Electronic Supplementary Information

Nanohybrids from NiCoAl-LDH Coupled with Carbon for Pseudocapacitors: Understanding the Role of Nano-structured Carbon

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Surface modification of MWCNT

The multiwall CNT with a diameter of 10–30 nm, a length of 1–2 μm, and a specific surface area of ca. 50 $m^2 g^{-1}$ were treated with a mixture of concentrated nitric acid and sulfuric acid (volume ratio 1:3) by refluxing in a round bottom flask at 100 ^oC for 4 h, then filtered and washed with deionized water to neutrality. Finally, the collected powder was dried at 80 ^oC for 12 h in the oven, yielding the modified CNT.

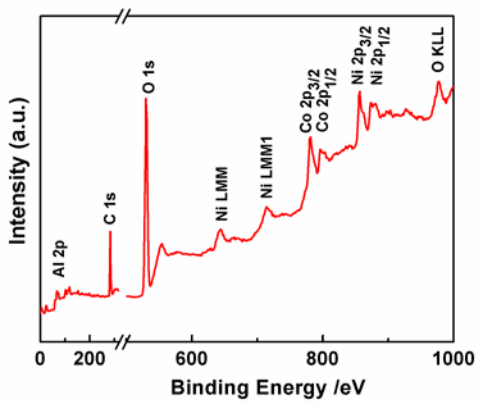

Fig. S1 XPS spectra of survey for the single NiCoAl-LDH sample.

Table S1 The relative atom contents of elements in NiCoAl-LDH nanosheet from XPS data.

Element	C	O	Al	Ni	Co
At%	39.90	46.87	3.93	4.78	4.52

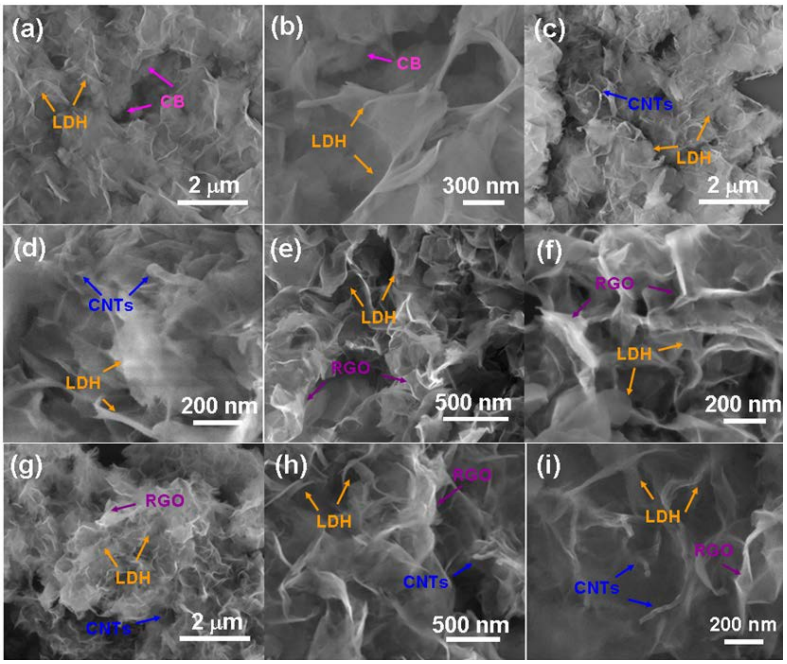

Fig. S2 FE-SEM images of the LDH-CB (a, b), LDH-CNT (c, d), LDH-RGO (e, f), and LDH-CNT/RGO nanohybrids(g, h, i).

Table S2 Pore structure parameters of the LDH-carbon nanohybrids.

Sample	Specific surface area (S_{BET}) (m^2 g^{-1})	Average pore size (nm)	Pore volume (cm^3 g^{-1})
LDH-CB	157	18	0.70
LDH-CNT	102	21	0.53
LDH-RGO	199	17	0.87
LDH-CNT/RGO	177	23	1.02

