Supporting Information

Pyridine-grafted nitrogen-doped carbon nanotubes achieving efficient electroreduction of CO_2 to CO within a wide electrochemical window

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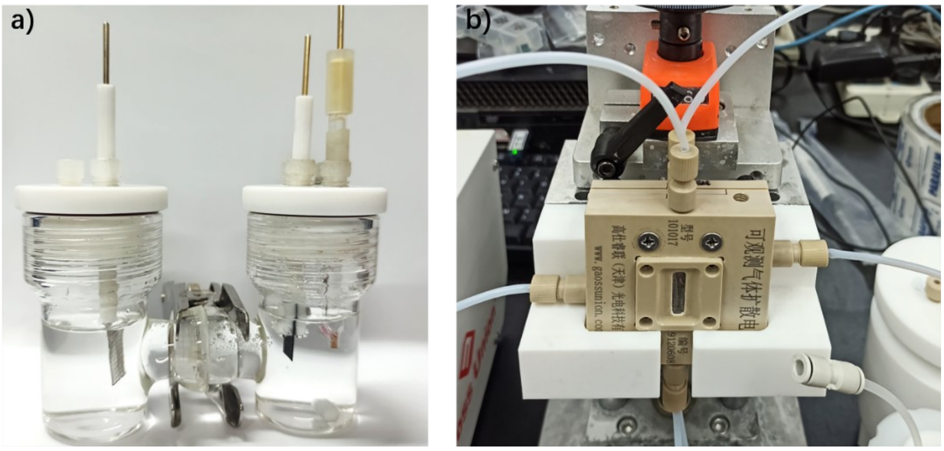

Fig. S1. Pictures of (a) H-type cell and (b) flow cell with an observable gas diffusion electrode.

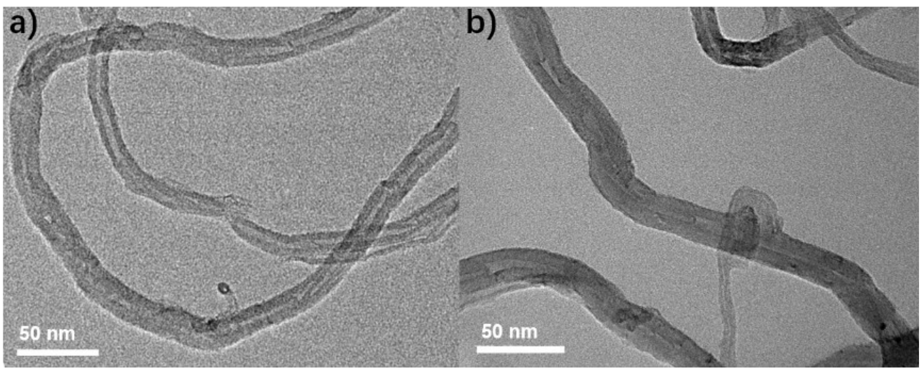

Fig. S2. TEM images of (a) N_4CNTs-800 and (b) Py-N_4CNTs-800.

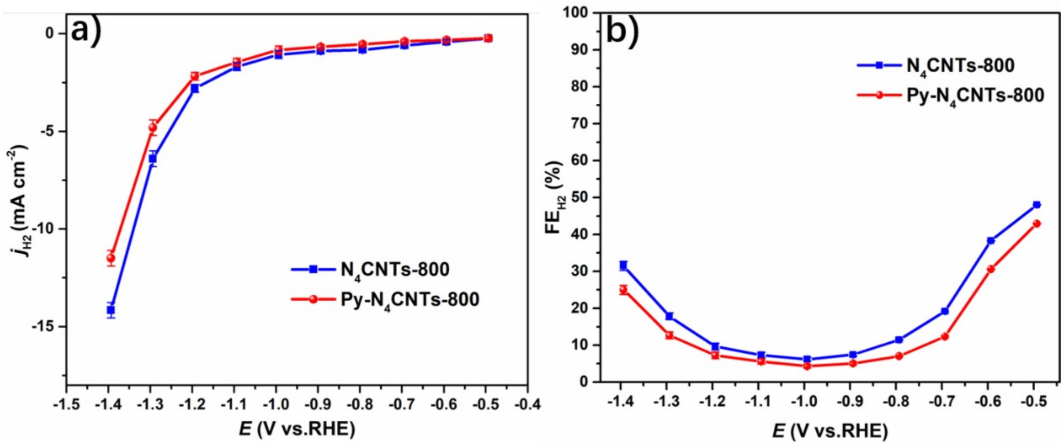

Fig.S3. (a) Partial current densities and (b) faradaic efficiencies of H_2 on N_4CNTs-800 and Py-N_4CNTs-800 at different potentials.

