

**Supplementary Figure S1.** CNT/CNP composite: *ca.* 10 µm CNTs distributed homogeneously in CNPs with initimate contact between CNT and CNP maintained.



**Supplementary Figure S2.** TEM of CNTs in N-Fe-CNT/CNP catalyst showing Fe nanoparticles encapsulated in several carbon nanoshells in nanotubes or individual particles; graphene defects are also shown.



**Supplementary Figure S3.** N 1s XPS spectra of N-Fe-CNT/CNP catalyst. Thin black line: measured data; thick dark blue line: fitted curve. Deconvoluted nitrogen peaks: pyridinic-N (398.3 eV, red line); pyrrolic-N (400.0 eV, blue line); graphitic-N (401.1 eV, green line).



**Supplementary Figure S4**. Steady-state ORR polarization plots recorded with N-Fe-CNT/CNP catalysts from six different syntheses. Disk loading 0.2 mg cm<sup>-2</sup>;  $E_{\frac{1}{2}} = 0.87(3) \pm 0.01(2)$ . Plot in green has been used in Fig. 2a in the manuscript.



**Supplementary Figure S5**. ORR Tafel plots in O<sub>2</sub>-saturated 0.1 M NaOH. (a) Plots for three different N-Fe-CNT/CNP catalyst loadings. (b) Comparison between Tafel plots recorded with N-Fe-CNT/CNP catalyst and Pt/C reference catalyst before the cycling test (Tafel slope calculated by linear regression method). The mass transport corrected ORR specific kinetic current density ( $i_{kinetic}$ ) was calculated by the following equation:  $i_{kinetic} = \frac{i_{measured} \times i_{limited}}{i_{limited} - i_{measured}}$ 



**Supplementary Figure S6.** Cyclic voltammograms in N<sub>2</sub>-saturated electrolyte recorded during the cycling durability test. (a) Pt/C catalyst (60  $\mu g_{Pt}$  cm<sup>-2</sup>). (b) N-Fe-CNT/CNP catalyst (1.0 mg cm<sup>-2</sup>).



**Supplementary Figure S7.** ORR polarization plots measured with Pt/C (60  $\mu$ g<sub>Pt</sub> cm<sup>-2</sup>) during cycling durability tests in O<sub>2</sub>-saturated 0.1 M NaOH in the potential range of 0.6-1.0 V *vs.* RHE. Scan rate 50 mV s<sup>-1</sup>.



**Supplementary Figure S8.** RDE chronoamperometric stability tests of N-Fe-CNT/CNP and Pt/C catalysts at different potentials. O<sub>2</sub>-saturated 0.1 M NaOH; 900 rpm; 25°C.



**Supplementary Figure S9**. Dependence of ORR activity and H<sub>2</sub>O<sub>2</sub> yield on catalyst loading. The percent H<sub>2</sub>O<sub>2</sub> yield was calculated from the disk current ( $I_D$ ), the ring current ( $I_R$ ), and the collection efficiency (N) using the following equation:  $x_{H_2O_2}(\%) = \frac{200I_R/N}{I_D + I_R/N}$ 

Samples	Atomic Content, %				
	C 1s	O 1s	N 1s	S 2p	Fe 2p
As-received BP 2000	95.07	4.72	-	0.21	-
N-Fe-CNT/CNP catalyst	92.57	4.01	3.09	0.24	0.08

Supplementary Table S1. Elemental composition of as-received BP 2000 and N-Fe-CNT/CNP catalyst.