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Effect of commercial activated carbons in sulfur cathodes on the electrochemical properties of lithium/sulfur batteries

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ABSTRACT

We prepared sulfur/active carbon composites via a simple solution-based process using the following commercial activated carbon-based materials: coal, coconut shells, and sawdust. Although elemental sulfur was not detected in any of the sulfur/activated carbon composites based on Thermogravimetric analysis, X-ray diffraction, and Raman spectroscopy, Energy-dispersive X-ray spectroscopy results confirmed its presence in the activated carbon. These results indicate that sulfur was successfully impregnated in the activated carbon and that all of the activated carbons acted as sulfur reservoirs. The sulfur/activated carbon composite cathode using coal exhibited the highest discharge capacity and best rate capability. The first discharge capacity at 1 C (1.672 A g^{-1}) was 1240 mAh g^{-1}, and a large reversible capacity of 567 mAh g^{-1} was observed at 10 C (16.72 A g^{-1}).

1. Introduction

Rechargeable Li/S batteries are regarded as one of the most attractive candidates for high-energy power sources because elemental sulfur is an inexpensive and abundant material with a high theoretical specific capacity of 1672 mAh g^{-1}. Li/S batteries exhibit a high theoretical energy density of 2600 Wh kg^{-1} [1–3]. This value is much greater than conventional lithium-ion battery. [4–6] However, Li/S batteries have not yet been commercialized because of the following problems: (1) poor cycle life [7–9] and high self-discharge [10–12] due to the dissolution of the lithium polysulfides in the electrolyte and (2) low utilization [7,10] and limited rate capability [13–16] of the sulfur cathodes due to the insulating nature of sulfur and lithium sulfide.

To overcome these problems, many researchers have studied novel cell components such as sulfur composites [14,17–20], trapping interlayers [16,21,22], Li/lithium polysulfide cells [23–25], and efficient electrolytes [26–28]. In particular, sulfur/mesoporous carbon composites can be prepared to infiltration of elemental sulfur into the nanopores of a conductive mesoporous carbon matrix. The sulfur/mesoporous carbon composite thus enhances both the electrical and ionic conductivity of the sulfur cathode while retarding the polysulfide shuttle phenomenon and accommodating volume variation of the cathode during cycling [17]. However, the preparation of the mesoporous carbon matrix requires a series of complex processes and the using of expensive materials [16,17,29]. Therefore, some researchers have prepared nanopore of carbon by activation process using inexpensive materials such as fruit seeds [15,30–33] and bugs [34]. However, most of nanoporous carbons were prepared in a Lab. It is necessary to further study for commercial needs. If the commercial carbon can be used, the commercialization of lithium/sulfur battery will be easier.

Activated carbon with nanopore is already commercialized and used a various fields. Activated carbon is a popular adsorbent for many applications, such as air and water cleaning and catalyst preparation for chemical and petrochemical industries, because of its high specific surface area, porosity, chemical inertness, and thermal stability. To prepare activated carbon, the most commonly used raw materials are coal (anthracite, bituminous, and lignite), coconut shells, wood (both soft and hard), peat, and petroleum-based residues. Also, global activated carbon market size is gradually increased recently.

However, there was no study on the infiltration of sulfur in commercial activated carbon using coal, coconut shells, and sawdust. In this study, we are investigating the possibility of

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commercial activated carbon for sulfur reservoir which can be used for lithium/sulfur battery. Also, various commercial activated carbons were used to fabricate sulfur/carbon composites as the cathode material for lithium/sulfur batteries, and their performance were evaluated.

2. Experimental

2.1. Synthesis of the sulfur/activated carbon composites

Activated carbon based on coal, coconut shells, and sawdust was purchased from the Dongyang Carbon Co. The sulfur/activated carbon composites were prepared with the various types of activated carbon. The activated carbons were heated in an argon-filled furnace at 1000 °C for 1 h, and the sulfur/activated carbon composites were prepared using a solution-based process [18]. For preparation of the sulfur/activated carbon composites, the sulfur powder was dissolved in a dimethyl sulfoxide (DMSO, Daejung Co.) solvent heated by a mantle to 90 °C. Next, the activated carbon powder was added into the solution and dispersed by stirring for 3 h. The color of the solution changed from yellow to black. The coal, coconut shells, and sawdust activated carbon materials were denoted SCoal, SCoconut, and SSawdust, respectively. The solution was cooled to room temperature and became transparent, which indicated recrystallization of sulfur in the activated carbon. The sulfur/activated carbon composite powder was washed with ethanol several times to remove the residual DMSO. The composite was then dried in vacuum at room temperature and finally heated in an oven at 100 °C for 24 h.

