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Simplified Fast Synthesis of Strong-Coupling Composite Supercapacitor Materials by One-Step Bipolar Pulse Electrodeposition

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Table S1 Comparison of preparation steps and time of various NiCoS/Ppy with different structure designs.

electrode materials	Step one (The required time)	Step two (The required time)	Step three (The required time)	references
This work	**NiCoS/PPy (2400 s)**			
$NiCo_2S_4$@PPy/CF	$NiCo_2S_4$/NF (12+14 h)	$NiCo_2S_4$@PPy/NF (90 s)		1
Co_9S_8@PPy@NiCo-LDH NTAs	Co_9S_8 on CC (10+6 h)	layer of PPy (3 h)	Electrodeposition NiCo-LDH	2
$NiCo_2S_4$@PPy	$NiCo_2S_4$ (6+7 h)	$NiCo_2S_4$@PPy (9 h)		3
PPy@$NiCo_2S_4$	PPy nanotubes (12 h)	PPy@SiO_2 NTs (3 h)	Ppy@$NiCo_2S_4$ core-shell (12+12 h)	4
PPy@$NiCo_2S_4$/GF	$NiCo_2S_4$/GF (8+12 h)	PPy@$NiCo_2S_4$/G (24 h)		5
$CoNi_2S_4$@PPy/N-3DG	$CoNi_2S_4$/N-3DG (4+10 h)	$CoNi_2S_4$@PPy/N-3DG (10 min)		6
NiCoS@PPy	PPy tubes (24 h)	ZIF-67@PPy (24 h)	NiCoS@Ppy (4 h)	7

NiCo-MOF@PPy	PPy nanotubes (24 h)	NiCo-MOF@Ppy (8 h)	8
$NiCo_2S_4$@PPy-Ni foam	$NiCo_2S_4$-Ni foam (8+12 h)	$NiCo_2S_4$@PPy-Ni foam (12 h)	9
CNS/PPy/CP	S@PPy	CNS/PPy/CP (4000 s)	10

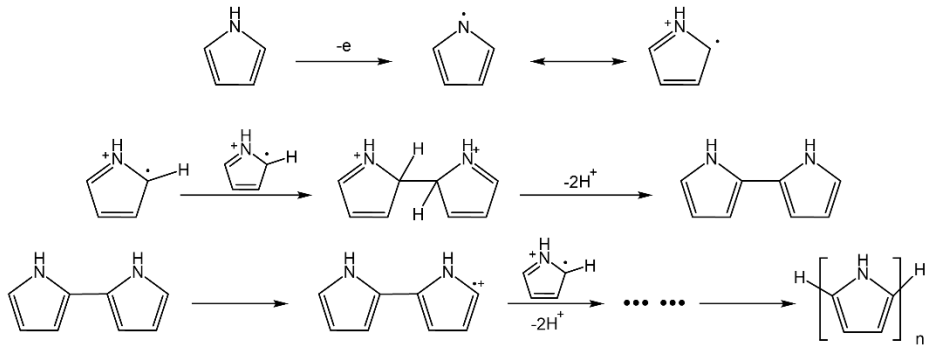

Fig. S1. Mechanism of electro polymerization of pyrrole.

