Supporting Information

Localized polyselenides with a graphene-coated polymer separator for high rate and ultralong life lithium-selenium batteries

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Experimental details

Graphene-Polymer Separator Fabrication

The graphene was fabricated by Deyang Carbonene Technology, using a process invented by Pei *et al.*[1] (Chinese patent No. CN201110282370.5). The graphene-polymer separator was fabricated by a vacuum-filtration process. Typically, the graphene was dispersed in ethanol to form a suspension with a concentration of 0.2 mg mL^{-1}, then the suspension was vacuum filtered onto one side of a commercial polypropylene separator (Celgard 2400), followed by air drying at 80℃ for 30 min. The graphene-polymer separator was then punched into circular disks.

Materials Characterization

The morphology and structure of the samples were characterized by using a scanning electron microscope (SEM) (FEI Nova NanoSEM 430, 15 kV) and a transmission electron microscope (TEM) (Tecnai F20, 200 kV). Raman measurements were performed using a Jobin Yvon LabRAM HR800, excited by a 632.8 nm He-Ne laser. X-ray diffraction (XRD) patterns were recorded on a Rigaku diffractometer using Cu Kα radiation.

Electrochemical Measurements

The cathode slurry was fabricated by mixing 70 wt% selenium as active material, 20 wt% Super P as conductive agent and 10 wt% polyvinylidene fluoride (PVDF) as binder in N-methyl-2-pyrolidone (NMP). The slurry was coated onto an Al foil followed by drying under vacuum at 100 ℃ for 24 h. The dried electrode was punched into circular disks with a diameter of 12 mm and the active material loading is about 4 mg cm^{-2}. CR-2025 type stainless steel coin cells were assembled with lithium metal foil as anode, selenium as cathode and a polypropylene polymer or an as-prepared graphene-polymer as separator. The electrolyte was prepared by dissolving lithium bis-trifluoromethanesulphonylimide (LITFSI, 99%, Acros Organics, 1 M) and lithium nitrate ($LiNO_3$, 99.9%, Alfa Asea, 0.1 M) in 1, 2-dimethoxyethane (DME, 99.5%, Alfa Asea) and 1,3-dioxolane (DOL, 99.5%, Alfa Asea) (1:1 ratio, by volume). A LAND galvanostatic charge–discharge instrument was used to perform electrochemical measurements. The C-rate used here was based on the theoretical specific capacity of selenium (678 mAh g^{-1}), the current density was varied from 0.339 to 3.39A g^{-1} referring to the mass loading of selenium in the cathode. The charge-discharge voltage range was 1.7-2.8 V. The CV test was performed using a VSP-300 multichannel potentiostat/galvanostat workstation operated in the voltage range of 1.7-2.8 V (*vs*. Li^+/Li) at a scan rate of 0.1mV s^{-1}.

