Electronic Supplementary Information

Printable Ultrathin Substrates Formed on a Concave/Convex Underlayer for Highly Flexible Membrane-Type Electrode Stickers

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<u>Fabrication of printable ultrathin polymer platelets on a handling substrate with</u> <u>circular concavities</u>

Depth: 2.8 µm Pitch: 20 µm



Figure S1: a, b) SEM images of circular concavities on silicon wafer chips after RIE etching (these images correspond to the step in Fig. 1b) and deposition of a 200-nm-thick SiO_2 layer (step Fig. 1c). c, d) SEM images of ultrathin square polymer islands (SU-8 in this image) after coating and patterning a polymer layer (step Fig. 1d) and etching the SiO_2 layer in HF solution (step Fig. 1e). The insets in (a-d) show the corresponding cross-sectional views.

Fabrication of printable ultrathin polymer platelets on a handling substrate with crossshaped concavities

Depth: 2.9 μm Pitch: 30 μm



Figure S2: a, b) SEM images of cross-shaped concavities on silicon wafer chips after RIE etching (these images correspond to the step in Fig. 1b) and deposition of a 200-nm-thick SiO_2 layer (step Fig. 1c). c, d) SEM images of ultrathin square polymer islands (SU-8 in this image) after coating and patterning a polymer layer (step Fig. 1d) and etching the SiO_2 layer in HF solution (step Fig. 1e). The insets in (a-d) show the corresponding cross-sectional views.

Alignment degree and transfer yield vs. concavity depth

Pitch: 20 µm



Figure S3: (Left column) Cross-sectional SEM images of silicon substrates with concavities of various sizes. (Middle column) Photographs of printable polymer island patterns after etching the sacrificial SiO_2 layer in HF solution. (Right column) Photographs of polymer island patterns on PDMS slabs after transfer printing.

Alignment degree and transfer yield vs. the pitch of circular concavities

Depth: 2.8 µm



Figure S4: (Left column) Photographs of printable polymer island patterns after etching the sacrificial SiO_2 layer in HF solution. (Right column) Photographs of polymer island patterns on PDMS slabs after transfer printing.

Alignment degree and transfer yield vs. the pitch of cross-shaped concavities

Depth: 2.9 µm



Figure S5: (Left column) Photographs of printable polymer island patterns after etching the sacrificial SiO_2 layer in HF solution. (Right column) Photographs of polymer island patterns on PDMS slabs after transfer printing.

Alignment degree and transfer yield vs. the pitch of circular concavities





Figure S6: (Top) Cross-sectional SEM images of silicon substrates with circular concavities. (Bottom left) Photographs of printable polymer island patterns after etching the sacrificial SiO₂ layer in HF solution. (Bottom right) Photographs of polymer island patterns on PDMS slabs after transfer printing.

Alignment degree and transfer yield vs. the pitch of circular concavities



Figure S7: (Top) Cross-sectional SEM images of silicon substrates with circular concavities. (Bottom left) Photographs of printable polymer island patterns after etching the sacrificial SiO_2 layer in HF solution. (Bottom right) Photographs of polymer island patterns on PDMS slabs after transfer printing.

Alignment degree and transfer yield vs. the pitch of cross-shaped concavities



Figure S8: (Top) Cross-sectional SEM images of silicon substrates with cross-shaped concavities. (Bottom left) Photographs of the printable polymer island patterns after etching the sacrificial SiO_2 layer in HF solution. (Bottom right) Photographs of polymer island patterns on PDMS slabs after transfer printing.

<u>Alignment degree and transfer yield vs. the thickness of the printable ultrathin polymer</u> <u>substrate</u>

Depth: 2.8 µm Pitch: 28 µm



Figure S9: (Left column) Photographs of printable polymer island patterns after etching the sacrificial SiO_2 layer in HF solution. (Middle column) Photographs of polymer island patterns on PDMS slabs after transfer printing. (Right column) Cross-sectional SEM images of polymer islands with cross-shaped convexities. (Bottom) Table showing the alignment degree and transfer yield vs. the thickness of the ultrathin polymer substrate.

