Electronic Supplementary Information

Co-doped Ni hydroxide and oxide nanosheet networks: Laser-Assisted Synthesis, Effective Doping, and Ultrahigh Pseudocapacitor Performance

Dewei Liang[a,b]*, Shouliang Wu*[a]*, Jun Liu*[a]*, Zhenfei Tian*[a]*, Changhao Liang*[a,b, *]

[a]Key Laboratory of Materials Physics and Anhui Key Laboratory of Nanomaterials and Nanotechnology, Institute of Solid State Physics, Hefei Institutes of Physical Science, Chinese Academy of Sciences, Hefei 230031, China.

[b]Department of Materials Science and Engineering, University of Science and Technology of China, Hefei 230026,China.

1. Preparation of pure $Ni(OH)_2$ nanosheet network

Pure $Ni(OH)_2$ nanosheet network was synthesized using a LAL technique. In a typical process, a nickel metal plate (99.99% purity) was fixed on a supporter in a vessel that filled with 15 mL 0.2 M $Na_2S_2O_3$ solution. Upon ablation of the metal plate, the vessel was rotated (10 rpm) by a motorized tunable stage. The metal plate was ablated using a Nd:YAG pulsed laser at 1064 nm wavelength with an energy density of 130 mJ per pulse and a pulse duration of 10 ns. After ablation for 20 min, the suspension was stored in a container and aged at room temperature for 1 week. After sufficient growth, the obtained gray-color precipitates were collected and dried in a vacuum chamber at 50 °C overnight (Fig. S1).

2. Characterization of pure $Ni(OH)_2$ nanosheet

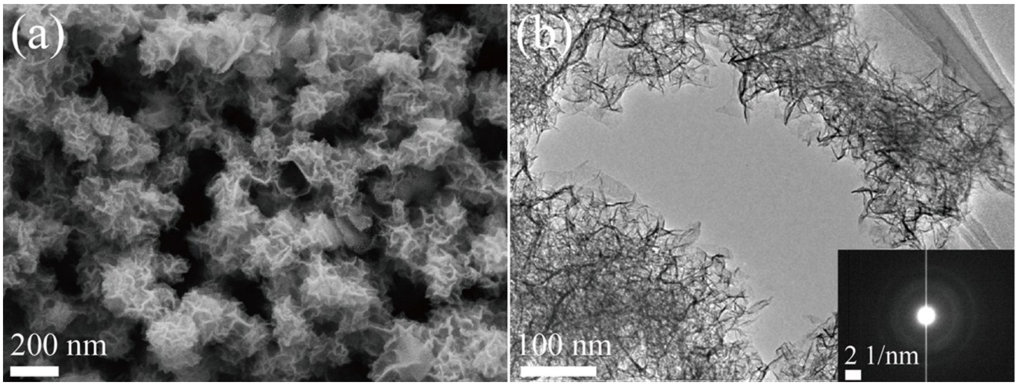


Fig. S1. (a) FESEM image, (b) TEM image and corresponding SAED pattern (inset) of pure $Ni(OH)_2$. The morphology of pure $Ni(OH)_2$ is very similar to CN-0.1, both of them are composed of nanosheet network. The diffused halo of the selected-area electron diffraction (SAED) pattern in the inset of Figure S1(b) also indicates the poor crystallinity and polycrystalline characteristics of the sample.[1]