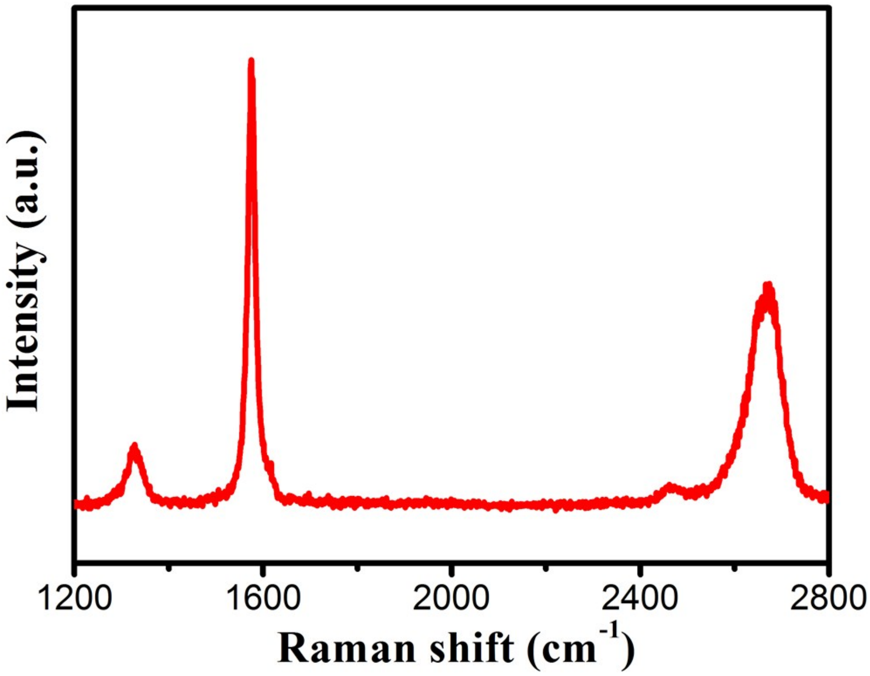

Fig. S1 Raman spectrum of the graphene used for the graphene-polymer separator.

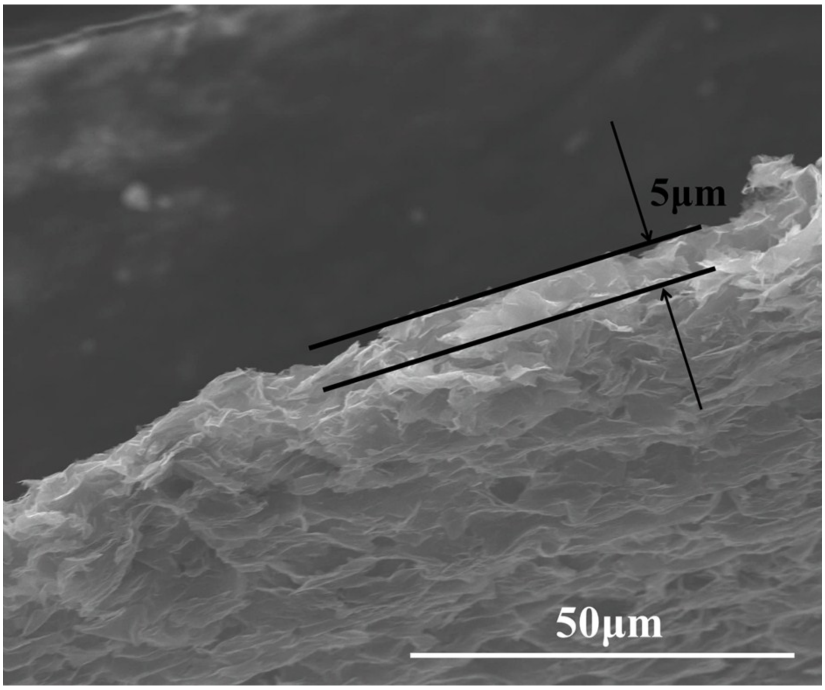

Fig. S2 Cross-sectional SEM image of the graphene layer in a graphene-polymer separator.

