## Controllable Synthesis of Layered K<sub>0.296</sub>Mn<sub>0.926</sub>O<sub>2</sub> to Assemble 2.4 V Aqueous Potassium-Ion Supercapacitors for Double High Device

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Notes

The authors declare no competing financial interest.

## **Figure captions**

**Figure.S1.** (a) (b) SEM images of the MnO<sub>2</sub> and as-prepared  $K_{0.296}Mn_{0.926}O_2$ , respectively; (c) AFM image of the as-prepared  $K_{0.296}Mn_{0.926}O_2$ ; (d) Obtained 3D image from the AFM image in Figure S1c.

Figure.S2. Powder XRD patterns of the commercial MnO<sub>2</sub>.

**Figure.S3.** XPS spectra analysis for the K 2p (a) and O 1s (b) of as-prepared  $K_{0.296}Mn_{0.926}O_2$ , respectively.

Figure.S4. XPS spectra of MnO<sub>2</sub>(a) and O 1s (b), respectively.

Figure.85. The EDX spectrum of  $K_{0.296}Mn_{0.926}O_2$ .

**Figure.S6.** (a) CV curves of commercial  $MnO_2$  at the scan rate between 10 and 80 mV s<sup>-1</sup>; (b) Galvanostatic charge-discharge curves of commercial  $MnO_2$  at the current densities between 1 and 5 A g<sup>-1</sup>.

Figure.S7. Density of states (DOS) of the MnO<sub>2</sub> and as-prepared K<sub>0.296</sub>Mn<sub>0.926</sub>O<sub>2</sub> using density functional theory (DFT)

**Figure.S8.** CV curves of MnO<sub>2</sub> and as-prepared  $K_{0.296}Mn_{0.926}O_2$  at the scan rate between 1 and 9 mV s<sup>-1</sup>, respectively.

**Figure.S9.** (a), (c) The contribution of the different energy storage mechanism of the  $MnO_2$  and  $K_{0.296}Mn_{0.926}O_2$  at 9 mV s<sup>-1</sup>; (b), (d) Percentage of diffusion-controlled and surface-capacitance for the  $MnO_2$  and  $K_{0.296}Mn_{0.926}O_2$  at various scan rates, respectively.

Figure S10. (a), (b) The SEM images of MnO<sub>2</sub> after cycling; (c), (d) EDS-mapping of Figure S9(b).

**Figure S11.** (a), (b) The SEM images of  $K_{0.296}Mn_{0.926}O_2$  after cycling.

**Figure S12.** The XRD spectrum of  $K_{0.296}Mn_{0.926}O_2$  after cycling.

**Figure.S13.** CV curves of  $MnO_2//AC$  at different potentials at a scan rate of 20 mV s<sup>-1</sup>.

**Figure.S14.** (a) CV curves of  $MnO_2//AC$  at the scan rate between 10 and 80 mV s<sup>-1</sup>; (b) Galvanostatic charge-discharge curves of  $MnO_2//AC$  at the current densities between 1 and 5 A g<sup>-1</sup>.

Figure. S15. (a), (b) The computational models of bulk  $MnO_2$  and  $K_{0.296}Mn_{0.926}O_2$ , respectively.

**Table S1.** The comparative table of morphology, specific surface areas and average pore diameters of our prepared products with previously

reported literature.



Figure.S1. (a) (b) SEM images of the  $MnO_2$  and as-prepared  $K_{0.296}Mn_{0.926}O_2$ , respectively; (c) AFM image of the as-prepared  $K_{0.296}Mn_{0.926}O_2$ ; (d) Obtained 3D image from the AFM image in Figure S1c.



Figure.S2. Powder XRD pattern of the commercial MnO<sub>2</sub>.



**Figure.S3.** XPS spectra analysis for the K 2p (a) and O 1s (b) of as-prepared  $K_{0.296}Mn_{0.926}O_2$ , respectively.



Figure.S4. XPS spectra of MnO<sub>2</sub>(a) and O 1s (b), respectively.



Figure.S5. The EDX spectrum of  $K_{0.296}Mn_{0.926}O_2$ .