Recycling test using a single silicon wafer for transfer printing

Depth: 4.0 µm Pitch: 30 µm



Figure S10: Top view and cross-sectional SEM images and photographs of the printable island patterns transferred onto PDMS.

Alignment of polymer island patterns before and after transfer printing using rollerand slab-type stamps

b)

Using flat-type stamp

Using roller-type stamp

- after HF & Cr deposition

2718236 um

a)

- after double transfer on an adhesive PET film									
PDMS type	Direction	After HF & Cr deposition	After double transfer on an adhesive PET film	Deformation Rate (%)					
Roller	X	2714.6 μm	2773.5 μm	2.20					
	у	2707.2 μm	2718.3 μm	0.41					
Flat	X	2714.5 μm	2714.6 μm	0.00					
	v	2723.9 µm	2723 7 um	-0.01					

Figure S11: (a, b) OM images of printable polymer island arrays on silicon handling substrates before transfer printing. (c, d) OM images of printed polymer island arrays on adhesive PET films using (c) roller-type and (d) slab-type PDMS stamps. For better contrast in the OM images, 100 nm Cr was deposited on the polymer islands.

solution	before			After 1 hour		
	control	circle	cross	control	circle	cross
Au etchant						
Cr etchant						
IPA						
MIF 500						
SU-8						
PR solution					ġ.	
						1 cm

Durability test with various chemicals

Figure S12: Durability test with various chemicals using square (500 μ m × 500 μ m) island arrays of polymer substrate. The depth and pitch are 3 μ m and 28 μ m, respectively.



Printable ultrathin polymer platelets on a handling substrate with circular concavities

Figure S13: a-d) SEM images of ultrathin square islands of (a) polyimide and (b-d) SU-8 after coating and patterning a polymer layer (step Fig. 1d). The insets in (a, b) show tilted and cross-sectional views, respectively, at high magnification. The thicknesses of the ultrathin polymer substrates in a, b, c, and d are $1.2 \mu m$, $1.4 \mu m$, $10.4 \mu m$, and $72.4 \mu m$, respectively.

<u>Process used for transfer printing of ultrathin polymer islands onto a curvilinear</u> <u>surface</u>



Figure S14: OM images of the transfer printing process using printable ultrathin polymer arrays with a square pattern onto a hemispherical convex PDMS membrane. a) Polymer island pattern transferred onto a flat PDMS membrane using a tensional stage. b, c) Both sides of the PDMS membrane after relaxing the tension. d) PDMS membrane with polymer pattern after removing the stage.

Lateral geometry of a highly flexible ultrathin serpentine metal electrode



Figure S15: Tiled OM images of an ultrathin metal electrode between two contact pads on a silicon handling wafer before removing the sacrificial layer.

Durability test of a highly flexible ultrathin serpentine metal electrode under 3dimensional tensile strain





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strain level was measured from the contour distance between two contact pads of an ultrathin electrode. (Left column) Photographs of a test kit for electrical connection under 3-dimensional tensile strain. (Middle and right columns) Cross-sectional photographs and top-view OM images of a stretchable metal electrode under various strain levels from 0 to 162%.

<u>A highly flexible ultrathin serpentine metal electrode after one cycle of 3-dimensional</u> <u>stretching</u>



Figure S17: a) Tiled OM images of an ultrathin metal electrode on stretchable adhesive tape before stretching. b) Tiled OM images of the same sample after one stretching test between 51% and 0 strain.

<u>A highly flexible ultrathin serpentine metal electrode after 20 cycles of 3-dimensional stretching</u>



Figure S18: OM images of an ultrathin metal electrode a) with no strain before stretching, b) under 51% strain and c) under no strain after 20 cycles of stretching. d) Resistance ratio of the ultrathin metal electrode as a function of various strain levels. The test was conducted for 20 cycles.