

## **Supporting information**

### **Encapsulated, High-performance, Stretchable Array of Stacked Planar Micro-supercapacitors as Waterproof Wearable Energy Storage Devices**

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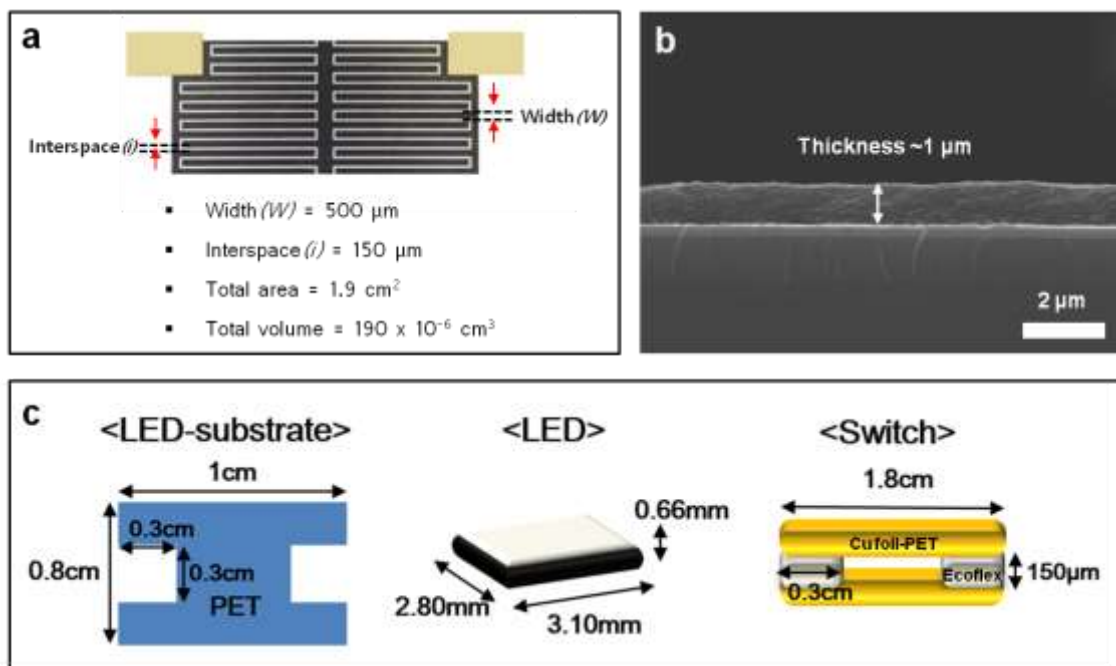
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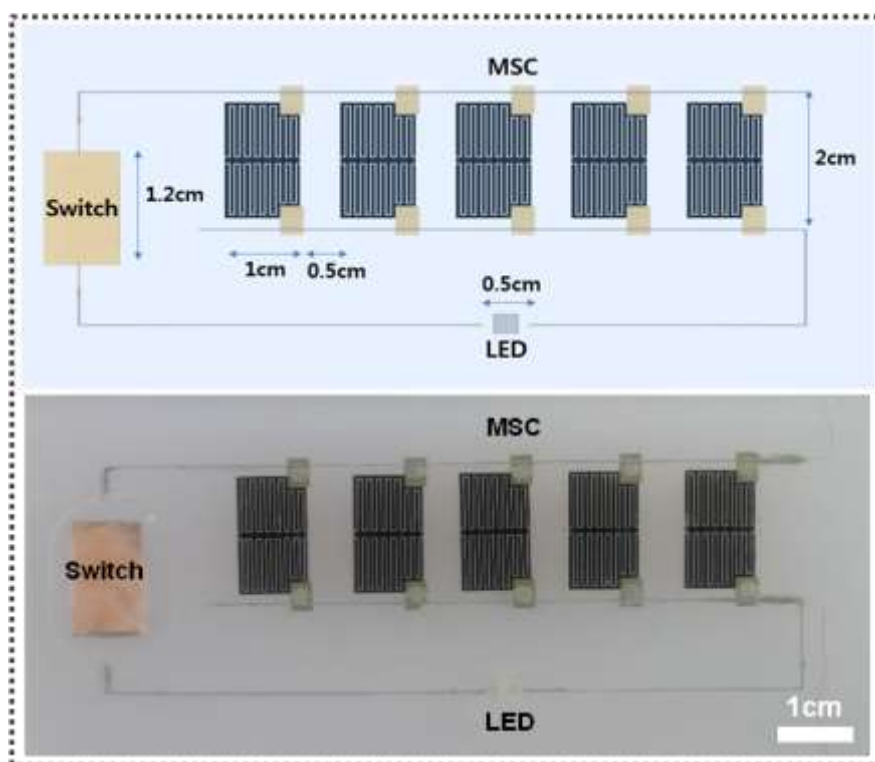
