Electronic Supplementary Information:

All-solid-state, origami-type foldable supercapacitor chip with integrated series circuit analogues

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Contents

Polarization of isolated electrodes (IEs)

Conventional series circuit systems are expressed by an equivalent circuit in Fig. S1a. At charging state, each capacitor units in series circuit (blue boxes in Fig. S1b) has a charge separation (Fig. S1b) therefore potential profile of the system will be generated as shown in Fig. S1c. The conventional series system includes the external lines connecting between each units (black line in Fig. S1b), which inevitably accompanies additional resistance and requires an unnecessary volume in the system.

In this research, we replaced two electrodes including different capacitor units and external line with just one electrode material (isolated electrodes (IEs)). In an equivalent circuit, the IEs are represented by red boxes in Fig. S1d. Through the equivalent circuit, IEs are completely isolated and only connected by sectionalized ion transferring papers (SITPs, yellow boxes in Fig. S1e). The narrow middle region of the IEs plays a role as an external line in a conventional series circuit. At charging, the IE materials should be polarized in horizontal direction according to the equivalent circuit in red boxes. In the manuscript, the horizontal direction is expressed by *x*-axis in Fig. 1. The polarization can be proved by the potential profile of the series circuit analogues system (Fig. S1f). From the potential profile, it is known that a limited potential in a conventional energy storage cell is placed between adjacent IEs. We measured the potential between neighboring IEs in the fully charged three IEs system (in the system, fully charged potential is 3.2 V). Each potential window between the neighboring IEs is about 0.8 V in Fig. S2, which proves that the IEs are polarized.

Supplementary figures

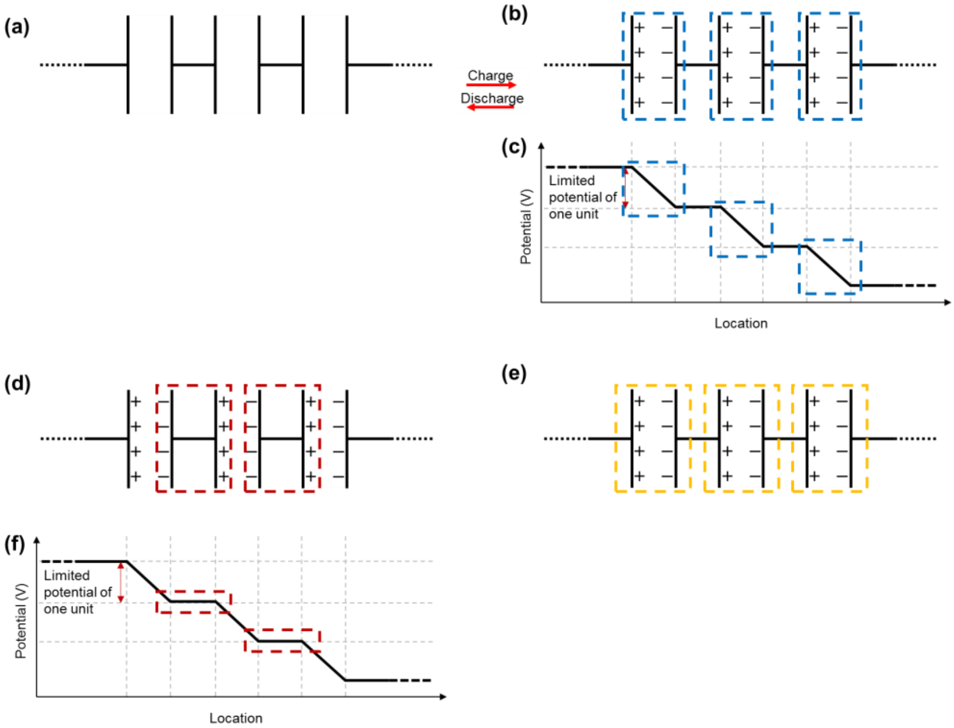

Fig. S1. (a) Discharged and (b) charged equivalent circuits of capacitor systems with series connection. (c) Potential diagram versus location (*x*-axis) of charged capacitor system with series connection. The blue boxes represent conventional energy storage units. (d) and (e) equivalent circuits of isolated electrodes (IEs) system. The red boxes represent IEs and the yellow boxes represent sectionalized ion transferring papers (SITPs). (f) Potential diagram versus location (*x*-axis) of IEs system. Potential gap between neighboring IEs are the same with the limited potential of conventional system.

