Supporting Information

Guanine-derived carbon nanosheets encapsulated Ni nanoparticles for efficient CO_2 electroreduction

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Supplementary Figures

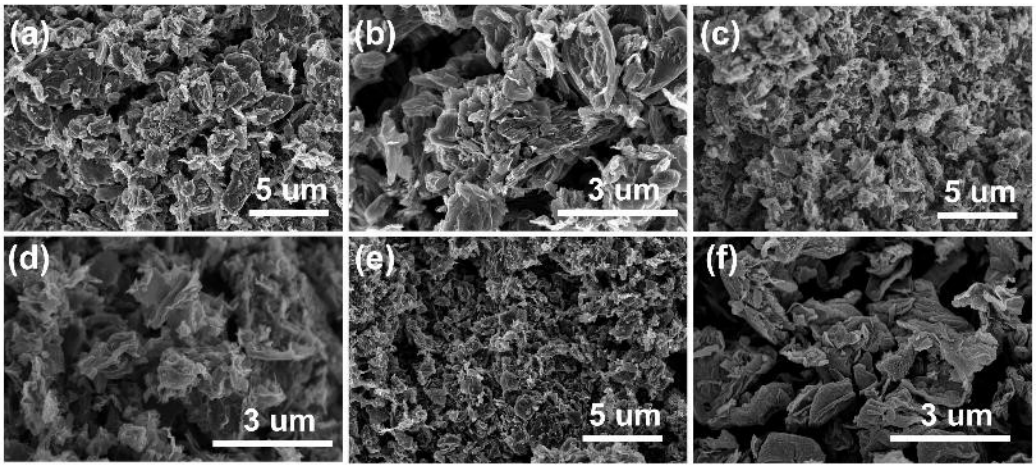

Fig. S1. SEM images of (a,b) Ni@NC-900, (c,d) Ni@NC-1100 and (e,f) NC-1000.

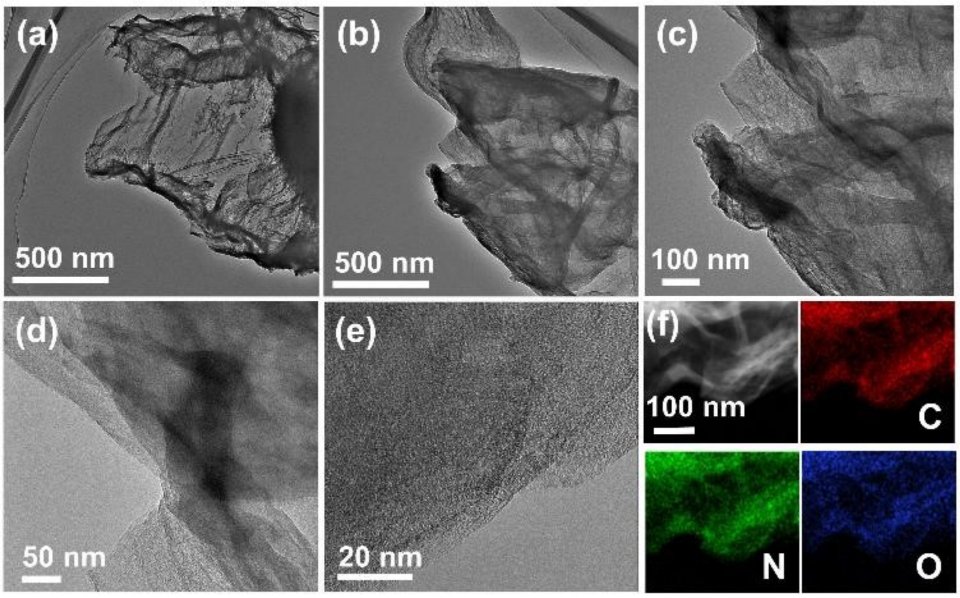

Fig. S2. TEM images of NC-1000.

