

Electronic Supplementary Information (ESI) for:

**Wet-Chemical Nitrogen-Doping of Graphene Nanoplatelets as
Electrocatalysts for Oxygen Reduction Reaction**

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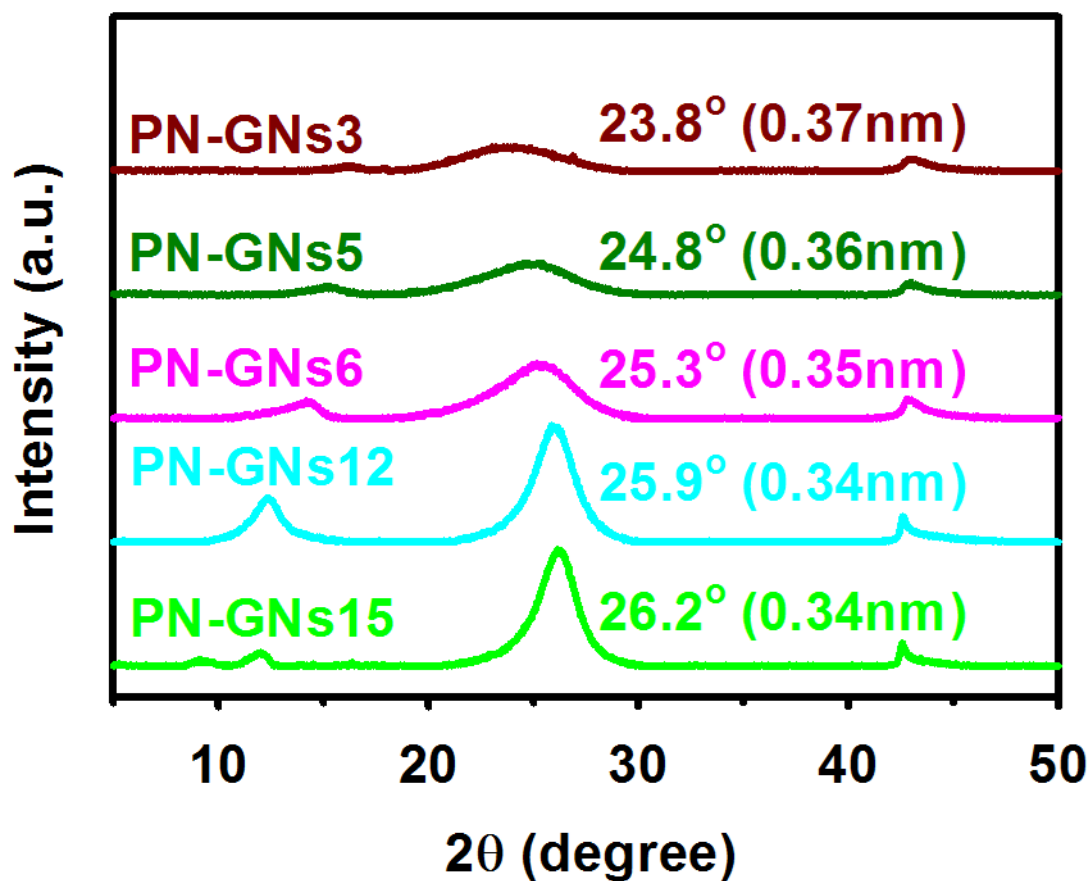


Figure S1. Magnified XRD diffraction patterns from the rectangle area in Figure 2d with the numbers of *d*-spacing in nanometer (nm) of PN-GNs3, PN-GNs5, PN-GNs6, PN-GNs12 and PN-GNs15.

