

Supplementary Information

One-step microwave synthesized core-shell structured selenium@carbon spheres as cathode materials for rechargeable lithium batteries

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Experimental

Materials Synthesis

Core-shell structured Se@C spheres were synthesized using SeO₂ (Sigma, 99.9%) as selenium source and sucrose (Aladdin, AR) as carbon source through a facile microwave method. Polyvinyl Pyrrolidone (PVP-1300000, Aladdin) was used as the dispersant. In a typical synthesis, 3 mmol SeO₂ and 1 g sucrose were dissolved in 30 ml of deionized water with various amounts of PVP. Then the clear solution was magnetically stirred for 30 min to ensure the homogeneity of all the constituents. After that, the mixture was transferred into the reaction vessel, which is a double-walled digestion vessel consisting of a Teflon TFM inner liner and cover surrounded by high strength vessel shell. The mixture was microwave-heated at 200 °C for 30 min and cooled down to room temperature naturally. The products were collected by centrifugation, washed with deionized water more than 10 times, and further dried with a freeze-drier (SCIENTZ-10N, China).

The microwave oven (2.45 GHz, maximum power 300 W) used was a focused single-mode microwave synthesis system (Discover, CEM, USA). The unique, circular single-mode cavity, which was equipped with an in-situ magnetic stirring and a water-cooled condenser outside, ensured that samples were in a homogenous highly dense microwave field. Temperature was accurately controlled by automatic adjusting of microwave power.

Materials Characterization

Elemental analyses were performed on a Vario EL III analyzer (Elementar). X-ray powder diffraction (XRD) patterns were taken using a Rigaku Ultima IV with Cu-K α radiation (λ = 1.54056 Å) operated at 40 kV and 200 mA with a scan speed of 6° min⁻¹ and a scan range of 10-80°. The morphology of samples were characterized by field emission scanning electron microscopy (SEM, JSM-6700) and transmission electron microscopy (TEM, JEOL JEM-2010). The elemental mapping results were examined with an energy dispersive spectrometer (EDS, Oxford) attached to the JEOL JEM-2010 TEM.

Electrochemical measurements

80 wt % of Se@C composites, 10 wt % of super P, 5 wt % of carboxyl methyl cellulose, 5 wt % of styrene-butadiene rubber and a proper amount of deionized water, were mixed through ball milling at 250 r/min for 4 h to form a well-dispersed slurry. Then the slurry was uniformly cast on an aluminium foil with the doctor blade method (at a height of 60 μ m). After evaporation of the solvent, the electrodes were further dried in a vacuum oven at 60 °C for 12 h and then were cut into discs with a diameter of 12 mm just prior to use. The active mass (selenium) loading was about 1-2 mg cm⁻². CR2025 type coin cells were assembled in an argon-filled glove box (O₂, H₂O < 1 ppm) to assess the electrochemical performance of Se@C materials. A solution of 1 M lithium bis(trifluoro-

methanesulfonyl)imide (LiTFSI) dissolved in 1, 3-dioxolane (DOL) / 1, 2-dimethoxyethane (DME) with a volume ratio of 1:1 was adopted as the electrolyte. Lithium foils and celgard 2320 were used as the anode and the separator, respectively. The cells were discharged and charged at various current densities in the voltage range of 1.0 - 3.0 V vs Li/Li⁺ on a LAND CT2001A battery test system (Wuhan, China). Cyclic voltammetry (CV) curves were recorded on an Autolab Electrochemical Workstation (ECO CHEMIE B.V, Netherlands) at 0.1 mV/s in the potential range of 1.0-3.0 V. All electrochemical measurements were carried out at ambient temperature.

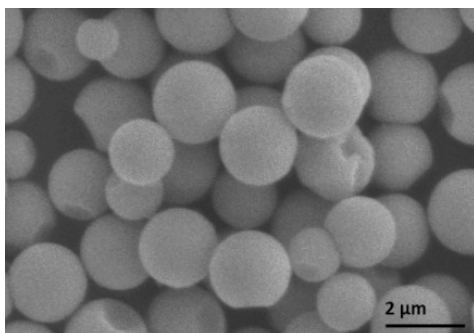


Figure S1. SEM image of Se@C composites synthesized with the soaking time of only 1 minute.

