Electronic Supplementary Information

Reduced graphene oxide/$Ni_{1-x}Co_xAl$-layered double hydroxide composites: preparation and high supercapacitor performance

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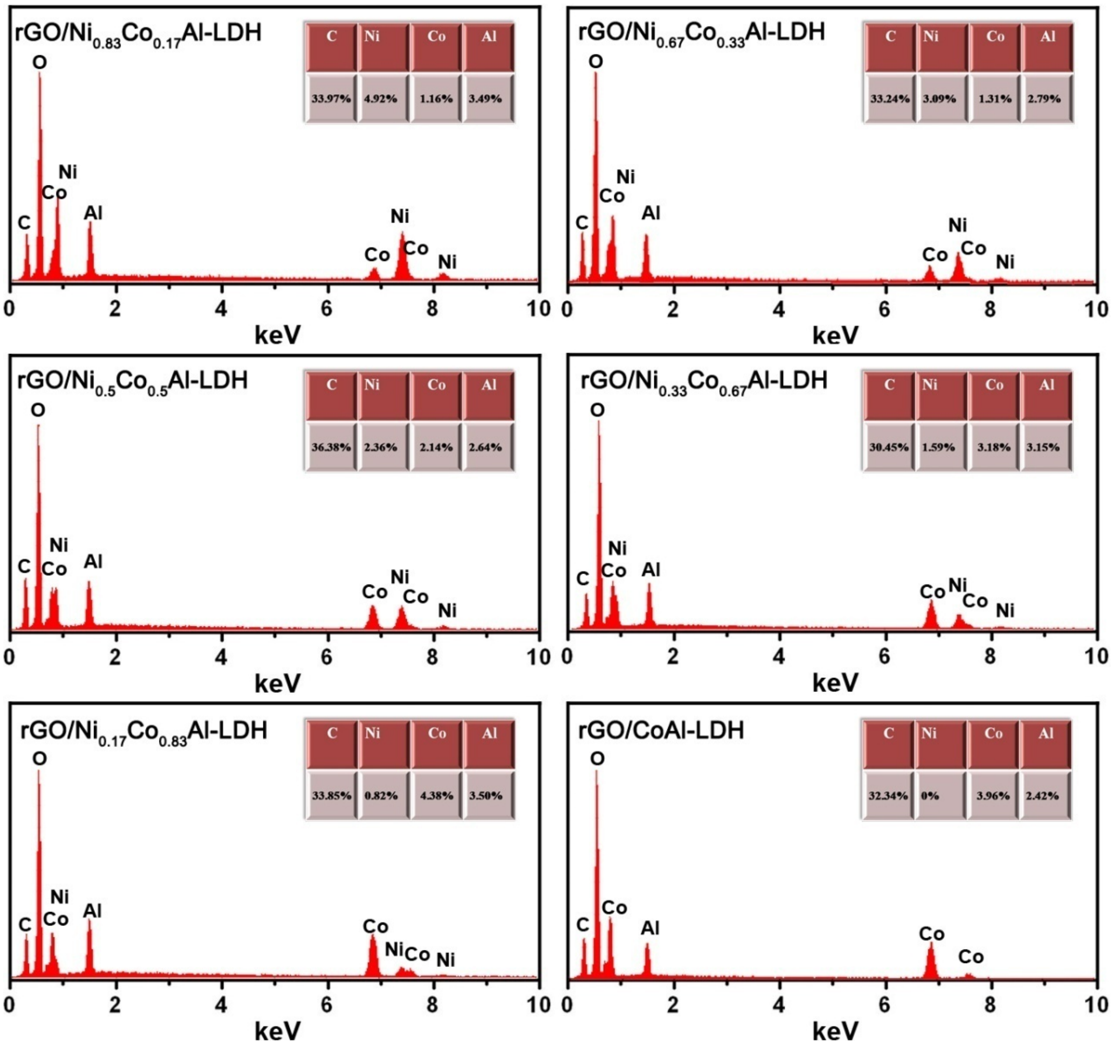

Fig. S1 EDS analysis of rGO/$Ni_{1-x}Co_x$Al-LDH with different Co doping.