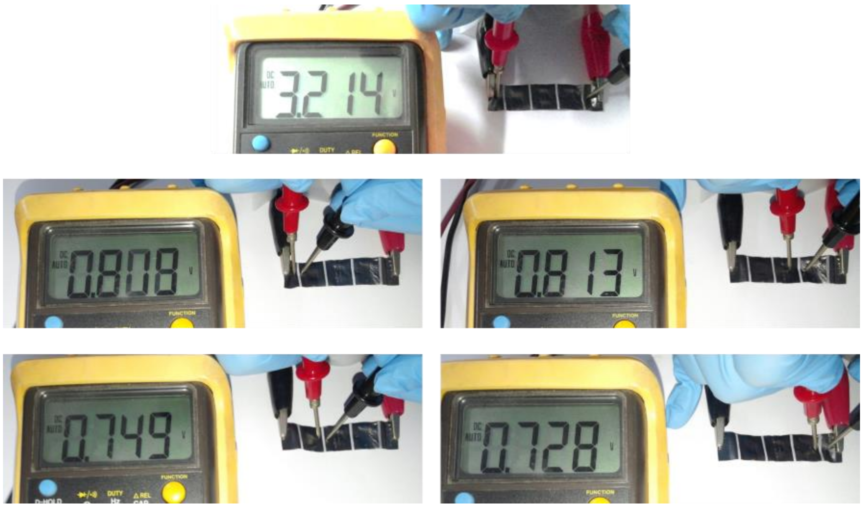

Fig. S2. Potential gap between near isolated electrodes (IEs) in fully charged three IEs system (Fully charged potential is about 3.2 V). Each potential windows between near IEs is about 0.8 V, which proves the IEs are polarized well.

