates after being used in the method of placing polymer pellets directly on the template microstructures and UPP, respectively. (**d**) The schematic dimensions of the micropore (through-hole) array. (**e**) and (**f**) Micropore array templates after being used in the method of placing polymer pellets directly on the template microstructures and UPP, respectively.

Table S1. The previous work in preparing hierarchical micropillar arrays through the template methods and its results.

NO. and mark	Technique	Primary structure		Secondary structure		Steps and time	Forming time	Preheating temperature of raw material (℃)	Forming environmental requirement	Raw material	Template material/type	Reference
		Width/ Diameter (μm)	Aspect ratio	Width/ Diameter (nm)	Aspect ratio							
1. ●	Nanoimprint lithography	3	4	200	10	Step1, Imprinting of microplilar array /Not reported. Step2, Spin-coating of poly(sodium 4 styrenesulfonate (PSS) /30 s. Step3, Second imprinting of fibrillar array /1000 s. Step4, Dissolution of PSS / (Not calculated).	＞1030 s	170	Not reported	Polycarbonate (PC)	(i) Silicon (Primary structure). (ii) Porous anode aluminum oxide (AAO) with a self-assembled monolayer of 1H,1H,2H,2H perfluorodecyltrichlorosilane (Secondary structure).	[45]
2. ●	Photo-nanoimprinting	2.6	6.2	360	1.2	Step1, Photo-nanoimprinting / Not reported Step2, Removal of the template / (Not calculated).	Not reported	Not reported	Not reported	Polyethylene terephthalate (PET)	AAO with a release layer	[46]
3. ●	Nanoimprinting and Modified laser swelling	58	0.5	500	2.4	Step1, Preparing the swelling polymer film /18.75 h (Not calculated). Step2, Nanoimprinting of the secondary pillar structure /1.5 min. Step3, Irradiating by the laser beam to form the primary structure /50 ms per structure.	＞1.5 min	Not reported	Not reported	UV-curable polymer and methyl red (MR)-doped PMMA	(i) No template (Primary structure). (ii) Polydimethylsiloxane (PDMS) (Secondary structure).	[47]
4. ●	Soft imprint technique and Directional photofluidization imprint lithography (DPIL)	40	1	500	1.2	Step1, Soft-imprinting of prestructured single-scale PDO 3 film /12 h. Step2, DPIL of the secondary structure /0.33 h.	12.33 h	Not reported	In vacuum	Polydisperse orange 3 (PDO 3)	PDMS	[48]

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		Width/ Diameter (μm)	Aspect ratio	Width/ Diameter (nm)	Aspect ratio							
5. ●	Nanoimprinting	0.28	21.4	110	5.5	Step1, Nanoimprinting of the hierarchical microstructures /20 min. Step2, Cooling to room temperature and demolding of the polymeric structures / (Not calculated).	20 min	175	In vacuum	Polycarbonate (PC)	Multi-tiered branched porous anodic alumina (PAA)	[49]
6. ■	Light-assisted soft molding	50	4	5	2	Poured the polymer mixture into the template and exposed in light to cure /14 h.	14 h	65	Vacuum of ~ 6×10^4 Pa	A mixture with 10:1 ratio of Sylgard 184 prepolymer and crosslinker	SU-8 photoresists	[50]
7. ■	Capillary force lithography (CFL) and Photo-patterning	50	0.2	1000	7.7	Step1, CFL of pillar array (Secondary structure) /8 min. Step2, Photo-patterning of micropillar array (Primary structure) /20 s. Step3, Solvent developing / (Not calculated).	8.3 min	95	Not reported	SU-8 photoresists	(i) PDMS (Secondary structure). (ii) Photomask (Primary structure).	[13]
8. ■	UV-assisted micromolding and Capillary molding	50	2	3000	6.7	Step1, UV-assisted micromolding of the primary structure / Not reported. Step2, Capillary molding of the secondary structure /24 h.	＞24 h	Room temperature	In vacuum	Polyurethanes (PU)	(i) Liquid silicone rubber (HS-II, Dow Corning) (Primary structure). (ii) Silicon (Secondary structure).	[51–53]
9. ■	UV-assisted capillary molding	30	0.7	300	16.7	Step1, UV-assisted capillary molding of micro-nano hierarchical pillar arrays /15 min. Step2, Baking on a hotplate at 100 ◦C for at least 15 min to dehydrate / (Not calculated).	15 min	Not reported	A hotplate with 100 ◦C	UV-curable prepolymer	PDMS	[54]

