



Selenium/carbon-rich core–shell composites as cathode materials for rechargeable lithium–selenium batteries



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HIGHLIGHTS

- The Se/C core–shell composites are synthesized by a one-step solution route.
- The Se/C composites exhibit good electrochemical properties than pristine selenium.
- The Se/C composites would be a promising cathode material for Li–Se batteries.

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ABSTRACT

Selenium/carbon-rich (Se/carbon-rich) core–shell composites are prepared by a one-step hydrothermal synthesis method as a cathode for rechargeable lithium batteries. The Se/carbon-rich composites are characterized and examined by X-ray diffraction (XRD), field emission scanning electron microscopy (SEM), transmission electron microscopy (TEM), and thermogravimetry (TGA) measurements. It is demonstrated from galvanostatic discharge/charge process that the Se/carbon-rich composites exhibit the discharge capacity of 558 mA h g^{−1} in the first cycle and maintain capacity of 181 mA h g^{−1} after 80 cycles at a rate of 0.5C, which is better than that of the pristine selenium. The result demonstrates that the unique core–shell structure is effective in suppressing the dissolution of polyselenides into the electrolyte and in maintaining high utilization of the active materials during the charge/discharge process. It provides a new selenium-based cathode material for rechargeable lithium batteries.

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1. Introduction

The rapidly developing market for the emerging plug-in hybrid vehicle and mobile electronics has prompted the urgent need for rechargeable batteries with high energy density. Despite the numerous advantages, the overall energy density of lithium ion batteries is limited by the low capacity of current cathode materials [1–3]. Therefore, cathode materials with high specific capacity have been extensively investigated. Among them, Lithium–sulfur (Li–S) battery has attracted extensive attention in the last two decades due to high theoretical capacity (1675 mA h g^{−1}) of sulfur cathode. However, the sulfur cathode still suffers from some issues [4–6]: (1) sulfur has low electronic conductivity; (2) high-order polysulfide intermediates are soluble in organic electrolytes; (3) sulfur undergoes large volume change during lithiation/delithiation. As a

congener of sulfur, selenium as a potential cathode material was studied in lithium batteries. Selenium has a theoretical gravimetric capacity of 675 mA h g^{−1} based on molecular weight of 78.86 g mol^{−1}, which is lower than sulfur (1672 mA h g^{−1}). However, its theoretical volumetric capacity of 3253 mA h cm^{−3} based on 4.82 g cm^{−3} is comparable to that of sulfur (1672 mA h g^{−1}). Moreover, selenium has higher electrical conductivity, approximately 20 orders of magnitude greater than sulphur [7–12]. The advantages of selenium make it an alternative promising cathode material in high-energy density lithium batteries for specific applications. However, similar to sulfur cathodes, one of the biggest challenges to selenium is the dissolution issue of high-order polyselenides, resulting in a rapid capacity decrease, low Coulombic efficiency [9].

To address these issues, many experiments have been made, which focus on enhancing the electrical conductivity of the cathode and suppressing the loss of soluble polyselenide intermediates during cycling. Various conductive porous carbons [9,10,13–19],

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porous metal oxide [20], conducting polymers [12,21], and graphenes [12,22] have been used as host materials for selenium cathode to improve the electrochemical performance of Li–Se batteries. Abouimrane et al. [7] conducted pioneering work on the use of Se and SeS_x as a cathode material for rechargeable Li-ion and Na-ion batteries. Wang et al. [9] synthesized the selenium-impregnated carbon composites containing 30 wt% selenium by infusing Se into mesoporous carbon at a temperature of 600 °C under vacuum, exhibiting good electrochemical performances. Kundu et al. [12] coated conducting polymer polypyrrole and graphene on selenium nanofiber to improve the electrochemical performance and minimize the polarization between charging and discharging. Most of these method employed by heating treatment to confine Se into the pores of absorbing materials, thus these composites can improve electrical conductivity and trap some polyselenides during cycling. However, these cathodes were prepared by cumbersome synthetic processes. To make the selenium encapsulated porous structure via the templates, multiple synthetic steps are needed.

In this study, we report the selenium/carbon-rich core–shell composites prepared by a one-step hydrothermal synthesis method. The outer carbon shell not only ensures high electronic conductivity of the composite, but provides a nanoscale capsule to confine selenium and polyselenides. As a cathode for rechargeable lithium batteries, it exhibits good cycling performance and high Coulombic efficiency.

