

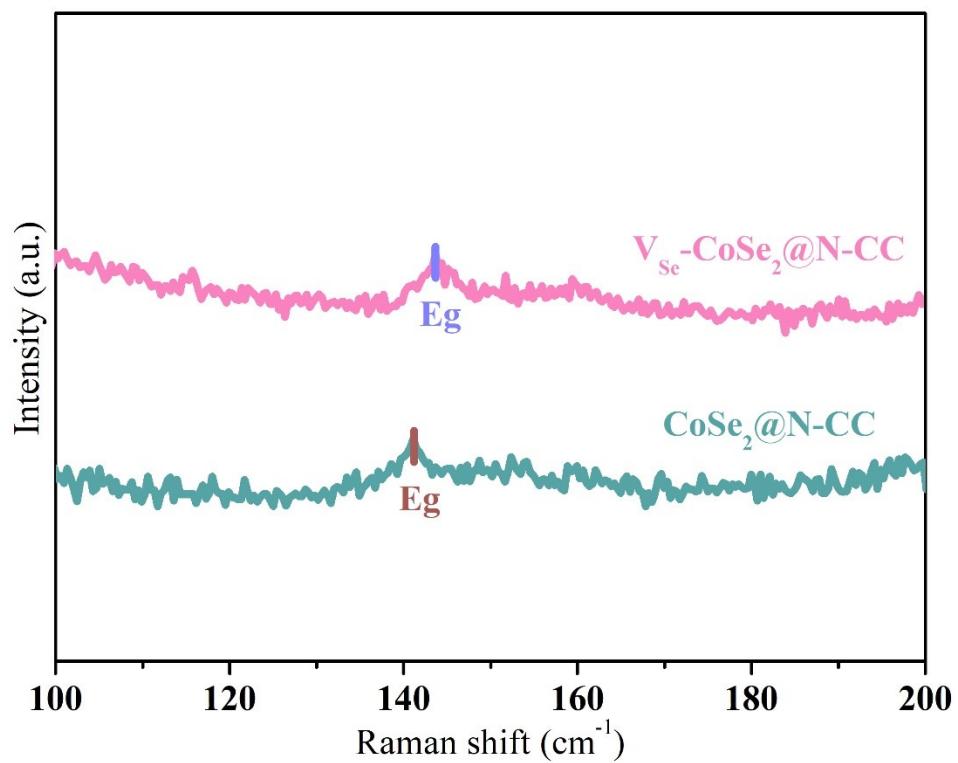
**Anionic Vacancy-dependent Activity of CoSe<sub>2</sub> with Tunable Interfacial Electronic Structure on N-doped Carbon Cloth Endowing Advanced Li-O<sub>2</sub> Battery**

Zhiqian Hou<sup>a,b</sup>, Chaozhu Shu<sup>a\*</sup>, Ruixin Zheng<sup>a</sup>, Chunhai Liu<sup>a</sup>, Zhiqun Ran<sup>a</sup>, Tingshuai Yang<sup>a</sup>, Peng Hei<sup>a</sup>, Qiang Zhang<sup>a</sup>, Jianping Long<sup>a\*</sup>

