

**Oxygen Reduction Reaction and Hydrogen Evolution Reaction Catalyzed by PdRu Nanoparticles
Encapsulated in Porous Carbon Nanosheets**

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Table S1. The summary of the ORR performance and size of the samples with different Pd-to-Ru ratios (The total metal mass loading was set as 10%).

Sample	Molar ratio of Pd: Ru	Size (nm)	E_{onset} (V vs. RHE)	$E_{1/2}$ (V vs. RHE)	Diffusion-limited current density @ +0.3 V (mA cm ⁻²)
Ru ₁₀₀ /CNs	1: 0	1.55 ± 0.57	0.841	0.764	-3.27
Pd ₃₃ Ru ₆₇ /CNs	1: 2	1.94 ± 0.40	0.877	0.791	-4.26
Pd ₅₀ Ru ₅₀ /CNs	1: 1	3.67 ± 0.96	0.903	0.799	-5.14
Pd ₆₇ Ru ₃₃ /CNs	2: 1	5.90 ± 1.02	0.895	0.804	-3.99
Pd ₁₀₀ /CNs	0: 1	3.76 ± 0.99	0.877	0.813	-3.43
Pd/C	/	/	0.915	0.816	-3.67

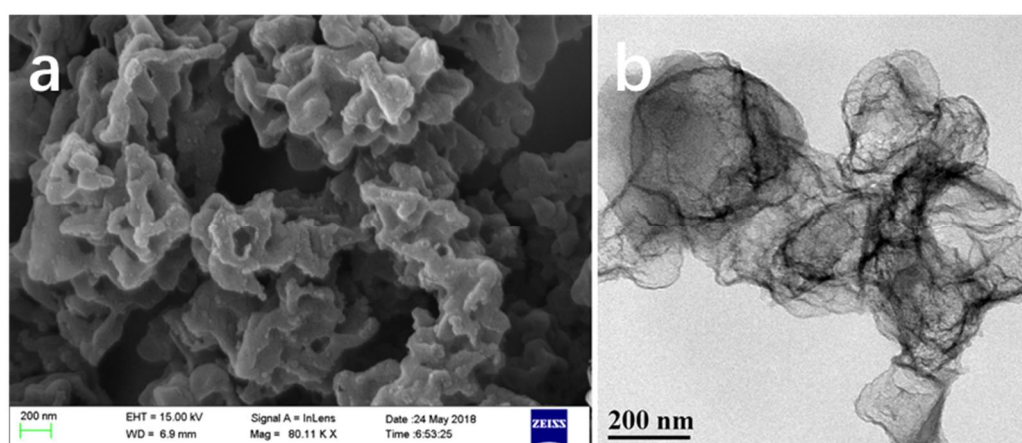


Figure S1. (a) The typical SEM and (b) TEM images of porous carbon nanosheets.

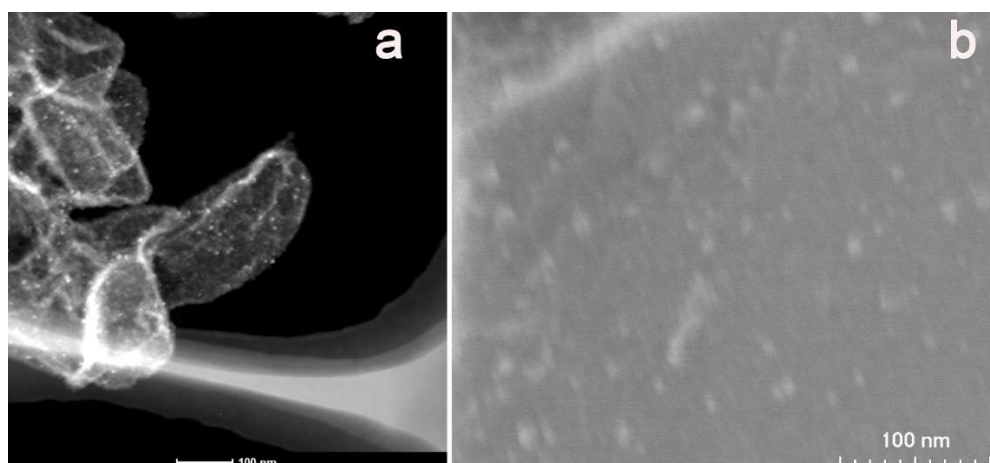


Figure S2. (a) The typical high-angle annular dark field-scanning tunneling electron microscopy (HAADF-STEM) and (b) Scanning electron microscopy (SEM) images of Pd₅₀Ru₅₀/CNs.

