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Effect of solvents on the electrochemical properties of binder-free sulfur cathode films in lithium–sulfur batteries

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The effects of solvents on the preparation of sulfur cathodes were investigated by fabricating binder-free sulfur electrode films using three different solvents: 1-methyl-2-pyrrolidinone (NMP), acetonitrile, and deionized water. These solvents are commonly employed to dissolve binders used to prepare sulfur cathodes for lithium–sulfur batteries. The sulfur electrode fabricated with NMP had a higher discharge capacity and longer cycle life than the ones fabricated with acetonitrile and deionized water. Better adhesion between the current collector and the sulfur electrode accounted for the improved capacity and cycle life of the battery. In addition, the stability of the electrode in the electrolyte was a result of the solubility of sulfur in the solvent. We thus concluded that the solvents used in the fabrication of sulfur electrodes had a positive influence on the electrochemical properties of Li–S batteries.

1. Introduction

Sulfur is an attractive cathode material for rechargeable lithium batteries. A lithium–sulfur (Li–S) battery has many advantages, such as a theoretical specific energy density up to 2600 Wh kg^{-1} [1–3]. However, sulfur is an electrical and ionic insulator. These properties result in some serious drawbacks such as low sulfur utilization and poor cycle property. Polysulfides have a high solubility in lithium battery electrolytes, which can cause fast capacity fading and a short cyclic life. To overcome these problems, researchers have focused on enhancing the electrical conductivity of the sulfur cathode to reduce the loss of soluble intermediates during cycling.

Sulfur electrodes consist of sulfur as the active material, a conducting material for the improvement of electrical conductivity, and a binder to enhance the adhesion between the different component layers. This improved adhesion between sulfur and its neighboring materials should improve the overall electrochemical performance in a Li–S cell. In previous studies, sulfur electrodes have been fabricated with various binders such as poly(vinylidene fluoride) (PVDF) [4–10], polyethylene oxide (PEO) [11–19], carboxymethyl cellulose–styrene-butadiene rubber (CMC–SBR) [20–22], polytetrafluoroethylene (PTFE) [23–29], gelatin [19,30], and β-cyclodextrin [31]. In addition, various solvents to dissolve these binders have been employed, for example, 1-methyl-2-pyrrolidinone (NMP) for dissolution of PVDF [4–10], acetonitrile (ACN) for PEO [11–19], and water for CMC–SBR [20–22], PTFE [23–29], gelatin [19,30], and β-cyclodextrin [31]. These binders occupied 10–30 wt% of the sulfur electrode with large quantities of solvent required for binder dissolution and mixing of materials. In the case of lithium ion battery, the solvent can not be a critical role because oxide cathode is very stable in solvent. However, sulfur is very reactive and can be dissolved in many solvents. Thus some of solvent can dissolve sulfur as well as binder. This phenomenon should become critical parameter for the no-binder sulfur electrode and high sulfur content with small amount of binder. For flexible or high energy density sulfur electrode, many researchers investigated binder-free electrodes [32–40]. For no-binder electrode, sulfur should attach on the current collector without binder and the effect of solvent is important. In addition, electrode with high sulfur content should have small inactive component such as binder or conducting agent and it is necessary to study the electrode with little inactive content. However, to the best of our knowledge, there has not been a study on how these solvents affect the sulfur electrodes themselves.

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In this study, we investigated how the use of different solvents affects the electrochemical performance of sulfur cathode films in lithium–sulfur batteries. To evaluate the impact of various solvents, we fabricated sulfur electrode films without a binder using NMP, ACN, and water; commonly used solvents for binders. Also, we evaluated electrochemical properties of the binder-free sulfur electrode for Li/S battery by directly paste the sulfur powder onto current collector without using binder. We labeled the binder-free sulfur electrodes prepared using NMP, ACN, and water as SNMP, SACN, and Swater, respectively. The electrochemical properties and mechanical stability of these electrodes were evaluated with particular attention to the role of the solvents used. The objective of this study is to determine whether the choice of binder solvent used to fabricate sulfur electrodes can positively enhance the performance of Li–S batteries.