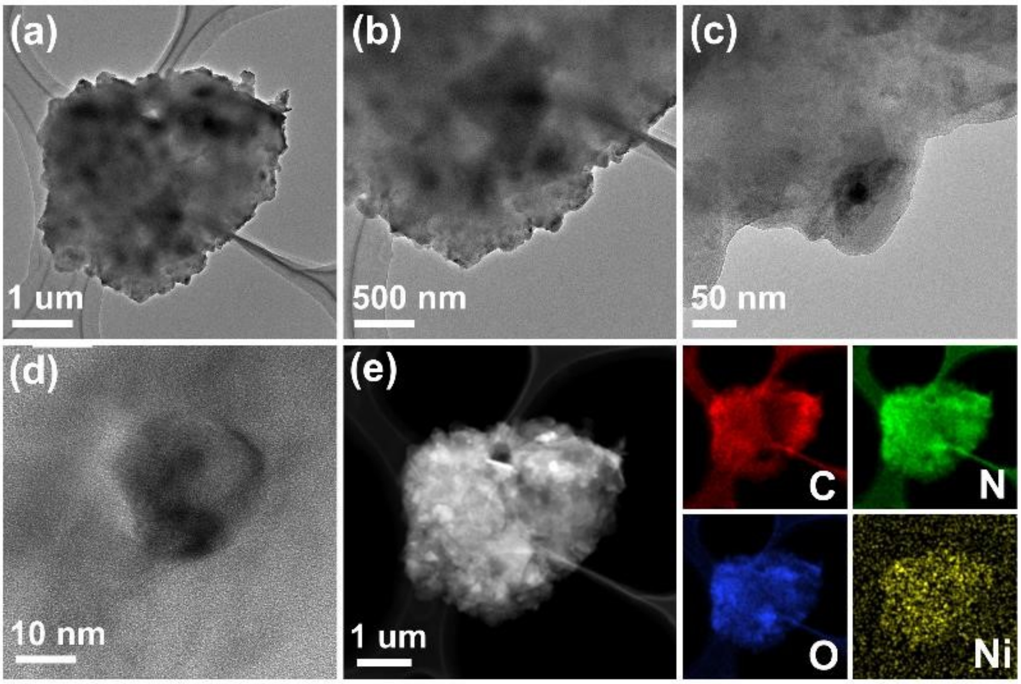

Fig. S3. TEM images of Ni@NC-900.

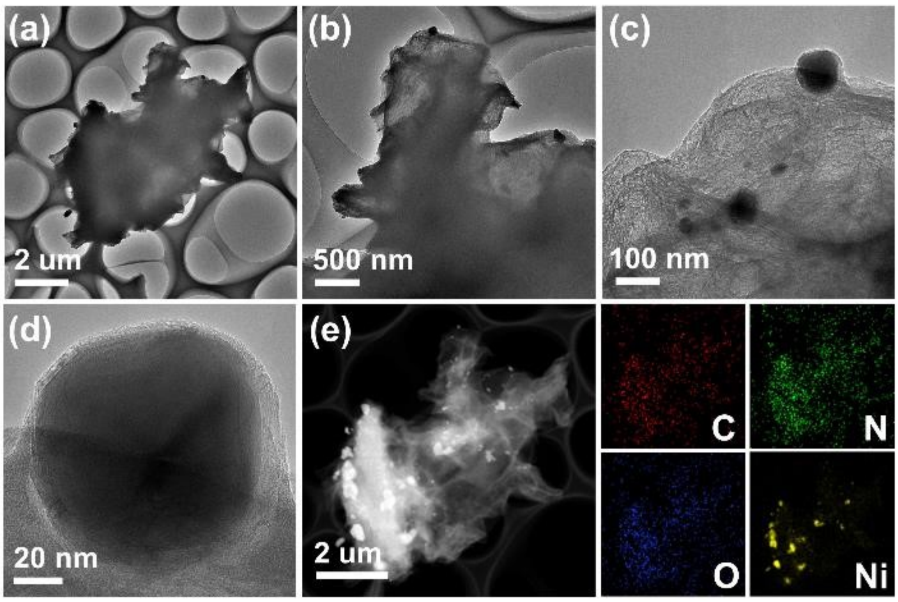

Fig. S4. TEM images of Ni@NC-1100.