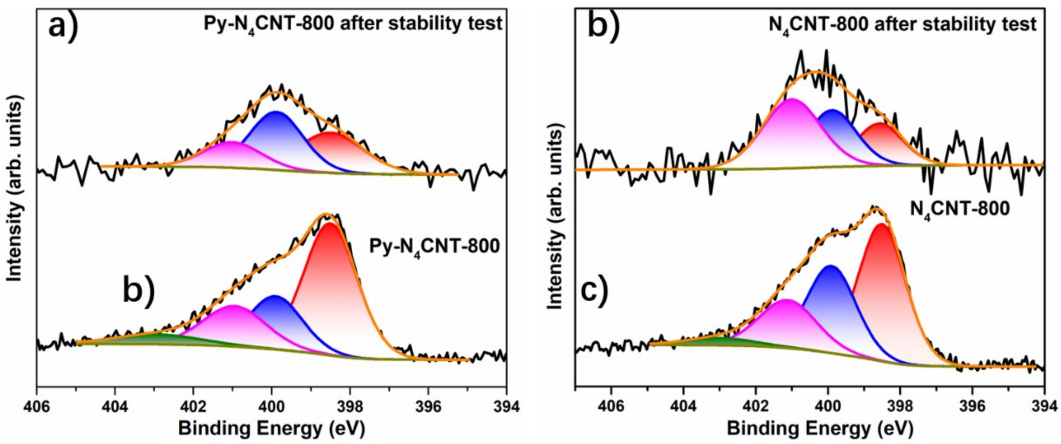

Fig.S4. (a) N 1s XPS spectra of Py-N_4CNTs-800 before and after the stability test. (b) N 1s XPS spectra of N_4CNTs-800 before and after stability test.

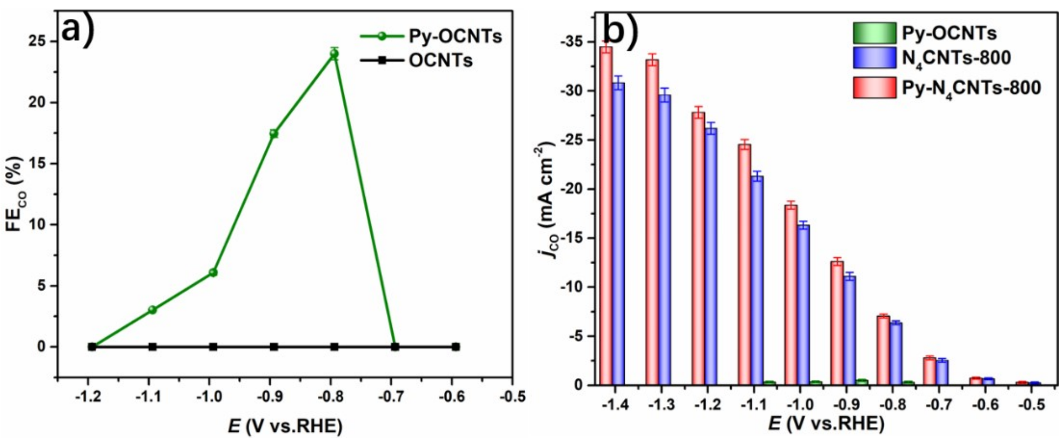

Fig. S5. (a) Faradaic efficiencies of CO on OCNTs and Py-OCNTs at different potentials. (b) Partial current densities of CO on Py-OCNTs, N_4CNTs-800 and Py-N_4CNTs-800 at different potentials.

