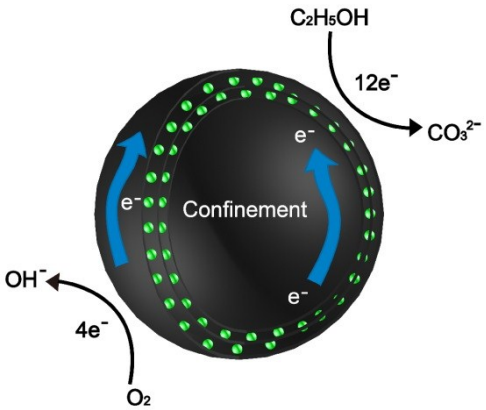
Supplementary Information

Facile synthesis of N-doped porous carbon encapsulated bimetallic PdCo as highly active and durable electrocatalysts for oxygen reduction and ethanol oxidation

Zheye Zhang, Shasha Liu, Xin Tian, Juan Wang, Pei Xu, Fei Xiao, and Shuai Wang*

Key laboratory of Material Chemistry for Energy Conversion and Storage, Ministry of Education, School of Chemistry and Chemical Engineering, Huazhong University of Science and Technology, Wuhan, 430074, China

Corresponding author: *E-mail: chmsamuel@mail.hust.edu.cn (S. Wang).



Scheme S1. Illustration of the bifunctional electrocatalyst of PdCo alloys confined in NPNCs.

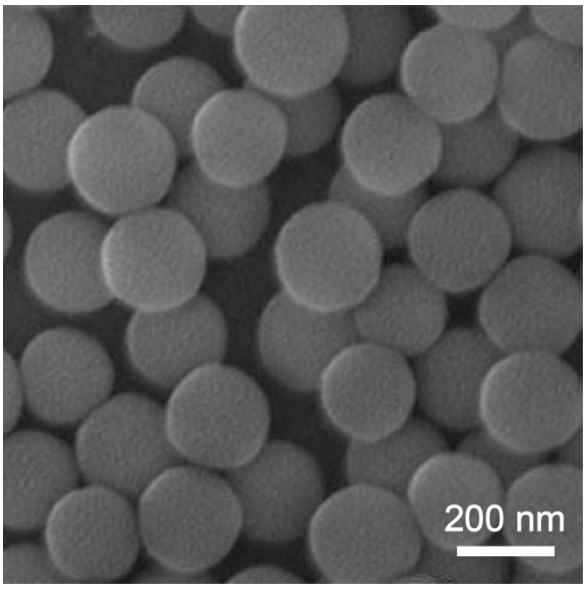


Figure S1. SEM image of SiO_2.

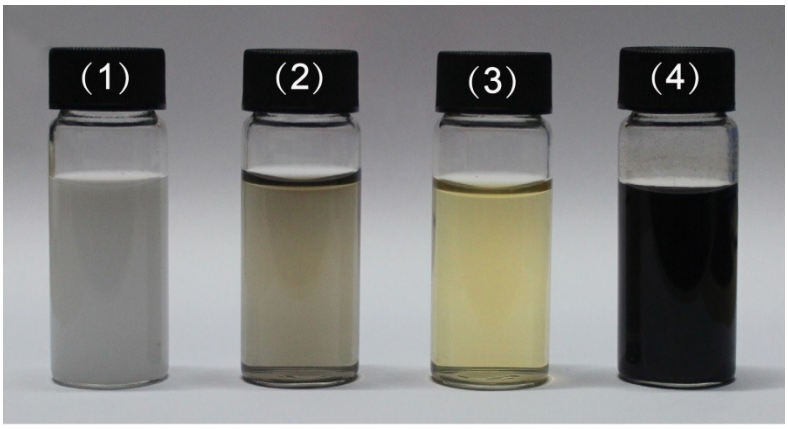


Figure S2. Photographs of SiO_2 dispersion (1), dopamine solution (2), K_2PdCl_4 and $Co(NO_3)_2 \cdot 6H_2O$ mixed solution (3), the mixture of (1), (2) and (3) with the addition of tris-(hydroxymethyl)aminomethan (4).

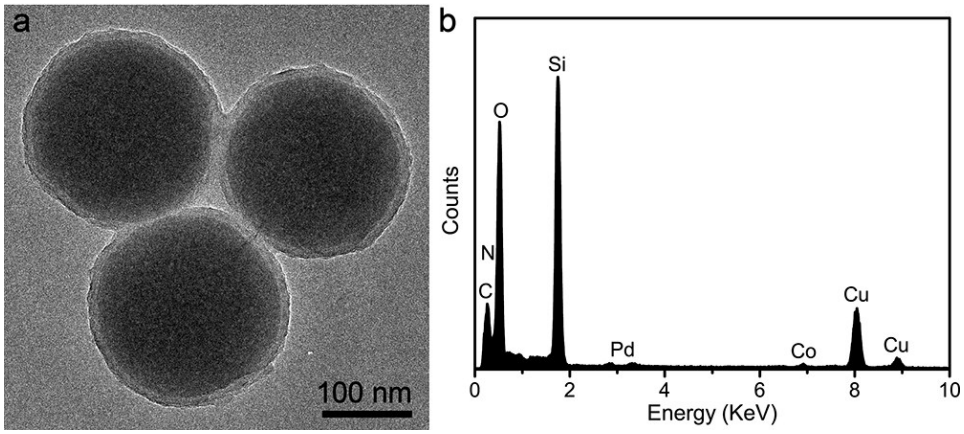


Figure S3. (a) TEM image and (b) EDX spectrum of SiO_2@$PdCl_4^{2-}$/Co^{2+}-PDA.