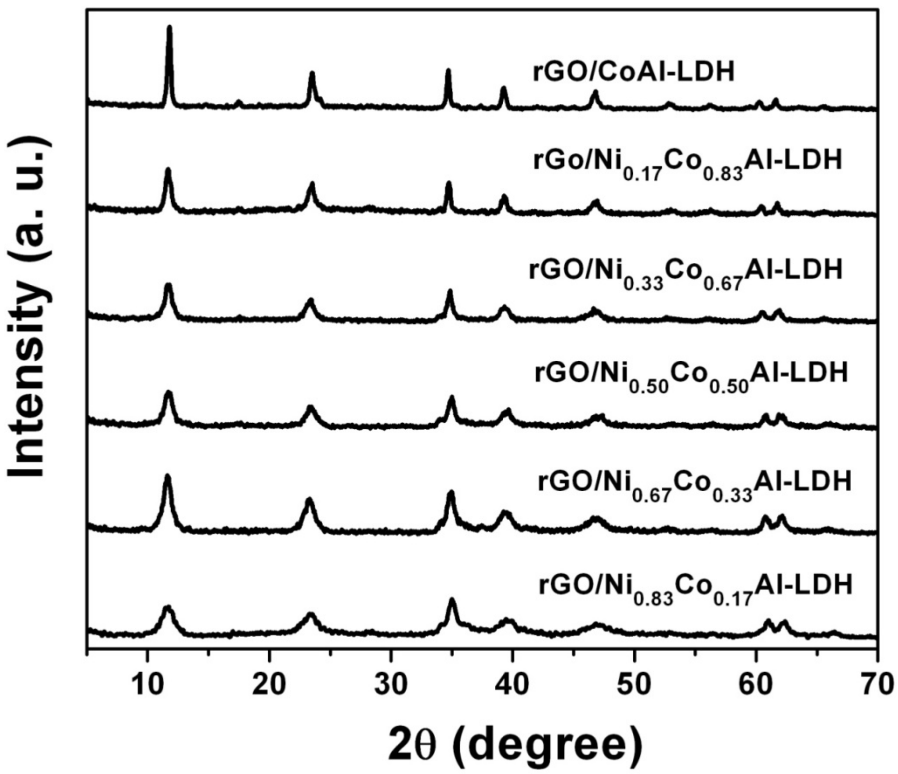

Fig. S2 The XRD patterns of rGO/$Ni_{1-x}Co_x$Al-LDH with different cobalt doping prepared by the hydrothermal process.

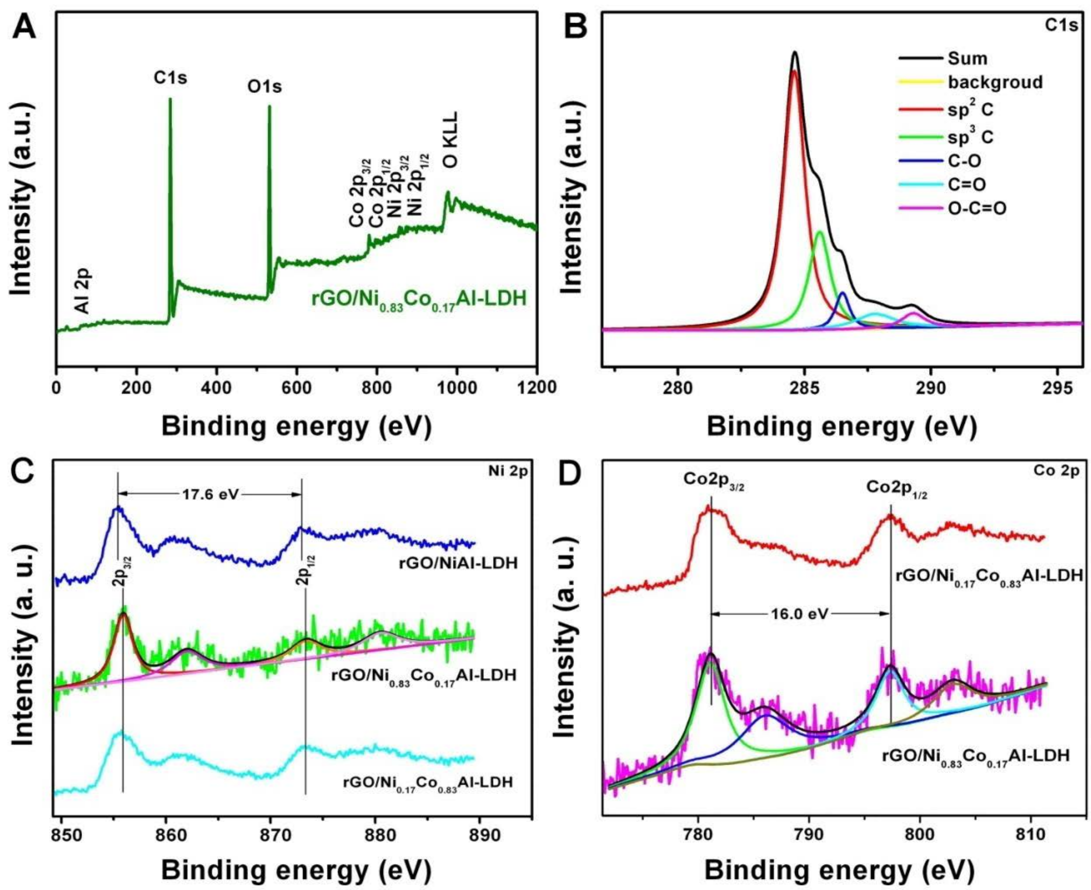

Fig. S3 Survey XPS spectrum (A); the core-level C 2p (B); the core-level Ni 2p (C) for undoped rGO/NiAl-LDH, RGL1 and RGL5; and the core level Co 2p (D) spectra of RGL1 and RGL5.

