

## An Advanced Design Concept of Mansion-like Freestanding Silicon Anodes with Improved Lithium Storage Performances

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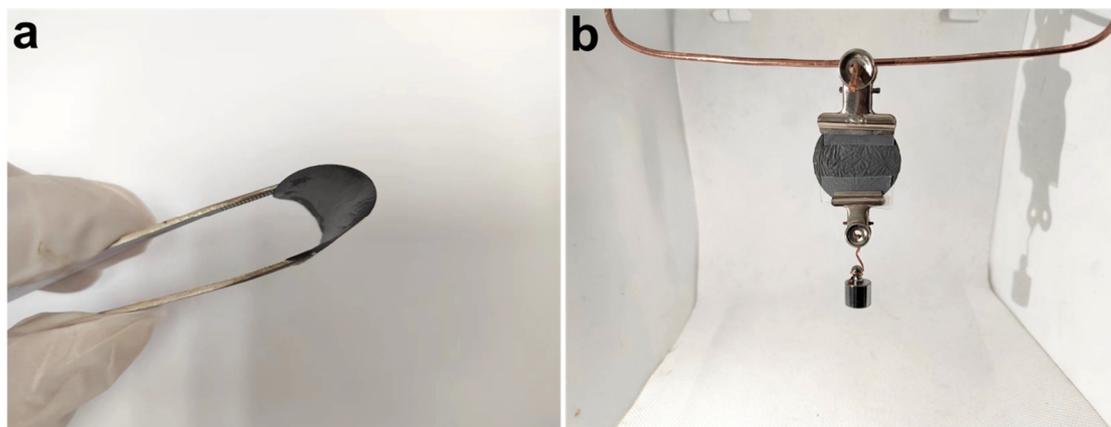
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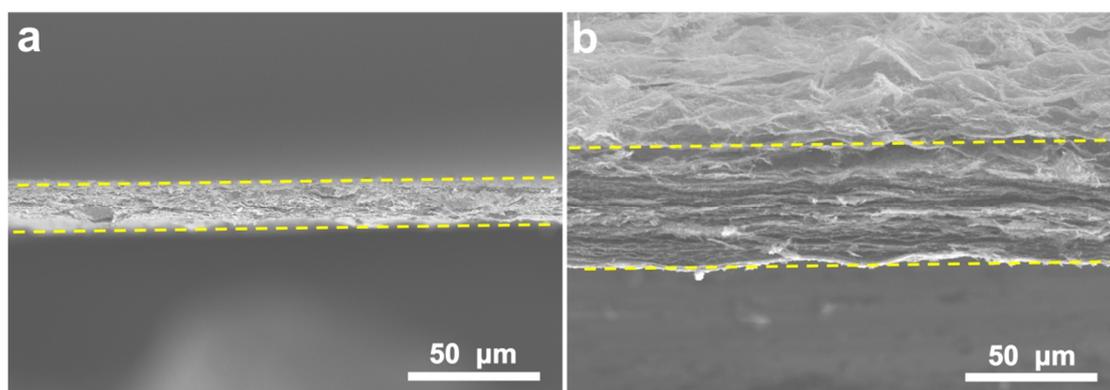
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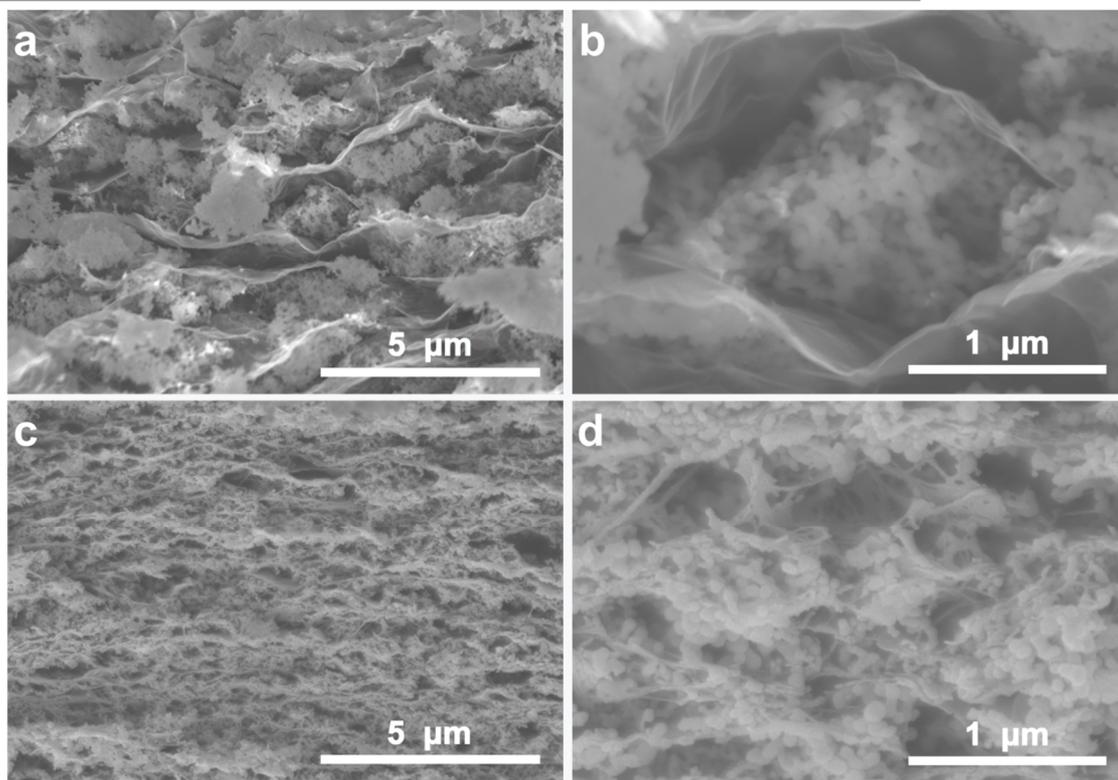
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**Figure S1.** Mechanical properties test of SSPBG electrode



