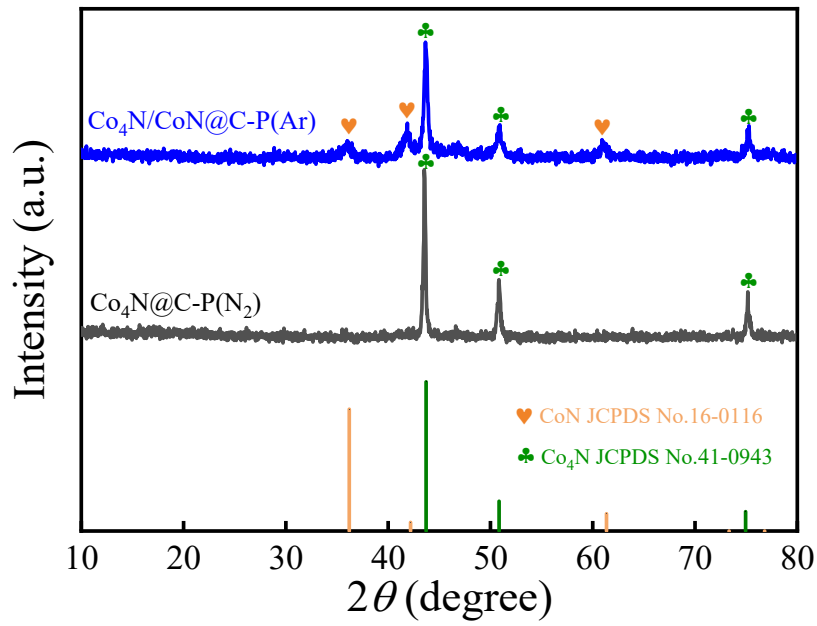


## **Supporting Information**

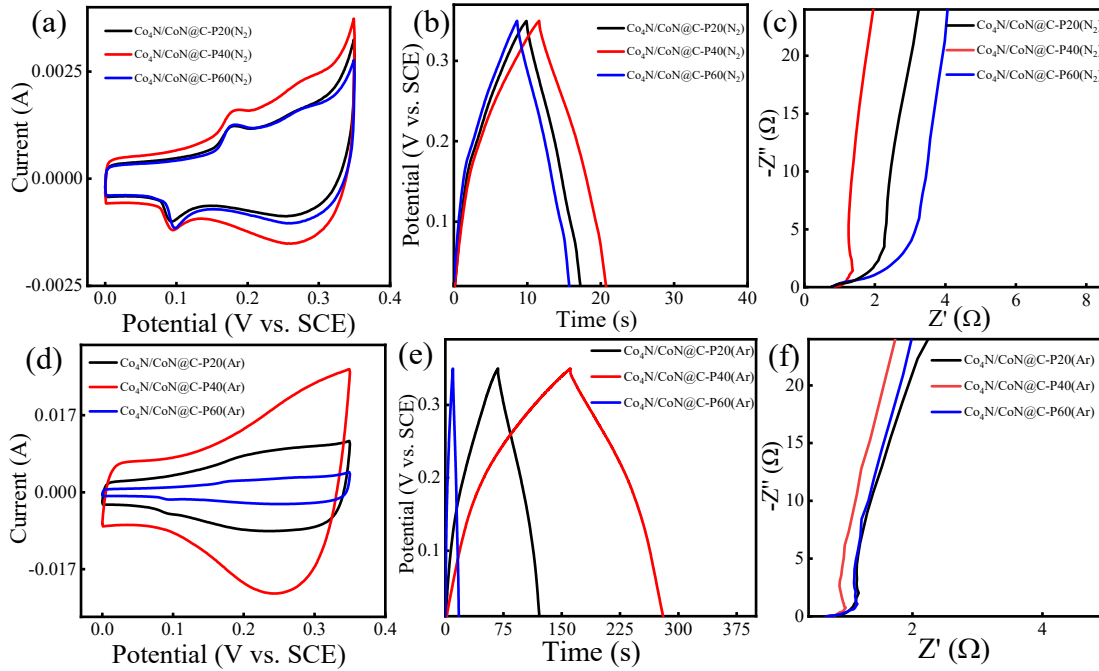
### **Plasma engineering of Co<sub>4</sub>N/CoN heterostructure for boosting supercapacitor performance**

Hong Li <sup>1</sup>, Yunzhe Ma <sup>1</sup>, Xulei Zhang <sup>2</sup>, Xiuling Zhang <sup>1</sup> and Lanbo Di <sup>1,3,4,\*</sup>

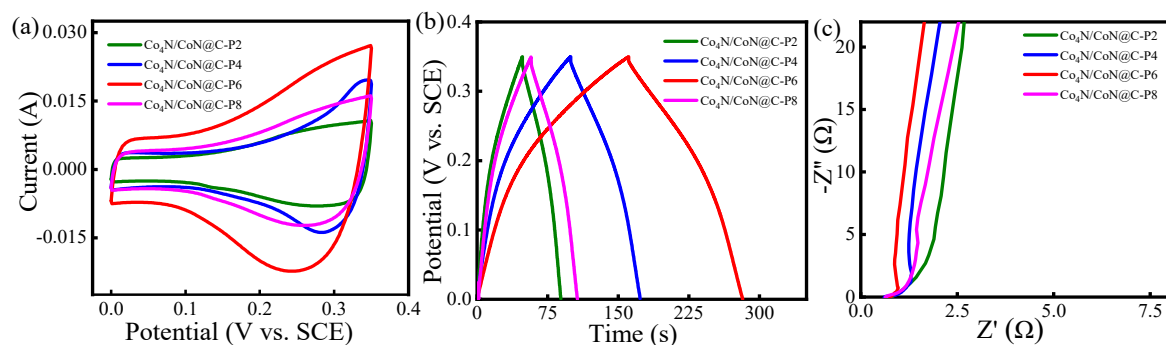
- 1 College of Physical Science and Technology, Dalian University, Dalian 116622, China
  - 2 Sunstone Energy Co., Ltd., Gansu 735100, China
  - 3 State Key Laboratory of Structural Analysis for Industrial Equipment, Dalian University of Technology, Dalian 116024, China
  - 4 Key Laboratory of Advanced Technology for Aerospace Vehicles of Liaoning Province, Dalian University of Technology, Dalian 116024, China
- \* Correspondence: dilanbo@163.com (L. B. Di).



