#### Supporting Information

#### High-content sulfur uniform embedded in mesoporous carbon: A new electrodeposition synthesis and outstanding lithium-sulfur battery cathode

Liyuan Zhang,<sup>a,b</sup> Hui Huang,<sup>a</sup> Yang Xia,<sup>a</sup> Chu Liang,<sup>a</sup> Wenkui Zhang,<sup>\*a</sup> Jianmin Luo,<sup>a</sup> Yongping Gan,<sup>a</sup> Jun Zhang,<sup>a</sup> Xinyong Tao,<sup>a</sup> Hong Jin Fan<sup>\*b</sup>

<sup>a</sup> College of Materials Science and Engineering, Zhejiang University of Technology, Hangzhou 310014, China. E-mail: msechem@zjut.edu.cn.

<sup>b</sup> School of Physical and Mathematical Sciences, Nanyang Technological University, Singapore 637371, Singapore. E-mail: fanhj@ntu.edu.sg



Figure S1. Experimental device of sulfur electrodeposited into mesoporous carbon.



Figure S2. Nitrogen adsorption-desorption isotherms of MC.



**Figure S3.** Current *versus* time curves of electrodeposition process controlled by constant potential of 1.0V (the MCSC-1.0 electrode *vs.* SCE)



**Figure S4.** (a-d) XPS C 1s spectrums of MCSC-1.4, MCSC-0.6, MC-S and MC. (e-g) S 2p spectrums of MCSC-1.4, MCSC-0.6 and MC-S.



**Figure S5.** TEM images of (a) MCSC-0.6 and (b) MCSC-1.4 with EDS carbon and sulfur mapping of the region.



**Figure S6.** CV curves of (a) MCSC-1.4, (b) MCSC-0.6, (c) MC-S and (d) pure Sulfur cathodes in the potential range of 1.9-2.6 V at 0.1 mV s<sup>-1</sup>.



**Figure S7.** (a) CV curves of MCSC-1.0 range in the potential range of 1.9-2.35 V at 0.1 mV s<sup>-1</sup>. (b) Long-term cycling performance of MCSC-1.0 cathode at 0.5 A  $g^{-1}$  in a potential range of 1.9-2.35 V.



Figure S8. Initial charge-discharge profiles of MCSC, MC-S and pure sulfur cathodes at 0.1 A  $g^{-1}$ 

Table S1. Characteristics	of various mesoporous	carbon-sulfur	composites	obtained
from different loading met	thods.			

_	Loading method	host material	Characteristic	Sulfur percentage	Capacity (Second cycle)	Cycling Stability	Refe renc e
	Electrodeposition	Mesoporous carbon	Powder carbon can be directly used. Sulfur can deposited in the small porous with self- limiting mechanism. The electrodeposition was conducted at a constant potential of 1.0V for 5h.	77wt.%	~1200 mAhg <sup>-1</sup> at 0.1 Ag <sup>-1</sup> depend on sulfur ~920 mAhg <sup>-1</sup> at 0.1 Ag <sup>-1</sup> depend on sample	Remains about 100% of second value after 200 cycles at 0.5 Ag <sup>-1</sup> .	
	Thermal diffusion	Carbon sphere	Heated at 149 °C for 6 h and kept at 300 °C for 2 h. High temperature and long time are cost.	42wt.%	~1000 mAhg <sup>-1</sup> at 0.04 Ag <sup>-1</sup> depend on sulfur ~420 mAhg <sup>-1</sup> at 0.1 Ag <sup>-1</sup> depend on sample	Remains about 80% of second value after 500 cycles at 0.4 Ag <sup>-1</sup> . Drop quickly in initial 200 cycles.	1
	Chemical deposition method + Thermal diffusion	Porous Carbon Nanotubes	Use chemical deposition method to grow sulfur on the surface of carbon and thermal diffusion method (155 °C for 12 h) to facilitate sulfur diffusion into the pore. Can not directly grow sulfur in the pore.	75wt.%	~1200 mAhg <sup>-1</sup> at 0.2 C depend on sulfur ~900 mAhg <sup>-1</sup> at 0.2 C depend on sample	Remains about 80% of second value after 300 cycles at 0.2C (~0.5 A g <sup>-1</sup> ) rate.	2
	Chemical deposition method	3D Graphene Nanosheet	Only use chemical deposition method. Sulfur particles with 50nm size can be found in TEM.	76.4wt.%	~1000 mAhg <sup>-1</sup> at 0.1 C depend on sulfur ~760 mAhg <sup>-1</sup> at 0.1 C depend on sample	Remains about 60% of second value after 200 cycles at 0.1C (~0.2 A g <sup>-1</sup> ) rate.	3

