

Electronic supplementary information (ESI)

**Highly Nitrogen-Doped Carbon Capsules: Scalable Preparation
and High-Performance Applications in Fuel Cells and Lithium
Ion Batteries**

Chuangang Hu, Ying Xiao, Yang Zhao, Nan Chen, Zhipan Zhang, Minhua Cao,

Liangti Qu*

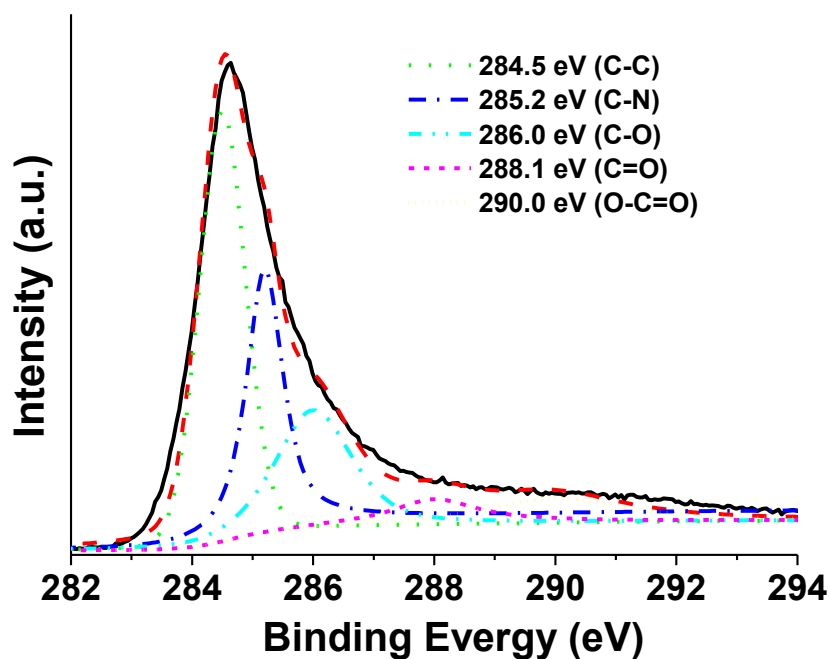


Fig. S1 High-resolution C 1s peaks of hN-CCs.

The emergence of the peak at 285.2 eV corresponds to C-N coordination and the peak at 284.5 eV is the C-C coordination (Fig. S1), which confirms the successful incorporation of N within carbon backbone.