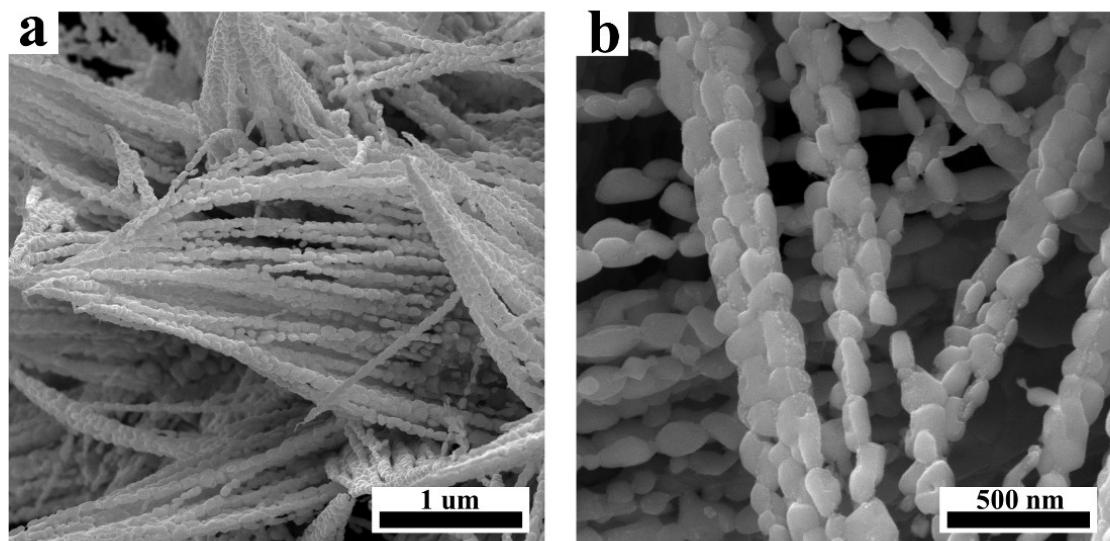
a. College of Materials and Chemistry & Chemical Engineering, Chengdu University of Technology, 1#, Dongsanlu, Exxianqiao, Chengdu 610059, Sichuan, P. R. China.

b. State Key Lab of High Performance Ceramics and Superfine microstructure, Shanghai Institute of Ceramics, Chinese Academy of Sciences, 1295 Dingxi Road, 200050 Shanghai, People's Republic of China.

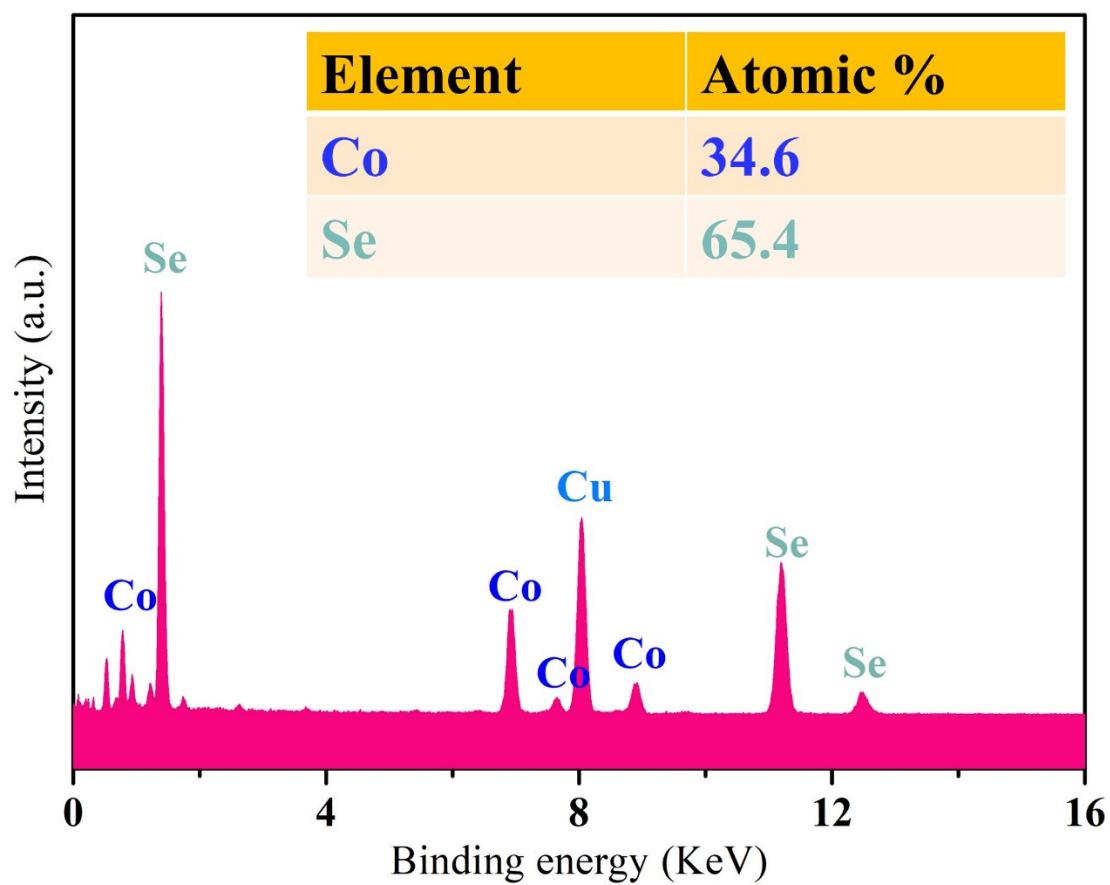
Email: czshu@imr.ac.cn (Chaozhu Shu); longjianping@cdut.cn (Jianping Long)



