## **Supplementary Information**

Flexible free-standing lithium-ion electrode based on robust layered

## assembly of graphene and Co<sub>3</sub>O<sub>4</sub> nanosheets

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Fig. S1. O1s spectrum of Co<sub>3</sub>O<sub>4</sub>/GS hybrid film.



Fig. S2. SEM images of pure Co<sub>3</sub>O<sub>4</sub>.



**Fig. S3.** Comparative cycle performance of electrodes with different content of  $Co_3O_4$  at a current density of 200 mA g<sup>-1</sup>. In the work,  $Co(OH)_2/GO$  mass ratio was varied as 1:2, 1:1 and 2:1 to optimize the lithium storage performance. The samples were named as F-Co<sub>3</sub>O<sub>4</sub>/GS(1:2), F-Co<sub>3</sub>O<sub>4</sub>/GS(1:1) and F-Co<sub>3</sub>O<sub>4</sub>/GS(2:1), respectively. The

sample of pure graphene film was named as F-GS. As shown in Fig. S3, it is obvious that F-Co<sub>3</sub>O<sub>4</sub>/GS(1:1) with Co<sub>3</sub>O<sub>4</sub> content of 75 % (determined by TG analysis) exhibited the highest specific capacity and the best cyclic stability (~1200 mAh g<sup>-1</sup> after 100 cycles). In contrast, for F-Co<sub>3</sub>O<sub>4</sub>/GS(1:2) and F-Co<sub>3</sub>O<sub>4</sub>/GS(2:1), the specific capacity decayed significantly and maintained only 876 and 517 mAh g<sup>-1</sup> after 100 cycles, respectively. The specific capacity of pure graphene free-standing and binder-free electrode was only ~230 mAh g<sup>-1</sup>. Graphene can improve the conductivity of the composite; however, its specific capacity is lower than that of Co<sub>3</sub>O<sub>4</sub>. Therefore, only with the optimal weight ratio of Co<sub>3</sub>O<sub>4</sub> to GS, the hybrid film can exhibit the best performance. Here, the optimal graphene content was 75 %. In the manuscript, all the detailed discussions were based on the optimized sample F-Co<sub>3</sub>O<sub>4</sub>/GS(1:1) with graphene content of 75 %.



Fig. S4. Coulombic efficiency of the  $Co_3O_4/GS$  free-standing electrode at a current density of 200 mA g<sup>-1</sup>.