

Supplementary Information

Shrinkage-transfer-assisted printing of microcircuits on fibers

Jiongke Jin¹, Mei Zou¹, Deyu Liu¹, Haoxuan Ma¹, Yingying Zhang^{1*}

¹Key Laboratory of Organic Optoelectronics and Molecular Engineering of the Ministry of Education, Department of Chemistry, Tsinghua University, Beijing 100084, PR China

Corresponding author: Yingying Zhang Email: yingyingzhang@tsinghua.edu.cn

Table of Content

Supplementary Fig. 1 | Ecoflex film platform used for shrinking EGaIn circuits

Supplementary Fig. 2 | Images of EGaIn microcircuits on Ecoflex films with different shrinkage ratios

Supplementary Fig. 3 | Transferred EGaIn microcircuits on PVA

Supplementary Fig. 4 | XPS of N 1s confirming nitrogen states in sericin and EGaIn/sericin

Supplementary Fig. 5 | SEM images comparing morphologies of EMPs in inks with and without sericin

Supplementary Fig. 6 | Effect of different shear times of blender on the size of EMPs

Supplementary Fig. 7 | Illustration of functional group changes on the substrate surface during the STAP process

Supplementary Fig. 8 | Photograph showing the mask placed on the pre-stretched Ecoflex substrate for plasma treatment and the screen printing

Supplementary Fig. 9 | Attachment of EMPs on the Ecoflex surface as a function of plasma treatment time

Supplementary Fig. 10 | Stress-strain curve of Ecoflex film

Supplementary Fig. 11 | Illustration of 4-stage of transfer-sintering.

Supplementary Fig. 12 | Force measurements between the EMP and different substrates

Supplementary Fig. 13 | Surface morphology changes during shrinkage-sintering and transfer-sintering of EGaIn microcircuits.

Supplementary Fig. 14 | Schematic illustration showing the shrinkage-induced resolution limit in the shrinkage process

Supplementary Fig. 15 | Side-view SEM images of microcircuits shrunk by 94% in area.

Supplementary Fig. 16 | Microscopic images of screens with the same pattern but different mesh sizes

Supplementary Fig. 17 | Differences in PVA-transferred EGaIn microcircuit caused by variations in screen mesh size

Supplementary Fig. 18 | EGaIn microcircuits with different widths

Supplementary Fig. 19 | Optical images of EGaIn microcircuits on fibers with different materials and diameters.

Supplementary Fig. 20 | Optical image of a fiber (diameter, 1.5 mm) with two layers of EGaIn microcircuits

Supplementary Fig. 21 | SEM images of the circuit-fiber interface at different temperature and humidity

Supplementary Fig. 22 | Influence of humidity and temperature on the mechanical stability of EGaIn microcircuits on fibers

Supplementary Fig. 23 | Evolution of brightness of luminance pixel with decrease of bending radius

Supplementary Fig. 24 | Applying STAP process using three typical commercial electronic inks.

Table S1 | Parameters of screens with different mesh counts, the resolution of as-printed EGaIn/sericin ink, and the resolution after shrinkage

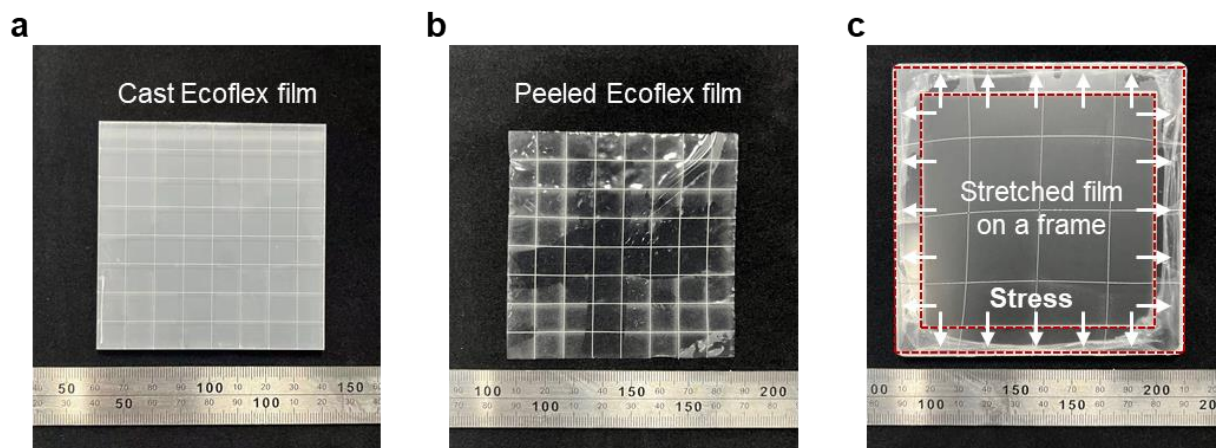
Table S2 | Conductivity of EGaIn microcircuits prepared with different pre-stretch ratio and EMPs coverage ratio

Supplementary Video 1. (Separate file) | Fabrication of stress-engineered 2D silicone platform

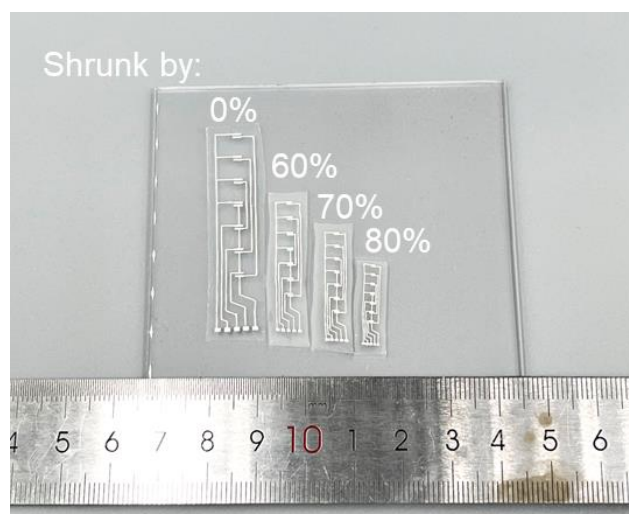
Supplementary Video 2. (Separate file) | Shrinkage-transfer-assisted printing (STAP)

Supplementary Video 3. (Separate file) | Differentiated hydrophilicity on the surface of Ecoflex film after patterned plasma treatment

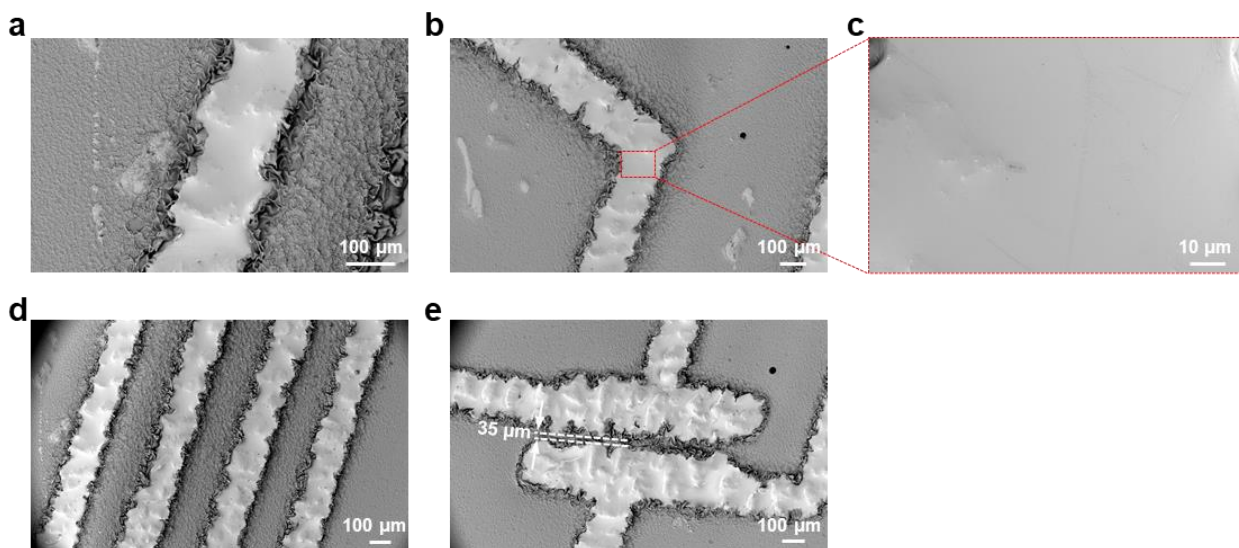
Supplementary Video 4. (Separate file) | Controllable electroluminescent pixel points on fiber