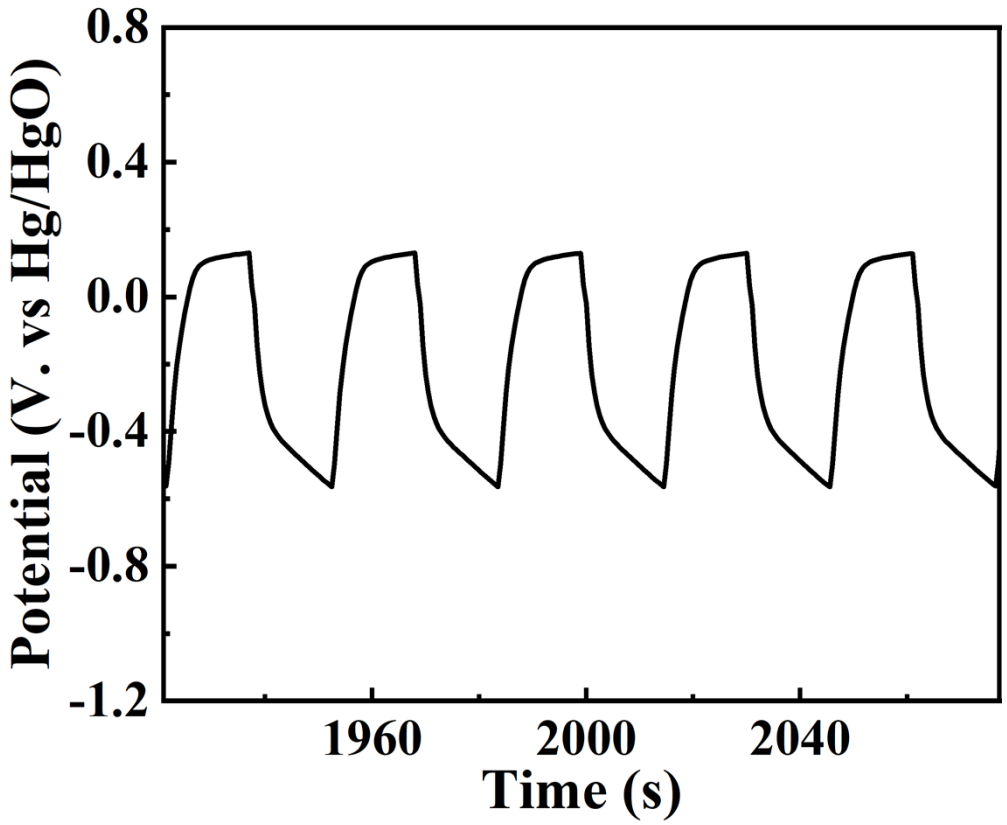

Fig. S2. The potential-time curve after stabilization of NCS/P-3.

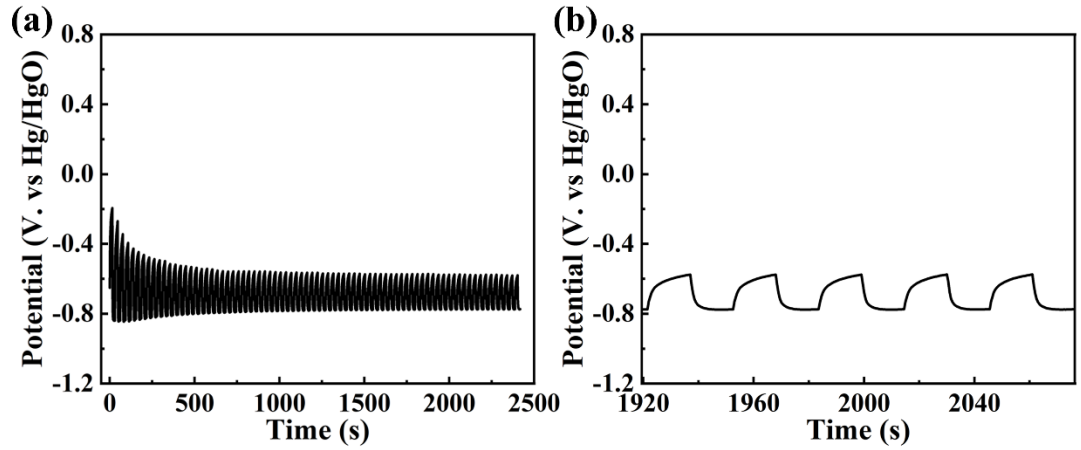

Fig. S3. The potential-time curve during the preparation of NCS/P-1 (a), and the potential-time curve after stabilization of NCS/P-1 (b).

