

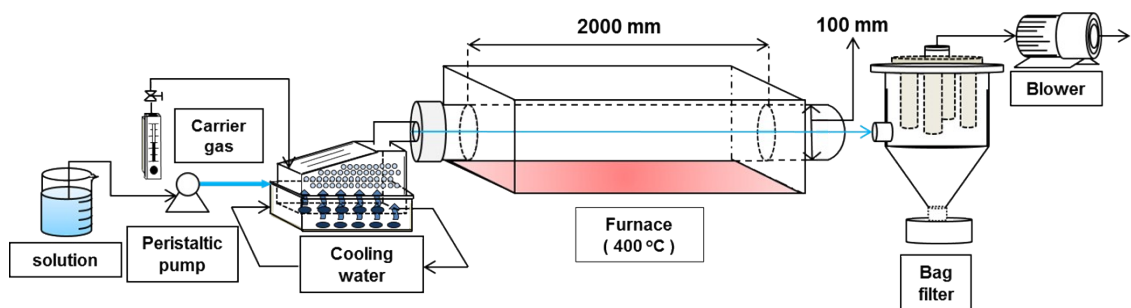
**Yolk-shell carbon microspheres with controlled yolk and void  
volumes and shell thickness and their application as a cathode  
material for Li-S batteries**

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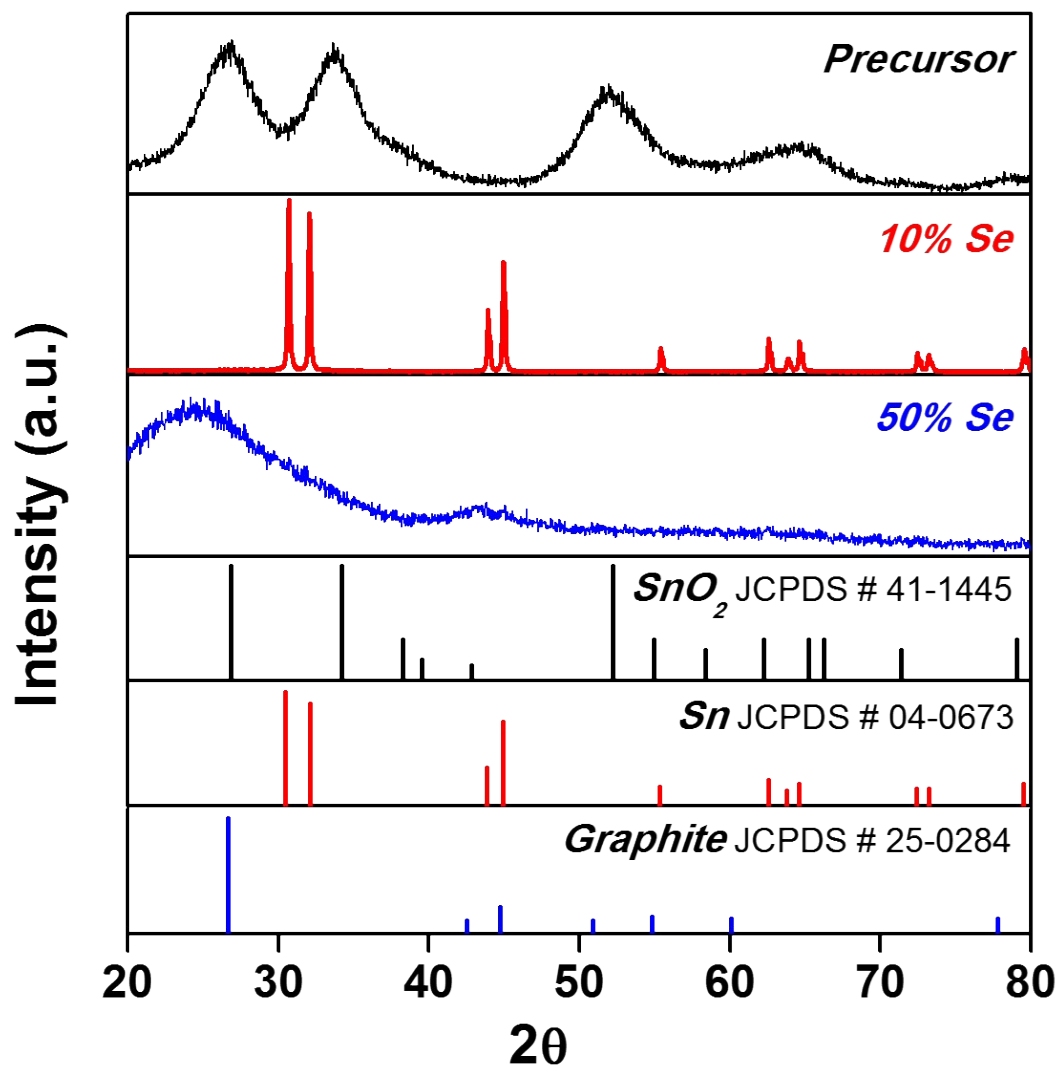
