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PdNi alloy decorated 3D hierarchically N, S co-doped macro–mesoporous carbon composites as efficient free-standing and binder-free catalysts for Li-O_2 battery

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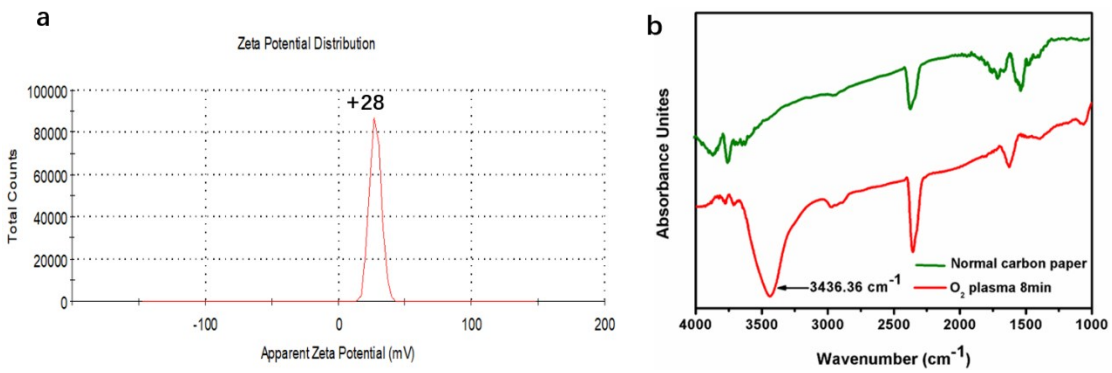

Figure S1. (a) The Zeta Potential in aqueous solution of amino-modified SiO_2 nanospheres, (b) FTIR spectra of normal and O_2 plasma treated carbon paper.

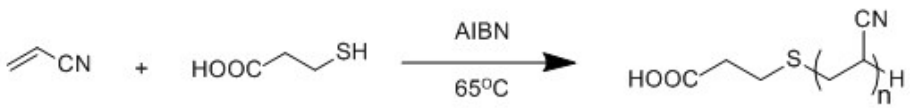

Figure S2. The synthesis route of ANT.

