## Electronic Supplementary Information

# Towards the upper bound of electrochemical performance of ACNT@polyaniline arrays as supercapacitors

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## Specific capacitance calculations of the PANI/ACNTs composites

The capacitances of the composites (CNT+PANI) and PANI were estimated from the CV curves by Equation (S1):[1]

$$C_{sp,\,comp} = \frac{\int_0^t i(t)dt}{m \times (U_2 - U_1)} \quad \text{(S1)}$$

where $C_{sp,comp}$ and $C_{sp,PANI}$ are the capacitance of the composite, the PANI and the ACNT, i is the current (A), $U_2$ and $U_1$ are the switching potentials (0.7 V and -0.2 V vs Ag/AgCl, respectively) and m is the mass of the composites.

The specific capacitance is produced by the pseudocapacitance and double layer capacitance of the PANI shell of the composite. Therefore, the contribution to the specific capacitance from only the PANI phase is calculated by Equation (S2)

$$C_{sp,\,PANI} = \frac{C_{sp,\,comp}}{PANI\%} \quad \text{(S2)}$$

where $C_{sp,PANI}$ is the specific capacitance based on the mass of the PANI and the PANI% is the weight percentage of the PANI in the composites.

## Characterizations of the supercapacitors

The measured cell capacitance (C), the specific capacitance of the entire cell ($C_{cell,sp}$) and the specific capacitance ($C_{sp}$) of the composite material on a single electrode was expressed by Eq. (S3, S4), where i is the current (A), t is the discharge time, U is the voltage (V) and $M_{cell}$ is the mass of the entire device, including the electrolyte and separator, and M is the total mass of the PANI/ACNTs composite materials on the cathode and anode.[2]

$$C = \frac{i \times t}{U} \quad \text{(S3)}$$

$$C_{cell,\,sp} = \frac{C}{M_{cell}} \text{ and } C_{sp} = \frac{4C}{M} \quad \text{(S4)}$$

The ESR, average specific energy ($E_{sp}$), average specific power ($P_{sp}$) and maximum specific power ($P_{max}$) based on the weight of the SC cells, including the wetted filtration paper, and the PANI/ACNTs composite material were calculated by Eq. (S5-8):[2, 3]

$$ESR = \frac{\Delta U}{2 \times i} \quad \text{(S5)}$$

$$E_{cell,\,sp} = \frac{C \times U^2}{2 \times M_{cell}} \text{ and } E_{sp} = \frac{C \times U^2}{2 \times M} \quad \text{(S6)}$$

$$P_{sp} = \frac{E_{sp}}{t} \quad \text{(S7)}$$

$$P_{max} = \frac{U^2}{4 \times ESR \times M} \quad \text{(S8)}$$

where ΔU is the iR drop shown in Figure 6c.

## References

1. H. Li, J. Wang, Q. Chu, Z. Wang, F. Zhang and S. Wang, *Journal of Power Sources*, 2009, **190**, 578-586.
2. C. Meng, C. Liu, L. Chen, C. Hu and S. Fan, *Nano Letters*, 2010, **10**, 4025-4031.
3. C. G. Liu, Z. N. Yu, D. Neff, A. Zhamu and B. Z. Jang, *Nano Letters*, 2010, **10**, 4863-4868.