Supporting Information

Electrostatic-induced Assembly of Graphene-encapsulated Carbon@Nickel-Aluminum Layered Double Hydroxide Core–Shell Spheres Hybrid Structure for High-energy and High-power-density Asymmetric Supercapacitor

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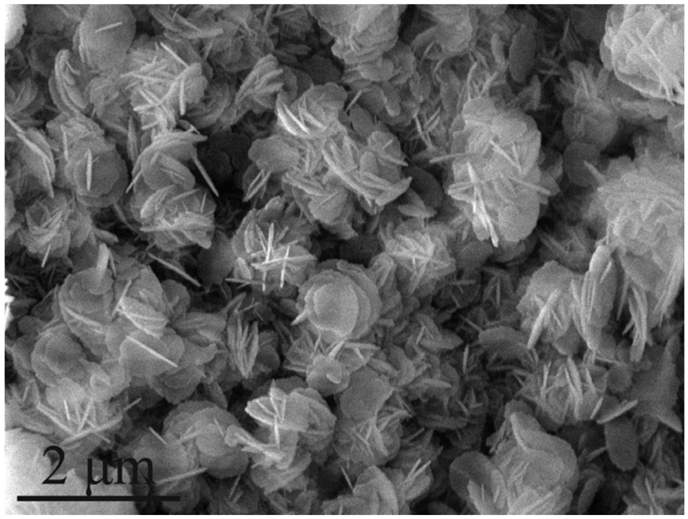