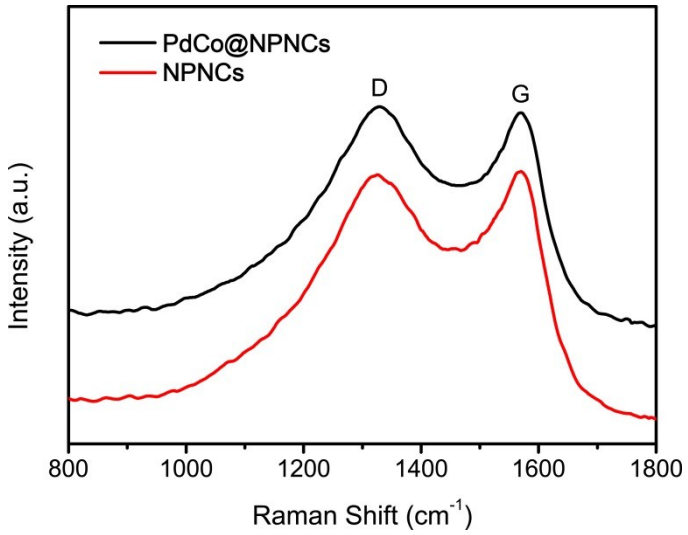


Figure S4. Raman spectra of PdCo@NPNCs and NPNCs.

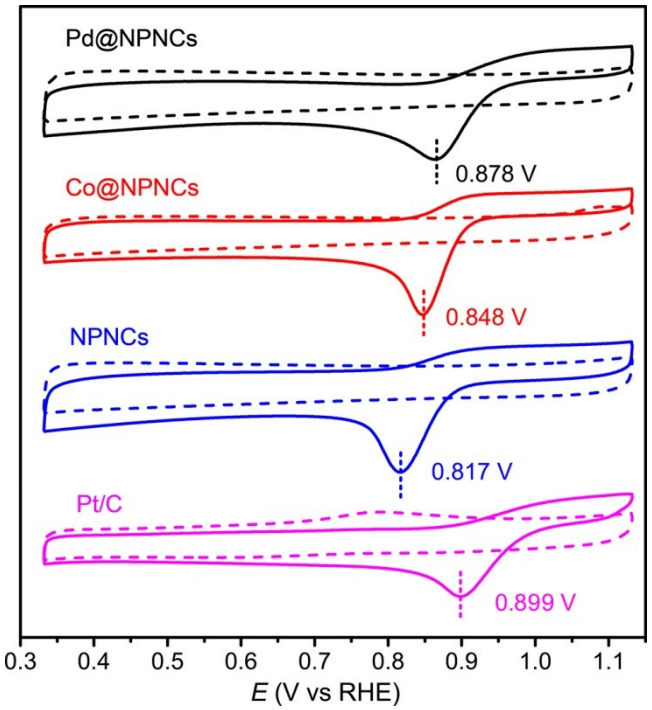


Figure S5. CV curves of Pd@NPNCs, Co@NPNCs, NPNCs and commercial Pt/C catalysts in 0.1 M KOH saturated with N_2 (dashed curves) and O_2 (solid curves).

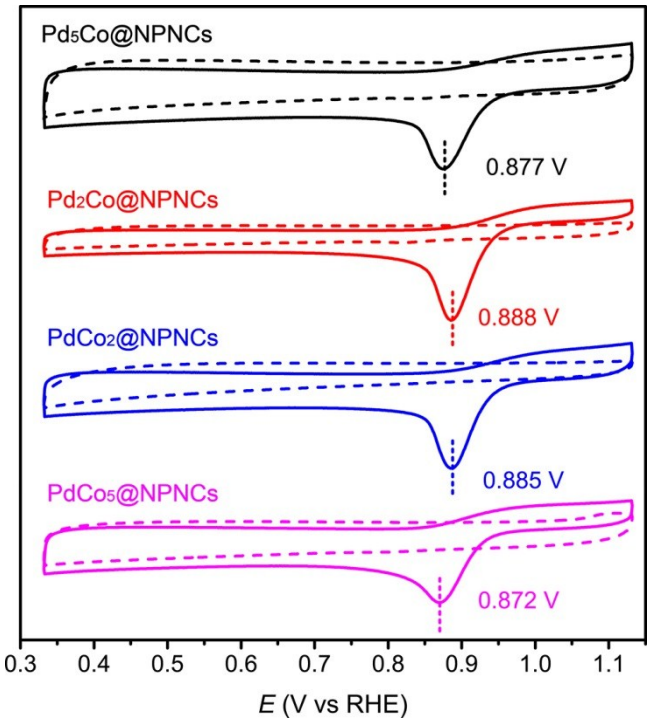


Figure S6. CV curves of Pd_xCo_y@NPNCs (x:y = 5:1, 2:1, 1:2, 1:5) in 0.1 M KOH saturated with N_2 (dashed curves) and O_2 (solid curves).

