



Enhancement of electrochemical performance of tin-based anode in lithium ion batteries by polyimide containing amino benzoquinone



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ABSTRACT

To study the effect of different polymer binders on the electrochemical performance of tin electrodes for rechargeable lithium-ion batteries, poly(vinylidene fluoride) (PVDF), conventional polyimide (PI-OB), and synthesized polyimide containing amino benzoquinone (PI-AQOB) were used as the polymer binders for electrodes consisting of commercial powdered Sn particles and Super P. PI-AQOB was converted from polyamic acid (PA-AQOB) synthesized from 2,5-bis(4,4'-oxydianiline)-1,4-benzoquinone (AQODA) and 4,4'-biphtalic dianhydride (BPDA) by condensation polymerization and characterized by Fourier-transform infrared analysis. Compared to the electrode employing the traditional PVDF binder, those with the PI-AQOB binder exhibited significantly enhanced electrochemical performance in terms of rate capability, specific capacity, and cycling behavior. PI-AQOB provided a high initial lithiation capacity of 1529 mAh/g at a current density of 50 mA/g. After 50 cycles, the PI-AQOB electrode maintained a higher specific capacity of 332 mAh/g than the Sn/PVDF electrode (only 65 mAh/g at a current density of 200 mA/g). Furthermore, the Sn/PI-AQOB electrode exhibited good volume restoration compared to the electrodes with Sn/PVDF and Sn/PI, as indicated by scanning electron microscopic analysis. The PI-AQOB binder increased the mechanical and adhesive strength of the electrode by suppressing pulverization of the Sn anode during expansion/contraction of Sn particles in the lithiation/delithiation process.

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1. Introduction

Lithium ion batteries (LIBs) have been widely used as the power source for various portable electronic devices, electric vehicles, and energy storage systems due to their higher operating voltage and energy density. The increasing demand for high-performance LIBs has prompted tremendous efforts to develop LIBs with high capacity, long cycle life, and fast charging rates [1–5]. Graphite is the active anode material most widely employed in conventional LIBs because of its low cost, low electrochemical potential, and excellent stability [6,7]. To achieve higher capacity and energy density, alternative anode materials that are superior to the graphite anode (theoretical capacity: 372 mAh/g) have been explored [8,9]. Metallic Sn is a promising alternative as an anode material for LIBs, where lithium can be reversibly alloyed/de-alloyed with Sn to obtain a high theoretical specific capacity ($\text{Li}_{4.4}\text{Sn}$: 992 mAh/g) [10–12]. However, Sn as an anode material suffers from severe vol-

ume change (>260%) during the alloying/dealloying process, which results in severe pulverization of the particles and mechanical damage to the electrodes, leading to a remarkable loss of capacity during the cycle life [13–18]. Several approaches have been proposed to curtail the severe volume change. These include (1) structural modifications such as using nano-sized or nanostructured materials [19,20], (2) incorporation with inactive materials or a matrix as a buffer to minimize the volume expansion [21–23], (3) potential control to form only partial alloys with Li^+ [24], and (4) use of new binders for better bonding between active materials, conducting carbon, and current collector.

The conventional binder, PVDF, has been used for both the anode and cathode due to its good electrochemical stability. However, the adhesion strength of PVDF to anode materials is not sufficient to accommodate the huge volume change, leading to poor cell performance. The effect of new binders on the performance of LIBs with metallic anode materials such as Si and Sn has been studied. Because metallic anode materials have exposed hydroxyl functional groups (-OH) on their surface, binders containing functional groups capable of polar bonding with the metallic surface have been studied. For example, alginic acid (AA) [25], polyacrylic acid (PAA)

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[26–28], carboxymethyl cellulose (CMC)/styrene butadiene rubber (SBR) [29], and polyimide (PI) have been reported to be effective binders for alloy-type (Si and Sn) electrodes. These binders have either carboxylate groups or imide groups that interact strongly with the surface of alloy particles during cycling. Notably, the aromatic PI binder has resulted in improved cell performance and suppressed collapse of electrode structures containing Si [30,31] and Sn [32] as anode materials due to the reactivity of the binder with Li^+ ions and its good thermal stability and mechanical and adhesive properties [33]. Ohta and coworkers [34] and Obrovac and coworkers [35] reported that PI binders modified with aromatic rings and functional groups gave rise to improved cell performance with highly uniform coverage of the metal surfaces.

In this study, we synthesize a new type of aromatic PI binder having amino benzoquinone that has high affinity to the surface of metals. The effects of the functional groups of the PI binder (PI-AQOB) on the electrochemical performance of a Sn particle anode are investigated by comparing with an untreated, conventional Sn particle anode employing PI-OB and PVDF binder, respectively. The

new PI binder results in improved initial capacity and cycle stability with accommodation of Sn expansion during alloying with Li.