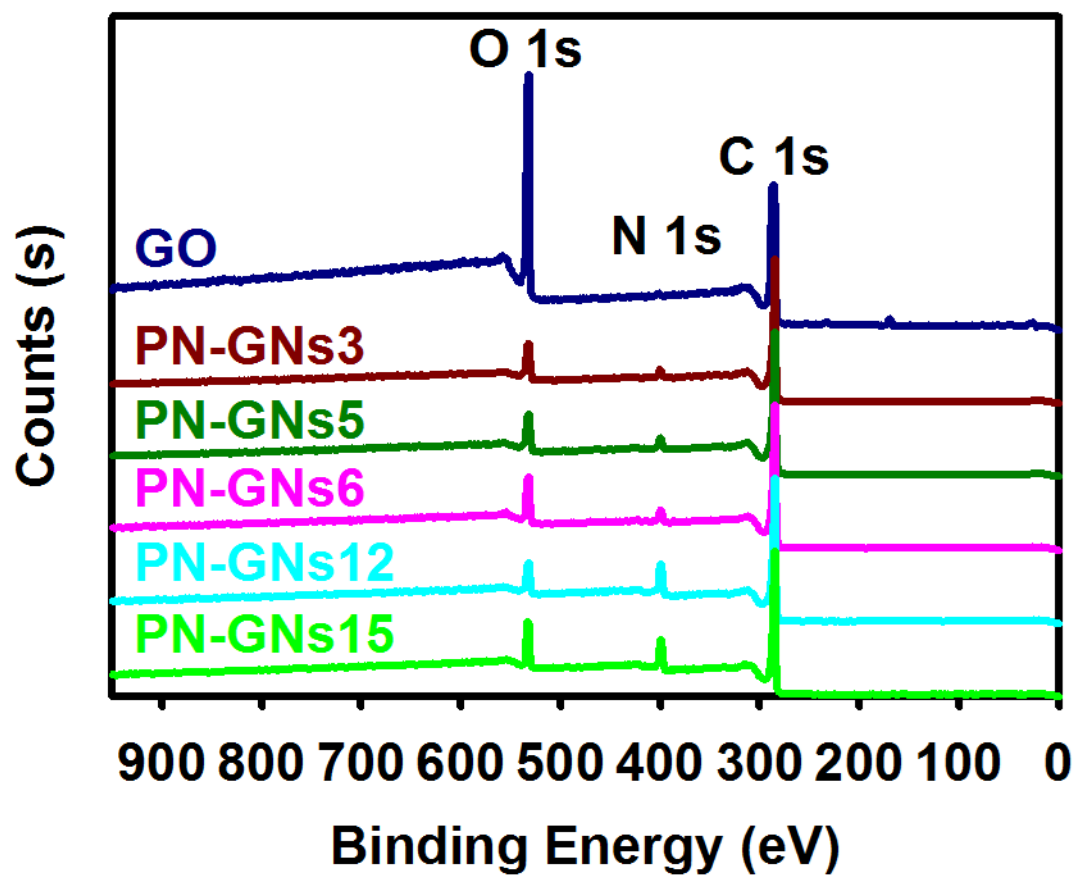


Figure S2. XPS survey spectra of GO, PN-GNs3, PN-GNs5, PN-GNs6, PN-GNs12 and PN-GNs15.