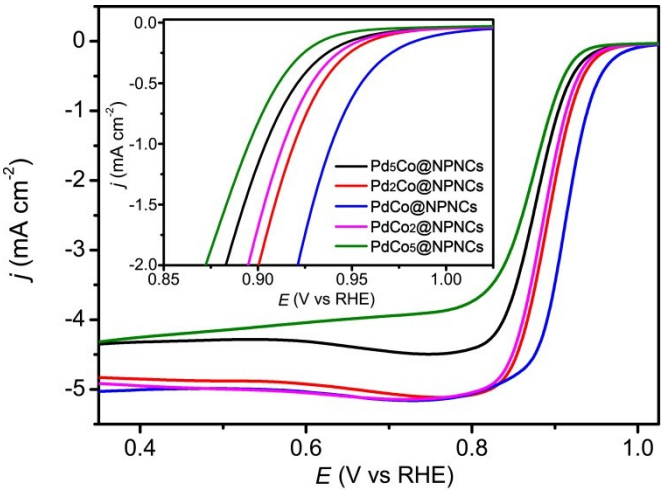


Figure S7. LSV curves of Pd_xCo_y@NPNCs (x:y = 5:1, 2:1, 1:1, 1:2, 1:5) in 0.1 M KOH saturated with O_2.

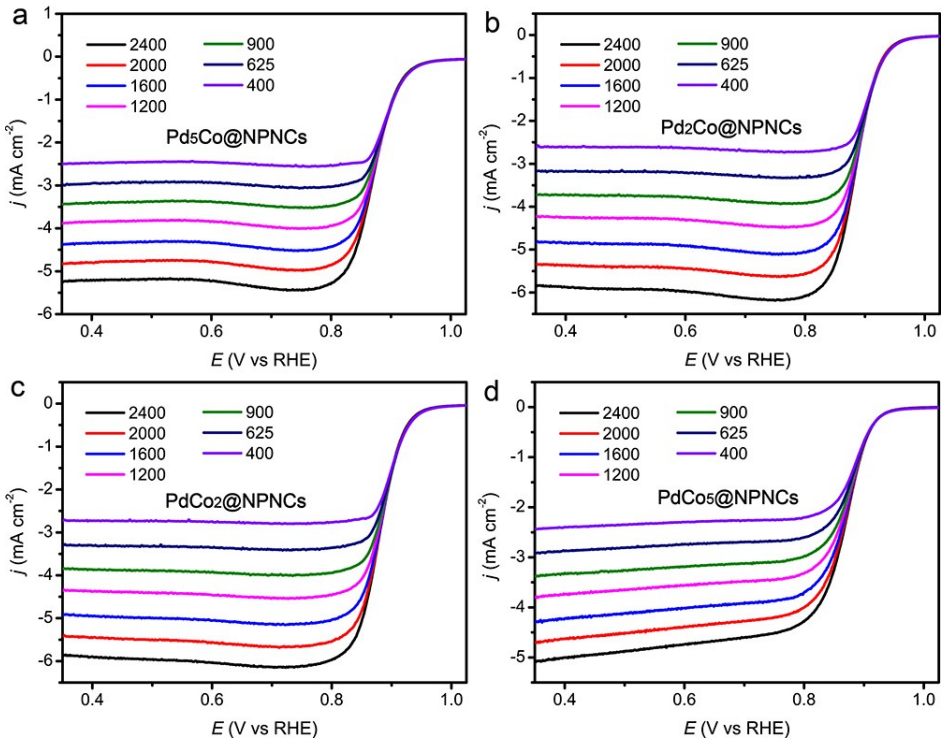


Figure S8. LSV curves of Pd_5Co@NPNCs (a), Pd_2Co@NPNCs (b), $PdCo_2$@NPNCs (c) and $PdCo_5$@NPNCs (d) in O_2-saturated 0.1 M KOH with various rotating speeds.

