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# Controllable crystal growth of NiCo-LDH nanostructure anchored on $KCu_7S_4$ nanowires via facile solvothermal method for supercapacitor application

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# 1. Experimental

## *1.1 Materials*

All reagents used in experiment were of analytical purity and directly used without any further purification. $KCu_7S_4$ NWs were prepared by water bath method, which was reported previously.

## *1.2 Preparation of $KCu_7S_4$ nanowires*

In a typical process, 3.87 g of NaOH and 5.11 g of KOH with a Na/K atomic ratio of 51.5: 48.5 were dissolved in 30 mL of distilled water to form a homogeneous solution with the concentration of $OH^-$ ions of~6 mol $L^{-1}$. Then, 3 mmol of $CuCl_2 \cdot 2H_2O$, 0.3 mL of ethylene diamine, 12 mmol of $Na_2S \cdot 9H_2O$ and 3 mL of hydrazine hydrate were added into the vessel sequentially. After thorough stirring, the vessel was sealed and kept at 80 °C in a water bath for 1 h. The taupe sponge-like products were finally collected by centrifugation and washed with distilled water until the centrifugal liquid become neutral, then washed with absolute ethanol several times and dried in a vacuum at 60 °C for 4 h for further characterization.

## *1.3 Material Characterization*

The morphology and elemental composition of as-prepared samples were monitored by focused ion beam (Zeiss Auriga FIB/SEM) and transmission electron microscopy (TEM ZEISS LIBRA200) equipped with an energy dispersive X-ray spectrometry (EDS). The X-ray diffractometer (XRD, D/max 1200, Cu Kα) was used for the analysis of chemical composition and crystal structure. The FTIR were performed to gain the surface structural information, and the chemical bonding states was detected by XPS.

## *1.4 Electrochemical Characterization*

To evaluate the electrochemical performance of as-prepared samples, the electrochemical testing was performed on CHI660E electrochemical workstation in a standard three-electrode system under ambient temperature. The preparation method of working electrode is as follows: the mixture slurry of active materials ($KCu_7S_4$@NiCo-LDHs , 70%), carbon black (20%), and polyvinylidene fluoride (PVDF, 10%) in N-methyl-2-pyrrolidone (NMP) was coated on the current collector (Ni foam, 1*1 $cm^2$), then the electrode was vacuum dried at 120 ℃ for 12 h. After calculation, the net mass of $KCu_7S_4$@NiCo-LDHs loaded on each working electrode was about 1.5–2.0 mg. The counter and reference electrodes were platinum plate and saturated calomel electrode (SCE), respectively, with 6 M KOH aqueous solution as the electrolyte. The prepared $KCu_7S_4$@NiCo-LDH electrodes were examined by cyclic

voltammetry (CV) method with scanning rate of 10-100 mV $s^{-1}$ in the potential range from 0-0.5 V. The alvanostatic charge-discharge (GCD) tests were performed at current density of 2-10 A $g^{-1}$ in the potential range of 0-0.4V. The electrochemical impedance spectroscopy (EIS) was conducted in the frequency range between 100kHz and 0.01Hz with a perturbation amplitude of 5 mV versus the open-circuit potential.

The mass specific capacitance ($C_m$, F $g^{-1}$) and battery-type specific capacity (Q, mAh $g^{-1}$) of the fabricated $KCu_7S_4$@NiCo-LDH electrodes were calculated based on the obtained GCD plots using the equation (1):

$$C_m = \frac{I\Delta t}{m\Delta V} \qquad (1)$$

$$Q = \frac{I\Delta t}{3.6m} \qquad (2)$$

where I is the constant discharge current (A), $\Delta$t is the discharge time (s), m is the mass (g) of active materials, and $\Delta$V is the discharging potential window (V).

