



A benzoquinone-based cathode for Li-organic batteries



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ABSTRACT

An attractive organic electrode material, poly (2-chloro-3,5,6-trisulfide-1,4-benzoquinone) (PCTB), as an active material for rechargeable lithium batteries was investigated. It was prepared by polymerization of chloranil through thioether bonds. Because the existence of C–Cl in the product, PCTB exhibits a superior lithium-storage performance with high potential. A positive-electrode that incorporated PCTB showed a discharge capacity of 138.65 mAh/g at 30 mA/g with an average voltage of 2.72 V.

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1. Introduction

Battery technology is now recognized as a main bottleneck of the development of consumer electronics, electric vehicles, and energy storage stations [1]. The situation is that conventional Li-ion batteries meet the ceiling of energy density because of the limited choices of inorganic intercalation cathodes, while Li-organic batteries are full of potential to achieve not only significant advantages in resource sustainability and environmental friendliness, but also excellent electrochemical performance including high energy density [2,3].

Considering the advantages of the organic active materials, the development of rechargeable organic batteries would be desirable. It has been expected that the capacity and voltage of the organic cathode can easily be improved by optimizing their chemical structure. However, the cyclability of organic molecules is generally poor, because most organic cathode materials are easily dissolved in the electrolyte. In order to suppress the dissolution, several investigations have been carried out. One of the most effective strategies is polymerization. Recent investigations revealed that quinone-based polymers along with the stable thioether bonds could achieve a high specific capacity and a good cycling stability simultaneously. Song and coworkers [4] reported the synthesis of poly (benzoquinonyl sulfide) (PBQS), and it was used as a lithium-ion battery cathode. Wu et al. [5] reported poly (2,5-dihydroxy-p-benzoquinonyl sulfide) as a novel sodium ion batteries anode material for the first time. Bitenc et al. [6] reported poly (hydroquinonyl-benzoquinonyl sulfide) (PHBQ) as an active mate-

rial in Mg and Li organic batteries the system of which retains more than 160 mAh/g after 340 cycles. Those investigations revealed that quinone-based polymers along with the stable thioether bonds could achieve a high specific capacity and a good cycling stability simultaneously.

Herein, we report on an attractive organic electrode material, poly (2-chloro-3,5,6-trisulfide-1,4-benzoquinone) (PCTB), as an active material for rechargeable lithium batteries.

2. Experimental

2.1. Materials

2,3,5,6-Tetrachloro-1,4-benzoquinone (TB) (Aladdin, 98%), Na₂S·9H₂O (Aladdin, 98%), N,N-Dimethylformamide (DMF)(Aladdin, 99.9%) were used without further purifications.

2.2. Materials synthesis

TB (1.23 g) reacted with Na₂S·9H₂O (4.80 g) in 20 mL aqueous solution, heated at 100 °C and stirred 3 h to obtain sodium salt. An additional mole of TB in DMF (20 mL) was added into sodium salt solution (after cooling to 90 °C), stirred 10 h. After filtration, the sample was washed with distilled water and ethanol, and then dried under vacuum at 120 °C to obtain PCTB product.

2.3. Characterization

Fourier transform infrared spectrometer (FTIR) was used to characterize the structure of the obtained sample. FTIR spectroscopy was recorded by using KBr pellet on a Nicolet 6700 in a