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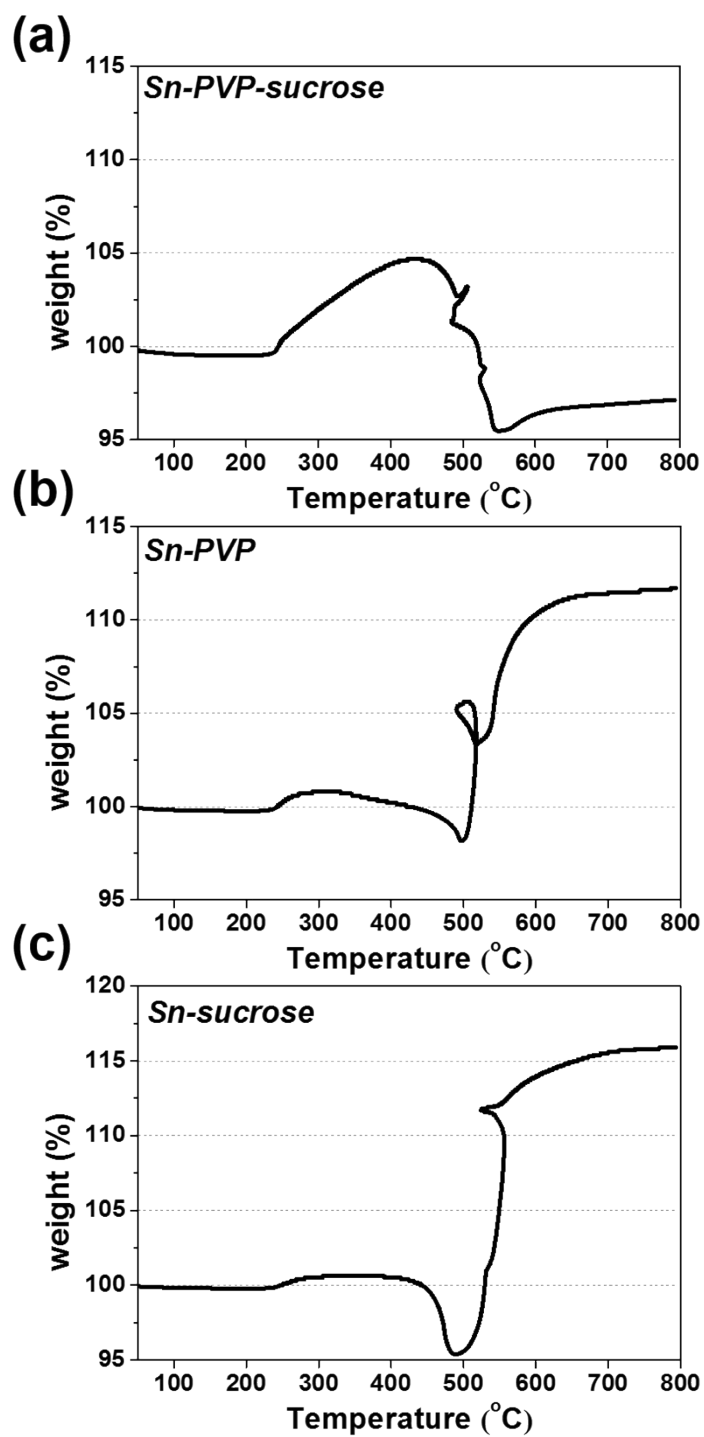
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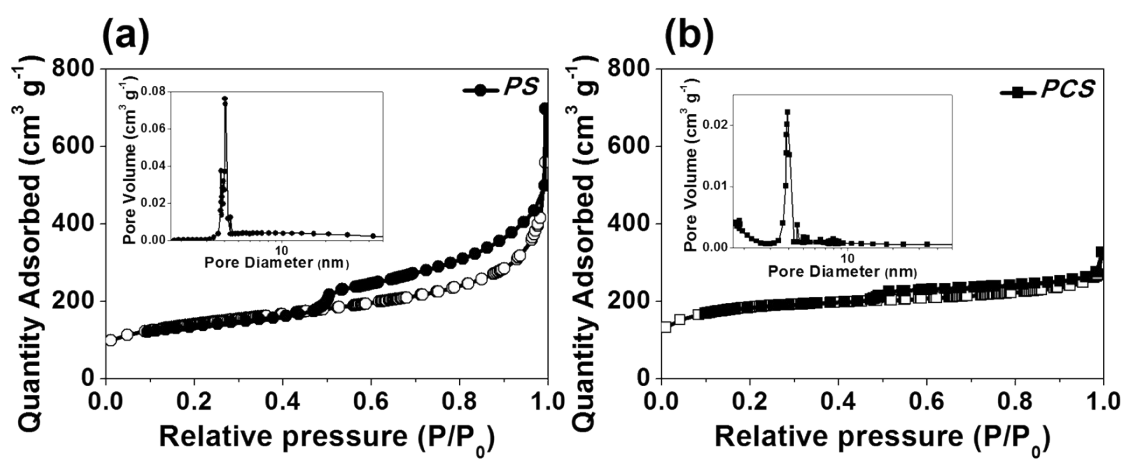
**Fig. S1.** Schematic diagram of the large-scale spray pyrolysis process applied in this study.