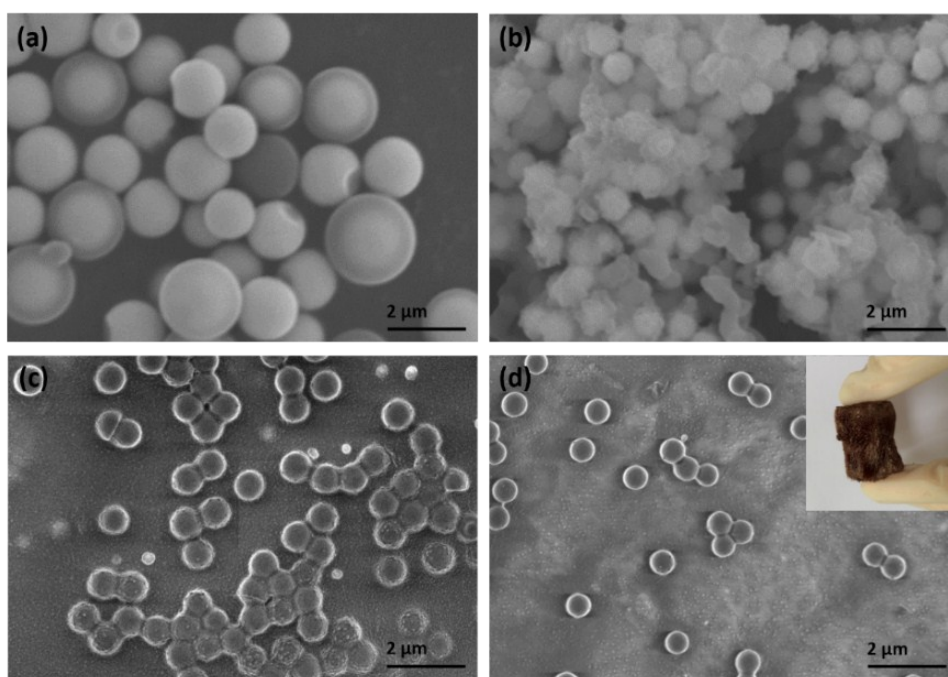


Figure S2. SEM images of Se@C composites synthesized using different amounts of PVP: (a) PVP free; (b) 1% PVP; (c) 3% PVP; (d) 5% PVP. The digital photo of the sample-5% PVP is presented in the inset of (d).

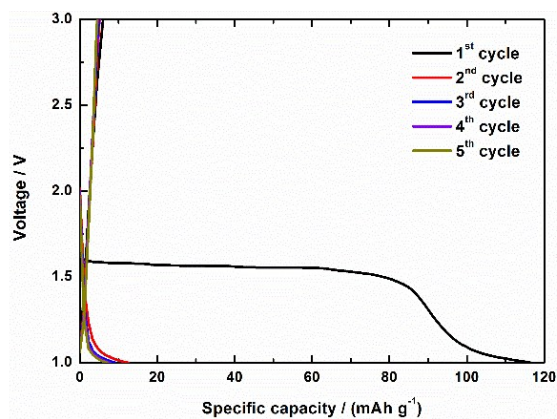


Figure S3. Discharge/charge profiles of the gel like Se@C composite.

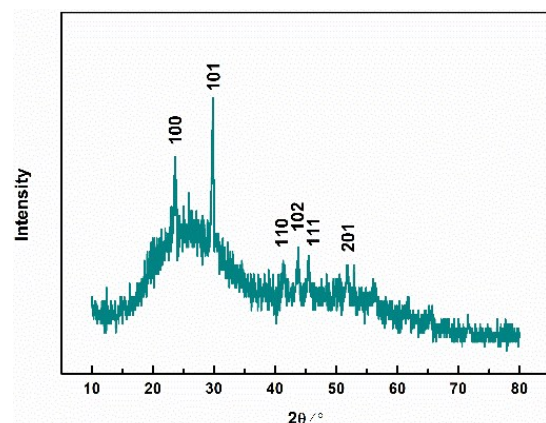


Figure S4. The XRD pattern of Se@C composite.