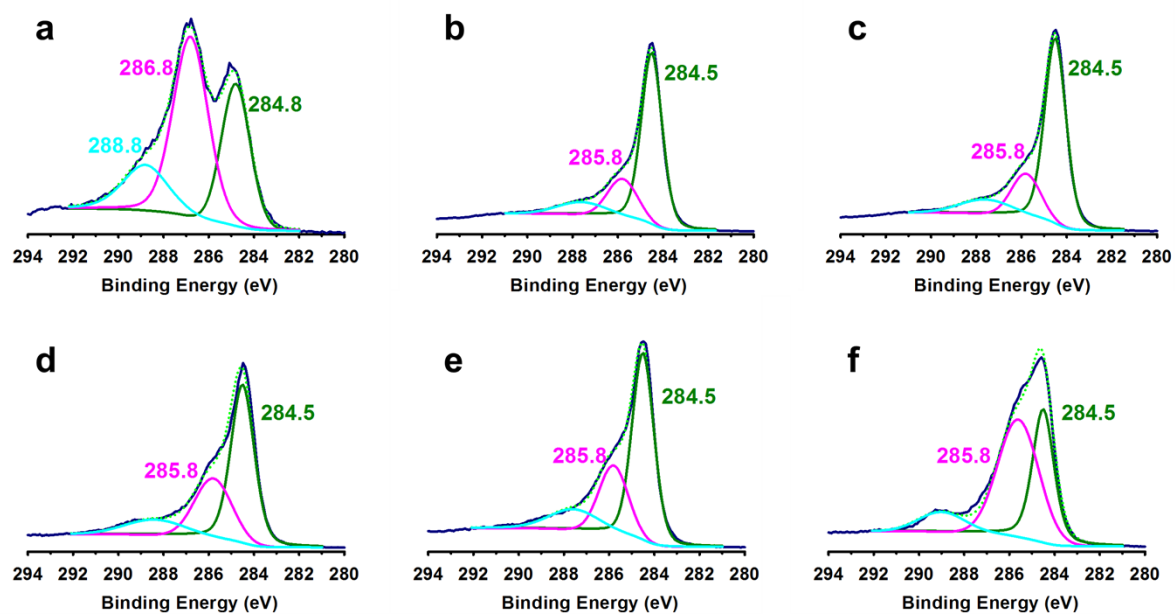


Figure S3. High-resolution XPS spectra of C 1s peaks: (a) GO; (b) PN-GNs3; (c) PN-GNs5; (d) PN-GNs6; (e) PN-GNs12; (f) PN-GNs15.

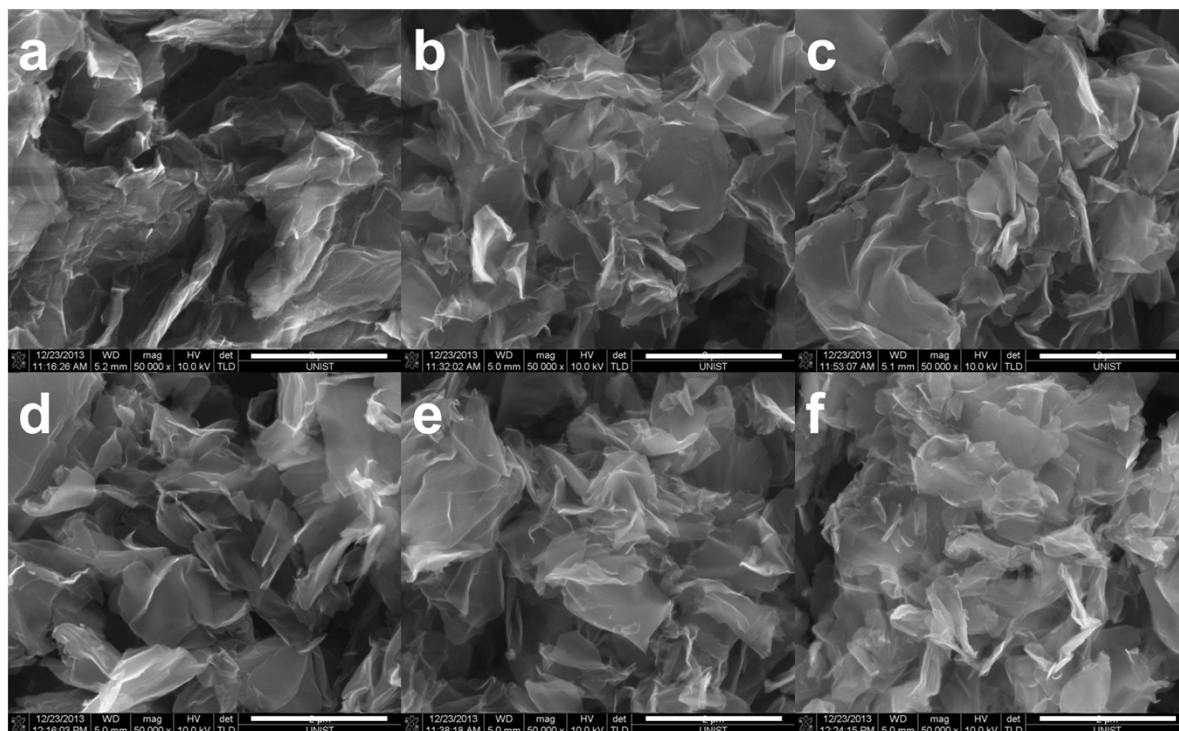


Figure S4. SEM images: (a) GO; (b) PN-GNs3; (c) PN-GNs5; (d) PN-GNs6; (e) PN-GNs12; (f) PN-GNs15. Scale bars are 2 μm.

