## **Supporting Information**

## Engineering carbon layer on yolk-shell bimetallic selenide microsphere boosts lithium storage as a high-performance anode

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Fig. S1 SEM images of (a, b) CoNi-glycerate, (c, d) CoNi-OH and (e, f) CoNi-OH@PDA.



Fig. S2 XRD patterns of CoNi-glycerate, CoNi-OH and CoNi-OH@PDA yolk shell structure.



Fig. S3 SEM images of (a, b) CoSe<sub>2</sub>/Ni<sub>3</sub>Se<sub>4</sub> and (c, d) s-CoSe<sub>2</sub>/Ni<sub>3</sub>Se<sub>4</sub>.



Fig. S4 Galvanostatic charge/discharge curves of (a)  $CoSe_2/Ni_3Se_4$  and (b) s- $CoSe_2/Ni_3Se_4$ .

## Table S1. Comparison of electrochemical properties of Co-based and Ni-based selenides carbon materials on LIBs

Electrode materials	Cycling performance	Rate Performance (mA h g <sup>-1</sup> @ A g <sup>-1</sup> )	Ref.
NiSe <sub>2</sub> /CoSe <sub>2</sub> /Graphene	< 245.6 mA h g <sup>-1</sup> after 500 cycles at 1 A g <sup>-1</sup>	544 @ 0.2 184 @ 2.0	Colloid. Surface. A, 2024, <b>688</b> , 133685.
CoSe-MS	156 mA h g <sup>-1</sup> after 100 cycles at 0.5 A g <sup>-1</sup>	-	ACS Appl. Mater. Interfaces, 2019, <b>11</b> , 11292-11297.
ZnSe-CoSe@NC/MX	469 mA h g <sup>-1</sup> after 80 cycles at 0.2 A g <sup>-1</sup>	477 @ 0.1 444 @ 0.2 335 @ 2.0	Electrochim. Acta, 2024, <b>487</b> , 144148.
CoSe/NC	310 mA h g <sup>-1</sup> after 500 cycles at 1 A g <sup>-1</sup>	-	Nanoscale Res. Lett., 2019, <b>14</b> , 385.
CoSe2@C	-	521 @ 0.1 468 @ 0.2 321 @ 1.0	Appl. Surf. Sci., 2019, 483, 85-90.
NiSe/rGO	378 mA h g <sup>-1</sup> after 50 cycles at 0.05 A g <sup>-1</sup>	364 @ 0.2 110@ 3.2	Materials, 2019, <b>12,</b> 3709.
NiSe/C	428 mA h g <sup>-1</sup> after 50 cycles at 0.1 A g <sup>-1</sup>	384 @ 0.2 299 @ 0.5	Electrochim. Acta, 2016, <b>208</b> , 238.
CoSe <sub>2</sub> /Ni <sub>3</sub> Se <sub>4</sub> @NC	$\begin{array}{c} 518 \text{ mA h } g^{-1} \text{ after } 100 \\ \text{cycles at } 0.2 \text{ A } g^{-1} \\ 319 \text{ mA h } g^{-1} \text{ after} \\ 500 \text{ cycles at } 1 \text{ A } g^{-1} \end{array}$	564 @ 0.2 129 @ 4.0	This Work



Fig. S5 SEM images of (a)  $s-CoSe_2/Ni_3Se_4$ , (b)  $CoSe_2/Ni_3Se_4$  and (c)  $CoSe_2/Ni_3Se_4$ @NC after 100 cycles at 200 mA g<sup>-1</sup>.



Fig. S6 CV curve with the pseudocapacitive fraction at a scan rate of 1.6 mV s<sup>-1</sup> of (a)  $CoSe_2/Ni_3Se_4@NC$ , (b)  $CoSe_2/Ni_3Se_4$  and (c) s- $CoSe_2/Ni_3Se_4$ .



Fig. S7 (a) The EIS plots of  $CoSe_2/Ni_3Se_4@NC$ ,  $CoSe_2/Ni_3Se_4$  and  $s-CoSe_2/Ni_3Se_4$  before cycling. (b) The EIS plots of  $CoSe_2/Ni_3Se_4@NC$  before and after 3 cycles.



**Fig. S8** The survey spectrum of XPS spectra for CoSe<sub>2</sub>/Ni<sub>3</sub>Se<sub>4</sub>@NC at different states (initial fully charge and discharge).