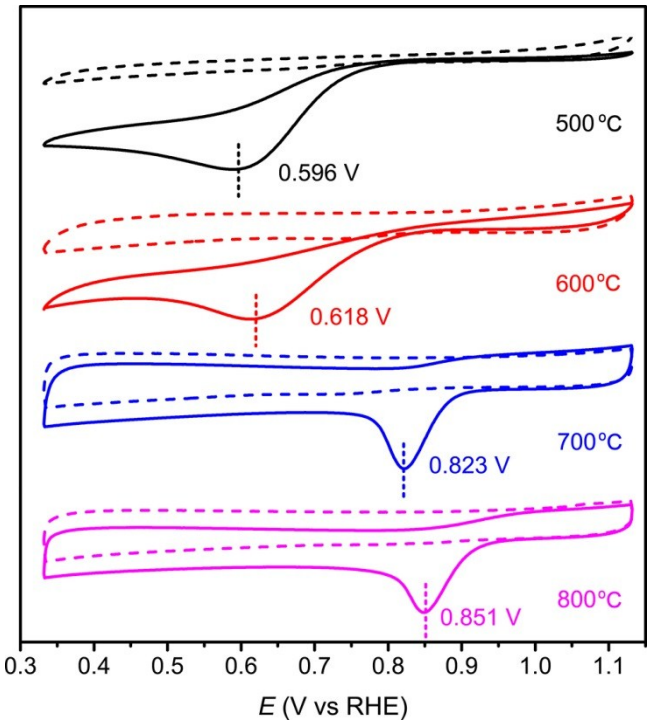


Figure S9. CV curves of PdCo@NPNCs obtained with different pyrolysis temperature (500 ^{o}C, 600 ^{o}C, 700 ^{o}C, 800 ^{o}C) in 0.1 M KOH saturated with N_2 (dashed curves) and O_2 (solid curves).

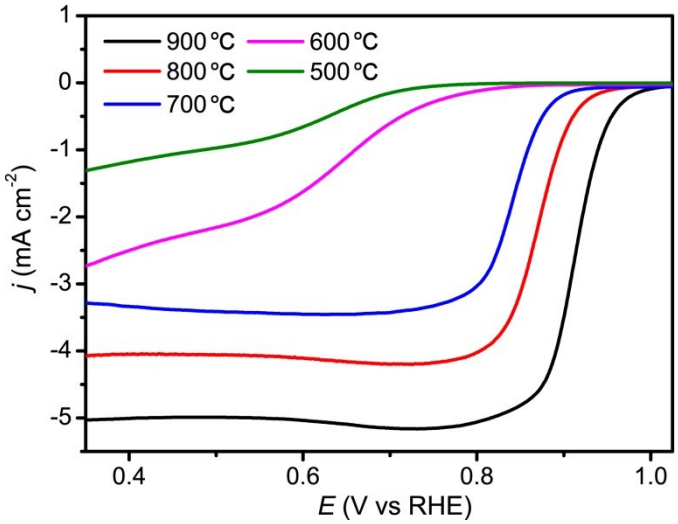


Figure S10. LSV curves of PdCo@NPNCs obtained at different pyrolysis temperature in 0.1 M KOH saturated with O_2.

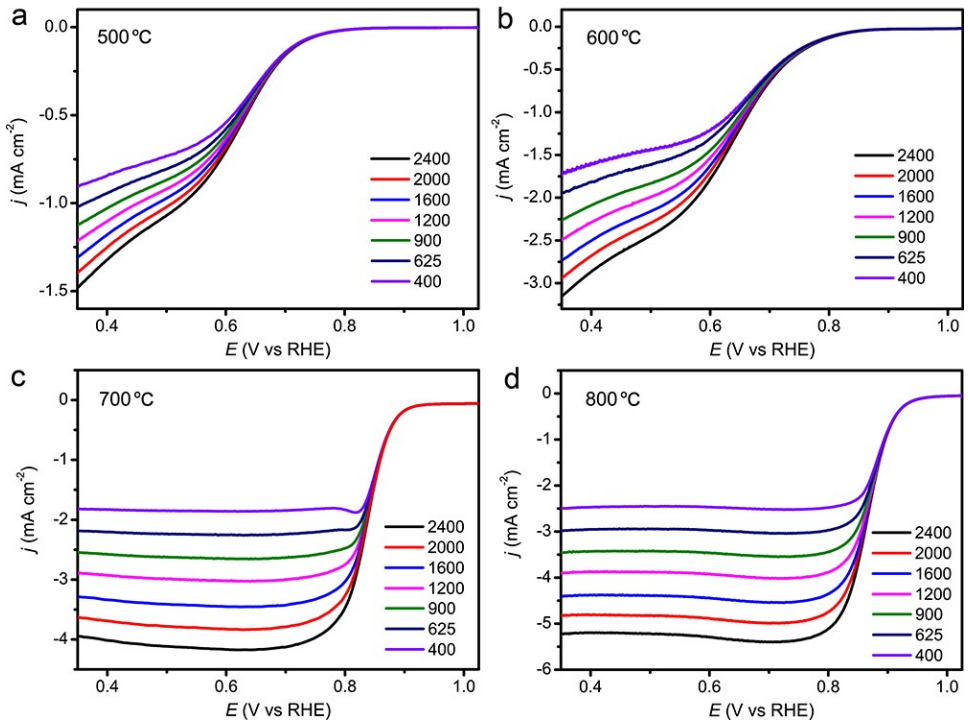


Figure S11. LSV curves of PdCo@NPNCs obtained at 500 °C (a), PdCo@NPNCs obtained at 600 °C (b), PdCo@NPNCs obtained at 700 °C (c) and PdCo@NPNCs obtained at 800 °C (d) in O_2-saturated 0.1 M KOH with various rotating speeds.

