Electronic Supplementary Information:

Transparent and ultra-bendable all-solid-state supercapacitors without percolation problems

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Supplementary Methods

1. Experiment

A positive photoresist layer was spin coated and developed on a PET substrate using a photomask (MA-6, Karl-suss). A Cr layer and Au layer were sequentially thermally evaporated onto the same layer to serve as an adhesion layer and a supercapacitor current collector was applied (ALPS-C03, alpha plus). Vertically oriented hierarchical MnO2 structure was electrodeposited on the surface of the Au layer. For electrodeposition, the platform was immersed in aqueous solutions of $Mn(NO_3)_2$ (0.02 M) and $NaNO_3$ (0.1 M) (WPG100, Wonatech). The platform was used as the working electrode, and a Pt electrode was used as the counter electrode, an Ag/AgCl electrode used as the reference electrode, and a constant current of 100 μA cm^{-2} was applied by means of a 10 min deposition time. The remaining photoresist was lifted off the photoresist with acetone followed by an overnight drying at room temperature and pressure. A H_3PO_4/PVA gel electrolyte was prepared by mixing PVA powder with water (1 g of PVA / 10 ml of H_2O), H3PO4 (0.8 g) together. The mixture was then heated to around 358K under vigorous stirring until the solution became clear. After cooling down and vaporizing about 48 h under ambient conditions, the electrolyte solidified to form an adhesive gel. After attaching the interdigitated electrode platform and gel membrane electrolyte, a transparent and flexible supercapcitor with interdigitated electrodes was finally produced.

2. Characterization

Magnified optical image image and SEM images of micro electrodes in the device were obtained using an optical microscope (HAL 100, Carl Zeiss) and field-emission scanning electron microscope (AURIGA, Carl Zeiss). UV-visible (UV-Vis) spectroscopy (Agilent

8453, HP) is used to determine the transmittance of each elements of device. CV and EIS measurements were carried out using a computer-controlled potentiostat (Iviumstat, Ivium) and C/D measurements were carried out using galvanostat (ZIVE sp2, ZIVE LAB).

3. Calculation of specific capacitance

The specific capacitances of symmetrical supercapacitors are calculated as follows. The capacitance (*C*) was calculated using the voltammetric discharge integrated from the cyclic voltammogram according to the following equation;[S1]

$$C = \frac{\int i\, dt}{\Delta E} = \frac{Q}{\Delta E} \qquad (1)$$

where *i* is the current (*A*), *t* the time (s) and ΔE is the potential window (*V*).

The specific capacitance (C_{sp}) is;[S2]

$$C_{sp} = \frac{4C}{M} \qquad (2)$$

where *M* is the total weight of both the positive and negative electrodes.

4. References

S1. Q. Lu, M. W. Lattanzi, Y. Chen, X. Kou, W. Li, X. Fan, K. M. Unruh, J. G. Chen and J. Q. Xiao, *Angew. Chem. Int. Ed.*, 2011, **50**, 6847.

S2. L. Demarconnay, E. Raymundo-Piñero and F. Béguin, *Electrochem. Commun.*, 2010, **12**, 1275; V. Khomenko, E. Raymundo-Piñero and F. Béguin, *J. Power Sources*, 2006, **153**, 183; J. H. Park and O. O. Park, *J. Power Sources*, 2002, **111**, 185.