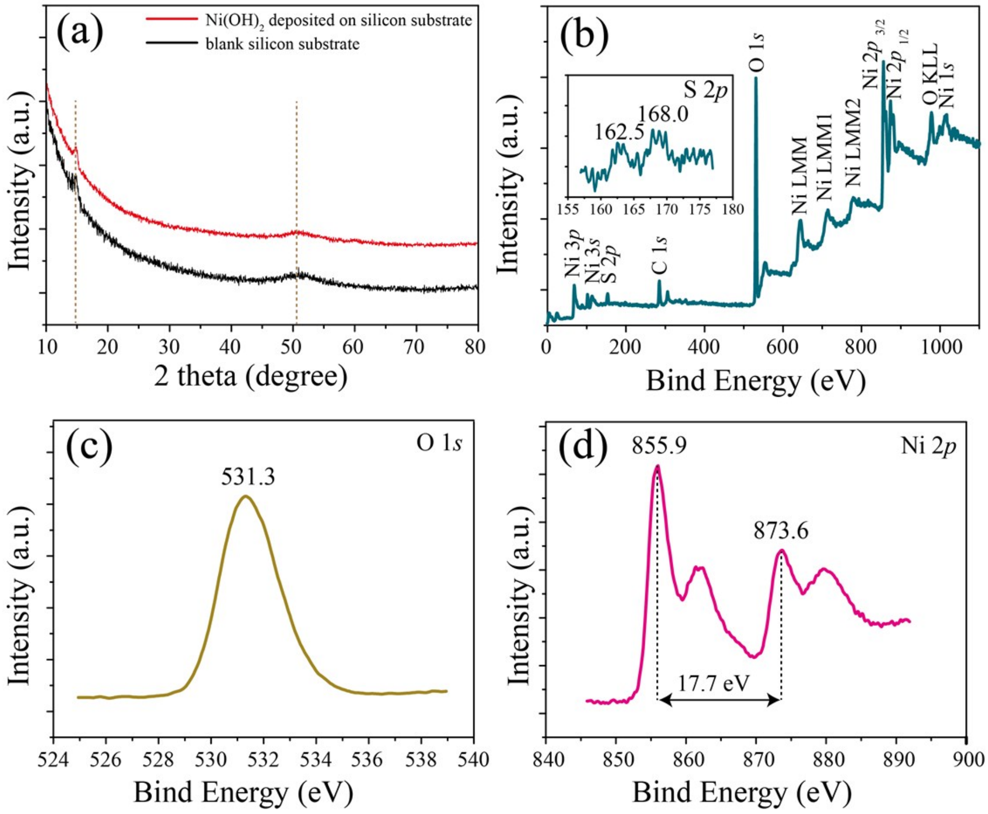
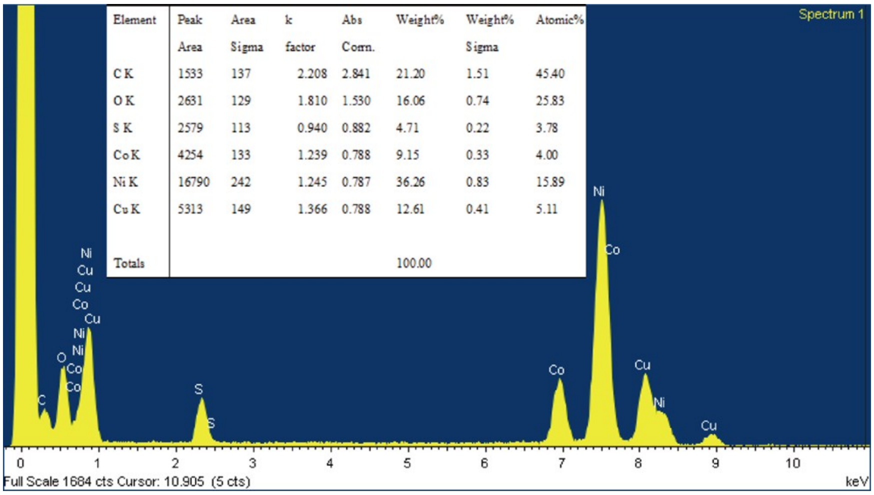


Fig. S2. (a) XRD spectra and (b) the survey XPS spectrum of pure $Ni(OH)_2$, and high-resolution spectra of (c) O1*s*, (d) Ni2*p*. As demonstrated by XRD measurement, the curve of the product shows no obvious sharp peak, indicating its amorphous structure. According to the *Handbook of X-ray Photoelectron Spectroscopy*, the S peak inset of Fig. S2(b) can be associated with the S 2*p* from the intercalated SO_4^{2-} group between the $Ni(OH)_2$ layers or the absorbed $S_2O_3^{2-}$ on the surface. The binding energy of 531.3 eV for O 1*s* can be mainly derived from the hydroxyl group (O-H) with a binding energy of ~531 eV [Fig. S2(c)]. Fig. S2(d) exhibits the high-resolution Ni $2p_{3/2}$ and Ni $2p_{1/2}$ peaks centered at 855.7 and 873.6 eV respectively, with a spin-energy separation of 17.9 eV, which is in good agreement with reported data for Ni^{2+}. These results together demonstrate the product is amorphous $Ni(OH)_2$.

3. Additional characterization of CN-0.1 and CN-0.2



Element	Peak Area	Area Sigma	k factor	Abs Corrn.	Weight%	Weight% Sigma	Atomic%
C K	1533	137	2.208	2.841	21.20	1.51	45.40
O K	2631	129	1.810	1.530	16.06	0.74	25.83
S K	2579	113	0.940	0.882	4.71	0.22	3.78
Co K	4254	133	1.239	0.788	9.15	0.33	4.00
Ni K	16790	242	1.245	0.787	36.26	0.83	15.89
Cu K	5313	149	1.366	0.788	12.61	0.41	5.11
Totals					100.00		

Fig. S3. EDX spectrum of sample CN-0.1. From the picture, we can see that the atomic ratio of the Co and Ni is approximately equal to 1:4.