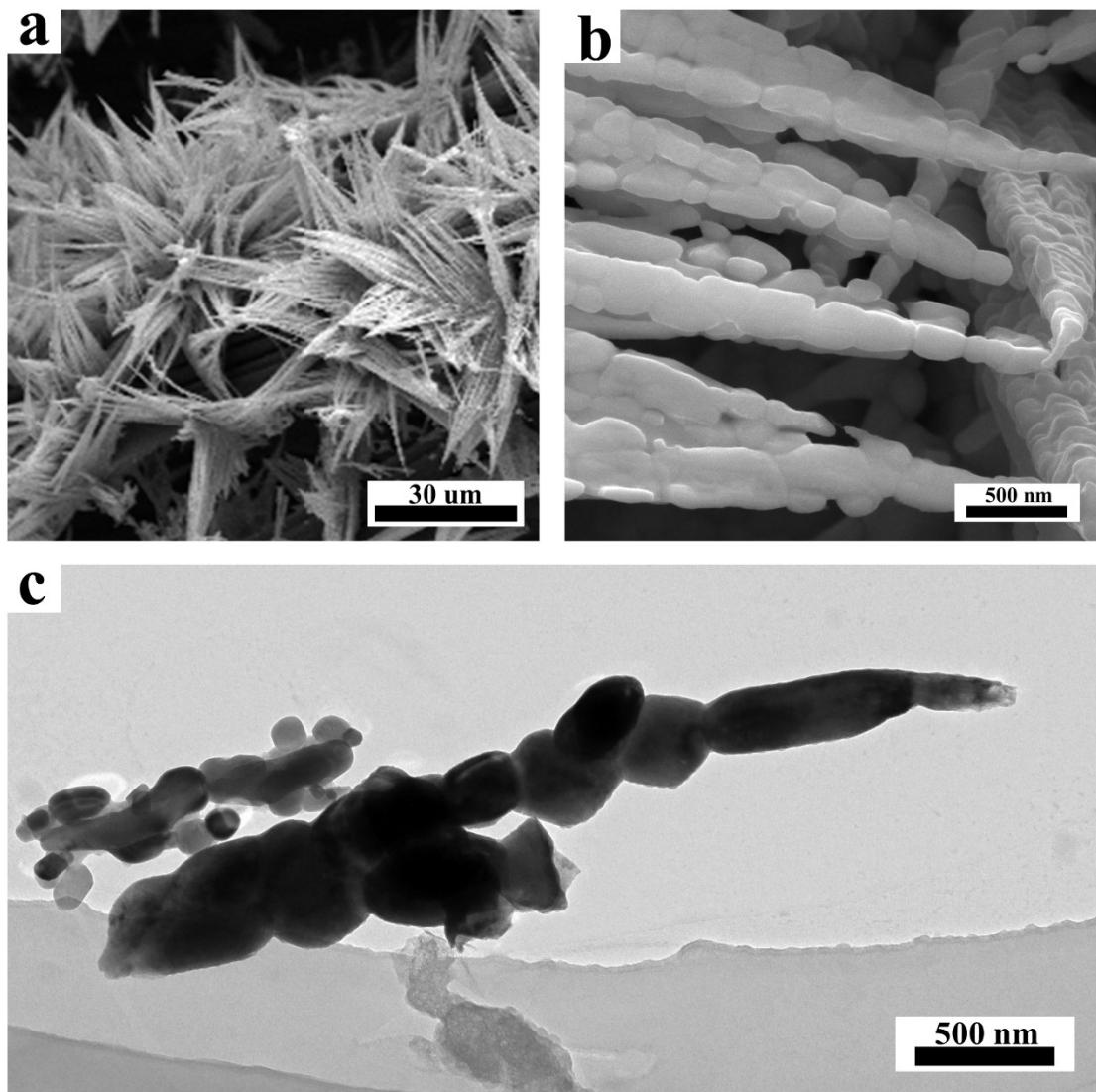
**Figure S1.** Raman spectra of  $\text{CoSe}_2\text{@N-CC}$  and  $\text{V}_{\text{Se}}\text{-CoSe}_2\text{@N-CC}$ .