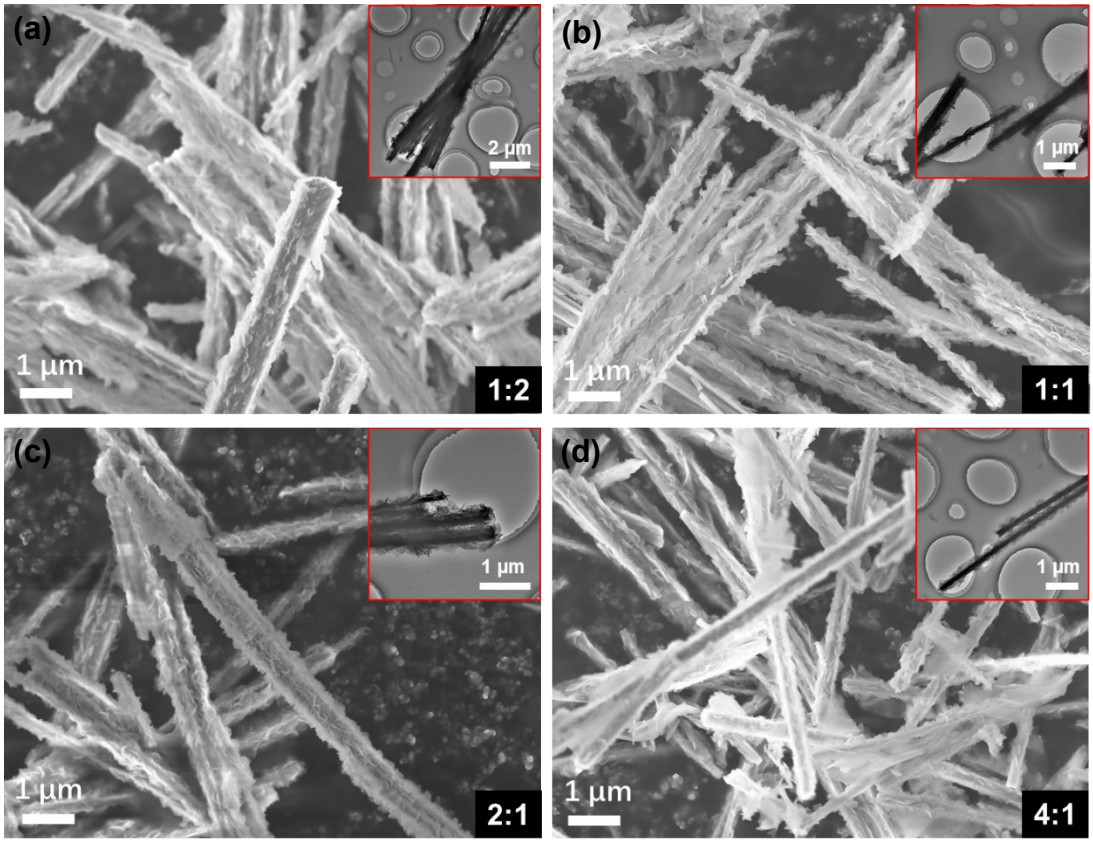


Fig S1. Typical SEM images of $KCu_7S_4$@NiCo-LDH with different Ni/Co ratio：(a) 1:2 (b) 1:2 (c) 2:1 (d) 4:1, the inset of SEM images shows the corresponding TEM images.

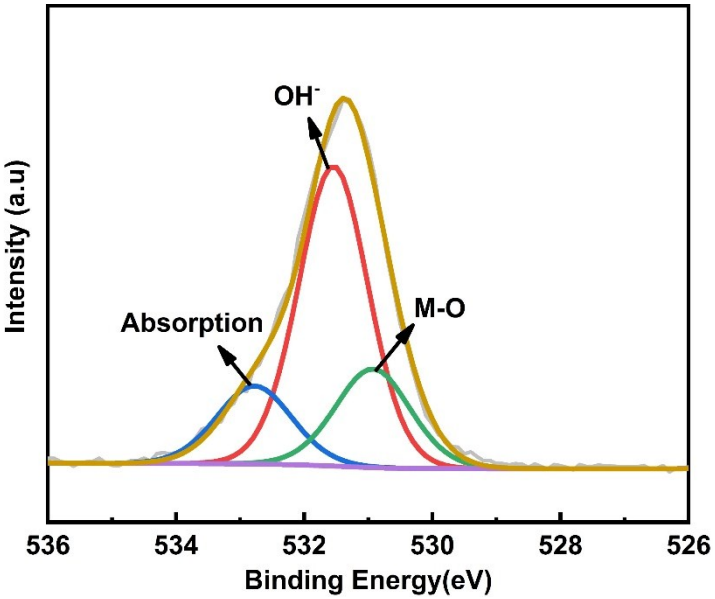

Fig S2. High-resolution XPS of O 1s.