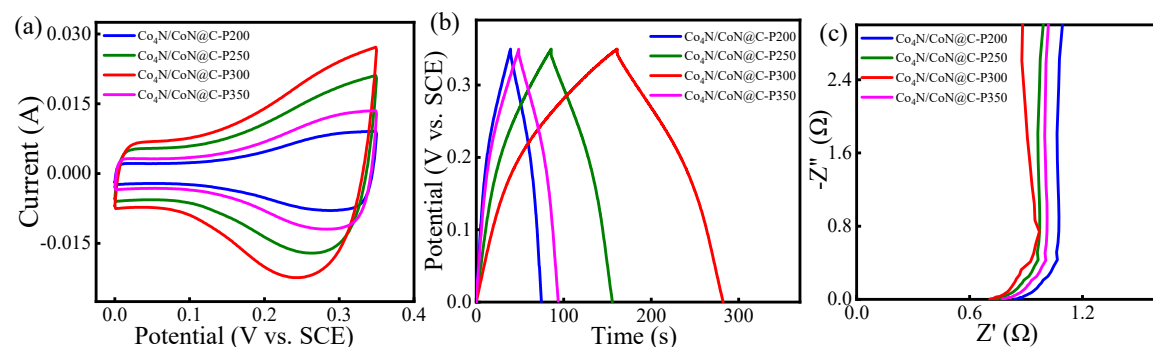
**Figure S1.** XRD patterns of  $\text{Co}_4\text{N}/\text{CoN}@C\text{-P}(\text{Ar})$  and  $\text{Co}_4\text{N}@C\text{-P}(\text{N}_2)$  prepared at 40 Pa, 300 W and discharge time of 6 min.



**Figure S2.** Electrochemical performance for different discharge atmosphere at a fixed power of 300 W and discharge time of 6 min, with different gas pressures of 20, 40, and 60 Pa. (a) CV curves at  $10 \text{ mV}\cdot\text{s}^{-1}$ , (b) GCD curves at  $1 \text{ A}\cdot\text{g}^{-1}$ , (c) EIS curves of  $\text{Co}_4\text{N}@C\text{-P}(\text{N}_2)$ ; (d) CV curves at  $10 \text{ mV}\cdot\text{s}^{-1}$ , (e) GCD curves at  $1 \text{ A}\cdot\text{g}^{-1}$ , (f) EIS curves of  $\text{Co}_4\text{N}/\text{CoN}@C\text{-P}(\text{Ar})$ .



**Figure S3.** Electrochemical performance of  $\text{Co}_4\text{N}/\text{CoN}@C\text{-P}(\text{Ar})$  prepared at 300 W and 40 Pa, with different discharge time of 2, 4, 6, and 8 min. (a) CV curves at  $10 \text{ mV}\cdot\text{s}^{-1}$ ; (b) GCD curves at  $1 \text{ A}\cdot\text{g}^{-1}$ ; (c) EIS curves.



**Figure S4.** Electrochemical performance of  $\text{Co}_4\text{N}/\text{CoN}@C\text{-P}(\text{Ar})$  prepared at 40 Pa and discharge time of 6 min, with different discharge powers of 200, 250, 300, and 350 W. (a) CV curves at  $10 \text{ mV}\cdot\text{s}^{-1}$ ; (b) GCD curves at  $1 \text{ A}\cdot\text{g}^{-1}$ ; (c) EIS curves.

**Table S1.** Equivalent circuit parameters obtained from Nyquist plots of Co-MFF,  $\text{CoN}/\text{Co}_4\text{N}@C$  and  $\text{Co}_4\text{N}/\text{CoN}@C\text{-P}$ .

	$R_s(\Omega)$	$R_{ct}(\Omega)$	CPE1	CPE2
Co-MFF	1.38	0.54	0.0434	0.1583
Error (%)	3.59	5.94	6.0140	0.8009
$\text{CoN}/\text{Co}_4\text{N}@C$	0.73	0.34	0.0451	0.0416
Error (%)	1.48	5.39	3.5010	0.5594
$\text{Co}_4\text{N}/\text{CoN}@C\text{-P}$	0.73	0.28	0.0510	0.0690
Error (%)	1.29	4.26	7.2620	0.7953

**Table S2.** The specific surface areas ( $S_{\text{BET}}$ ), pore volumes ( $V_{\text{p}}$ ), and pore diameters ( $D_{\text{p}}$ ) of Co-MFF, CoN/Co<sub>4</sub>N@C, and Co<sub>4</sub>N/CoN@C-P.

	$S_{\text{BET}} (\text{m}^2 \cdot \text{g}^{-1})$	$V_{\text{p}} (\text{cm}^3 \cdot \text{g}^{-1})$	$D_{\text{p}} (\text{nm})$
Co-MFF	65.0	0.1905	3.777
CoN/Co <sub>4</sub> N@C	17.7	0.0405	3.382
Co <sub>4</sub> N/CoN@C-P	7.0	0.0740	42.661