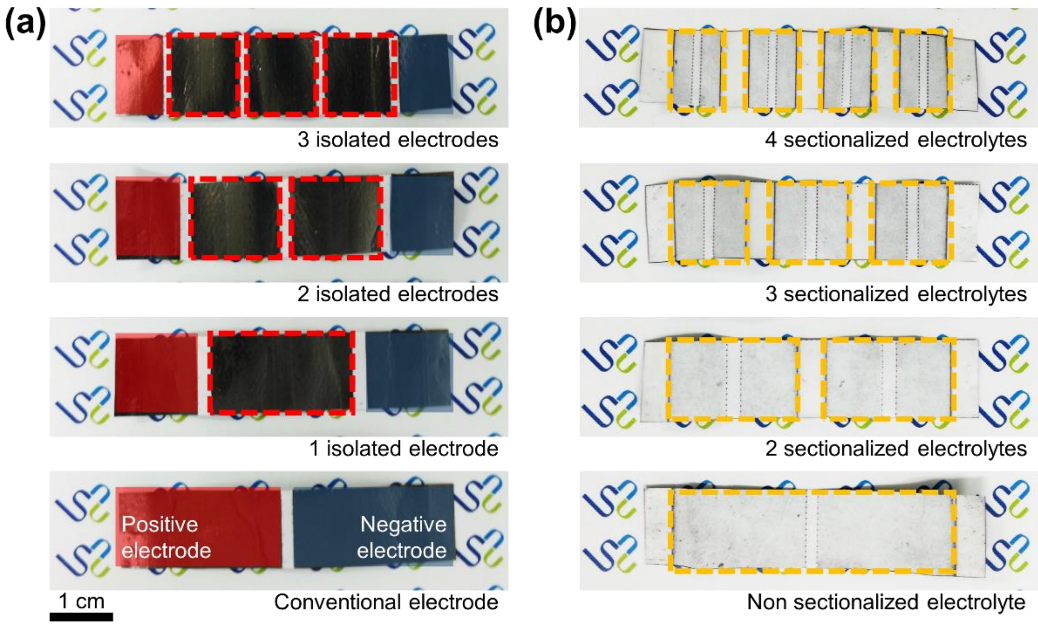

Fig. S3 Photographs of IEs systems. (a) Front sides of conventional type (without IE) and IEs systems with 1, 2 and 3 IEs, respectively. The red boxes enclose each IEs. (b) Back sides of the conventional type (without IE) and IEs systems with 1, 2 and 3 IEs, respectively. The sectionalized electrolytes are absorbed on the back side of the system and the yellow boxes represent each of the sectionalized electrolytes.

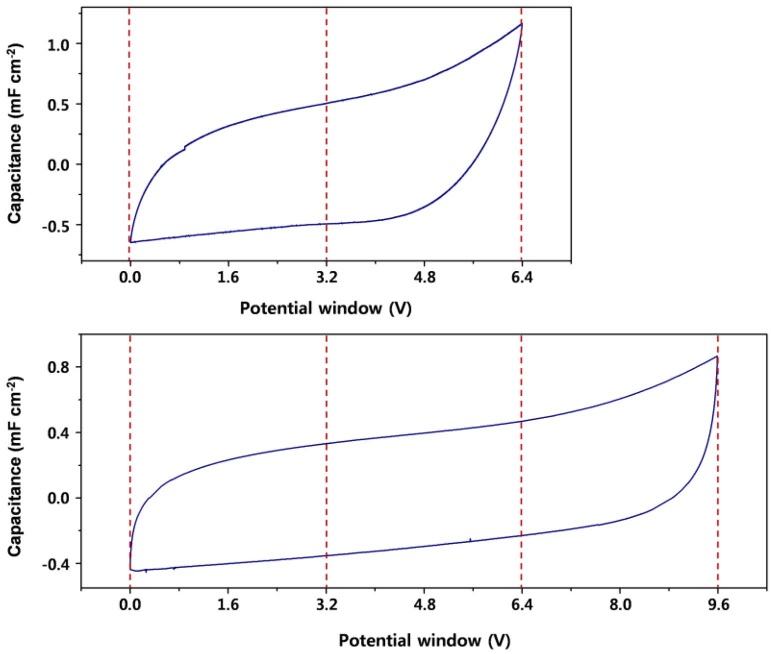

Fig. S4. Cyclic voltammograms (CV) of isolated electrode (IEs) energy storage systems with (a) 7 and (b) 11 IEs. Scan rate is 5 mV s^{-1}. The system have 6.4 V and 9.6 V of potential window respectively, which proved the linearly increasing potential window with the number of IEs based on the same area systems.