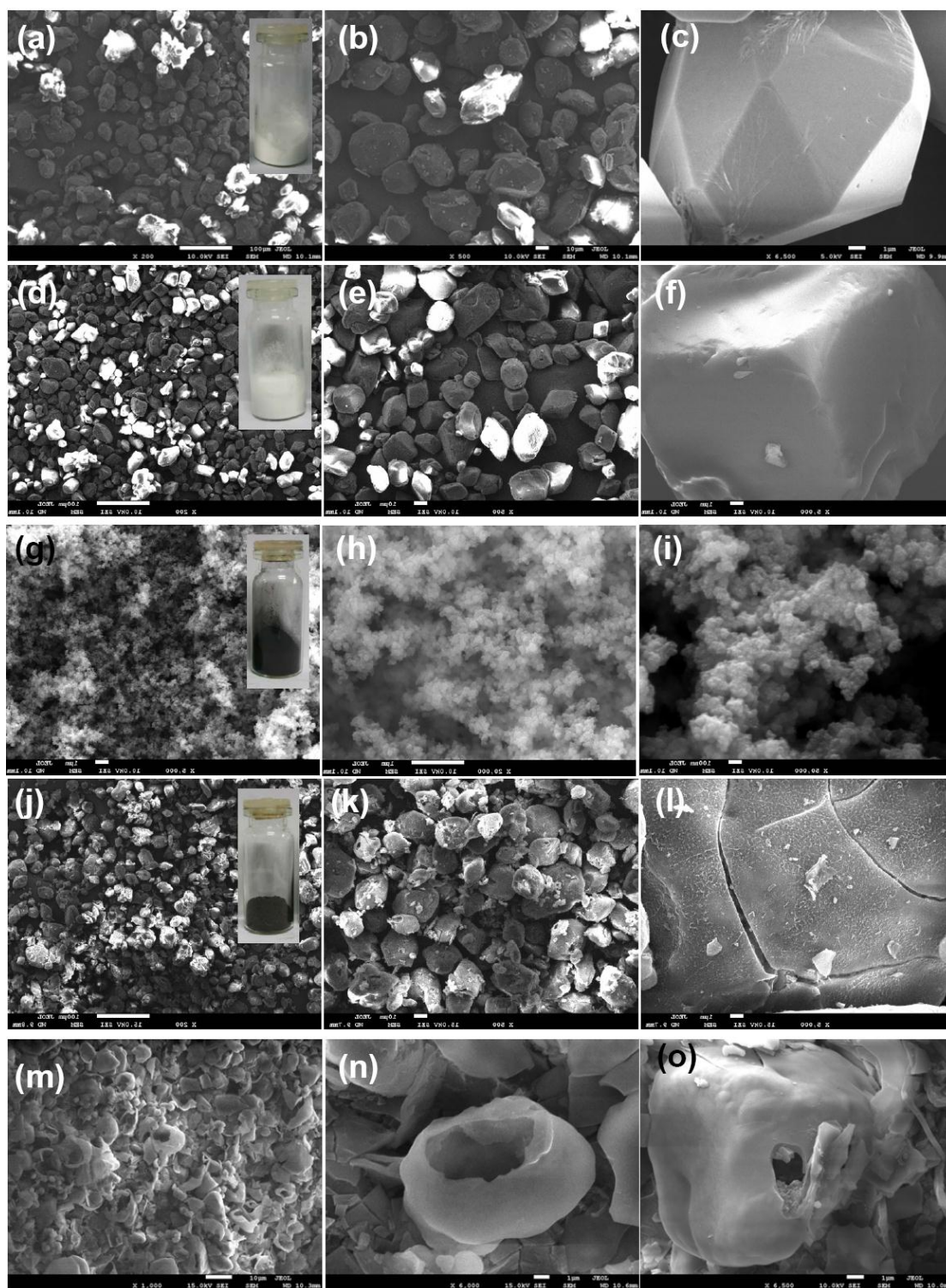


Fig. S2 (a–c) SEM images of the melamine particles at different magnifications. (d–f) SEM images of melamine particles after solvothermal treatment at 140 °C. (g–i) SEM images of glyoxal treated solvothermally. (j–l) SEM images melamine particles after solvothermal treatment with glyoxal at 140 °C. (m–o) SEM images of pre-capsules. Scale bars: a,d,j, 100 μ m; b,e,k,m, 10 μ m; c,f,g,h,l,n,o, 1 μ m; i, 100nm.

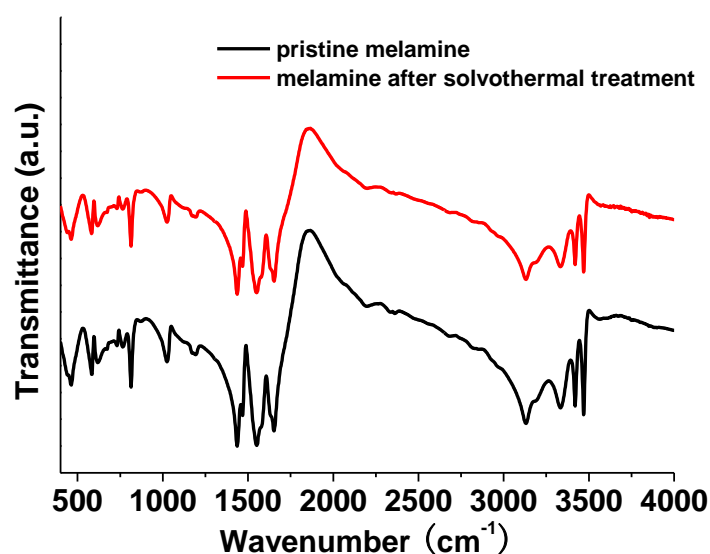


Fig. S3 FT-IR spectra of the melamine before and after solvothermal treatment at 140 °C for 4h under the same synthesis procedure as typical experiment.

