Supporting Information

Achieving high-performance nitrate electrocatalysis with PdCu nanoparticles confined in nitrogen-doped carbon coralline

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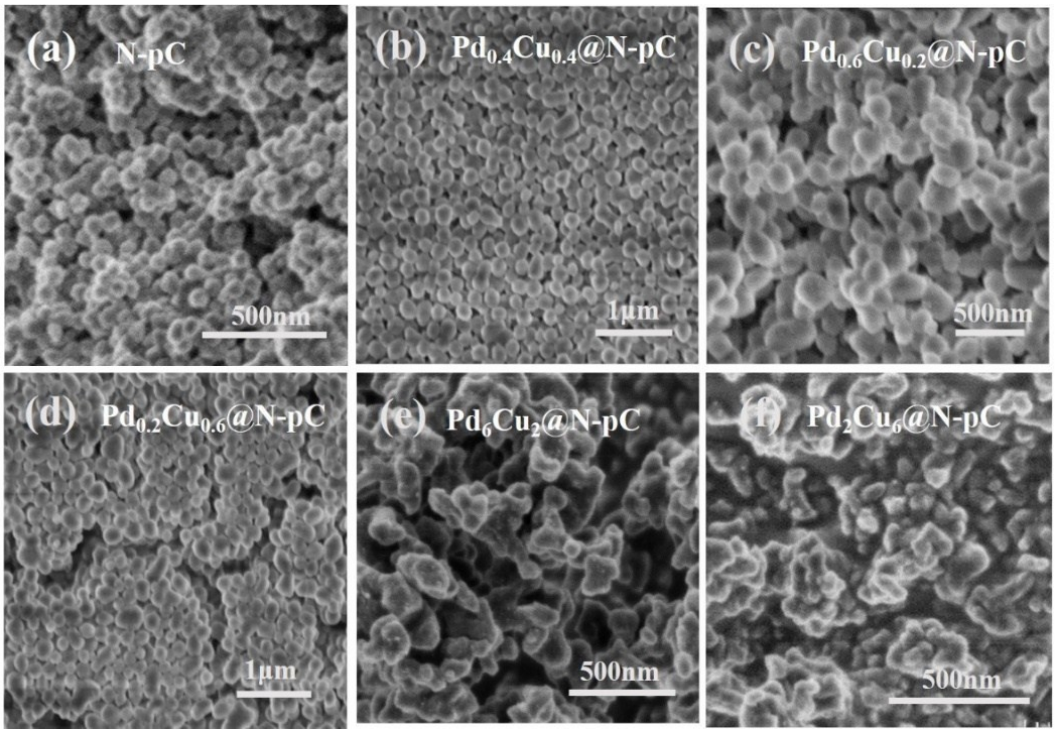

Fig. S1 SEM images of the prepared catalysts with different PdCu alloy loading amount: (a) N-pC: without metal salts; (b) $Pd_{0.4}Cu_{0.4}$@N-pC: 0.4 wt% $PdCl_4^{2+}$ + 0.4 wt% Cu^{2+}; (c) $Pd_{0.6}Cu_{0.2}$@N-pC: 0.6 wt% $PdCl_4^{2+}$ + 0.2 wt% Cu^{2+}; (d) $Pd_{0.2}Cu_{0.6}$@N-pC: 0.2 wt% $PdCl_4^{2+}$ +0.6 wt% Cu^{2+} ; (e) Pd_6Cu_2@N-pC: 6 wt% $PdCl_4^{2+}$ + 2 wt% Cu^{2+}; (f) Pd_2Cu_6@N-pC: 2 wt% $PdCl_4^{2+}$ + 6 wt% Cu^{2+}.