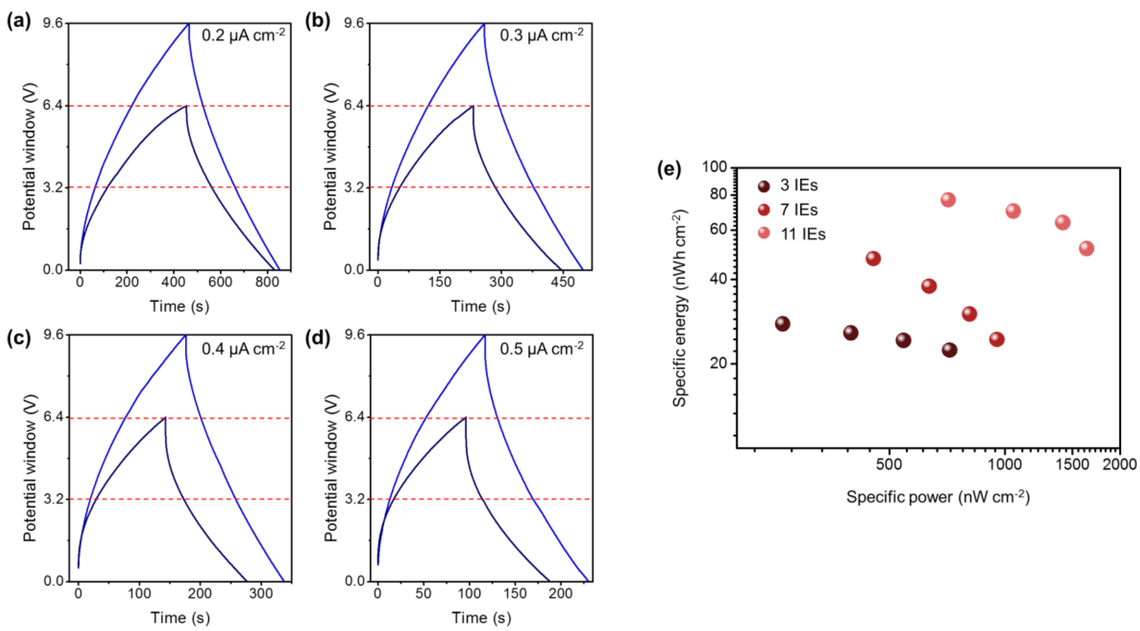

Fig. S5. Galvanostatic charge/discharge (C/D) curves of system with 7 and 11 isolated electrodes (IEs) with same-sized substrate. The specific currents are (a) 0.2, (b) 0.3, (c) 0.4 and (d) 0.5 μA cm^{-2}, respectively. (e) Ragon plot of IEs system with 3, 7 and 11 IEs.

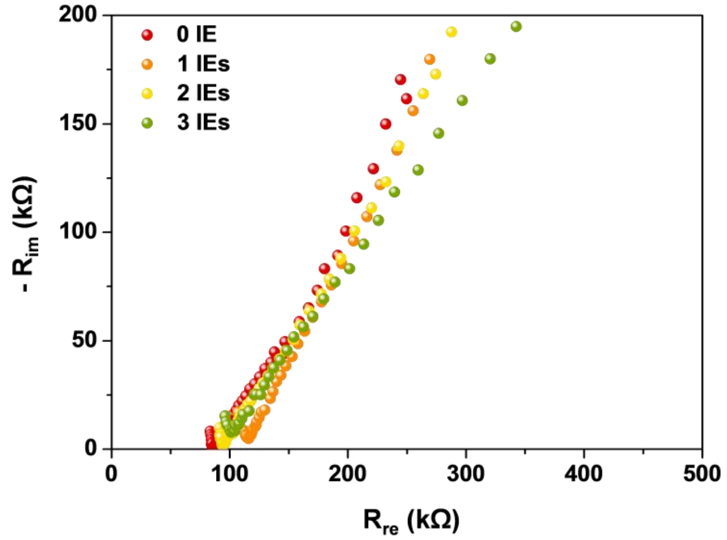

Fig. S6. Nyquist impedance plots of energy system without isolated electrode (IE) and with 1, 2 and 3 IEs. Applied potential is 0.4, 0.8, 1.2 and 1.6 V, respectively. Ac amplitude is 10 mV and frequencies of 100 mHz – 50 kHz. The curves for the various IEs systems have tangent line that intersects the real axis, which caused by synergetic effect of capacitance and mass transfer resistance caused the electrode system with vertical formation.[S1-S4]