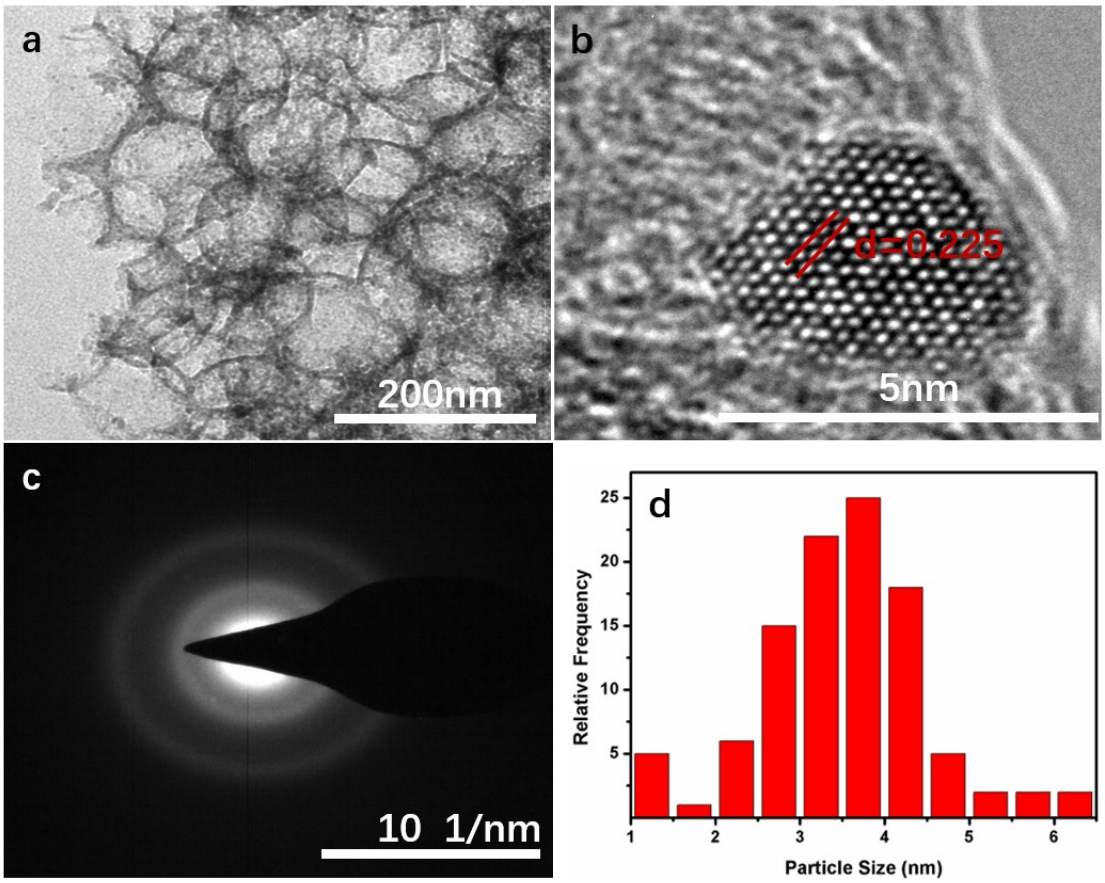

Figure S3. (a) TEM and (b) HRTEM images; (c) selected area electron diffraction (SAED) pattern and (d) the particle size distribution of Pd-NSMmC/CP.

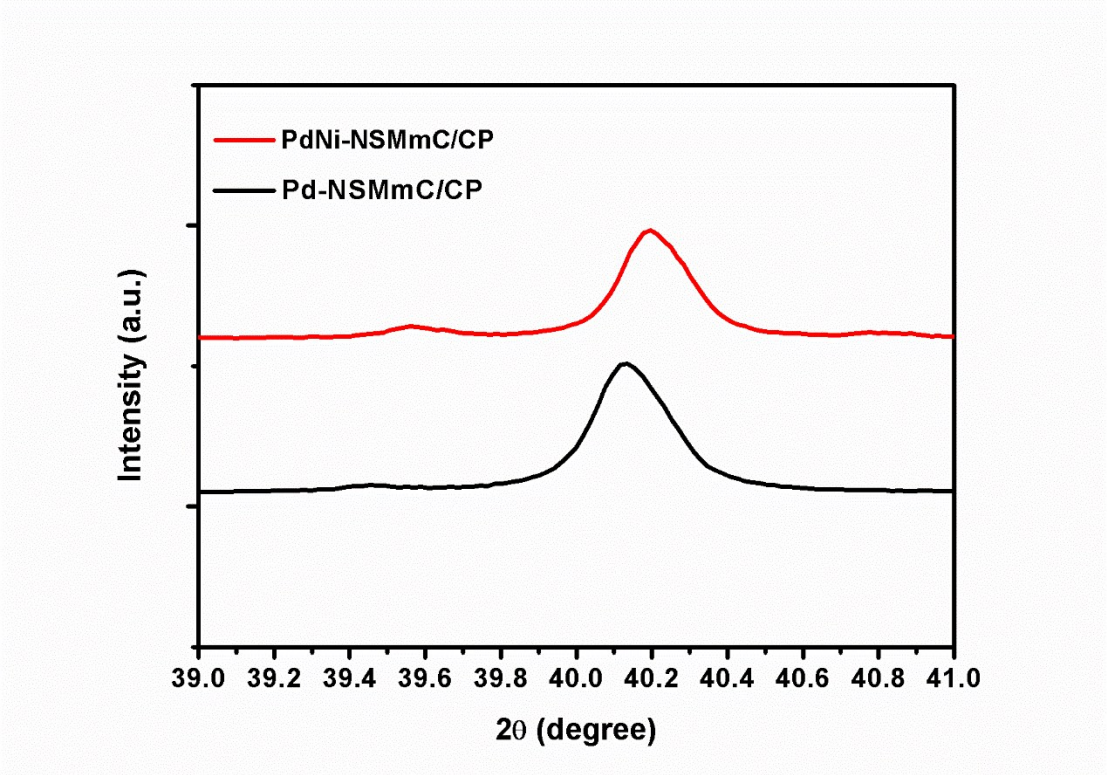

Figure S4. The enlarged XRD pattern of the (111) peak.