Figure S1. SEM image of CLH

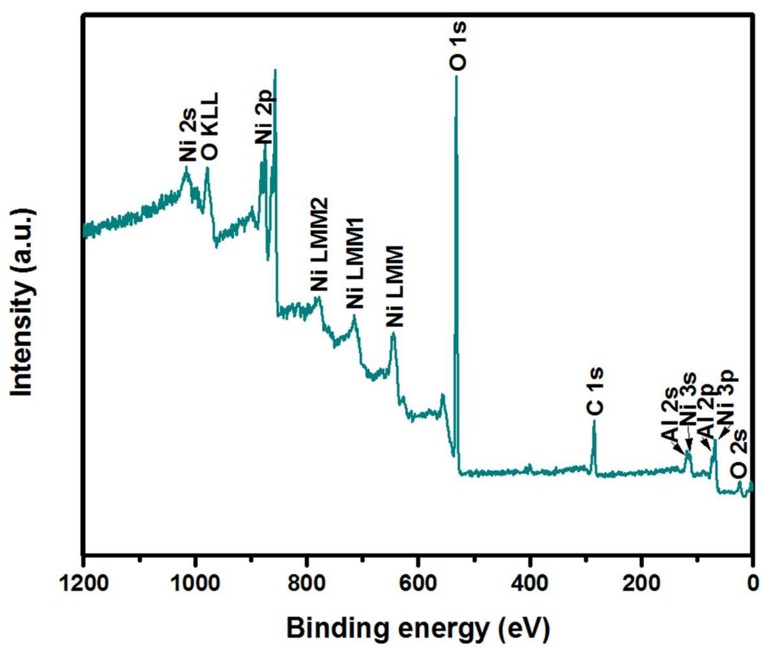

Figure S2. XPS survey spectra of CLS

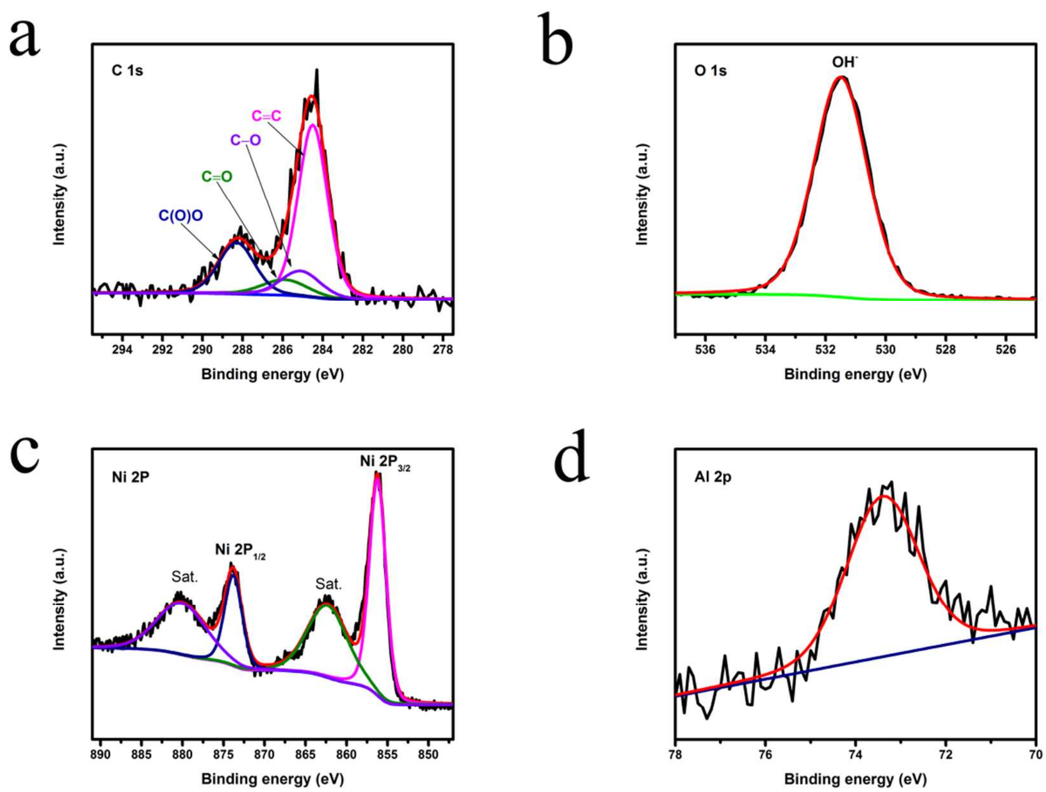

Figure S3. High-resolution XPS spectrum of C 1s (a), O1s (b), Ni 2p (c), and Al 2p (d).

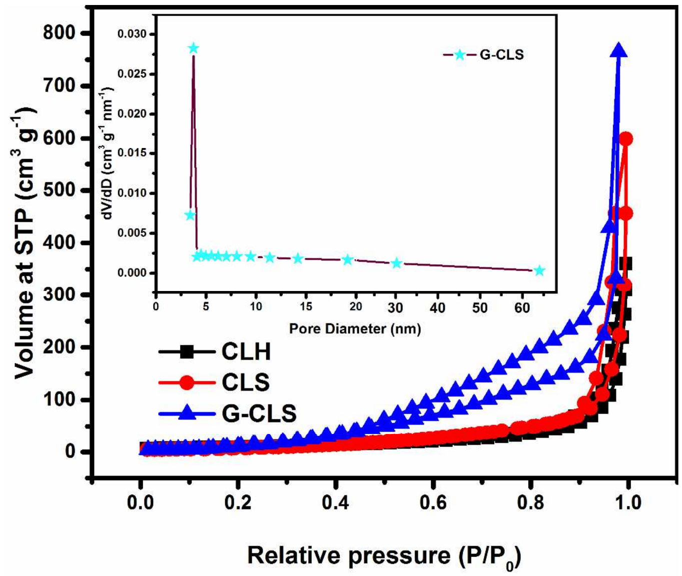

Figure S4. Nitrogen adsorption-desorption isotherms of CLH, CLS and G-CLS, and pore size distribution (inset) of G-CLS

