SUPPORTING INFORMATION

Fabricating Multifunctional Nanoparticle Membranes by a Fast Layer-by-Layer Langmuir-Blodgett Process: Application in Lithium-sulfur Batteries

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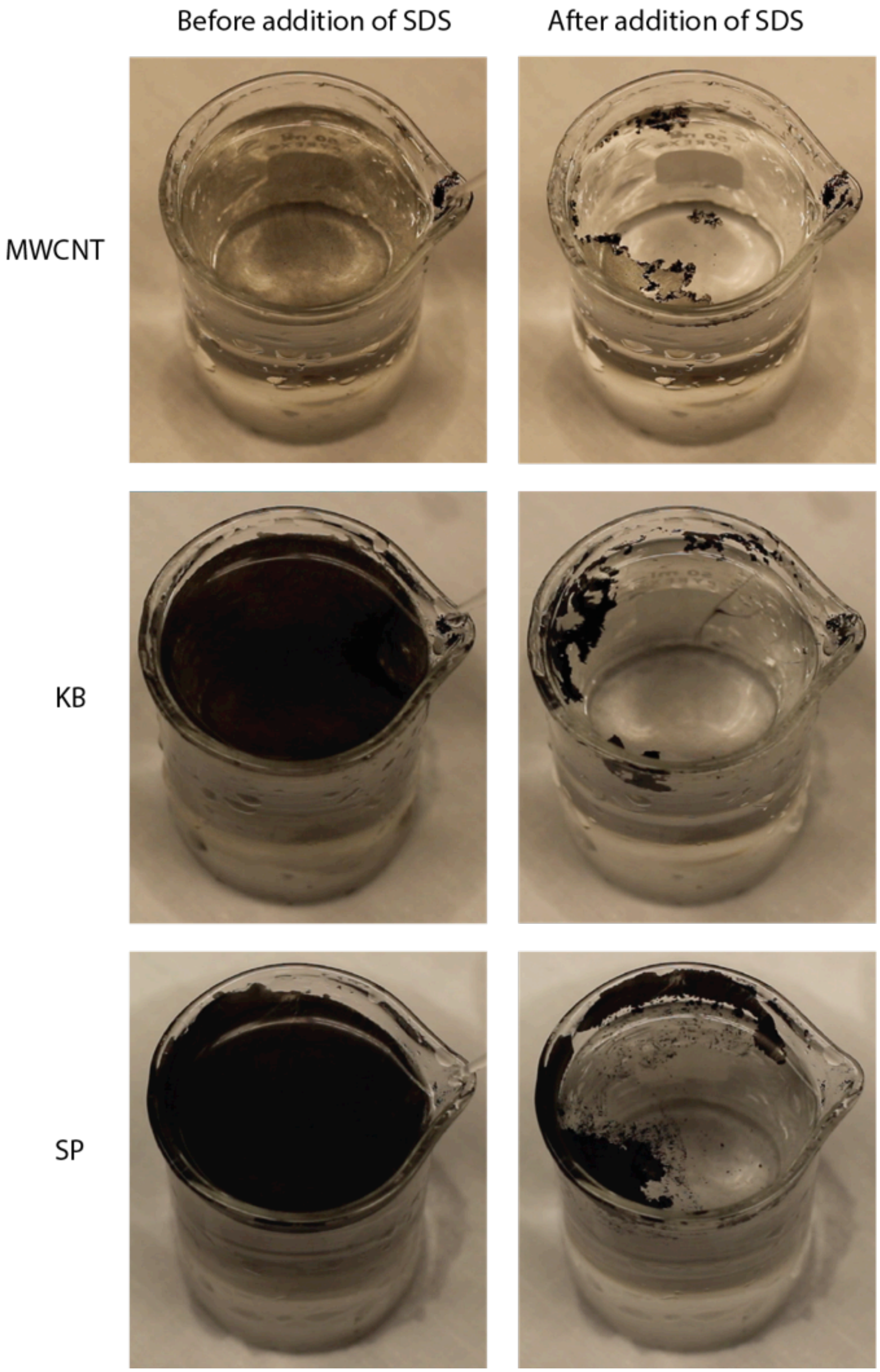

Fig. S1: Collapsing self-assembled LB film in the presence of SDS surfactant. The self-assembled MWCNT, KB, and SP films are formed at the surface of water using LBS coating method, and images of the films before and after adding a one-drop of SDS surfactant at the neck of the beaker are shown.

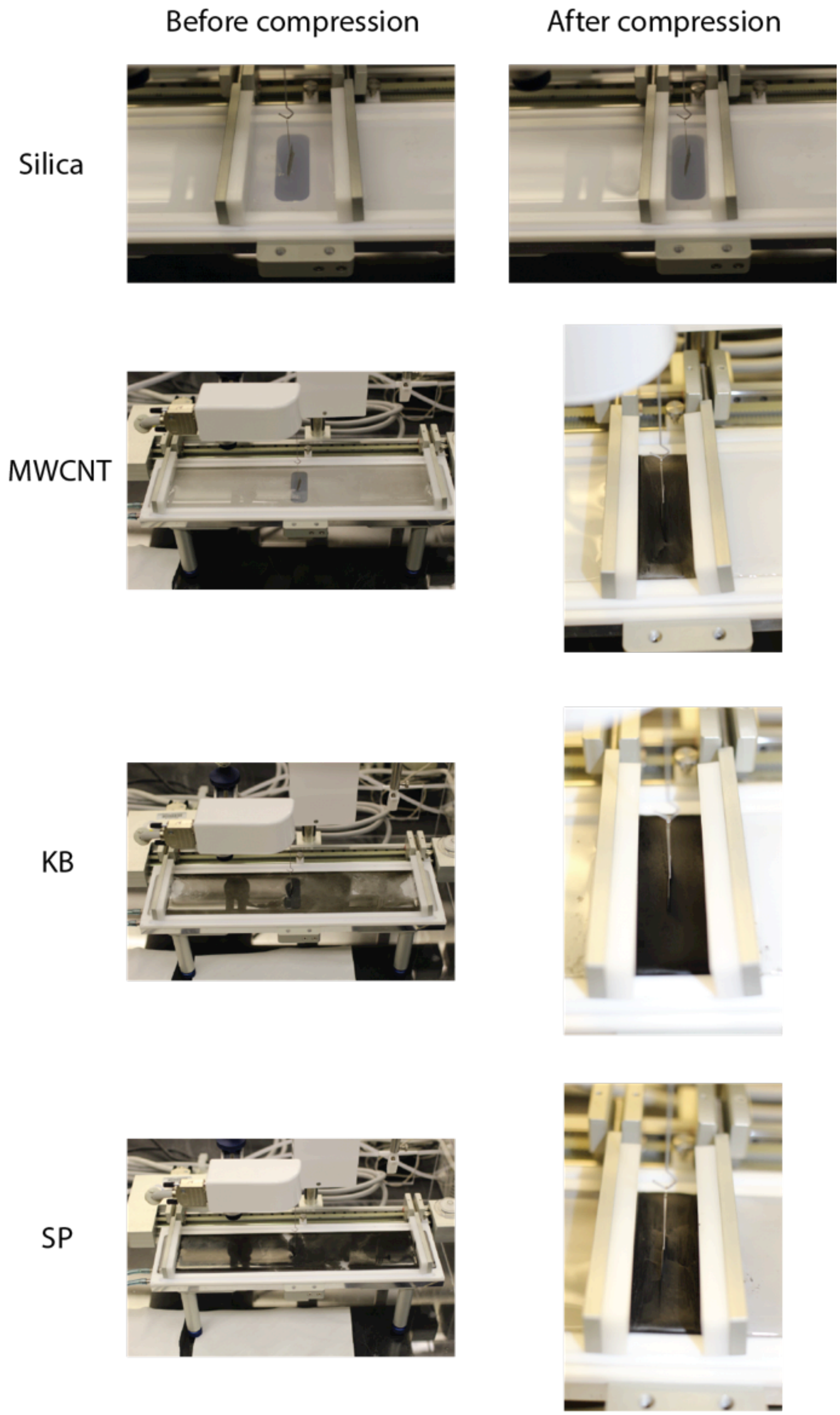

Fig. S2: Langmuir-Blodgett trough experiment for surface pressure measurements. Images of compressing silica nanospheres, MWCNT, KB, and SP are shown. Note that no materials are lost during the compression of the LB films, and folding of the films is observed instead of particles sinking.

