

Supporting Information for

## Highly Reversible Li–Se Batteries with Ultra-Lightweight N,S-Codoped Graphene Blocking Layer

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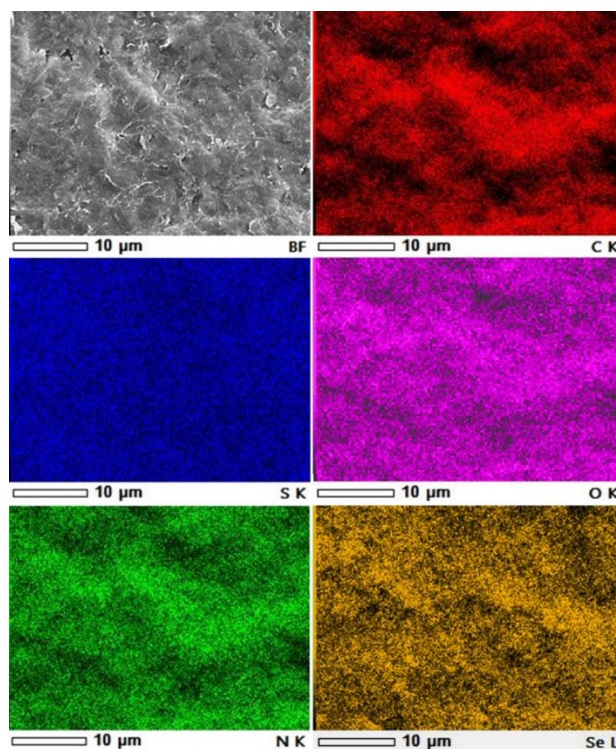
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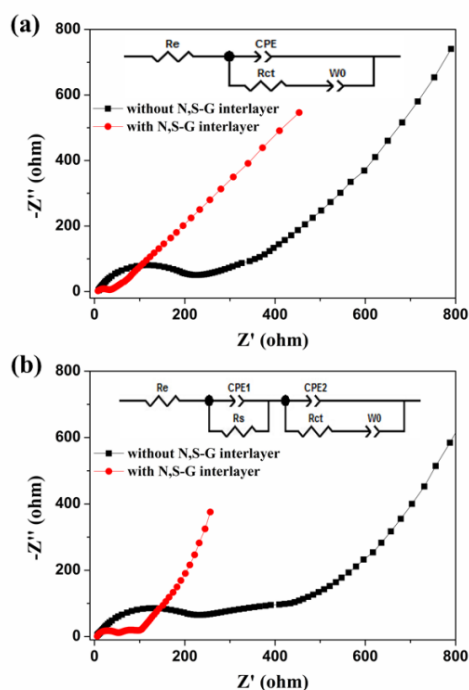
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### Supplementary Figures



**Fig. S1** SEM images of the N,S-G interlayer after 500 cycles and the corresponding element mapping of C, S, O, N, Se



**Fig. S2** Nyquist plots and equivalent circuit (inset) of the polyselenides catholyte with and without N,S-G interlayer: **a** before cycling and **b** after 500 cycles

**Table S1** Electrochemical performances comparison with different carbon-Se composites cathode and carbon interlayer

Sample name	Selenium content in the cathode (%)	Se loading (mg cm <sup>-2</sup> )	Long-cycle performances	Rate capability	References
polyselenides catholyte with N,S-G interlayer	~79 (including N,S-G weight)	~5.0	330.7 mAh g <sup>-1</sup> after 500 cycles at 1 C	301.4 mA h g <sup>-1</sup> at 4 C	This work
Se/porous carbon microcubes composite	40	1.0-1.5	231.4 mAh g <sup>-1</sup> after 460 cycles at 0.5 C	218.1 mAh g <sup>-1</sup> at 5 C	[1]
Se-impregnated hollow carbon microspheres	41.7	0.72	525 mAh g <sup>-1</sup> after 1000 cycles at 0.74 C	496 mAh g <sup>-1</sup> at 2.97 C	[2]
Se/CNT@microporous carbon composite	50.2	2.0	~200 mAh g <sup>-1</sup> after 1000 cycles at 1 C	~150 mAh g <sup>-1</sup> at 5 C	[3]
Graphene-CNT@Se	51	1.6	504.3 mA h g <sup>-1</sup> after 150 cycles at 0.2 C	436.4 mAh g <sup>-1</sup> at 4 C	[4]
Se@hollow-core nitrogen-doped carbon nanobelts	62.5	3.0	~453.2 mAh g <sup>-1</sup> after 1000 cycles at 1 C	~450 mAh g <sup>-1</sup> at 2.37 C	[5]
Graphene-encapsulated selenium/polyaniline core-shell nanowire	52.9	3.0	461.7 mAh g <sup>-1</sup> after 200 cycles at 0.2 C	510.9 mAh g <sup>-1</sup> at 2 C	[6]
Porous carbon nanofibers-selenium	52.3	0.8	516 mAh g <sup>-1</sup> after 900 cycles at 0.74 C	306 mAh g <sup>-1</sup> at 5.9 C	[7]