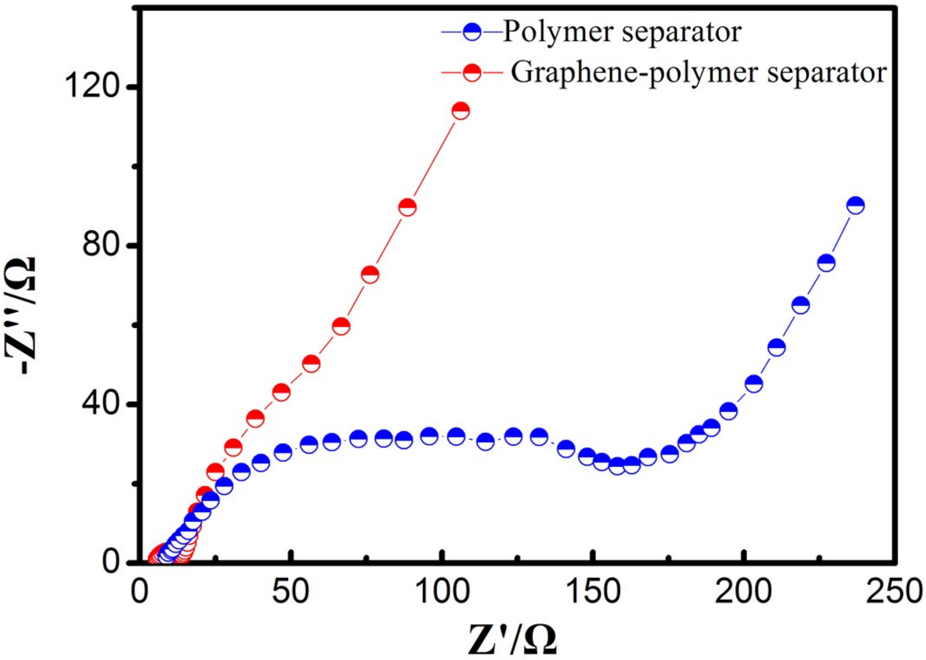

Fig. S3 Electrochemical impedance spectra of Li-Se cells with different separators recorded after 10 cycles (inset is the magnified plot for the cell with a graphene-polymer separator).

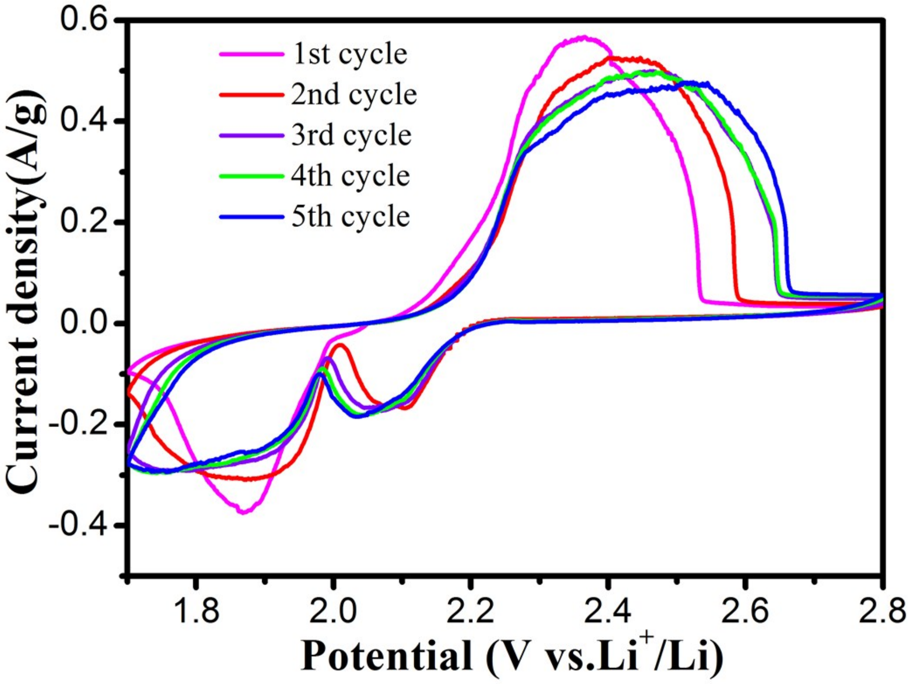

Fig. S4 CV curves of the cell with a polymer separator at a scan rate of 0.1 mV s^{-1} for five cycles.