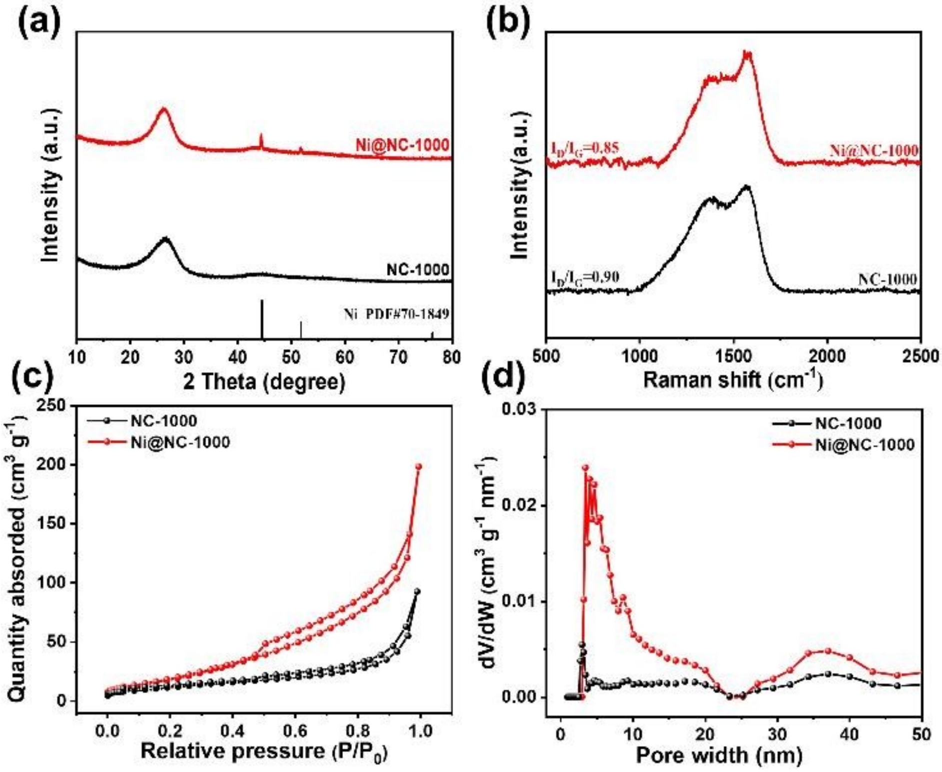

Fig. S5. (a) XRD spectra. (b) Raman spectra. (c) N_2 adsorption-desorption isotherms. (d) Pore size distribution obtained by the BJH method of Ni@NC-1000 and NC-1000.

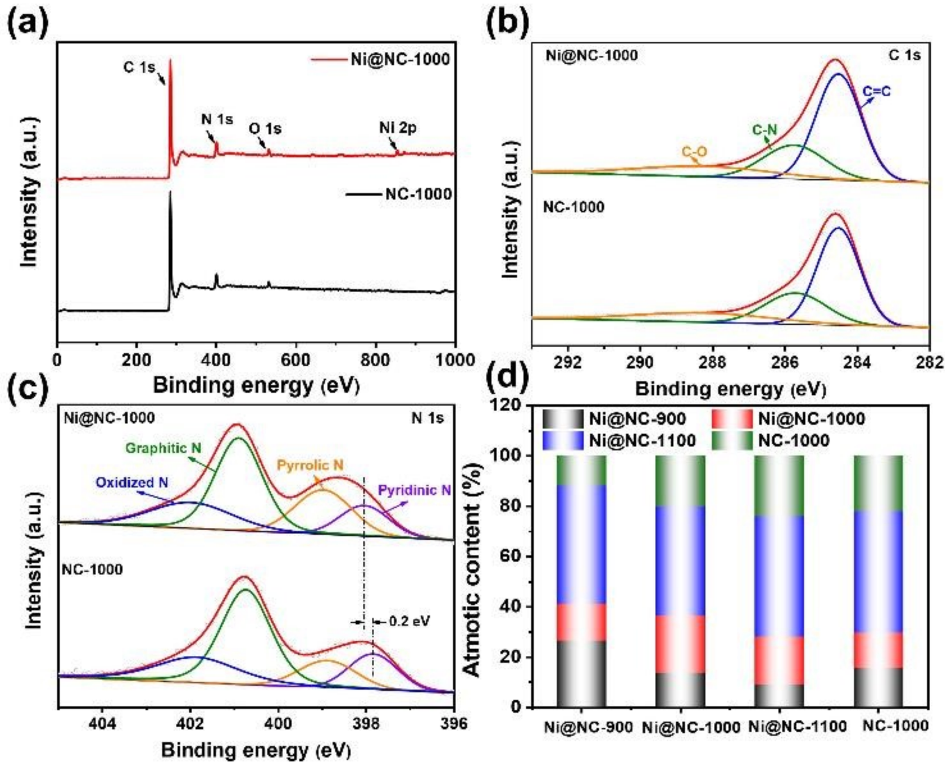

Fig. S6. (a) XPS survey spectra. (b) High-resolution C 1s XPS spectra. (c) High-resolution N 1s XPS spectra. (d) The relative contents of different-type N species in various catalysts.