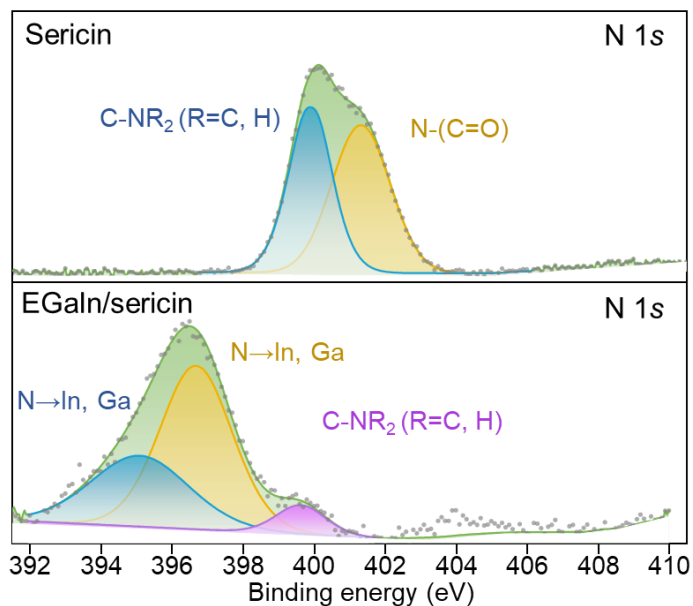
Supplementary Fig. 1. Ecoflex film platform used for shrinking EGaIn circuits. (a) Ecoflex was cast onto an acrylic plate to prepare a stretchable platform (the $1\text{ cm} \times 1\text{ cm}$ grid here is to help to illustrate the uniformity of the stretchable platform, which will not present during actual shrink process). (b) Ecoflex film peeled off from the acrylic plate. (c) Uniformly stretched Ecoflex film fixed onto an acrylic frame, ready for circuit construction on its surface.



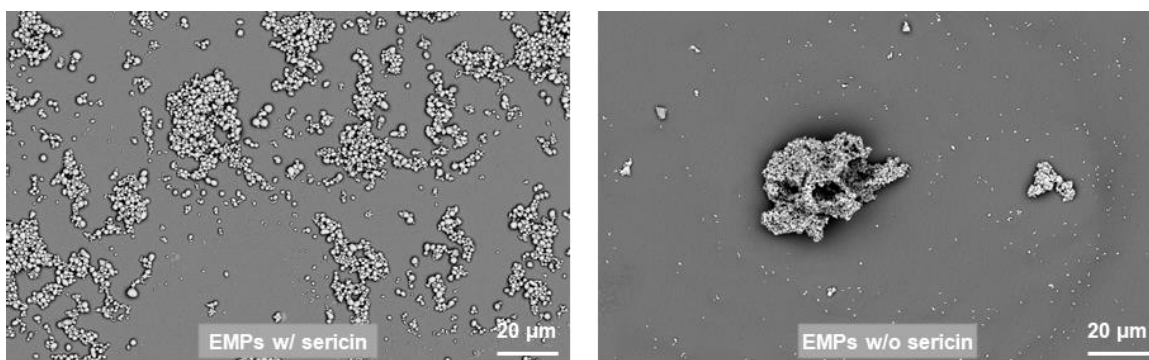
Supplementary Fig. 2. Images of EGaIn microcircuits on Ecoflex films with different shrinkage ratios. From left to right, shrunk by 0%, 60%, 70%, 80% in area.



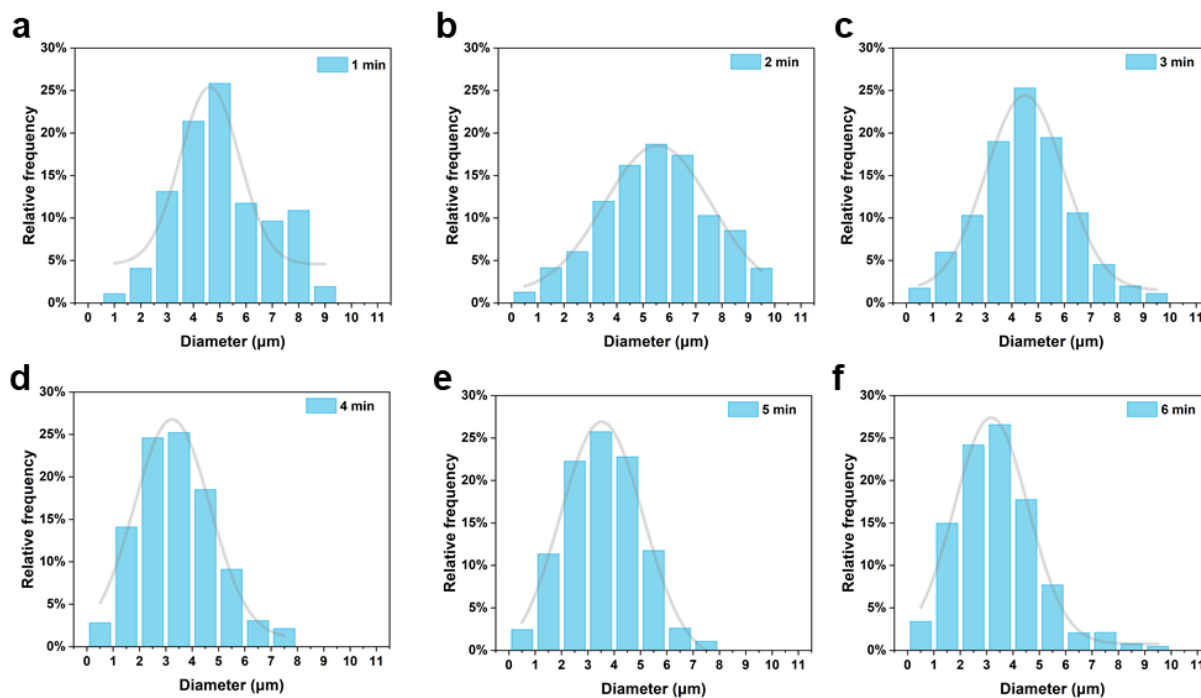
Supplementary Fig. 3. Transferred EGaIn microcircuits on PVA. (a) A single line. (b) Sharp corner of the circuit. (c) Enlarged view of the circuit surface. (d) Multiple lines. (e) Small gaps (35 μm).