2.2. Electrochemical measurement

The cathode was prepared by mixing 60 wt.% of the sulfur/activated carbon composite, 20 wt.% of Super-P (TIMCAL), and 20 wt.% of poly(vinylidene fluoride) (Mw 23,500, Alfa Aesar) dissolved in *N*-methyl-2-pyrrolidone (NMP, Aldrich) to form a slurry, which was then pasted on Al foil substrate. The cathode electrode was dried in oven at 60 °C for 24 h. Li metal was used as a counter electrode, and a microporous polyethylene membrane (Celgard 2400) was used as the separator. The electrolyte was 0.5 M bis(trifluoromethane)sulfinimide lithium (Aldrich) dissolved in 1,2-dimethoxyethane and 1,3-dioxolane (4:1 by volume ratio).

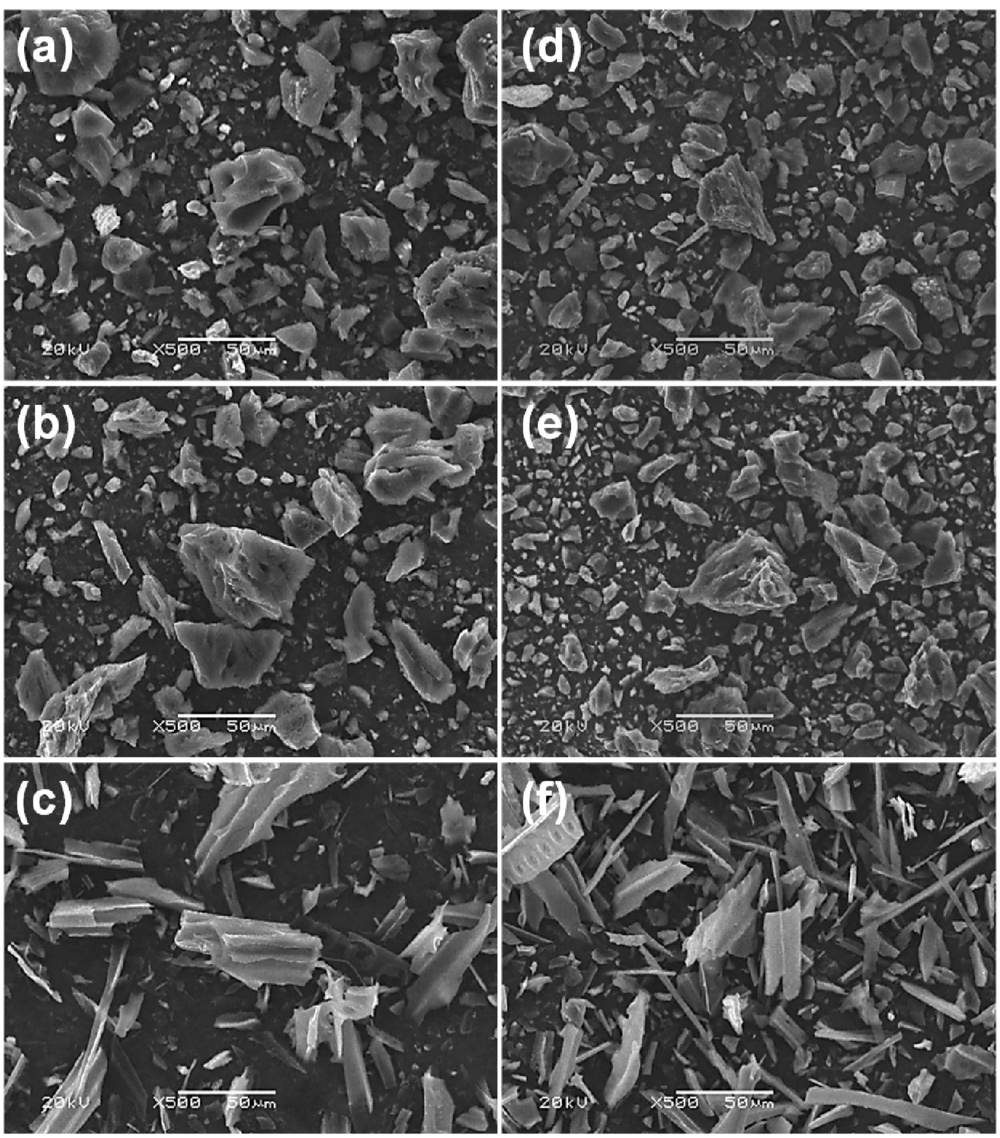

Fig. 1. SEM images of the sulfur/activated carbon composite powders prepared using coal, coconut shells, and sawdust: (a) SCoal composite, (b) SCoconut composite, and (c) SSawdust composite.