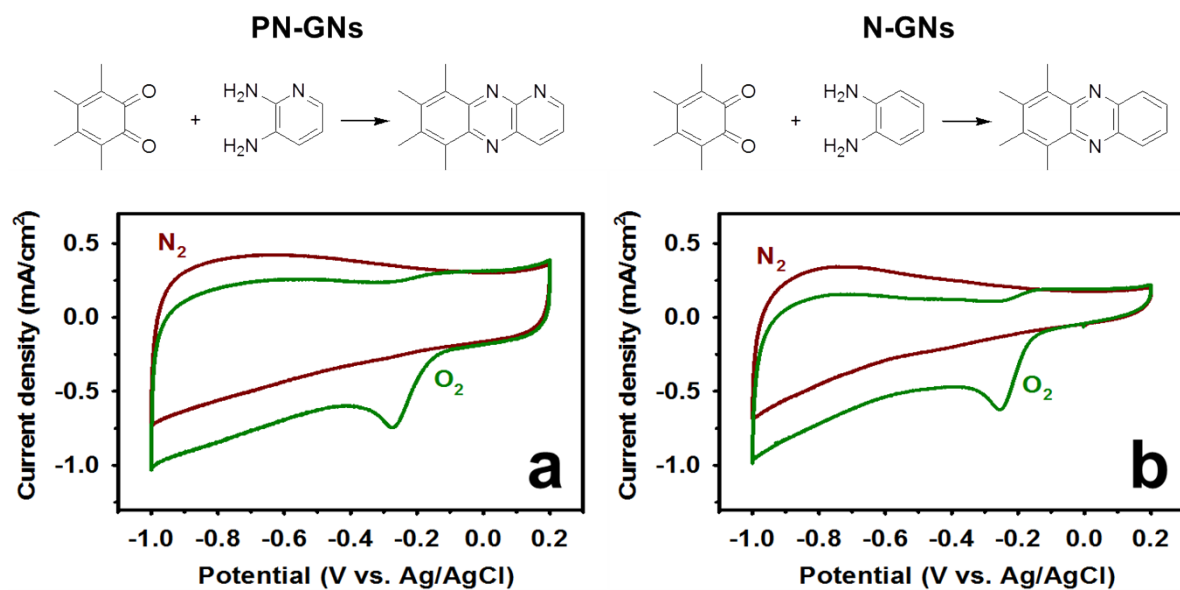


Figure S5. Reaction scheme and cyclic voltammograms (CV) of samples on glassy carbon (GC) electrodes in N_2 - and O_2 - saturated 0.1 M aqueous KOH solution at a scan rate of 10 $mV s^{-1}$: (a) PN-GNs6; (b) N-GNs.

