

**Multiscale honeycomb structured activated carbon from nitrogen containing
mandarin peel: High-performance supercapacitors with extreme cycling
stability**

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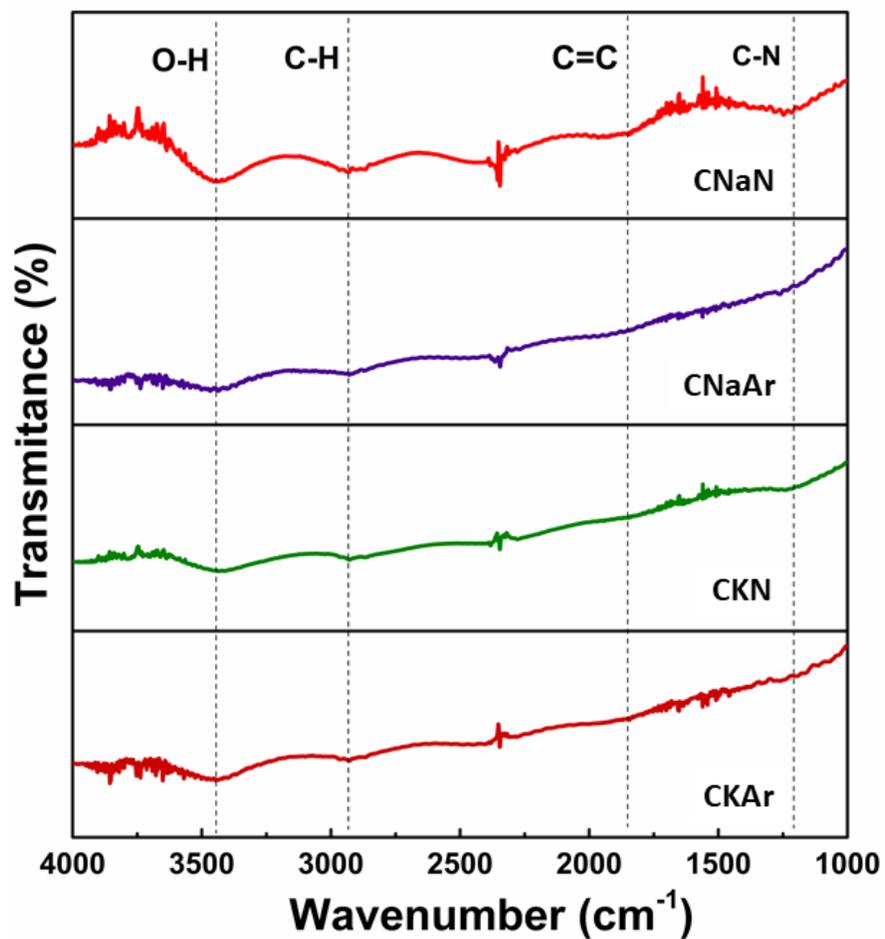


Figure S1. FTIR spectra of activated carbon samples.

The chemical bonding structures have been analyzed using the FTIR spectrum and given in figure S1. Four main peaks were identified and have been indexed corresponding to O-H, C-H, C=C and C-N bonds.

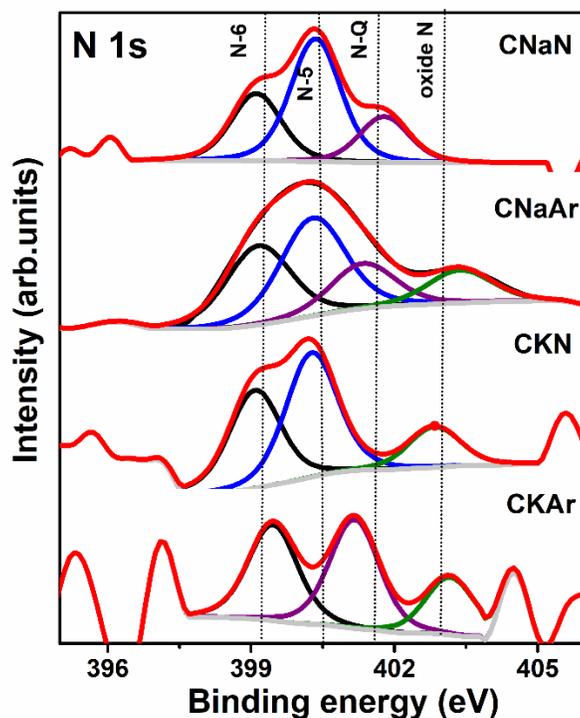


Figure S2. N1s Core level XPS spectra of as-prepared samples

N1s spectra show the presence of nitrogen in the carbon rings in all the samples prepared from mandarin peels confirm the natural inheritance and their respective elemental atomic ratio is given in Table S1.