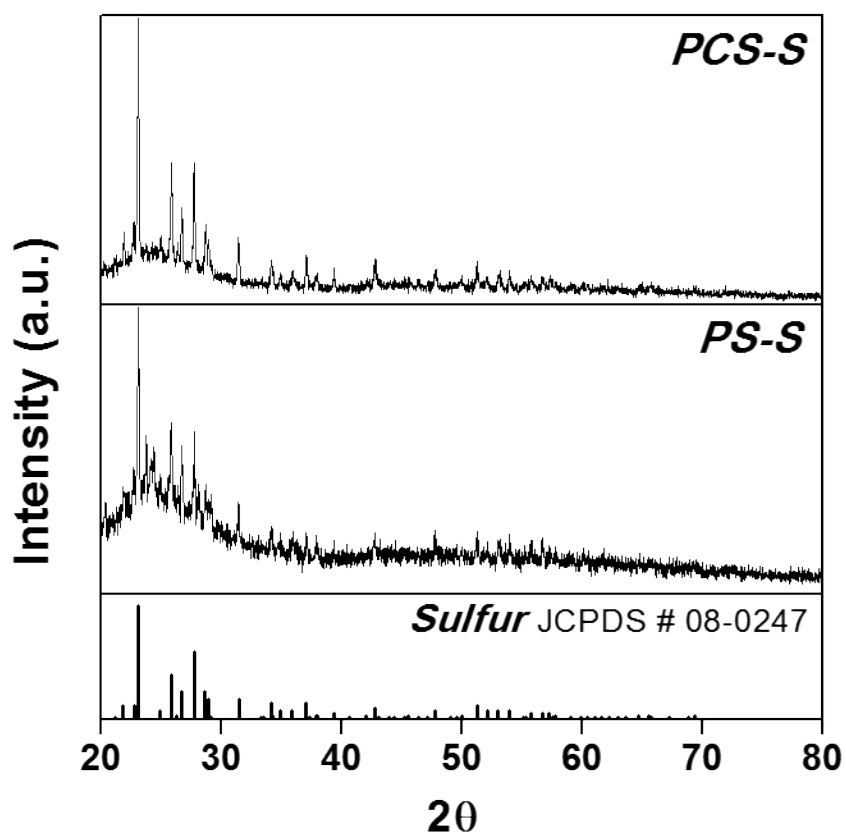
**Fig. S2.** XRD patterns of the SnO<sub>2</sub>/carbon-carbon core-shell-structured microspheres prepared from the spray solution containing Sn oxalate and PVP before and after post-treatment with different quantities of Se.



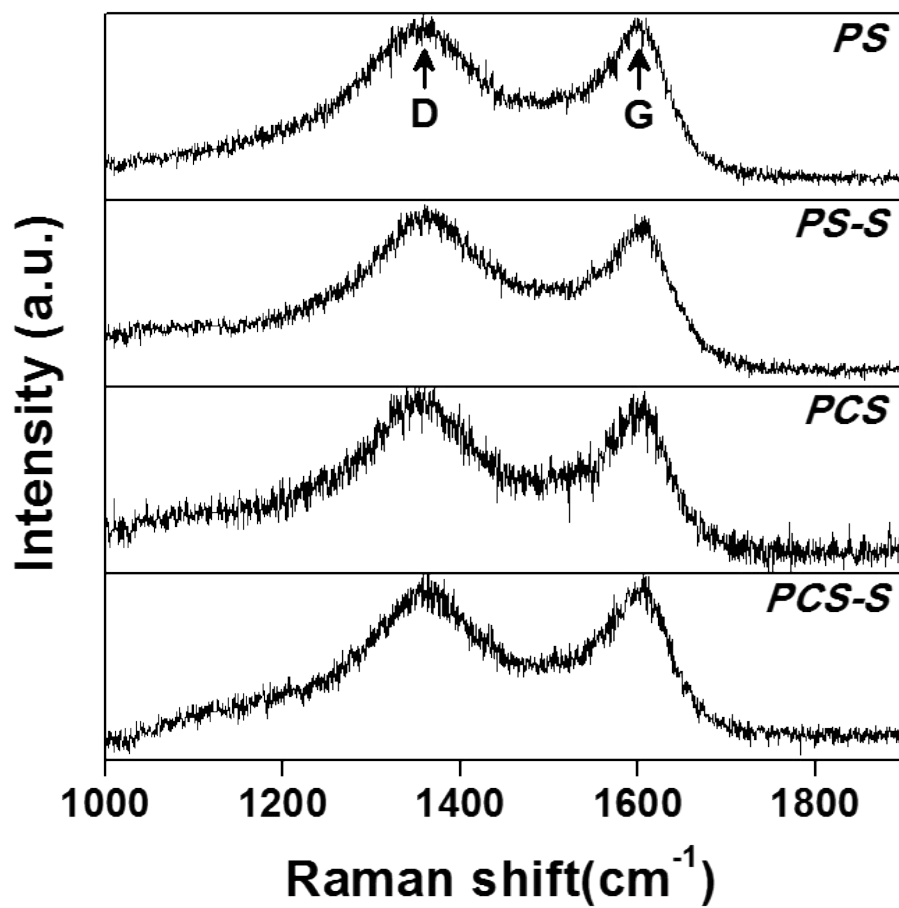
**Fig. S3.** TG curves of the Sn-C microspheres prepared from the spray solution with (a) both PVP and sucrose, (b) PVP alone, and (c) sucrose alone as carbon sources.



