A universal *in situ* strategy for charging supercapacitors

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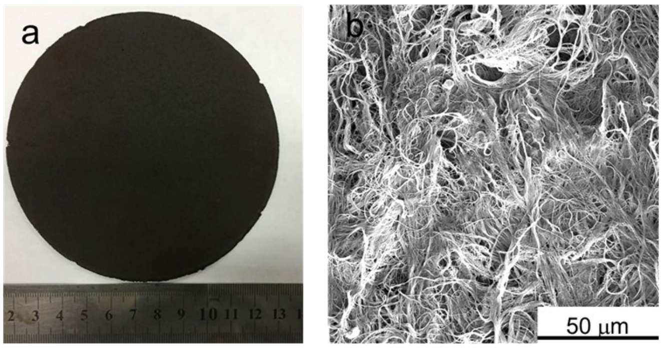

Fig. S1 (a) Digital photographs of free-standing PANI/CNT thin film. (b) Low magnification SEM image of PANI/CNT composites.

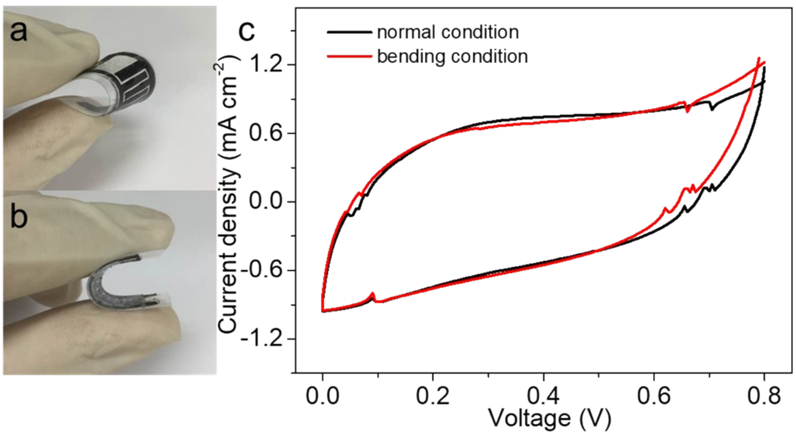

Fig. S2 (a)-(b) Digital photographs of self-charging SC in bending state. (c) CV curves of SC in normal condition and bending condition (180°).

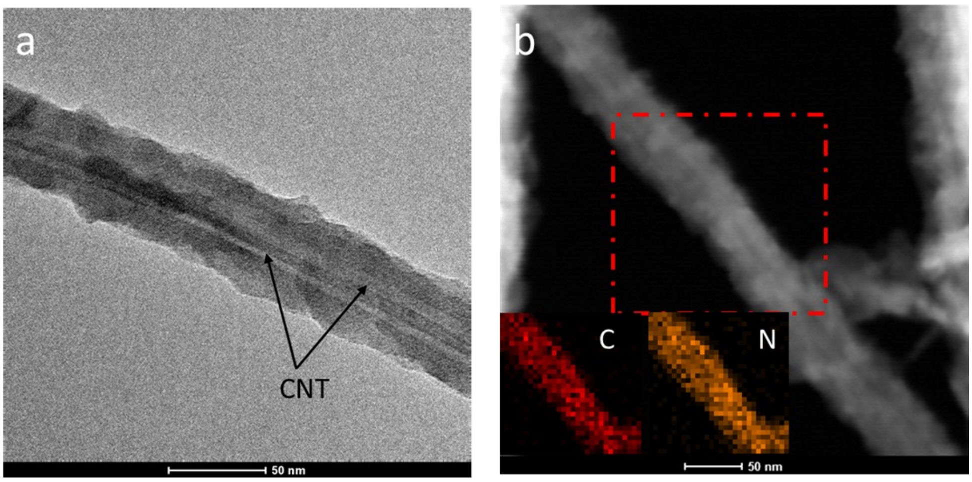

Fig. S3 (a) High resolution transmission electron microscopy (TEM) shows the wrapping of PANI on CNTs. (b) Corresponding energy dispersive spectroscopy (EDS) of selected area of a TEM image. The insets show the EDS results of C and N elements where the N elements origin from aniline (C_6H_7N).