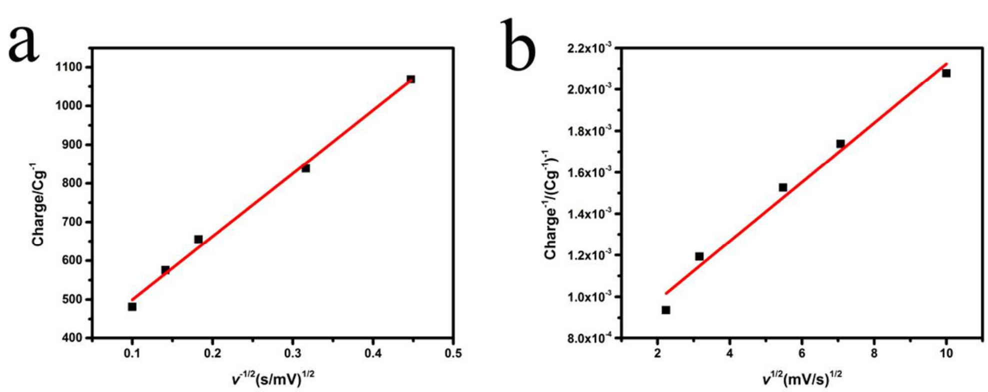

Figure S5. The relationship of (a) charge Q *vs* $v^{-1/2}$ and (b) Q^{-1} *vs* $v^{1/2}$

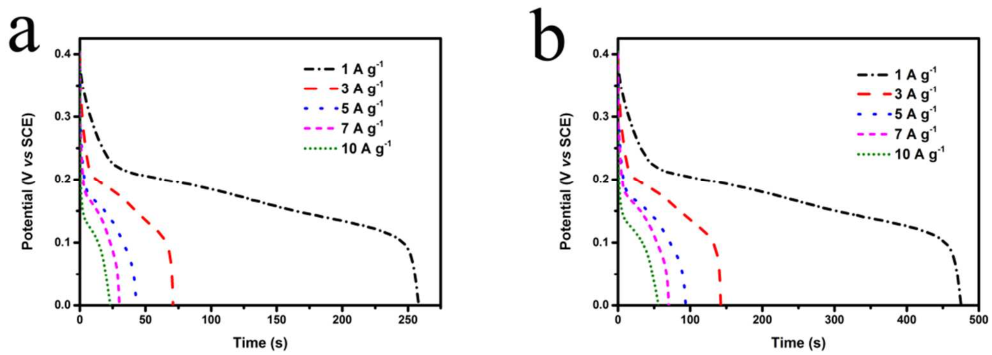

Figure S6. Discharge curves of CLH (a) and CLS (b) at various densities.

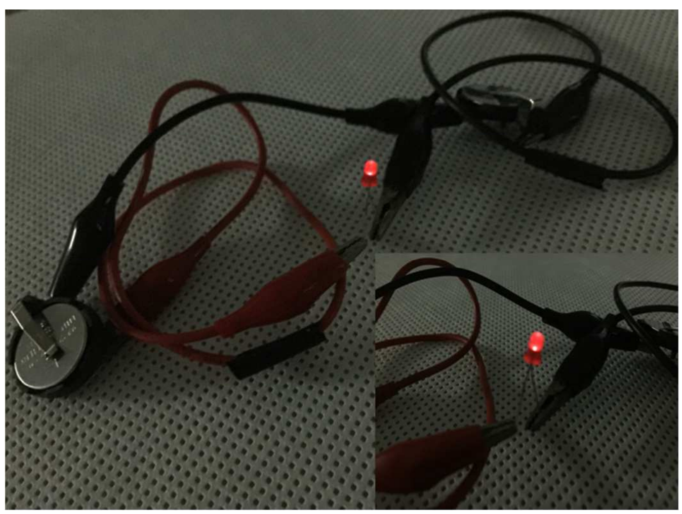

Figure S7. A red LED powered by two G-CLS//rGO ASC devices.

Table S1 Comparison of the electrochemical performance of the reported LDHs and graphene based electrodes in recent three years and the present work.