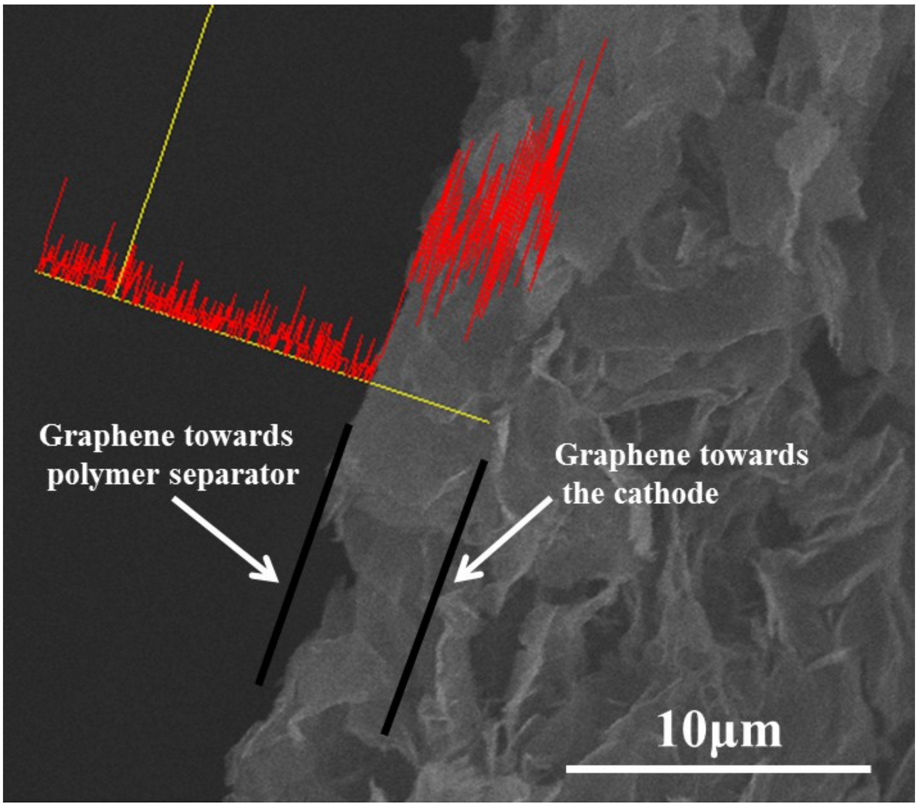

Fig. S5 Cross-sectional elemental line scan of Se on the graphene interlayer after cycling.

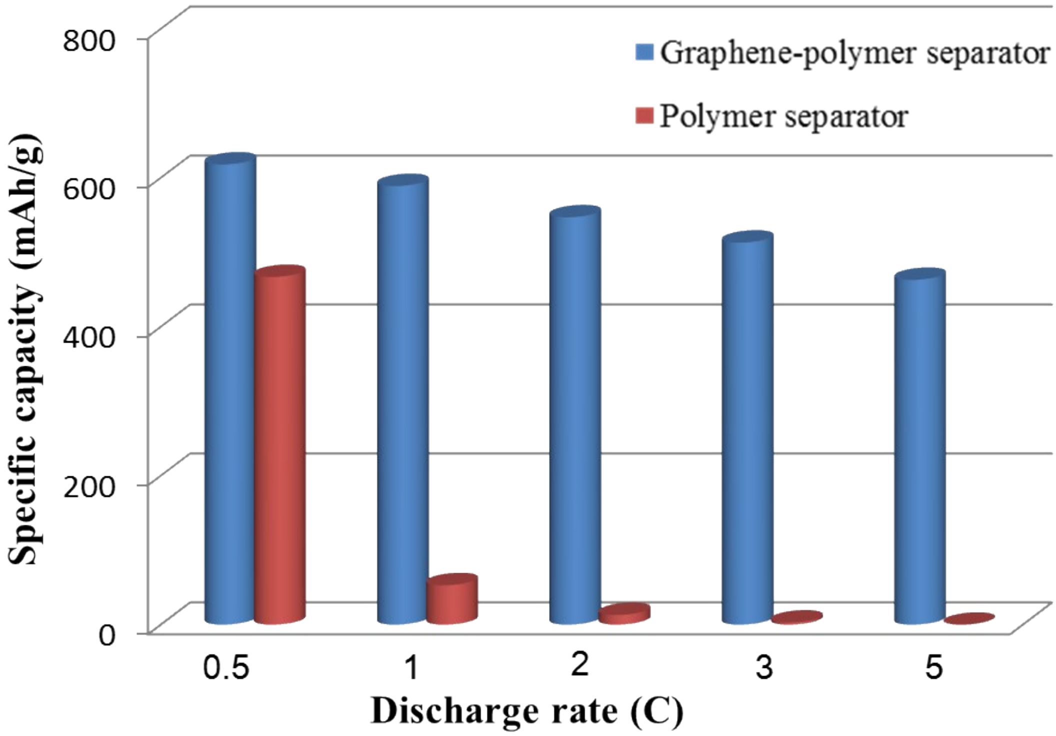

Fig. S6 Rate performance of Li-Se cells with different separators at different rates.

