# **Supporting Information**

## Hierarchical Foam of Exposed Ultrathin Nickel Nanosheets Supported on Chainlike Ni-Nanowires

## and the Derivative Chalcogenide for Enhanced Pseudocapacitance

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**Figure S1.** The complementary TEM photos of Ni nanofoam with altered nucleation and growth rate. And some Ni lamellae (with visibly exposed sharp edge directly facing the viewer) are even thinner than 5 nm.



Figure S2. The evolution of XRD spectra of the Ni nanofoam under air annealing.



Figure S3 Energy dispersive X-ray spectrum (EDS) of the nanostructured NiS<sub>2</sub>.



**Figure S4.** TGA curves of the as-prepared  $NiS_2$  (300 °C sulfidation sample) in  $N_2$  and air atmospheres, respectively. Desulfurization process by thermal decomposition occurs under a nitrogen atmosphere, and an oxidative desulfurization process under air calcination.



Figure S5. The charge/discharge curves of NiS<sub>2</sub> at a current density of  $1 \text{ A g}^{-1}$ .



**Figure S6.** A sketch of the tailor-made steel autoclave for high-pressure sulfidation of e.g. the as-synthesized Ni-nanofoam. An enough amount of sublimed sulfur powder (i.e. 2-3 g) is deposited in the bottom of a 25-mL steel autoclave, the Ni-nanofoam with volume of 1-2 cm<sup>3</sup> contained in a narrow-mouth glass bottle is located on a porous steel scaffold in the autoclave, then a soft folded Al-foil is pressed and tightly sealed as a gasket. After transferred into a program-controlled muffle furnace, the high-pressure sulfur vapor forms [Ref 1,2] and the sulfidation of metals happens. One of the advantages is that it offers smaller sulfur molecules [Ref 3] and higher rate of diffusion and thus a more effective reaction process, yet with no sulfur residue on the product surface.

[1] B. Meyer. 'Elemental Sulfur,' Chem. Rev., 1976, 76, 367-388.

[2] W. A. West, A. W. C. Menzies. 'The Vapor Pressures of Sulphur between 100° and 550° with related Thermal Data,' *J. Phys. Chem.*, **1928**, *33*, 1880-1892.

[3] S. Xin, L. Gu, N.-H. Zhao, Y.-X. Yin, L.-J. Zhou, Y.-G. Guo, L.-J. Wan. 'Smaller Sulfur Molecules Promise Better Lithium–Sulfur Batteries,' *J. Am. Chem. Soc.*, **2012**, *134*, 18510-18513.

#### S1. Crystallographic Structures

Ni: face-centered cubic (fcc) structure (Nickel, syn, JCPDS card no. 04-0850, Fm-3m (225),  $a_0=b_0=c_0=3.524$  Å). NiO: cubic structure (Bunsenite, syn, JCPDS card no. 47-1049, Fm-3m (225),  $a_0=b_0=c_0=4.177$  Å). NiS<sub>2</sub>: cubic structure (Vaesite, JCPDS card no. 11-0099, Pa-3 (205),  $a_0=b_0=c_0=5.670$  Å) Data based on *ICDD/JCPDS PDF Retrievals [Level-1 PDF, Sets 1-51 (04/25/07)]* 

## S2. Reference on 'Vaesite $(NiS_2)$ '

Kerr, P. F. (1945) Cattierite and vaesite: new Co–Ni minerals from the Belgian Congo. *Amer. Mineral.*, *30*, 483-497. (http://www.minsocam.org/ammin/AM30/AM30\_483.pdf)

Vaesite NiS<sub>2</sub>, © 2001-2005 Mineral Data Publishing, version 1 (<u>http://rruff.info/doclib/hom/vaesite.pdf</u>) Vaesite Mineral Data (<u>http://webmineral.com/data/Vaesite.shtml</u>) Vaesite mineral information and data (<u>http://www.mindat.org/min-4133.html</u>)

Last accessed on 03/12/1013

## **S3. Extended Reading Interests**

Metal Aerogels (<u>http://www.aerogel.org/?p=932</u>) Metal Oxide Aerogels (<u>http://www.aerogel.org/?p=44</u>) Semiconducting Metal Chalcogenide Aerogels (<u>http://www.aerogel.org/?p=560</u>) *Last accessed on 05/12/1013*