Se/chitosan-derived hierarchical porous carbon	35	1.37-1.77	537.6 mAh g <sup>-1</sup> after 100 cycles at 0.2 C	325.2 mAh g <sup>-1</sup> at 4.7 C	[8]
Se/CMK-3	39.2	2.0	600 mAh g <sup>-1</sup> after 50 cycles at 0.1 C	—	[9]
Se/carbon aerogel	44.8	1.5-2.0	309 mAh g <sup>-1</sup> after 100 cycles at 0.5 C	301 mAh g <sup>-1</sup> at 5 C	[10]
Se/porous carbon sphere	56.4	2.0	417 mAh g <sup>-1</sup> after 1200 cycles at 1 C	384.75 mAh g <sup>-1</sup> at 20 C	[11]
Se/nitrogen-doped microporous carbon spheres	35	2.0	570 mAh g <sup>-1</sup> after 350 cycles at 0.5 C	200 mAh g <sup>-1</sup> at 5 C	[12]
Core-shell structured selenium@ carbon spheres	54.4	1.0-2.0	300 mAh g <sup>-1</sup> after 100 cycles at 0.1 C	447 mAh g <sup>-1</sup> at 4.7 C	[13]
Selenium in nitrogen-containing hierarchical porous carbon	45.0	1.0	267 mAh g <sup>-1</sup> after 100 cycles at 1 C	~300 mAh g <sup>-1</sup> at 5 C	[14]
Selenium/microporous carbon nanofiber composite	40	1.0-1.2	400 mAh g <sup>-1</sup> after 2000 cycles at 1 C	420 mAh g <sup>-1</sup> at 5 C	[15]
Se@Carbide-derived carbon	46.5	1.0	~450 mAh g <sup>-1</sup> after 150 cycles at 0.2 C	310 mAh g <sup>-1</sup> at 2 C	[16]
Se@mesoporous carbon/ graphene	49.6	1.2	385 mAh g <sup>-1</sup> after 1300 cycles at 1 C	274 mAh g <sup>-1</sup> at 3 C	[17]
Se@erylene-3,4,9,10-tetracarboxylic dianhydride derived carbon	43.2	1.2	430 mAh g <sup>-1</sup> after 250 cycles at 0.15 C	280 mAh g <sup>-1</sup> at 1.78 C	[18]
Hollow carbonized polyaniline spheres/selenium composites	32.96	1.5-2.0	298.7 mAh g <sup>-1</sup> after 100 cycles at 0.5 C	246.8 mAh g <sup>-1</sup> at 2 C	[19]
Selenium/micro-mesoporous carbon sphere nanocomposite	48	2.0	540 mAh g <sup>-1</sup> after 100 cycles at 0.1 C	430 mAh g <sup>-1</sup> at 5 C	[20]
Selenium@mesoporous carbon	38.4	0.55-0.71	306.9 mAh g <sup>-1</sup> after 100 cycles at 0.5 C	275.9 mAh g <sup>-1</sup> at 2 C	[21]
Cellulose based carbon interlayer	80 (without carbon interlayer weight) 64%	1.6	282 mAh g <sup>-1</sup> after 100 cycles at 0.5 C	—	[22]
Se@reduced graphene oxide with CNT interlayer	(without CNT interlayer weight) 52%	2.0	265 mAh g <sup>-1</sup> after 200 cycles at 0.2 C	250 mAh g <sup>-1</sup> at 5 C	[23]
Graphene-coated polymer separator	(including graphene weight)	4.0	331 mAh g <sup>-1</sup> after 1000 cycles at 0.5 C	~3400 mAh g <sup>-1</sup> at 5 C	[24]

**Table S2** Impedance parameters of the Li-Se batteries calculated according to the equivalent circuits

Sample name	Cycle number	Resistance		
		$R_e$ ( $\Omega$ )	$R_s$ ( $\Omega$ )	$R_{ct}$ ( $\Omega$ )
Without N,S-G interlayer	Before cycling	3.75	—	38.64
	500 <sup>th</sup> cycles	5.87	34.23	65.92
With N,S-G interlayer	Before cycling	7.90	—	176.80
	500 <sup>th</sup> cycles	4.93	195.70	479.10

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