Ref.	Electrode materials	Electrolyte	Specific capacitance (F/g)	Rate performance	Capacitance retention (cycles)
This work	Graphene encapsulated Carbon shphere@NiAl LDH	6 M KOH	1710.5 (1 A/g)	86.4% (10 A/g)	77.9% (8000)
1[1]	Electrostatic self-assembly rGO/CoAl LDH	6 M NaOH	825 (1 A/g)	62.3% (8 A/g)	89.3% (4000)
2[2]	NiAl LDH/graphene	3 M KOH	915 (2 A/g)	16.8% (10 A/g)	95% (1500)
3[3]	NiAl LDH/rGO	6 M KOH	1630 (1 A/g)	49.4% (8 A/g)	65% (2000)
4[4]	3D hierarchical rGO LDH	1 M KOH	621 (1 A/g)	65.5% (8 A/g)	92.4% (5000)
5[5]	Porous CoAl LDH/graphene	6 M KOH	479.2 (1 A/g)	71.2% (10 A/g)	93.9% (2000)
6[6]	CoAl LDH/graphene 3D porous hybrid aerogel	6 M KOH	640 (1 A/g)	47.7% (20 A/g)	97% (10000)
7[7]	Porous NiCo LDH/rGO	3 M KOH	1911 (2 A/g)	76.9% (20 A/g)	74% (1000)
8[8]	GNS/NiCoAl-LDH	6 M KOH	1962 (1 A/g)	60.1% (10 A/g)	96.2% (2000)
9[9]	rGO/Ni1−xCoxAl LDH	6 M KOH	1902 (1 A/g)	74.8% (10 A/g)	62% (1500)
10[10]	3D NiCoMn LDH/rGO	2 M KOH	912 (1 A/g)	81.4% (10 A/g)	63.3% (5000)
11[11]	CoAl LDH nanosheets/rGO	2 M KOH	1296 (1 A/g)	42.7% (10 A/g)	90.5% (1000)
12[12]	CoAl LDH/graphene nanosheets	6 M KOH	974.1 (10 mV s^{-1})	30.1% (100 mV s^{-1})	94% (3000)
13[13]	NiCo LDH nanoflakes/graphene sheets	6 M KOH	1980.7 (1 A/g)	64.4% (15 A/g)	102.9% (1500)
14[14]	Ultrathin NoCoAl LDH/rGO	6 M KOH	1544 (1 A/g)	70% (40 A/g)	100% (2000)
15[15]	LDH monolayer/graphene	6 M KOH	379.7 (1 mA/cm^2)	81.1% (10 mA/cm^2)	88.1% (1000)
16[16]	CoMn LDH/rGo	2 M KOH	1635 (1 A/g)	71% (10 A/g)	100% (10000)

Table S2 Fittted values of R_s, R_{ct}, C *dl*, W, and C *ps* through CNLS fitting of the EIS based on the proposed equivalent circuit in Figure 5e

Electrodes	R_s (Ω)	R_{ct} (Ω)	*C dl* (F)	*W*(Ω)	*C ps* (F)
CLH	1.69	1.634	0.007545	1.214	3.678
CLS	1.53	0.896	0.006443	1.029	3.754
G-CLS	0.86	0.662	0.01049	0.7328	2.865

REFERENCES

(1) Zhong, Y. Y.; Liao, Y. Q.; Gao, A. M.; Hao, J. N.; Shu, D.; Huang, Y. L.; Zhong, J.; He, C.; Zeng, R. H., Supercapacitive Behavior of Electrostatic Self-Assembly Reduced Graphene Oxide/CoAl-Layered Double Hydroxides Nanocomposites. *J Alloy Compd* **2016,** *669*, 146-155.

(2) Wimalasiri, Y.; Fan, R.; Zhao, X. S.; Zou, L., Assembly of Ni-Al Layered Double Hydroxide and Graphene Electrodes for Supercapacitors. *Electrochim Acta* **2014,** *134*, 127-135.

(3) Li, M.; Cheng, J. P.; Fang, J. H.; Yang, Y.; Liu, F.; Zhang, X. B., NiAl-Layered Double Hydroxide/Reduced Graphene Oxide Composite: Microwave-Assisted Synthesis and Supercapacitive Properties. *Electrochim Acta* **2014,** *134*, 309-318.

(4) Hao, J. H.; Yang, W. S.; Zhang, Z.; Lu, B. P.; Ke, X.; Zhang, B. L.; Tang, J. L., Facile Synthesis of Three Dimensional Hierarchical Co–Al Layered Double Hydroxides on Graphene as High-Performance Materials for Supercapacitor Electrode. *J Colloid Interface Sci***2014,** *426*, 131–136.