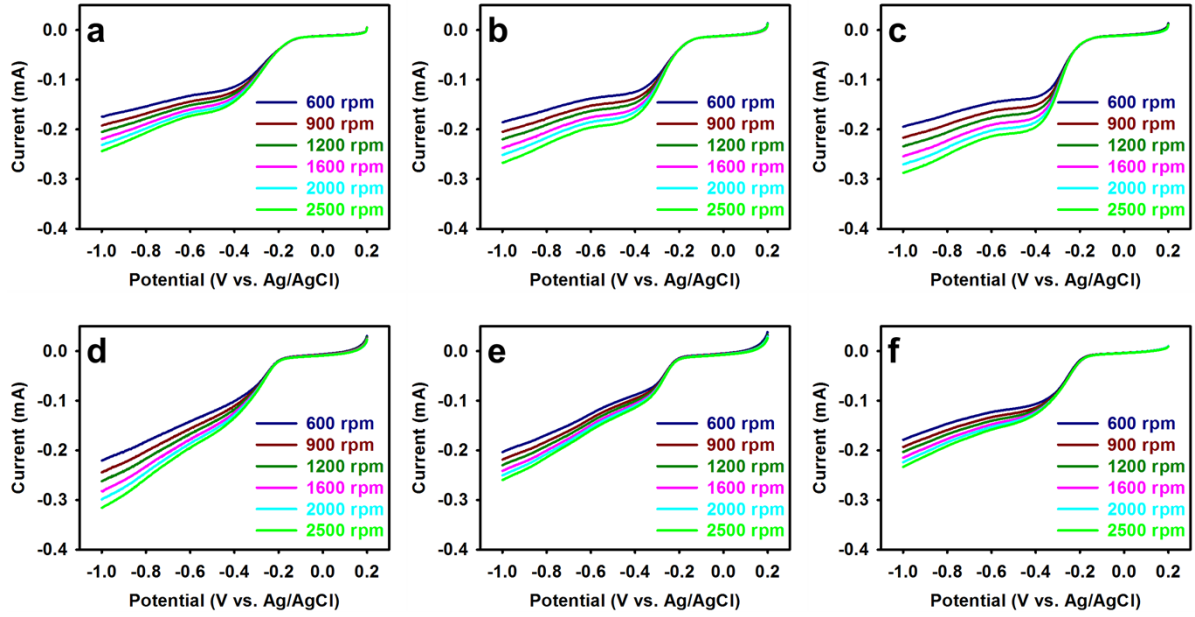


Figure S6. RDE voltammograms in O_2 - saturated 0.1 M aqueous KOH solution with a scan rate of 10 mVs^{-1} at different rotating rates of 600, 900, 1200, 1600, 2000 and 2500 rpm: (a) PN-GNs3; (b) PN-GNs5; (c) PN-GNs6; (d) PN-GNs12; (e) PN-GNs15; (f) N-GNs.

Calculation of electron transferred number (n)

The kinetic analysis for ORR was conducted according to Koutecky-Levich plots:

$$\frac{1}{j} = \frac{1}{j_k} + \frac{1}{B\omega^{0.5}} \quad (1)$$

where j_k is the kinetic current and B is Levich slope which is given by:

$$B = 0.2nF(D_{O_2})^{2/3} \nu^{-1/6} C_{O_2} \quad (2)$$

The constant 0.2 is generally adopted when the rotation speed is expressed in rpm. n is the number of electrons transferred for the reduction of one O_2 molecule, F is the Faraday constant ($F = 96485 \text{ C/mol}$), D_{O_2} is the diffusion coefficient of O_2 ($D_{O_2} = 1.9 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}$), ν

is the kinematics viscosity for KOH ($\nu = 0.01 \text{ cm}^2 \text{ s}^{-1}$) and C_{O_2} is concentration of O_2 in the solution ($C_{\text{O}_2} = 1.2 \times 10^{-6} \text{ mol cm}^{-3}$).

According to equations (1) and (2), the number of electrons transferred (n) can be obtained from the slope of Koutecky-Levich plot of i^{-1} vs. $\omega^{-1/2}$.