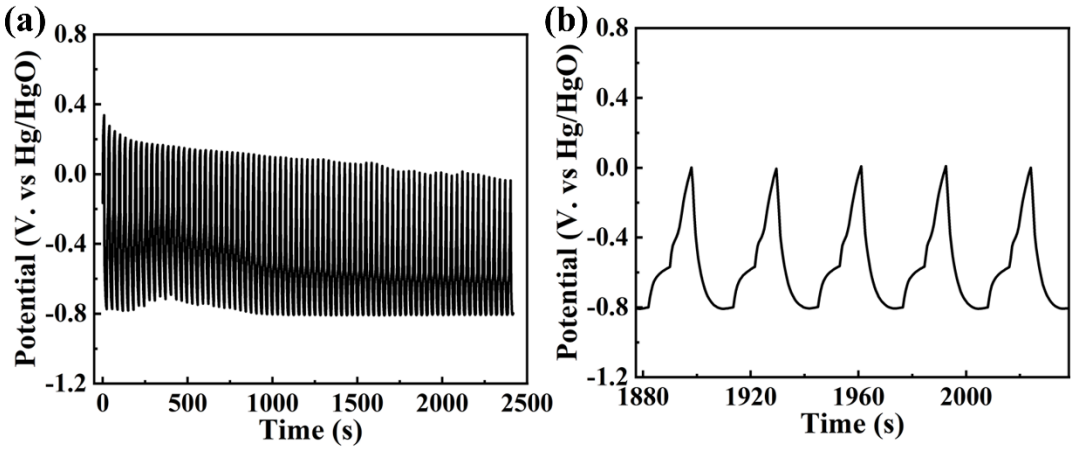

Fig. S4. The potential-time curve during the preparation of NCS/P-2 (a), and the potential-time curve after stabilization of NCS/P-2 (b).

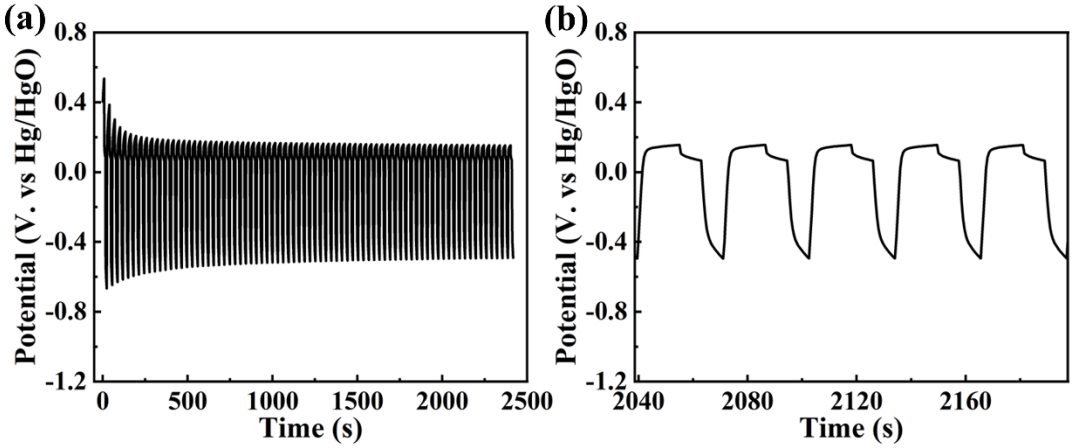

Fig. S5. The potential-time curve during the preparation of NCS/P-4 (a), and the potential-time curve after stabilization of NCS/P-4 (b).

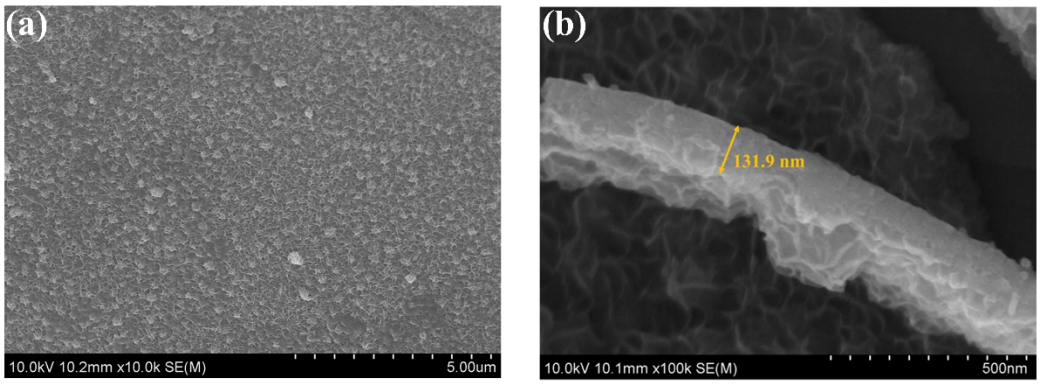

Fig. S6. SEM images of (a) NCS/P-1, SEM section images of (b) NCS/P-1.