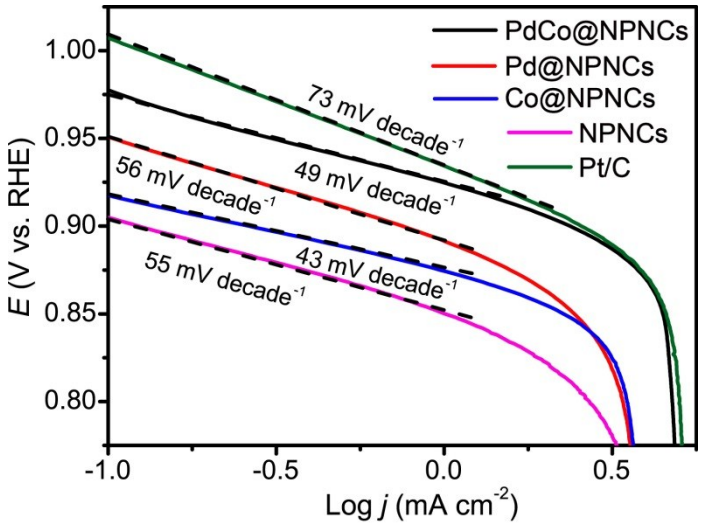


Figure S12. The Tafel curves of the PdCo@NPNCs, Pd@NPNCs, Co@NPNCs, NPNCs and Pt/C catalysts.

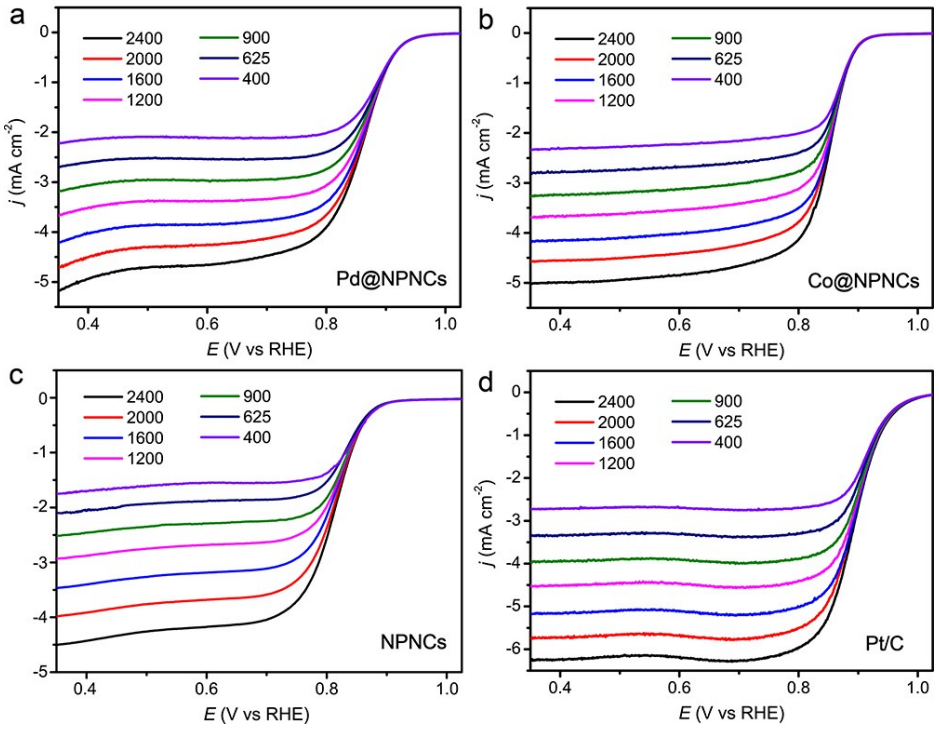


Figure S13. LSV curves of Pd@NPNCs (a), Co@NPNCs (b), NPNCs (c) and Pt/C (d) in O_2-saturated 0.1 M KOH with various rotating speeds.

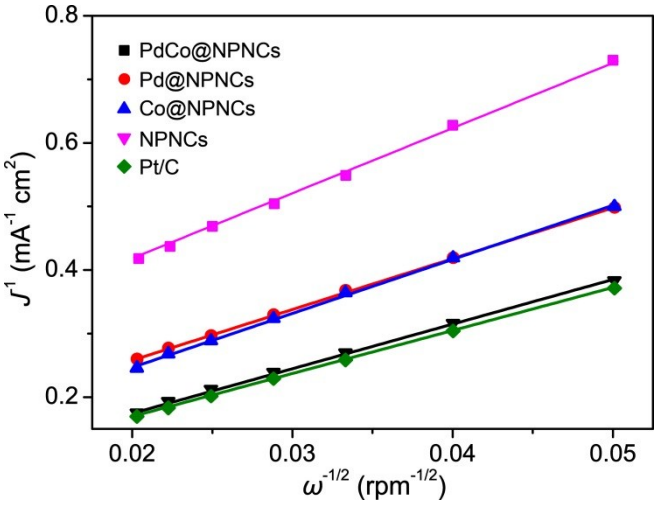


Figure S14. K–L plots for PdCo@NPNCs, Pd@NPNCs, Co@NPNCs, NPNCs and Pt/C catalysts at 0.8 V.

