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Synthesis of Three-Dimensional Mesoporous Cu-Al Layered Double Hydroxide/g-C_3N_4 Nanocomposites on Ni-foam for Enhanced Supercapacitors with Excellent Long-Term Cycling Stability

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Experimental

Synthesis of Ni-foam@Cu-Al LDH/g-C_3N_4 composite

The Cu-Al LDH and g-C_3N_4 composite on Ni-foam was prepared by a hydrothermal method with molar ratio 2:1 of Cu and Al, respectively. Typically, 0.144 g of $Cu(NO_3)_2.3H_2O$ (0.02 mol), 0.112 g of $Al(NO_3)_2.9H_2O$ (0.01 mol) and 0.027 g of urea (0.15 mol) were dissolved in 30 mL distilled water separately and stirred for 30 min. After this all solutions were mixed and again stirred for 1 h to form a clear mixed solution. The solution thus prepared was transferred into a 100 mL Teflon-lined stainless-steel autoclave. Then the Ni-foam@g-C_3N_4 was immersed into the above solution. The autoclave was sealed and heated at 130 ^{0}C for 4 hours. Then, the as-prepared nanocomposite was washed with distilled water and ethanol for several times and dried at 100 ^{0}C for 2 hours (denoted as Ni-foam@Cu-Al LDH/g-C_3N_4 2:1). To analyze the effect of molecular ratio, another one sample was again prepared under the same process of Ni-foam@Cu-Al LDH/g-C_3N_4 – 2:1 using the molecular ratio 1:2 of Cu and Al, respectively (denoted as Ni-foam@Cu-Al LDH/g-C_3N_4 – 1:2).

Results and discussion

Structure and morphology of the composites

The XRD patterns of Ni-foam, Ni-foam@g-C_3N_4 tested at different times are shown in Figure S1a and Figure S1b. The corresponding peaks of g-C_3N_4 on Ni-foam@g-C_3N_4 on both did not appear. This may be due the low weight loading of g-C_3N_4 on Ni-foam. But, the decreased peak height of Ni on Ni-foam@g-C_3N_4 comparison to Ni-foam indicated that the g-C_3N_4 was successfully deposited on Ni-foam during electrodeposition.