Table S1. The atomic ratio of elements in as prepared carbon samples

Sample name	C (%)	O (%)	Na (%)	N (%)
CKAr	91.66	8.03	-	0.31
CKN	88.15	11.36	-	0.49
CNaAr	87	9.54	2.6	0.77
CNaN	86.72	10.11	2.71	0.44

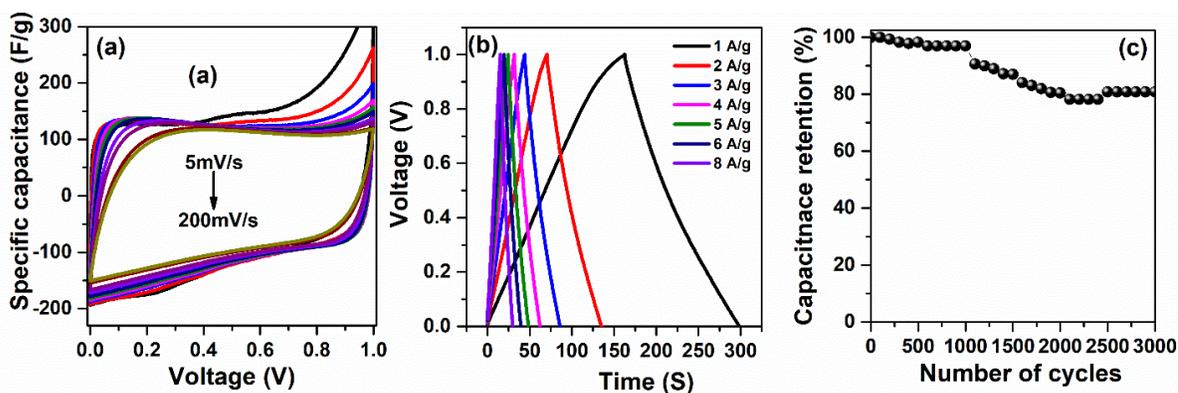


Figure S3. (a) CV curves at various scan rates, (b) GCD profiles at a various current rate and (c) cycling stability of CKAr

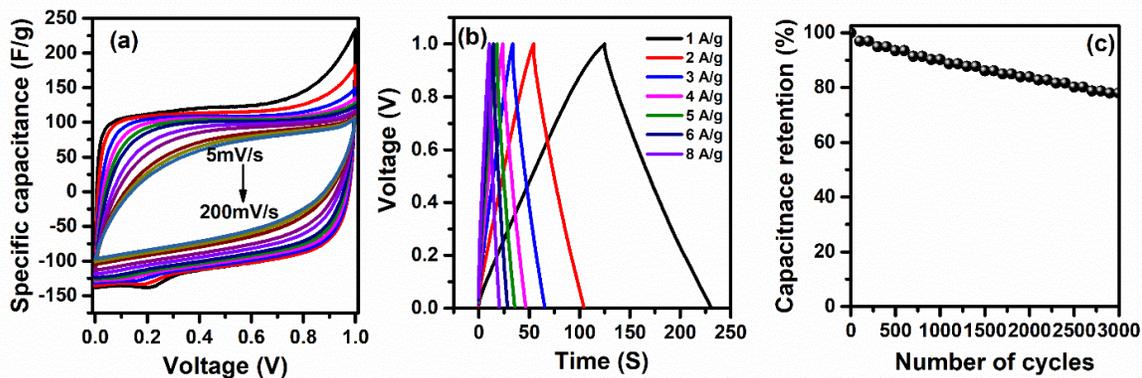


Figure S4. (a) CV curves at various scan rates, (b) GCD profiles at a various current rate and (c) cycling stability of CNAr

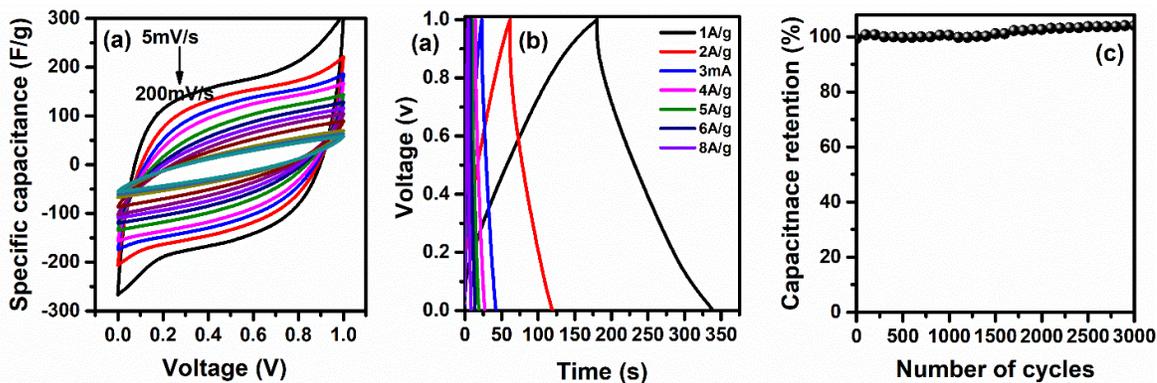


Figure S5. (a) CV curves at various scan rates, (b) GCD profiles at a various current rate and (c) cycling stability of CKAr samples CNAN