As revealed in Figure S2, the melamine particles before (Fig. S2a–c) and after (Fig. S2d–f) solvothermal treatment at 140 °C for 4h have similar surface character and FT-IR spectroscopy (Fig. S3), indicating no change occurred during this process. However, when only the glyoxal was employed as precursors, small particles were yield during the solvothermal process (Fig. S2g–i). Once the mixture of glyoxal and melamine was treated solvothermally, melamine still keeps the particle shape but with a surface coating layers (Fig. 4a&b, Fig. S2j–l), which could be attributed to the deposition of glyoxal on melamine particles. After washing away the excess melamine with hot water, capsules were obtained (Fig. S2m–o). Further FT-IR investigation reveals the possible formation of amide compounds via the reaction between glyoxal and melamine (Fig. 4d).

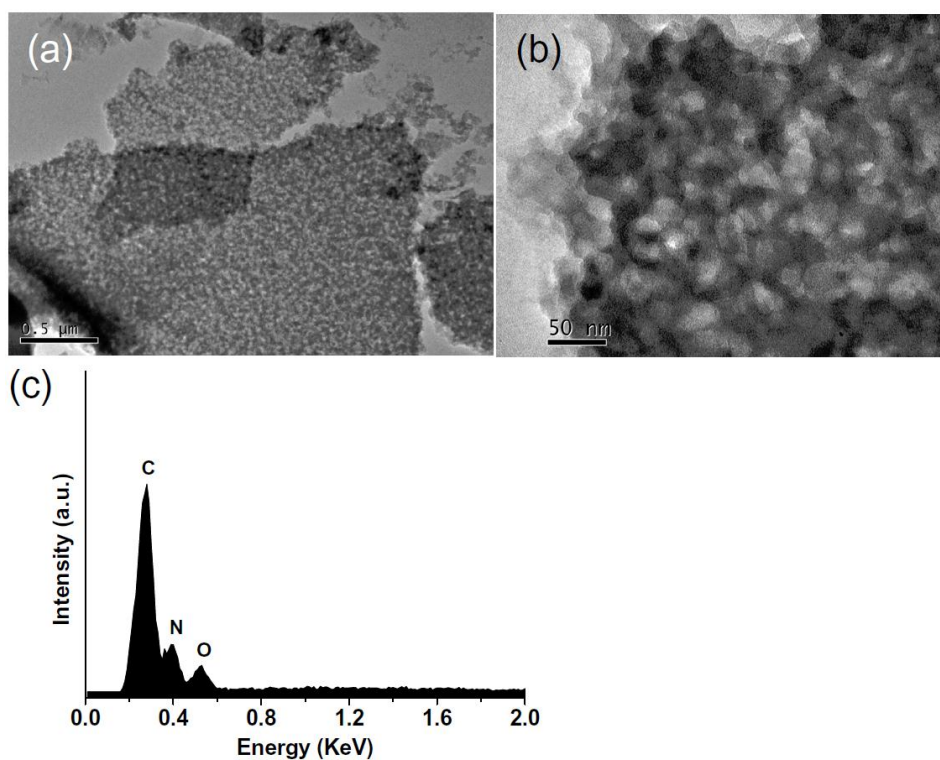


Fig. S4 (a,b) TEM images of the obtained pre-capsules at different magnifications. (c) The corresponding energy dispersive spectroscopy (EDS) spectroscopy.

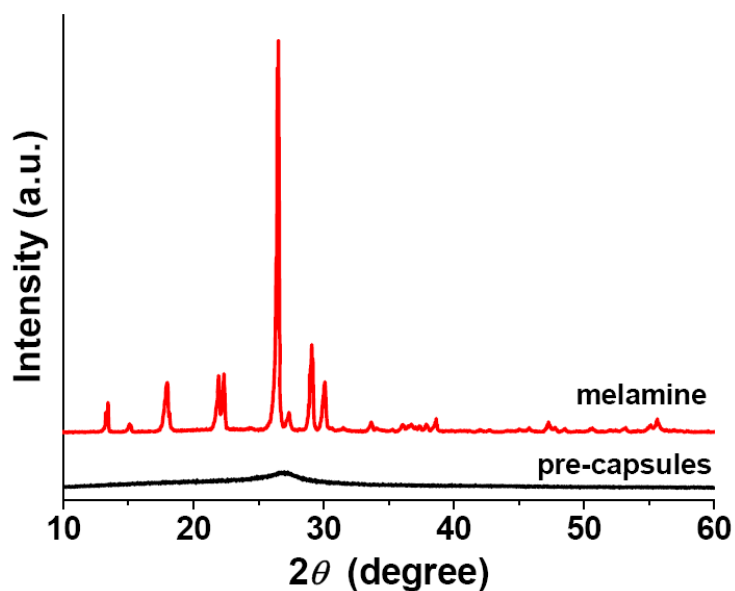


Fig. S5 XRD pattern of melamine after solvothermal treatment at 140 °C and pre-capsules.