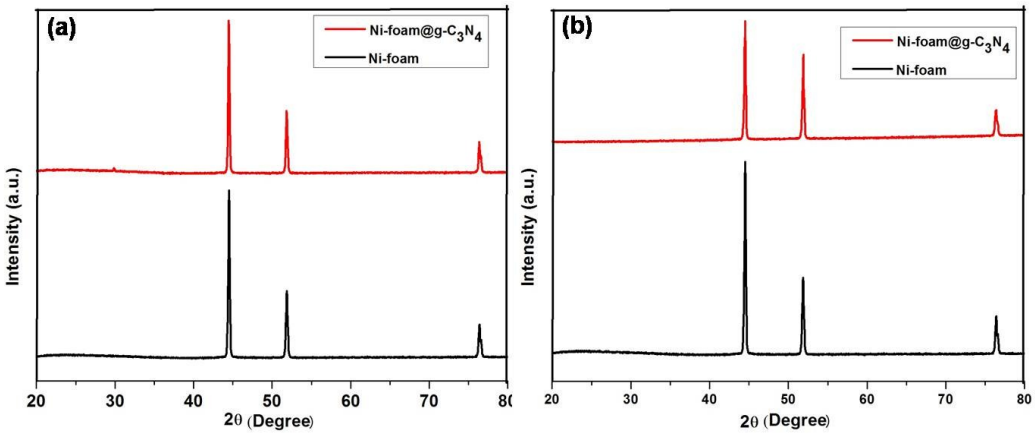

Figure S1: XRD patterns of the Ni-foam, and Ni-foam@g-C_3N_4

Electrochemical measurements

To evaluate the macroscopic electrochemical surface reactions at the electrode materials of the supercapacitor, we first carried out CV measurements of both samples. Figure S2a shows the CV curves of Ni-foam@Cu-Al LDH/g-C_3N_4 – 1:2 and Ni-foam@g-C_3N_4, Ni-foam@Cu-Al LDH/g-C_3N_4 – 2:1 at 50 mVs^{-1}. The CV curve of Ni-foam@Cu-Al LDH/g-C_3N_4 – 2:1 have well defined oxidation and reduction peaks in comparison to Ni-foam@Cu-Al LDH/g-C_3N_4 – 1:2, which represents the better pseudocapacitive characteristics of Ni-foam@Cu-Al LDH/g-C_3N_4 – 2:1 than Ni-foam@Cu-Al LDH/g-C_3N_4 – 1:2 electrode. The oxidation peaks were observed at 0.43 and 0.38 mV for Ni-foam@Cu-Al LDH/g-C_3N_4 – 1:2 and Ni-foam@Cu-Al LDH/g-C_3N_4 – 2:1, respectively. Correspondingly, the reduction peaks were 0.13 and 0.08 V for Ni-foam@Cu-Al LDH/g-C_3N_4 – 1:2 and Ni-foam@Cu-Al LDH/g-C_3N_4 – 2:1, respectively. In accordance with the CV curves, the remarkable difference in electrochemical surface reaction between these two samples can be observed. The Ni-foam@Cu-Al LDH/g-C_3N_4 – 2:1 exhibited significantly larger rectangular curve, which is due to a well hetero-junction formed between Cu-Al LDH and g-C_3N_4 sheets than Ni-foam@Cu-Al LDH/g-C_3N_4 – 1:2. Here, the peak current observed in Ni-foam@Cu-Al LDH/g-C_3N_4 – 2:1 nanocomposite is higher than other. The specific capacitances of samples from the CV curves were

calculated using equation 1. The mass of active electrode for both samples was calculated from the weight difference of Ni-foam before and after loading g-C_3N_4, and Cu-Al LDH, which were approximately same for Ni-foam@Cu-Al LDH/g-C_3N_4 – 1:2 and Ni-foam@Cu-Al LDH/g-C_3N_4 – 2:1. The Figure S2b shows the corresponding calculated specific capacitance for both samples, which were 530.3012 and 770.98 Fg^{-1} for Ni-foam@Cu-Al LDH/g-C_3N_4 – 1:2 and Ni-foam@Cu-Al LDH/g-C_3N_4 – 2:1, respectively. Here, the specific capacitance obtained from Ni-foam@Cu-Al LDH/g-C_3N_4 – 2:1 was significantly higher than the specific capacitance obtained from Ni-foam@Cu-Al LDH/g-C_3N_4 – 1:2, which may be due to the better pseudocapacitive characteristics and well hetero-junction formed between Cu-Al LDH and g-C_3N_4 sheets in Ni-foam@Cu-Al LDH/g-C_3N_4 – 2:1 than Ni-foam@Cu-Al LDH/g-C_3N_4 – 1:2. This result demonstrates that the molecular ratio of Cu and Al has momentous effect on the electrochemical performance of the nanocomposites.

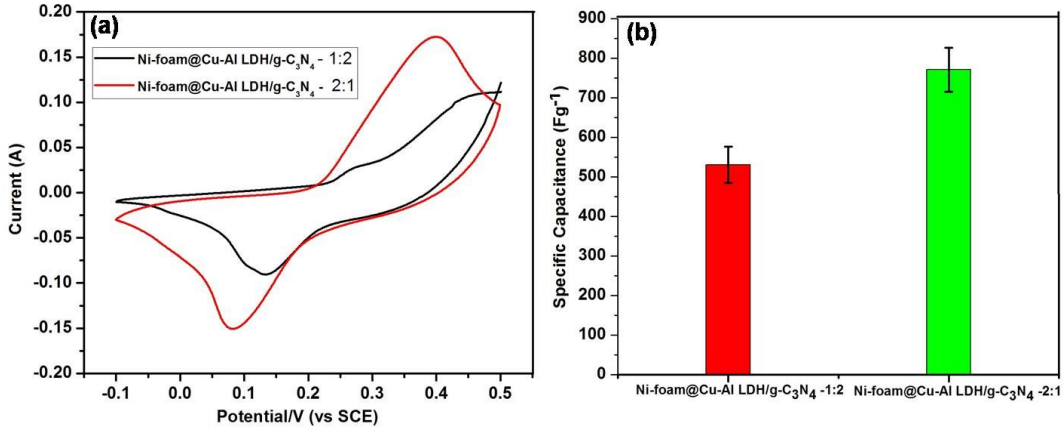

Figure S2: CV curves of the samples at scan rate of 50 mVs^{-1} in 6 M KOH electrolyte (a), Specific capacitance calculated from CV curves (b)