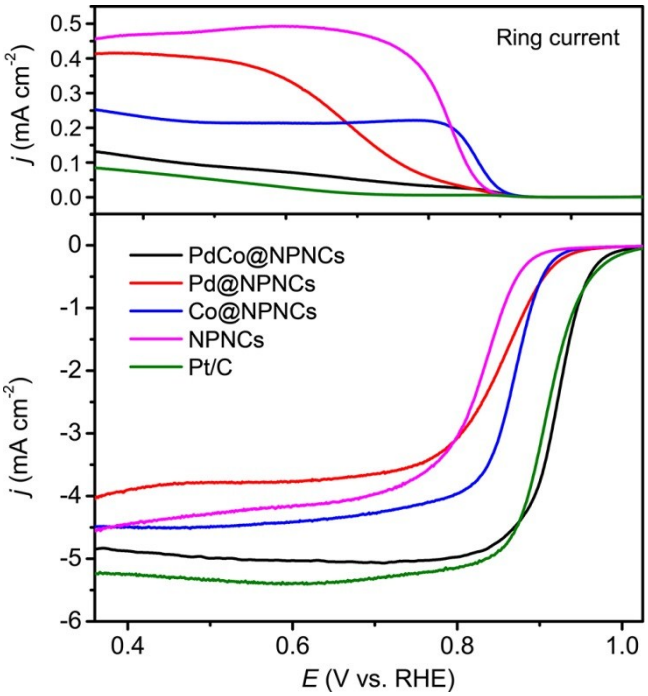


Figure S15. RRDE tests (1600 r.p.m.) of PdCo@NPNCs, Pd@NPNCs, Co@NPNCs, NPNCs and Pt/C catalysts for ORR in 0.1 M KOH saturated with O_2.

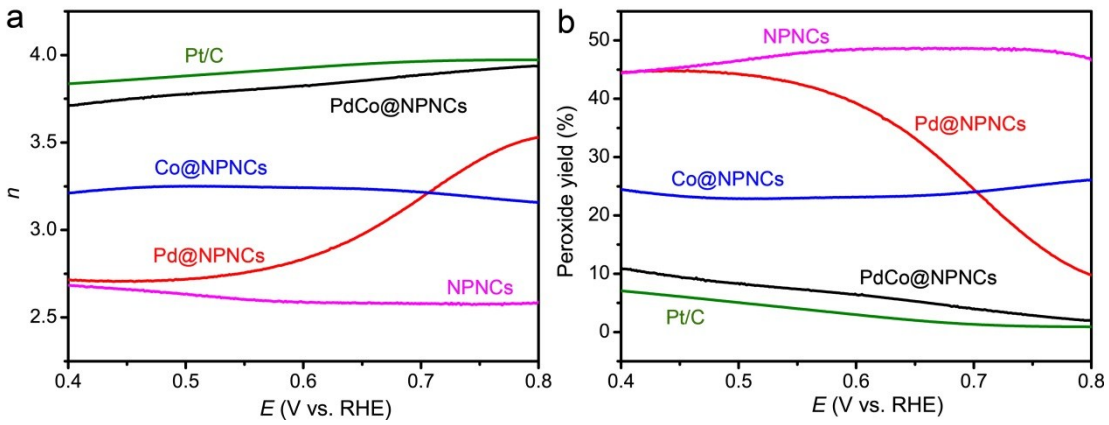


Figure S16. The number of electron transfer (a) and the percentage of peroxide in the total oxygen reduction products (b) at various catalysts based on the RRDE result.

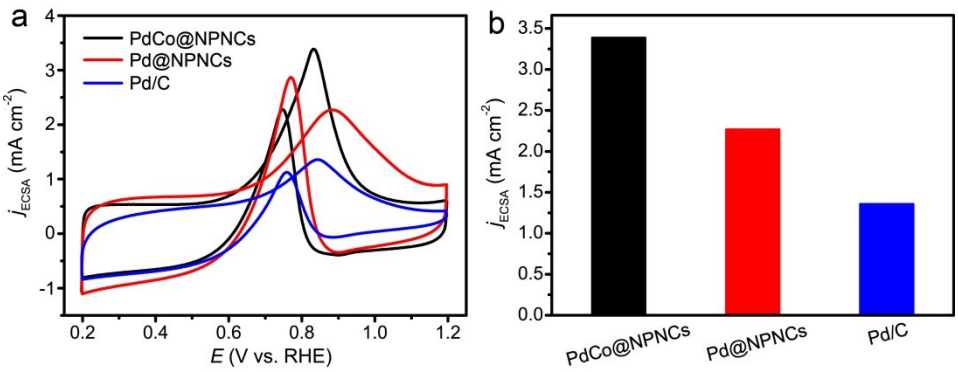


Figure S17. (a) CV curves of PdCo@NPNCs, Pd@NPNCs and commercial Pd/C catalysts in 1.0 M KOH + 1.0 M C_2H_5OH at 50 mV s^{-1} (the current densities all are normalized to the ECSAs of catalysts). (b) The comparison of ECSA peak current densities of PdCo@NPNCs, Pd@NPNCs and Pd/C in 1.0 M KOH + 1.0 M C_2H_5OH at 50 mV s^{-1}.