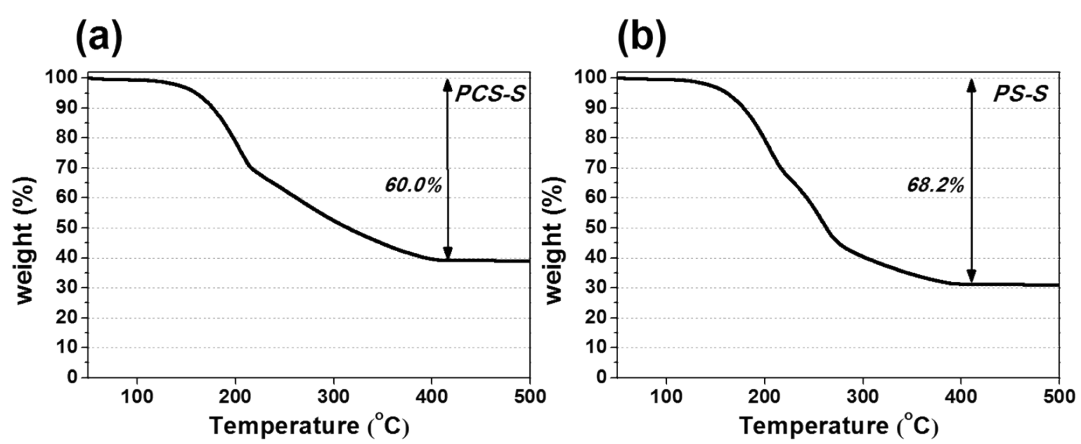
**Fig. S4.** Nitrogen adsorption and desorption isotherms and pore size distributions of the (a) PS and (b) PCS microspheres.



