Supporting Information for

# A Generic Strategy to Create Mechanically Interlocked Nanocomposite/Hydrogel Hybrid Electrodes for Epidermal Electronics

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# **Supplementary Figures**



Fig. S1 (a) SEM image showing the microstructures of salicylic acid microrods. (b) Histograms of length and width distributions. Gaussian fits to the distributions (smooth curves) yield  $11.4 \pm 2.7 \mu m$  for the length and  $3.7 \pm 1.3 \mu m$  for the width



Fig. S2 Uniaxial stress-strain curves of SEBS/salicylic acid composite and the corresponding microfoam



Fig. S3 180°-peeling force *versus* displacement for SEBS microfoam thermally laminated on SEBS substrate. The corresponding interfacial toughness is  $1776.5 \text{ Jm}^{-2}$ 



**Fig. S4** Uniaxial stress–strain curve of the SEBS elastomer (left) and PDA/PAM hydrogel (right). The SEBS elastomer has a modulus of 1.3 MPa, a fracture strain of 941%, and a toughness of 1716 MJ m<sup>-3</sup>. The hydrogel has a modulus of 0.54 KPa, a fracture strain of 952%, and a toughness of 1.063 MJ m<sup>-3</sup>



**Fig. S5** Cross-sectional SEM images of microfoam-attached SEBS film (left) and mechanically interlocked hydrogel/microfoam hybrid (right)



**Fig. S6** (a) SEM image of the SEBS microfoam attached to the SEBS substrate. (b) SEM image showing the residual surface of the interlocked hybrid after peeling off the hydrogel layer. The microfoam remains on the substrate and is partially filled with the hydrogel



Fig. S7 SEM image showing SEBS microfoams with different porosity. In the sacrificial template synthesis, the porosity is modulated by the weight ratio ( $\phi$ ) between salicylic acid and SEBS. Scale bars: 10  $\mu$ m



**Fig. S8** Young's Modulus and fracture strain of SEBS sponge with different porosity. The porosity is modulated by the weight ratio ( $\phi$ ) between salicylic acid and SEBS elastomer



**Fig. S9** (a) 180°-peeling force *versus* displacement for tackified microfoam bonded to different substrates, including EVA, PMMA-PnBA, and PET. (b) Corresponding interfacial toughness



**Fig. S10** 180°-peeling force *versus* displacement curves for interlocked EVAmicrofoam/hydrogel hybrids on EVA and SEBS substrates



Fig. S11 Capacitive current *versus* scan rate of Ag NW nanocomposite electrodes with/without microfoam attachment



Fig. S12 Change in the resistance of the hybrid electrode during 1000 stretch-relaxation cycles to 50% strain



Fig. S13 180°-peeling force *versus* displacement curves for PDA-PAM hydrogel on porcine skin. The interfacial adhesion toughness is determined as  $\sim 20 \text{ J m}^{-2}$ 



**Fig. S14** Skin-electrode contact impedance from multiple human subjects at selected frequencies of 10 (left), 100 (middle), and 1000 (right) Hz. The contact impedance decreases in the order of nanocomposite, Ag/AgCl gel, and interlocked hybrid electrodes



Fig. S15 Four-channel EMG waveforms acquired from the epidermal sensing sleeve in response to different hand gestures

## **Supplementary Video**

Video S1 Dynamic peeling process of the hydrogel layer from the interlocked hybrid revealed by optical microscopy