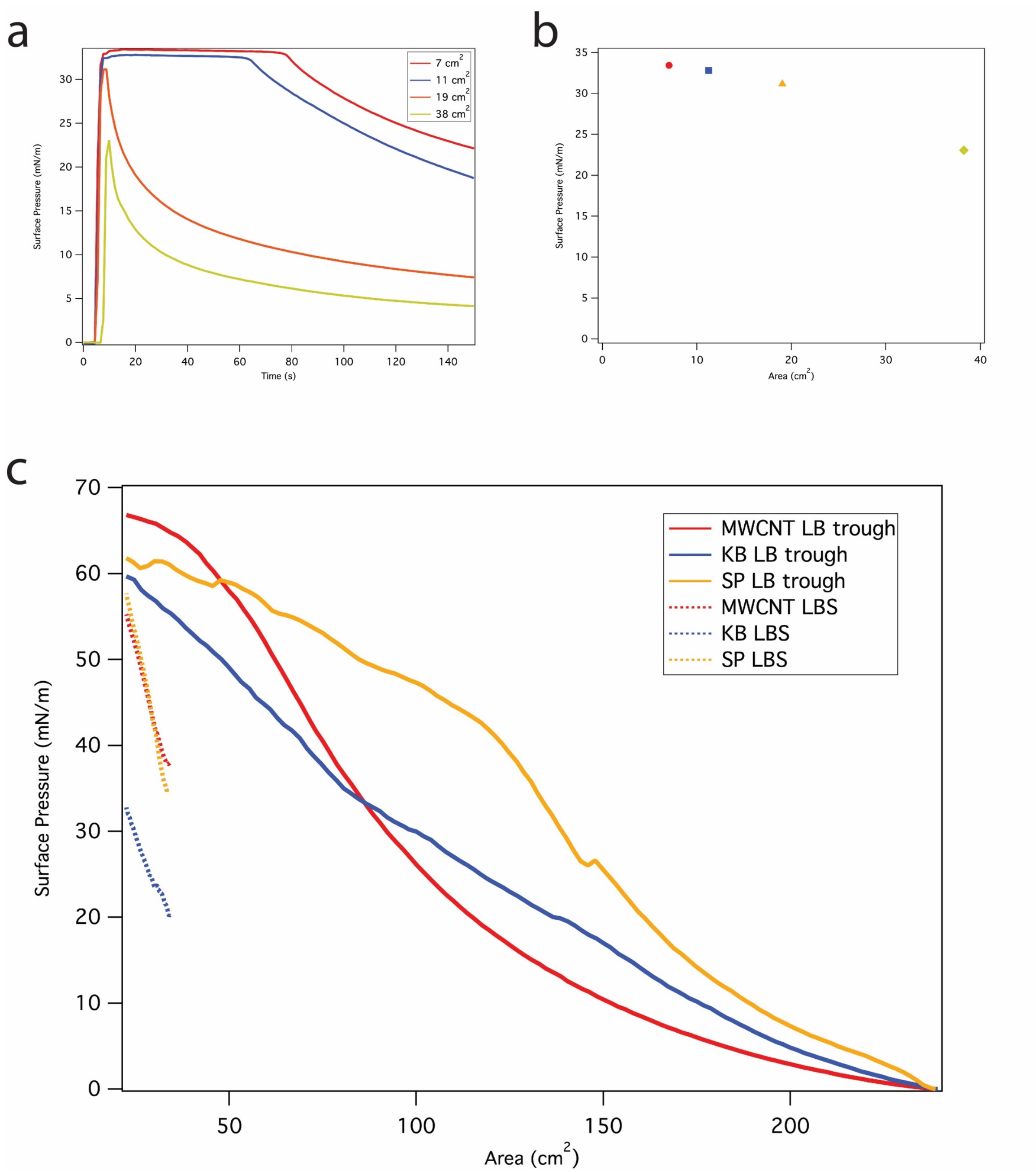

Fig. S3: SDS surfactant and carbon particles surface pressure profiles. (a) Surface pressures of SDS surfactant versus time for the surface area of 7 cm^2, 11 cm^2, 19 cm^2, and 38 cm^2. (b) Maximum surface pressures exerted by the surfactant for the surface area of 7 cm^2, 11 cm^2, 19 cm^2, and 38 cm^2. (c) MWCNT, KB, and SP surface pressure profiles obtained from conventional LB trough and LBS methods.

