

Cyclability of sulfur/dehydrogenated polyacrylonitrile composite cathode in lithium-sulfur batteries

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Introduction

Dehydrogenated polyacrylonitrile (S/DPAN composite) is a promising cathode for lithium-sulfur (Li-S) batteries. This cathode exhibits higher specific discharge capacity and better cyclability in comparison with sulfur/carbon system [1-3]. Nonetheless, capacity fading still can be observed. Understanding the reasons for capacity fading is essential for designing a better cathode composite. In this study, characterization techniques such as scanning electron microscopy (SEM), energy dispersive X-ray spectroscopy (EDX), elemental analysis (CHNS) and electrochemical impedance spectroscopy (EIS) were used to investigate the change of cathode properties with cycle number.

Experimental

Sulfur and polyacrylonitrile (PAN) were mixed in a ball mill. The resulting mixture was dried in a vacuum oven at 50°C for 3 h and then heat treated at 300°C for 3 h in argon.

The crystalline phase of the samples was studied using X-ray diffraction (XRD) technique equipped with Cu-K α radiation. The composite surface morphology was examined using a field emission scanning electron microscopy (FE-SEM) equipped with energy dispersive X-ray spectroscopy (EDX). Sulfur content of the samples was analyzed using an elemental analyzer (CHNS).

The electrochemical performance of the S/DPAN composite cathode was investigated using coin-type cells (CR2025). Each cell was composed of a lithium metal negative electrode and a S/DPAN composite positive electrode, separated by a microporous polypropylene film. 1 M LiPF₆ in a mixed solvent of ethylene carbonate (EC), dimethyl carbonate (DMC), and diethylene carbonate (DEC) with a 1:1:1 weight ratio was used as the liquid electrolyte. The prepared cathode comprised 80 wt% S/DPAN with 10 wt% ketjenblack and polyvinylidene fluoride (PVdF) binder. The average cathode material loading was 10-12 mg per electrode (12.7-15.3 mg cm⁻²). The cells were tested galvanostatically on a multi-channel battery tester between 1 and 3 V at 0.2 C charge-discharge rate (current density = 334.4 mA g⁻¹). At the end of the 10th, 20th and 50th charge stage, the cells were disassembled. The cathodes were washed by dimethyl carbonate (DMC) and ethanol, and then vacuum dried at 50°C for 3 h. Finally, a part of the composite particles was removed from the nickel foam by tweezers and used for SEM observation and elemental analysis.

Measurements of electrochemical impedance spectroscopy (EIS) and cyclic voltammetry (CV) were carried out using a multi-channel potentiostat. The amplitude of the AC signal was 10 mV in the frequency range from 1 MHz to 0.1 Hz. CV was performed between 1 and 3 V vs. Li⁺/Li^o at a scan rate of 0.1 mV s⁻¹. All electrochemical measurements were conducted at ~ 25°C.

Results and Discussion

Using the cathode, which have material loading three times that of the reported [1,3], the electrochemical

performance was carried out. Higher sulfur loading means that the cyclability fading is more obvious. **Figure 1** shows the charge-discharge profiles of the sample. The first discharge curve consists of two plateaus, at 2.31 V and 1.3 V. However, the second discharge profile is a single plateau centered at 1.7 V. The average discharge voltage is 1.85 V at the 10th cycle. All charge profiles have a single plateau, which is centered at ~ 2.3 V at the 2nd and 10th cycle. The polarization significantly increases, and the charge and discharge capacities decrease with cycle number. Cyclability of the cell is shown in **Fig. 2**. A reversible capacity retention of 66 % can be achieved after 50 cycles at 0.2 C. It is noted here that the coulombic efficiency is almost 100 %, meaning that there is no significant shuttle effect occurring during 50 cycles. The details and the further development of this work will be presented in the meeting.

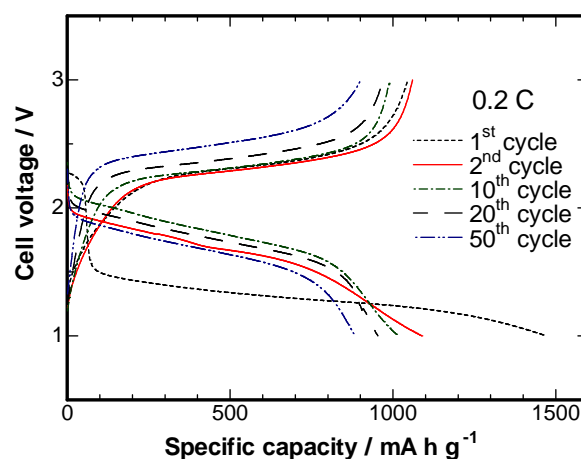


Fig. 1 Charge-discharge profiles of the Li-S battery at different cycles at 0.2 C.

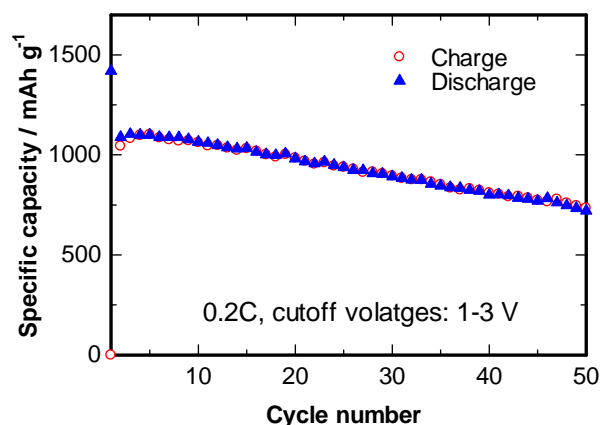


Fig. 2 Cyclability of the Li-S battery at 0.2 C.

References

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