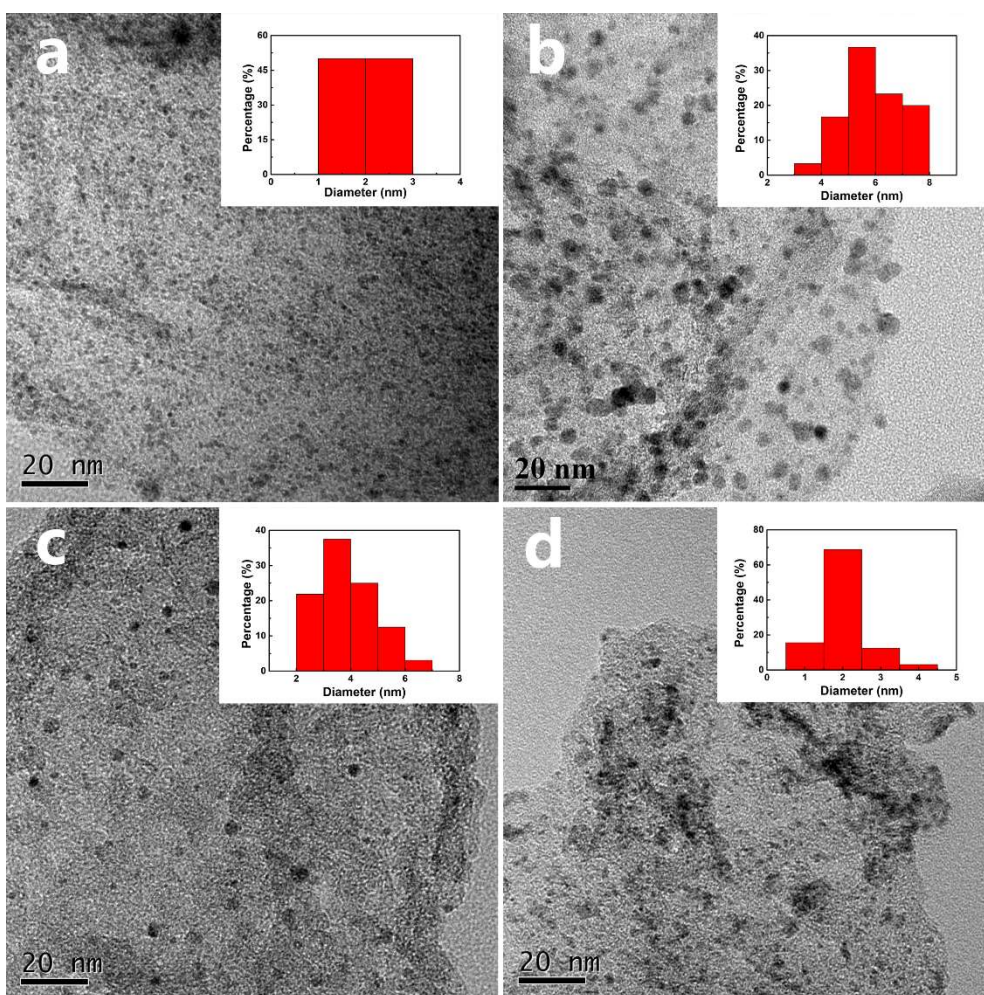


Figure S3. The representative TEM images of (a) Pd₃₃Ru₆₇/CNs (1.94 ± 0.40 nm), (b) Pd₆₇Ru₃₃/CNs (5.90 ± 1.02 nm), (c) Pd₁₀₀/CNs (3.76 ± 0.99 nm) and (d) Ru₁₀₀/CNs (1.55 ± 0.57 nm). (Inset is the corresponding size distribution histogram).