**Fig. S5.** XRD patterns of the PS-S and PCS-S microspheres.

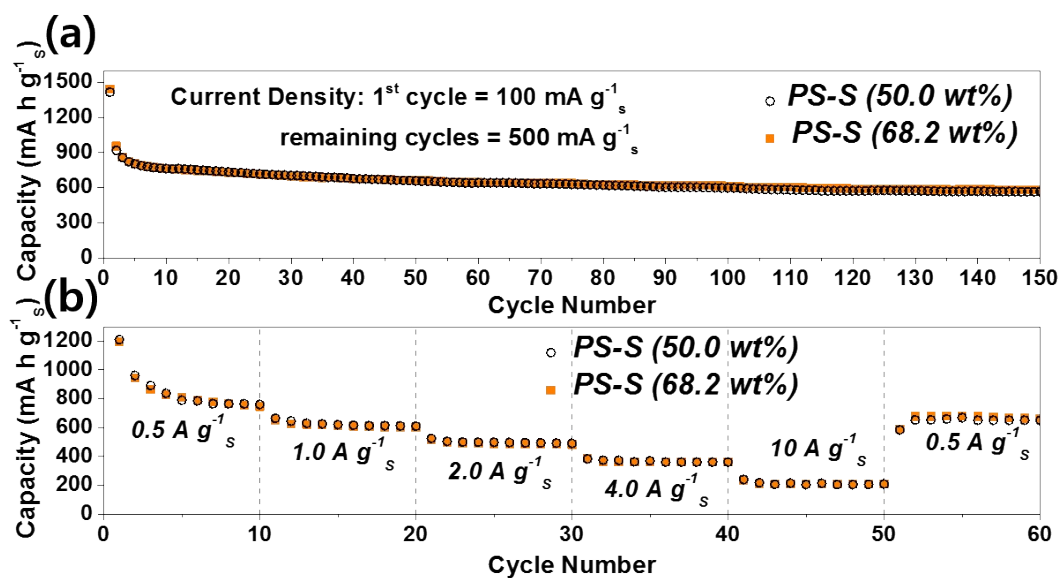


**Fig. S6.** Raman spectra of PS, PS-S, PCS, and PCS-S microspheres.

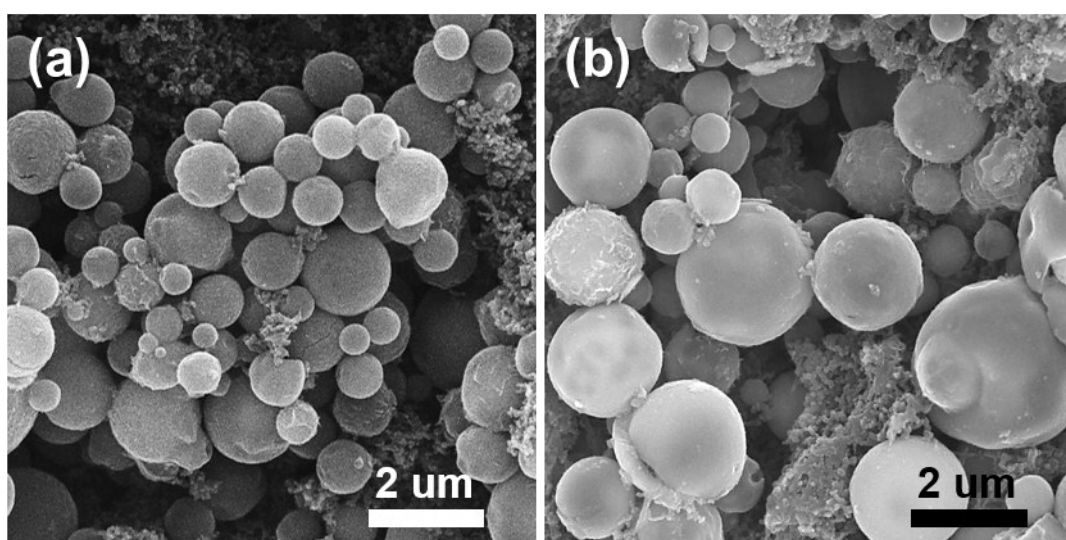


**Fig. S7.** TG curves of (a) PCS-S and (b) PS-S microspheres.