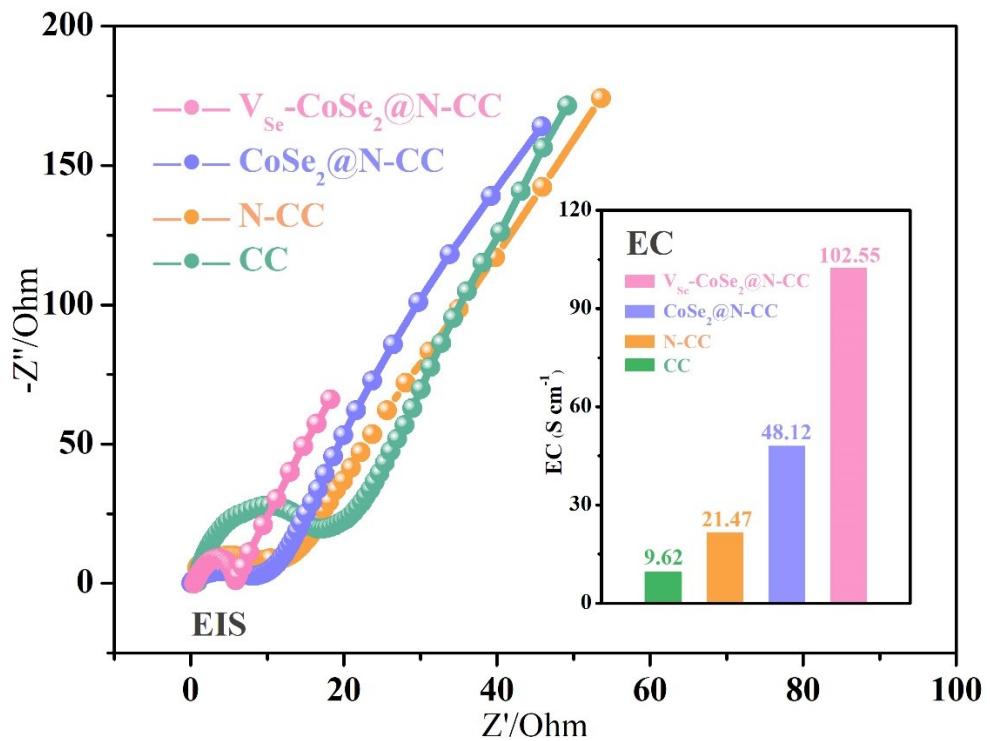
**Figure S2.** (a) and (b) FESEM images of the CoSe<sub>2</sub>@N-CC.



