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Application of diatomite as an effective polysulfides adsorbent for lithium-sulfur batteries

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ARTICLE INFO

Article history:

Received 24 July 2017

Revised 7 September 2017

Accepted 12 September 2017

Available online xxx

Keywords:

Lithium-sulfur batteries

Diatomite

Adsorbent

Cycle performance

ABSTRACT

The lithium-sulfur batteries show the great potential to be the most promising candidate for high energy applications. However, the shuttling of soluble polysulfides deteriorates the battery performance tremendously. To suppress the diffusion of soluble polysulfides, diatomite that has abundant natural three-dimensional ordered pores is incorporated into the cathode to trap polysulfides. The composite cathode material (S-DM-AB for short), including sulfur (S), diatomite (DM), and acetylene black (AB) is prepared by an impregnation method. For comparison, another composite cathode material (S-AB for short) including sulfur and acetylene black is also prepared by the same method. The battery with S-DM-AB composite cathode material delivers a discharge capacity of 531.4 mAh/g after 300 cycles at 2 C with a capacity retention of 51.6% at room temperature. By contrast, the battery with S-AB composite cathode material delivered a capacity of only 196.9 mAh/g with a much lower capacity retention of 18.6% under the same condition. The addition of diatomite in the cathode is proved to be a cheap and effective way to improve the life time of the lithium sulfur batteries.

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1. Introduction

Lithium-sulfur batteries are highly promising power devices due to the high theoretical specific capacity (1672 mAh/g) of sulfur and energy density (2500 Wh/kg) of the lithium-sulfur battery, which far exceed the currently used commercial lithium-ion batteries [1,2]. What is more, sulfur is a low cost, environmentally friendly and naturally abundant element distributed all over the world. However, lithium-sulfur batteries still suffer from several shortcomings to hinder the commercialization, such as the poor conductivity of sulfur ($\sim 10^{-30}$ S/cm), large volume change ($\sim 80\%$) between the charge and discharge products, and particularly the shuttle effect arising from the diffusion of the soluble long-chain polysulfides (Li_2S_n , $4 \leq n \leq 8$) [3,4]. The soluble long-chain polysulfides in the ether based liquid electrolyte generating from the first step of the discharge process, would migrate between the lithium anode and sulfur electrode, leading to the low utilization of active material, poor coulombic efficiency and rapid capacity degradation [1-5].

Recently, various strategies have been performed to suppress polysulfides shuttling in electrolyte, including: (1) incorporating or encapsulating sulfur within carbon materials [6-15], metal com-

plexes [16-30] and conductive polymers [31-34]; (2) coating the separator or cathode with a functional film [35-41]; (3) inserting an interlayer between the separator and sulfur electrode [42-44]; (4) application of single ion conductor via electric static repulsion [45,46]; (5) exploration of functional binders [47-49]. In addition, the introduction of additive is also an effective method to restrain polysulfides from diffusion [50]. Some nano-sized metal oxides, such as $\text{Mg}_{0.8}\text{Cu}_{0.2}\text{O}$ [51], $\text{Mg}_{0.6}\text{Ni}_{0.4}\text{O}$ [52,53], Al_2O_3 [54,55], TiO_2 [56], La_2O_3 [57] and V_2O_5 [58,59], were added to the sulfur cathode to adsorb polysulfides, and they showed positive effect to increase the cycling stability of lithium-sulfur batteries by suppressing the diffusion of soluble polysulfides. However, the synthesis of these nano-sized metal oxide additives requires high cost and complex preparation process.

Diatomite is a kind of biological sedimentary minerals with abundant natural pores. It is mainly comprised of SiO_2 with small concentrations of impurities including Fe_2O_3 , CaO , MgO and organics [60,61]. Owing to the ordered porous structure, large surface area, good chemical stability, excellent sorption ability and cheap price, diatomite has been widely used to adsorb heavy metals and organic chemicals [62,63]. Herein, diatomite is incorporated into the cathode to adsorb polysulfides to improve the life time of lithium sulfur batteries. A blank sulfur electrode without diatomite is also prepared for battery test. The electrochemical results confirmed that both the capacity retention and coulombic efficiency of the lithium sulfur battery are improved with the addition of diatomite to the sulfur/acetylene black composite cathode.

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49 2. Experimental

50 2.1. Preparation of sulfur cathode materials

51 The sulfur (S)-diatomite (DM)-acetylene black (AB) composite
52 was prepared by an impregnation method. In detail, stoichiomet-
53 ric amounts of acetylene black and diatomite were mixed together
54 at first, and then a solution of sublimed sulfur in an appropriate
55 amount of carbon disulfide was later added followed by ultrasonic
56 dispersion. Carbon disulfide was slowly evaporated at room tem-
57 perature under stirring followed by vacuum dry at 60 °C for 24 h to
58 get the S/DM/AB mass ratio of 6:3:1. The S-AB composite with the
59 S/AB mass ratio of 6:4 was also synthesized by the same method.