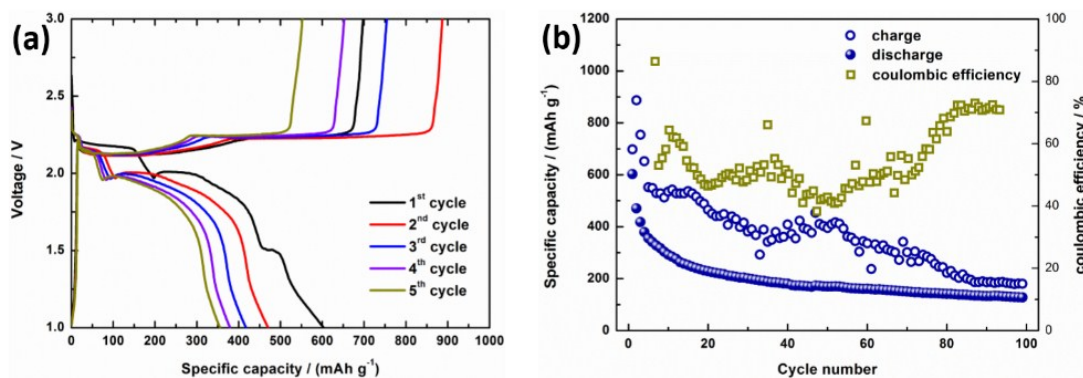


Figure S5. Discharge/charge profiles (a) and cycling performance & Coulombic efficiency (b) of pristine Se.

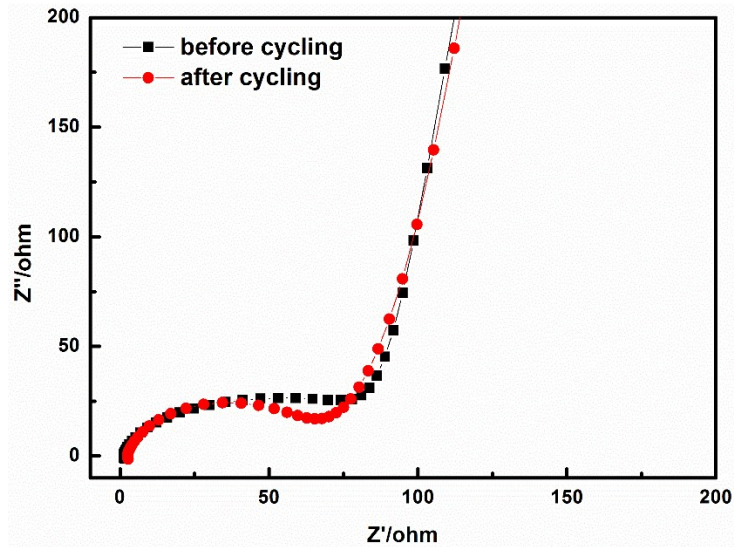


Figure S6. The Nyquist plots of the cells before and after 50 cycles at 0.1 C. The impedance spectra are composed of a depressed semicircle at high frequency and an inclined line at low frequency. The diameter of the semicircle is used to determine the charge transfer resistance of the cell and the inclined line reflects the Li ion diffusion into the active materials.