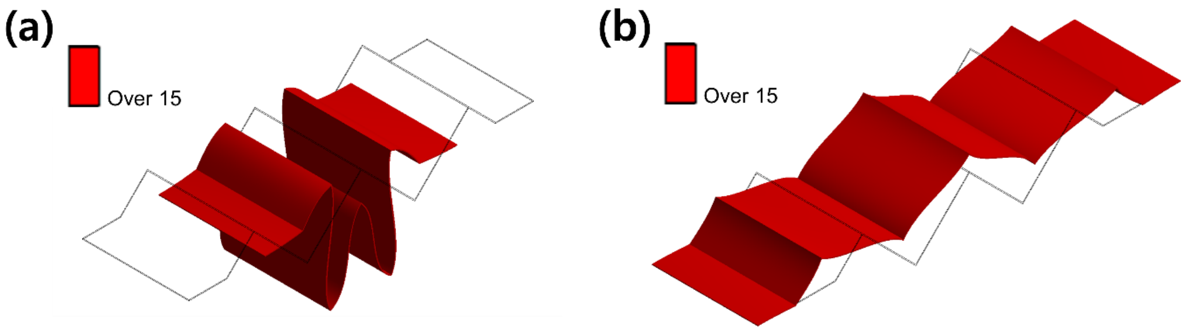

Fig. S7. Safety factor analysis of origami-based 7 folding 3 isolated electrodes (IEs) system. (a) - 60 % compressive and (b) 30% tensile decomposition states.

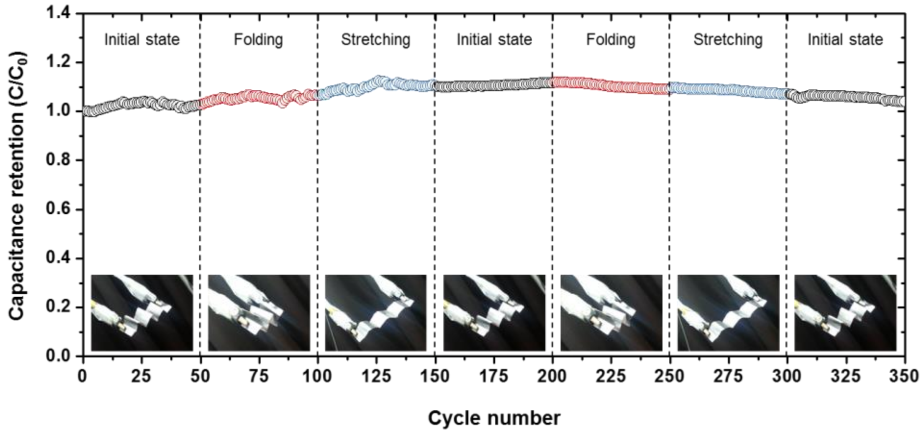

Fig. S8. Capacitance retention *versus* cycle number for the origami-type three isolated electrodes (IEs) system septuple-folded in a zigzag formation at various deformation states.

Supplementary table

Table S1 Total energy in eV for each molecular structure component and the thermodynamic decomposition voltage of poly(vinyl alcohol) (PVA) at half and full decomposition by density functional theory (DFT) calculation.[7]

Component	Energy [eV]	Decomposition state	Decomposition voltage [V]
$-CH_2-CHOH-$	-39.78	Half	1.18
$-CH-CH-$	-24.97	Full	0.80
$-CH_2-CHOH-CH-CH-$	-64.36		
H_2O	-14.01		

Reference

S1. O. E. Barcia, E. D'Elia, I. Frateur, O. R. Mattos, N. Pebere and B. Tribollet, *Electrochim*. *Acta* 2002, **47**, 2109-2116.

S2. W. G. Pell and B. E. Conway, *J*. *Electroanal*. *Chem*. 2001, **500**, 121-133.

S3. J. P. Meyers, M. Doyle, R. M. Darling and J. Newman, *J*. *Electrochem*. *Soc*. 2000, **147**, 2930-2940.

S4. H. K. Song, Y.-H. Jung, K.-H. Lee and L. H. Dao, *Electrochim*. *Acta* 1999, **44**, 3513-3519.