The oxidation states of Ni and Co can be determined by the further analyses of XPS spectra (Fig. S3C and D). In the Ni 2p XPS spectrum of rGO/NiAl-LDH (Fig. S3C), the two major peaks around 873.5 and 855.9 eV corresponding to Ni $2p_{1/2}$ and Ni $2p_{3/2}$ with a spin-energy separation of 17.6 eV are characteristics of Ni^{2+} in LDHs.[1] The Ni 2p spectra of rGO/$Ni_{0.83}Co_{0.17}$Al-LDH and rGO/$Ni_{0.17}Co_{0.83}$Al-LDH show the much similar shape and spin-energy separation to those of rGO/NiAl-LDH, indicating the doping of Co has not influenced the oxidation states of Ni. And Ni also keeps in the divalent state in all the samples.

A similar case is observed in Co 2p XPS spectra (Fig. S3D), which has the same characteristic peaks at 781.3 and 797.3 eV corresponding to the $2p_{3/2}$ and $2p_{1/2}$ spin-orbit peaks, and same spin-energy separation (16.0 eV) of Co^{2+}.[2] The results indicate the change of Co doping content also has not affected the oxidation states of Co, which may be due to the much similar ion radius of the doped Co ion. It is believed that the coexisting Ni^{2+} and Co^{2+} metal ions in rGO/NiCoAl-LDH composites are electrochemical active sites, which results in high electrochemical performance of graphene based composite.[3]

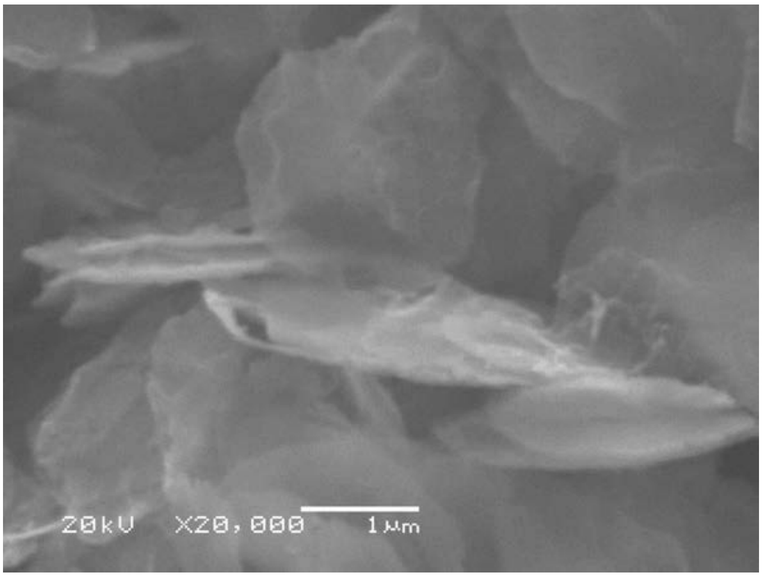

Fig. S4 SEM image of rGO/CoAl-LDH obtained by the similar process without Ni doping.

Table S1 EDS analysis of Co, Ni, Al and C in rGO/Ni_xCo_yAl-LDHs obtained with different molar ratios of $Ni(NO_3)_2.6H_2O$ and $Co(NO_3)_2.6H_2O$

Ni:Co	C	Ni	Co	Al
5:1	33.97%	4.92%	1.16%	3.49%
2:1	33.24%	3.09%	1.31%	2.79%
1:1	36.38%	2.36%	2.14%	2.64%
1:2	30.45%	1.59%	3.18%	3.15%
1:5	33.85%	0.82%	4.38%	3.50%
0	32.34%	0	3.96%	2.42%

Table S2 The theoretical values of Ni/Co molar ratios in rGO/Ni_xCo_yAl-LDH and the measured values by the ICP analysis

Samples	theoretical value (Ni: Co)	measured value (Ni: Co)	measured value (Ni: Co): Al
rGO/$Ni_{0.83}Co_{0.17}$Al-LDH	5	4.98	3.64
rGO/$Ni_{0.67}Co_{0.33}$Al-LDH	2	2.09	3.19
rGO/$Ni_{0.50}Co_{0.50}$Al-LDH	1	1.01	3.50
rGO/$Ni_{0.33}Co_{0.67}$Al-LDH	0.5	0.54	3.25
rGO/$Ni_{0.17}Co_{0.83}$Al-LDH	0.2	0.20	2.83

References

1 X.-L. Huang, X. Zhao, Z.-L. Wang, L.-M. Wang and X.-B. Zhang, *J. Mater. Chem.*, 2012, **22**, 3764.

2 J. W. Lee, T. Ahn, D. Soundararajan, J. M. Ko and J.-D. Kim, *Chem. Commun.*, 2011, **47**, 6305.

3 Yang, C. Yu, X. Fan, Z. Ling, J. Qiu, Y. Gogotsi, *J. Mater. Chem. A*, 2013, **1** 1963.