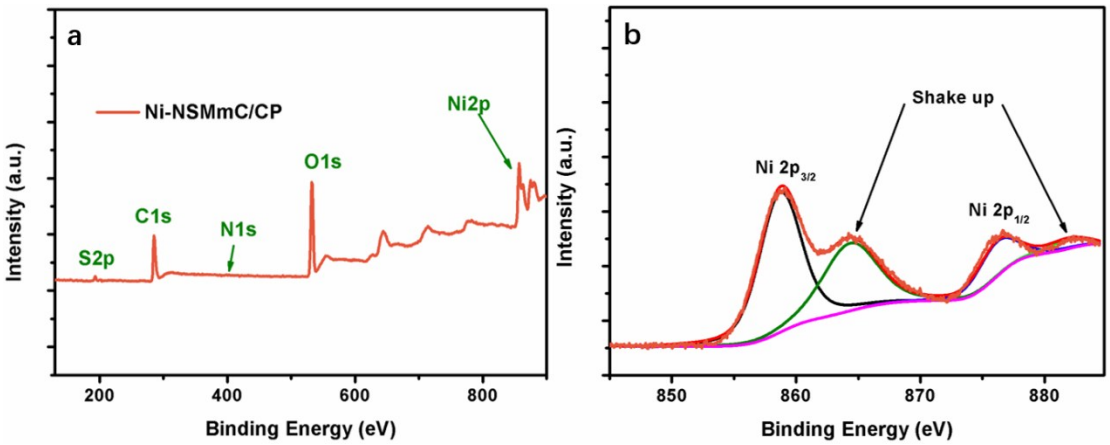

Figure S5. (a) XPS survey spectrum of Ni-NSMmC/CP; (b) Ni2P XPS spectrum.