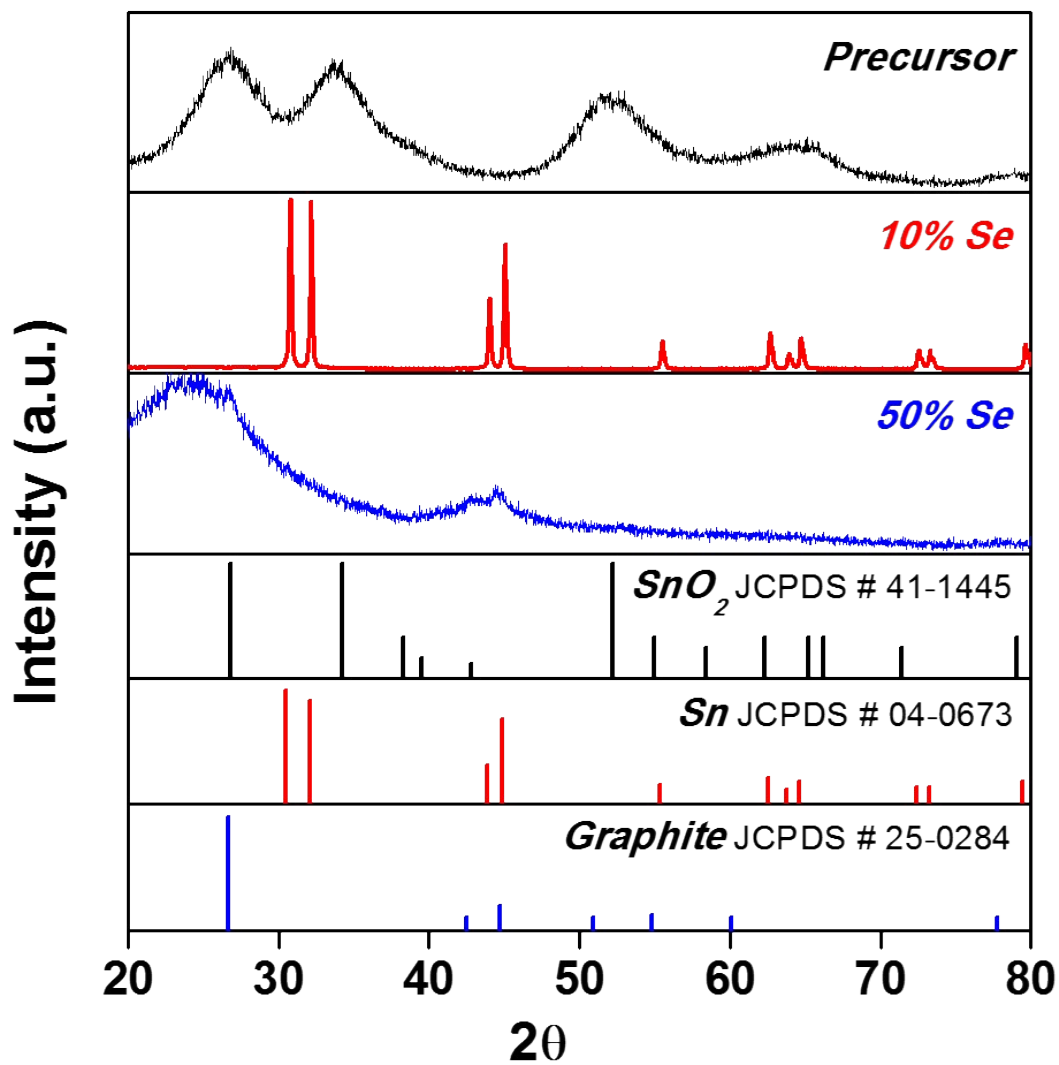




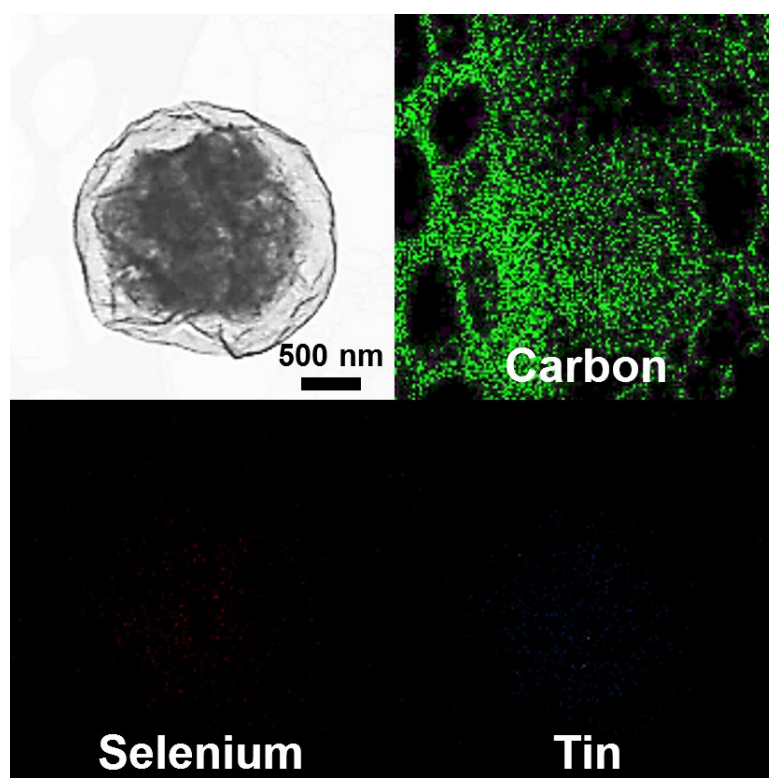
**Fig. S8.** Electrochemical properties of PS-S microspheres with 50.0 and 68.2 wt% sulfur: (a) cycling performances and (b) rate performances.