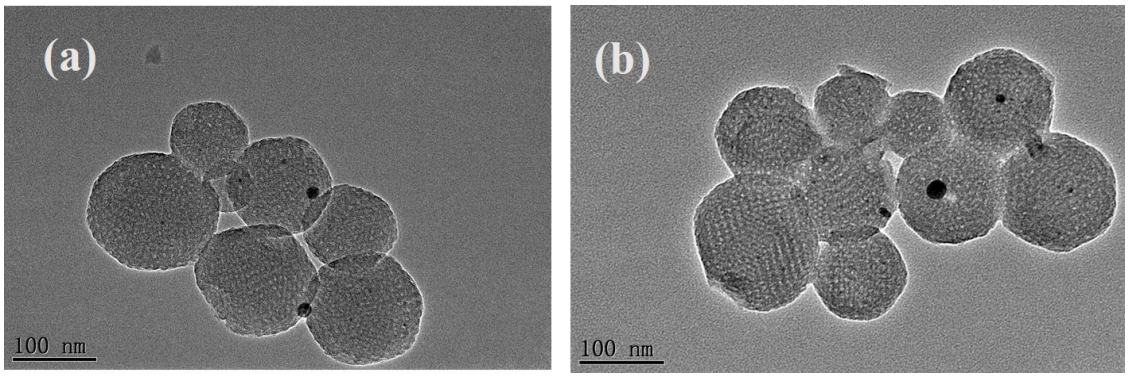

Fig. S2 TEM image of catalyst $Pd_{0.4}Cu_{0.4}$@pC without adding melamine: 4 wt% $PdCl_4^{2+}$ + 4 wt% Cu^{2+} . The preparation process is the same as Pd_4Cu_4@N-pC, except that no melamine is added.

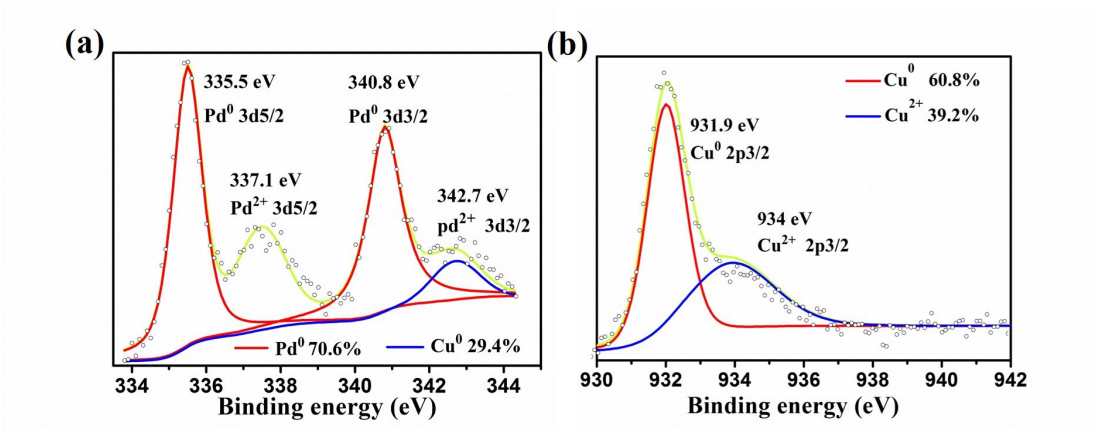

Fig. S3 Pd 3d XPS spectra and (c) Cu 2p XPS spectra of Pd_4Cu_4@N-pC.