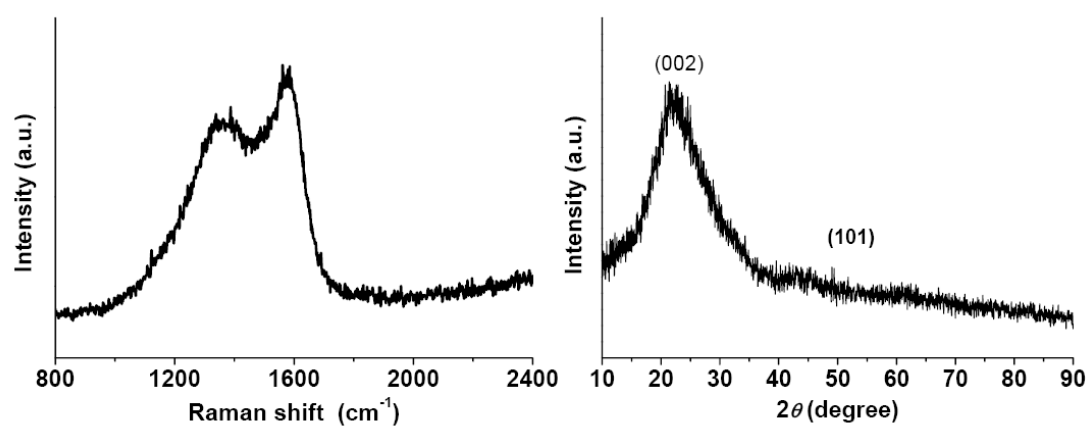


Fig. S6 (a) Raman spectrum, (b) XRD pattern of the CG.

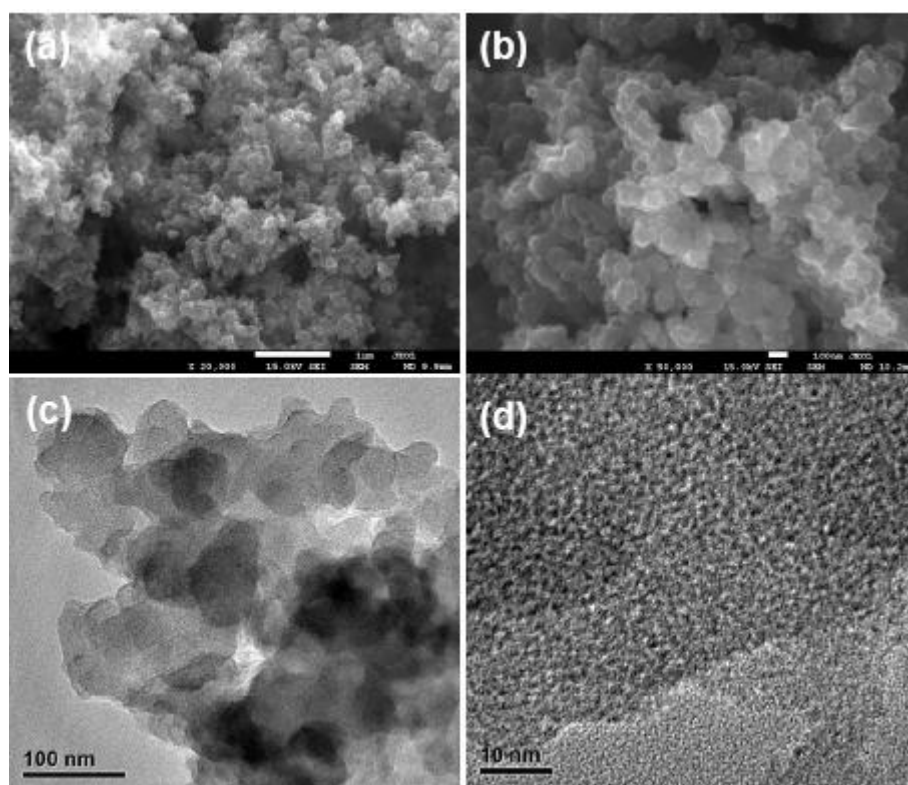


Fig. S7 (a, b) SEM images of CG at different magnifications, (c, d) TEM and HR-TEM images of CG.