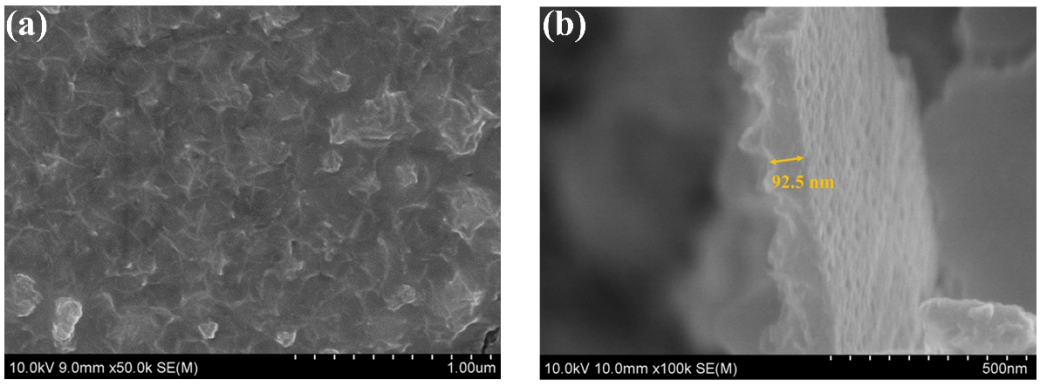

Fig. S7. SEM images of (a-b) NCS/P-2, SEM section images of (c) NCS/P-2.

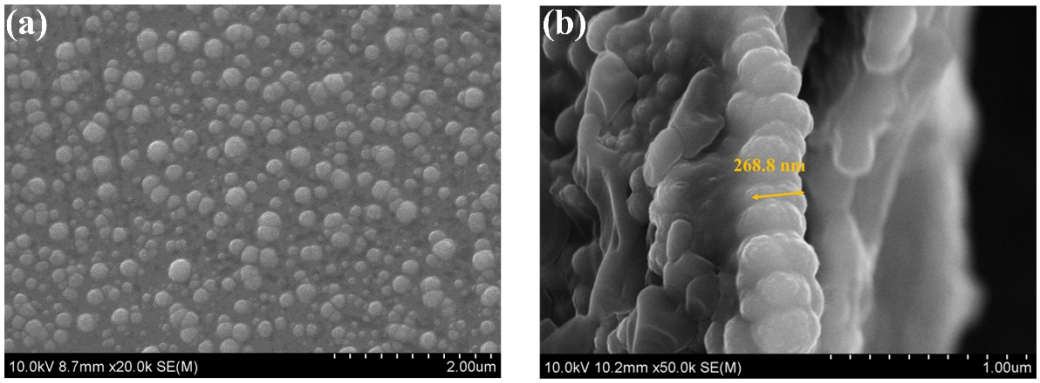

Fig. S8. SEM images of (a) NCS/P-4, SEM section images of (b) NCS/P-4.

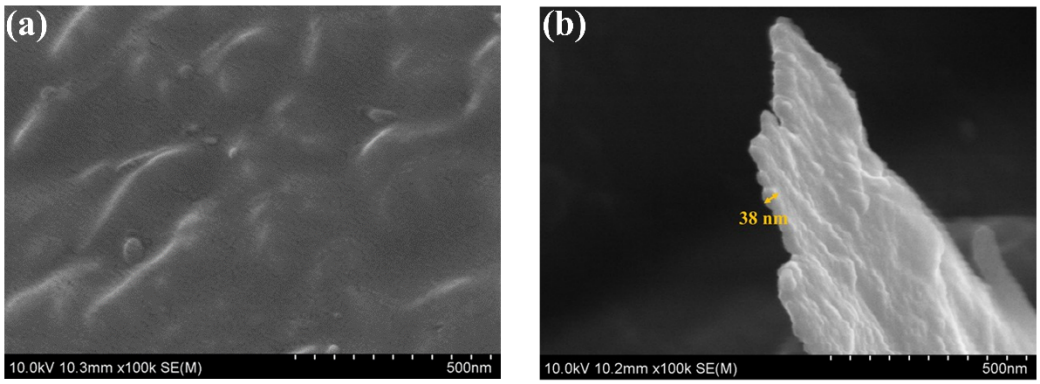

Fig. S9. SEM images of (a) Pure PPy, SEM section images of (b) Pure PPy.

