## **Supporting Information**

## Construction of cobalt oxyhydroxide nanosheets with rich oxygen vacancies as high-performance lithium-ion batteries anodes

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Fig. S1 SEM images of  $Co(OH)_2$  (a) and  $Co_3O_4$  (b); TEM images of  $Co(OH)_2$  (c) and  $Co_3O_4$  (d).



Fig. S2 XRD patterns of  $Co(OH)_2$  and  $Co_3O_4$ .



Fig. S3 XPS spectrum of CoOOH nanosheets.



Fig. S4 (a) TG profile of CoOOH nanosheets; (b) TGA profile of CoOOH nanosheets.



Fig. S5 Cycling performance of CoOOH, Co(OH)<sub>2</sub>, and Co<sub>3</sub>O<sub>4</sub> electrodes at 3 A  $g^{-1}$ .

Anode materials	Current density (mA g <sup>-1</sup> )	Cycle number	Capacity retention	Initial coulombic efficiency (%)	Refs
			$(mAh g^{-1})$		
Co <sub>3</sub> O <sub>4</sub> nanosheets	200	100	1717	~68	[1]
CoOOH in Lithium–Sulfur Battery	0.1C (about 100)	100	1199.4		[2]
The composite materials of mica	225	70	650		[3]
flake and cobalt oxide					
The yolk–shell Co <sub>3</sub> O <sub>4</sub> /C	200	120	1100	~71	[4]
N-Doped Carbon/Cobalt Ferrite	3000	300	410	~78	[5]
Hybrid Nanocomposites					
Nanoarchitectured Co <sub>3</sub> O <sub>4</sub> /reduced	1C(about 1000)	200	513.9		[6]
graphene oxide					
Co <sub>3</sub> O <sub>4</sub> nanotubes	100	80	380	~58.7	[7]
nanoparticles-assembled Co <sub>3</sub> O <sub>4</sub>	200	50	1340	~73	[8]
microspheres					
Oxidizing solid Co into hollow	200	500	871.5	~66.7	[9]
Co <sub>3</sub> O <sub>4</sub>					
Co <sub>2</sub> (OH) <sub>2</sub> CO <sub>3</sub> nanowires and rGO	200	150	1380	~71.7	[10]
films					
CoOOH nanosheets	200	300	1588	~90	This
					work

**Table S1.** Comparison of cycling stability and initial coulombic efficiency of CoOOH anode with previously reported cobalt oxide anodes for LIBs.



**Fig. S6** (a) Nyquist plots of CoOOH, Co(OH)<sub>2</sub>, and Co<sub>3</sub>O<sub>4</sub> electrodes (insets show the equivalent circuits for EIS fitting and the detail of Nyquist plots); (b) The relationship between  $I_{\text{peak}}$  and  $v^{1/2}$  for CoOOH electrode; (c) The relationship between  $I_{\text{peak}}$  and  $v^{1/2}$  for Co<sub>3</sub>O<sub>4</sub> electrode.



Fig. S7 SEM and High-resolution TEM images of CoOOH (a, d),  $Co(OH)_2$  (b, e), and  $Co_3O_4$  (c, f) electrodes after 300 cycles.



**Fig. S8** Kinetic analysis of the electrochemical behavior of the  $Co_3O_4$  electrode versus Li<sup>+</sup>/Li. (a) CV curves at various scan rates ranging from 0.1 to 1.5 mV s<sup>-1</sup>; (b) Determination of b values using the relationship between the peak current and scan rate according to the voltammograms in (a); (c) Contribution ratios of the capacitive and diffusion-controlled effects at various scan rates; (d) The relationship between I<sub>p</sub> and v<sup>1/2</sup>.



**Fig. S9** Kinetic analysis of the electrochemical behavior of the Co(OH)<sub>2</sub> electrode versus Li<sup>+</sup>/Li. (a) CV curves at various scan rates ranging from 0.1 to 1.0 mV s<sup>-1</sup>; (b) Determination of b values using the relationship between the peak current and scan rate according to the voltammograms in (a); (c) Contribution ratios of the capacitive and diffusion-controlled effects at various scan rates; (d) The relationship between I<sub>p</sub> and v<sup>1/2</sup>.



Fig. S10 The relationship between  $I_{p}$  and  $v^{1/2}$  for the CoOOH electrode.



Fig. S11 A 3 × 3 supercell of the CoOOH (001) crystal surface including 3 atomic layers model.



Fig. S12 The model of the process for adsorption of Li<sup>+</sup> in O active sites, desorption of Li<sub>2</sub>O and formation of VO.



**Fig. S13** The model of the process for adsorption of Li<sup>+</sup> in a OH active site and formation of LiH or LiOH; desorption of LiH and formation of CoOOH–VO–VH.



Fig. S14 The model of the process for adsorption of  $Li^+$  in the O active site of the VH; desorption of  $Li_2O$  and formation of CoOOH–VO–VH–VO.

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