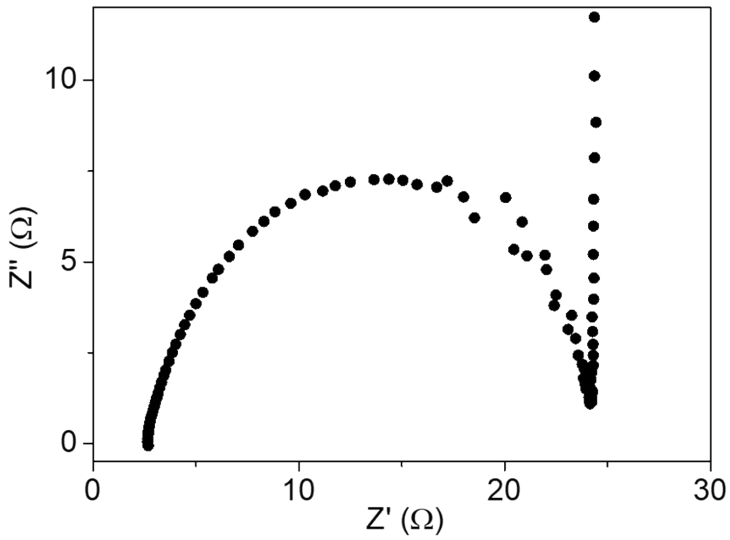

Fig. S4 Electrochemical impedance spectroscopy of PANI/CNT electrodes in 1 M H_2SO_4 solution.

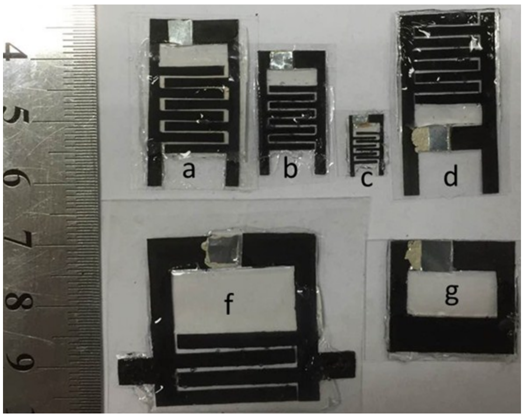

Fig. S5 Digital photographs of self-charging device with different (a)-(b) sizes and (d)-(g) designs. Fig. S5g shows a sandwich type self-charging SC.

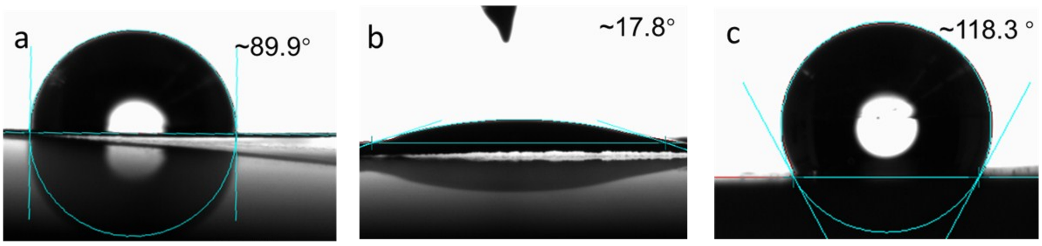

Fig. S6 Contact angles of (a) Zn foil, (b) PANI/CNT composite and (c) silicone rubber.