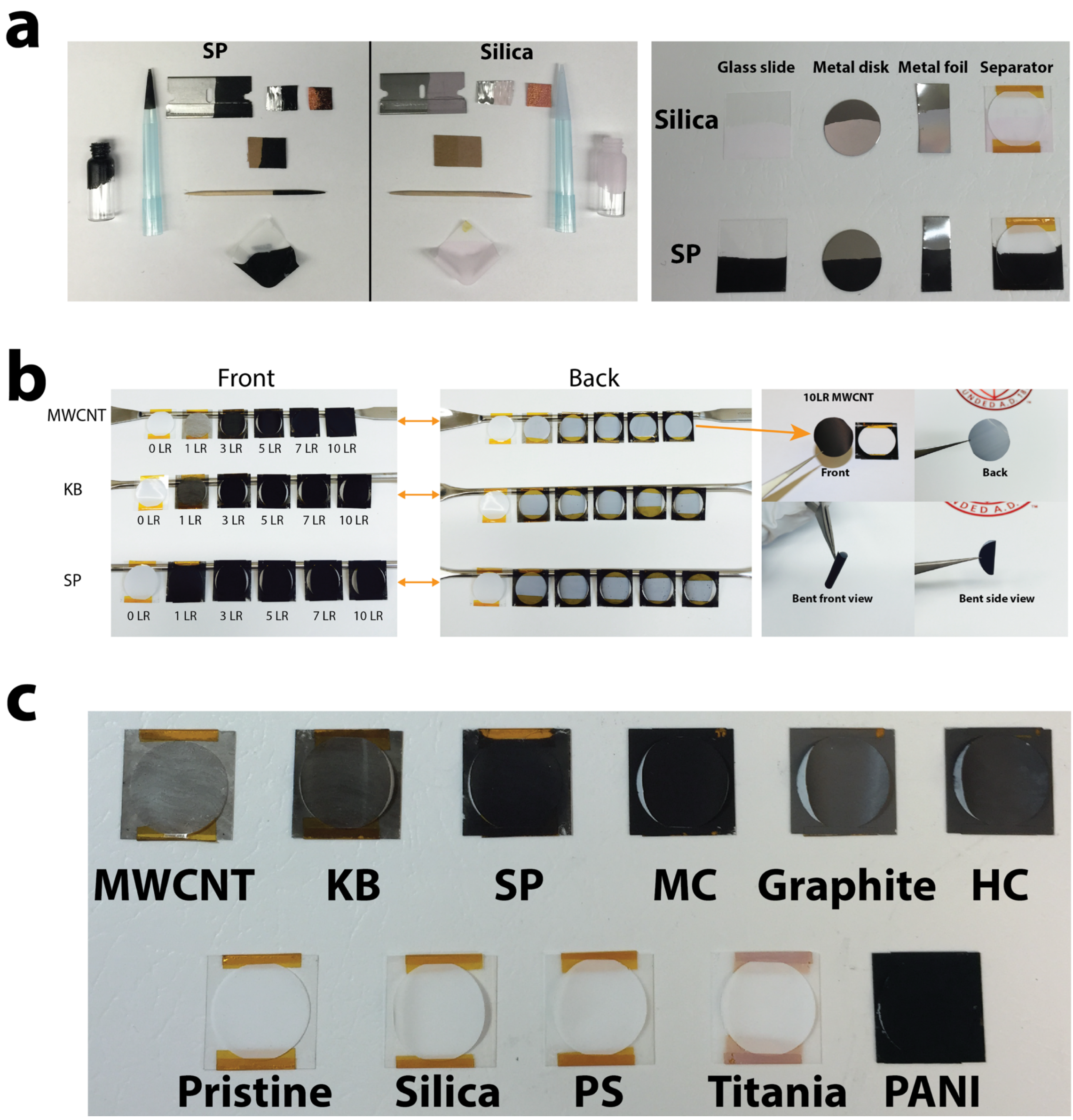

Fig. S4: Demonstrating versatility of the LB coating methods. (a) Images of fluorescent 1μm silica nanosphere and SP carbon coatings on various nonuniform (left) and uniform (right) substrates using LBS coating method. Note that fluorescent silicon and SP carbon are used to enhance the visibility of the coatings on the substrates. (b) Demonstration of single-sided and layer-by-layer separator coating (left) and mechanical strength of the single component coating (right). (c) Images of single coating layer of various materials on the separator – Multi-walled carbon nanotube (MWCNT), Ketjen Black carbon (KB), Super P carbon (SP), Microporous carbon (MC), Graphite, Hard carbon (HC), 350 nm silica nanospheres, 1μm polystyrene spheres (PS), titania nanopowder, and polyaniline (PANI).

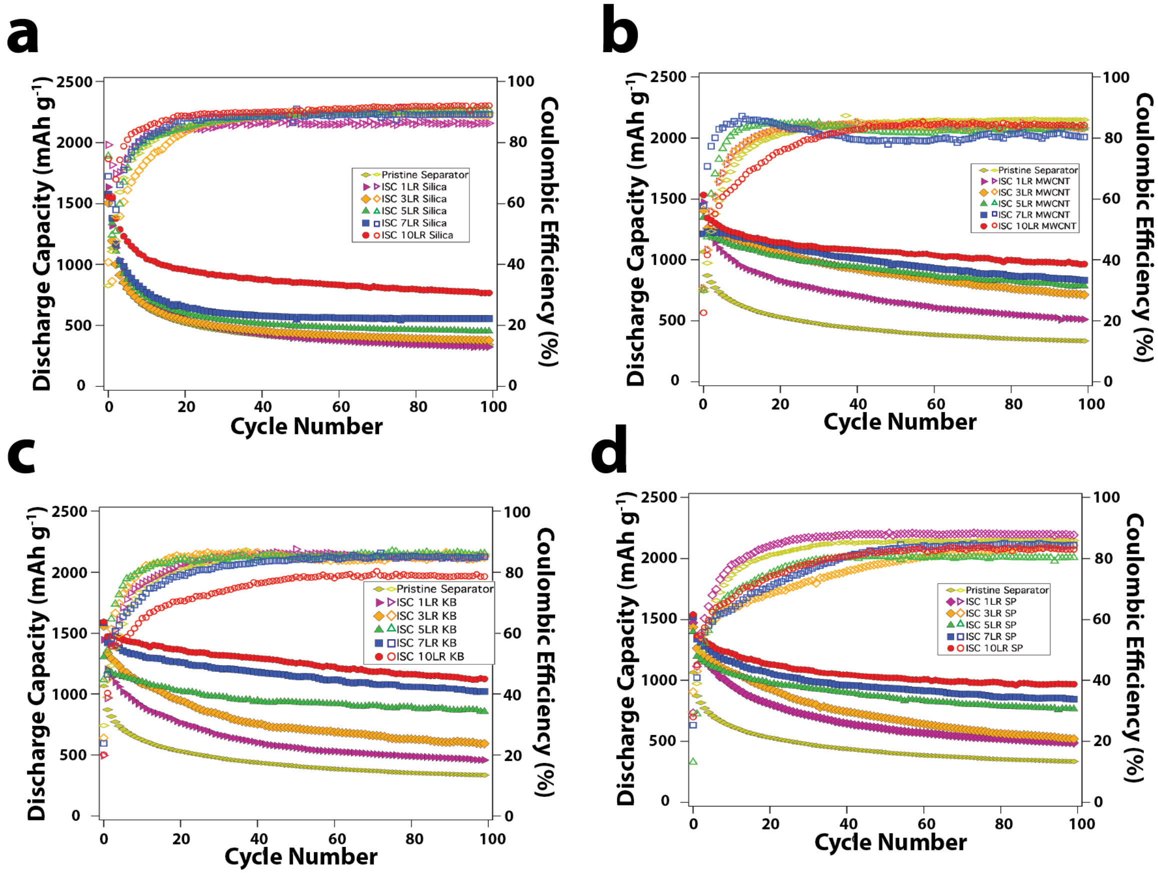

Fig. S5: Electrochemical performances of KB and SP carbon coated separators with ISC. Electrochemical performance of zero to ten coating layers of (a) Silica nanospheres, (b) MWCNT, (c) KB and (d) SP coated separators Li-S cells at 0.2C for the silica and 0.5 C for the carbon coatings.

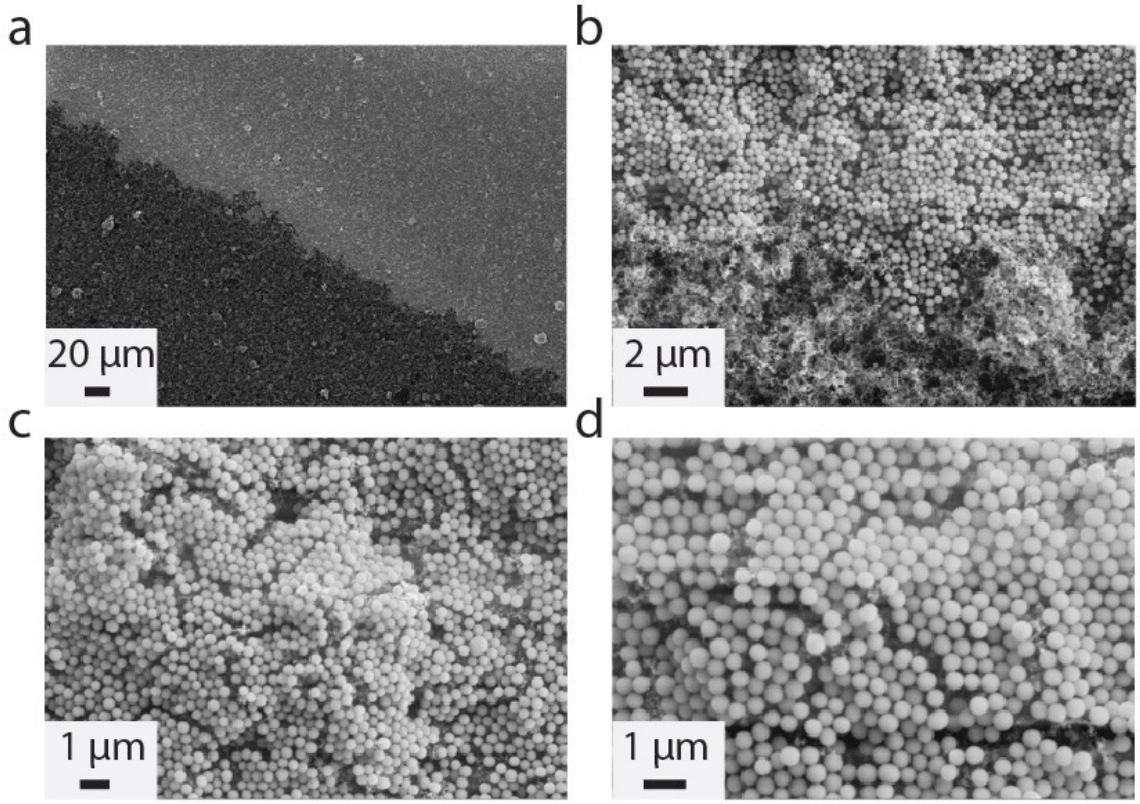

Fig. S6: SEM images of a monolayer of silica nanospheres coated on MWCNT-SP layer. (a) Silica-SP layer boundary region, (b) wide-view, and (c) close-view. A monolayer of silica coating on top of the SP-MWCNT layer (c, d).