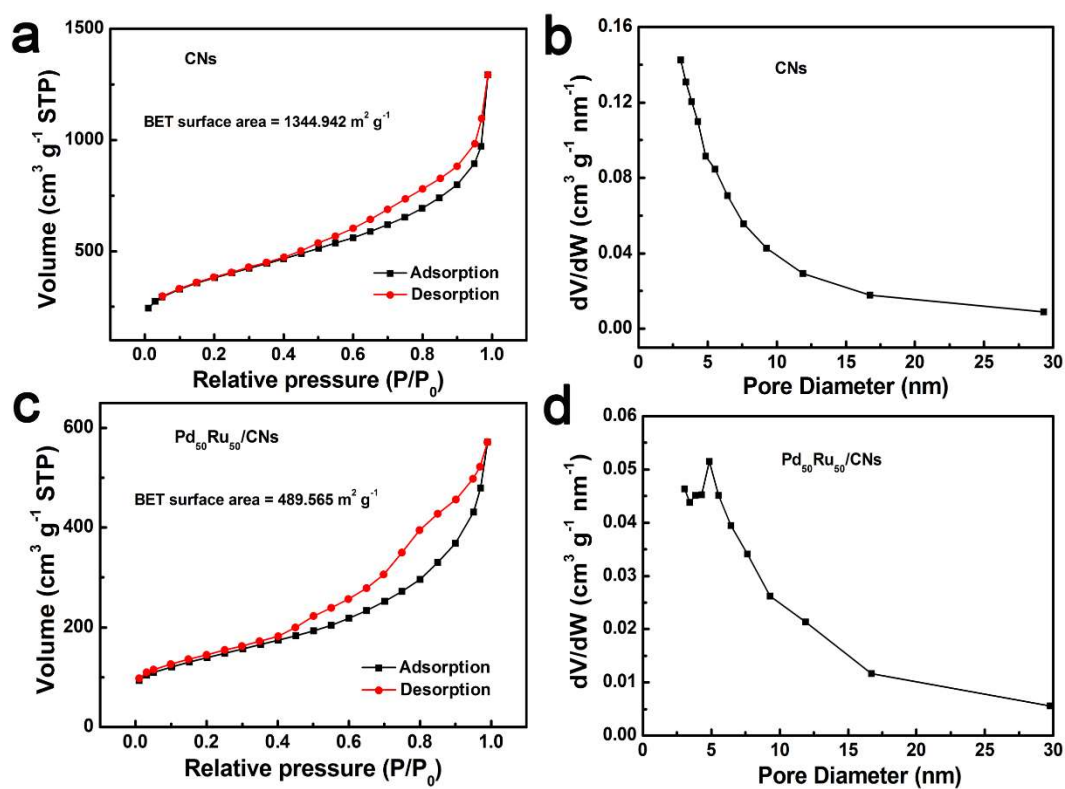


Figure S4 (a, c) Nitrogen adsorption/desorption isotherms at 77 K and (b, d) the corresponding pore-size distribution of CNs and $\text{Pd}_{50}\text{Ru}_{50}/\text{CNs}$.

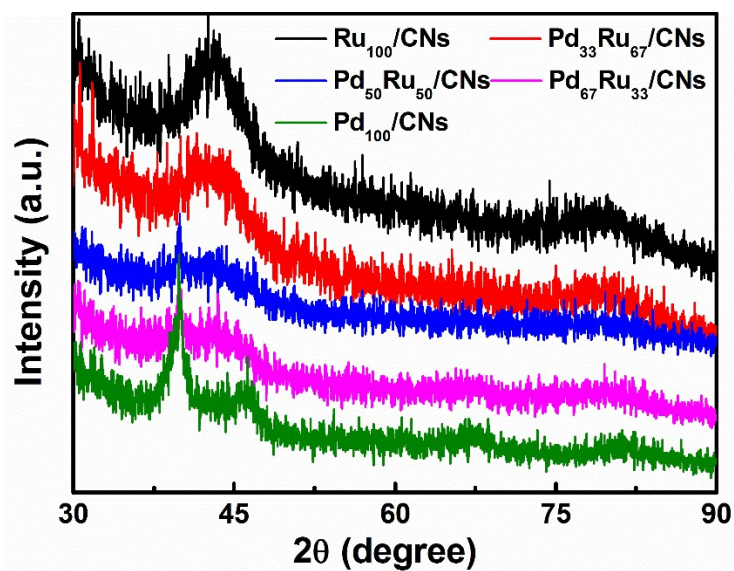


Figure S5. The XRD patterns of all the samples.