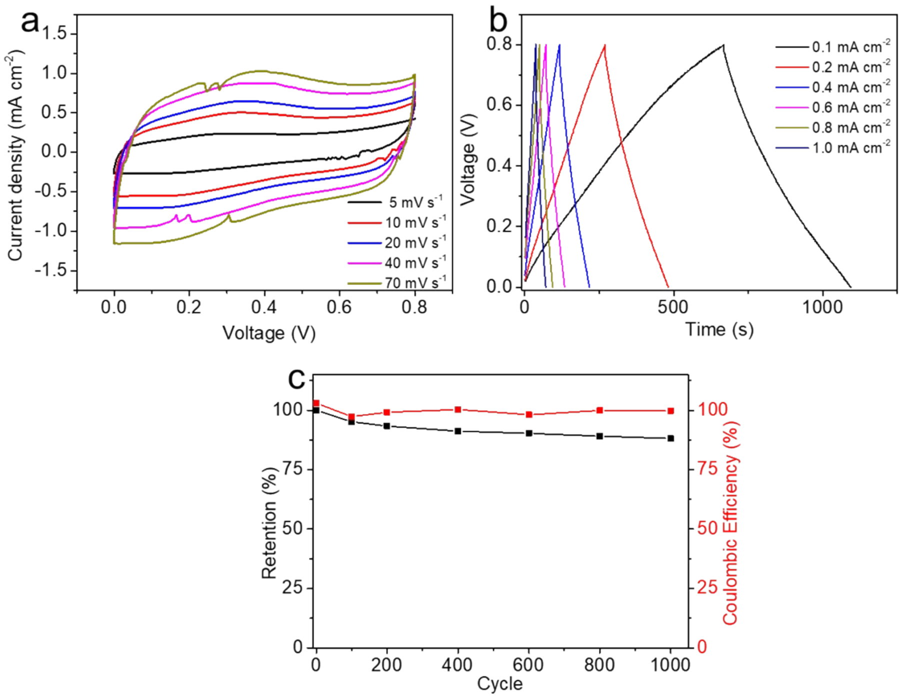

Fig. S7 (a) CV curves of SCs at different scan rate. (b) GCD curves of SCs at different current density. (c) Cycling performance of SCs at the current density of 0.4 mA cm^{-2}.

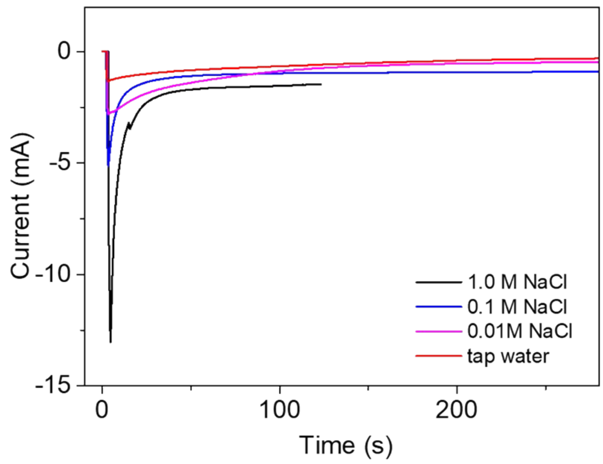

Fig. S8 Output current of the Zn-air cell when PANI/CNT and Zn foil were put into NaCl solutions with different concentration and tap water. The immersed area of each electrode is 1.5×0.5 cm^2. The average V_{OC} of Zn-air cell are 1.21 V, 1.15V, 1.10 V and 1.08 V using 1.0 M NaCl, 0.1 M NaCl, 0.01 M NaCl and tap water as electrolyte respectively.

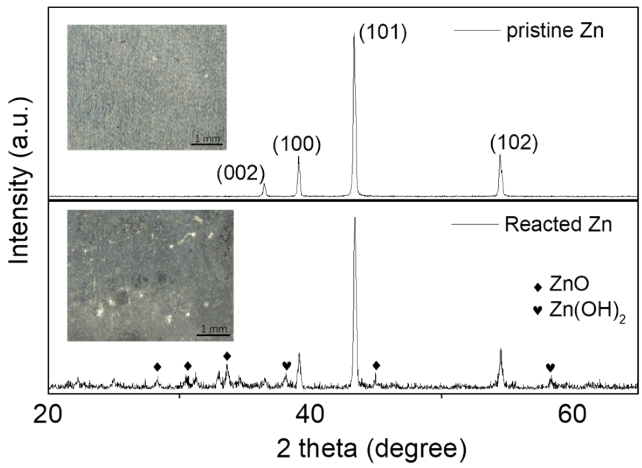

Fig. S9 XRD pattern of (a) pristine Zn foil and (b) reacted Zn foil. The inset shows the optical images of the surface of Zn foil before and after reaction.

