



A metal nitride interlayer for long life lithium sulfur batteries

Jin-Lei Qin^{a,b}, Huiyou Zhao^a, Jia-Qi Huang^{b,c,*}

^a Department of Materials Science and Engineering, China University of Mining and Technology (Beijing), Beijing 100083, China

^b Advanced Research Institute of Multidisciplinary Science, Beijing Institute of Technology, Beijing 100081, China

^c CAS Key Laboratory of Carbon Materials, Institute of Coal Chemistry, Chinese Academy of Sciences, Taiyuan 030001, Shanxi, China

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With the growing concerns on global energy crisis and the greenhouse effect, the exploration on renewable energy and related emerging energy conversion and storage technologies are highly interested [1]. Lithium sulfur (Li-S) battery system receives great attention for its high theoretical specific energy (2600 Wh/kg), which is deemed to be a promising candidate as next generation high energy density batteries [2]. Yet, the practical application of the Li-S batteries is limited by the notorious shuttle effect of lithium polysulfides (LiPSs) between cathode and anode [3,4]. Among diverse strategies that devoted to suppress the shuttle effect, a functional interlayer between the cathode and separator has been proposed to sterically obstruct and chemically bind LiPSs, kinetically favoring LiPSs interconversion [5–7]. For instance, He et al. employed an ultrathin interlayer composed of $\text{Li}_4\text{Ti}_5\text{O}_{12}$ and carbon nanofiber to effectively retard shuttle of polysulfides [8]. Simultaneously, the functional interlayer formed by the reduced graphene oxide and activated carbon is proposed by Zhang and coworkers can capture and reuse polysulfide species in Li-S batteries [9].

Recently, Wang and co-workers from Hunan University for the first time demonstrated the metal nitrides derived from lay-

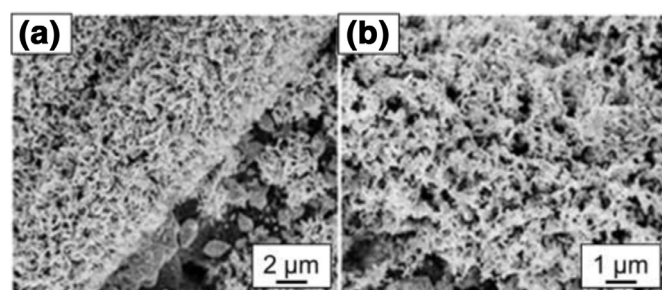


Fig. 1. SEM images of nanoparticle-stacked Co_2N and Ni_3N prepared by NH_3 annealing of LDHs on carbon paper [10].

ered double hydroxides (LDHs) as a self-supporting interlayer inserted independently between sulfur cathode and PP separator, endowing the cell with long cycling life, excellent cycle stability, and high Coulombic efficiency retention in working Li-S batteries [10]. Nanoparticle-stacked nitride material (Co_2N and Ni_3N) was prepared by annealing LDHs on carbon paper (CP) under NH_3 atmosphere at 400 °C (named as N-LDH-400/CP). The formed N-LDH-400/CP functional interlayer possesses the following attractive attributes: (1) large specific surface area to effectively absorb polysulfides; (2) hydrophilic metal-O groups to anchor polysulfides on the polar surface; (3) good electric conductivity to re-utilize the polysulfides anchored on the interlayer; (4) the structure to retain electrolyte for excellent electrochemical performance.

* Corresponding author at: Advanced Research Institute of Multidisciplinary Science, Beijing Institute of Technology, Beijing 100081, China.

E-mail address: [jqhuang@bit.edu.cn](mailto:jquang@bit.edu.cn) (J.-Q. Huang).

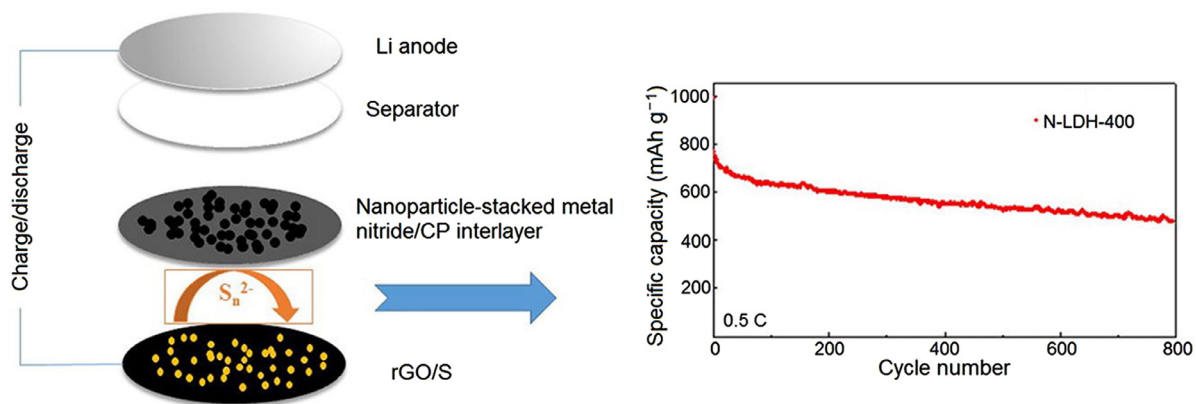


Fig. 2. (a) Schematic diagram of the Li-S battery with N-LDH-400/CP interlayer; (b) Long cycle performance of a Li-S battery with N-LDH-400/CP interlayer [10].

Specifically, the surface of transition metal nitrides on CP formed an oxide passivation layer with exposing abundant active sites and adsorption sites, which exhibited excellent chemical stability and hindered the diffusion of polysulfides by chemical and physical adsorption. Additionally, Co_2N and Ni_3N nanoparticles retain large enough inter-particle space to store electrolyte (Fig. 1). Accordingly, the N-LDH-400/CP interlayer between the rGO-S cathode and PP (Celgard 2400) (Fig. 2a) exhibited stable cycle performance. As demonstrated in Fig. 2(b), the discharge capacity was 764.6 mAh g^{-1} at the first cycle and maintained 62.4% after 800 cycles.

The metal nitrides derived from LDHs with above advantages significantly improved both capacity and cycling stability of Li-S cells, demonstrating the importance of the rational design of cell configuration and interfacial properties. Furthermore, the employed $\text{Co}_2\text{N}/\text{Ni}_3\text{N}/\text{CP}$ interlayer derived from LDH is highly inspiring, which may arouse great interest in exploring emerging functional interlayers by accurately regulating active sites for advanced energy storage devices.

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