**Figure S2.** Cross-sectional SEM images of SSPBG with controllable thickness.



**Figure S3.** Cross-sectional SEM images of (a-b) SSG and (c-d) SSPB.

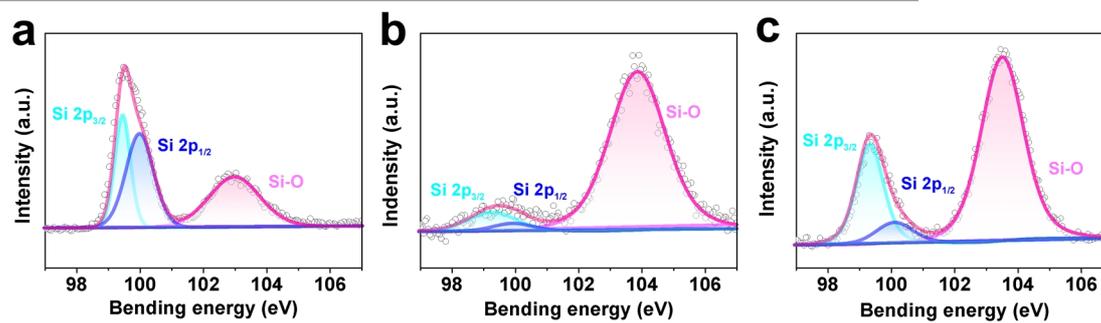
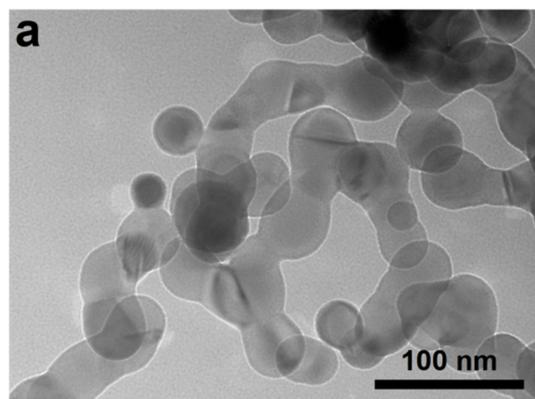


Figure S4. Si 2p spectrum of (a) Si NPs, (b) SSPB and (c) SSG.