2. Experimental

2.1. Materials

4,4'-Oxydianiline (ODA, >98%) and 4,4'-bipthalic dianhydride (BPDA, >98%) were purchased from TCI. 1,4-Benzoquinone ($\geq 98\%$) and 1-methyl-2-pyrrolidinone (NMP, anhydrous 99.5%) were obtained from Aldrich. Battery electrolyte comprising 1 M lithium hexafluorophosphate (LiPF_6) in ethylene carbonate (EC)/propylene carbonate (PC) (1:1 v/v) was purchased from Soulbrain. Fluoroethylene carbonate (FEC) as an electrolyte additive was obtained from Fousung. The electrolyte comprised 1 M LiPF_6 in EC/PC with 5 wt.% FEC containing less than 20 ppm moisture. BPDA was recrystallized from acetic anhydride and dried at 130°C . ODA, 1,4-benzoquinone, NMP, and the Sn nanoparticles were used without further purification. The Sn nanoparticles were purchased from Sigma-Aldrich. The particle size specified by the manufacturer was <150 nm with

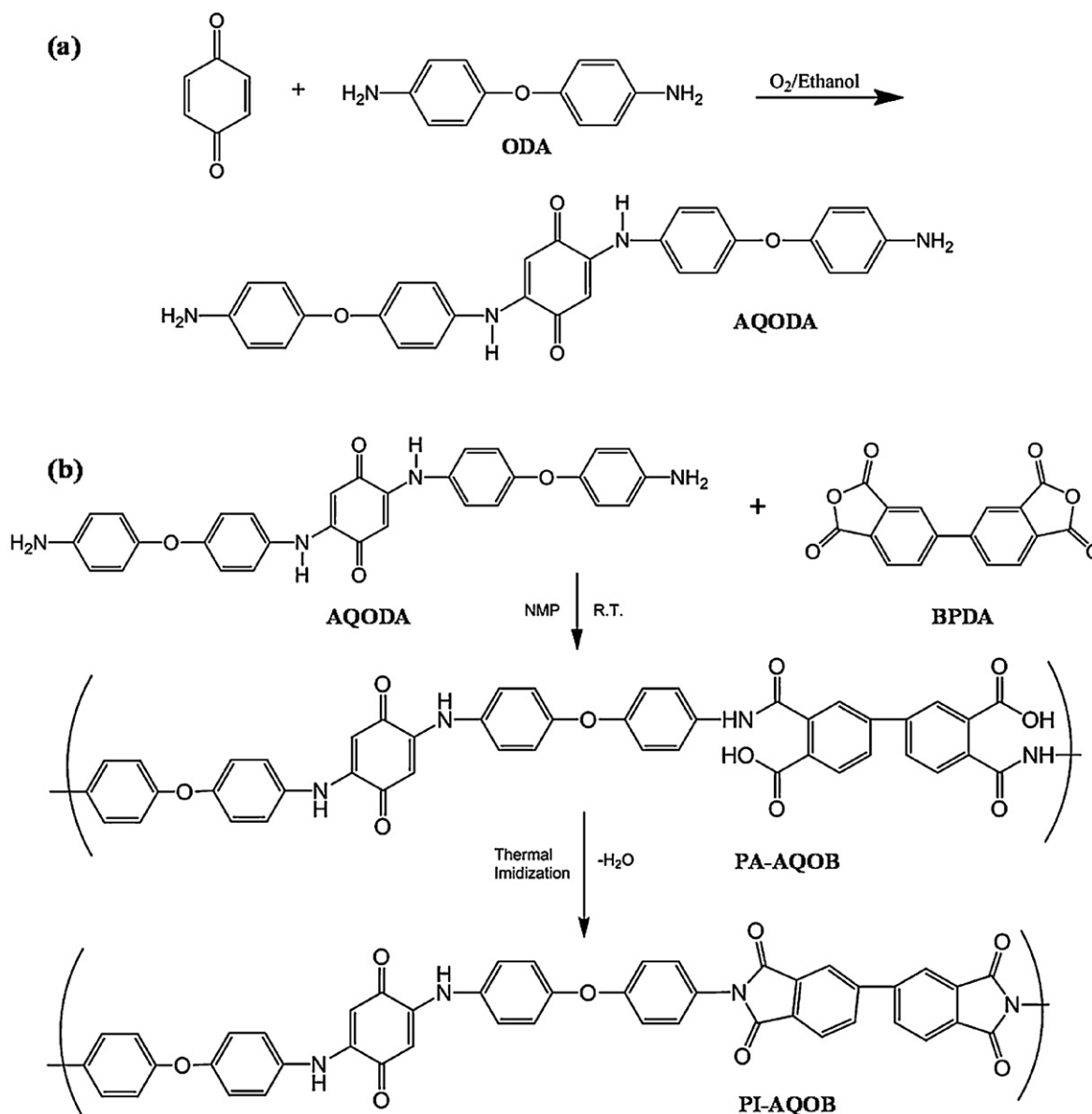


Fig. 1. Synthesis of (a) 2,5-bis(4,4'-oxydianiline)-1,4-benzoquinone (AQODA) and (b) polyimide (PI-AQOB).

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