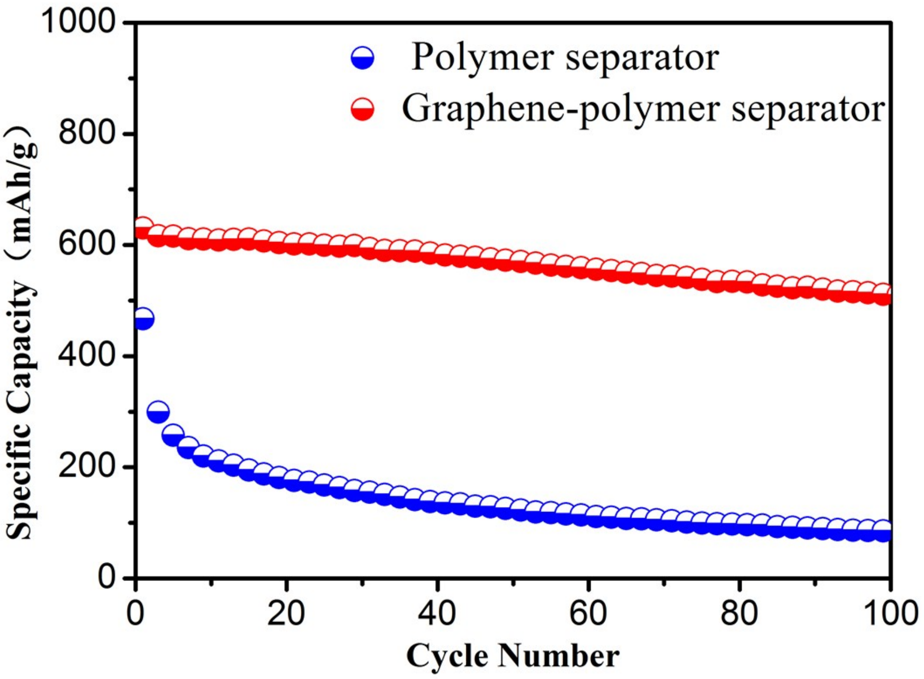

Fig. S7 Cycling stability of a Li-Se cell at 0.5C.

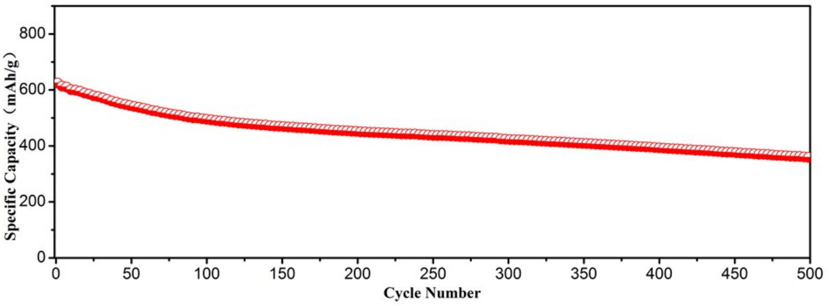

Fig. S8 Cycling stability of a Li-Se cell with a graphene-polymer separator at 1C.

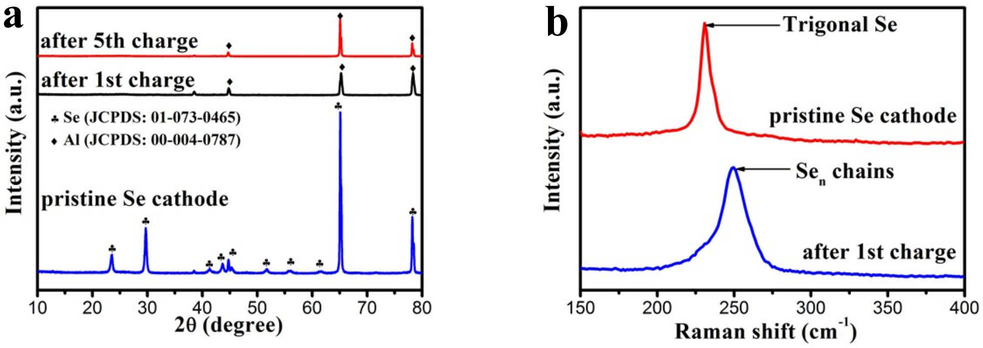

Fig. S9 (a) XRD patterns and (b) Raman spectra of the Se cathode at different charge states.

1 W. C. Ren and H. M. Cheng, *Nat. Nanotechnol.*, 2014, **9**, 726-730.