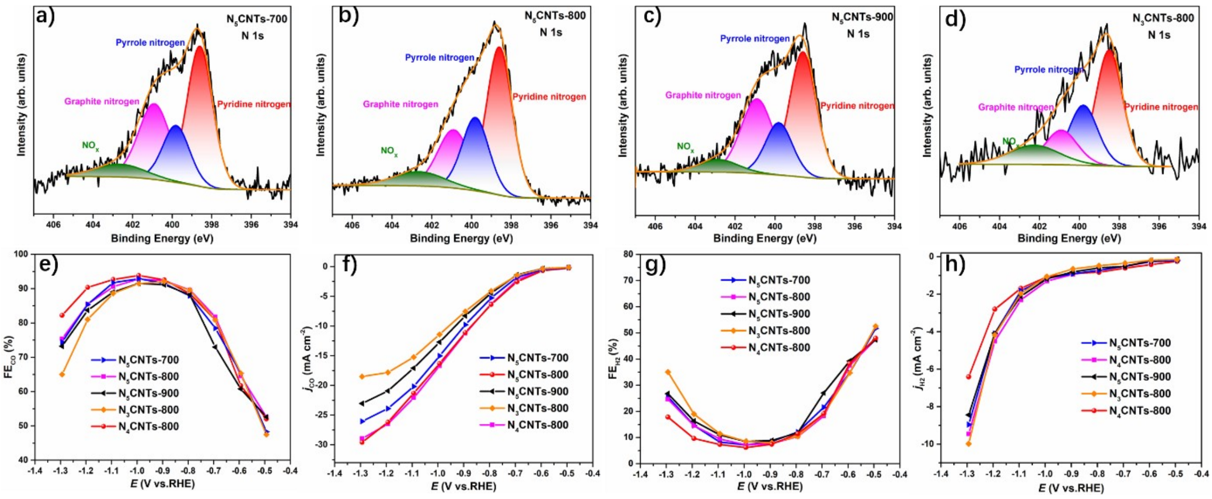

Fig. S6. (a) N 1s XPS spectra of N_5CNTs-700, (b) N_5CNTs-800 (c)N_5CNTs-900 and (d) N_3CNTs-800. (e)Faradaic efficiencies of CO, (f) partial current densities of CO, (g) faradaic efficiencies of H_2 and (h) partial current densities of H_2 on N_4CNTs-800, N_5CNTs-700, N_5CNTs-800, N_5CNTs-900 and N_3CNTs-800 at different potentials.

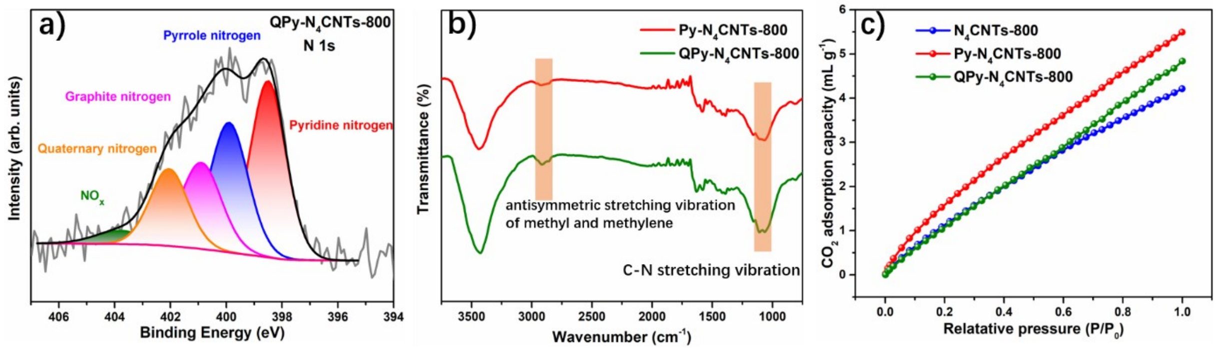

Fig. S7. (a) N 1s XPS spectrum of QPy-N_4CNTs-800. (b) FT-IR spectra Py-N_4CNTs-800 and QPy-N_4CNTs-800. (c) CO_2 adsorption isotherms of N_4CNTs-800, Py-N4CNTs-800 and QPy-N_4CNTs-800.

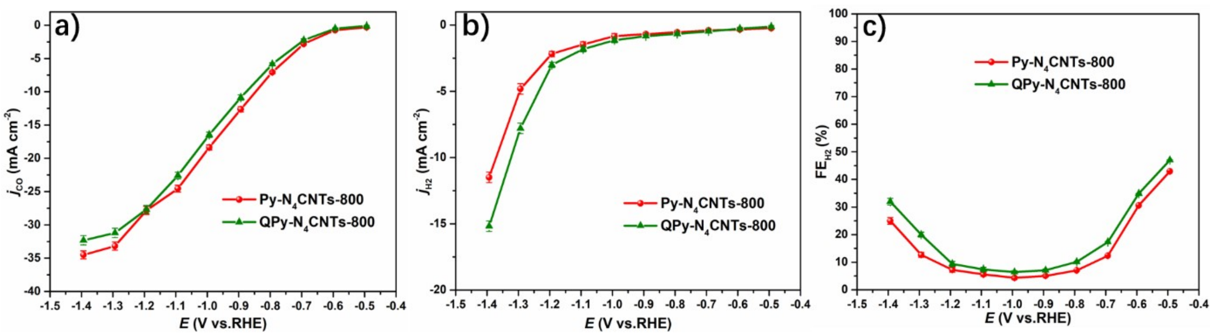

Fig. S8. (a) Partial current densities of CO, (b) partial current densities of H_2 and (c) faradaic efficiencies of H_2 on Py-N_4CNTs-800 and QPy-N_4CNTs-800 at different potentials.

