## **Supporting Information**

## Ultrathin Petal-like NiAl Layered Double oxide/sulfide Composites as Advanced Electrode for High-performance Asymmetric Supercapacitor

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Table S1. Elements composition (at%) of the NiAl LDO/LDScomposites analyzed by XPS

Sample	Element					
	Ni	Al	0	S	С	
NiAl LDO/LDS	31.65	8.76	35.71	5.06	18.82	



**Fig. S1.** (a) XRD pattern of NiAl LDO/LDS composites at different sulfidation temperature, and the corresponding SEM images of (b) 400 °C, (c) 600 °C and (d) 700 °C.



Fig. S2. XPS pattern of the oxygen element in the NiAl LDO/LDS composites.



Fig. S3. (a) TEM image, (b) AFM image, and (c) height profile of the NiAl LDO/LDS composites.



Fig. S4. Cycling performance and Coulombic efficiency of the NiAl LDH and NiAl LDO/LDS composites at a current density of 5 A  $g^{-1}$ .

Table S2. EIS fitting parameters of NiAl LDH and NiAl LDO/LDSelectrodes

Sample	$R_{s}(\Omega)$	C <sub>dl</sub> (F)	$R_{ct}(\Omega)$	W (Ω)	<b>C</b> <sub>F</sub> ( <b>F</b> )
NiAl LDH	0.665	0.0024	12.53	2.452	1.94
NiAl LDO/LDS	0.499	0.0058	0.308	2.612	3.52

Materials	Current density	Specific capacitance	Ref.
	(A g <sup>-1</sup> )	(F g <sup>-1</sup> )	
CoNi <sub>0.5</sub> LDH	1	1938	[1]
Ni <sub>1-x</sub> Co <sub>x</sub> Al-LDH	1	1902	[2]
CNTs/NiAl-LDH	1	694	[3]
MXene/NiAl LDH	1	1061	[4]
NiCo <sub>2</sub> O <sub>4</sub> @NiCoAl-	1	1814.24	[5]
LDH			
Co <sub>3</sub> O <sub>4</sub> @NiCoAl-	1	1104	[6]
LDH			
Ni <sub>3</sub> S <sub>2</sub>	1	1670	[7]
Ni <sub>3</sub> S <sub>2</sub> @Ni(OH) <sub>2</sub> /G	5.1	1037.5	[8]
NiO/Ni <sub>3</sub> S <sub>2</sub>	1	2153	[9]
MnCo <sub>2</sub> S <sub>4</sub>	1	2067	[10]
NiS <sub>2</sub>	1	1020.2	[11]
NiS <sub>2</sub> /ZnS	1	1198	[12]
Nickel sulfides/MoS <sub>2</sub>	0.5	757	[13]
Ni <sub>3</sub> S <sub>2</sub> /NiS	2	1158	[14]

 Table S3. Comparison of the electrochemical performance of the NiAl

LDO/LDS composites with the reported materials in literature

## References

[1] M.F. Warsi, I. Shakir, M. Shahid, M. Sarfraz, M. Nadeem, Z.A. Gilani, Conformal coating of cobalt-nickel layered double hydroxides nanoflakes on carbon fibers for high-performance electrochemical energy storage supercapacitor devices, Electrochimica Acta, 135 (2014) 513-518.

[2] J. Xu, S. Gai, F. He, N. Niu, P. Gao, Y. Chen, P. Yang, Reduced graphene oxide/Ni<sub>1-</sub>  $_xCo_x$  Al-layered double hydroxide composites: preparation and high supercapacitor performance, Dalton Transactions, 43 (2014) 11667-11675.

[3] C. Bai, S. Sun, Y. Xu, R. Yu, H. Li, Facile one-step synthesis of nanocomposite based on carbon nanotubes and Nickel-Aluminum layered double hydroxides with high cycling stability for supercapacitors, Journal of colloid and interface science, 480 (2016) 57-62.

[4] Y. Wang, H. Dou, J. Wang, B. Ding, Y. Xu, Z. Chang, X. Hao, Three-dimensional porous MXene/layered double hydroxide composite for high performance supercapacitors, Journal of Power Sources, 327 (2016) 221-228.

[5] X. He, Q. Liu, J. Liu, R. Li, H. Zhang, R. Chen, J. Wang, Hierarchical NiCo<sub>2</sub>O<sub>4</sub>@ NiCoAl-layered double hydroxide core/shell nanoforest arrays as advanced electrodes for high-performance asymmetric supercapacitors, Journal of Alloys and Compounds, 724 (2017) 130-138.

[6] X. Li, Z. Yang, W. Qi, Y. Li, Y. Wu, S. Zhou, S. Huang, J. Wei, H. Li, P. Yao, Binderfree Co<sub>3</sub>O<sub>4</sub>@NiCoAl-layered double hydroxide core-shell hybrid architectural nanowire arrays with enhanced electrochemical performance, Applied Surface Science, 363 (2016) 381-388.

[7] B. Yang, L. Yu, Q. Liu, J. Liu, W. Yang, H. Zhang, F. Wang, S. Hu, Y. Yuan, J. Wang, The growth and assembly of the multidimensional hierarchical  $Ni_3S_2$  for aqueous asymmetric supercapacitors, CrystEngComm, 17 (2015) 4495-4501.

[8] W. Zhou, X. Cao, Z. Zeng, W. Shi, Y. Zhu, Q. Yan, H. Liu, J. Wang, H. Zhang, One-step synthesis of  $Ni_3S_2$  nanorod@Ni(OH)<sub>2</sub> nanosheet core-shell nanostructures on a three-dimensional graphene network for high-performance supercapacitors, Energy & Environmental Science, 6 (2013) 2216-2221.

[9] S. Liu, S.C. Lee, U.M. Patil, C. Ray, K.V. Sankar, K. Zhang, A. Kundu, J.H. Park, S.C. Jun, Controllable sulfuration engineered NiO nanosheets with enhanced capacitance for high rate supercapacitors, J Mater Chem A, 5 (2017) 4543-4549.

[10] S. Liu, S.C. Jun, Hierarchical manganese cobalt sulfide core-shell nanostructures for high-performance asymmetric supercapacitors, Journal of Power Sources, 342 (2017) 629-637.

[11] Y. Ruan, J. Jiang, H. Wan, X. Ji, L. Miao, L. Peng, B. Zhang, L. Lv, J. Liu, Rapid self-assembly of porous square rod-like nickel persulfide via a facile solution method for high-performance supercapacitors, Journal of Power Sources, 301 (2016) 122-130.

[12] G.-C. Li, M. Liu, M.-K. Wu, P.-F. Liu, Z. Zhou, S.-R. Zhu, R. Liu, L. Han, MOFderived self-sacrificing route to hollow  $NiS_2/ZnS$  nanospheres for high performance supercapacitors, RSC Advances, 6 (2016) 103517-103522.

[13] X. Yang, L. Zhao, J. Lian, Arrays of hierarchical nickel sulfides/MoS<sub>2</sub> nanosheets supported on carbon nanotubes backbone as advanced anode materials for asymmetric supercapacitor, Journal of Power Sources, 343 (2017) 373-382.

[14] W. Li, S. Wang, L. Xin, M. Wu, X. Lou, Single-crystal  $\beta$ -NiS nanorod arrays with a hollow-structured Ni<sub>3</sub>S<sub>2</sub> framework for supercapacitor applications, Journal of Materials Chemistry A, 4 (2016) 7700-7709.