Symmetrical supercapacitor performances have been studied and are given in Figures S3, S4, S5 for CKAr, CNaAr and CNaN respectively. The samples heat treated in the nitrogen atmosphere show better specific capacitance. Samples activated in NaOH show enhanced intercalation capacitance due to sodium intercalation in the carbon network. It has a higher specific capacitance and a good capacitance retention after 3000 cycles.

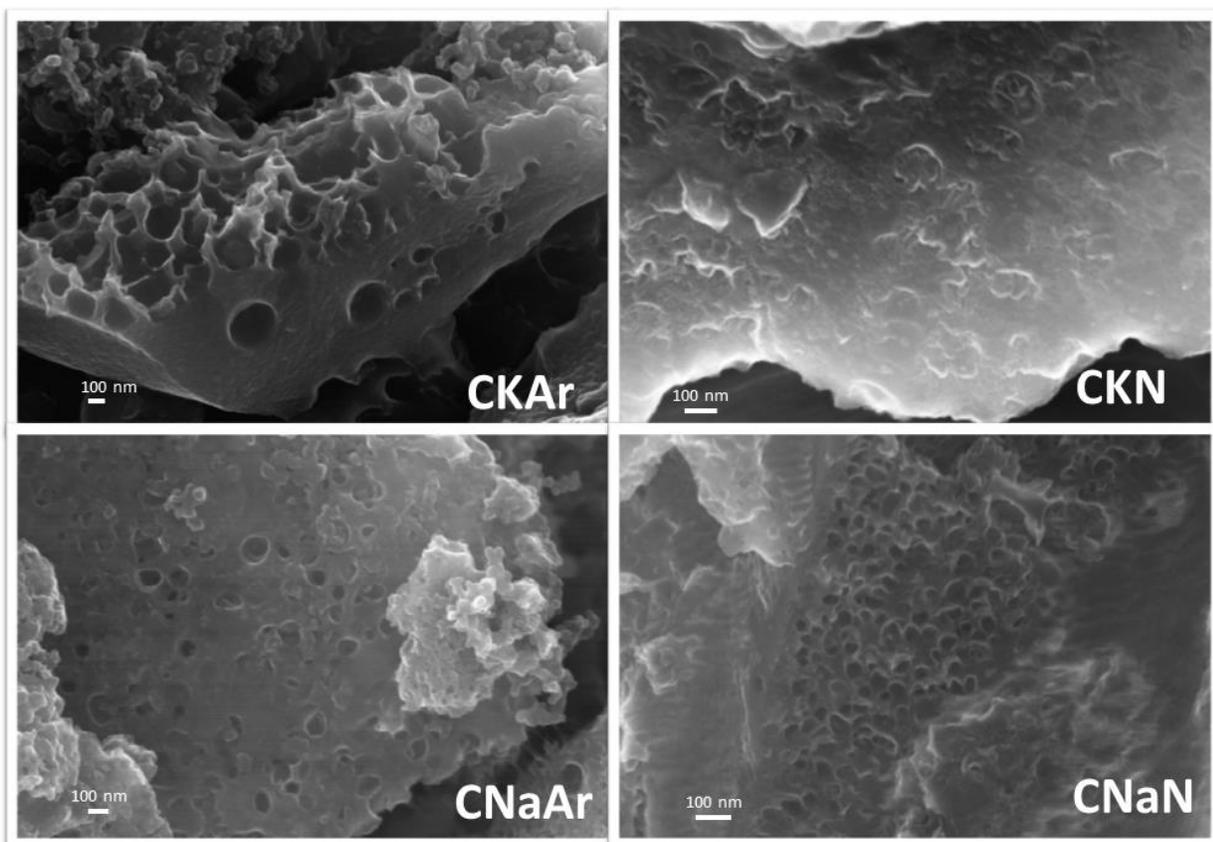


Figure S6. FE-SEM images of samples collected from cycled supercapacitor devices

Figure S6 show the FE-SEM images of charging/discharging cycled carbon samples. It reveals that the honeycomb structured are well maintained even after a long-term stability test due to the potassium metal intercalation during cycling. It confirms that the honeycomb structured carbon would be a better option in practical supercapacitor application with a longer lifespan.