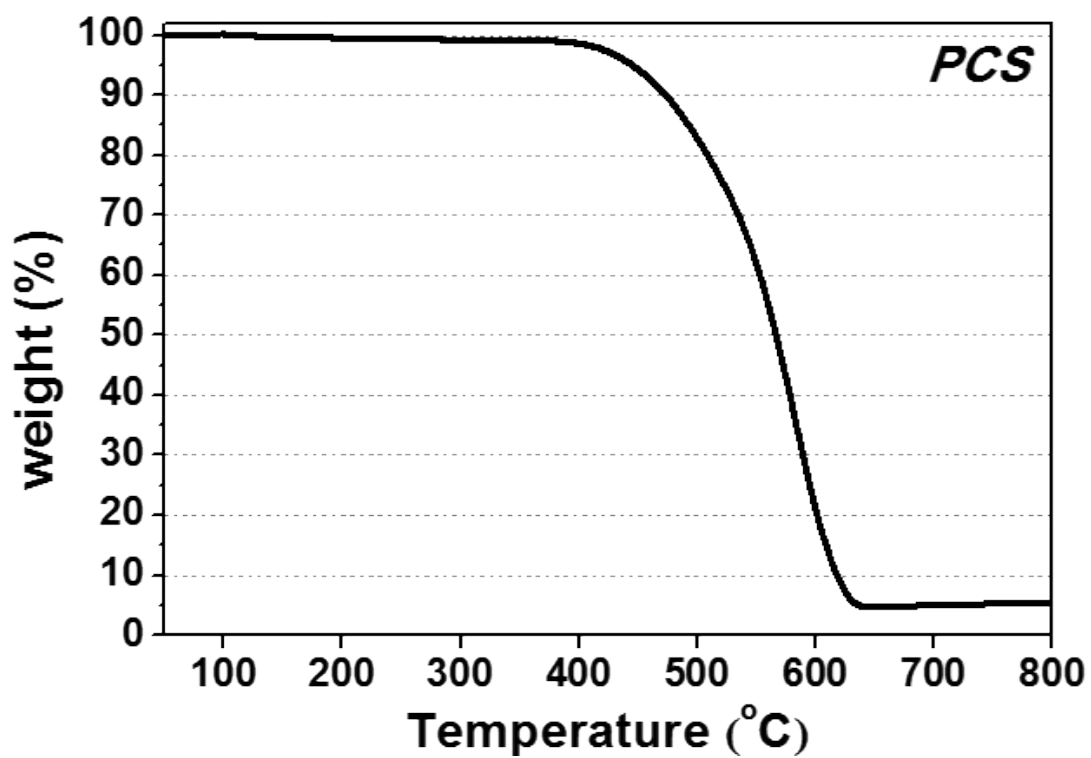
**Fig. S9.** Morphologies of the (a) PCS-S and (b) PS-S microspheres obtained after the 50<sup>th</sup> cycle.



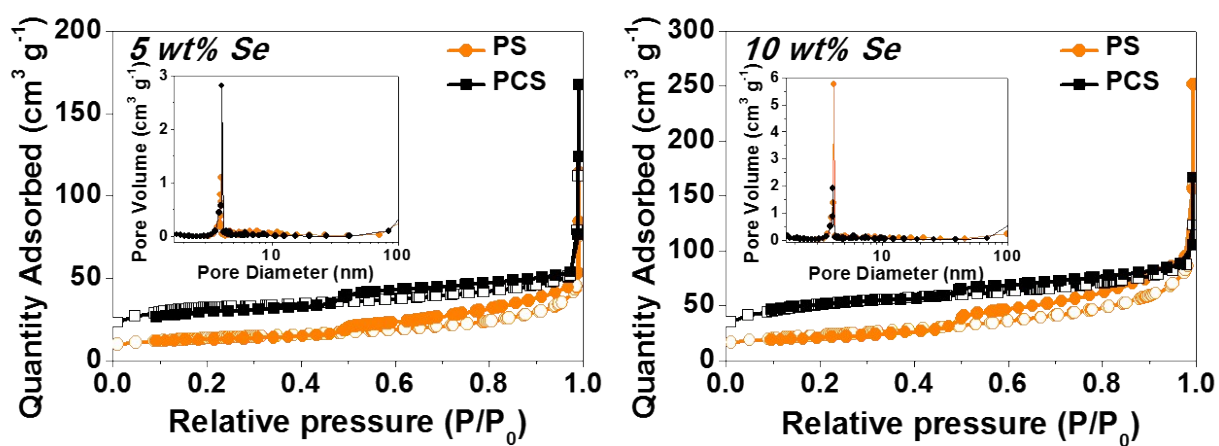
**Fig. S10.** XRD patterns of the SnO<sub>2</sub>/carbon-carbon core-shell-structured microspheres prepared from the spray solution containing Sn oxalate, PVP, and sucrose before and after post-treatment with different quantities of Se.



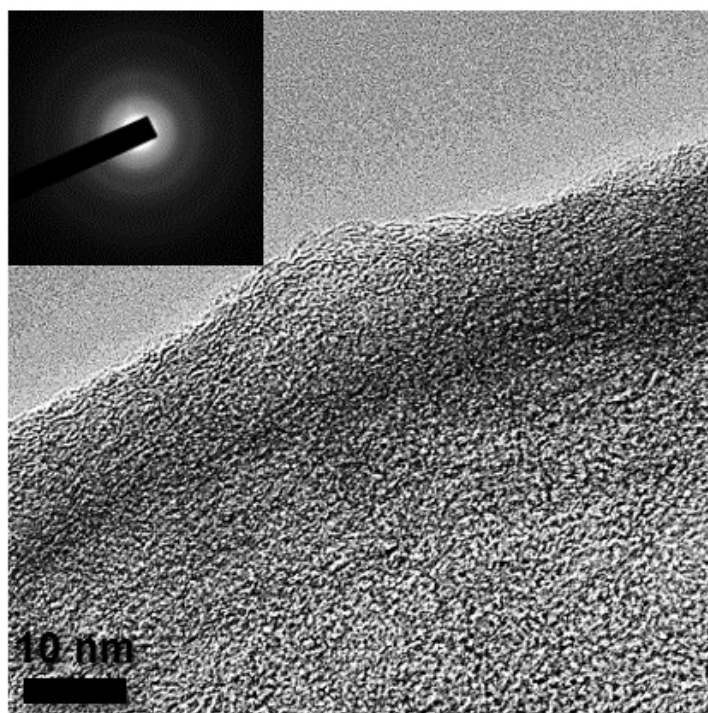
**Fig. S11.** TEM and dot-mapping images of the carbon yolk-shell microspheres prepared from the spray solution containing Sn oxalate, PVP, and sucrose.