NO. and mark	Technique	Primary structure Width/ Diameter (μm)	Primary structure Aspect ratio	Secondary structure Width/ Diameter (nm)	Secondary structure Aspect ratio	Steps and time	Forming time	Preheating temperature of raw material (℃)	Forming environmental requirement	Raw material	Template material/type	Reference
10. ■	2-step UV assisted capillary molding	5	5	475	5.9	Step1, Spin-coating of PUA on the flexible PET substrate (Not calculated). Step2, UV-assisted capillary molding of the microhairs /50 s. Step3, Second UV-assisted capillary molding of nanohairs on the preformed microhairs /10 s.	60 s	Not reported	Not reported	Soft UV-curable polyurethane acrylate (PUA)	(i) PDMS (Primary structure). (ii) PUA (Secondary structure).	[55]
11. ⬢	Thermal-assisted capillary molding	0.38	2.4	100	1.8	Step1, Capillary molding of hierarchical PS nanohairs /3 h. Step2, Removal of the AAO template / (Not calculated).	3 h	185	In vacuum	Polystyrene (PS)	AAO	[56]
12. ⬢	Infiltration forming (Capillary molding)	10	7	60	8.3	Step1, Spin-coating of low-viscosity polymeric solutions (10 wt% PMMA) on the top surface of the template /30 s. Step2, Capillary molding of the hierarchical microstructures /1 h. Step3, Dissolving the template / (Not calculated).	60.5 min	120	Not reported	Low-viscosity polymeric solutions (10 wt% PMMA)	A bonded template of the micro- and nanoporous alumina membranes	[57]
13. ⬢	Soft molding	20	3	7000	2.9	Poured PU onto the PDMS mold and allowed it to cure at 75 °C for 72 h /72 h.	72 h	75	Not reported	Polyurethane (PU)	PDMS	[58]
14. ⬢	Nanomolding	5	2	600	9	Step1, Molding the liquid polymer under vacuum /4 h. Step2, Etching away the both membrane / (Not calculated).	4 h	65	In vacuum	PDMS	A membrane with self-organized high aspect ratio polycarbonate micro/nano-pores	[59]
15. ▲	Hot-embossing	15	2	100	11	Hot-embossing and demolding /10 min.	10 min	150	Vacuum of 1 Pa	Thermoplastic COP polymer (Zeonor 1060R)	Solvay MD700 UV cured polymer	[60]

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		Width/ Diameter (μm)	Aspect ratio	Width/ Diameter (nm)	Aspect ratio							
16. ▲	Hot-pressing	5	3.5	600	8	Step1, Melting the PP and filling the pores in the membrane /40-50 min. Step2, Etching the membranes to realize the polypropylene fibers /1 h (Not calculated).	40-50 min	200	Not reported	polypropylene (PP)	Polycarbonate (PC)	[61]
17. ⬟	Injection molding	18.5	1	50	10	Injection molding /Not reported.	Not reported	255	Mold temperature 50 °C	Polypropylene (PP)	(i) Aluminum (Primary structure). (ii) AAO (Secondary structure).	[62]
18. ⬟	Injection molding	40	2	50	2.5	Injection molding /Not reported.	Not reported	255	Mold temperature 70 °C	Polypropylene (PP)	(i) Aluminum (Primary structure). (ii) AAO (Secondary structure).	[63]
19. ⬟	Injection molding	200	1.5	400	12	Injection molding /95 s	95 s	Not reported	Mold temperature 178 °C	Polypropylene (PP)	(i) Etched plate (Primary structure). (ii) AAO with anti-sticking layer (Secondary structure).	[64]
20. ⬟	Ultrasonic forming	20.9	1.2	600	1.3	Ultrasonic forming of the hierarchical microstructures /9 s.	9 s	Room temperature	None	Polyethylene (PE)	Nickel nano-micro mold	[23]
21. ★	Ultrasonic plasticizing and pressing	48.5	4.1	250	20	Step1, Ultrasonic plasticizing and pressing /5.3 s. Step2, Removal of residual template /10 min (Not calculated).	5.3 s	Room temperature	None	Polypropylene (PP)	(i) 304 stainless steel (Primary structure). (ii) AAO (Secondary structure).	This work

Note: The forming time in the table does not include the heating and cooling time of the mold, the heating time of raw materials, and the process time that can process a large number of workpieces simultaneously.