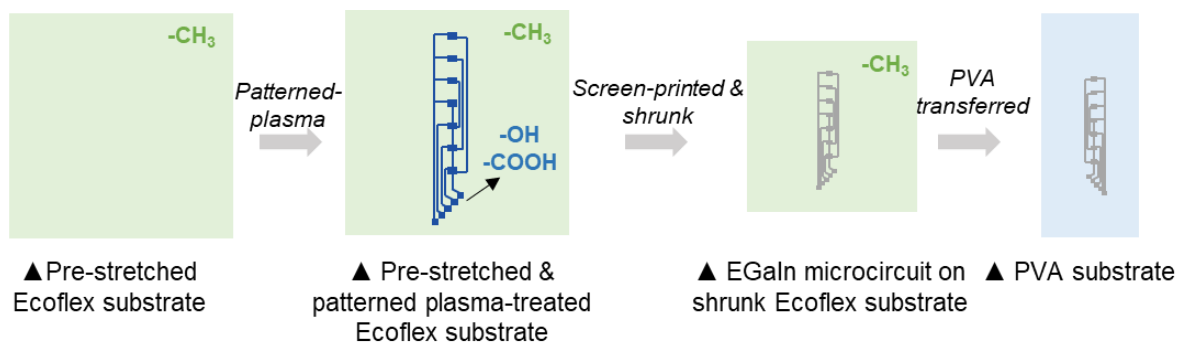
Supplementary Fig. 4. XPS of N 1s confirming nitrogen states in sericin and EGaIn/sericin.



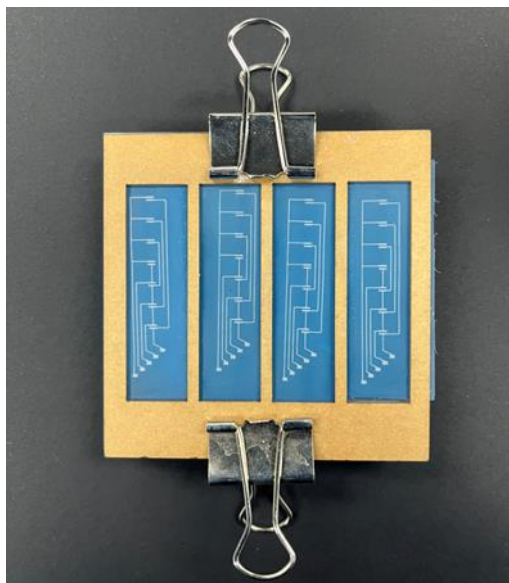
Supplementary Fig. 5. SEM images comparing morphologies of EMPs in inks with and without sericin, after being placed in an atmospheric environment at 60 °C for 30 min.



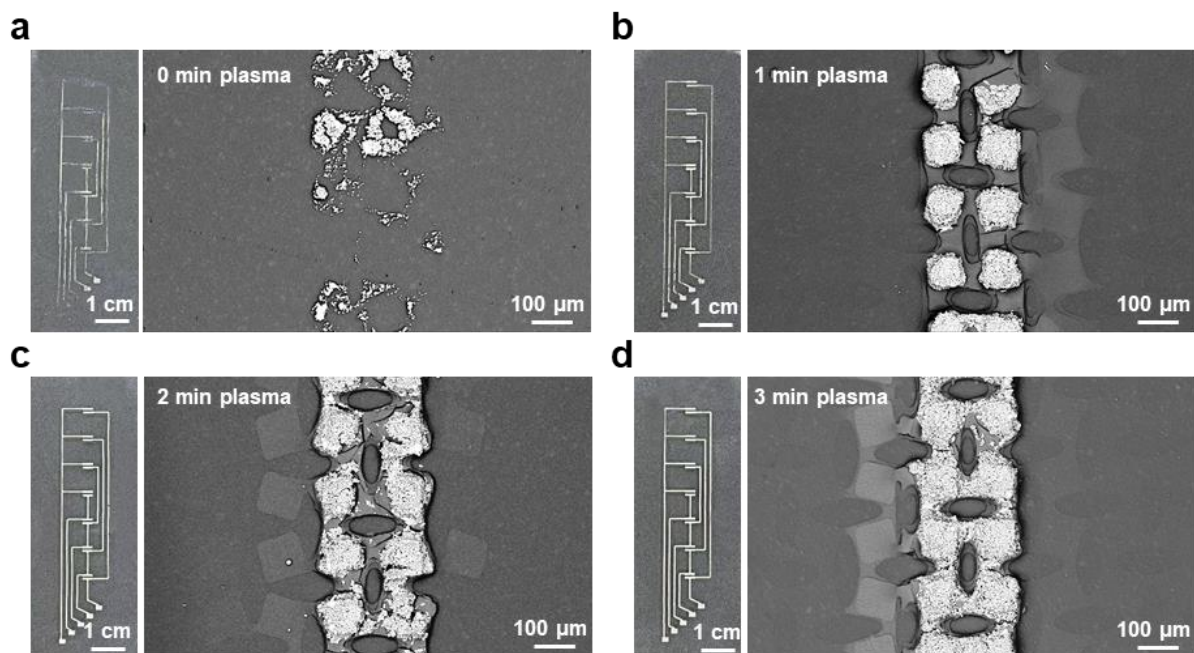
Supplementary Fig. 6. Effect of different shear times of blender on the size of EMPs. The n values (number of particles analyzed) are 962, 2536, 1852, 4342, 4760, and 5516 for 1 min to 6 min, respectively.



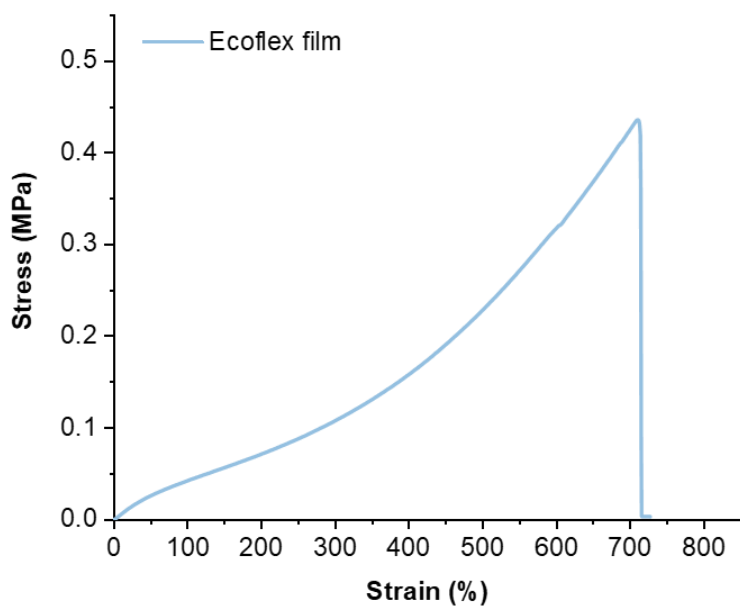
Supplementary Fig. 7. Illustration of functional group changes on the substrate surface during the STAP process. The patterned plasma-treated Ecoflex substrate was prepared by attaching a patterned screen on the substrate.



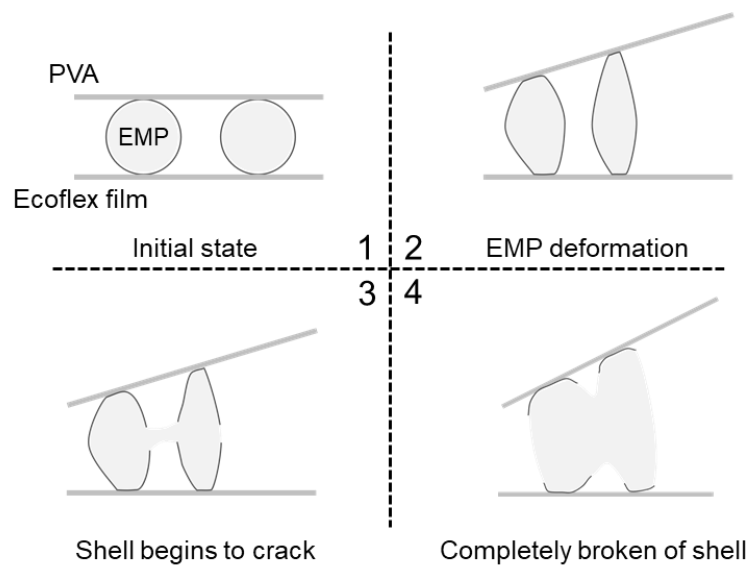
Supplementary Fig. 8. Photograph showing the mask placed on the pre-stretched Ecoflex substrate for plasma treatment and the screen printing.