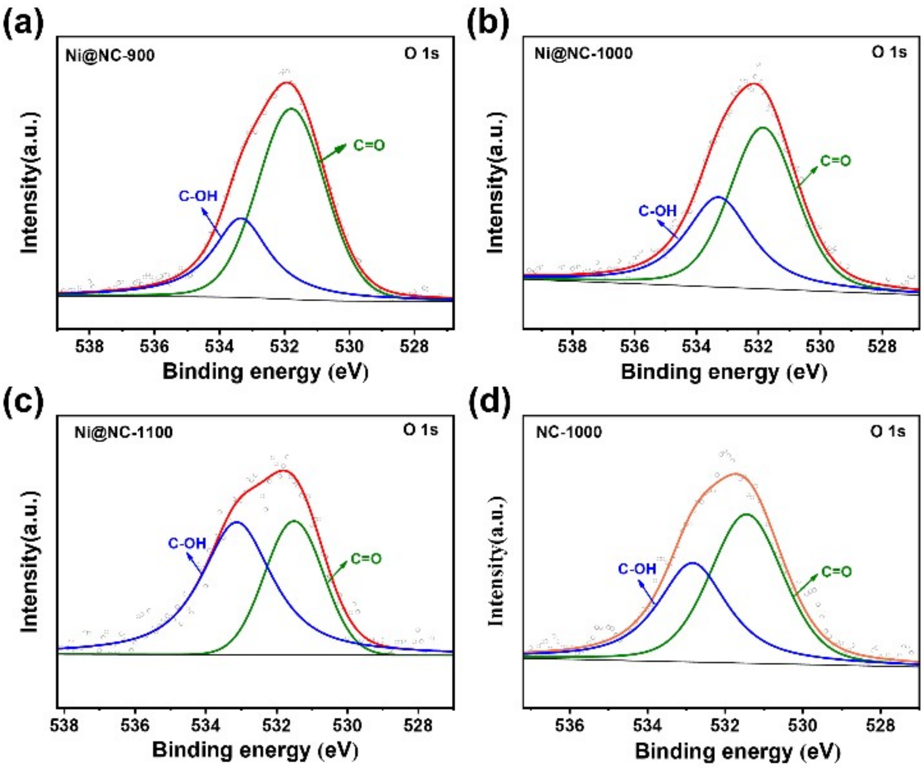

Fig. S7. High-resolution O 1s XPS spectra of (a) Ni@NC-900, (b) Ni@NC-1000, (c) Ni@NC-1100, (d) NC-1000.