2. Experimental

2.1. Preparation of binder-free electrodes

Sulfur (Aldrich Co.), Super-P® conductive carbon black (Aldrich Co.), and a solvent were mixed using a planetary ball mill at 300 rpm for 3 h. The weight ratio of sulfur to Super-P® was 3:1. The three solvents used were water, ACN (CH_3CN, Junsei Chemical Co.),

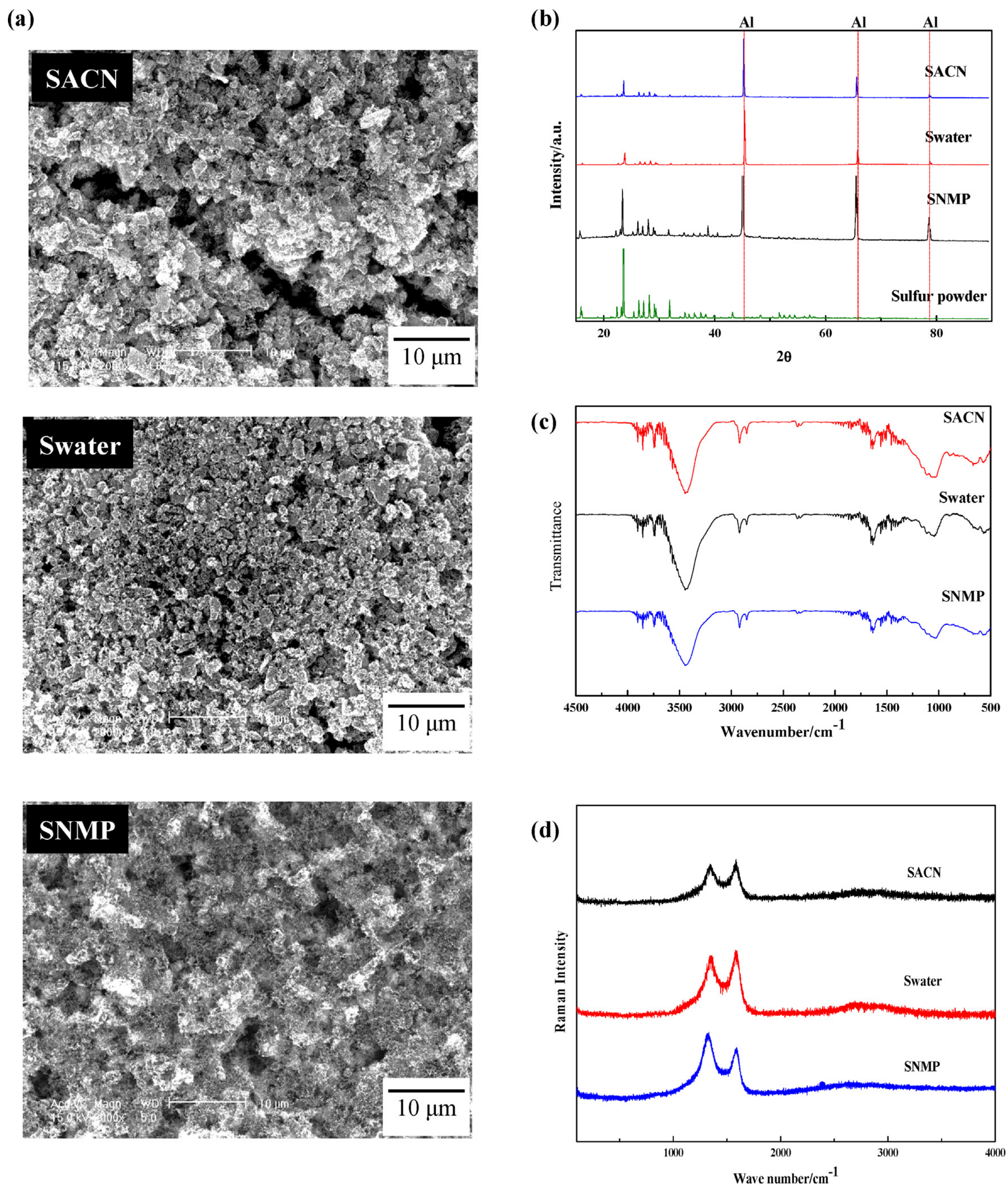

Fig. 1. Characteristics of the binder-free sulfur electrodes prepared with acetonitrile (ACN), water, and 1-methyl-2-pyrrolidinone (NMP): (a) SEM images, (b) XRD patterns, (c) FTIR spectra, and (d) Raman spectra.