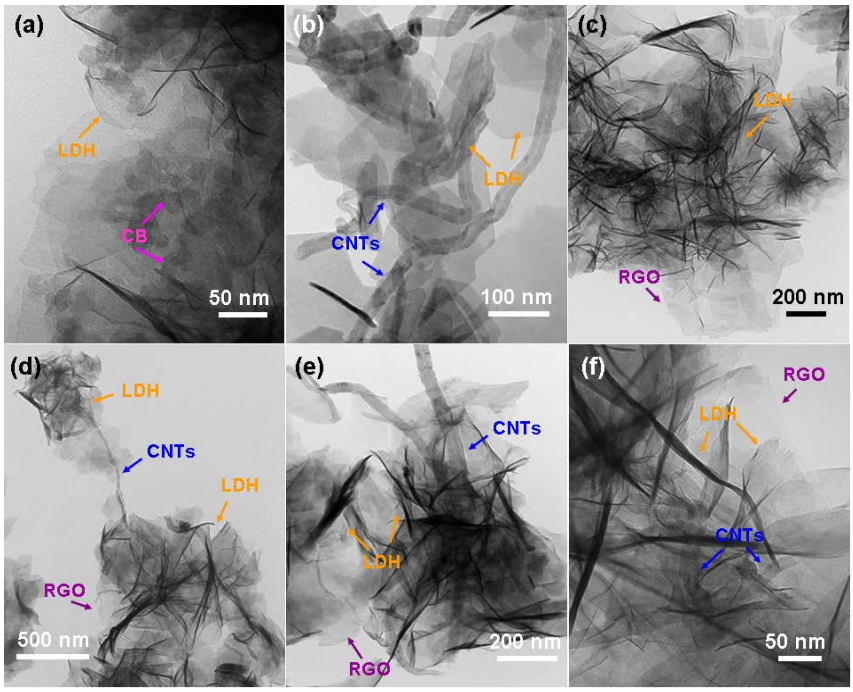

Fig. S3 TEM images of (a) the LDH-CB, (b) LDH-CNT, (c) LDH-RGO, and (d) (e) (f) LDH-CNT/RGO nanohybrids.

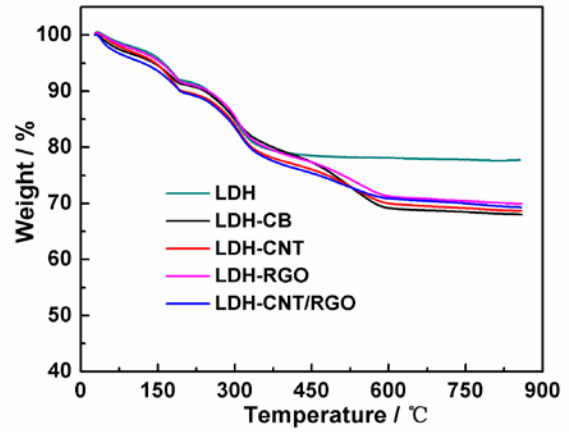

Fig. S4 TGA curves of the LDH, LDH-CB, LDH-CNT, LDH-RGO, and LDH-CNT/RGO nanohybrids, in which it can be clearly seen that a weight loss of nanohybrids mainly occurs between 350 and 600 ^{o}C in comparison to that of the single LDH sample, corresponding to the oxidation and decomposition of carbon species in air. On the basis of this, the weight fraction of carbon species in the LDH-carbon nanohybrids is calculated and exhibited in Table S2. It is also noted that the weight fraction of carbon species in the LDH-carbon nanohybrids has relatively small changes and varies in a range of 10-13 wt%. This will also be in favor of having insight into the effect of carbon structure and properties within electrode matrix on the electrochemical activity.

Table S3 The mass fraction of carbon species in the LDH-carbon nanohybrids.

Sample	LDH	LDH-CB	LDH-CNT	LDH-RGO	LDH-CNT/RGO
Weight retention rate (from 25 to 850℃)	77.7%	68.0%	68.6%	69.9%	69.3%
Mass fraction of carbon	0	12.5%	11.7%	10.0%	10.8%

Table S4 Specific capacitance of the samples at various current densities.

Current density ($A\ g^{-1}$)	LDH[1] ($F\ g^{-1}$)	LDH-CB ($F\ g^{-1}$)	LDH-CNT ($F\ g^{-1}$)	LDH-RGO ($F\ g^{-1}$)	LDH-CNT/RGO ($F\ g^{-1}$)
1	950	1060	1003	936	1188
10	232	498	546	437	850
Retention rate	24%	47%	54%	47%	72%

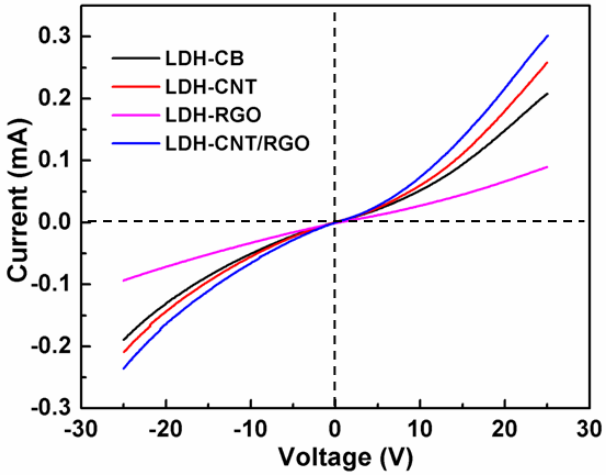

Fig. S5 The current–voltage (*I-V*) curve plots of LDH-CB, LDH-CNT, LDH-RGO, and LDH-CNT/RGO nanohybrids, in which it can be found that the currents increase in turn and follow the order of LDH-GO, LDH-CB, LDH-CNT, and LDH-CNT/RGO nanohybrids under the same applied voltages, suggesting that the electrical conductivity of nanohybrids is ever-increasing. This will be favorable for fast transferring electrons to the current collector and is also why the LDH-CNT/RGO nanohybrids demonstrate the excellent electrochemical performance.

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