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Al_2O_3-doped ZnO coating of carbon nanotubes as cathode material for lithium-sulfur batteries

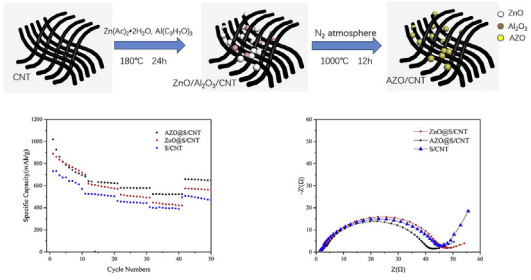
Tianye Ma[a], Mengmeng Liu[a], Tao Huang[a,**], Aishui Yu[b,*]

[a] Laboratory of Advanced Materials, Fudan University, Shanghai, 200438, China
[b] Department of Chemistry, Shanghai Key Laboratory of Molecular Catalysis and Innovative Materials, Institute of New Energy, Collaborative Innovation Center of Chemistry for Energy Materials, Fudan University, Shanghai, 200438, China

HIGHLIGHTS

- Al_2O_3-doped ZnO enhances the conductivity of cathode materials.
- A high-temperature solid-state reaction maintains the crystal lattice of ZnO.
- Carbon nanotubes provide steady structure.
- Al_2O_3-doped ZnO composites lead to high battery capacity.

GRAPHICAL ABSTRACT



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ABSTRACT

Currently, lithium-sulfur (Li-S) batteries become increasingly attractive because of their high theoretical energy density and high theoretical specific capacity. In this work, high-sulfur-loading Al_2O_3 doping of a ZnO-modified carbon nanotube (AZO@S/CNT) composite is synthesized via the hydrothermal method and subsequent high-temperature solid-state reaction. Al_2O_3-doped ZnO (AZO) induces a strong chemical interaction between polysulfides to restrain the "shuttle effect." In addition, we use a novel method to enhance the electronic conductivity of the cathode material. The Al_2O_3 doping of ZnO can increase the electron density in ZnO, and thus enhance its electronic conductivity. The initial discharge capacity of the cathode with 60-wt.% S reaches 1100 mAh g^{-1} and the recycle capacity still remains 700 mAh g^{-1} after 200 cycles at 0.2 C with a capacity fade rate of 0.18% per cycle, offering a potential candidate for practical application of high-energy-density Li-S batteries in the future.

1. Introduction

With the increasing demands of electronic equipment, batteries with high energy density and long-term durability are required [1,2]. Lithium-sulfur (Li-S) batteries have become increasingly attractive because of their high theoretical energy density of 2600 W h kg^{-1} and high theoretical specific capacity of 1675 mAh g^{-1}, confirming Li-S batteries as one of the energy storage systems with the most potential in the future [1,3,4]. Moreover, S has many advantages, such as having abundant reserves on the Earth, nontoxicity, and environmental friendliness, which make it an excellent material for battery cathodes [5]. However, Li-S batteries suffer from a few problems that significantly hinder its widespread practical application. First, the low electrical conductivity of S and discharge products result in the rapid

* Corresponding author. Department of Chemistry, Shanghai Key Laboratory of Molecular Catalysis and Innovative Materials, Institute of New Energy, Collaborative Innovation Center of Chemistry for Energy Materials, Fudan University, Shanghai, 200438, China.
** Corresponding author. Laboratory of Advanced Materials, Fudan University, Shanghai, 200438, China.
E-mail address: asyu@fudan.edu.cn (A. Yu).

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Fig. 1. AZO/CNT synthesis procedure.