**Figure S5.** TEM image of Si NPs.

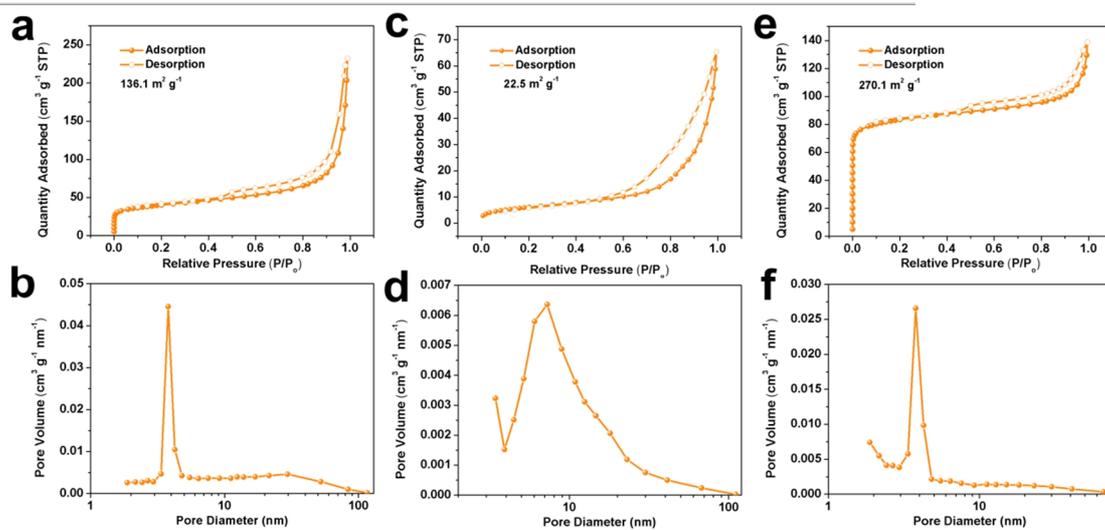
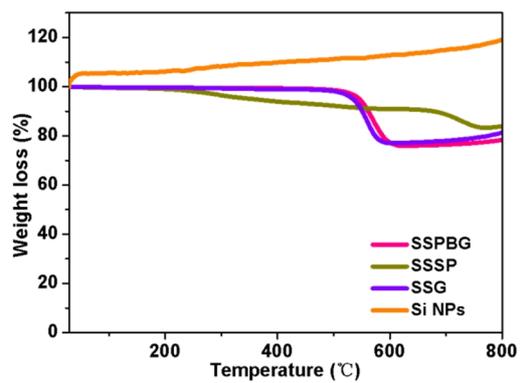
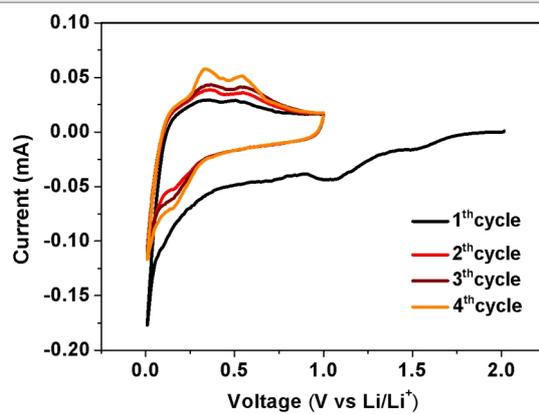


Figure S6.  $N_2$  adsorption-desorption isotherms of (a) SSPBG, (c) SSG and (e) SSPB. Pore size distribution of (b) SSPBG, (d) SSG and (f) SSPB.



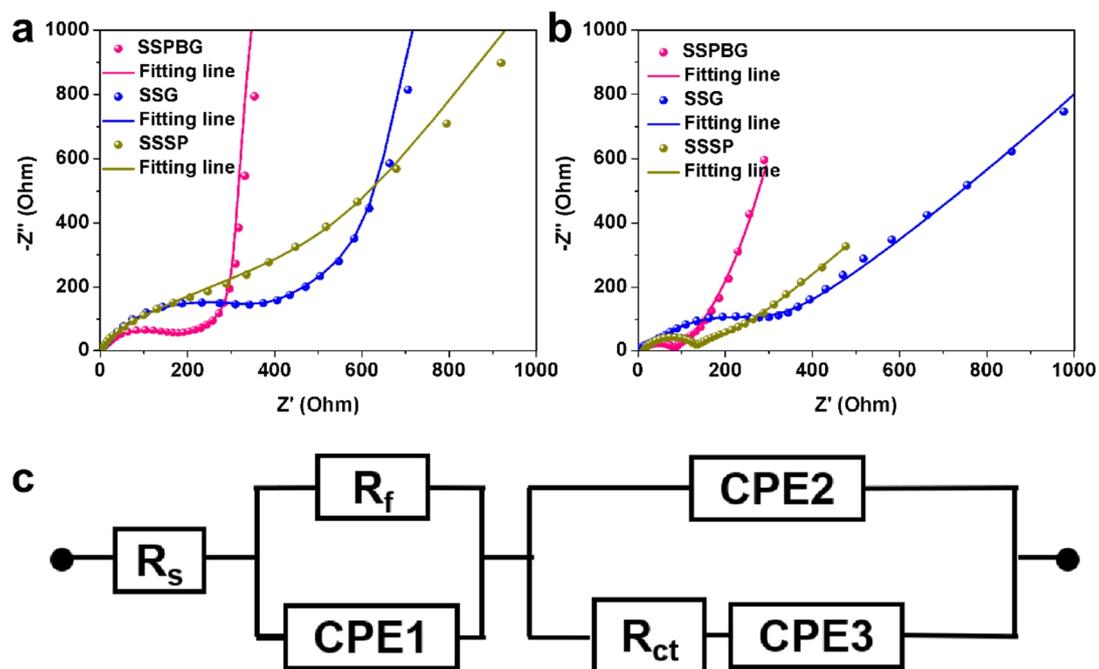
**Figure S7.** TGA curves of SSPBG and control electrodes obtained under 800 °C for 2 h at a heating rate of 10 °C/min in air atmosphere.