Hydrotherma method	al Graphene oxide	Toxic CS <sub>2</sub> is used as a solvent of sulfur. The mixture is sealed in a Teflon-lined stainless-steel autoclave and treated at 160 °C for 10 h. High temperature and long time are cost.	65wt.%	~800 mAhg <sup>-1</sup> at 0.2 C depend on sulfur ~520 mAhg <sup>-1</sup> at 0.2 C depend on sample	Remains about 45% of second value after 100 cycles at 0.5C (~0.8 A g <sup>-1</sup> ) rate. (Sample without an Al <sub>2</sub> O <sub>3</sub> coating)	4
Thermal diffus	ion Carbon	After thermal diffusion method, use toxic CS <sub>2</sub> to wash sample to remove excess external sulfur. About 14.1wt.% (depend on sample) of sulfur lost after washed.	44.2wt.% S-BMC/S-70- W	~1000 mAhg <sup>-1</sup> at 1 C depend on sulfur ~440 mAhg <sup>-1</sup> at 1 C depend on sample	Remains about 83% of second value after 100 cycles at 1C (~1.7 A g <sup>-1</sup> ) rate. (Sample S-BMC/S- 70-W)	5
		By ma				
		nip		~1040 mAbg-1 at		
Vapor-infusio method	on Mesoporous carbon	ulat ing	69.73wt.%	0.5 C depend on sulfur ~730 mAhg <sup>-1</sup> at 0.5 C depend on sample	Remains about 91% of initial value after 100 cycles at 0.5C (~0.8 A g <sup>-</sup> <sup>1</sup> ) rate.	6
		mg				
		the				
		met				