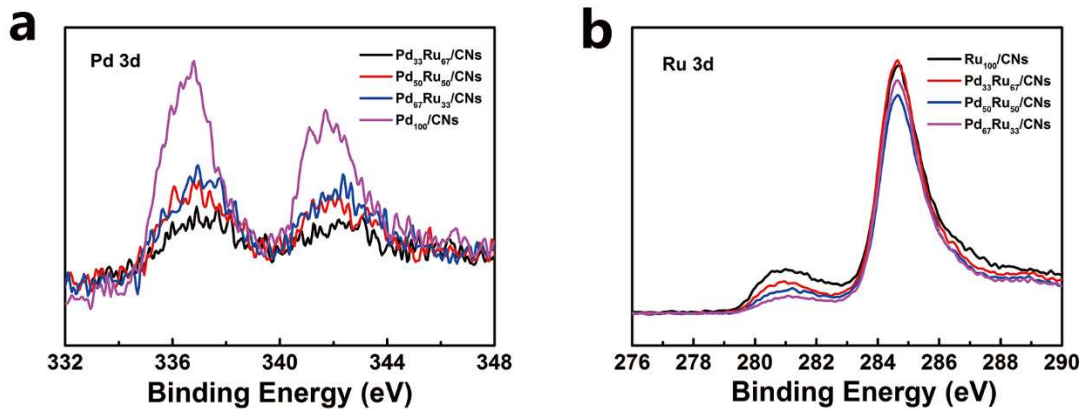


Figure S6. The high-resolution XPS spectra of the (a) Pd3d and (b) Ru3d electrons in the series of samples.

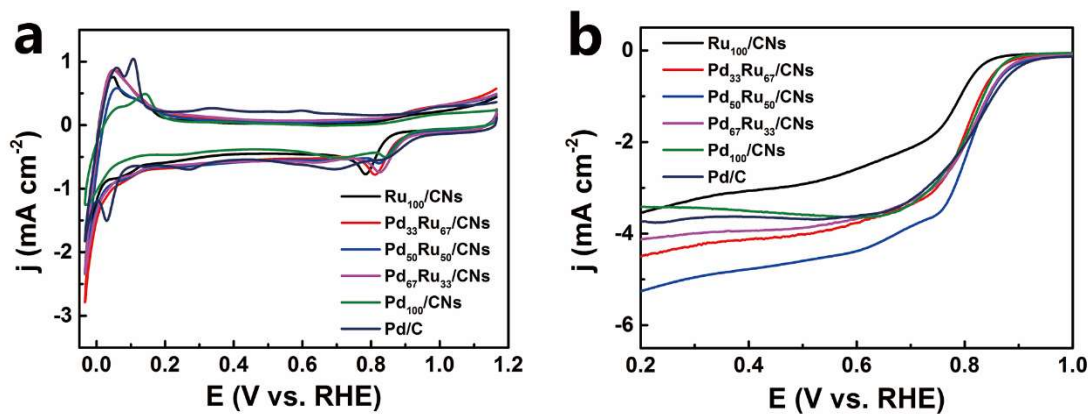


Figure S7. (a) The CV curves and (b) RRDE voltammograms of all the $\text{Pd}_x\text{Ru}_{100-x}/\text{CNs}$ alloyed samples and Pd/C in O_2 -saturated 0.1 M KOH solution.