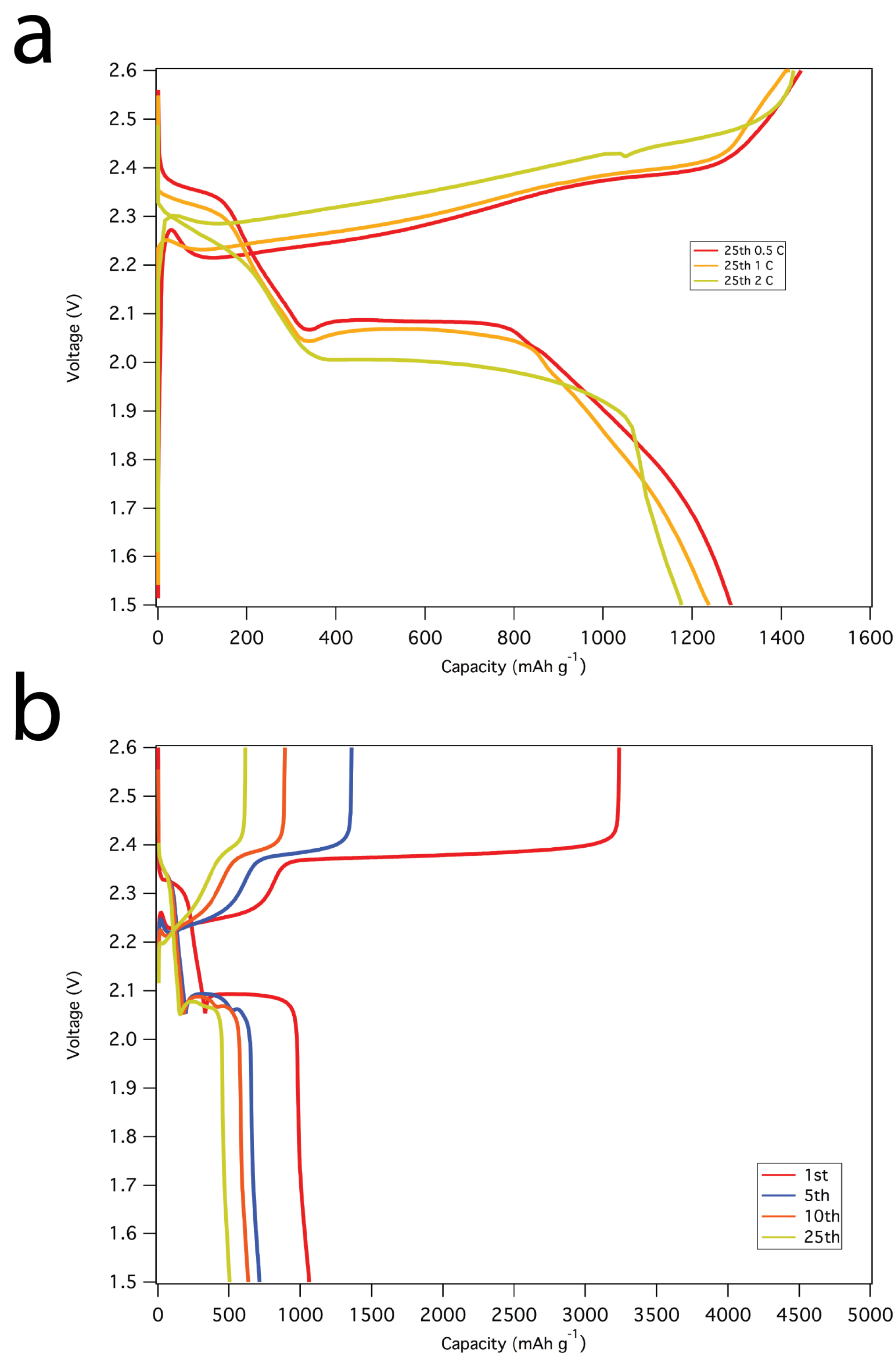

Fig. S7: Voltage profiles of Li-S cells with/without clip coated separator and ISC. (a) Discharge-charge voltage profiles of pristine Li anode Li-S cell with the clip coated separator and ISC at 25th cycle for various C rates. (b) Discharge-charge voltage profiles. of pristine Li anode Li-S cell with the pristine separator and ISC at 0.5 C for various cycles.

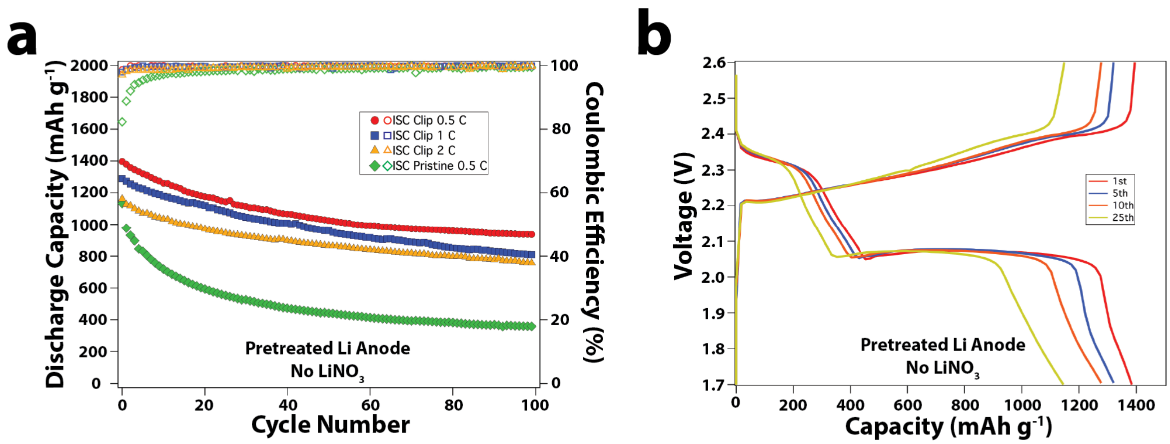

Fig. S8: Electrochemical performances of clip coated separators with pretreated Li anode and ISC. (a) Cycling performance of the pretreated Li anode Li-S cell with/without the clip coated separator and with ISC at three different C rates. (b) Discharge-charge voltage profiles of the pretreated Li anode Li-S cell with the clip coated separator and with ISC at 0.5 C. Note that $LiNO_3$ is not present in the electrolyte.