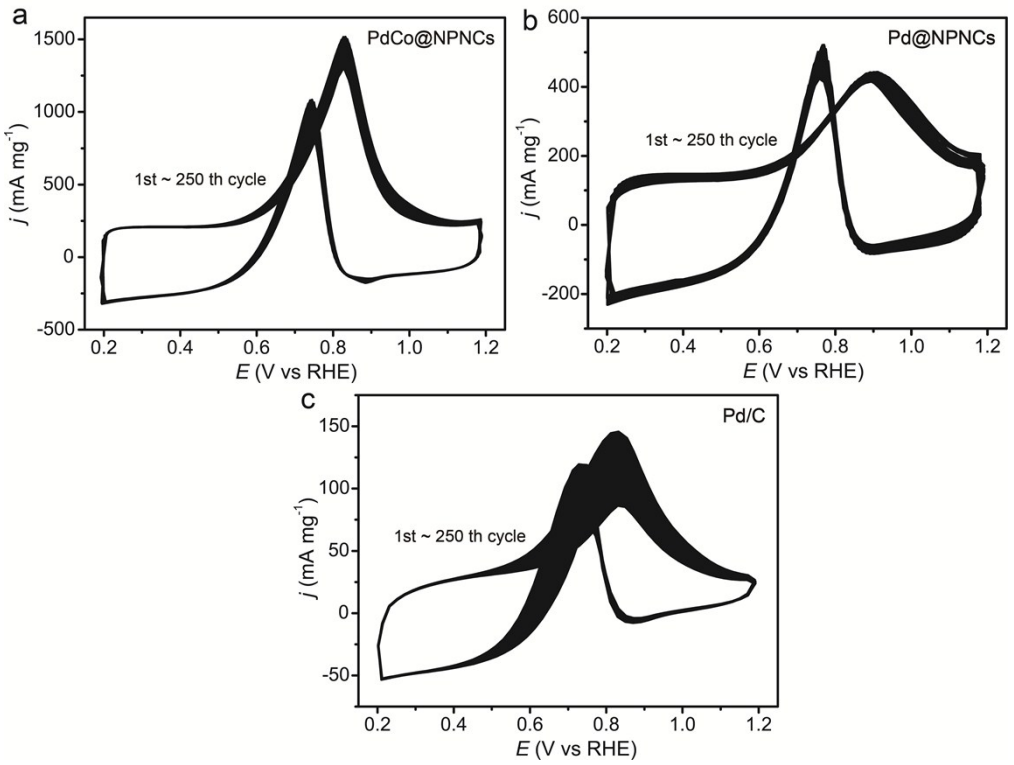


Figure S18. CV curves of PdCo@NPNCs (a), Pd@NPNCs (b) and Pd/C (c) from 1 st to 250 th cycle in solution of 1.0 M KOH + 1.0 M C_2H_5OH at 50 mV s^{-1}.

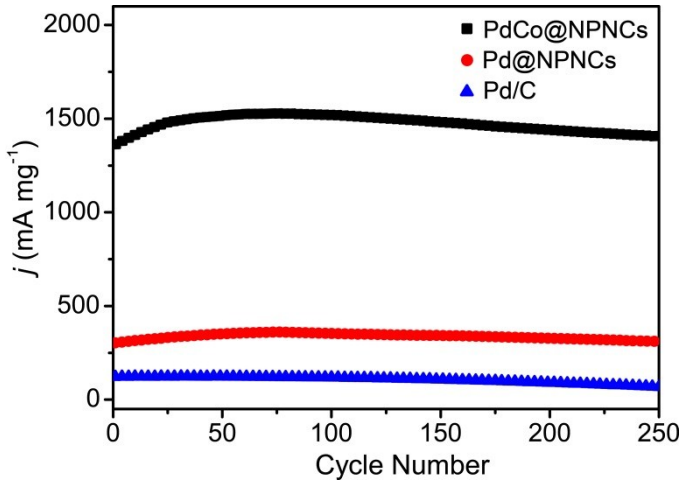


Figure S19. The cycling stability of peak current densities of PdCo@NPNCs, Pd@NPNCs and commercial Pd/C catalysts with increasing cycles.