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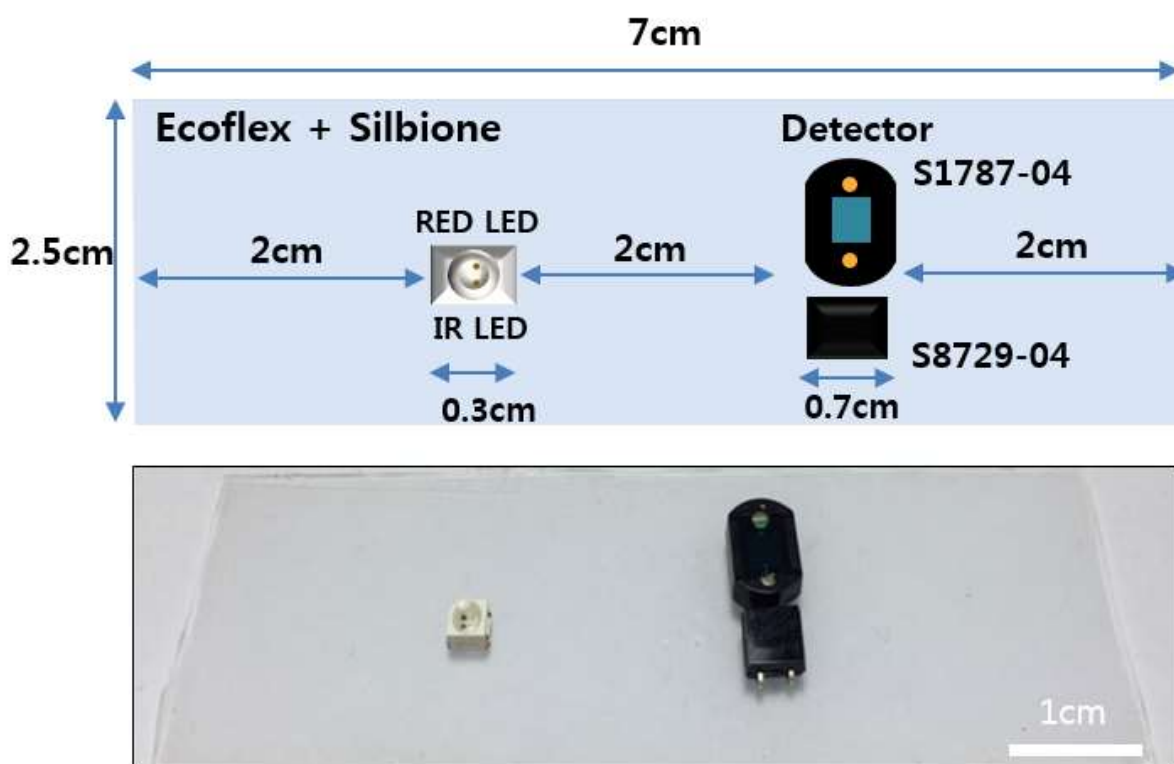
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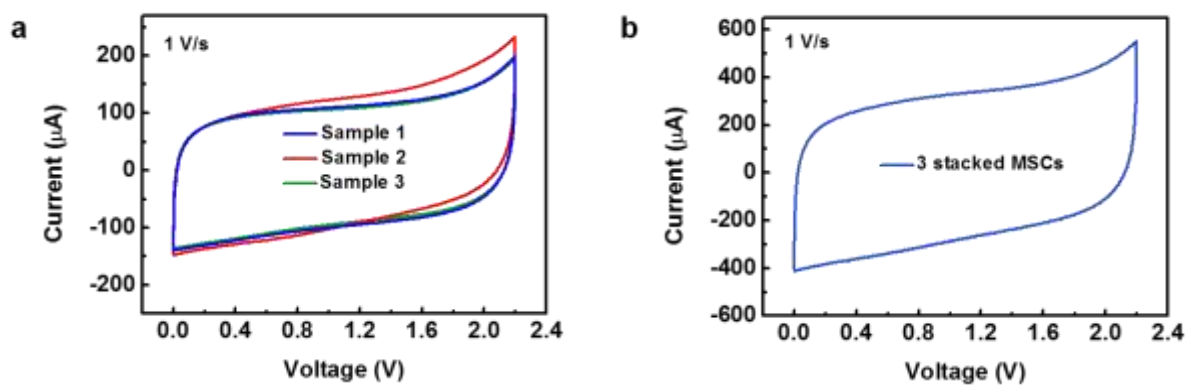
**Figure S1.** (a) Specific dimensions of the fabricated MSC. (b) Cross-sectional SEM image of spray-coated MWNT film. (c) Dimensions of LED-substrate, LED, and switch.