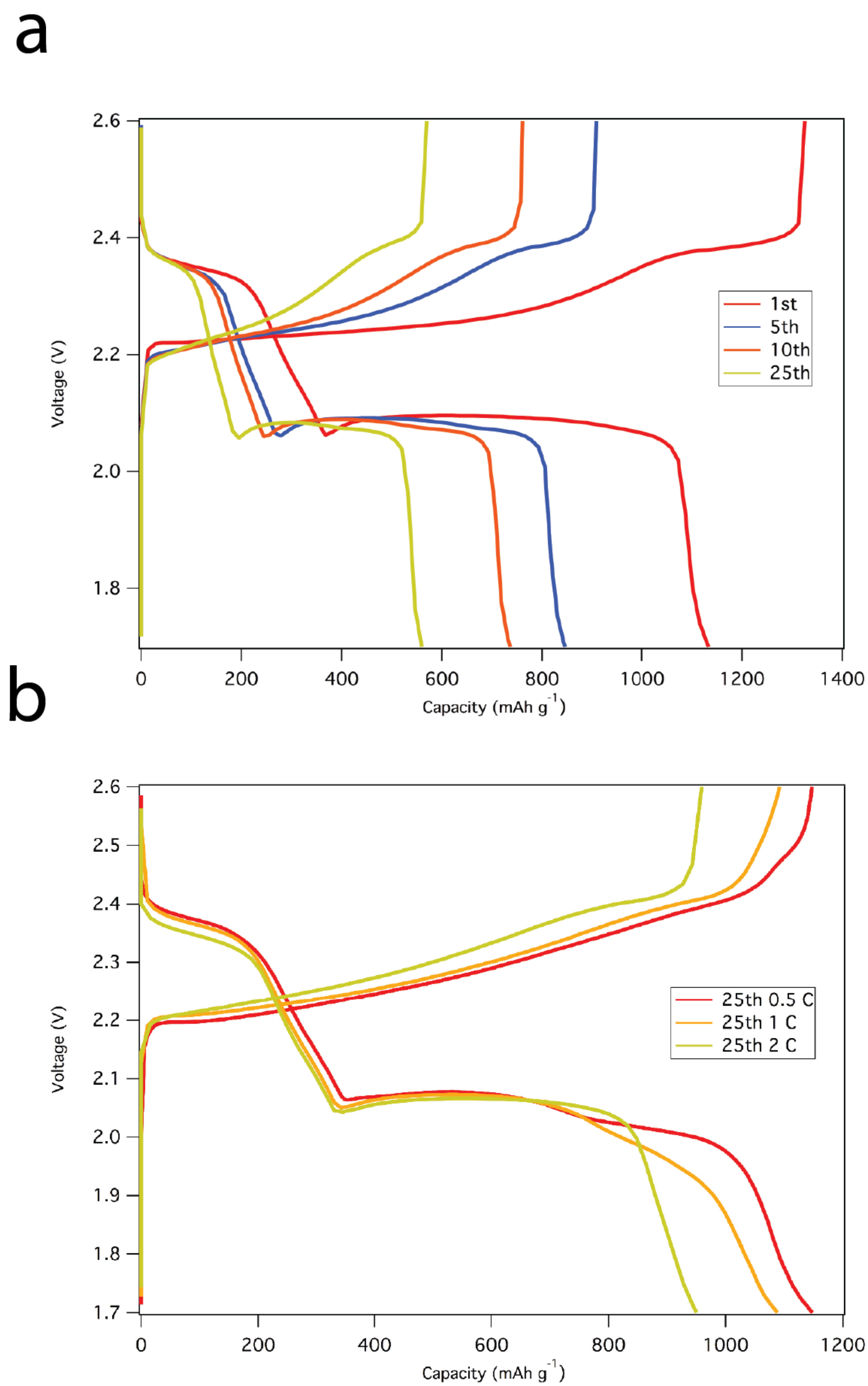

Fig. S9: Discharge-charge voltage profiles of clip coated separator Li-S cell with the pretreated Li anode and with ISC for (a) various cycles at 0.5C and (b) various C rates at 25th cycle.

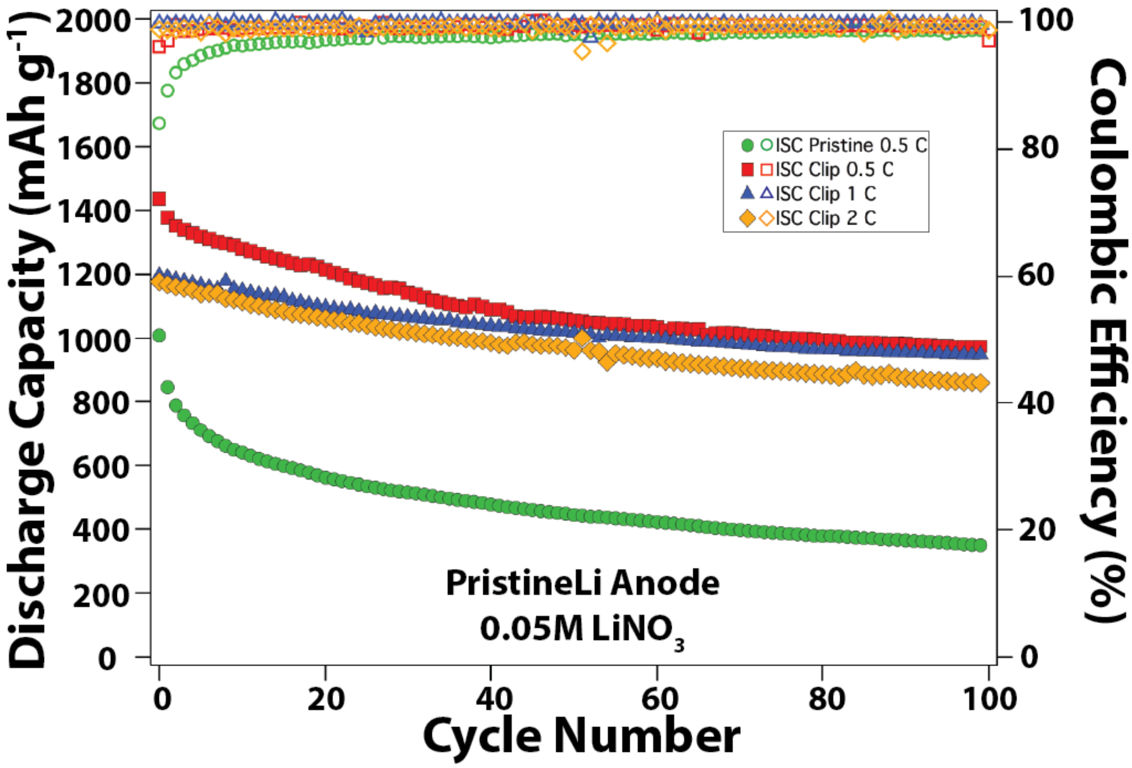

Fig. S10: Electrochemical performances of clip coated separators with pristine Li anode, ISC, and $LiNO_3$ in the electrolyte. Cycling performance of pristine Li anode of the Li-S cell with/without the clip coated separator and with ISC at three different C rates with 0.05M LiNO3 in the electrolyte.