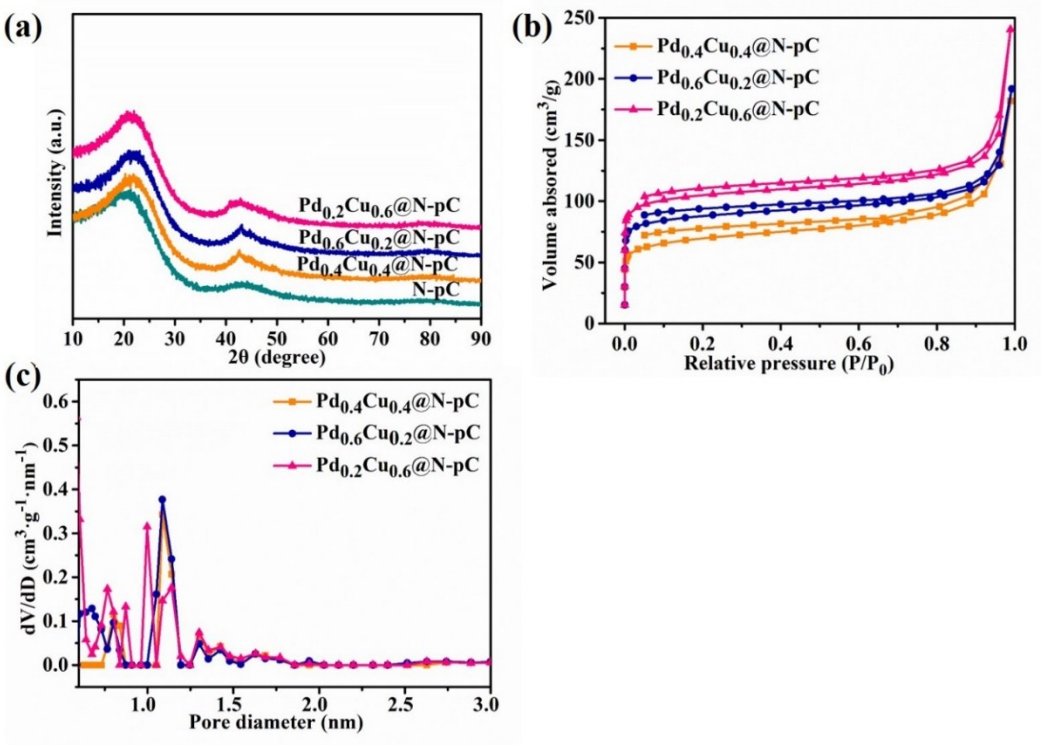

Fig. S4 (a) XRD patterns, (b) nitrogen sorption isotherms, (c) pore size distribution curves of $Pd_{0.4}Cu_{0.4}$@N-pC, $Pd_{0.6}Cu_{0.2}$@N-pC, and $Pd_{0.2}Cu_{0.6}$@N-pC.

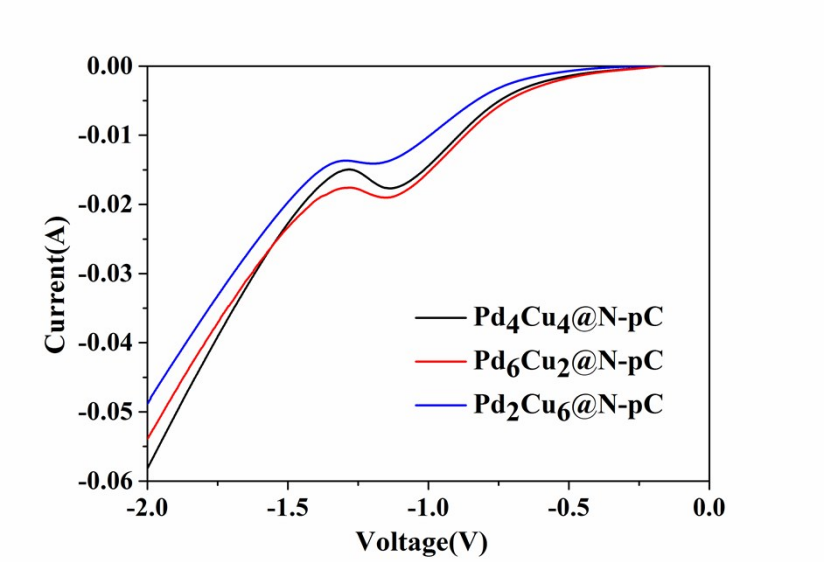

Fig. S5 LSV patterns of Pd_4Cu_4@N-pC 、 Pd_6Cu_2@N-pC. Pd_2Cu_6@N-pC.

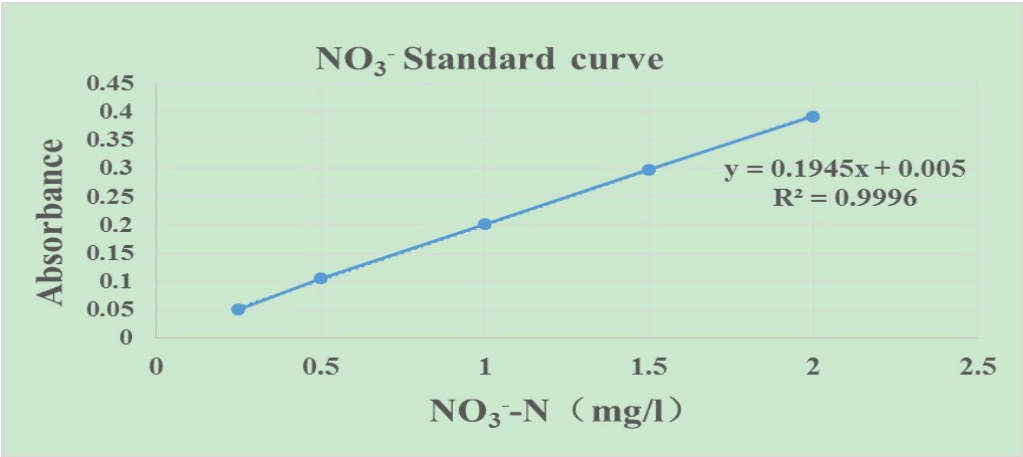

Fig. S6 Standard curve for NO_3^- to calculate the concentration of electrolyte after electrocatalysis.