(5) Zhang, L. J.; Hui, K. N.; Hui, K. S.; Lee, H., Facile Synthesis of Porous CoAl-Layered Double Hydroxide/Graphene Composite with Enhanced Capacitive Performance for Supercapacitors. *Electrochim Acta* **2015,** *186*, 522-529.

(6) Zhang, A. L.; Wang, C.; Xu, Q. J.; Liu, H. M.; Wang, Y. G.; Xia, Y. Y., A Hybrid Aerogel of Co-Al Layered Double Hydroxide/Graphene with Three-Dimensional Porous Structure as A Novel Electrode Material for Supercapacitors. *Rsc Adv* **2015,** *5* (33), 26017-26026.

(7) Cai, X. Q.; Shen, X. P.; Ma, L. B.; Ji, Z. Y.; Xu, C.; Yuan, A. H., Solvothermal Synthesis of NiCo-Layered Double Hydroxide Nanosheets Decorated on RGO Sheets for High Performance Supercapacitor. *Chem Eng J* **2015,** *268*, 251-259.

(8) He, F.; Hu, Z. B.; Liu, K. Y.; Guo, H. J.; Zhang, S. R.; Liu, H. T.; Xie, Q. L., Facile Fabrication of GNS/NiCoAl-LDH Composite as An Advanced Electrode Material for High-Performance Supercapacitors. *J Solid State Electr* **2015,** *19* (2), 607-617.

(9) Xu, J.; Gai, S. L.; He, F.; Niu, N.; Gao, P.; Chen, Y. J.; Yang, P. P., Reduced Graphene Oxide/$Ni_{1-x}Co_x$Al-Layered Double Hydroxide Composites: Preparation and High Supercapacitor Performance. *Dalton T* **2014,** *43* (30), 11667-11675.

(10) Li, M.; Cheng, J. P.; Liu, F.; Zhang, X. B., 3D-architectured Nickel-cobalt-manganese Layered Double Hydroxide/Reduced Graphene Oxide Composite for High-Performance Supercapacitor. *Chem Phys Lett* **2015,** *640*, 5-10.

(11) Huang, Z. C.; Wang, S. L.; Wang, J. P.; Yu, Y. M.; Wen, J. J.; Li, R., Exfoliation-restacking Synthesis of CoAl-Layered Double Hydroxide Nanosheets/Reduced Graphene Oxide Composite for High Performance Supercapacitors. *Electrochim Acta* **2015,** *152*, 117-125.

(12) Kim, Y.; Kim, S., Direct Growth of Cobalt Aluminum Double Hydroxides on Graphene Nanosheets and the Capacitive Properties of the Resulting Composites. *Electrochim Acta* **2015,** *163*, 252-259.

(13) Yan, T.; Li, R. Y.; Li, Z. J., Nickel-Cobalt Layered Double Hydroxide Ultrathin Nanoflakes Decorated on Graphene Sheets with A 3D Nanonetwork Structure as Supercapacitive Materials. *Mater Res Bull* **2014,** *51*, 97-104.

(14) Zheng, C. H.;Yao, T.; Xu, T. R.; Wang, H. A.; Huang, P. F.; Yan, Y.; Fang, D. L., Growth of Ultrathin Ni-Co-Al Layered Double Hydroxide on Reduced Graphene Oxide and Superb Supercapacitive Performance of the Resulting Composite. *J Alloy Compd* **2016,** *678*, 93–101.

(15) Du, M.; Yin, X. S.; Tang, C. H.; Huang, T. J.; Gong, H., Takovite-derived 2D Ni/Al Double Hydroxide Monolayer and Graphene Hybrid Electrodes for Electrochemical Energy Storage Applications with High Volumetric Capacitance. *Electrochim Acta* **2016,** *190*, 521-530.

(16) Li M.; Chen J. P.; Wang J.; Liu F.; Zhang X. B., The Growth of Nickel-Manganese and Cobalt-Manganese Layered Double Hydroxides on Reduced Graphene Oxide for Supercapacitor. *Electrochim Acta* **2016,** *206*, 108–115.