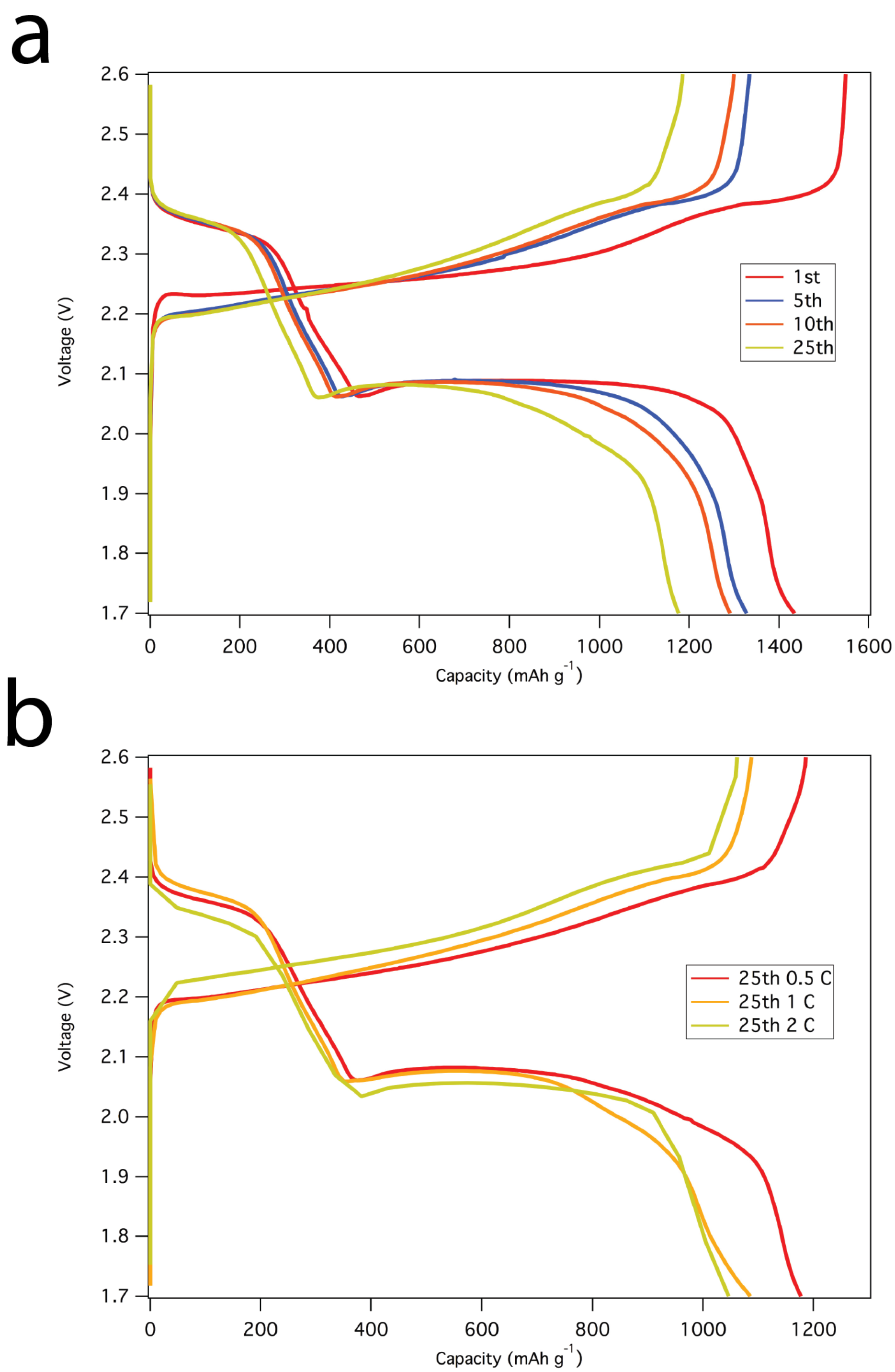

Fig. S11: Cycling performance and voltage profiles of Li-S cells with clip coated separator, ISC, and 0.05M $LiNO_3$ in the electrolyte. (a) Discharge-charge voltage profiles of clip coated separator Li-S cell with 0.05M $LiNO_3$ added in the electrolyte and with ISC for various cycles at 0.5 C. (b) Discharge-charge voltage profiles of the clip coated separator Li-S cell with 0.05M $LiNO_3$ in the electrolyte and with ISC at 25th cycle at various C rates.

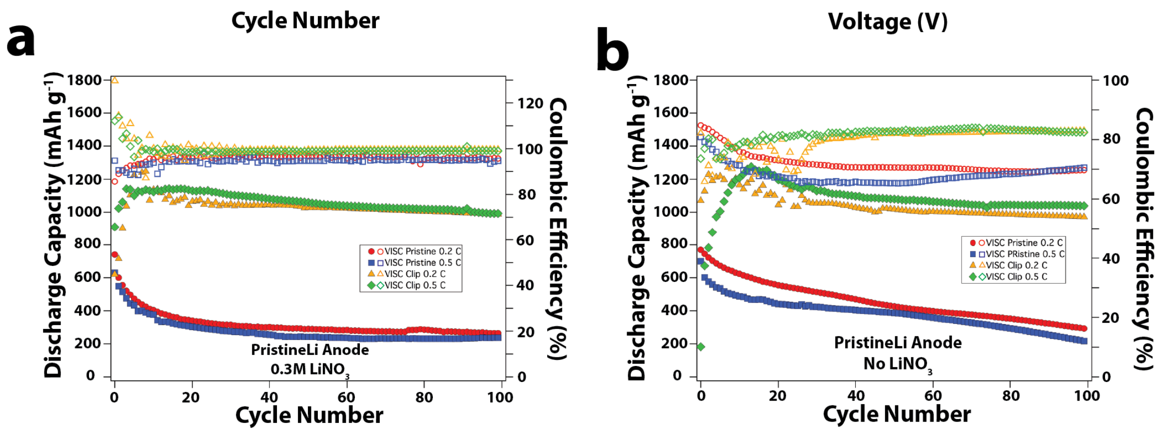

Fig. S12: Electrochemical performances of clip coated separators with pristine Li anode, VISC, and with/without $LiNO_3$ in the electrolyte. (a) Cycling performance of the pristine Li anode Li-S cells with/without the clip coated separator, with VISC, and 0.3M LiNO3 in the electrolyte at two different C rates. (b) Cycling performance of the pristine Li anode Li-S cells with/without the clip coated separator and with VISC at two different C rates without LiNO3 in the electrolyte.

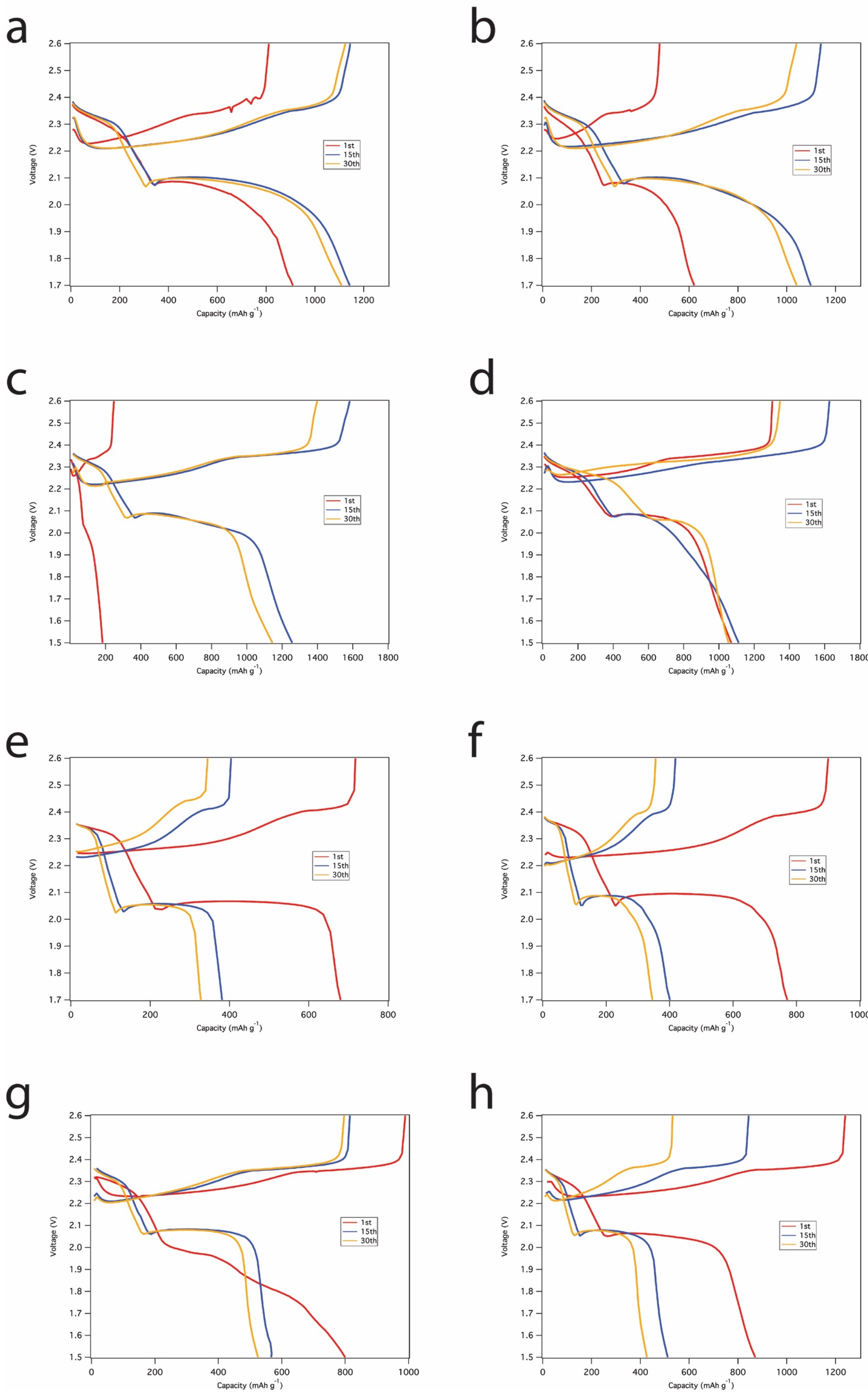
a
b
c
d
e
f
g
h
Voltage (V)
Capacity (mAh g^{-1})
1st
15th
30th