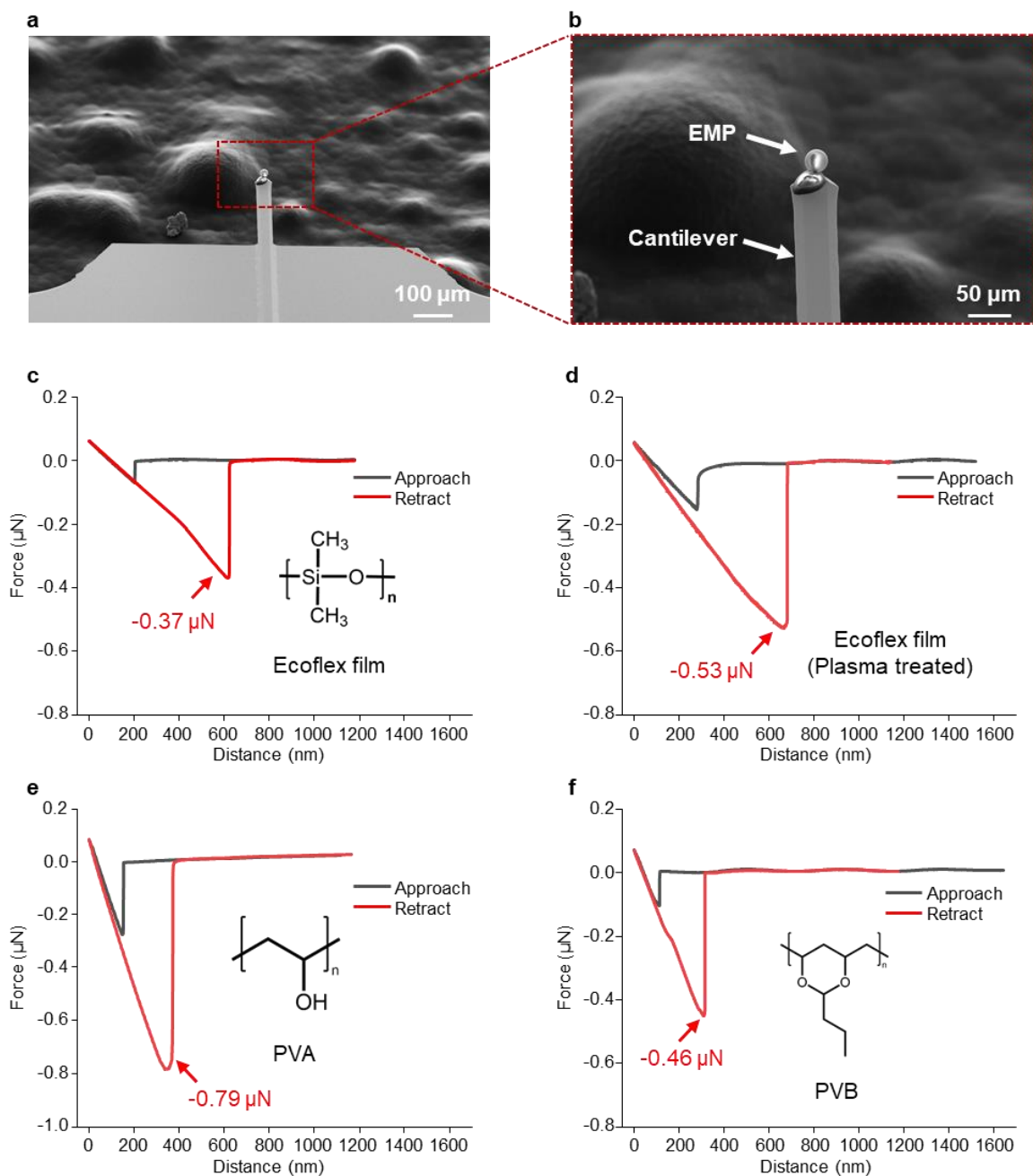
Supplementary Fig. 9. Attachment of EMPs on the Ecoflex surface as a function of plasma treatment time.



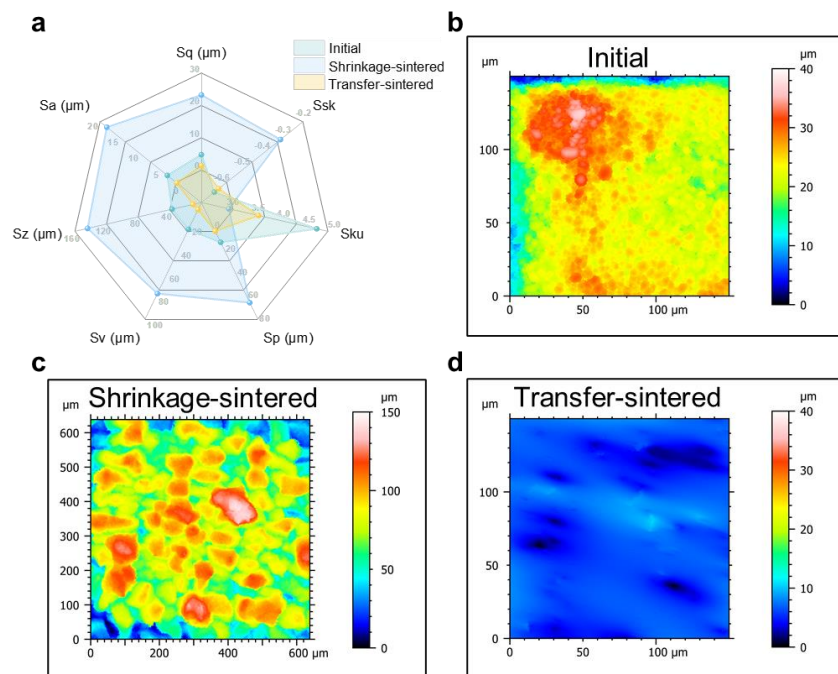
Supplementary Fig. 10. Stress-strain curve of Ecoflex film.



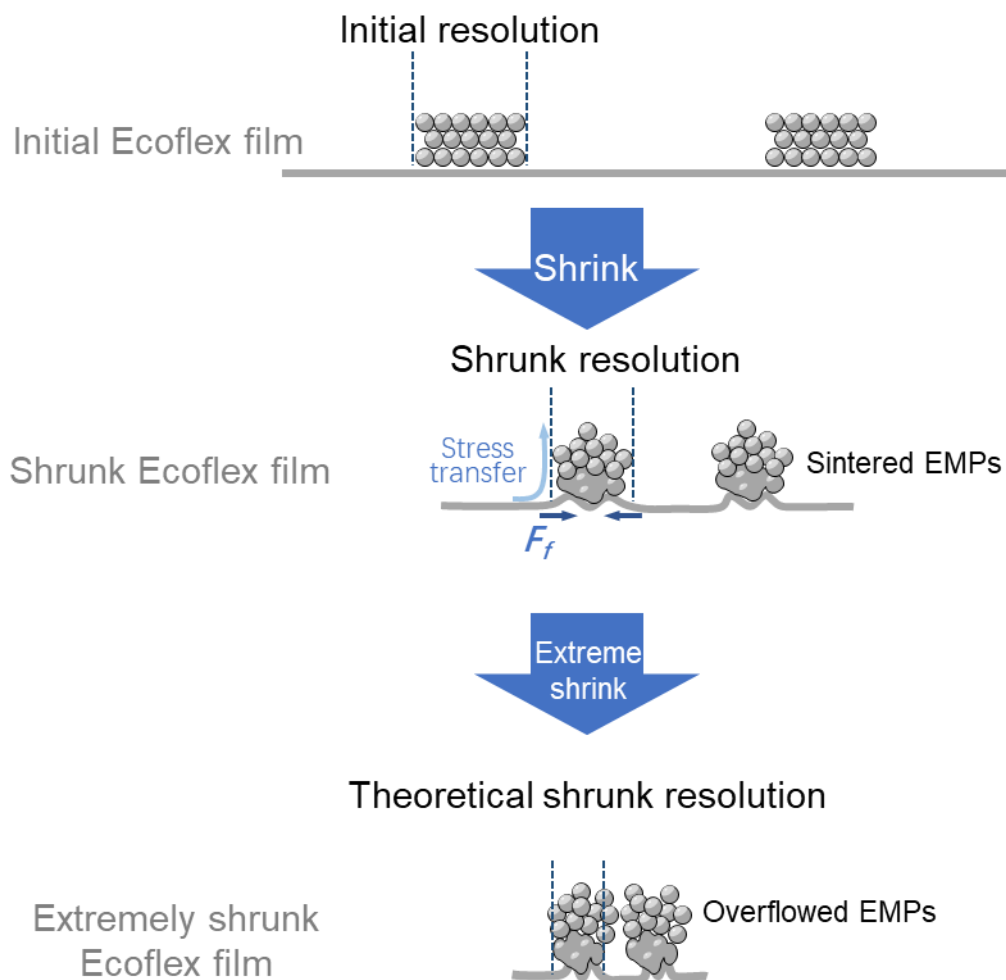
Supplementary Fig. 11. Illustration of 4-stage of transfer-sintering.



