
Dewei Liang, Shouliang Wu, Jun Liu, Zhenfei Tian, Changhao Liang

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.

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 Na₂S₂O₃ 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.

**Fig. S2.** (a) XRD spectra and (b) the survey XPS spectrum of pure Ni(OH)$_2$, and high-resolution spectra of (c) O1s, (d) Ni2p. 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 2p from the intercalated SO$_4^{2-}$ group between the Ni(OH)$_2$ layers or the absorbed S$_2$O$_3^{2-}$ on the surface. The binding energy of 531.3 eV for O 1s 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

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