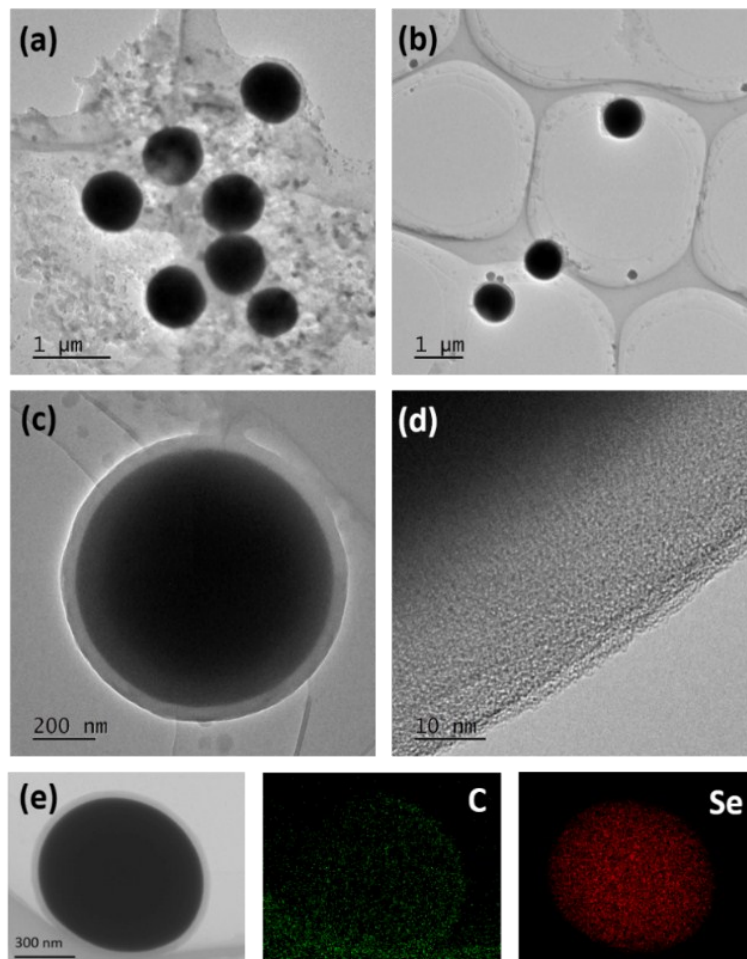


Figure S7. TEM images (a-c), HRTEM image (d) and bright-field (BF)-STEM image (e) and corresponding

C, Se elemental mapping of Se@C cathode materials after 1st discharge.

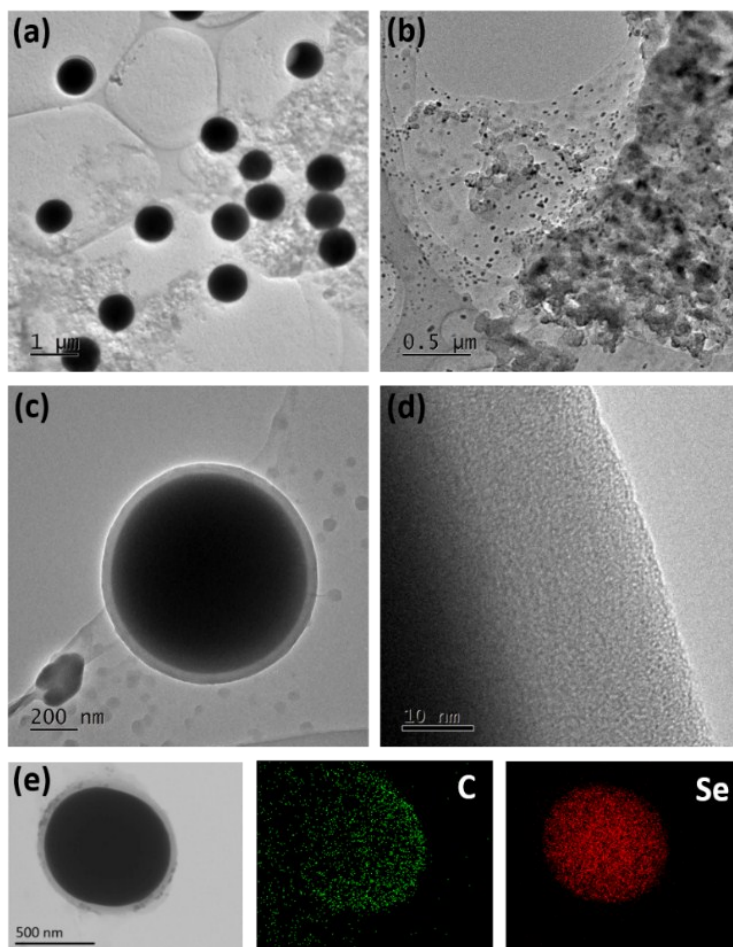
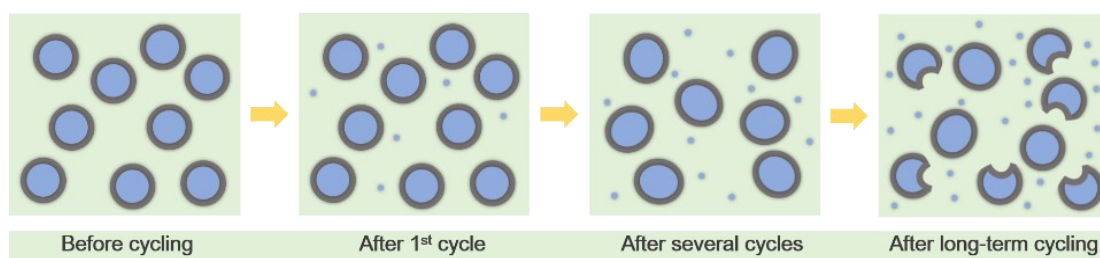


Figure S8. TEM images (a-c), HRTEM image (d) and bright-field (BF)-STEM image (e) and corresponding C, Se elemental mapping of Se@C cathode materials after 5th fully charged cycles.



Scheme S1. Schematics of morphology change of Se@C cathodes during cycling.