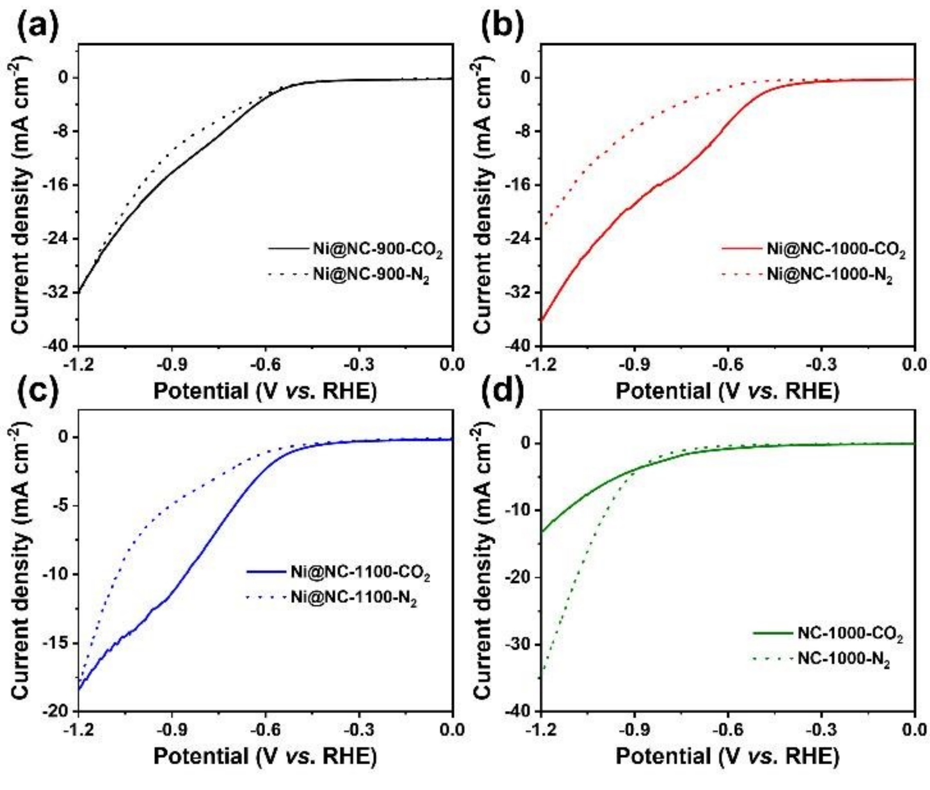

Fig. S8. LSV curves of (a) Ni@NC-900, (b) Ni@NC-1000, (c) Ni@NC-1100 and (d)NC-1000 in N_2/CO_2-saturated 0.5 M $NaHCO_3$.

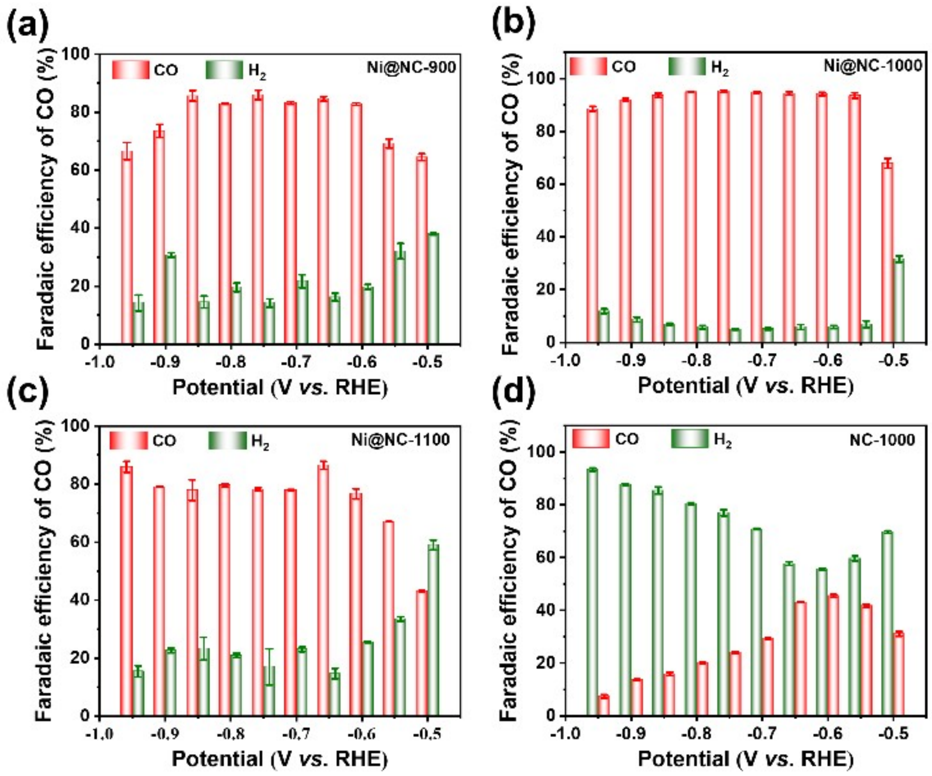

Fig. S9. FE_{CO} and FE_{H2} of (a) Ni@NC-900, (b) Ni@NC-1000, (c) Ni@NC-1100, (d) NC-1000 at different applied potentials in CO_2-saturated 0.5 M $NaHCO_3$.