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Supplementary Figures

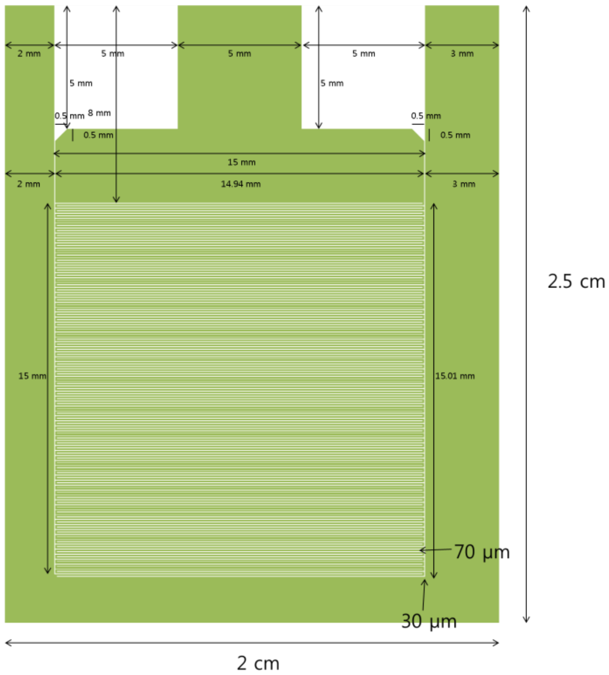

Fig. S1. Pattern design and linear dimensions of an interdigitated electrode system on a transparent and flexible supercapacitor without percolation. With 150 interdigitated electrodes, each microelectrode was 30 μm in width and 1.5 cm in length. The electrodes were interspaced by a distance of 70 μm. The occupied area of the transparent electrode was 1.5 cm × 1.5 cm. This area can be increased up to an area limit of the electrodeposition method (size of the counter electrode).

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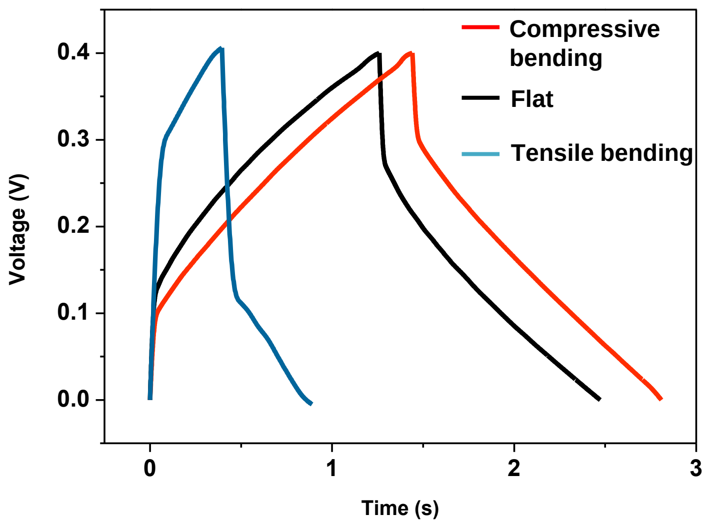

Fig. S2. Typical shapes of charge/discharge curve were observed for the different bending states of transparent and ultra-bendable supercapacitor with interdigitated patterned electrode system; flat, compressive and tensile bending state. Current density is 150μA cm^{-2} and bending radius is 1.5 mm (curvature is 6.7 cm^{-1}).

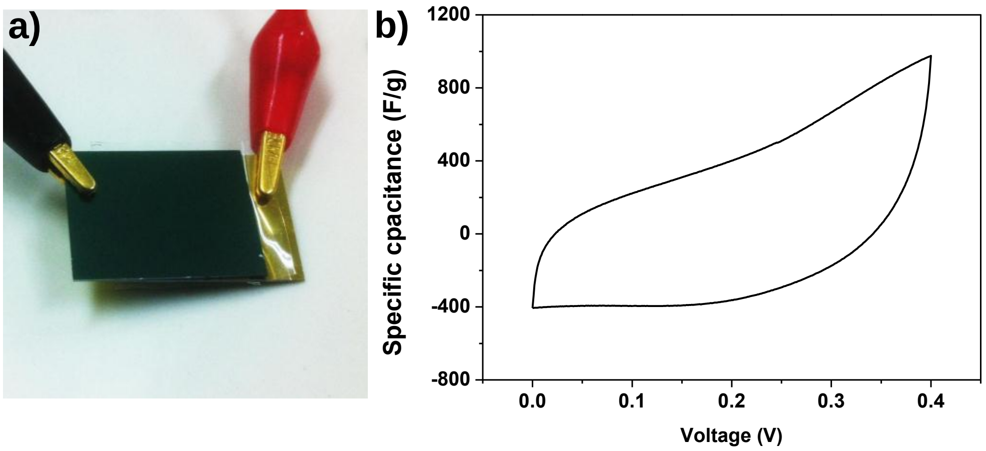

Fig. S3. a) Photograph and b) Cyclic voltammogram for a conventional sandwich type supercapacitor based on the same hierarchical MnO_2 structure.