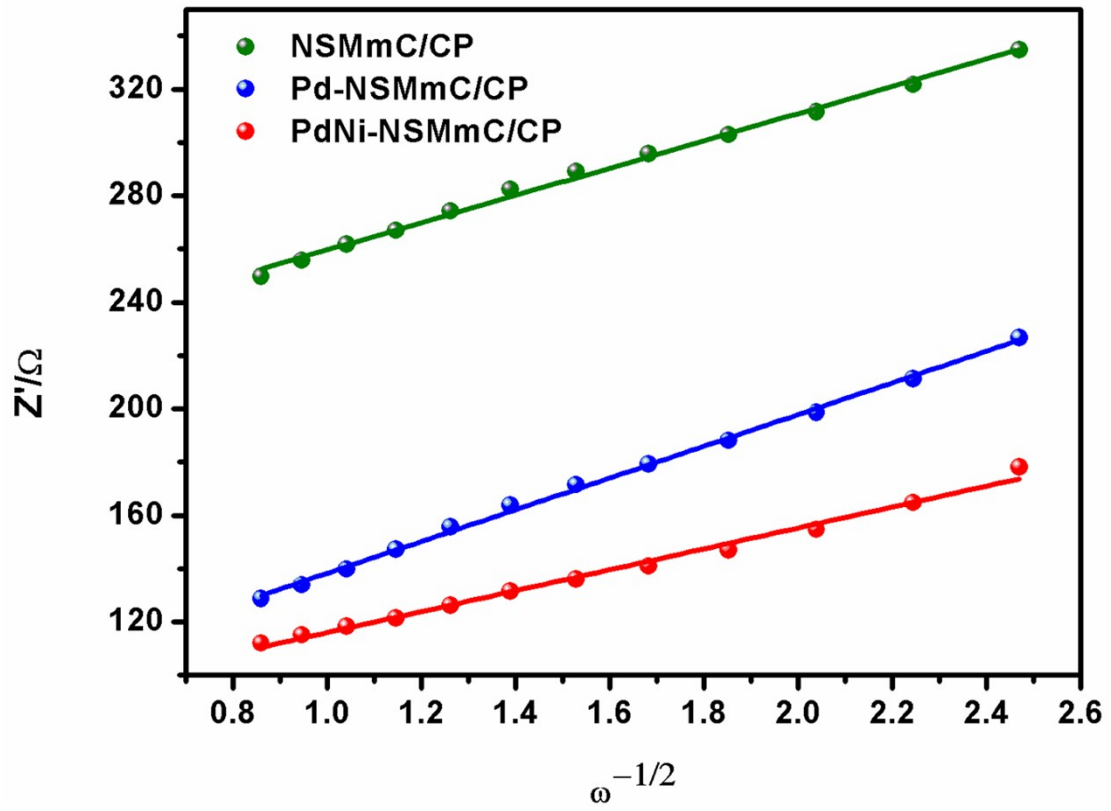

Figure S6. The relationship between Z´and $\omega^{-1/2}$ of O_2 electrode.

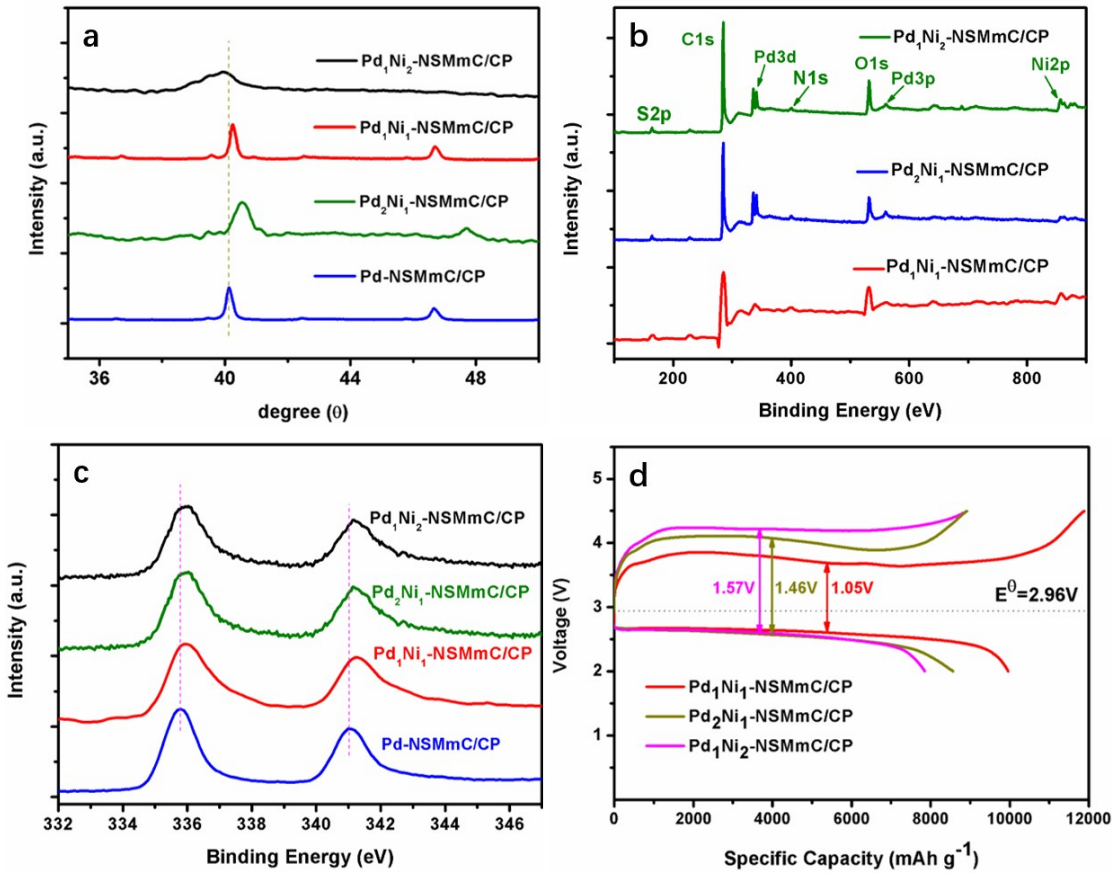

Figure S7. (a) XRD patterns, (b) XPS spectra, (c) Raman spectra and (d) charge/discharge profiles of Pd_xNi_y-NSMmC/CP-based composites.