2. Experimental section

2.1. Material synthesis

The synthesis process for Se/carbon-rich core–shell composites is illustrated in Fig. 1. All the reagents were analytical grade. 0.07 g of sodium selenite (Sigma, 99.9% metals basis) was dissolved in 40 ml of ultrapure water containing 0.08 g of CTAB (Aladdin, 99.9% metals basis). Then 0.8 g of ascorbic acid (Aladdin, 99.9% metals basis) was added to the solution under vigorous stirring. The colour of the dispersion changed from colourless to red suggesting formation of the amorphous selenium. Then the mixture was transferred into a Teflon-lined autoclave with 60 ml capacity, followed by hydrothermal treatment at 200 °C for 24 h in an oven. After

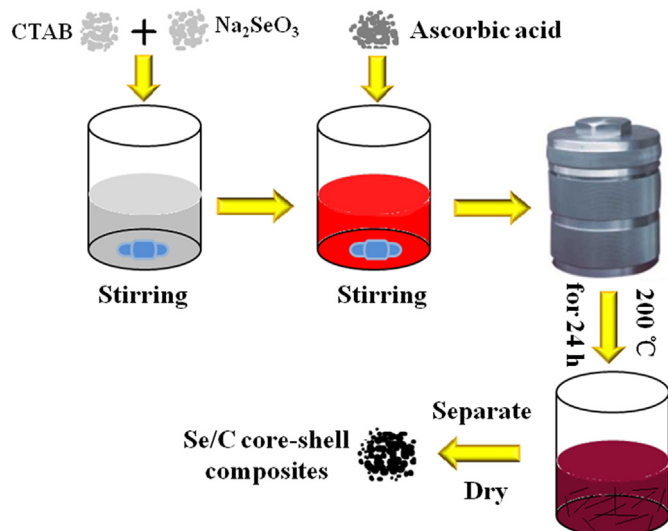
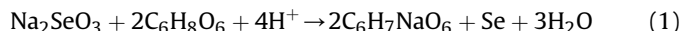


Fig. 1. Schematic illustration for the preparation process of Se/carbon-rich core–shell composites.

cooled to room temperature, the final product was collected by centrifugation and washed with distilled water and ethanol several times. Finally, the products were dried in vacuum at 80 °C for 12 h. The experiment treats sodium selenite with ascorbic acid to obtain Se [Eq. (1)]. Ascorbic acid was employed as both reductant and carbon source to form a carbon shell.



2.2. Materials characterization

The overall morphology of the samples was examined with scanning electron microscope (SEM, Sirion 200) and transmission electron microscopy (TEM, Tecnai G2 20ST). The elements on the surface of the samples were identified by energy-dispersive X-ray spectroscopy (EDX). Powder X-ray diffraction (XRD, Rigaku3014) using Cu K α radiation was employed to identify the crystalline phase of Se/carbon-rich core–shell composites. Thermogravimetric analysis (TGA, SDTQ600) was conducted in determining the selenium content in the composites.

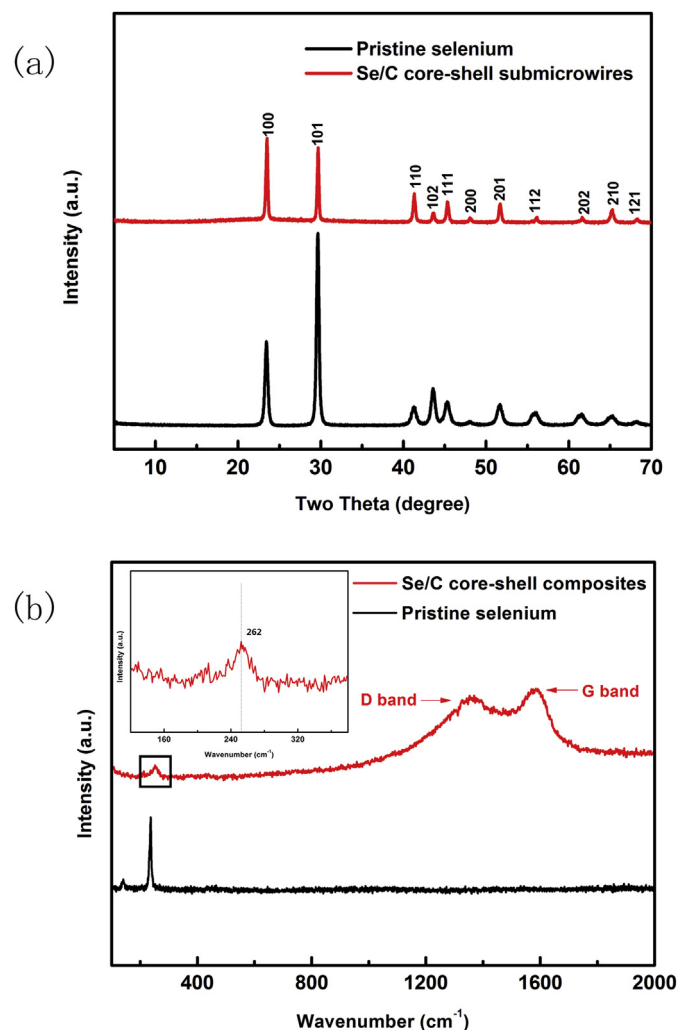


Fig. 2. (a) X-ray diffraction patterns and (b) Raman spectra of the Se/carbon-rich core–shell composites and the pristine selenium.

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