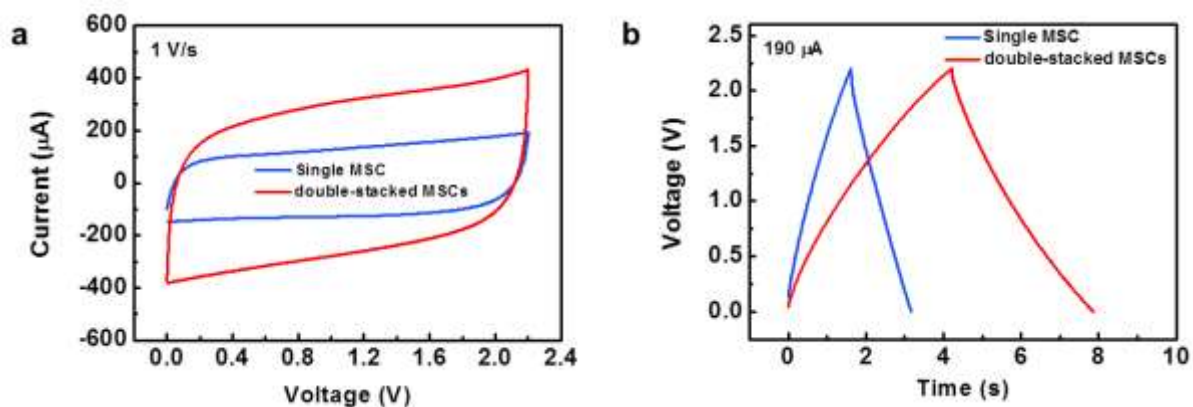
**Figure S2.** Schematic (top) and optical image (bottom) of MSC,  $\mu$ -LED, and switch integrated onto stretchable substrate.



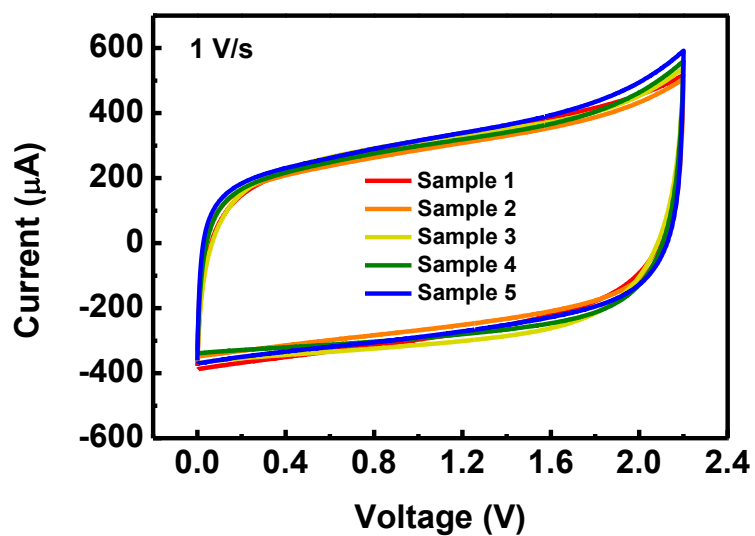
**Figure S3.** Specific dimensions of LEDs and detectors for oximetry.