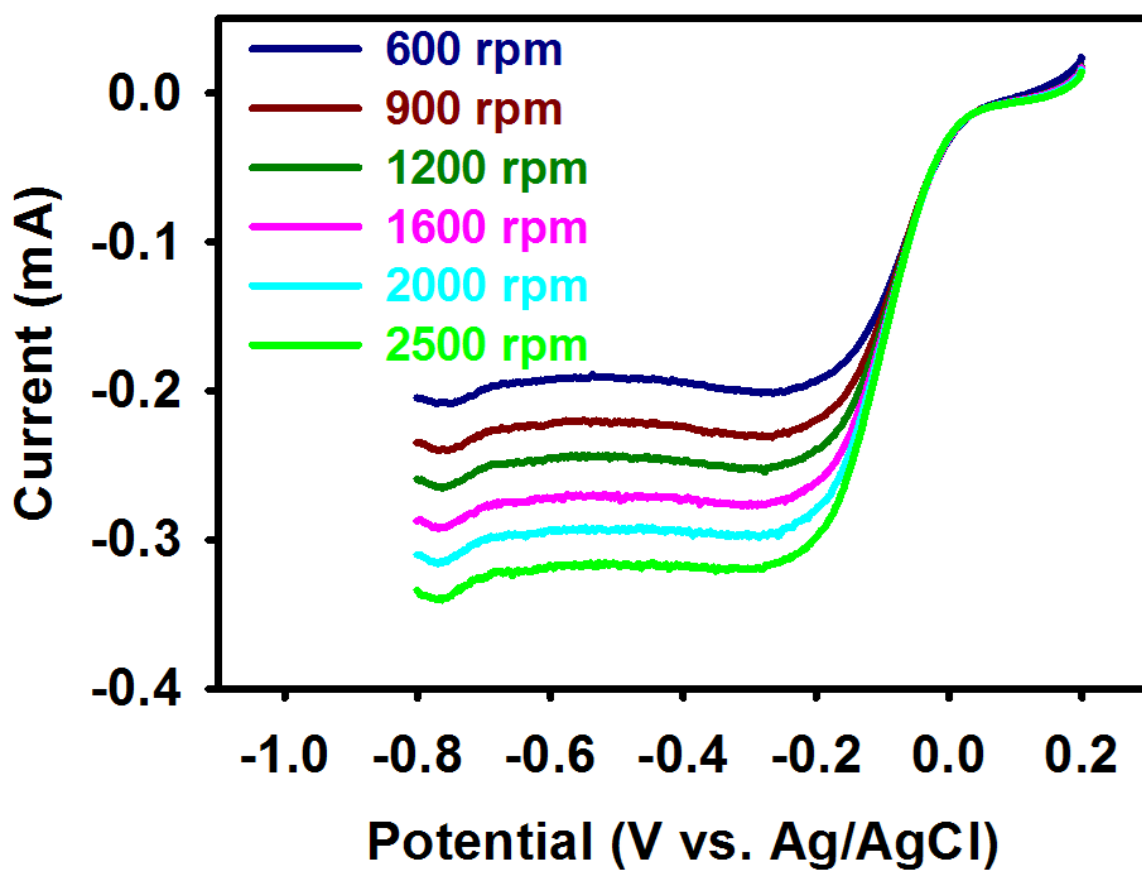


Figure S7. RDE voltammograms of Pt/C in O_2 - saturated 0.1 M aqueous KOH solution with a scan rate of 10 mVs^{-1} at different rotating rates of 600, 900, 1200, 1600, 2000 and 2500 rpm.

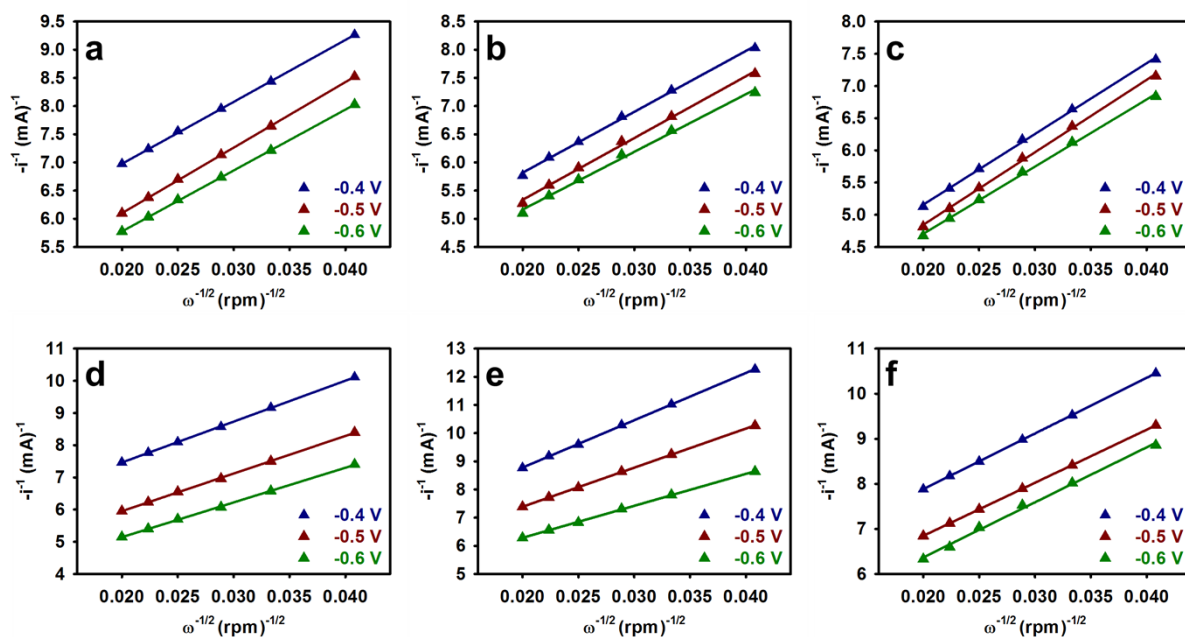


Figure S8. Koutechy-Levich plots derived from RDE measurements at different electrode potentials (blue line: -0.4 V, brown line: -0.5 V and green line: -0.6 V): (a) PN-GNs3; (b) PN-GNs5; (c) PN-GNs6; (d) PN-GNs12; (e) PN-GNs15; (f) N-GNs.