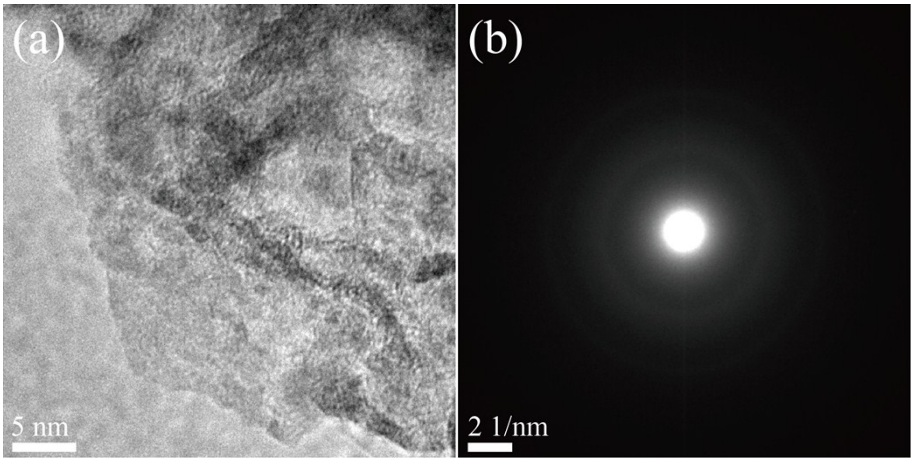


Fig. S4. (a) HRTEM image and (b) SAED pattern of sample CN-0.2. No obvious lattice fringe can be found in the HRTEM image, together with the diffused halo of the selected-area electron diffraction (SAED) pattern, these results suggest that the products synthesized in this way tend to be a low crystalline or amorphous structure.

4. Electrochemical test of pure $Ni(OH)_2$

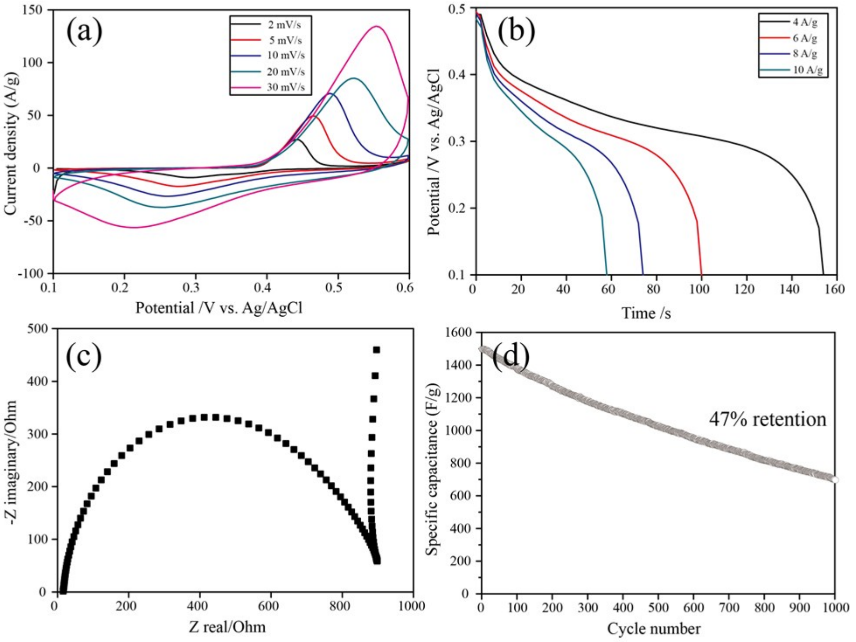


Fig. S5. (a) CV curves of pure $Ni(OH)_2$ at scan rate of 2, 5, 10, 20 and 30 mV/s; (b) galvanostatic discharging curve of pure $Ni(OH)_2$ at current densities of 4, 6, 8 and 10 A/g; (c) Nyquist plots of pure $Ni(OH)_2$; (d) cycling performance of pure $Ni(OH)_2$ at a discharge current density of 6 A/g. Cyclic voltammetry (CV), electrochemical impedance spectroscopy (EIS) and galvanostatic charge/discharge measurements were conducted in 1 M KOH solution.

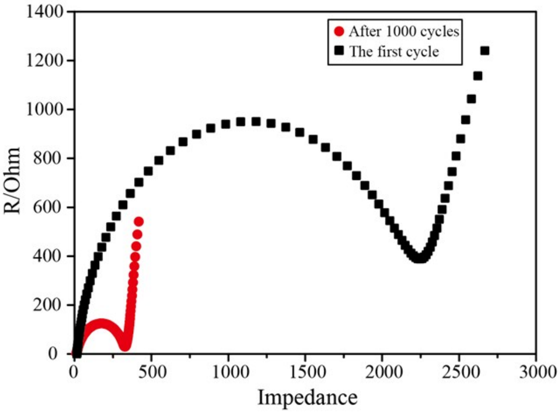


Fig. S6. Nyquist plots of the sample CN-0.2 before and after 1000 cycles.

References

[1] Wang, C. H.; Zhang, X.; Xu, Z. T. Ethylene Glycol Intercalated Cobalt/Nickel Layered Double Hydroxide Nanosheet Assemblies with Ultrahigh Specific Capacitance: Structural Design and Green Synthesis for Advanced Electrochemical Storage. *ACS Appl. Mater. Interfaces* 2015, 7, 19601-19610.