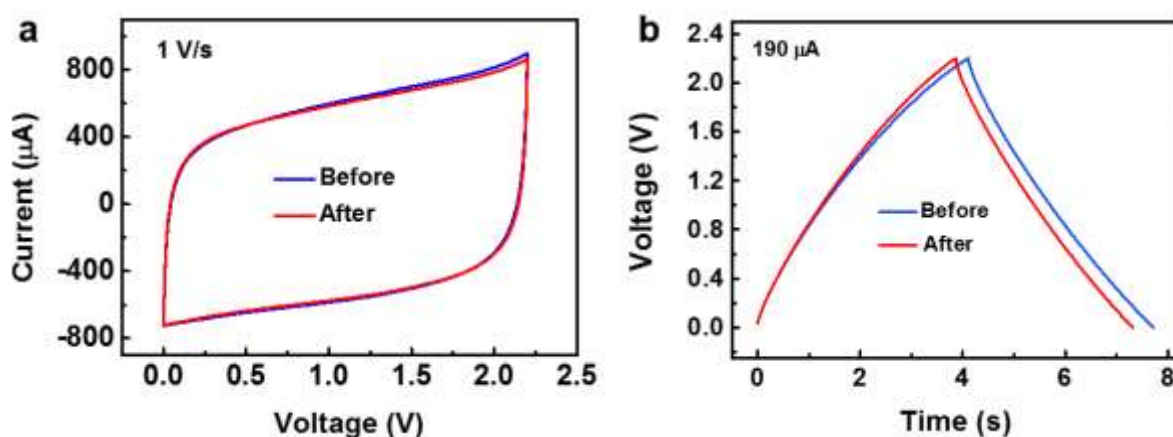
**Figure S4.** (a) Cyclic voltammetry curves of three different MSCs at a scan rate of 1 V/s. (b) Cyclic voltammetry curve of three stacked MSCs at a scan rate of 1 V/s.



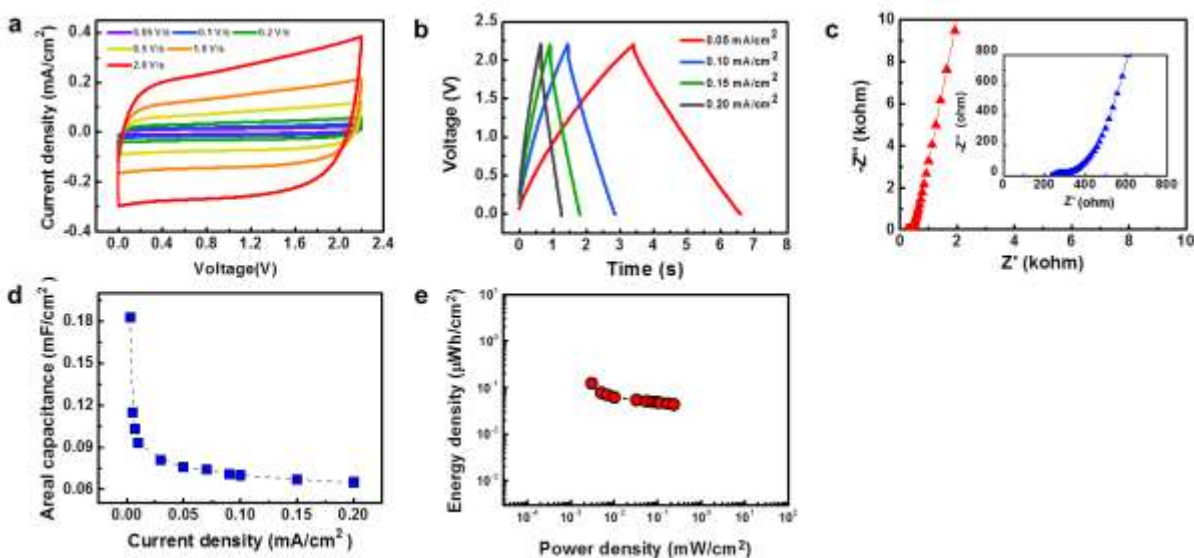
**Figure S5.** Electrochemical performance of single MSC (blue) and double-stacked MSCs (red). (a) CV curves measured at a scan rate of 1 V/s. (b) Galvanostatic charge-discharge curves at a current of 190  $\mu\text{A}$ .