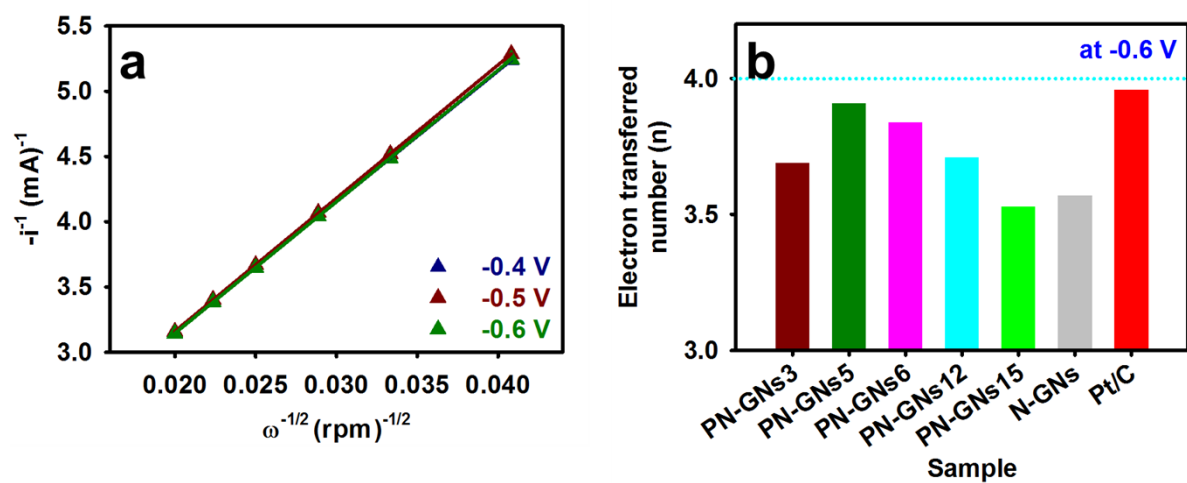


Figure S9. (a) Koutechy-Levich plots derived from RDE measurements of Pt/C at different electrode potentials (blue line: -0.4 V, brown line: -0.5 V and green line: -0.6 V); (b) electron transferred number (n) of all samples at -0.6 V.

Table S1. Elemental analyses of graphite, GO, PN-GNs3, PN-GNs5, PN-GNs6, PN-GNs12, PN-GNs15 and N-GNs

Materials	C	O	H	N	Sum	C/O	C/N
Graphite	98.74	BDL ^a	0.11	BDL ^a	98.86	∞^b	∞^b
GO	52.21	43.34	2.02	BDL ^a	97.57	1.61	∞^b
PN-GNs3	75.12	17.69	1.69	3.27	97.77	5.66	26.72
PN-GNs5	74.33	17.12	1.71	4.82	97.98	5.79	17.99
PN-GNs6	72.89	15.63	1.93	6.49	96.94	6.22	13.10
PN-GNs12	70.18	13.38	2.02	11.64	97.22	6.99	7.03
PN-GNs15	68.19	12.31	2.09	15.28	97.87	7.39	5.21
N-GNs	79.01	10.49	1.62	6.45	97.57	10.04	14.29

^a BDL = Below detection limit

^b ∞ = Unlimited

Table S2. Capacitance (F/g) of samples on glassy carbon (GC) electrodes in N₂-and O₂-saturated 0.1 M aqueous KOH solution at a scan rate of 10 mV s⁻¹

Sample	Capacitance (F/g) in O₂	Capacitance (F/g) in N₂
PN-GNs3	130.1	120.2
PN-GNs5	132.5	120.6
PN-GNs6	139.7	122.5
PN-GNs12	129.4	118.6
PN-GNs15	108.5	104.8
N-GNs	97.3	81.4
Pt/C	88.8	82.3

Table S3. Onset potentials and limiting diffusion current at -0.6 V of all samples on a rotating disk electrode at a rotation speed of 1600 rpm and a scan rate of 10 mV s⁻¹

	PNGNs3	PNGNs5	PNGNs6	PNGNs12	PNGNs15	N-GNs	Pt/C
Onset	-0.17	-0.19	-0.20	-0.20	-0.21	-0.22	0.01
potential (V)							
Current	-0.16	-0.18	-0.19	-0.18	-0.15	-0.14	-0.27
(mA)							

Table S4. Electron transferred numbers (n) of all samples from RDE measurements at different electrode potentials of -0.4 V, -0.5 V and -0.6 V

Sample	-0.4 V	-0.5 V	-0.6 V
PN-GNs3	3.63	3.43	3.70
PN-GNs5	3.70	3.65	3.91
PN-GNs6	3.64	3.55	3.84
PN-GNs12	3.15	3.42	3.71
PN-GNs15	2.38	3.88	3.53
N-GNs	3.24	3.40	3.57
Pt/C	4.00	3.92	3.96