## al oxi de part icle size and por osit у, holl

OW car bon sph eres wit h hig h spe cifi

#### С surf ace area of 648 m 2 g **□**1 , 3 nm

ave rag e por e dia met er (Fig ure S2a

,b), and larg e inte rnal voi d spa ce (Fig

ure 1 a) wer e faci lely crea ted. In the fina

### 1 step of the synt hesi S, we take adv anta

ge of the rela tive ly low subl ima tion tem

per atur e of sulf ur to infu se gas eou S

sulf
ur
into
the
car
bon
sup
port
pres
ent
in

one co mp art me nt of a dua 1-CO mp

#### art me nt seg me nte d tub e. [19] Thi

S met hod olo gу faci litat es fast , effi

cien t, and con troll ed infu sion of ele me

ntal sulf ur into the host car bon por ous stru

ctur e and yiel ds part icle S wit h tap

den sity of aro und 0.8 2 gc m  $\Box 3$ 

The rma 1 gra vim etri C anal ysis (Fig ure

S3) sho WS that app roxi mat ely 35 % sulf

ur can be inc orp orat ed in the part icle

s in
a
sing
le
pas
s,
and
that
by
thre
e

pas ses (i.e. rep eat exp osu res of the por

ous car bon part icle s to sulf ur vap or By

ma nip ulat ing the met al oxi de part icle

size and por osit у, holl OW car bon sph eres

wit h hig h spe cifi C surf ace area of

#### 648 m 2 g **□**1 , 3 nm ave rag e por e

## dia met er (Fig ure S2a ,b), and larg e inte

## rnal voi d spa ce (Fig ure 1 a) wer e faci

lely crea ted. In the fina 1 step of the synt

## hesi S, we take adv anta ge of the rela tive

ly low subl ima tion tem per atur e of sulf ur

to
infu
se
gas
eou
S
sulf
ur
into
the
car

bon sup port pres ent in one CO mp art me

## nt of a dua 1co mp art me nt seg me

nte
d
tub
e. [19] Thi
1 111 S
met
hod
olo
gу

faci litat es fast , effi cien t, and con troll

ed infu sion of ele me ntal sulf ur into the

host car bon por ous stru ctur e and yiel ds

part icle S wit h tap den sity of aro und

0.8 2 gc m  $\Box 3$ The rma 1 gra vim

etri
C
anal
ysis
(Fig
ure
S3)
sho
WS
that
app

roxi mat ely 35 % sulf ur can be inc orp

orat ed in the part icle s in a sing le pas

S, and that by thre e pas ses (i.e. rep eat

exp osu res of the por ous car bon part icle

		s to				
		sulf				
		ur				
		vap				
		<b>OT</b> By exposure of the				
		porous carbon particles to sulfur vapor. A lot of sulfur vapor will be cost.				
Thermal diffusion	Mesoporous carbon	Heated at 160 °C for 24 h and kept at 200 °C for 10 min with under flowing argon gas (50 cm <sup>3</sup> s <sup>-1</sup> ).	63wt.%	~1070 mAhg <sup>-1</sup> at 0.3 C (initial) depend on sulfur ~670 mAhg <sup>-1</sup> at 0.3 C depend on sample	Remains about 65% of initial value after 300 cycles at 0.3C (~0.5 A g <sup>-1</sup> ) rate.	7
Thermal diffusion	Mesoporous carbon	Toxic CS <sub>2</sub> is used as a solvent of sulfur. Carbon is then added to the solution and allowed CS <sub>2</sub> evaporation under stirring. The mixture was then heated at 155 °C for 8 h. Evaporate of CS <sub>2</sub> is very dangerous.	75wt.%	~1150 mAhg <sup>-1</sup> at 0.084 A g <sup>-1</sup> (initial) depend on sulfur ~860 mAhg <sup>-1</sup> at 0.084 A g <sup>-1</sup> depend on sample	Remains about 87% of fifth value (first stable cycle) after 200 cycles at 0.084 A g <sup>-1</sup> .	8

#### Notes and references

- 1 B. Zhang, X. Qin, G. R. Lia, X. P. Gao, *Energ. Environ. Sci.*, 2010, **3**, 1531-1537.(Cited 622)
- 2 J. S. Lee, J. Jun, J. Jang, A. Manthiram, *Small*, 2017, DOI: 10.1002/smll.201602984.
- 3 Z. Zhang, L. Kong, S. Liu, G. Li, X. Gao, *Adv. Energy Mater.*, 2017, DOI: 10.1002/aenm.201602543
- 4 M. Yu, W. Yuan, C. Li, J. Hong, G. Shi, J. Mater. Chem. A, 2014, 2, 7360-7366.

Jçrg Schuster, Guang He, Benjamin Mandlmeier, Taeeun Yim, Kyu Tae Lee, Thomas Bein,\* and Linda F. Nazar

# Jçrg Schuster, Guang He, Benjamin Mandlmeier, Taeeun Yim, Kyu Tae Lee, Thomas Bein,\* and Linda F. Nazar

- 5 J. Schuster, G. He, B. Mandlmeier, T. Yim, K. T. Lee, T. Bein, L. F. Nazar, Angew. Chem., 2012, **124**, 3651-3655.
- N. Jayaprakash, J. Shen, S. S. Moganty, A.Corona, L. A. Archer, *Angew. Chem.*, 2011, 123, 6026-6030. (Cited 944)

- 7 H. Wang, C. Zhang, Z. Chen, H. K. Liu, Z. Guo, Carbon, 2015, 81, 782-787.
- 8 T. Xu, J. Song, M. L. Gordin, H. Sohn, Z. Yu, S. Chen, D. Wang, ACS Appl. Mater. Interfaces, 2013, 5, 11355-11362.