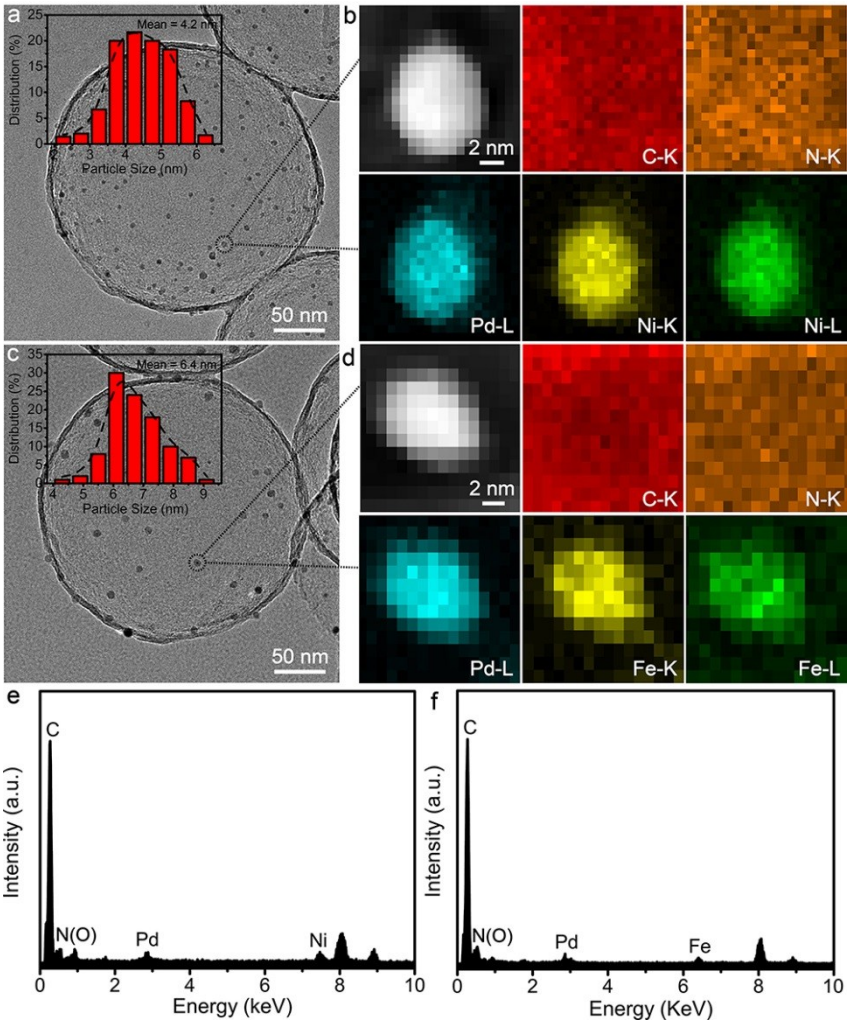


Figure S20. (a) Typical TEM image of PdNi@NPNCs. Inset: the corresponding particle size distribution histogram of bimetallic PdNi NPs. (b) EDX mapping images of bimetallic PdCo NPs. (c) Typical TEM image of PdFe@NPNCs. Inset: the corresponding particle size distribution histogram of bimetallic PdFe NPs. (d) EDX mapping images of bimetallic PdFe NPs. (e,f) EDX spectrum of PdNi@NPNCs (e) and PdFe@NPNCs (f).

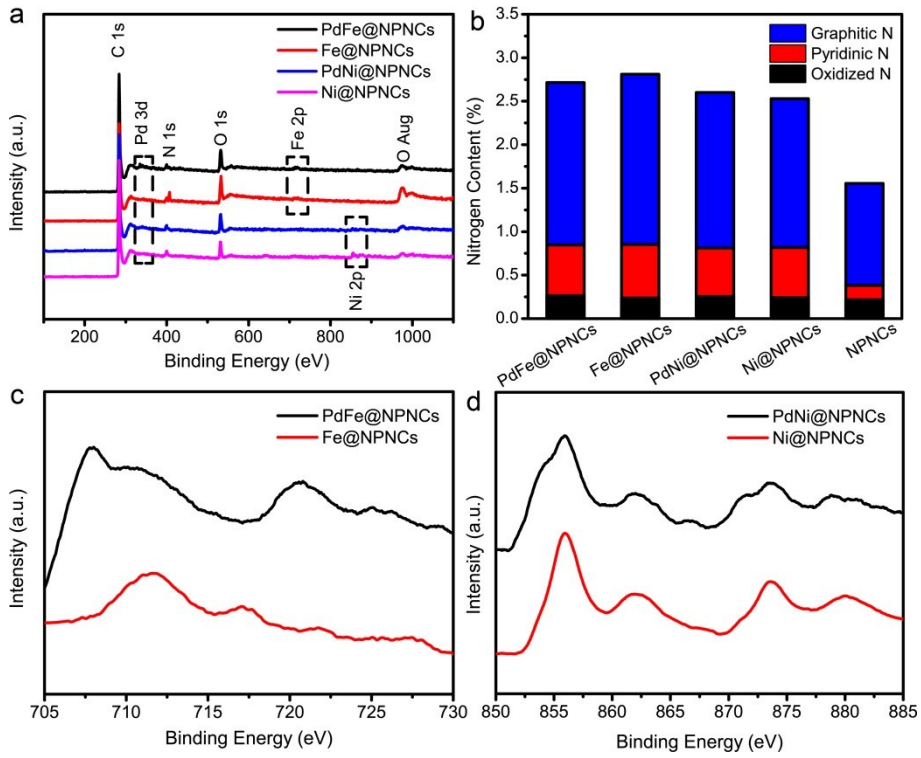


Figure S21. (a) XPS spectra of the surface chemical composition of PdFe@NPNCs, Fe@NPNCs, PdNi@NPNCs and Ni@NPNCs. (b) The concentrations of different N species in PdFe@NPNCs, Fe@NPNCs, PdNi@NPNCs, Ni@NPNCs and NPNCs. (c) XPS spectra of Fe 2p performed on the PdFe@NPNCs and Fe@NPNCs. (d) XPS spectra of Ni 2p performed on the PdNi@NPNCs and Ni@NPNCs.