Directly Embedded $Ni_3S_2/Co_9S_8@S$ -Doped Carbon Nanofiber Networks as Free-standing Anode for Lithium-ion Batteries

Zizhou He,^a Hui Guo,^{a h} Jed D LaCoste,^a Ryan A Cook,^a Blake Hussey,^a Xu Zhang,^c Daniel Dianchen Gang,^c Ji Hao,^d Liang Chen,^e Peter Cooke,^f Hui Yan,^b Ling Fei *^a

- a. Department of Chemical Engineering, Institute for Materials Research and Innovations, University of Louisiana at Lafayette, Lafayette, LA 70504, United States E-mail: ling.fei@louisiana.edu
- b. Department of Chemistry, University of Louisiana at Lafayette, Lafayette, LA 70504, United States
- c. Department of Civil Engineering, University of Louisiana at Lafayette, Lafayette, LA, 70504, United States
- d. National Renewable Energy Laboratory, Materials Science Center, Golden, CO 80401, United States
- e. New Jersey Institute to Technology, Newark, NJ 07102, United States
- f. Core University Research Resources Laboratory, New Mexico State University, Las Cruces, NM 88003, United States
- g. School of Chemical Engineering, Zhengzhou University, Zhengzhou 450001, P.R. China



Figure S1. TGA analysis of all the samples under air atmosphere

To further confirm the composition of the as-prepared samples, we performed a TGA analysis under air atmosphere. The process involved combustion of carbon, oxidation-reduction reaction of metal sufides:

Metal sulfide + Carbon nanofibers + $O_2 \rightarrow$ Metal oxide + CO_2 + SO_2

Where the metal sulfide composites were oxidized to NiO and Co_3O_4 . Based on this formulation the weight percentages of Co_9S_8 , Ni_3S_2/Co_9S_8 , and Ni_3S_2 in the composites are calculated to be 50.7%, 41.5%, and 39.5%, respectively



Figure S2. (a) nitrogen N_2 adsorption-desorption isotherms (b) pore-size distribution of the electrodes



Figure S3. SEM image of $Ni_3S_2/Co_9S_8@S$ -CNFs anode after 200 cycles at a current density of 1 A g⁻¹



Figure S4. The relationship plot of Z' versus $\omega^{-1/2}$ at low-frequency region.

The Li diffusion kinetics was analyzed via EIS by using the following equation:

$$D = \frac{R^2 T^2}{2A^2 n^2 F^4 C^2 \sigma^2}$$
 Equation S1

Where R is the gas constant, T represents the temperature, A is the surface area of electrode, F is the Farady constant, *n* is the number of electrons per molecule attending the charge-discharge reaction, C is the concentration of lithium ion in the electrode, and σ is the slop of the line Z ' - $\omega^{-1/2}$. Based on the equation, the lithium ions diffusion coefficient is inversely proportional to σ (shown in Figure S4).