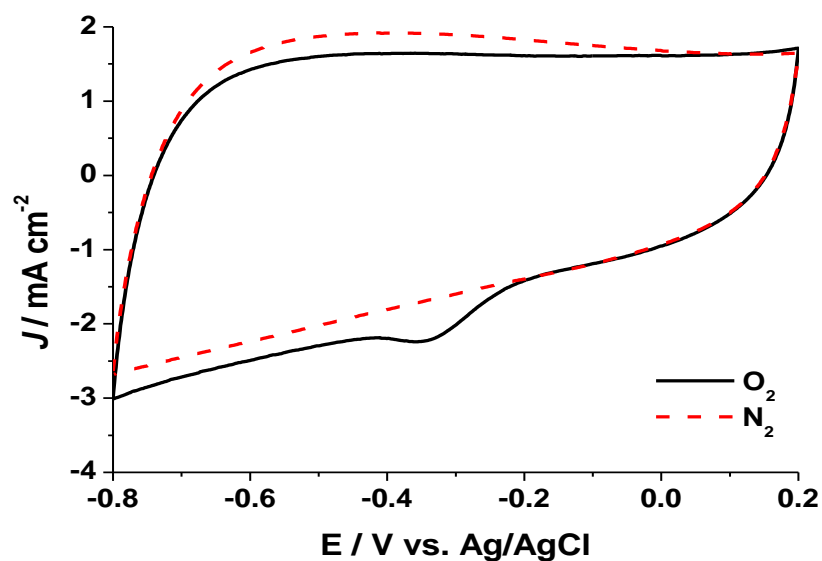


Fig. S8 CV performed for CG in O₂ and N₂-saturated 0.1 M KOH solution at a scan rate of 50 mV s⁻¹.

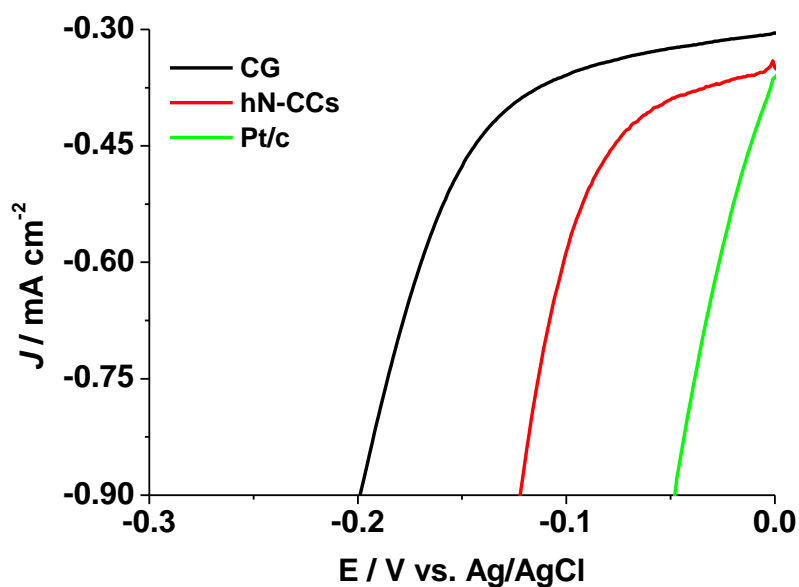


Fig. S9 The enlargement of the curves in Fig. 5b within the range of - 0.0 V to - 0.3 V.