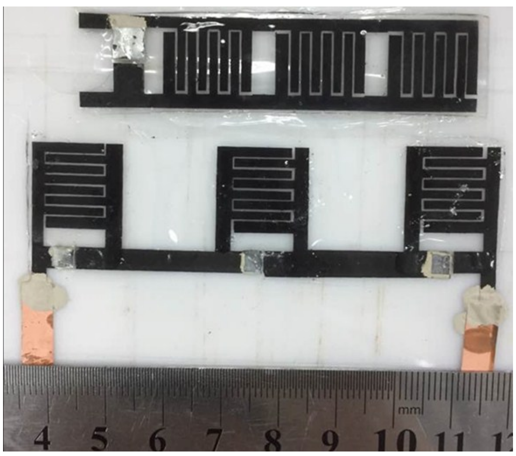

Fig. S10 Digital photographs of self-charging SCs combined in parallel and series.

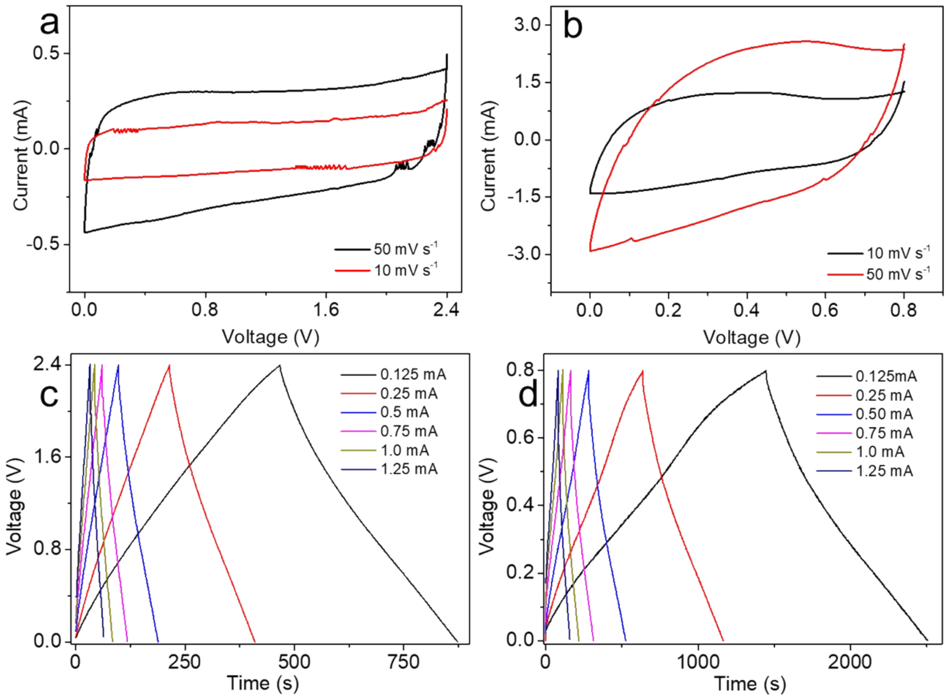

Fig. S11 CV curves of three SCs assembled in series (a) and parallel (b) at different scan rates. GCD curves of three SCs assembled in series (c) and parallel (d) at different currents.