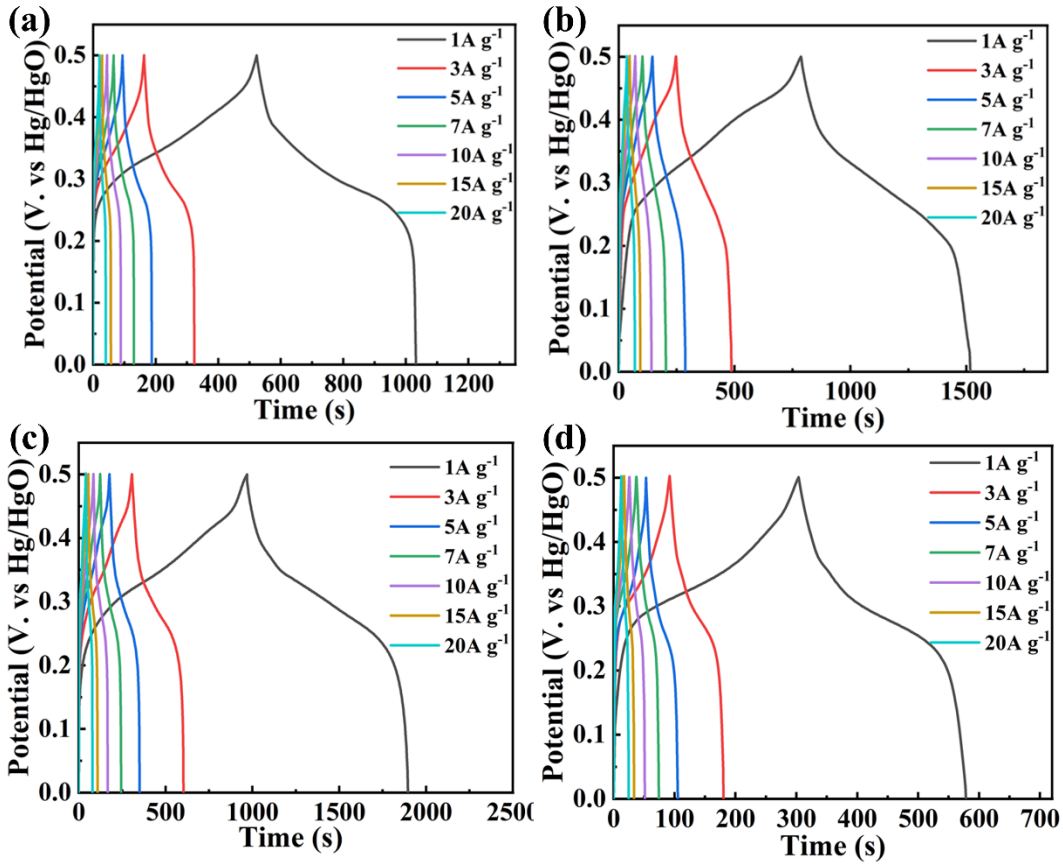

Fig. S10. The GCD at various current density of (a) NCS/P-1, (b) NCS/P-2, (c) NCS/P-3, (d) NCS/P-4.