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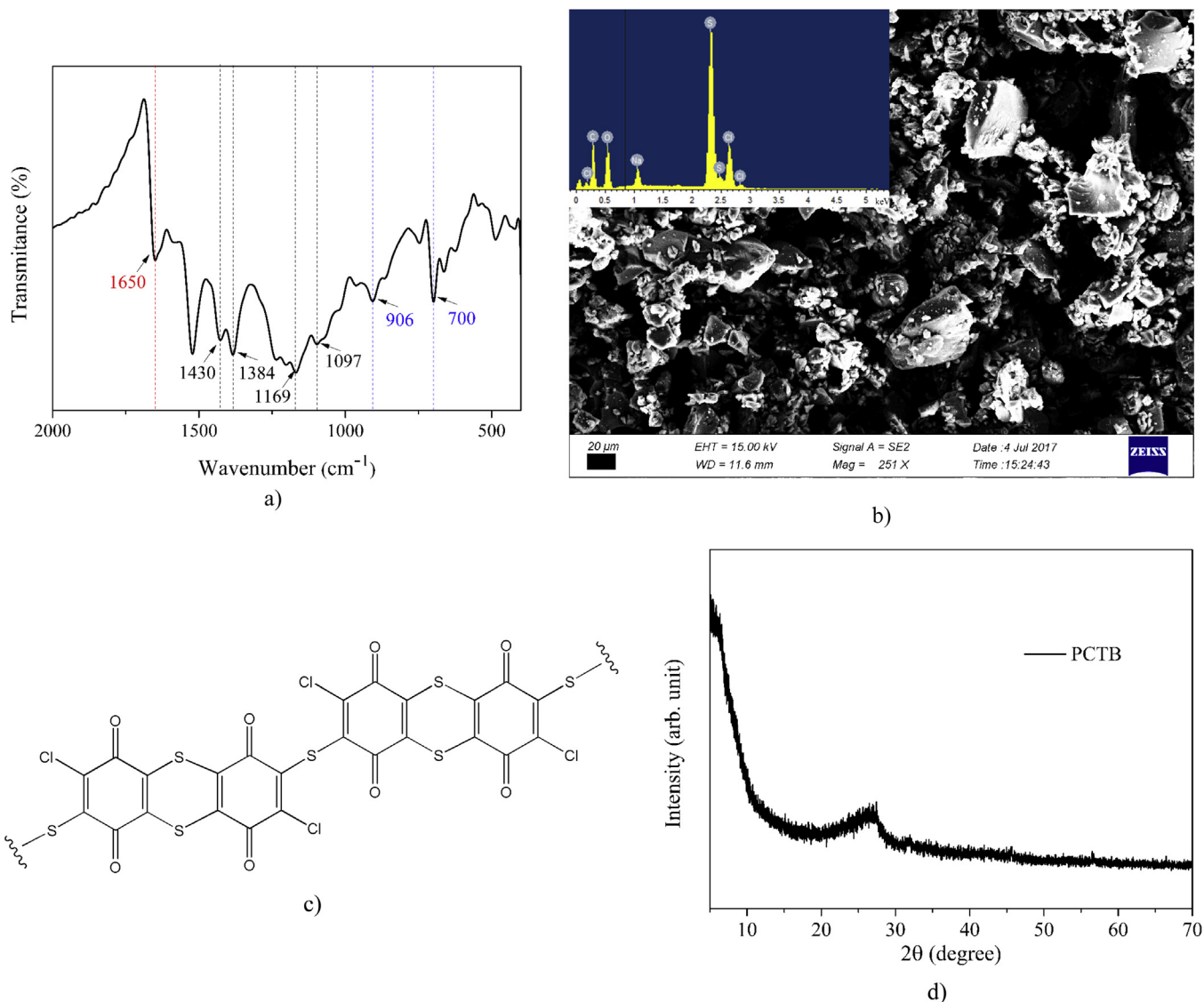


Fig. 1. a) FTIR of PCTB, b) SEM and EDX pattern of PCTB, c) proposed structure of PCTB sample, d) XRD spectra of PCTB.

wavenumber range of 400–2000 cm^{-1} . Energy dispersive X-ray spectroscopy (EDX) and the surface morphology of the particle were observed using SEM (SIGMA 300). Elemental analysis (EA) of CHNSO, and Cl were conducted on Vario EL cube (Elementar, Germany) and ICS-2000 ion chromatography system (Dionex, America). X-ray diffractometry spectroscopy (XRD) were conducted on a D8-FOCUS X-ray diffractometer (Bruker, Germany) with $\text{Cu K}\alpha$ ($\lambda = 1.54 \text{ \AA}$) radiation.

2.4. Electrochemical measurement

CR2016 coin-type cells were used for cycling tests. The cathode film contained active material (50 wt%), conductive carbon (30 wt %) and poly (vinylidene fluoride) binder (PVDF, 20 wt%), with aluminum mesh as current collector. The electrolyte was 1 M LiPF_6 solution (in EC: DMC/1:1 in volume ratio). The cells were cycled on a Land battery test system (CT2001A, China). Cyclic voltammetry (CV) measurements were conducted on a three-electrode cell (Li/Li/PCTB) on Interface 1000E electrochemical workstation (Gamry, America). The electrochemical impedance spectroscopies (EIS) was measured using a frequency response analyser (CHI

660D, shanghai) with an electrochemical interface in the frequency range from 10^5 to 10^{-2} Hz at an amplitude of 10 mV.

3. Results and discussion

The chemical structure of the as-prepared sample was characterized by FTIR. As shown in Fig. 1a, the strong absorption band near 1650 cm^{-1} corresponds to the stretching vibration of the $\text{C}=\text{O}$ [6]. The peak at 1430, 1384, 1169 and 1097 cm^{-1} can be assigned to the $\text{C}-\text{S}$ [7], indicating the successful formation of the $\text{C}-\text{S}-\text{C}$ during the polymerization process. Compared with TB, it is noticed that the absorption peaks at 906 and 700 cm^{-1} do not completely disappear, which means that still exist of $\text{C}-\text{Cl}$ in the product [8]. The relatively low potential of the organic electrodes limited the overall energy and power density while incorporated into a full device. By adding electron withdrawing groups, the potential which the organic cathode accepts an electron is increased [3]. So the existence of $\text{C}-\text{Cl}$ in the products may be the reason why its oxidation and reduction potential higher than poly(2,3-dithiobenzophenone) [7].

The resulting PCTB was in the form of black powder and exhibited irregular morphology with an average size in the micron range

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