Fig. S13: Voltage profiles of Li-S cells with clip coated separator and VISC. (a) Discharge-charge voltage profiles of clip coated separator Li-S cell with VISC and 0.3M $LiNO_3$ added in the electrolyte for various cycles at 0.5 C. (b) Discharge-charge voltage profiles of clip coated separator Li-S cell with VISC and 0.3M $LiNO_3$ added in the electrolyte for various cycles at 0.2 C. (c) Discharge-charge voltage profiles of clip coated separator Li-S cell with VISC for various cycles at 0.5 C. (d) Discharge-charge voltage profiles of clip coated separator Li-S cell with VISC for various cycles at 0.2 C. (e) Discharge-charge voltage profiles of pristine separator Li-S cell with VISC and 0.3M $LiNO_3$ added in the electrolyte for various cycles at 0.5 C. (f) Discharge-charge voltage profiles of pristine separator Li-S cell with VISC and 0.3M $LiNO_3$ added in the electrolyte for various cycles at 0.2 C. (g) Discharge-charge voltage profiles of pristine separator Li-S cell with VISC for various cycles at 0.5 C. (h) Discharge-charge voltage profiles of pristine separator Li-S cell with VISC for various cycles at 0.2 C.

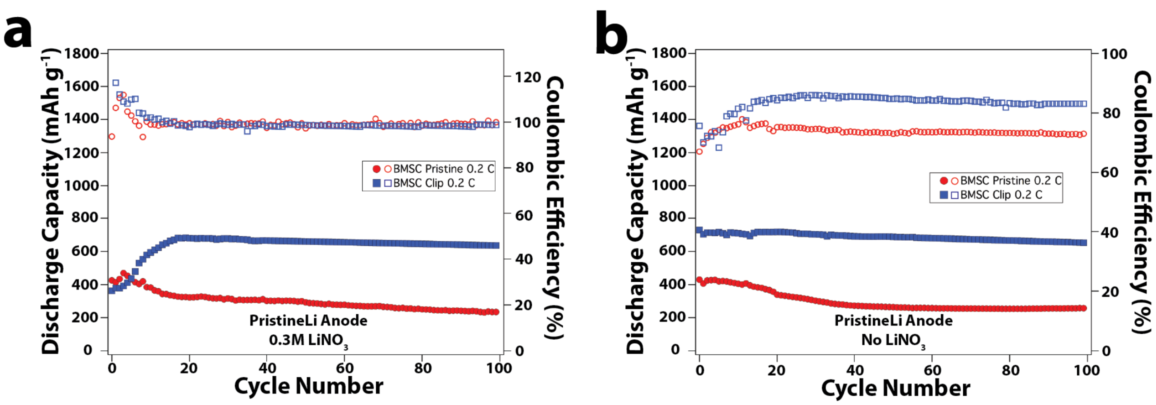

Fig. S14: Electrochemical performances of clip coated separators with pristine Li anode, BMSC, and with/without $LiNO_3$ in the electrolyte. (a) Cycling performance of the pristine Li anode Li-S cells with/without the clip coated separator, with BMSC, and 0.3M LiNO3 in the electrolyte at 0.2 C. (b) Cycling performance of the pristine Li anode Li-S cells with/without the clip coated separator and with BMSC at 0.2 C without LiNO3 in the electrolyte.

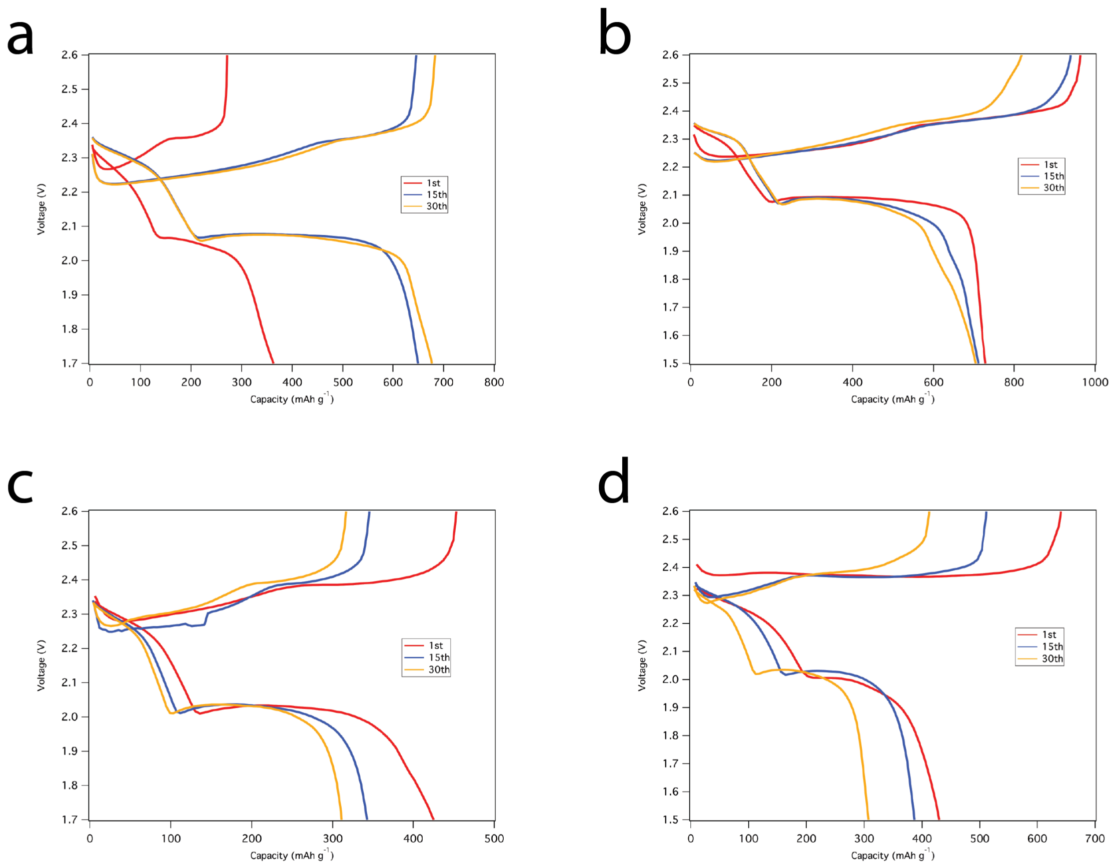

Fig. S15: Voltage profiles of Li-S cells with clip coated separator and BMSC. (a) Discharge-charge voltage profiles of clip coated separator Li-S cell with BMSC and 0.3M $LiNO_3$ added in the electrolyte for various cycles at 0.2 C. (b) Discharge-charge voltage profiles of clip coated separator Li-S cell with BMSC for various cycles at 0.2 C. (c) Discharge-charge voltage profiles of pristine separator Li-S cell with BMSC and 0.3M $LiNO_3$ added in the electrolyte for various cycles at 0.2 C. (d) Discharge-charge voltage profiles of pristine separator Li-S cell with BMSC for various cycles at 0.2 C.

