Supporting Information

Rapid Prototyping Heterostructured Organic Microelectronics using Wax Printing, Filtration and Transfer

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Table S1: Standard PEDTO:PSS patterning methods in comparison with the here described

 wax printing assisted a-PEDOT:PSS microgel filtration

| Patterning Technique | Patterning Resolution | Fabrication time | Fabrication cost | Multilayer stacking | conductivity |
|--|--|------------------------|------------------|-----------------------------------|---|
| Wax printing microgel filtration | ~100 µm | fast | low | simple manual paste/peeling | ${\sim}25~\Omega~sq^{-1}$ |
| Photolithography based patterning | 5 μm* ¹ / 10 μm ² | slow | high | possible | ${\sim}60~\Omega~sq^{-12}$ |
| Screen Printing | $\sim 100 \ \mu m^{3}$ | fast | low | challenging | - |
| Inkjet Printing | 5 μm* ^{1/} 44μm ⁴ | Intermediate - slow | low | challenging | 45 Ω sq ^{-1 4} |
| 3D Printing | 30 µm ⁵ | intermediate | intermediate | Possible | 28 S cm ⁻¹ as hydrogel / 155 S cm ⁻¹ in dry state ⁵ |

* Inkjet printed but relies on photolithography patterned hydrophobic surface.



Figure S1. A) AFM height and phase images of pristine PEDOT:PSS films and films of acidified PEDOT:PSS with roughness average (Ra) and roughness root mean square (Rq). B) Sheet resistance of the films obtained by filtering different concentrations of a-PEDOT:PSS volumes per wax free membrane area, and subsequently treated in methanol and water.

Self-healing PEDOT:PSS micropatterns

It has previously been observed that dried PEDOT:PSS films can partially re-dispersed in water. For films with higher thickness, this process allows the edges of the films to solubilize and migrate under water treatment, therefore filling up the cracks and cuts on the film.⁶ Dried, acidified PEDOT:PSS films retained the ability to be re-dispersed and therefore can be healed by water. As shown in Figure S2, we made a cut on a bar-bell a-PEDOT:PSS electrode and disconnected it. After dropping 2 μ L of water onto the cut, the edges became immediately blurry as the film started to swell into microgel and re-solubilize. Meanwhile, we observed a recovery of the film conductivity, which suggested that the mobilized gel particles had reconnected and formed a conducting pathway. Real-time monitoring of the current at 10 mV DC bias showed that the resistance of the film was relatively stable until the film was dry, upon which the resistance recovered to the level before cut. Further, the gel nature of acidified PEDOT:PSS enables it to be stretchable in the hydrated state.



Figure S2. Healing of acidified PEDOT:PSS. (A) to (F) Cutting a patterned a-PEDOT:PSS electrode and healing it with water. (G) Real time current response of the electrode as showed in (A) to (F). (H) Zoom in of (B), showing the cut. (I) Zoom in of (F), showing the healed cut.

Strechable a-PEDOT:PSS micropatterns

As shown in Figure S3, we patterned a-PEDOT:PSS into two basic shapes: barbell and circle. We stretched the barbell electrode along the axis direction. Unlike pristine PEDOT:PSS, which shows cracking at less than 10% of strain, the barbell electrode only cracked at over 30% of strain along the stretching direction. We then stretched the circle shaped pattern bilaterally, this results in a non-uniform stretching of a-PEDOT:PSS. While the barbell cracked into stripes, the round circle broke into fish scale-like cracks, which sustained a strain of over 70% along one stretching direction.



Figure S3. Stretching of patterned a-PEDOT:PSS microgel. (A) to (E) Unilateral stretching of an a-PEDOT:PSS bar bell showing the breaking of the barbell electrode at the "bar" region under around 30% strain. (F) to (J) Non-uniform stretching of round a-PEDOT:PSS electrodes showing the breaking at more than 70% strain along the lateral direction. The white arrows indicate the pulling directions.



Figure S4. (A) CV scanning of an a-PEDOT:PSS bar-bell electrode at different scan rate. (B) Calibration curve for estimating the surface area of a-PEDOT:PSS barbell electrodes using Randels-Sevick equation.⁷



Figure S5. LUHMES cells on a-PEDOT:PSS electrodes on tape. Fluorescent image stained

with phalloidin (green) and Hoechst (blue). Cells were fixed on differentiation day 5.

References

- 1. H. Sirringhaus, T. Kawase, R. Friend, T. Shimoda, M. Inbasekaran, W. Wu and E. Woo, *Science*, 2000, **290**, 2123-2126.
- 2. S. Ouyang, Y. Xie, D. Wang, D. Zhu, X. Xu, T. Tan, J. DeFranco and H. H. Fong, Journal of Polymer Science Part B: Polymer Physics, 2014, **52**, 1221-1226.
- 3. M. Zabihipour, R. Lassnig, J. Strandberg, M. Berggren, S. Fabiano, I. Engquist and P. A. Ersman, *npj Flexible Electronics*, 2020, **4**, 1-8.
- 4. I. Basak, G. Nowicki, B. Ruttens, D. Desta, J. Prooth, M. Jose, S. Nagels, H.-G. Boyen, J. D'Haen and M. Buntinx, *Polymers*, 2020, **12**, 2915.
- 5. H. Yuk, B. Lu, S. Lin, K. Qu, J. Xu, J. Luo and X. Zhao, *Nature Communications*, 2020, **11**, 1604.
- 6. S. Zhang and F. Cicoira, *Advanced Materials*, 2017, **29**, 1703098.
- 7. N. Elgrishi, K. J. Rountree, B. D. McCarthy, E. S. Rountree, T. T. Eisenhart and J. L. Dempsey, *Journal of chemical education*, 2018, **95**, 197-206.