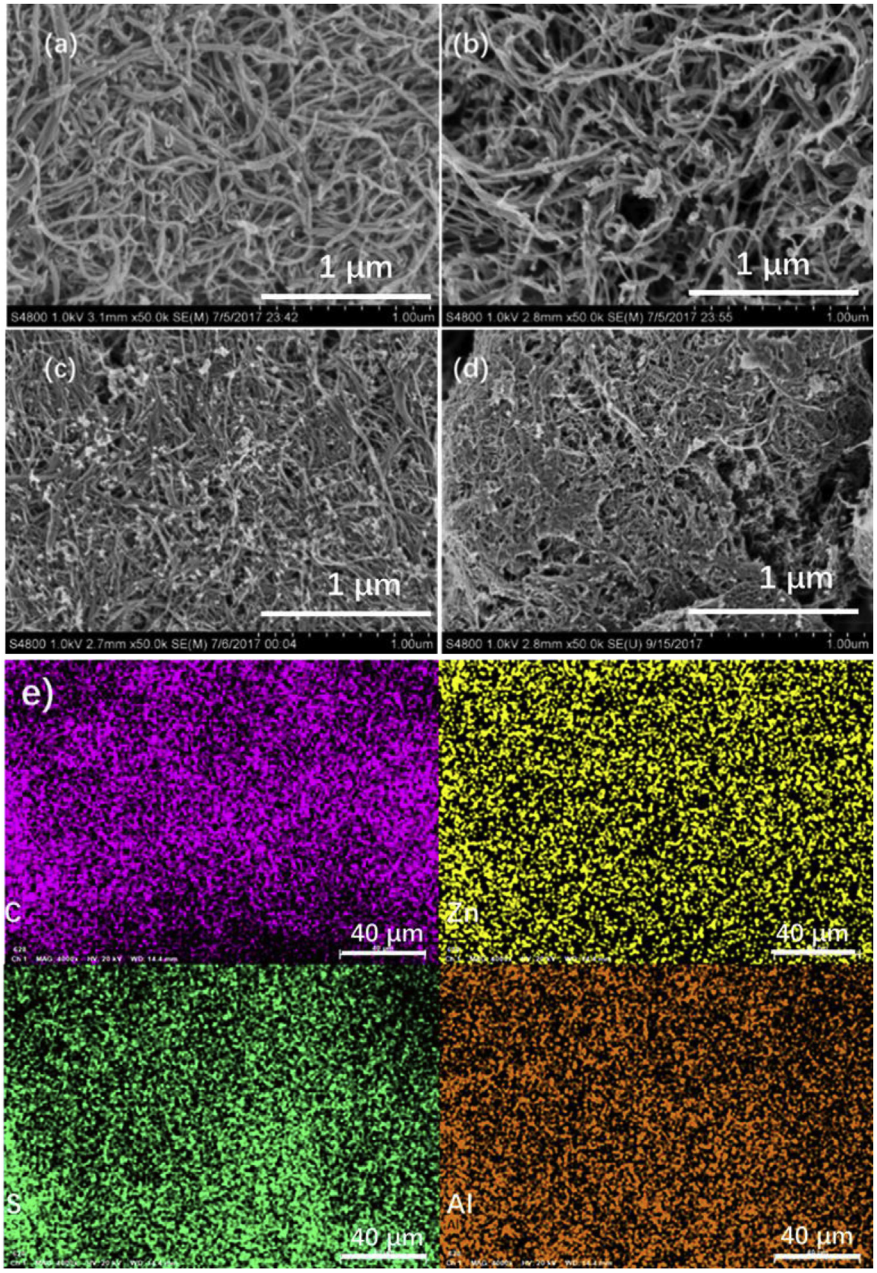

Fig. 2. SEM images of (a) CNT, (b) ZnO/CNT, (c) AZO/CNT, and (d) AZO@S/CNT; (e) SEM mapping pictures of C, Zn, S, and Al in AZO@S/CNT.

degradation of rate capacity [6]. Second, the intermediate polysulfides dissolve easily in the electrolyte, which further results in low Coulombic efficiency during long-term cycling [7]. In addition, the volume expansion during the charge/discharge process destroys the electrode structure of Li-S batteries.

Many strategies have been used over the years to solve the above-mentioned problems. It is well known, as an ideal host, carbon materials can provide steady conductive structure in volume changes during the charge/discharge process. Many carbon hosts with novel, elaborate structures have been designed and utilized, such as porous carbons [6,8–10], (reduced) graphene oxide [11], and carbon nanotubes (CNTs) [12,13]. Compared with porous C, CNTs have long-range conductivity and graphene has the disadvantage of low conductivity while in an upright position. CNTs tend to be the ideal matrix because of their low weight, high conductivity, and long-range conductivity. Major improvements were achieved by employing advanced CNTs. However, because C is an intrinsic non-polarity material, and polysulfides are a polarity material, the connection between C and polysulfides on the surface is very weak because it is physical absorption [7], with which it is difficult to efficiently restrain the “shuttle effect” of lithium