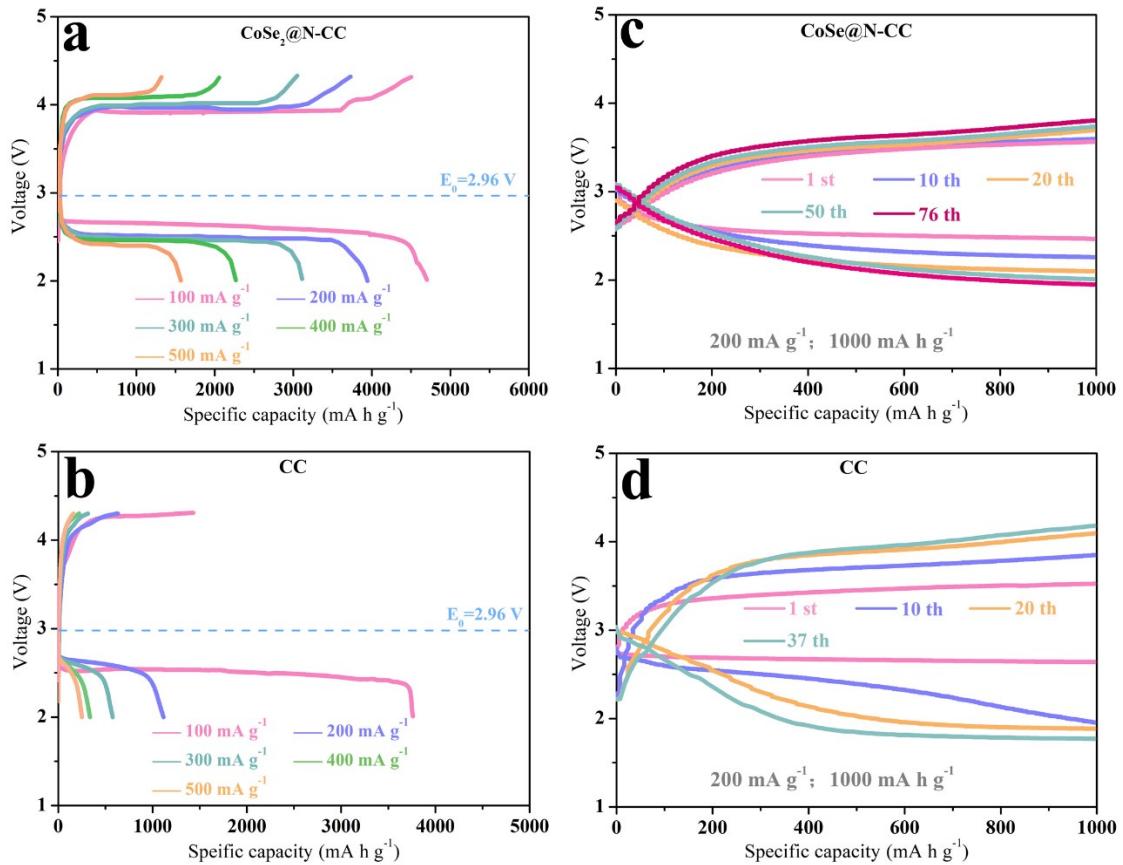
**Figure S3.** EDS spectrum of CoSe<sub>2</sub>@N-CC.



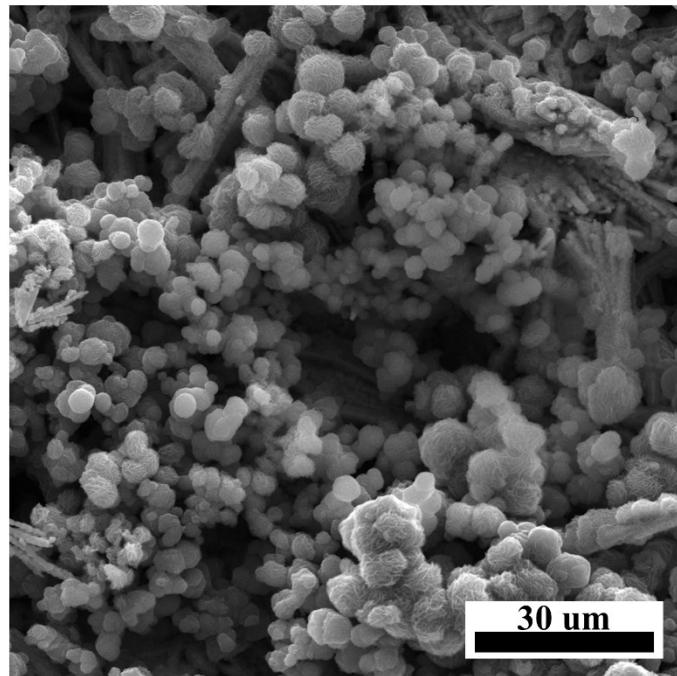
**Figure S4.** FESEM images of  $\text{V}_{\text{Se}}\text{-CoSe}_2@\text{N-CC}$  integrated electrode with low (a) and (b) high magnification after 24 h of drastic sonication; (c) the typical corresponding TEM image.