60 2.2. Quantitative determination of the adsorption capacity of 61 diatomite and acetylene black for polysulfides

62 The Li_2S_6 solution with the concentration of 5 mmol/L was pre-
63 pared referring to literature [64]. The sulfur and Li_2S with a molar
64 ratio of 5:1 were added into the 1,2-dimethoxyethane (DME) solu-
65 tion in an argon-filled glove box firstly. After stirring vigorously for
66 24 h, the dark yellow Li_2S_6 solution was obtained. In order to de-
67 termine the exact adsorption capacity of diatomite and acetylene
68 black for polysulfides, 50 mg of diatomite and 50 mg of acetylene
69 black were separately added into two bottles filling with 5 mL of
70 the Li_2S_6 solution in each in the argon protected glove box. The

suspensions were treated with magnetic stirring for 6 h followed
71 by standing for 24 h to deposit the adsorbents completely. The
72 upper solution was sucked out and transferred into quartz tube
73 for UV-Vis analysis. The concentration of the Li_2S_6 solution af-
74 ter adsorption is calculated using the equation according to Beer-
75 Lambert law,
76

$$\frac{A_1}{A_2} = \frac{c_1}{c_2} \quad (1)$$

where A_1 is the absorbance of the pristine Li_2S_6 solution, c_1 is
77 the concentration of the pristine Li_2S_6 solution (5 mmol/L), A_2 is
78 the absorbance of the Li_2S_6 solution after adsorption, and c_2 is the
79 concentration of the Li_2S_6 solution after adsorption. Next, the ad-
80 sorption capacity can be calculated using the following equation,
81
82

$$Q_{\text{ads}} = \frac{(c_1 - c_2)VM}{m} \quad (2)$$

where V is 5 mL, M is the molecular weight of Li_2S_6 , m is the mass
83 of adsorbent, and Q_{ads} is the adsorption capacity (g/g).
84

85 2.3. Material characterization

86 The specific surface area and the pore structure of the di-
87 atomite were measured by a mercury intrusion porosimeter (Au-
88 toPore IV9500, Micromeritics). The polysulfide solutions before
89 and after adsorption experiments were quantitatively analyzed by

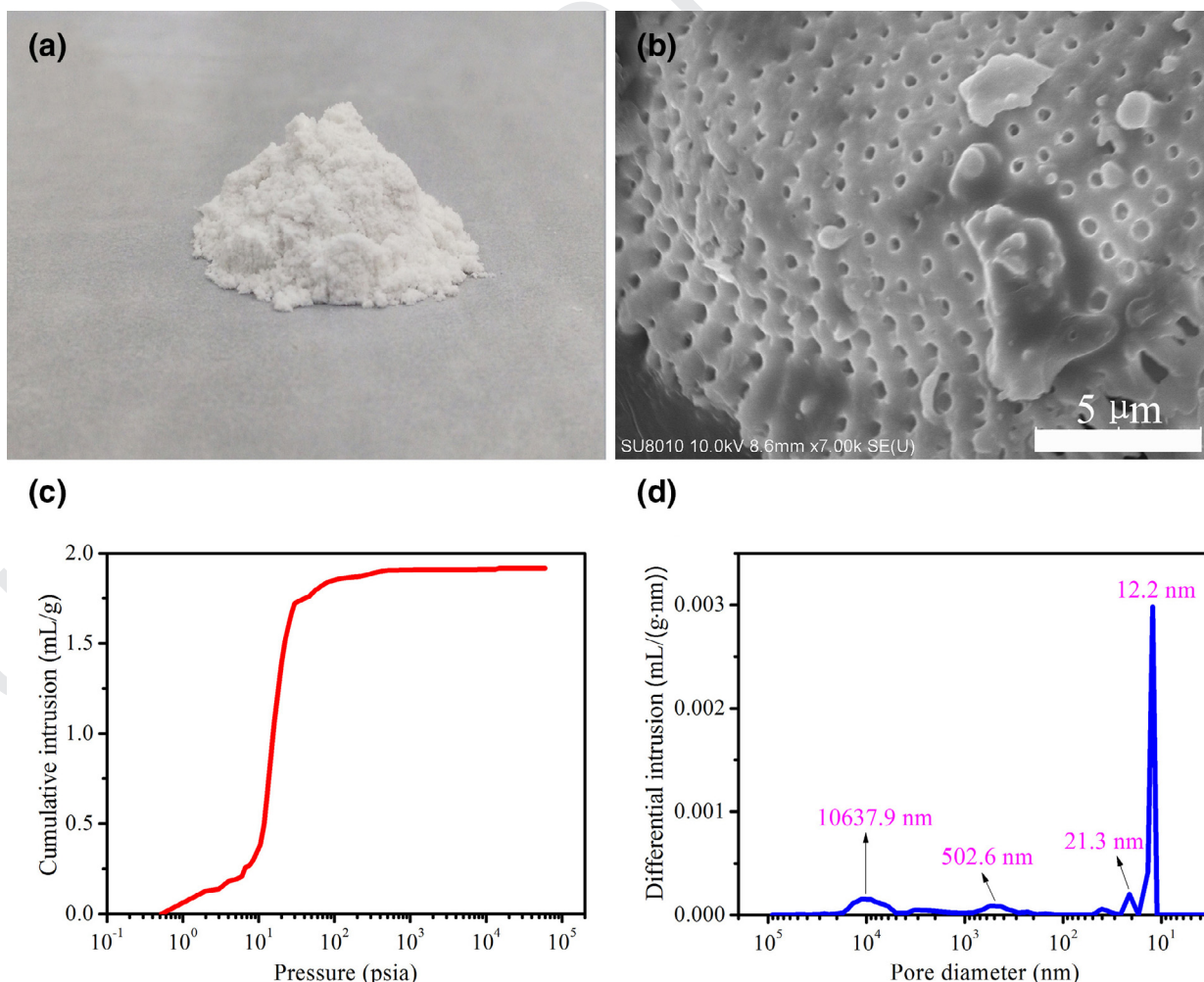


Fig. 1. (a) Optical image of diatomite powder, (b) FE-SEM image of diatomite particle, (c) the cumulative mercury intrusion of diatomite powder, (d) pore size distribution of diatomite powder.

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