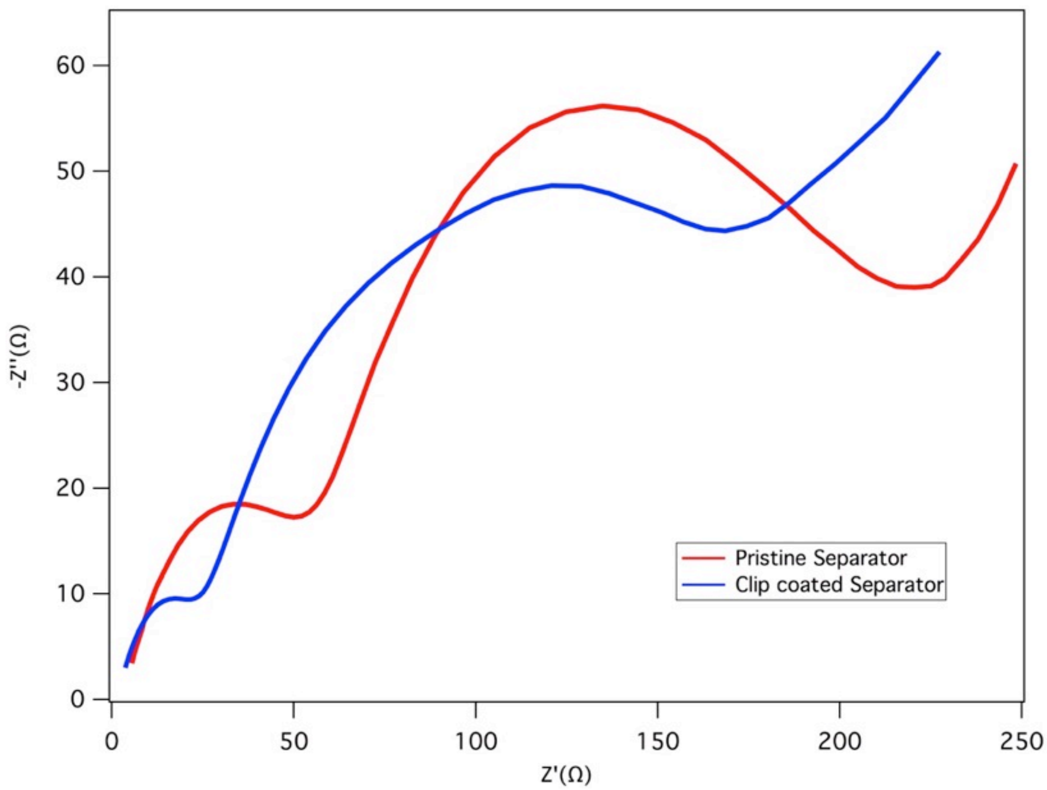

Fig. S16: AC impedance spectroscopy analysis of the clip coated separator and pristine separator Li-S cells with ISC.

Separator	Anode	Cathode	Electrolyte	Cycle Number	C rate	Capacity (mAh g^{-1})	CE (%)
Clip	Li	ISC	1M LiTFSI DME:DOL	100	0.5	1250	90
Clip	Li	ISC	1M LiTFSI DME:DOL	250	0.5	1180	89
Clip	Li	ISC	1M LiTFSI DME:DOL	100	1	1080	91
Clip	Li	ISC	1M LiTFSI DME:DOL	100	2	960	91
Pristine	Li	ISC	1M LiTFSI DME:DOL	100	0.5	340	85
Clip	Pretreated Li	ISC	1M LiTFSI DME:DOL	100	0.5	1000	99.9
Clip	Pretreated Li	ISC	1M LiTFSI DME:DOL	100	1	830	99.9
Clip	Pretreated Li	ISC	1M LiTFSI DME:DOL	100	2	770	99.8
Pristine	Pretreated Li	ISC	1M LiTFSI DME:DOL	100	0.5	350	99.1
Clip	Li	ISC	1M LiTFSI DME:DOL 0.05M $LiNO_3$	100	0.5	1000	98.5
Clip	Li	ISC	1M LiTFSI DME:DOL 0.05M $LiNO_3$	100	1	980	99.1
Clip	Li	ISC	1M LiTFSI DME:DOL 0.05M $LiNO_3$	100	2	870	99.3
Pristine	Li	ISC	1M LiTFSI DME:DOL 0.05M $LiNO_3$	100	0.5	348	98.3
Clip	Li	VISC	1M LiTFSI DME:DOL	100	0.2	1000	82
Pristine	Li	VISC	1M LiTFSI DME:DOL	100	0.2	290	69
Clip	Li	VISC	1M LiTFSI DME:DOL	100	0.5	1060	83
Pristine	Li	VISC	1M LiTFSI DME:DOL	100	0.5	220	70
Clip	Li	VISC	1M LiTFSI DME:DOL	250	0.5	1010	82
Clip	Li	VISC	1M LiTFSI DME:DOL 0.3M $LiNO_3$	100	0.2	1105	99.5
Pristine	Li	VISC	1M LiTFSI DME:DOL 0.3M $LiNO_3$	100	0.2	300	95.6
Clip	Li	VISC	1M LiTFSI DME:DOL 0.3M $LiNO_3$	100	0.5	1100	99.6
Pristine	Li	VISC	1M LiTFSI DME:DOL 0.3M $LiNO_3$	100	0.5	270	94.9

Clip	**Li**	**BMSC**	**1M LiTFSI DME:DOL**	**100**	**0.2**	**660**	**85**
Pristine	**Li**	**BMSC**	**1M LiTFSI DME:DOL**	**100**	**0.2**	**260**	**73**
Clip	**Li**	**BMSC**	**1M LiTFSI DME:DOL 0.3M $LiNO_3$**	**100**	**0.2**	**640**	**98.4**
Pristine	**Li**	**BMSC**	**1M LiTFSI DME:DOL 0.3M $LiNO_3$**	**100**	**0.2**	**230**	**99.3**
MWCNT (10LR)	**Li**	**ISC**	**1M LiTFSI DME:DOL**	**100**	**0.5**	**960**	**84**
MWCNT (7LR)	**Li**	**ISC**	**1M LiTFSI DME:DOL**	**100**	**0.5**	**830**	**81**
MWCNT (5LR)	**Li**	**ISC**	**1M LiTFSI DME:DOL**	**100**	**0.5**	**790**	**83.2**
MWCNT (3LR)	**Li**	**ISC**	**1M LiTFSI DME:DOL**	**100**	**0.5**	**710**	**83**
MWCNT (1LR)	**Li**	**ISC**	**1M LiTFSI DME:DOL**	**100**	**0.5**	**330**	**83**
KB (10LR)	**Li**	**ISC**	**1M LiTFSI DME:DOL**	**100**	**0.5**	**1130**	**80**
KB (7LR)	**Li**	**ISC**	**1M LiTFSI DME:DOL**	**100**	**0.5**	**1030**	**85**
KB (5LR)	**Li**	**ISC**	**1M LiTFSI DME:DOL**	**100**	**0.5**	**850**	**87**
KB (3LR)	**Li**	**ISC**	**1M LiTFSI DME:DOL**	**100**	**0.5**	**590**	**85**
KB (1LR)	**Li**	**ISC**	**1M LiTFSI DME:DOL**	**100**	**0.5**	**460**	**86**
SP (10LR)	**Li**	**ISC**	**1M LiTFSI DME:DOL**	**100**	**0.5**	**960**	**83**
SP (7LR)	**Li**	**ISC**	**1M LiTFSI DME:DOL**	**100**	**0.5**	**830**	**85**
SP (5LR)	**Li**	**ISC**	**1M LiTFSI DME:DOL**	**100**	**0.5**	**760**	**81**
SP (3LR)	**Li**	**ISC**	**1M LiTFSI DME:DOL**	**100**	**0.5**	**510**	**85**
SP (1LR)	**Li**	**ISC**	**1M LiTFSI DME:DOL**	**100**	**0.5**	**480**	**88**
Silica (10LR)	**Li**	**ISC**	**1M LiTFSI DME:DOL**	**100**	**0.2**	**780**	**93**
Silica (7LR)	**Li**	**ISC**	**1M LiTFSI DME:DOL**	**100**	**0.2**	**550**	**91**
Silica (5LR)	**Li**	**ISC**	**1M LiTFSI DME:DOL**	**100**	**0.2**	**480**	**89**
Silica (3LR)	**Li**	**ISC**	**1M LiTFSI DME:DOL**	**100**	**0.2**	**380**	**89**
Silica (1LR)	**Li**	**ISC**	**1M LiTFSI DME:DOL**	**100**	**0.2**	**330**	**88**

Table S1: LiS electrochemical performance summary table.