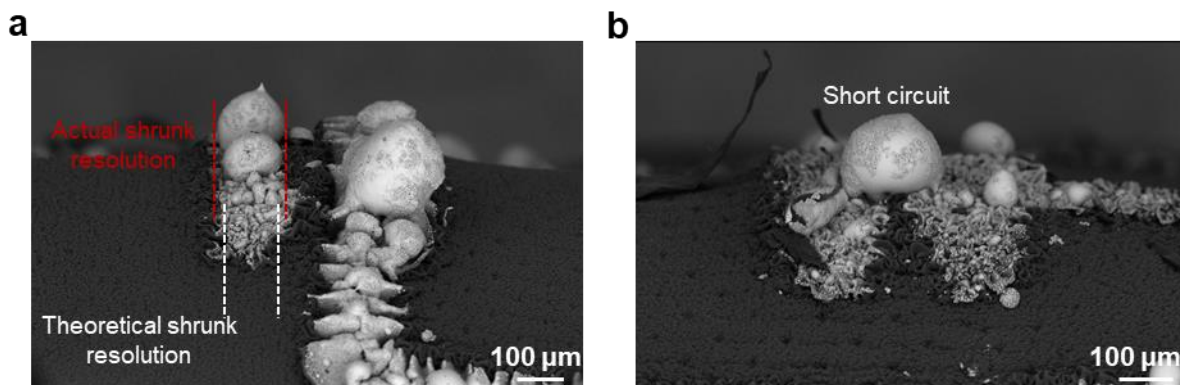
Supplementary Fig. 12. Force measurements between the EMP and different substrates. (a) SEM image of an EMP attached to an AFM probe. (b) Magnified view of the probe shown in (a). (c-f) Approach-retract force curves acquired between the EMP and a pristine Ecoflex film (c), an Ecoflex film treatment with 3 min plasma (d), a PVA film (e), and a polyvinyl butyral (PVB) film (f).



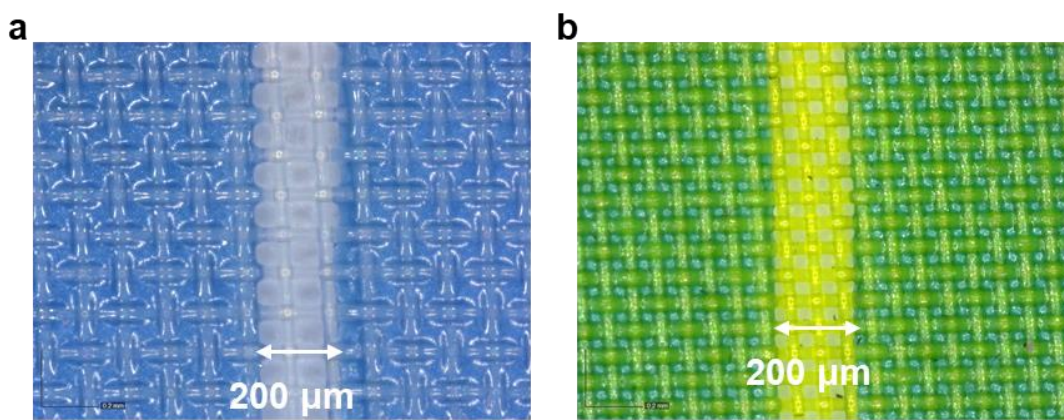
Supplementary Fig. 13. Surface morphology changes during shrinkage-sintering and transfer-sintering of EGaIn microcircuits. (a) Statistical analysis of surface morphology parameters, including S_q (root mean square height, representing the standard deviation of height values), S_{sk} (skewness, indicating the asymmetry of height distribution), S_{ku} (kurtosis, reflecting peak sharpness), S_p (maximum peak height), S_v (maximum pit depth), S_z (maximum height, the sum of S_p and S_v), and S_a (arithmetic mean height). (b) Initial EGaIn circuit on Ecoflex. (c) Shrinkage-sintered EGaIn microcircuit on Ecoflex. (d) Transfer-sintered EGaIn microcircuit on PVA.



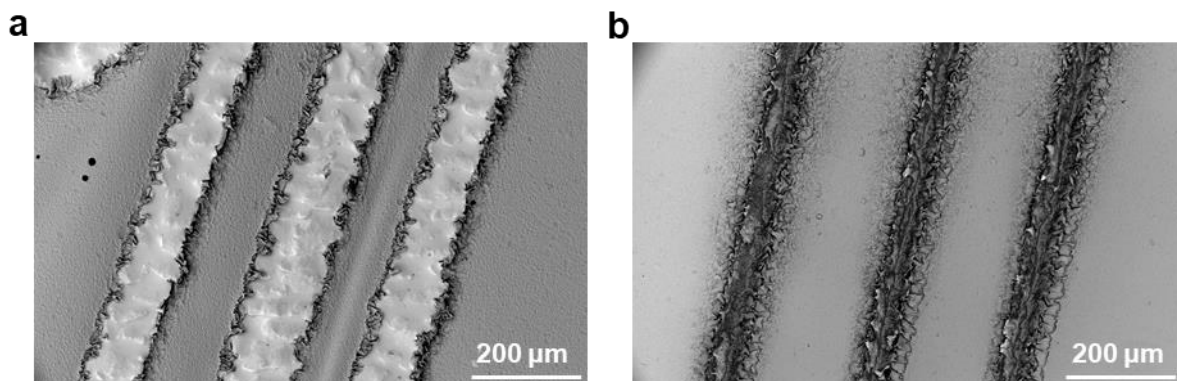
Supplementary Fig. 14. Schematic illustration showing the shrinkage-induced resolution limit in the shrinkage process. Excessive shrinkage causes overflow of EMP ink beyond the pattern boundaries, thereby limiting the minimum achievable resolution.