Electrochemical calculations for symmetric supercapacitors.

- 1) The Gravimetric specific capacitance for a single electrode was calculated from the Galvanostatic charge-discharge profile using the formula (1)

$$C_{sp} = \frac{2I}{(dV/dt)m} \quad (1)$$

Where I is the current (A), dV/dt is the slope of the discharge curve without ohmic loss and m is the mass in gram of the active material in an electrode

The Energy density (E , Whkg⁻¹) and power density (P , Wkg⁻¹) was calculated from equations (2) and (3) given below

$$E = \frac{1}{2} \cdot C_{sp} \cdot V^2 \cdot \frac{1}{4} \cdot \frac{1}{3.6} \quad (2)$$

$$P = \frac{E}{t} \cdot 3600 \quad (3)$$

Where V is the cell voltage after the ohmic drop (V) and t is the discharge time (h).

Table S2. Comparison of symmetrical supercapacitor performance of various carbon materials

Carbon materials	Electrolyte	Voltage (V)	Specific capacitance	Energy and power density (Whkg ⁻¹ and Wkg ⁻¹)	Mass (mg/cm ²) and active area (cm ²)	Ref.
Auricularia biomass derived carbon	1M H ₂ SO ₄	1 V	256 F/g @ 1 A/g	8.9 and 250	0.7 and 1.13	¹
Soybean derived carbon	1M H ₂ SO ₄	1.1 V	261 F/g @ 0.2 A/g	12 and 2000	6and 0.785	²
Carbon nanosheets from sodium gluconate	1M H ₂ SO ₄	1 V	140 F/g @ 150 A/g	26 and 15000	5	³
nitrogen-doped porous carbon derived from lecithin	1 M KOH	1 V	156 F/g @ 0.5 A/g	18 and 500	1.15 and 1.5	⁴
Porous Carbon Derived from PolyHIPE	1M H ₂ SO ₄	1 V	221 F/g @ 0.5A/g	32.7 and 373.2	4 and 0.5	⁵
Recycled jute to carbon	3 M KOH	1 V	185 F/g @ 0.5 A/g	-	4 and 1	⁶
Hierarchical Porous Carbon from Lignin-Derived Byproducts	6 M KOH	1 V	312 F/g @ 1 A/g	8.8 and 1300	5	⁷
Interconnected Phosphorus and Nitrogen Co-doped Porous	6 M KOH	1 V	265 F/g @ 0.5 A/g	9 and 100	4.15	⁸
Multiscale Pore Network Carbon	3 M KOH	1 V	376.5 F/g @ 1 A/g	10.4 and 200	2.5 and 1	⁹
Soybean Root-Derived Hierarchical Porous Carbon	6 M KOH	1 V	276 F/g @ 0.5 A/g	10 and 120	2 and 1	¹⁰
Multiscale honeycomb structured activated carbon	3 M KOH	1 V	348 F/g @ 1 A/g	10.92 and 240	2 and 1	This work

References:

1. Z. Zhu, H. Jiang, S. Guo, Q. Cheng, Y. Hu and C. Li, *Sci Rep*, 2015, **5**, 15936.
2. G. A. Ferrero, A. B. Fuertes and M. Sevilla, *Sci Rep*, 2015, **5**, 16618.
3. A. B. Fuertes and M. Sevilla, *ACS Appl Mater Interfaces*, 2015, **7**, 4344-4353.
4. M. Demir, S. K. Saraswat and R. B. Gupta, *RSC Adv.*, 2017, **7**, 42430-42442.
5. W. Hu, F. Xie, Y. Li, Z. Wu, K. Tian, M. Wang, L. Pan and L. Li, *Langmuir*, 2017, **33**, 13364-13375.
6. C. Zequine, C. K. Ranaweera, Z. Wang, P. R. Dvornic, P. K. Kahol, S. Singh, P. Tripathi, O. N. Srivastava, S. Singh, B. K. Gupta, G. Gupta and R. K. Gupta, *Sci Rep*, 2017, **7**, 1174.
7. L. Zhang, T. You, T. Zhou, X. Zhou and F. Xu, *ACS Appl Mater Interfaces*, 2016, **8**, 13918-13925.
8. J. Jin, X. Qiao, F. Zhou, Z. S. Wu, L. Cui and H. Fan, *ACS Appl Mater Interfaces*, 2017, **9**, 17317-17325.
9. F. Zhang, T. Liu, M. Li, M. Yu, Y. Luo, Y. Tong and Y. Li, *Nano Lett*, 2017, **17**, 3097-3104.
10. N. Guo, M. Li, Y. Wang, X. Sun, F. Wang and R. Yang, *ACS Appl Mater Interfaces*, 2016, **8**, 33626-33634.