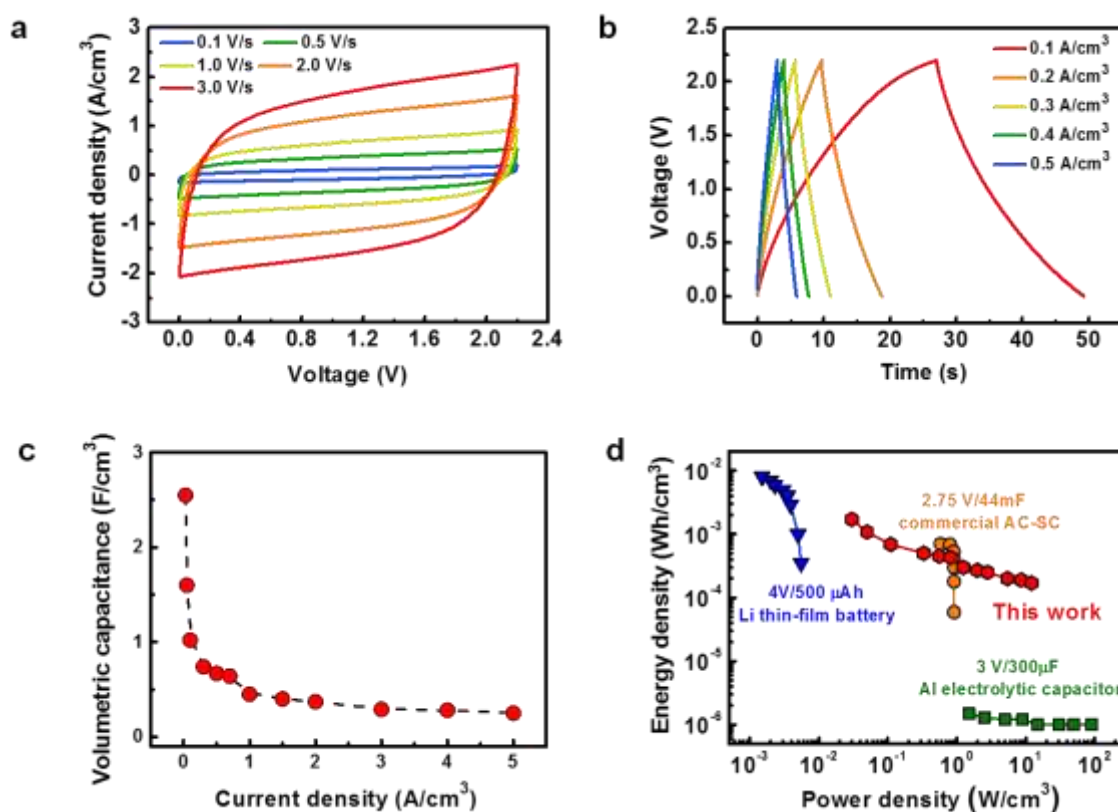
**Figure S6.** CV curves of five different double-stacked MSCs for integration onto stretchable substrate at a scan rate of 1 V/s.



**Figure S7.** Electrochemical performance of double-stacked MSCs before (blue) and after embedding (red) in stretchable polymer substrate. (a) CV curves measured at a scan rate of 1 V/s (b) Galvanostatic charge-discharge curves at a current of 190  $\mu\text{A}$ .