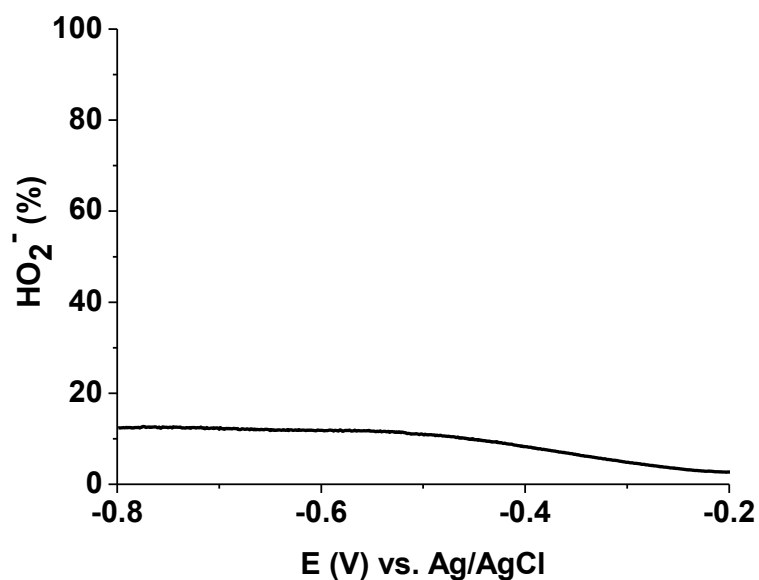


Fig. S10 The peroxide percentage (%HO₂⁻) according to linear sweep voltammograms (LSVs) obtained for hN-CCs at 1600 rpm and 5 mV s⁻¹.

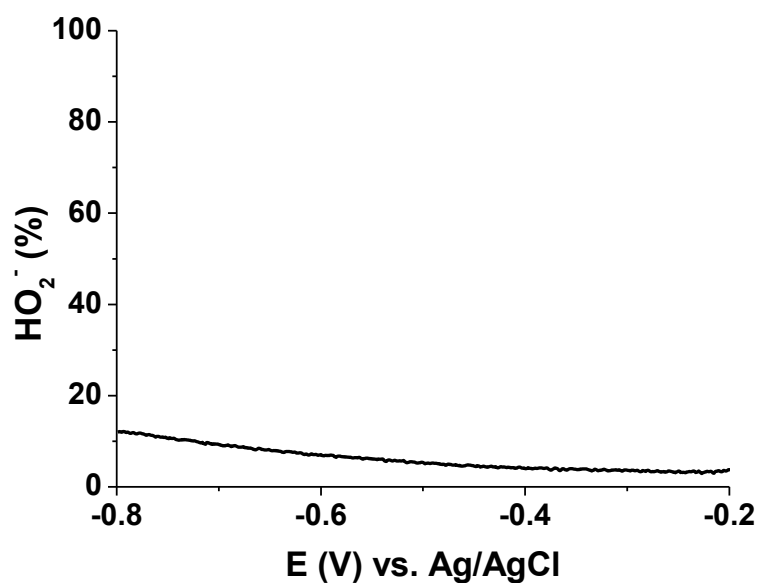


Fig. S11 The peroxide percentage (%HO₂⁻) according to linear sweep voltammograms (LSVs) obtained for E-TEK 20% Pt/C at 1600 rpm and 5 mV s⁻¹.

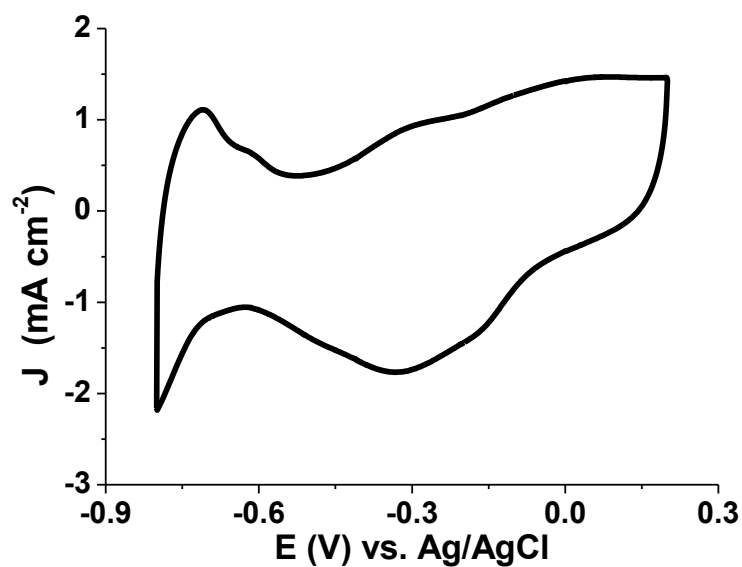


Fig. S12 The CV of Pt/C catalyst in O₂-saturated 0.1 M KOH solution at a scan rate of 50 mV s⁻¹.