**Figure.S6.** (a) CV curves of commercial  $MnO_2$  at the scan rate between 10 and 80 mV s<sup>-1</sup>; (b) Galvanostatic charge-discharge curves of commercial  $MnO_2$  at the current densities between 1 and 5 A g<sup>-1</sup>.



Figure.S7. Density of states (DOS) of the  $MnO_2$  and as-prepared  $K_{0.296}Mn_{0.926}O_2$  using density functional theory (DFT)



**Figure.S8.** (a), (b) CV curves of  $MnO_2$  and as-prepared  $K_{0.296}Mn_{0.926}O_2$  at the scan rate between 1 and 9 mV s<sup>-1</sup>, respectively.



**Figure.S9.** (a), (c) The contribution of the different energy storage mechanism of the  $MnO_2$  and  $K_{0.296}Mn_{0.926}O_2$  at 9 mV s<sup>-1</sup>; (b), (d) Percentage of diffusion-controlled and surface-capacitance for the  $MnO_2$  and  $K_{0.296}Mn_{0.926}O_2$  at various scan rates, respectively.



Figure S10. (a), (b) The SEM images of MnO<sub>2</sub> after cycling; (c), (d) EDS-mapping of Figure S9(b).



Figure S11. (a), (b) The SEM images of  $K_{0.296}Mn_{0.926}O_2$  after cycling.



Figure S12. The XRD spectrum of  $K_{0.296}Mn_{0.926}O_2$  after cycling.



Figure.S13. CV curves of MnO<sub>2</sub>//AC at different potentials at a scan rate of 20 mV s<sup>-1</sup>.



**Figure.S14.** (a) CV curves of  $MnO_2//AC$  at the scan rate between 10 and 80 mV s<sup>-1</sup>; (b) Galvanostatic charge-discharge curves of  $MnO_2//AC$  at the current densities between 1 and 5 A g<sup>-1</sup>.



Figure. S15. (a), (b) The computational models of bulk  $MnO_2$  and  $K_{0.296}Mn_{0.926}O_2$ , respectively.

 Table S1. The comparative table of morphology, specific surface areas and average pore diameters of our prepared products with previously reported literature.

Samples	Morphology	Specific surface area (m <sup>2</sup> g <sup>-1</sup> )	Average pore diameters (nm)	Device	Energy and power density	Reference
nanoporous carbon@K <sub>0.5</sub> Mn <sub>2</sub> O <sub>4</sub>	nanosheets	/	/	NCMO//NC	60 Wh kg <sup>-1</sup> at 1200 W kg <sup>-1</sup>	[30]
ultrathin MnO <sub>2</sub>	nanosheets	/	/	MnO <sub>2</sub> /carbo n fiber//graph ene/carbon fiber	27.2 Wh kg <sup>-1</sup> at 979.7 W kg <sup>-1</sup>	[31]
MnO <sub>2</sub> -CNT nanocomposite	nanoparticles	/	/	MnO <sub>2</sub> - CNT//activa ted carbon	25 Wh kg <sup>-1</sup> at 500 W kg <sup>-1</sup>	[32]
MnO <sub>2</sub> /graphene nano- composites	nanoparticles	332	/	MnO <sub>2</sub> /graph ene//graphe ne	35.2 Wh kg <sup>-1</sup> at 7.4 kW kg <sup>-1</sup>	[33]
graphene/MnO <sub>2</sub> composite	nanoparticles			graphene/M nO <sub>2</sub> //activat ed carbon nanofibers	51.1 Wh kg <sup>-1</sup> at 198 kW kg <sup>-1</sup>	[34]
graphene-MnO <sub>2</sub>	nanorods	43.8	2-5	graphene- MnO <sub>2</sub> //dens ely stacked graphene	62.4 Wh kg <sup>-1</sup> at 180 kW kg <sup>-1</sup>	[35]
K <sub>0.296</sub> Mn <sub>0.926</sub> O <sub>2</sub>	nanoparticles	52.4	3.7	K <sub>0.296</sub> Mn <sub>0.92</sub> <sub>6</sub> O <sub>2</sub> //AC	70 Wh kg <sup>-1</sup> at 1200 kW kg <sup>-1</sup>	This work