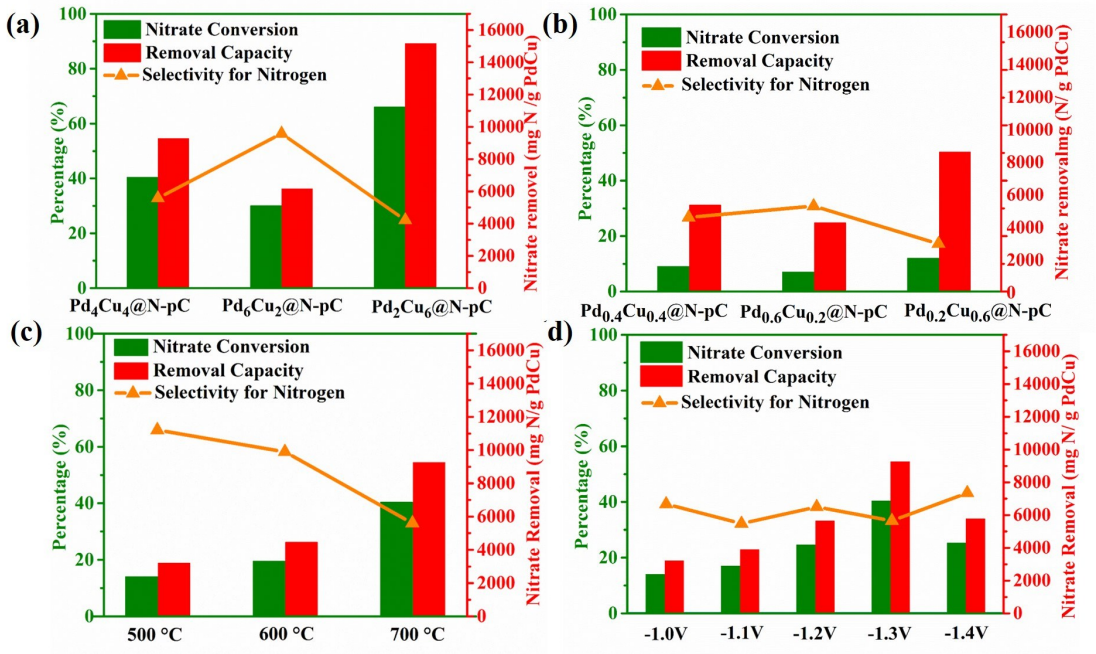

Fig. S7 The results including nitrate conversion, removal capacity and selectivity for nitrogen of nitrate reduction of (a) Pd_4Cu_4@N-pC, Pd_6Cu_2@N-pC, and Pd_2Cu_6@N-pC, and (b) $Pd_{0.4}Cu_{0.4}$@N-pC, $Pd_{0.6}Cu_{0.2}$@N-pC, and $Pd_{0.2}Cu_{0.6}$@N-pC; the detail investigation the nitrate electro-catalysis ability of Pd_4Cu_4@N-pC coralline-like nanostructures (c) at different calcination temperatures of 500 °C, 600 °C, and 700 °C, and (d) under different reduction voltage of -1.0 V, -1.1 V, -1.2 V, -1.3 V and -1.4 V. All composites were reduced after 12 h at -1.3 V.

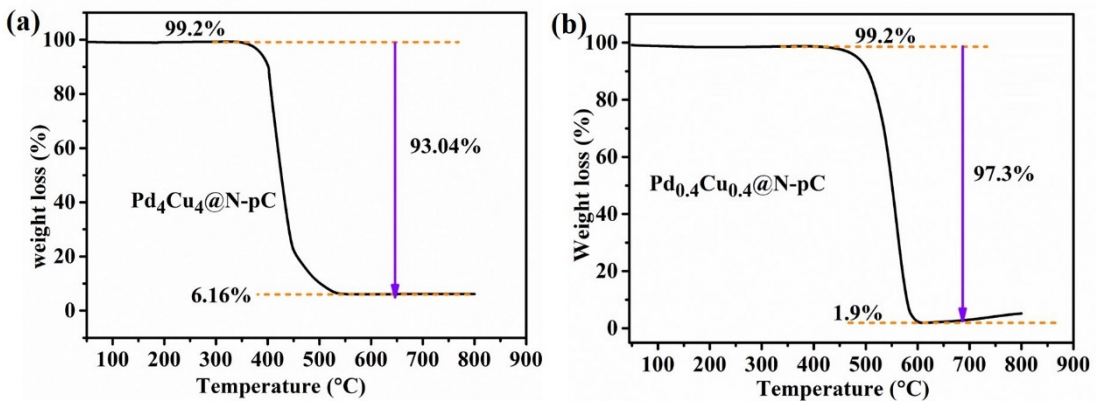

Fig. S8 Thermal gravimetric analysis (TGA) curves of (a) Pd_4Cu_4@N-pC and (b) $Pd_{0.4}Cu_{0.4}$@N-pC, respectively.