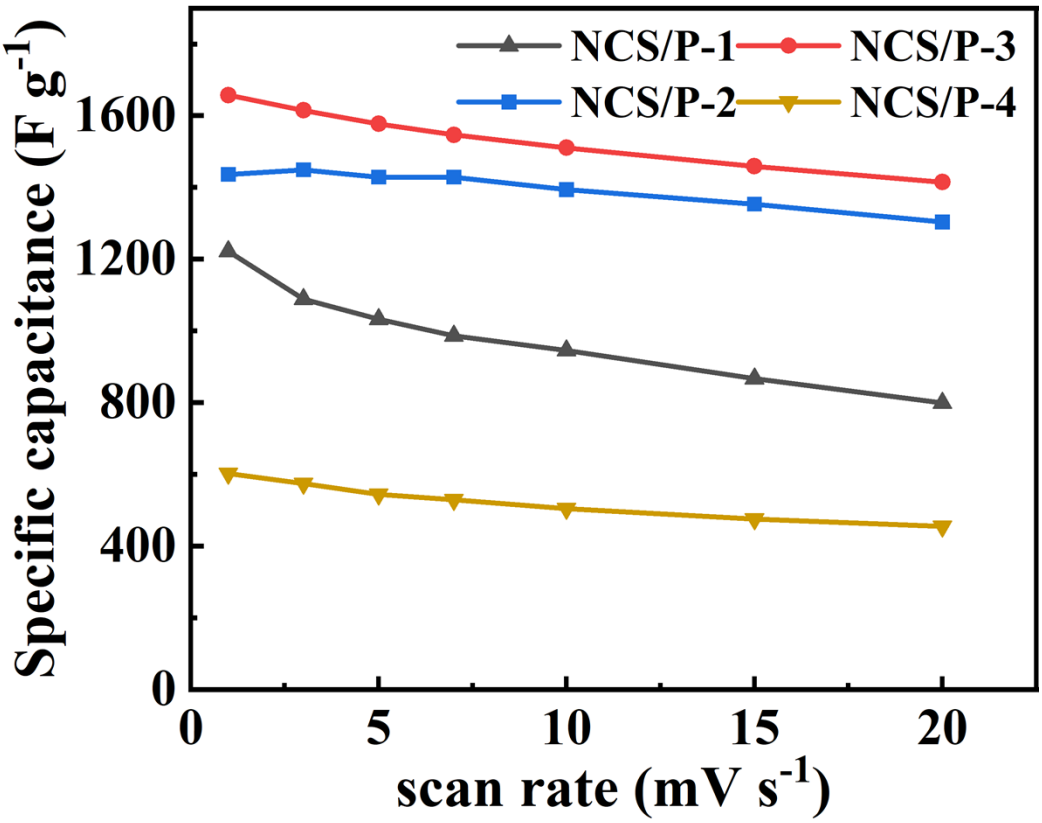

Fig. S11. The specific capacitance of electrodes at various scan rates.

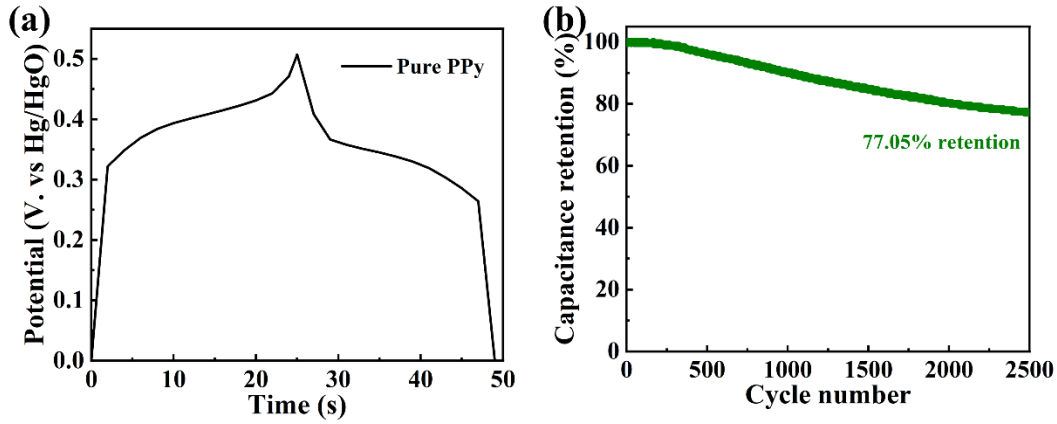

Fig. S12. The GCD at 5 A g^{-1} of pure PPy electrode (a), and the cycling performance of pure PPy electrode at the current density of 10 A g^{-1} (b).

