## Electronic Supplementary Information

## One-pot Synthesized Molybdenum Dioxide-Molybdenum Carbide Heterostructures Coupled with 3D Holey Carbon Nanosheets for Highly Efficient and Ultrastable Cycling Lithium-Ion Storage

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Fig. S1 TGA and DTA curve of MoO<sub>2</sub>/C samples.

The TGA analysis of MoO<sub>2</sub>/C nanocomposites was carried out to calculate the MoO<sub>2</sub>, Mo<sub>2</sub>C and carbon content in MoO<sub>2</sub>/Mo<sub>2</sub>C/C nanocomposites. In order to keep carbon content consistent, the MoO<sub>2</sub>/C and MoO<sub>2</sub>/Mo<sub>2</sub>C/C samples were prepared from the same precursors, just with different carbonation temperature. A 32.66 % weight of carbon and 69.34 % weight of MoO<sub>2</sub> were obtained according to the TGA results of MoO<sub>2</sub>/C nanocomposites in Fig. S3. The weight of carbon of ca. 32.66 % in MoO<sub>2</sub>/Mo<sub>2</sub>C/C samples was extrapolated. Then the content of MoO<sub>2</sub> and Mo<sub>2</sub>C was further calculated through the reaction as follows:

$$2MoO_2 + 3C = Mo_2C + 2CO_2$$
 (1)

$2MoO_2 + O_2 = 2MoO_3$ (2)	ļ

 $Mo_2C + 4O_2 = 2MoO_3$  (3)



Fig. S2 SEM images of MoO<sub>2</sub>/Mo<sub>2</sub>C/C precursors before carbonation processes (a, b); SEM images

of  $MoO_2+Mo_2C$  mixtures blended together by grinding (c, d).



Fig. S3 Charge/discharge curves of  $MoO_2/Mo_2C/C$  electrodes with vary current densities from 0.1to 10.0 A g<sup>-1</sup>.



Fig. S4 Cycling performance of  $MoO_2/C$  (a) and  $Mo_2C/C$  (b) electrodes at 0.1 A g<sup>-1</sup>.



Fig. S5 Fitting curves of  $i/v^{1/2}$  responses plotted against  $v^{1/2}$  for cathodic (a) and anodic scans (b).



Fig. S6 XPS survey of the as-prepared  $MoO_2/Mo_2C/C$  electrodes after 200 cycles at the current density of 0.1 A g<sup>-1</sup>.



Fig. S7 Density of states of  $MoO_2$  (a) and  $Mo_2C$  (b).



Fig. S8 A unite cell of  $MoO_2$  (a) and corresponding differential electron density distribution (b).



Fig. S9 A unite cell of Mo<sub>2</sub>C (a) and corresponding differential electron density distribution (b).



Fig. S10 EIS curve of MoO<sub>2</sub> + Mo<sub>2</sub>C electrode before cycling.