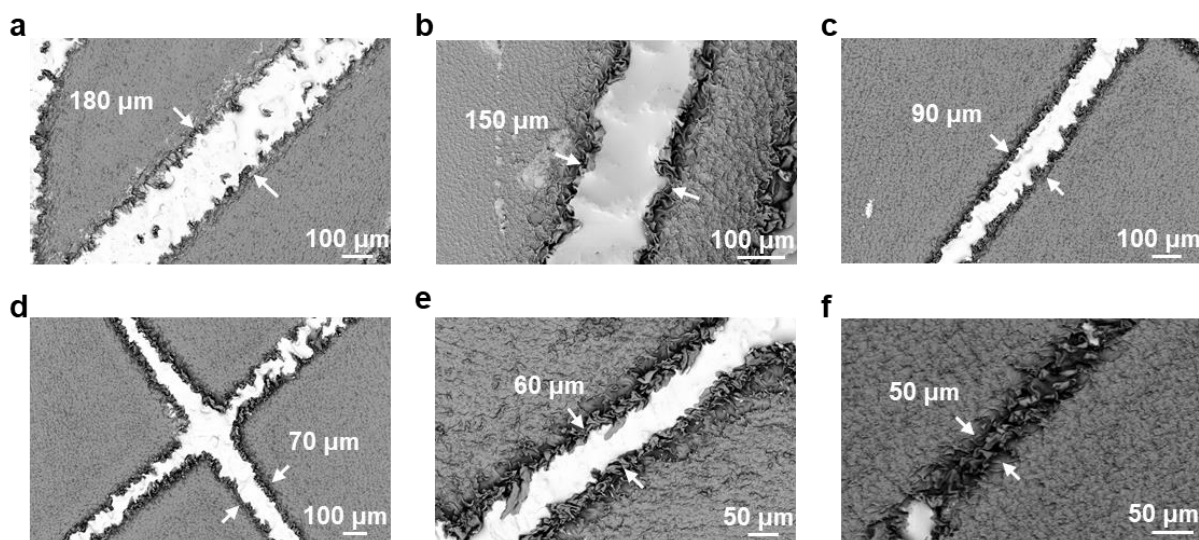
Supplementary Fig. 15. Side-view SEM images of microcircuits shrunk by 94% in area. (a) The actual line width becomes larger than the designed width due to EMP overflow. **(b)** Short-circuit behavior observed in the capacitor.



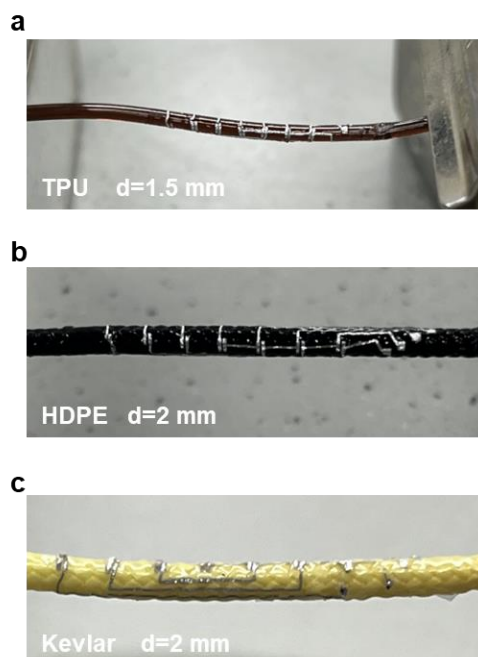
Supplementary Fig. 16. Microscopic images of screens with the same pattern but different mesh sizes. (a) 200 mesh, with an EMPs coverage ratio of 26%. **(b)** 300 mesh, with an EMPs coverage ratio of 16%.



Supplementary Fig. 17. Differences in PVA-transferred EGaIn microcircuit caused by variations in screen mesh size. (a) 200 mesh, with an EMP coverage ratio of 26%. (b) 300 mesh, with an EMP coverage ratio of 16%. (Pre-stretch ratio: 75% for both)



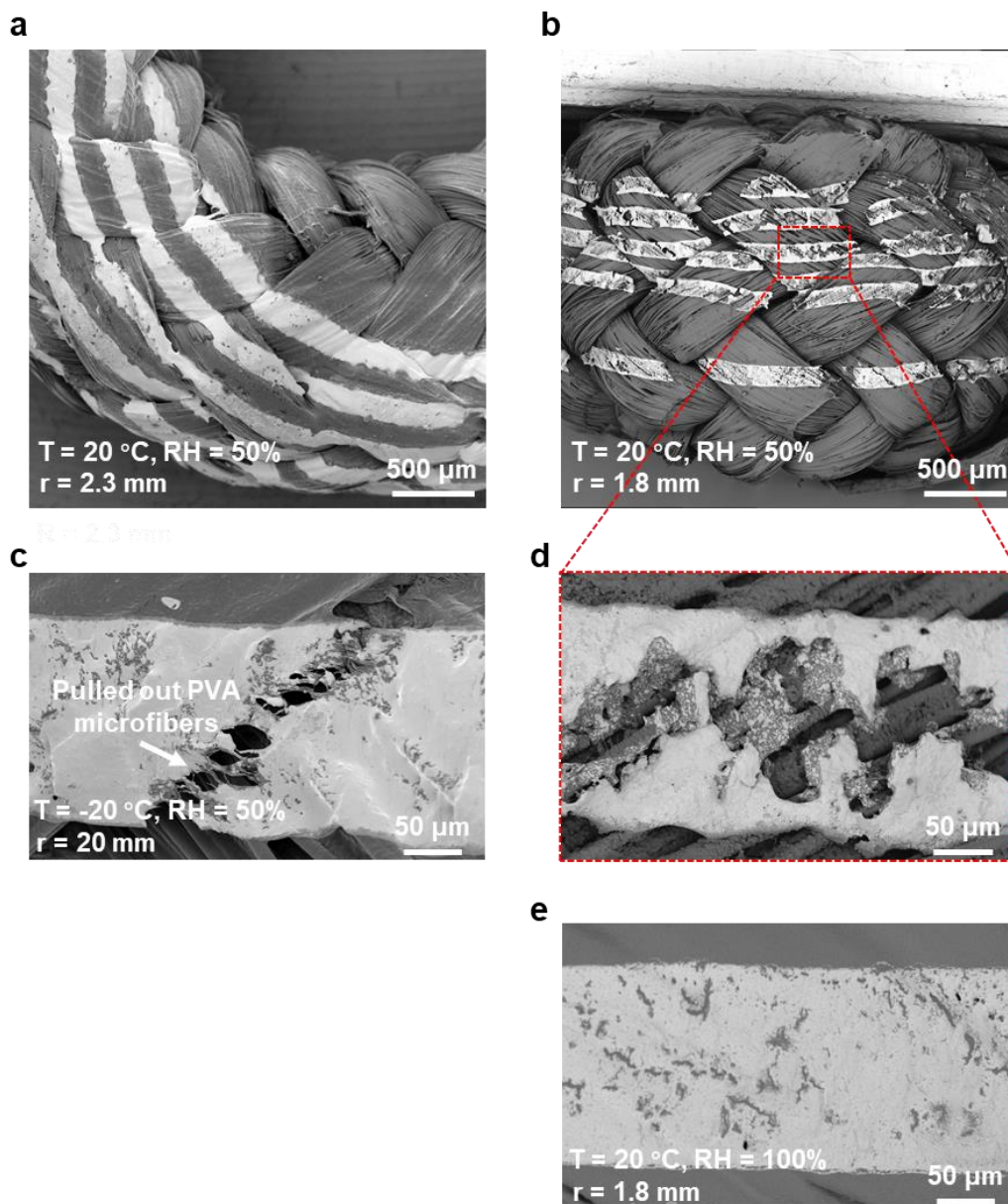
Supplementary Fig. 18. EGaIn microcircuits with different widths. (a) 180 μm , (b) 150 μm , (c) 90 μm , (d) 70 μm , (e) 60 μm , (f) 50 μm . The resolution limit of screen-printed patterns restricts the passage of EMPs, thereby limiting the size of EGaIn microcircuits.