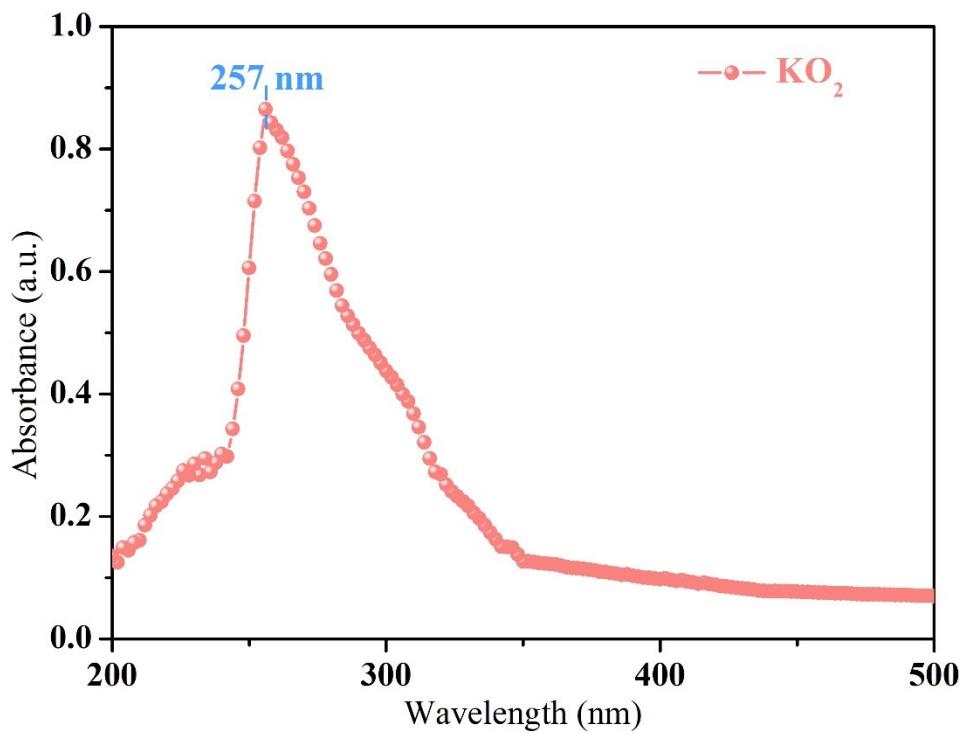
**Figure S5.** Electrochemical impedance spectroscopy (EIS) of  $\text{V}_{\text{Se}}\text{-CoSe}_2@\text{N-CC}$ ,  $\text{CoSe}_2@\text{N-CC}$  N-CC, and CC; the illustration is the corresponding electronic conductivity.



