Electronic supplementary information (ESI)

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

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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 solvothermral treatment at 140 °C. (g–i) SEM images of glyoxal treated solvothermally. (j–l) SEM images melamine particles after solvothermral treatment with glyoxal at 140 °C. (m–o) SEM images of pre-capsules. Scale bars: a,d,j, 100 μm; b,e,k,m, 10 μm; c,f,g,h,l,n,o, 1 μm; 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 O$_2$ and N$_2$-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 (%$\text{HO}_2^-$) according to linear sweep voltammograms (LSVs) obtained for hN-CCs at 1600 rpm and 5 mV s$^{-1}$.

**Fig. S11** The peroxide percentage (%$\text{HO}_2^-$) 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 O$_2$-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.

<table>
<thead>
<tr>
<th>Samples</th>
<th>N/C atomic ratio</th>
<th>Technique</th>
<th>Onset potential (V)</th>
<th>References</th>
</tr>
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<tbody>
<tr>
<td>hN-CCs</td>
<td>ca. 13%</td>
<td>CV, RDE</td>
<td>-0.06</td>
<td>This work</td>
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<td>N-doped graphene</td>
<td>ca. 8 %</td>
<td>CV, RDE</td>
<td>-0.1</td>
<td>1</td>
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<td>Nanoporous N doped carbon</td>
<td>ca. 3 %</td>
<td>RDE</td>
<td>-0.15</td>
<td>2</td>
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<tr>
<td>N-doped graphene</td>
<td>ca. 4 %</td>
<td>CV, RDE</td>
<td>-0.2</td>
<td>3</td>
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<tr>
<td>Graphitic C$_3$N$_4$/carbon composite</td>
<td>unknown</td>
<td>CV, RDE</td>
<td>-0.18</td>
<td>4</td>
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<tr>
<td>N-doped carbon nanotubes</td>
<td>ca. 3.3 %</td>
<td>RDE</td>
<td>-0.15</td>
<td>5</td>
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<tr>
<td>N-doped carbon nanotubes</td>
<td>ca. 3 %</td>
<td>CV, RDE</td>
<td>-0.20</td>
<td>6</td>
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<tr>
<td>N-doped graphene supported Fe$_3$O$_4$ nanoparticles</td>
<td>ca. 3.5 %</td>
<td>CV, RDE</td>
<td>-0.19</td>
<td>7</td>
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References and notes