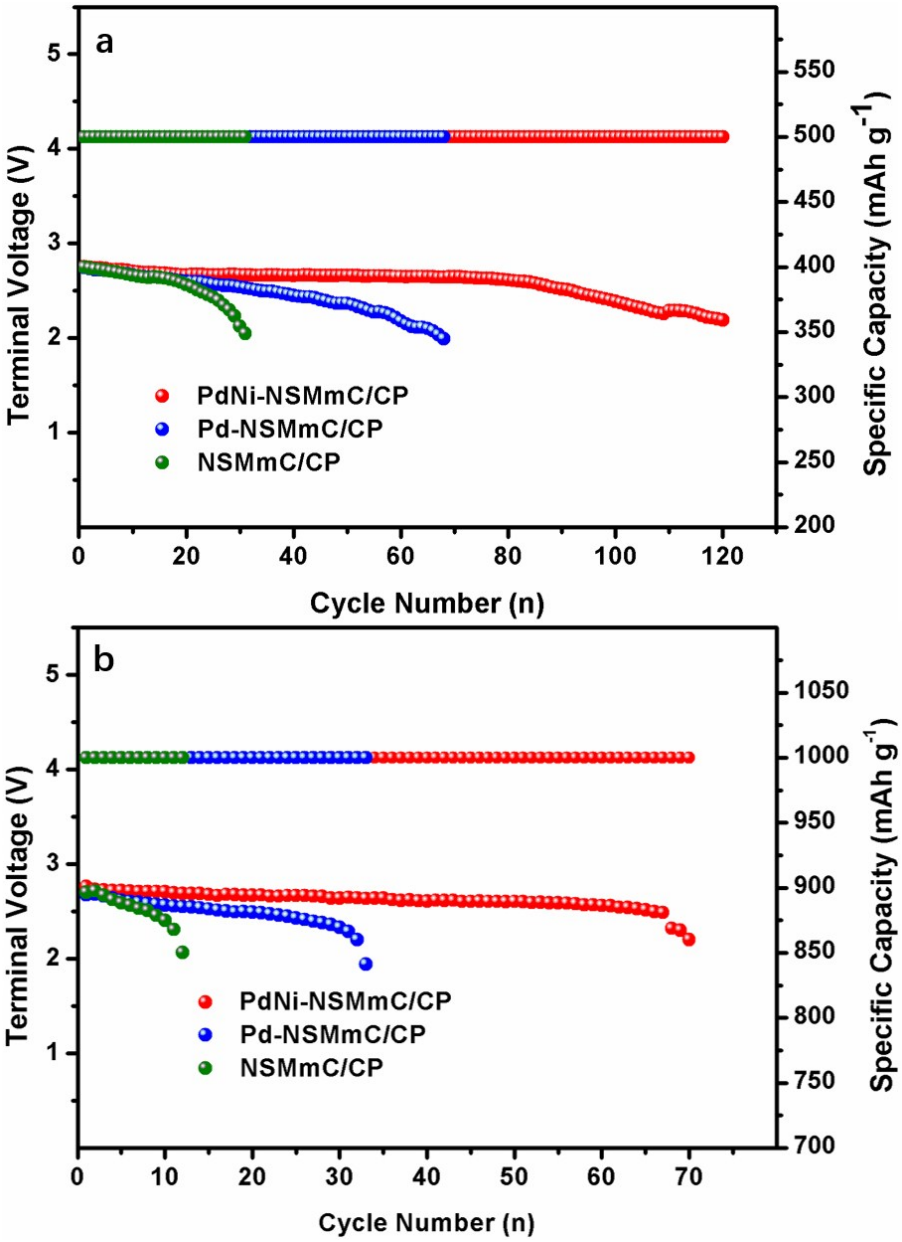

Figure S8. Cycle performance of the PdNi-NSMmC/CP, Pd-NSMmC/CP and NSMmC electrodes at current density of 300mA g^{-1} with limited capacity of (a) 500mAh g^{-1} (b)1000mAh g^{-1}.

