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# ACCEPTED MANUSCRIPT

### NiCo<sub>2</sub>S<sub>4</sub>/Co<sub>3</sub>S<sub>4</sub> Heterogeneous Double-Shelled nanocages for

#### high-performance electrochemical energy storage

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<sup>*a*</sup>College of Machinery and Automation, Zhejiang Sci-Tech University, Hangzhou 310018, China <sup>*b*</sup>Key Laboratory of Advanced Textile Materials and Manufacturing Technology of the Ministry of Education, College of Materials and Textiles, Zhejiang Sci-Tech University, Hangzhou 310018, China **Abstract:** zeolitic imidazolate framework-67 (ZIF-67)-derived yolk-shelled ZIF-67/Ni–Co layered double hydroxides were further vulcanized, forming heterogeneous double-shelled cage-in-cage NiCo<sub>2</sub>S<sub>4</sub>/Co<sub>3</sub>S<sub>4</sub>. As-synthesized double-shelled nanocages were characterized by XRD, SEM and TEM. As active materials for water-based electrochemical energy storage devices, NiCo<sub>2</sub>S<sub>4</sub>/Co<sub>3</sub>S<sub>4</sub> showed relatively high discharge capacity and cycling stability, large current adaptability and reversibility. These could be due to the cage-in-cage hollow nanostructure and high electrochemical activity of transition metal sulfide.

Keywords: NiCo<sub>2</sub>S<sub>4</sub>/Co<sub>3</sub>S<sub>4</sub>; Nanocages; Microstructure; Energy storage and conversion

#### 1. Introduction

Electrochemical Energy Storage Devices (EESDs) with higher specific capacity, faster charge-discharge rate and longer cycle life have attracted tremendous research interests for various applications. They can substitute for the fossil fuel energy as a rechargeable and environmentally friendly power sources including secondary batteries and electrochemical capacitors [1-3]. Recently, transition metal sulfides have been investigated as potential active materials for EESDs because they show higher electrical conductivity and richer redox

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