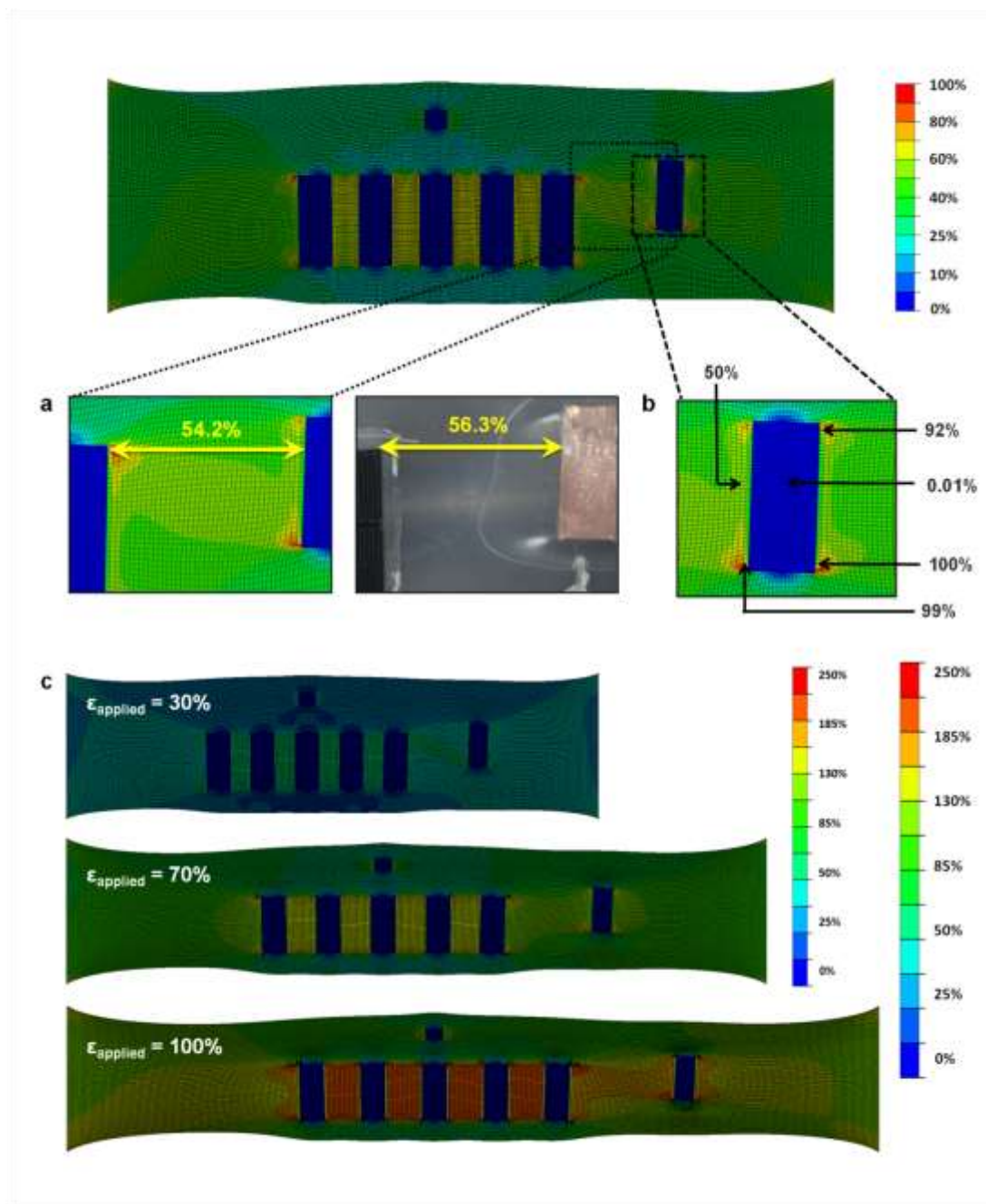


**Figure S8.** Electrochemical performance of single micro-supercapacitor. (a) Cyclic voltammetry curves at various scan rates from 0.05 V/s to 2 V/s. (b) Galvanostatic charge/discharge curves at current density of 0.05, 0.1, 0.15 and 0.2 mA/cm<sup>2</sup>. (c) Nyquist impedance plot at frequency range from 1 MHz to 100 mHz. The ESR value is 234  $\Omega$ . (d) Areal capacitance at various current densities from 0.003 mA/cm<sup>2</sup> to 0.2 mA/cm<sup>2</sup>. (e) Ragone plot of areal energy density vs. areal power density.



**Figure S9.** Electrochemical performance of double-stacked MSC array integrated on the stretchable Ecoflex substrate. (a) Cyclic voltammetry curves at various scan rates from 0.01 V/s to 3 V/s. (b) Galvanostatic charge/discharge curves at current density of 0.1, 0.2, 0.3, 0.4, and 0.5 A/cm³. (c) Volumetric capacitance at various current densities from 0.03 A/cm³ to 5 A/cm³. (d) Ragone plots of volumetric energy density vs. power density.