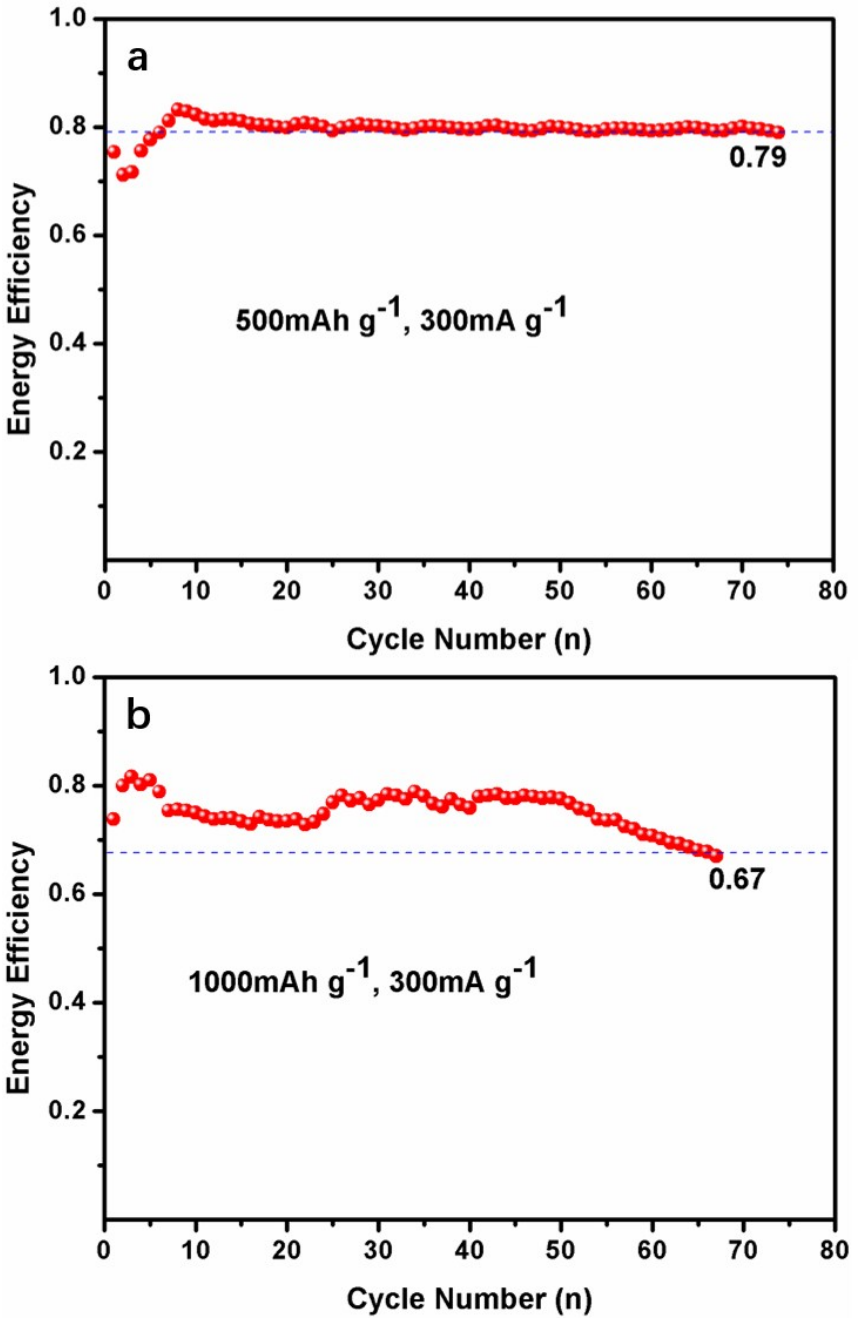

Figure S9. Energy conversion efficiency changes with cycling for PdNi-NSMmC/CP.