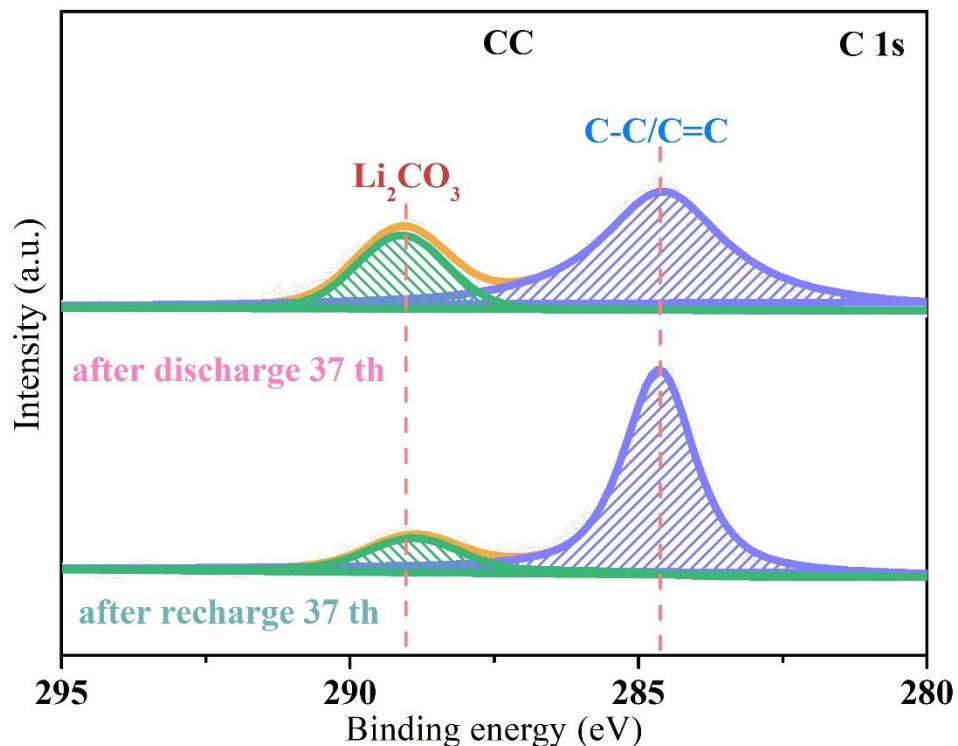
**Figure S6.** The rate performance of Li-O<sub>2</sub> cells based on (a) CoSe<sub>2</sub>@N-CC, and (b) CC electrodes at various current density; the discharge/charge curves of Li-O<sub>2</sub> batteries with (c) CoSe<sub>2</sub>@N-CC, and (d) CC electrodes at a current density of 200  $\text{mA g}^{-1}$  with a restricted capacity of 1000  $\text{mA h g}^{-1}$ .



**Figure S7.** SEM image of V<sub>Se</sub>-CoSe<sub>2</sub>@N-CC electrode after fully discharge.



**Figure S8.** Ultraviolet-visible spectrum of  $\text{KO}_2$  dissolved in TEGDME solution.



**Figure S9.** Typical XPS high-resolution spectrum of C 1s with the CC electrode after first (a) and 37 th (b) discharge/charge.

**Table S1.** The electrochemical performance comparison of V<sub>Se</sub>-CoSe<sub>2</sub>@N-CC oxygen electrode with transition metal chalcogenides electrodes.

Catalysts	Current density (mA g <sup>-1</sup> )	Discharge capacity (mA h g <sup>-1</sup> )	Cycle number (cycles)	Ref.
<b>MoS<sub>2</sub></b>	100	1250	50	[1]
<b>Co<sub>3</sub>S<sub>4</sub></b>	100	5917	25	[2]
<b>Ni<sub>3</sub>S<sub>2</sub></b>	400	3264	116	[3]
<b>NiCo<sub>2</sub>S<sub>4</sub></b>	150	14173	101	[4]
<b>CoSe<sub>2</sub>/CoO</b>	100	1500	30	[5]
V <sub>Se</sub> - <b>CoSe<sub>2</sub>@N-CC</b>	100	6089	More than 490	This work

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**Table S2.** The electrochemical performance of different types of oxygen electrode.

Catalysts	Current density	Cycle Performance	First Discharge	Ref.
			Capacity (mA h g <sup>-1</sup> ) (Cycles)	
<b>Co<sub>3</sub>O<sub>4</sub>/CNTs</b>	100 mA g <sup>-1</sup>	116	4331 mA h g <sup>-1</sup>	[1]
<b>2D Co<sub>3</sub>S<sub>4</sub> nanosheets</b>	100 mA g <sup>-1</sup>	25	5917 mA h g <sup>-1</sup>	[2]
<b>CoS<sub>2</sub>/RGO</b>	100 mA g <sup>-1</sup>	20	3000 mA h g <sup>-1</sup>	[3]
<b>MnCo<sub>2</sub>O<sub>4</sub> microspheres</b>	200 mA g <sup>-1</sup>	50	4861 mA h g <sup>-1</sup>	[4]
<b>NiO</b>	100 mA g <sup>-1</sup>	40	1260 mA h g <sup>-1</sup>	[5]
<b>V<sub>Se</sub>-CoSe<sub>2</sub>@N-CC</b>	100 mA g <sup>-1</sup>	>490	6089 mA h g <sup>-1</sup>	This work

## Reference

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