Supporting Information

Pyridinic N Doped Graphene: Synthesis, Electronic Structure, and Electrocatalytic Property

Zhiqiang Luo\textsuperscript{1,2}, Sanhua Lim\textsuperscript{2}, Zhiqun Tian\textsuperscript{2}, Jingzhi Shang\textsuperscript{1}, Linfei Lai\textsuperscript{1,2}, Brian MacDonald\textsuperscript{3}, Chao Fu\textsuperscript{1}, Zexiang Shen\textsuperscript{1}, Ting Yu\textsuperscript{1,4*}, and Jianyi Lin\textsuperscript{2*}

1.Division of Physics and Applied Physics, School of Physical and Mathematical Sciences, Nanyang Technological University, Singapore, 637371
2.Applied Catalysis, Institute of Chemical and Engineering Sciences, Singapore, 627833
4.Department of Physics, Faculty of Science, National University of Singapore, Singapore, 117542

Calculation of electron transfer numbers in ORR:

The electron transfer numbers were calculated using Koutecky-Levich equation:\textsuperscript{S1}

\[
i^-1 = i_k^{-1} + i_{lev}^{-1};
\]

\[
i_k = nFkO_2C_{O_2}\Gamma_{catalyst};
\]

\[
i_{lev} = 0.62nFC_{O_2}DO_2^{2/3}v^{-1/6}w^{1/2};
\]

\[
i^-1 = A + 1/(Bw^{1/2});
\]

\[
B = 0.62nFC_{O_2}DO_2^{2/3}v^{-1/6},
\]

where, \(i\) is the total current density; \(i_k\) is the kinetic current density; \(i_{lev}\) is the levich current density; \(n\) is the electron transfer numbers; \(F\) is the Faraday constant (96500 C mol\(^{-1}\)); \(\Gamma_{catalyst}\) is the catalyst loading; \(kO_2\) is the rate constant for oxygen reduction; \(C_{O_2}\) is concentration of O\(_2\) dissolved in 0.1 M KOH solution (1.2 x 10\(^{-6}\) mol cm\(^{-3}\)); \(DO_2\) is the diffusion coefficient of O\(_2\) in 0.1 M KOH solution (1.9x10\(^{-5}\) cm\(^{2}\) s\(^{-1}\)); \(w\) is the rotation rate (rad s\(^{-1}\)) and \(v\) is the kinetic viscosity of the water (0.01 cm\(^2\) s\(^{-1}\)). The slope of the plot of reciprocal current \(i^-1\) versus the reciprocal square root of rotation rate \((w^{-1/2})\) gives B values. The value of \(n\) can be calculated from B by using above parameters.\textsuperscript{S1}
Figure S1. Optical image of single layer graphene transferred onto a SiO_2/Si substrate. The inset shows a photograph of the single layer graphene transferred onto a SiO_2/Si substrate.
Figure S2. Raman imaging of CN$_x$ graphene synthesized with the NH$_3$/He flow rate of 12 sccm (GN12). (a) D band intensity, (b) 2D band intensity, (c) G band height, (c) D’ band height.
Figure S3. Raman spectra of the CNₓ graphene synthesized with the NH₃/He flow rate of 12 sccm (GN12). Spectrum (a) and spectrum (b) corresponds to the A and B spots in Figure S1, respectively.

Figure S4. Overlay imaging of CN⁻ and C₂⁻ mass spectra images shown in Figure 3. N rich area is demonstrated with the red color.
Figure S5. TOF-SIMS mass spectra in linear scale from the A and B spots indicated in Figure S4.
Figure S6. (a) XPS survey scan of CNₓ graphene synthesized with the NH₃/He flow rate of 6 sccm (GN6), (b) N1s core-level spectrum.

Reference