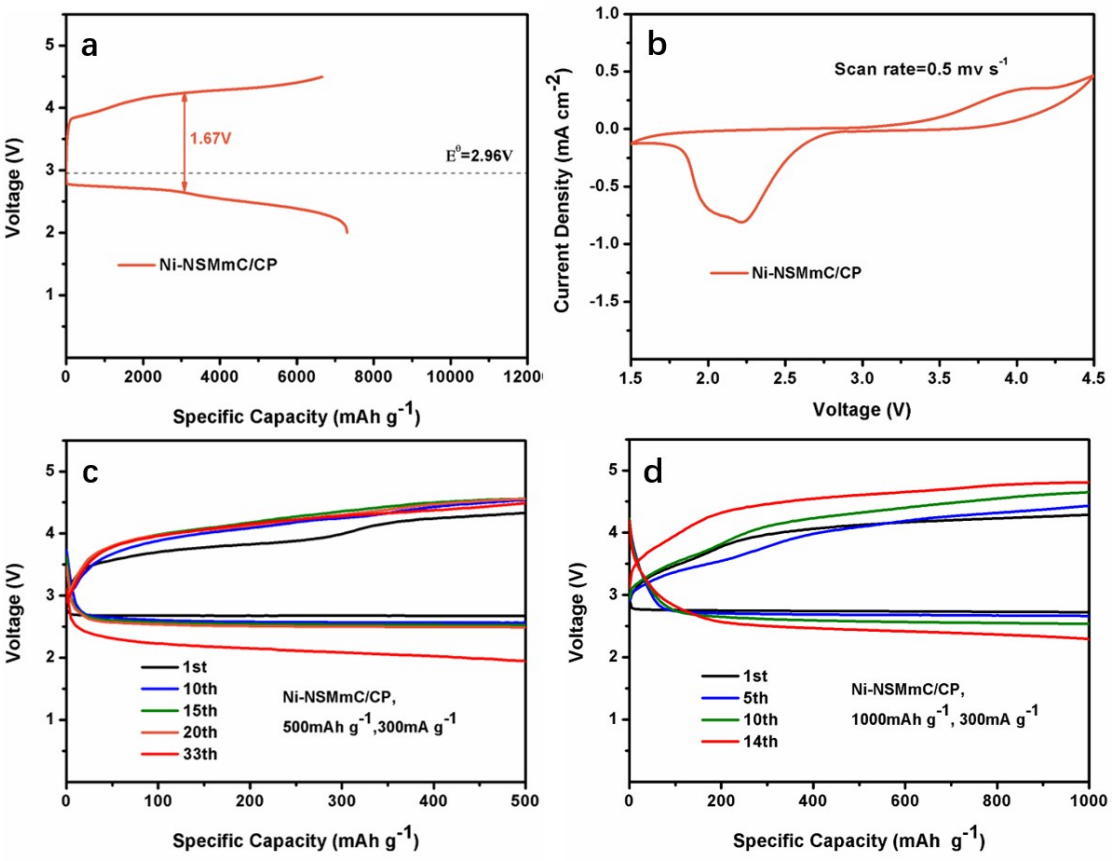

Figure S10. (a) The initial discharge/charge profiles of Ni-NSMmC/CP cathode with the voltage range of 2.0-4.5 V at a current density of 300mAh g^{-1}. (b) CV curve of Ni-NSMmC/CP at a scan rate of 0.5 mV s^{-1}. Cycling performance of Ni-NSMmC/CP cathode at a current density of 300 mA g^{-1} with a limited capacity of (c) 500 mAh g^{-1} and (d) 1000 mAh g^{-1}.

Table S1. Contents of Pd and Ni determined by ICP-AES.

	Pd (mg/sample)	Ni (mg/sample)
PdNi-NSMmC/CP	0.0542	0.0178
Pd-NSMmC/CP	0.0651	/
NSMmC/CP	/	/

Table S2. Comparison of Li-O_2 battery properties of PdNi-NSMmC/CP cathode with those of representative state-of-the-art cathodes reported in literature.