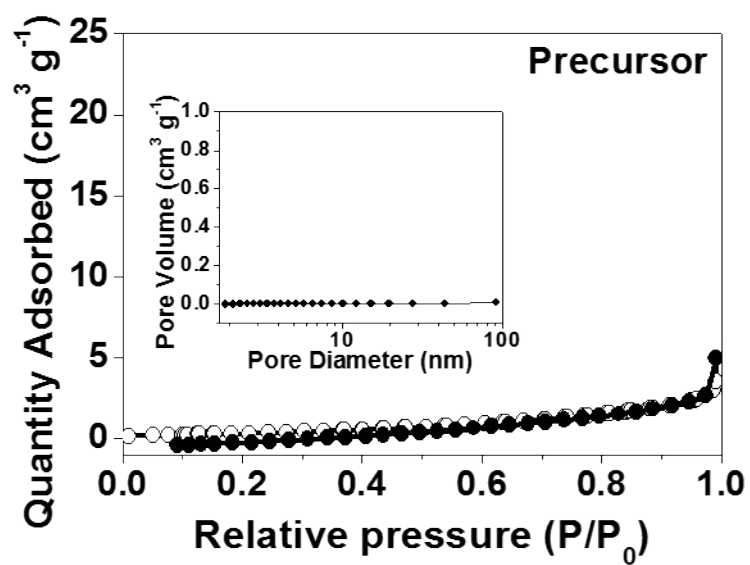
**Fig. S12.** TG curve of the carbon yolk-shell microspheres prepared from the spray solution containing Sn oxalate, PVP, and sucrose.



**Fig. S13.** Nitrogen adsorption and desorption isotherms of the SnO<sub>2</sub>/carbon-carbon core-shell-structured microspheres after post-treatment with different quantities of Se.



**Fig. S14.** High resolution TEM image and SAED pattern of the carbon yolk-shell microspheres prepared from the spray solution containing Sn oxalate and PVP.



**Fig. S15.** Nitrogen adsorption and desorption isotherms of the SnO<sub>2</sub>/carbon-carbon core-shell-structured microspheres prepared from the spray solution containing Sn oxalate, PVP, and sucrose.



**Table S1.** Li-ion storage properties of the carbon materials as cathode materials for Li-S batteries reported in the previous literatures.

Morphology [preparation method]	S content [wt%]	Current density	Initial $C_{\text{dis}}$ [mA h g <sup>-1</sup> ] <sub>s</sub>	Discharge capacity [mA h g <sup>-1</sup> ] <sub>s</sub>	Cycle number	Ref.
hollow carbon nano sphere [direct carbonization]	61	837.5 mA g <sup>-1</sup> (0.5 C)	1043	967	100	[22]
polydopamine-coated, nitrogen-doped, hollow carbon [silica template]	65	1003 (0.6 C)	740	630	600	[24]
multi-shelled hollow carbon nanospheres [aqueous emulsion approach]	86	167.3 mA g <sup>-1</sup> (0.1 C)	1350	1250	200	[25]
hierarchical porous carbon [spray pyrolysis]	46	4020 mA g <sup>-1</sup> (2.4 C)	700 (5 <sup>th</sup> cycle)	539	500	[27]
porous hollow carbon spheres [template strategy]	50.2	83.75 mA g <sup>-1</sup> (0.05 C)	1450	1357	50	[28]
hollow-in-hollow carbon spheres [template-assisted]	70	1000 mA g <sup>-1</sup>	1080 (3 <sup>rd</sup> cycle)	780	300	[38]
tube-in-tube carbon nanostructure [SiO <sub>2</sub> template]	71	2000 mA g <sup>-1</sup>	659	647	200	[39]
double-shelled hollow carbon [hard template]	64	167.5 mA g <sup>-1</sup> (0.1 C)	~1000	690	100	[40]
Yolk-shell carbon microspheres [spray pyrolysis]	60	500 mA g <sup>-1</sup>	908 (2 <sup>nd</sup> cycle)	600	150	This work