Supporting information

Biomass-derived C/N co-doped Ni(OH)$_2$/Ni$_x$S$_y$ with sandwich structure for supercapacitors

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Synthesis of reduced graphene oxide hydrogel (rGH)
The graphene oxide (GO) suspension was prepared according to the Hummers’ method. Firstly, 1.5 mL GO suspension (5 mg g$^{-1}$) was transformed into glass tube. Then five tubes were transformed into the 100 mL Teflon-lined autoclave containing 13 mL deionized water and maintained at 150 $^\circ$C for 12 h. After the autoclave naturally cooled down to room temperature, the obtained rGH was immersed into distilled water for dialysis.

Synthesis of C/N co-doped nickel sulfides with different ratio of egg white to nickel nitrate
This synthesis process is the same as the preparation of C/N-Ni$_x$S$_y$, and the only difference is the proportions of raw materials. The feeding mass of egg white is 13 g. According to the different mass of nickel nitrate: 0.10, 0.12, 0.14, 0.2, 0.24, and 0.26 g, these obtained products are donated as C/N-Ni$_x$S$_y$-1, C/N-Ni$_x$S$_y$-2, C/N-Ni$_x$S$_y$-3, C/N-Ni$_x$S$_y$-4, C/N-Ni$_x$S$_y$-5, and C/N-Ni$_x$S$_y$-6, respectively.

![Fig. S1](image1) (a) SEM images of C/N-Ni$_x$S$_y$, and TEM images of (b) Ni$_x$S$_y$ nanoparticles and (c) bulk carbon in C/N-Ni$_x$S$_y$. 

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Fig. S2 SEM images of pure Ni(OH)$_2$.

Fig. S3 SEM image and the corresponding EDX mapping images of Ni, S, C, N and O elements in C/N-Ni(OH)$_2$/Ni$_x$S$_y$. 
Fig. S4 XRD patterns of product C/N-Ni$_x$S$_y$.

Fig. S5 XRD patterns of C/N-Ni$_x$S$_y$-1, C/N-Ni$_x$S$_y$-2, C/N-Ni$_x$S$_y$-3, C/N-Ni$_x$S$_y$-4, C/N-Ni$_x$S$_y$-5, and C/N-Ni$_x$S$_y$-6.

According to the XRD patterns of C/N-Ni$_x$S$_y$ and C/N-Ni$_x$S$_y$-1 to C/N-Ni$_x$S$_y$-6 (Fig. S3 and S4), the main components of products obtained without adding ammonia aqueous are Ni$_x$S$_y$. This result could further confirm that the egg white could be used as sulfur source to obtain Ni$_x$S$_y$. 
**Fig. S6** Standard XRD patterns of Ni$_x$S$_y$ in C/N-Ni(OH)$_2$/Ni$_x$S$_y$.

**Fig. S7** XPS spectra of C/N-Ni(OH)$_2$/Ni$_x$S$_y$, C/N-Ni$_x$S$_y$, and Ni(OH)$_2$: (a) survey spectra, and (b) O 1S spectra.
As shown in Fig. S8, Raman spectrum was also measured to characterize the C/N-Ni(OH)$_2$/Ni$_x$S$_y$ composite. The weak D and G bands can be observed, which attribute to $A_{1g}$ vibration mode of disordered carbon and $E_{2g}$ vibration mode of ordered graphitic carbon respectively, confirming the existence of carbon. Besides, the peaks around 509.1 and 1085.3 cm$^{-1}$ could be attributed to nickel hydroxide, and the peak around 321 cm$^{-1}$ could be attributed to the presence of nickel sulfide. In general, both the XPS and Raman spectra further confirm the successfully C/N co-doping and the formation of Ni(OH)$_2$ and Ni$_x$S$_y$ in C/N-Ni(OH)$_2$/Ni$_x$S$_y$.

Fig. S9 SEM image of rGH.
Fig. S10 (a) Galvanostatic charge-discharge curves and (b) cycling stability of C/N-Ni(OH)$_2$/Ni$_x$S$_y$ at different current densities.

Fig. S11 (a) CV curves at different scan rates and (b) galvanostatic charge-discharge curves at different current densities of rGH.

Fig. S12 (a) CV curves at different scan rates and (b) galvanostatic charge-discharge curves at different current densities of C/N-Ni(OH)$_2$/Ni$_x$S$_y$/rGH. (c) The capacitances at different current densities, and (d) Nyquist plots of C/N-Ni(OH)$_2$/Ni$_x$S$_y$/rGH, C/N-Ni$_x$S$_y$/rGH, and Ni(OH)$_2$/rGH.