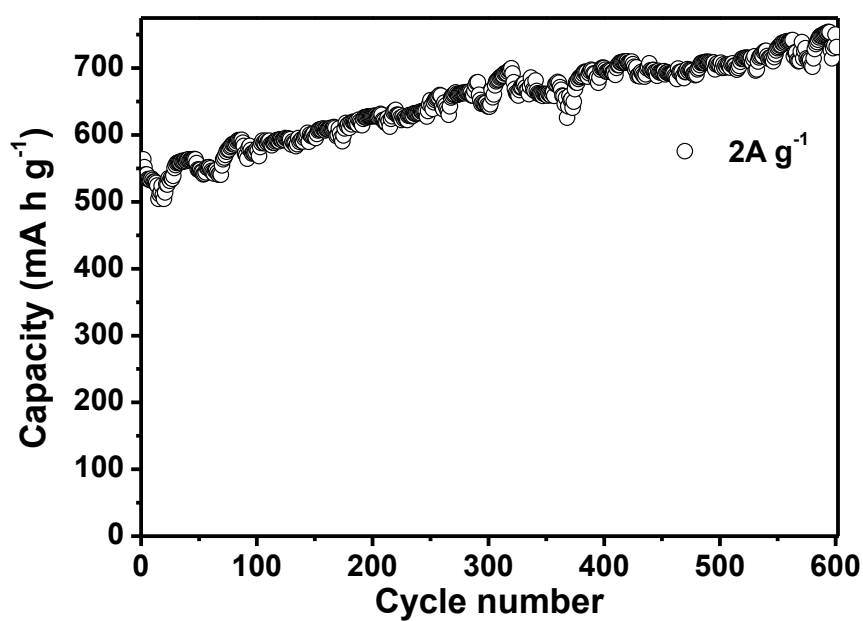


Fig. S13 The cycle performance of the hN-CCs at the current density of 2 A g⁻¹.

Table S1. Comparison of ORR electrocatalytic performance of hN-CCs with some carbon-based ORR catalysts in alkaline solution reported previously.

Samples	N/C atomic ratio	Technique	Onset potential (V)	References
hN-CCs	ca. 13%	CV, RDE	-0.06	This work
N-doped graphene	ca. 8 %	CV, RDE	-0.1	1
Nanoporous N doped carbon	ca. 3 %	RDE	-0.15	2
N-doped graphene	ca. 4 %	CV, RDE	-0.2	3
Graphitic C ₃ N ₄ /carbon composite	unknown	CV, RDE	-0.18	4
N-doped carbon nanotubes	ca. 3.3 %	RDE	-0.15	5
N-doped carbon nanotubes	ca. 3 %	CV, RDE	-0.20	6
N-doped graphene supported Fe ₃ O ₄ nanoparticles	ca. 3.5 %	CV, RDE	-0.19	7

References and notes

- S1 Z. Lin, M. Song, Y. Ding, Y. Liu, M. Liu and C. Wong, *Phys. Chem. Chem. Phys.*, 2012, **14**, 3381.
- S2 Y. Q. Sun, C. Li and G. Q. Shi, *J. Mater. Chem.*, 2012, **22**, 12810.
- S3 L. T. Qu, Y. Liu, J. B. Baek and L. M. Dai, *ACS Nano*, 2010, **4**, 1321.
- S4 J. Liang, Y. Zheng, J. Chen, J. Liu, D. Hulicova-Jurcakova, M. Jaroniec and S. Z. Qiao, *Angew. Chem. Int. Ed.*, 2012, **51**, 1.
- S5 Z. Chen, D. Higgins and Z. W. Chen, *Carbon*, 2010, **48**, 3057.
- S6 N. Alexeyeva, E. Shulga, V. Kisand, I. Kink and K. Tammeveski, *J. Electroanal. Chem.*, 2010, **648**, 169.
- S7 Z. S. Wu, S. B. Yang, Y. Sun, K. Parvez, X. L. Feng and K. Müllen, *J. Am. Chem. Soc.*, 2012, **134**, 9082.