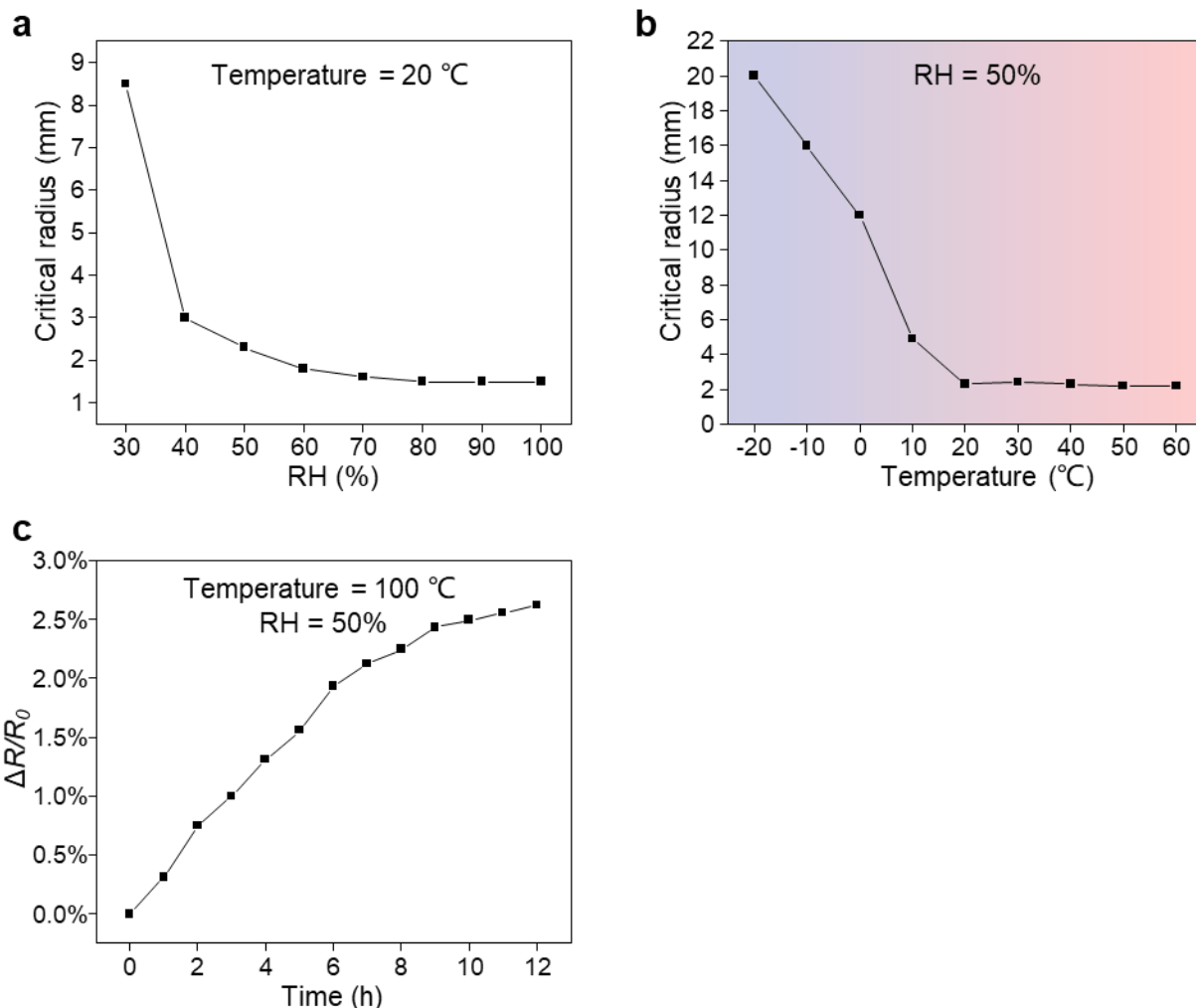
Supplementary Fig. 19. Optical images of EGaIn microcircuits on fibers with different materials and diameters. (a) Thermoplastic polyurethane (TPU), smooth surface, diameter 1.5 mm. (b) High-density polyethylene (HDPE), rough surface, diameter 2 mm. (c) Kevlar, rough surface, diameter 2 mm.



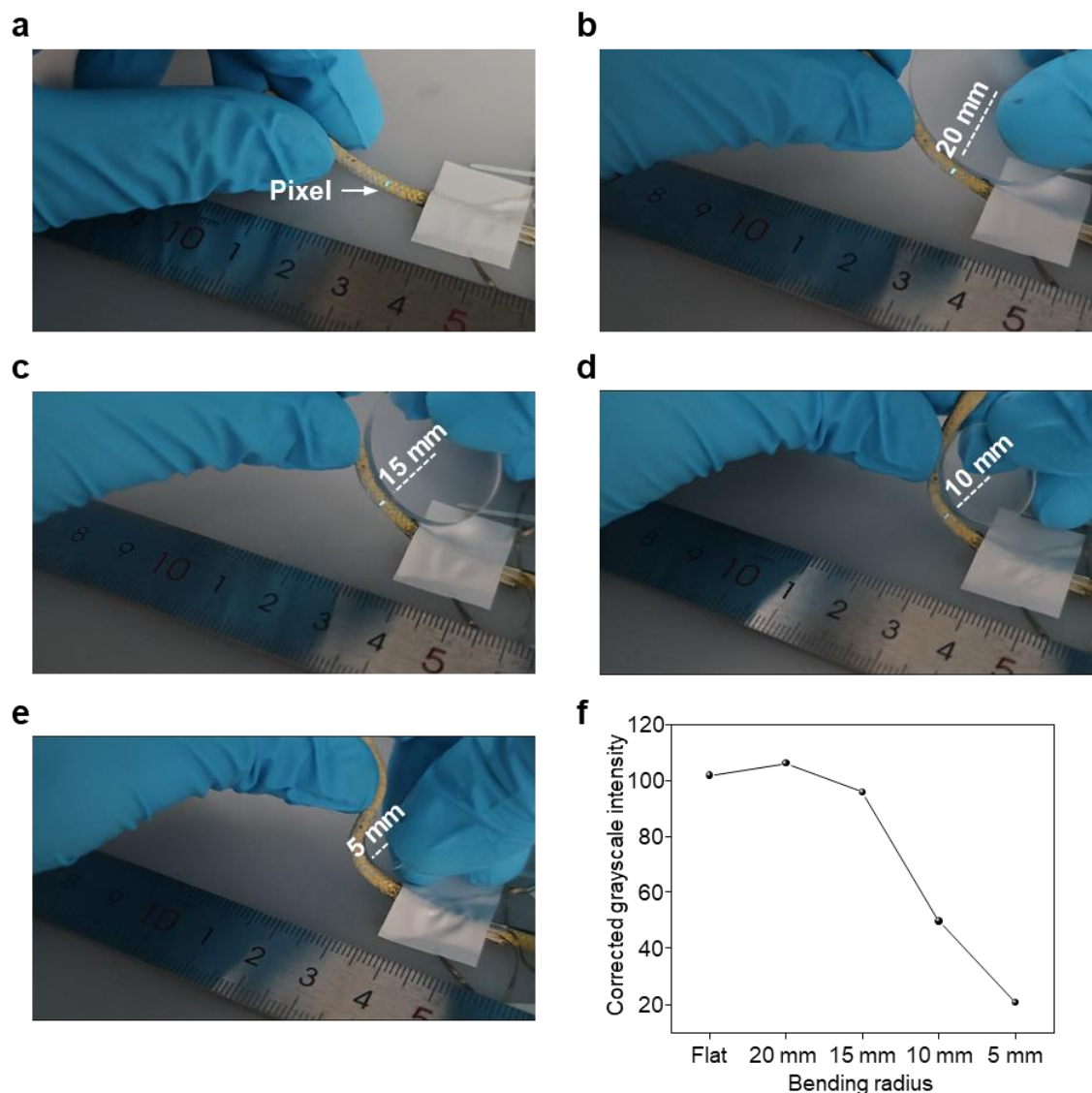
Supplementary Fig. 20. Optical image of a fiber (diameter, 1.5 mm) with two layers of EGaIn microcircuits.