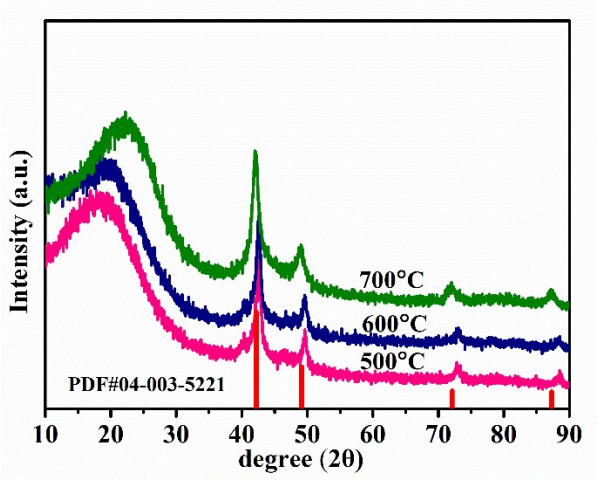

Fig. S9 XRD patterns of Pd_4Cu_4@N-pC coralline nanostructures obtained at different calcination temperatures of 500 °C, 600 °C, and 700 °C.

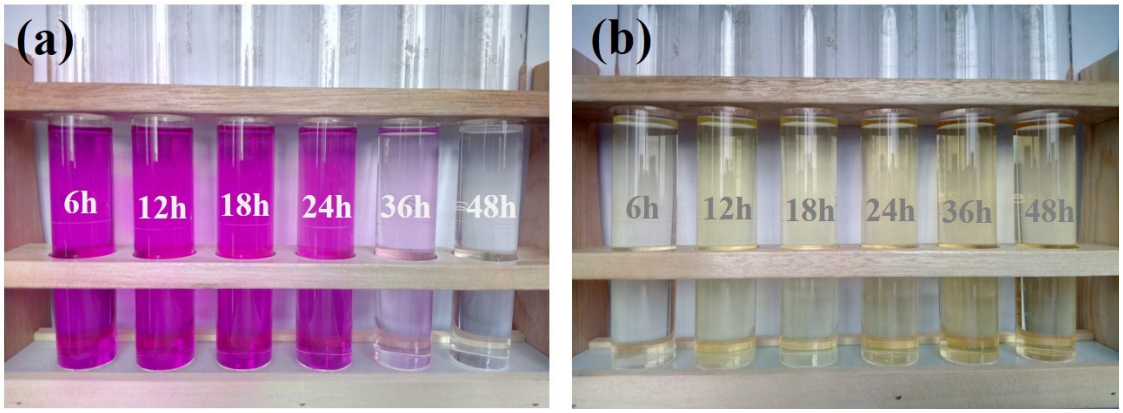

Fig. S10 Testing the respective solutions of NO_2^- and NH_4^+ from electrocatalytic corresponding points from 6 h to 48 h .

Table S1: Physicochemical properties of Pd_xCu_y@N-pC coralline nanostructures.

Sample	Pore diameter[a] (nm)	Pore volume[b] ($cm^3\ g^{-1}$)	S_{BET}[c] ($m^2\ g^{-1}$)	S_{micro}[d] ($m^2\ g^{-1}$)
N-pC	1.97	0.26	524.1	440.1
Pd_4Cu_4@N-pC	2.28	0.25	444.8	368.3
Pd_6Cu_2@N-pC	2.6	0.21	470.6	372.7
Pd_2Cu_6@N-pC	2.99	0.19	455.2	338.1
$Pd_{0.4}Cu_{0.4}$@N-pC	3.02	0.17	318.8	241.4
$Pd_{0.6}Cu_{0.2}$@N-pC	2.25	0.18	323.9	256
$Pd_{0.2}Cu_{0.6}$@N-pC	2.63	0.21	323.6	248.5

[a] Pore diameter were obtained by using Barrett-Joyner- Halenda (BJH) model from the adsorption branches of isotherms.
[b] Pore volume were evaluated at a relative pressure P/P_0 of 0.995.
[c] BET surface areas, S_{BET}, were determined by common Brunauer-Emmett-Teller (BET) method.
[d] T-plot micropore surface.

Table S2. Leaching atomic concentration of Pd_4Cu_4@N-pC electrode for electro-catalysis test at the respective electrocatalytic time points.

Reaction time (h)	Cu concentration (ppb)	Pd concentration (ppb)
6	0.15	0.03
12	0.15	0.03
18	0.07	0.02
24	0.08	0.03
36	0.05	0.01
48	0.06	0.02