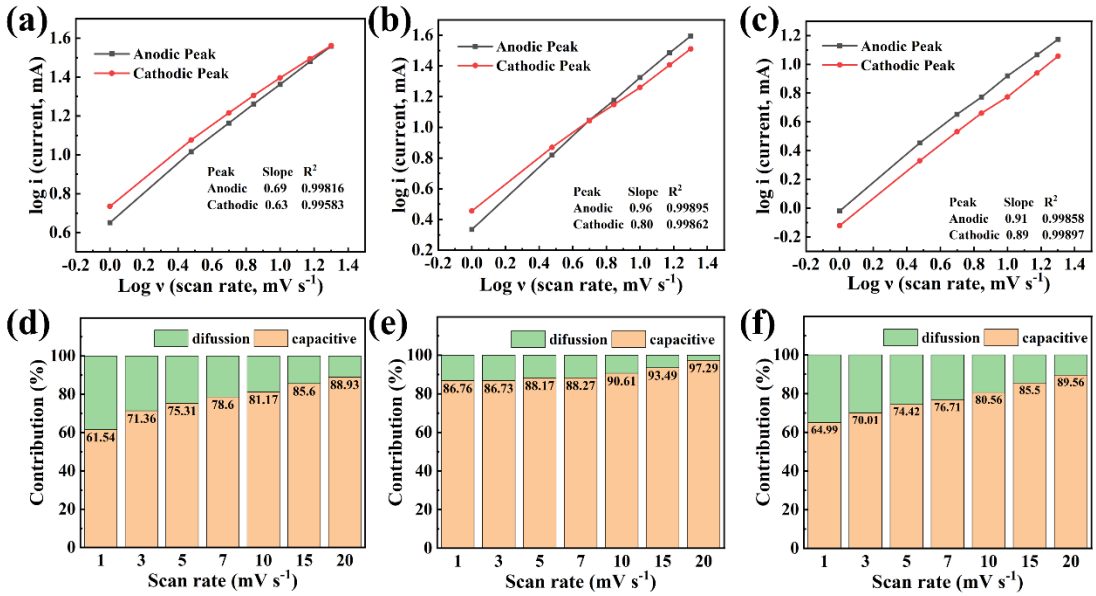

Fig. S13. Log (i) as a function of log (v) of (a) NCS/P-1, (b) NCS/P-2, (c) NCS/P-4. The contribution fractions of the capacitive and diffusion-controlled processes of (d) NCS/P-1, (e) NCS/P-2, (f) NCS/P-4.

Reference：

1. M. Yan, Y. Yao, J. Wen, L. Long, M. Kong, G. Zhang, X. Liao, G. Yin and Z. Huang, *ACS Appl Mater Interfaces*, 2016, **8**, 24525-24535.
2. L. Wang, S. K. Li, F. Z. Huang, X. Y. Yu, M. J. Liu and H. Zhang, *Journal of Power Sources*, 2019, **439**.
3. J. Li, Y. J. Zou, B. Li, F. Xu, H. L. Chu, S. J. Qiu, J. Zhang, L. X. Sun and C. L. Xiang, *Ceramics International*, 2021, **47**, 16562-16569.
4. Y. Y. Zheng, J. Xu, X. S. Yang, Y. J. Zhang, Y. Y. Shang and X. Y. Hu, *Chemical Engineering Journal*, 2018, **333**, 111-121.
5. Y. F. Zhu, F. F. Wang, H. L. Zhang, X. B. Lv, Z. F. Hu, H. Han, X. Y. Fan, J. Y. Ji and X. D. Guo, *Journal of Alloys and Compounds*, 2018, **747**, 276-282.

6. B. Liu, R. W. Mo, D. Z. Kong, Y. Wang and H. Y. Yang, *Flatchem*, 2017, **6**, 1-10.

7. X. Y. Zhao, Q. X. Ma, K. Tao and L. Han, *Acs Applied Energy Materials*, 2021, **4**, 4199-4207.

8. Y. X. Liu, Y. Z. Wang, Y. J. Chen, C. Wang and L. Guo, *Applied Surface Science*, 2020, **507**.

9. X. Wu, L. Meng, Q. Wang, W. Zhang and Y. Wang, *Chemical Engineering Journal*, 2019, **358**, 1464-1470.

10. X. Li, W. Yan, S. Guo, Y. Liu, J. Niu, L. Yin and Z. Wang, *Electrochimica Acta*, 2021, **387**.