Materials	Current density	Overpotential (V)	Limited capacity/cycles (mAh g^{-1})/ times	1st discharge capacity (mAh g^{-1})	Products Morphology	Ref.
Pd/Co3O4 Nanoclusters	0.1 mA cm^{-2}	1.17	300 /70	～1840	nanoparticles	1
Pd NDs -GNPs	200mA g^{-1}	0.89	500/30	～3000	nanosheet-like	2
PtAu/HMCMS	100mA g^{-1}	1.28	1000/75	～6000	film-like	3
Pd/PNCNF	100mA g^{-1}	1.47	1000/50	～10080	Sheet-like	4
Ru/GA	0.1mA cm^{-2}	1.25	500/50	～12000	toroidal	5
CBC/Ru Nanofibers	200 mA g^{-1}	1.26	500/27	~ 2750	-	6
Ru/hCNCs	0.08mA cm^{-2}	1.15	500/79	～8000	toroid-like	7
Au-MnO2	100mA g^{-1}	1.36	1000/60	～5760	leaf-like	8
Au@CST	400mA g^{-1}	1.79	500/112	～5000	thin-layered	9
Pt-HGNs	100mA g^{-1}	-	1000/55	～5600	toroid shape	10
Pd/Co3O4 nanosheets	100mA g^{-1}	1.46	300/72	～1500	nanoparticles	11
wheat-like Ag–Mn3O4	0.02mA cm^{-2}	0.8	1000/50	～5000	–	12
PdNi-NSMmC/CP	300mA g^{-1}	1.05	500/120, 1000/70	～9960	Cage-like	This work

Reference

1. L. Leng, X. Zeng, H. Song, T. Shu, H. Wang and S. Liao, *Journal of Materials Chemistry A*, 2015, **3**, 15626-15632.
2. S. J. Ye, D. Y. Kim, D. W. Kim, O. O. Park and Y. Kang, *Journal of Materials Chemistry A*, 2016, **4**, 578-586.
3. M. Lu, D. Chen, C. Xu, Y. Zhan and J. Y. Lee, *Nanoscale*, 2015, **7**, 12906-12912.
4. J. Wang, L. Liu, S. Chou, H. Liu and J. Wang, *Journal of Materials Chemistry A*, 2017, **5**, 1462-1471.
5. J. Jiang, P. He, S. Tong, M. Zheng, Z. Lin, X. Zhang, Y. Shi and H. Zhou, *NPG Asia Materials*, 2016, **8**, e239-e239.
6. S. Tong, M. Zheng, Y. Lu, Z. Lin, X. Zhang, P. He and H. Zhou, *Chemical communications*, 2015, **51**, 7302-7304.
7. L. Wang, Z. Lyu, L. Gong, J. Zhang, Q. Wu, X. Wang, F. Huo, W. Huang, Z. Hu and W. Chen, *ChemNanoMat*, 2017, **3**, 415-419.
8. M. Lu, J. Qu, Q. Yao, C. Xu, Y. Zhan, J. Xie and J. Y. Lee, *ACS applied materials & interfaces*, 2015, **7**, 5488-5496.
9. F. Tu, J. Hu, J. Xie, G. Cao, S. Zhang, S. A. Yang, X. Zhao and H. Y. Yang, *Advanced Functional Materials*, 2016, **26**, 7725-7732.
10. F. Wu, Y. Xing, X. Zeng, Y. Yuan, X. Zhang, R. Shahbazian-Yassar, J. Wen, D. J. Miller, L. Li, R. Chen, J. Lu and K. Amine, *Advanced Functional Materials*, 2016, **26**, 7626-7633.
11. Y. Ren, S. Zhang, H. Li, X. Wei and Y. Xing, *Applied Surface Science*, 2017, **420**, 222-232.
12. J. Qu, M. Lu, C. Xu, B. Ding, Y. Zhan, J. Yang and J. Y. Lee, *Nanoscale*, 2014, **6**, 12324-12327.