Table S1. The types and content of nitrogen in the prepared catalysts from XPS spectra.

Samples	N 1s (at.%)	Pyridine N (at.%)	Pyrrole N (at.%)	Graphite N (at.%)	NOx (at.%)	Quaternary N (at.%)
N_4CNTs-800	4.37	2.04	1.29	0.87	0.17	/
N_3CNTs-800	3.26	1.46	0.81	0.52	0.46	/
N_5CNTs-700	4.26	1.88	0.80	1.26	0.33	/
N_5CNTs-800	4.45	2.02	1.05	0.99	0.38	/
N_5CNTs-900	3.98	1.75	0.73	1.25	0.25	/
Py-N_4CNTs-800	5.38	2.85	1.15	1.03	0.34	/
QPy-N_4CNTs-800	5.35	1.84	1.44	1.02	0.20	0.86

Table S2. Summary of electrocatalysts studied for CO_2ER to CO.

Samples	Electrolyte	Potential (V vs. RHE)	FE_{CO} (%)	j_{CO} (mA cm^{-2})	Stability (h)	FE_{CO}>90 (V vs. RHE)	Ref.
Py-N_4CNTs-800	0.1 M $KHCO_3$	-0.99	96%	18.4	19.3	-0.79 ~ -1.19	This work
NSHCF900	0.1 M $KHCO_3$	-0.7	94%	~96	36	-0.7	7
N-GRW	0.5 M $KHCO_3$	-0.40	87.6%	~3.08	10	/	16
CNPC-1100	0.1 M $KHCO_3$	-0.6	92%	~0.8	8	-0.6	17
1D/2D NR/CS-900	0.5 M $KHCO_3$	-0.45	94.2%	~1	30	-0.45 ~ -0.55	24
NCNT-NH_3	0.5 M $NaHCO_3$	-0.8	94.5%	~14	40	-0.6 ~ -0.9	26
NF-C-950	0.1 M $KHCO_3$	-0.6	90%	1.9	40	-0.6	29
NCNT-3-700	0.5 M $NaHCO_3$	-0.9	90%	5	60	-0.9	30
NPC-900	0.5 M $KHCO_3$	-0.67	95%	2.3	10	-0.67	31
DG	0.1 M $KHCO_3$	-0.6	84%	1.3	10	/	32
NDAPC	0.1 M $KHCO_3$	-0.9	83%	~3.8	8	/	33
DPC-NH_3-950	0.1 M $KHCO_3$	-0.6	95.2%	2.71	24	-0.5 ~ -0.7	34
NS-CNSs	0.5 M $KHCO_3$	-0.55	85.4%	2.4	20	/	35
WNCNs	0.1 M $KHCO_3$	-0.6	84%	1.15	8	/	36
NS-C-900	0.1 M $KHCO_3$	-0.6	92%	2.63	20	-0.6	37
NG-800	0.1 M $KHCO_3$	-0.58	85%	~1.53	5	/	55
SZ-HCN	0.1 M $KHCO_3$	-0.6	93%	~4.65	20	-0.6	56
NRMC-900	0.1 M $KHCO_3$	-0.6	80%	2.9	10	/	57
D-C-1100	0.1 M $KHCO_3$	-0.6	94.5%	~0.9	10	-0.6 ~ -0.7	58
CN-H-CNTs	0.1 M $KHCO_3$	-0.5	88%	~0.4	7	/	59
Ag/CNT COOH	0.1 M $KHCO_3$	-1.1	83%	~17	10	/	60
AgNPs@PAM	0.1 M $KHCO_3$	-0.89	97.2%	~22	48	-0.79 ~ -1.09	61
Au/Py-CNTs-O	0.1 M $KHCO_3$	-0.59	93%	~7	10	-0.58 ~ -0.98	62
H-Zn-NPs	0.1 M $KHCO_3$	-0.96	94.2%	~5	12	-0.76 ~ -1.06	63