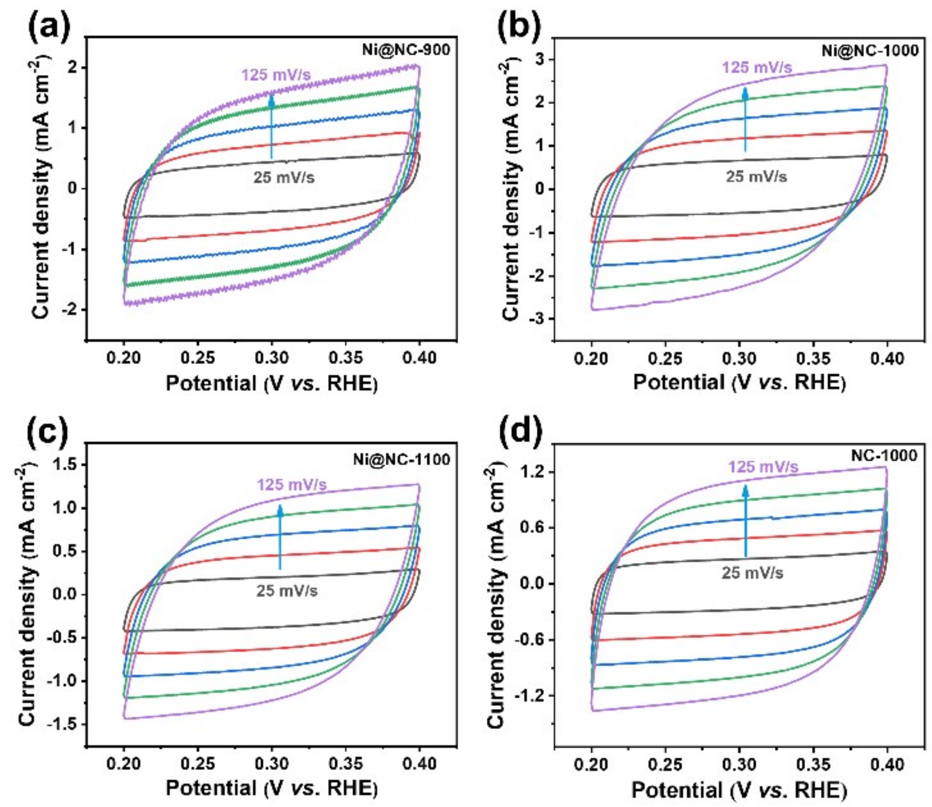

Fig. S10. Cyclic voltammetry curves of (a) Ni@NC-900, (b) Ni@NC-1000, (c) Ni@NC-1100, (d) NC-1000 in 0.5 M CO_2-saturated $NaHCO_3$ solution.

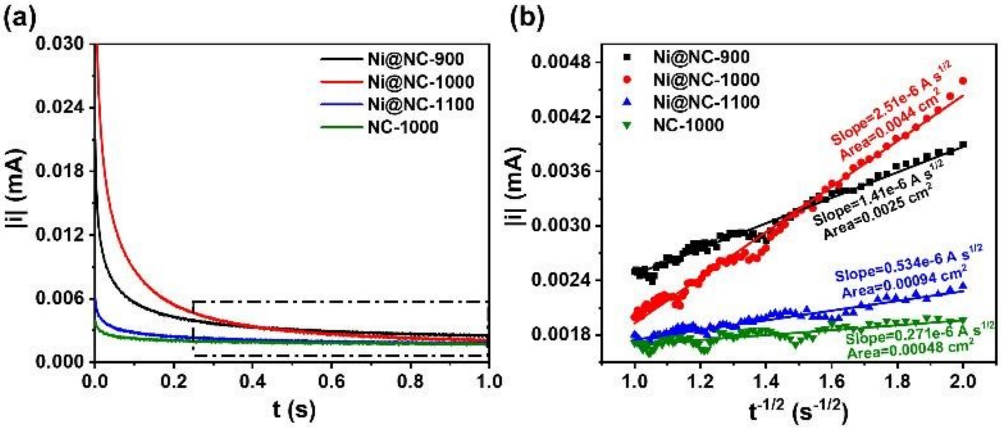

Fig. S11. (a) Chronoamperometric response recorded in 0.1 M KCl (containing 5 mM $K_3Fe(CN)_6$) after stepping the potential from 0.8 to 0.1 V *vs.* Ag/AgCl over Ni@NC-X and NC-1000. (b) Linearized plot based on the Cottrell equation for Ni@NC-X and NC-1000 (The first 250 ms were omitted because of double layer charging effects)

The ECSAs of all samples were obtained using the Cottrell equation based on the literature.[1, 2] The Cottrell equation is as follows:

$$i = \frac{nFAC\sqrt{D}}{\sqrt{\pi t}}$$

where i is the current, n = 1, D = 4.34 × 10^{-6} cm^2 s^{-1}, F = 96485 C mol^{-1}, A is ECSA, and C is the concentration of $K_3Fe(CN)_6$ (5 mM).