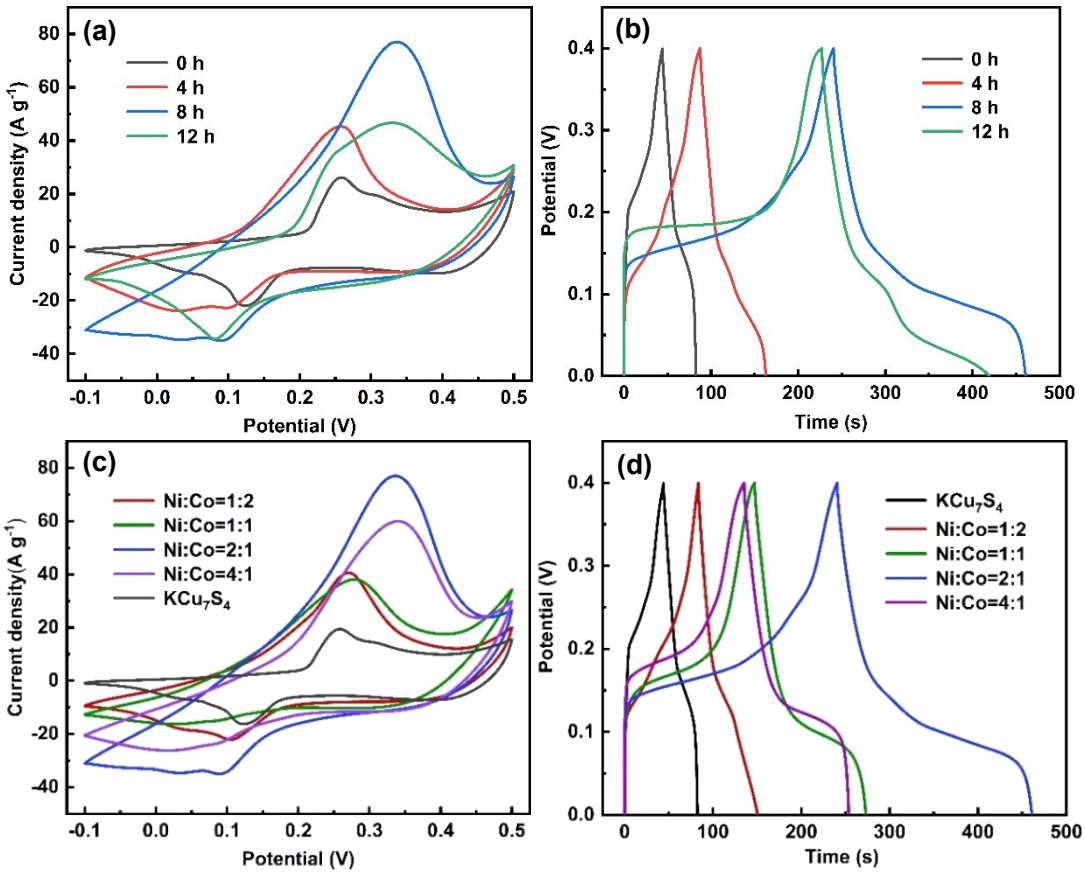


Fig S3. (a) Comparison of the CV curves of $KCu_7S_4$@NiCo-LDH electrodes with different reaction time at 50 mV $s^{-1}$ and (b) the corresponding GCD curves comparison at a charge current density of 2 A $g^{-1}$; (c) Comparison of the CV curves of $KCu_7S_4$@NiCo-LDH electrodes with different ratios of Ni/Co at 50 mV $s^{-1}$ and (d) the corresponding GCD curves comparison at a charge current density of 2 A $g^{-1}$;