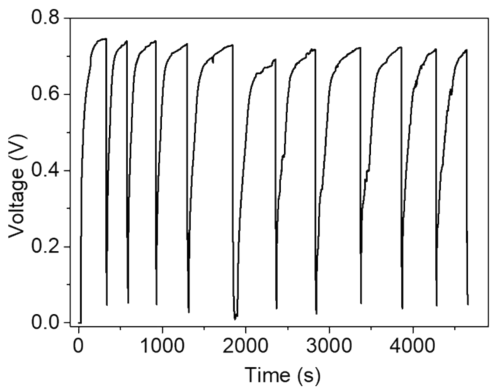

Fig. S12 Verification of $CaCl_2$ solution as electrolyte for Zn-air cell/SC hybrid device. The device was charged in a touch mode that filter paper and Zn foil (inset in Fig. 4d) were pressed together by a swap soaked with 1 M $CaCl_2$ solution. The device was discharged by powering a small motor.

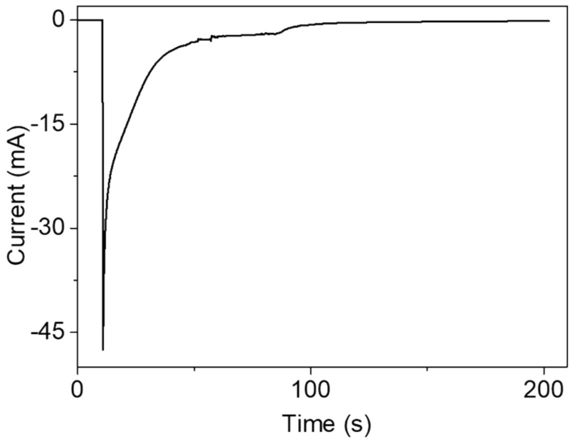

Fig. S13 Short-circuit current of moisture-charging SC after being charged (RH=88%).

Table S1 Comparison of SC charged *in situ* and externally

Power source	Current (mA)	Charging range (V)	Time (s)
	0.125	0.025-0.8	484
external	0.5	0.073-0.8	88
	1.25	0.167-0.8	26
integrated	/	0.012-0.8/1.0	373/968

Chemical Reactions for Zn-air cell

Anode: $Zn\text{-}2e^- \rightarrow Zn^{2+}$ $Zn^{2+} + 2OH^- \rightarrow Zn(OH)_2 \rightarrow ZnO + H_2O$ (S1)

Cathode： $O_2 + 4e^{-+} \rightarrow 4(OH)^-$ (S2)

Overall: $2Zn + O_2 \rightarrow 2ZnO$ (S3)

The nominal cell voltage is 1.65 V but it is 1.35-1.45 V in practical cells.[1-3]

Equations for calculating electrochemical performances

$$C_m = \frac{I \cdot \Delta t}{S \cdot \Delta U} \quad \text{(S4)}$$

$$P = V_{OC} \cdot I_{SC} / 4 \quad \text{(S5)}$$

$$E = C \cdot V_{OC}{}^2 / 2 = V_{OC} \cdot Q / 2 \quad \text{(S6)}$$

$$Q = \int I_{SC} \cdot dt \quad \text{(S7)}$$

Where C_m is the areal capacitance, I is the discharging current, Δt is the discharging time, S is the effective area and ΔU is the potential range during the discharging process. P is the power of SPSC when powering a motor. P is the maximum power density of self-charging SC and V_{OC} and I_{SC} are the open circuit voltage and short circuit current of SC after being in situ charged. E is the energy stored in SC and Q is the charge stored in SC.

References:

1. J. Goldstein, I. Brown and B. Koretz, *J. Power Sources*, 1999, **80**, 171-179.
2. D. U. Lee, P. Xu, Z. P. Cano, A. G. Kashkooli, M. G. Park and Z. Chen, *J. Mater. Chem. A*, 2016, **4**, 7107-7134.
3. J. Fu, R. Liang, G. Liu, A. Yu, Z. Bai, L. Yang and Z. Chen, *Adv. Mater.*, 2018, 1805230.