The specific testing method is as follows: Firstly, We applied the potential from 0.8 to 0.1 V (*vs.* Ag/AgCl) in 0.1 M KCl (containing 5 mM $K_3Fe(CN)_6$) purged with N_2 and recorded the chronoamperometric response within 1 s, as shown in Fig. R8 (a). Then we ploted a graph of i and $t^{-1/2}$ and fit it to obtain the slope, as shown in Fig. R8 (b). The ECSAs of all catalysts can be calculated by slope. According to the results, the ECSAs of Ni@NC-900, Ni@NC-1000, Ni@NC-1100 and NC-1000 were assessed to be 0.0025, 0.0044, 0.00094 and 0.00048 cm^2, respectively.

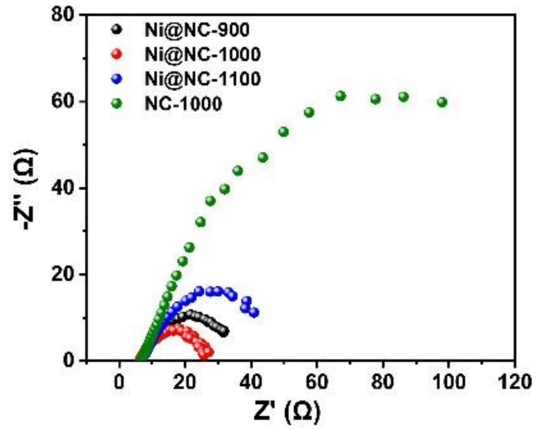

Fig. S12. The EIS plots of Ni@NC-X and its comparison sample NC-1000.

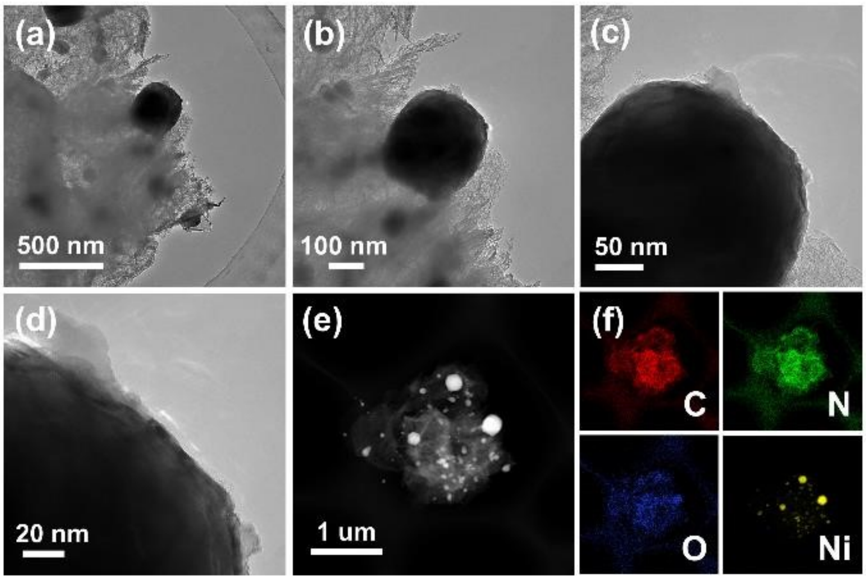

Fig. S13. TEM images of spent Ni@NC-1000.

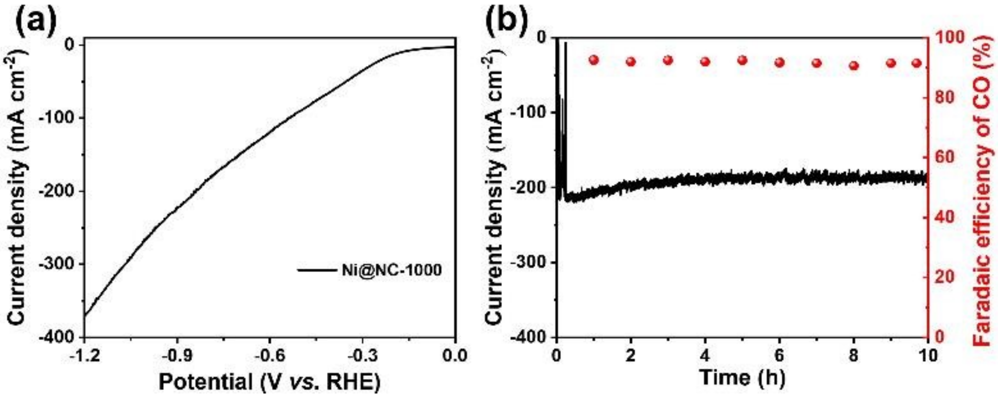

Fig. S14. Flow cell measurement of Ni@NC-1000 at -0.9 V (vs. RHE) in 1 M KOH.