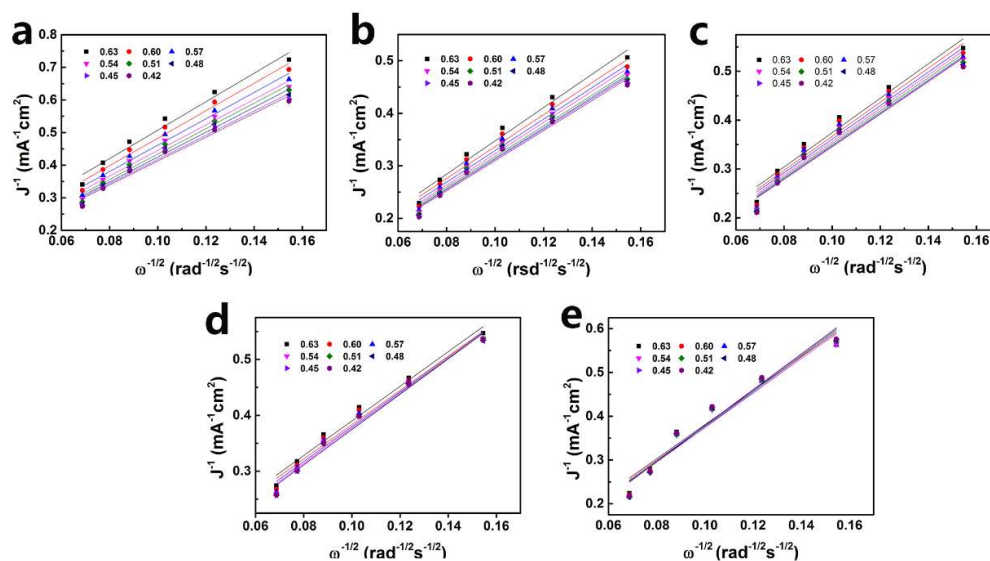


Figure S8. The Koutecky-Levich (K-L) plots of (a) $\text{Ru}_{100}/\text{CNs}$, (b) $\text{Pd}_{33}\text{Ru}_{67}/\text{CNs}$, (c) $\text{Pd}_{67}\text{Ru}_{33}/\text{CNs}$, (d) $\text{Pd}_{100}/\text{CNs}$ and (e) Pd/C.

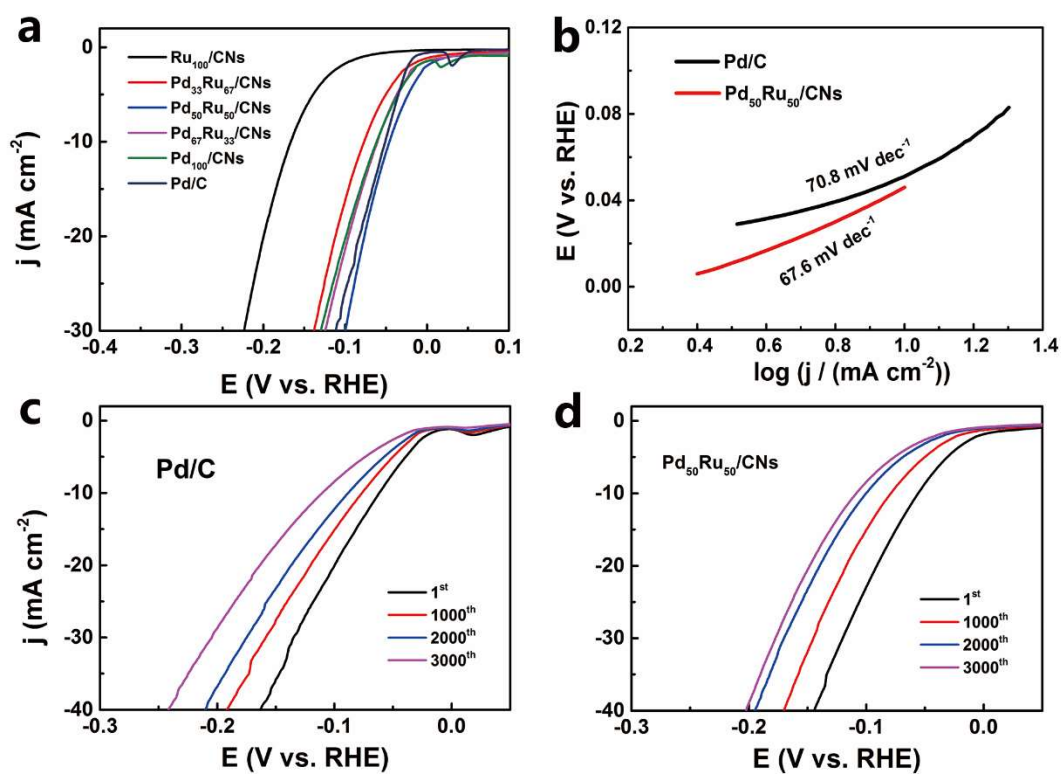


Figure S9. HER activity curves (a) of PdRu alloy CNs and Pd/C in 0.5 M H₂SO₄ with scan rate of 10 mV s⁻¹. The corresponding Tafel plots (b) of the Pd₅₀Ru₅₀/CNs and Pd/C catalyst. Polarization curves after continuous potential sweeps of Pd/C (c) and Pd₅₀Ru₅₀/CNs (d) at 100 mV s⁻¹ in 0.5 M H₂SO₄.