The volumetric capacitance ( $C_v$ ) of five parallel connected double-stacked MSCs is obtained using the equation  $C_v = C_{\text{cell}}/V_{\text{cell}}$ , where  $V_{\text{cell}}$  is the total volume of 10 MSCs. The total area of a single MSC is calculated to be 1.9 cm², which includes the interspace interval of 150 μm, and total thickness of the current collector (55 nm), and the MWNT film, excluding the electrolyte. The thickness of the spray coated MWNT film is roughly 1 μm; the resultant total volume of a single MSC is roughly  $190 \times 10^{-6}$  cm³. The calculated energy density was more than two times greater than that of commercial AC-SC<sup>1</sup>, and 1,000 times higher than that of the Al electrolytic capacitor<sup>1</sup>; the power density was more than 10 times greater than that of the commercial Ac-SC<sup>1</sup>, and roughly 1,000 times higher than that of a Li thin film battery<sup>2</sup>, respectively.



**Figure S10.** (a) FEM analysis between MSC and a switch integrated onto stretchable substrate with optical image. (b) FEM analysis near switch. (c) FEM analysis under different strains (30%, 70%, and 100%).



## Methods

According to the modified Beer-Lambert law, the absorbance at wavelength  $\lambda$  ( $A_\lambda$ ) is given by<sup>3</sup>

$$A_\lambda = -\ln(T) = -\ln\left(\frac{I}{I_0}\right), \quad (1)$$

where  $T$  represents the fraction of transmitted light that passes through a sample, and where  $I_0$  and  $I$  are the incident and transmitted light intensities, respectively. In this experiment, we used two LEDs [ $\lambda = 670$  nm (red) and 870 nm (infrared)] and two photodetectors to receive signals from a finger. The ratio of the absorbance ( $R$ ) for red and infrared light can be calculated to obtain the peripheral capillary oxygen saturation ( $SpO_2$ ) using the following equation.<sup>4</sup>

$$\begin{aligned} R &= \frac{A_{red}}{A_{ired}} = \frac{(\epsilon_{HbO_2,red} \cdot c_{HbO_2} + \epsilon_{Hb,red} \cdot c_{Hb}) \cdot \Delta l}{(\epsilon_{HbO_2,ired} \cdot c_{HbO_2} + \epsilon_{Hb,ired} \cdot c_{Hb}) \cdot \Delta l} \\ &= \frac{\epsilon_{HbO_2,red} \cdot SpO_2 + \epsilon_{Hb,red} \cdot (1 - SpO_2)}{\epsilon_{HbO_2,ired} \cdot SpO_2 + \epsilon_{Hb,ired} \cdot (1 - SpO_2)} \end{aligned} \quad (2)$$

Here,  $\Delta l$  represents the thickness of the heartbeat-added arterial layer, which is assumed to be same for both wavelengths.  $\epsilon_{HbO_2,red}$  and  $\epsilon_{HbO_2,ired}$  are the molar absorptivity of oxy-hemoglobin for red ( $\lambda = 670$  nm) and infrared ( $\lambda = 870$  nm) wavelengths, respectively.

Finally,  $SpO_2$  can be calculated using the following equation.

$$SpO_2 = - \frac{\epsilon_{Hb,red} - R \cdot \epsilon_{Hb,ired}}{(\epsilon_{HbO_2,red} - \epsilon_{Hb,red}) - R \cdot (\epsilon_{HbO_2,ired} - \epsilon_{Hb,ired})} \quad (3)$$



## Reference

1. El-Kady, M. F.; Strong, V.; Dubin, S.; Kaner, R. B.; Laser Scribing of High-Performance and Flexible Graphene-Based Electrochemical Capacitors. *Science*. **2012**, 335, 1326-1330.
2. Pech, D.; Brunet, M.; Durou, H.; Huang, P.; Mochalin, V.; Gogotsi, Y.; Taberna, P.-L.; Simon, P.; Ultrahigh-Power Micrometre-Sized Supercapacitors Based on Onion-Like Carbon. *Nat. Nanotechnol.* **2010**, 5, 651-654.
3. Obrig, H.; Wenzel, R.; Kohl, M.; Horst, S.; Wobst, P.; Steinbrink, J.; Thomas, F.; Villringer, A.; Near-Infrared Spectroscopy: Does It Function in Functional Activation Studies of the Adult Brain?. *International Journal of Psychophysiology*. **2000**, 35, 125-142.
4. Lochner, C. M.; Khan, Y.; A. Pierre, A. C. Arias. All-Organic Optoelectronic Sensor for Pulse Oximetry. *Nat. Commun.* **2014**, 5, 5745