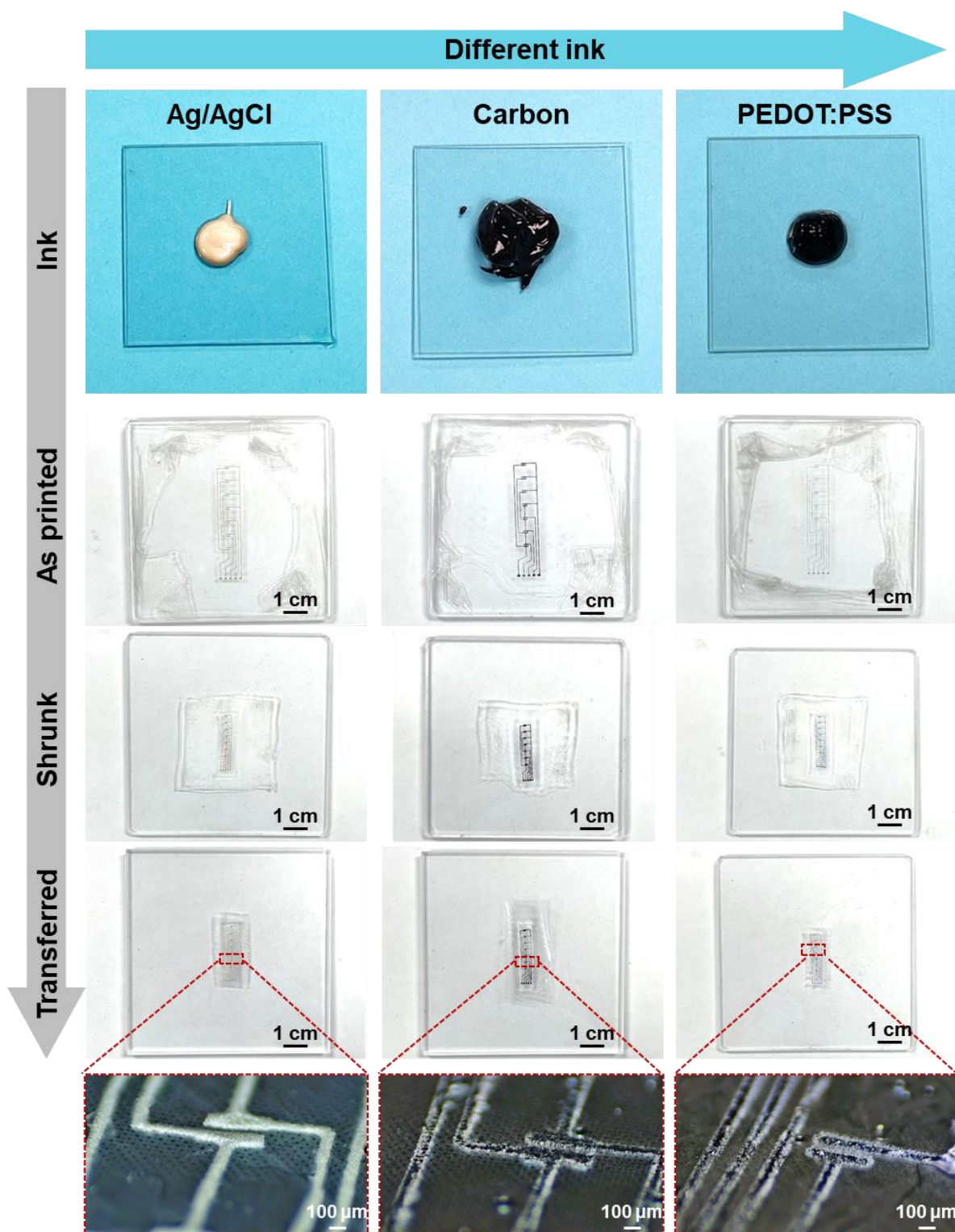
Supplementary Fig. 21. SEM images of the circuit-fiber interface at different temperature and humidity. (a) Interfacial morphology at 20 °C and 50% relative humidity (RH). Bending radius: 2.3 mm. (b) Morphology of the interface after bending-induced failure under the same environment conditions as in (a). Bending radius: 1.8 mm. (c) Image of the fracture at -20 °C and 50% RH, showing PVA microfibrils at the failure cross-section, indicating PVA-dominated interfacial failure. (d) Magnified view of (b), revealing abundant EGaIn speckles at the fractured region, indicating plastic deformation of the PVA. (e) Intact interface at bending radius of 1.8 mm under 20 °C and 100% RH, in contrast to the fractured interface in (d).



Supplementary Fig. 22. Influence of humidity and temperature on the mechanical stability of EGaIn microcircuits on fibers. (a) Influence of RH on the critical radius at 20 °C. Critical radius is defined as the bending radius at which $\Delta R/R_0$ begins to exceed 200%. (b) Influence of temperature on the critical radius at 50% RH. (c) Resistance variation of the microcircuit during prolonged thermal exposure at 100 °C and RH of 50%. All samples were stored under the corresponding environmental conditions for 3 h to ensure equilibrium.



Supplementary Fig. 23. Evolution of brightness of luminance pixel with decrease of bending radius. (a-e) Optical images of a single EL pixel on the fiber device bent to different curvature radius ((a) flat, (b) 20 mm, (c) 15 mm, (d) 10 mm, (e) 5 mm). (f) Corrected grayscale intensity extracted from a fixed region of interest (ROI) for each pixel. Pixel brightness was quantified using ImageJ by measuring the mean grayscale value and subtracting a nearby background region to remove illumination variations.



Supplementary Fig. 24. Applying STAP process using three typical commercial electronic inks. The inks include Ag/AgCl ink (purchased from Prtronic), carbon ink (purchased from Loctite), and PEDOT:PSS ink (purchased from Prtronic), showing consistent printing, ~80%

shrinkage, and successful transfer, confirming the compatibility of the STAP process with other inks.

Table S1. Parameters of screens with different mesh counts, the resolution of as-printed EGaIn/sericin ink, and the resolution after shrinkage (shrunk by 80% in area).

Mesh count (/inch)	EMPs coverage ratio (%)	Resolution (μm)	Resolution-after-extreme shrink (μm)
420	12	Discontinuous	Discontinuous
300	16	136	63
250	21	160	74
200	26	202	95
180	29	245	122
150	34	272	128
120	41	285	138
100	47	300	148
80	54	325	180
60	63	420	221

Table S2. Conductivity of EGaIn microcircuits prepared with different pre-stretch ratio and EMPs coverage ratio.

EMPs coverage ratio (%)	Pre-stretch ratio							
	0%	75%	150%	225%	300%	375%	450%	525%
63.2	0	0.25	0.36	0.39	0.42	0.46	0.47	0.48
53.7	0	0.13	0.18	0.20	0.24	0.29	0.32	0.33
47.4	0	0.06	0.10	0.13	0.18	0.21	0.21	0.24
41.1	0	0/28.8	0.05	0.09	0.15	0.20	0.21	0.21
33.8	0	0/28.3	0/28.5	0.02	0.08	0.17	0.19	0.21
29.2	0	0/27.6	0/28.5	0.01	0.05	0.11	0.16	0.17
26.3	0	0/27.2	0/28.2	0/28.4	0.02	0.02	0.03	0.04
20.6	0	0	0/27.9	0/28.2	0/28.2	0/28.4	0/28.4	0/28.5
16.4	0	0	0	0/27.3	0/27.3	0/27.5	0/27.5	0/27.5
12.4	0	0	0	0	0	0	0	0

Note: Corresponding to the measured electrical conductivity ($\times 10^5 \text{ S m}^{-1}$) after shrinkage. A single non-zero value indicates conductivity obtained by shrinkage alone. Entries in the form “0/x” denote circuits that remained non-conductive after shrinkage but became conductive ($x \times 10^5 \text{ S m}^{-1}$) after transfer.