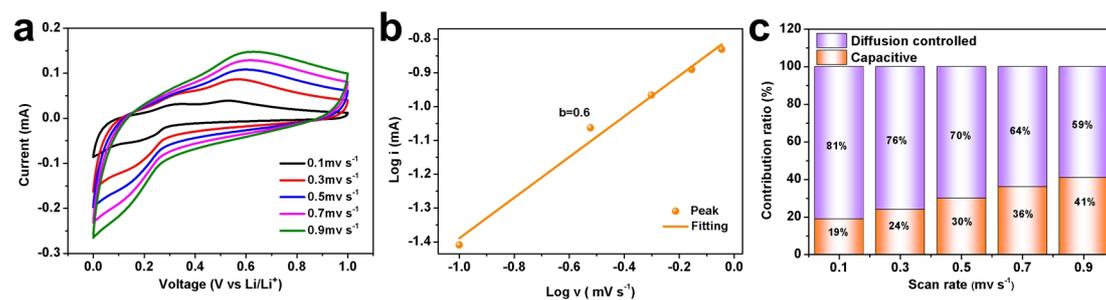
**Figure S8.** Typical CV curve of SSPBG.



**Figure S9.** Photograph showing electrical conductivity of SSPBG (88.2  $\Omega$ ) and SSG (170.3  $\Omega$ ).



**Figure S10.** Nyquist plots of SSPBG, SSG and SSSP obtained from electrochemical impedance spectroscopy (EIS) measurement for (a) fresh electrodes and (b) electrodes after 100 cycles. (c) equivalent circuit.



**Figure S11.** (a) CV curves of SSPBG electrodes at different scanning rates. (b) Relation between log(peak current) and log(scanning rate). (c) Contribution ratio of capacitive and diffusion-controlled charge storage at different scanning rates.

**Table S1.** Weight Distribution of Si in SSPBG, SSG and SSSP

	(%)	(630 °C)	(770 °C)	X <sub>Si</sub> (%)
SSPBG	W <sub>Si</sub>	113	/	67
	W <sub>C</sub>	0	/	33
	W <sub>SSBG</sub>	76	/	/
SSG	W <sub>Si</sub>	113	/	68
	W <sub>C</sub>	0	/	32
	W <sub>SG</sub>	77.2	/	/
SSSP	W <sub>Si</sub>	/	117	66
	W <sub>C</sub>	/	0	34
	W <sub>SSSP</sub>	/	78	/

**Table S2.** Fitted Electrochemical Impedance Value.

Sample	Rs/Ohm	Rf/Ohm	Rct/Ohm
SSPBG	2.119	122.3	381
SSG	1.67	1135	459
SSSP	2.236	211.9	1051
SSPBG after cycling	2.557	62.7	74.4
SSG after cycling	2.896	45.95	289.3
SSSP after cycling	12.67	413.3	181.8

**Table S3.** Literature on Self-supporting Si-based Anodes for Lithium-ion Batteries

Sample	Discharge capacity (mAh g <sup>-1</sup> )	Current density (mA g <sup>-1</sup> )	After n <sup>th</sup> cycle	Ref.
CNTs/Si	1000	1000	50	[1]
CNTs/Si NPs	1510	840	100	[2]
PAN/Si/Ni	910	1000	100	[3]
Si NPs/porous carbon nanofibers	870	100	100	[4]
Si NPs/carbon nanofibers	869	400	480	[5]
Si NPs/graphene	1390	2000	200	[6]
Graphene/CNTs/Si	808	420	60	[7]
Si NPs/porous carbon/carbon black/CNTs	765	200	100	[8]
Si@SiO <sub>2</sub> /bacterial cellulose/graphene	901	2000	500	This work

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