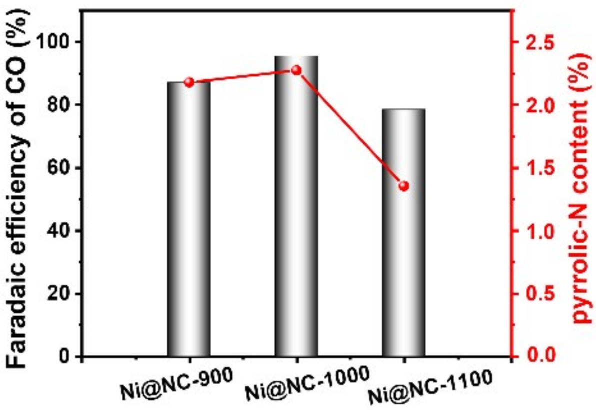

Fig. S15. The relationship between the content of pyrrolic N in catalysts and the corresponding CO_2RR activity.

Supplementary Tables

Table S1. Summary of BET results for Ni@NC-X and NC-1000 catalysts.

Catalyst	BET Surface Area (m^2/g)	Adsorption Pore Diameter (nm)	Pore Volume (cm^3/g)
Ni@NC-900	63	9.9	0.16
Ni@NC-1000	77	10.1	0.31
Ni@NC-1100	65	10.2	0.18
NC-1000	44	12.1	0.14

Table S2. The contents of carbon, nitrogen, oxygen and nickel elements in various catalysts.

Catalyst	C wt%	N wt%	O wt%	Ni wt%
Ni@NC-900	81.88	14.17	3.45	0.5
Ni@NC-1000	87.66	9.41	2.38	0.56
Ni@NC-1100	90.95	6.7	1.75	0.6
NC-1000	88.54	8.71	2.5	-

Table S3. The relative contents of four major N species in various catalysts.

N Species / Catalyst	Pyridinic N (%)	Pyrrolic N (%)	Graphitic N (%)	Oxidized N (%)
Ni@NC-900	26.5	14.5	47.1	11.9
Ni@NC-1000	13.6	22.8	43.4	20.2
Ni@NC-1100	8.9	19.1	47.8	24.2
NC-1000	15.5	14	48.3	22.2

Table S4. Summary of CO_2RR performance from recent literature

Catalyst	Electrolyte	FE_{CO} (%)	Potential (V vs. RHE)	j_{CO} (mA cm^{-2})	Ref
Ni@NC-1000	0.5 M $NaHCO_3$	95.6	-0.75	12.3	This work
Ni-NCN	0.5 M $KHCO_3$	96.6	-0.83	9.8	[3]
Ni-SAC@NC	0.5 M $KHCO_3$	95.0	-0.6	5.7	[4]
NiSA@N_3-C	0.5 M $KHCO_3$	96	-0.83	18.87	[5]
Ni-NC/NHCSs-600	0.5 M $NaHCO_3$	98.57	-0.87	14.2	[6]
Ni@N-C	0.5 M $KHCO_3$	90	-0.8	17	[7]
Ni@N-C/rGO 4,4'-bipy	0.5 M $KHCO_3$	88	-0.97	20	[8]
Ni-NC(HPU)	0.5 M $KHCO_3$	91	-0.8	24.7	[9]
SA-NiNG-NV	0.5 M $KHCO_3$	96	-0.7	10	[10]
NC-CNTs (Ni)	0.1 M $KHCO_3$	90	-0.8	7.5	[11]
Ni, N-C-800	0.1 M $KHCO_3$	94.8	-0.86	18.2	[12]
Ni-N-C-NH_4Cl	0.5 M $KHCO_3$	98	-0.62	8.5	[13]
Ni SAs	0.5 M $KHCO_3$	97	-0.8	6.8	[14]

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