

## Hydrogel-derived VPO<sub>4</sub>/porous carbon framework for enhanced lithium and sodium storage

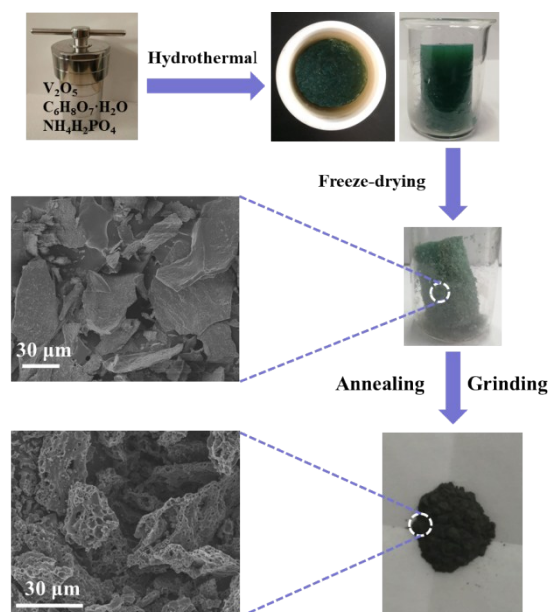
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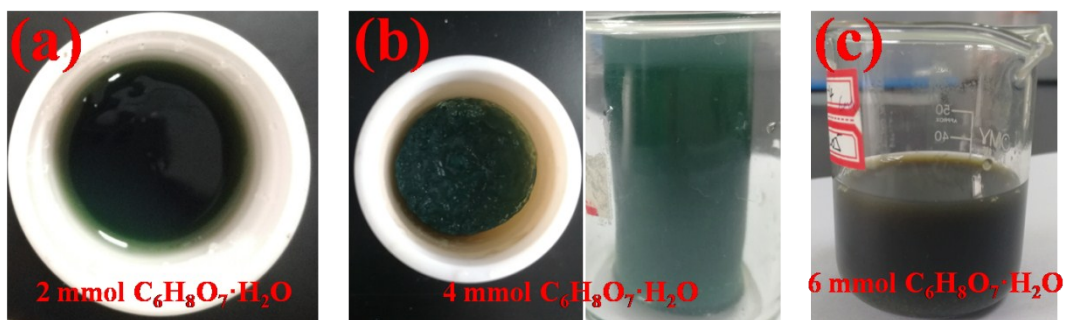
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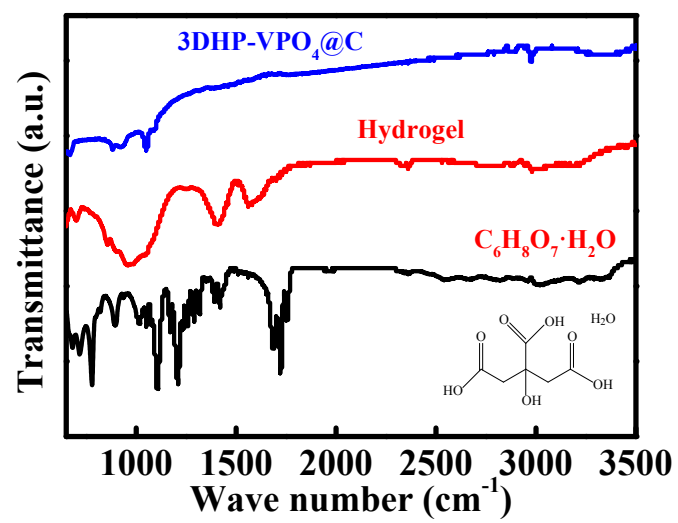
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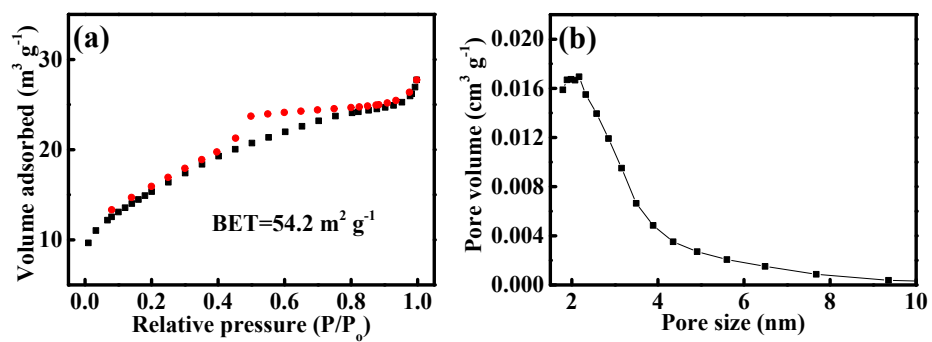
**Fig. S1.** Digital photographs and FESEM images of precursor sample material after freeze-drying and 3DHP-VPO<sub>4</sub>@C.



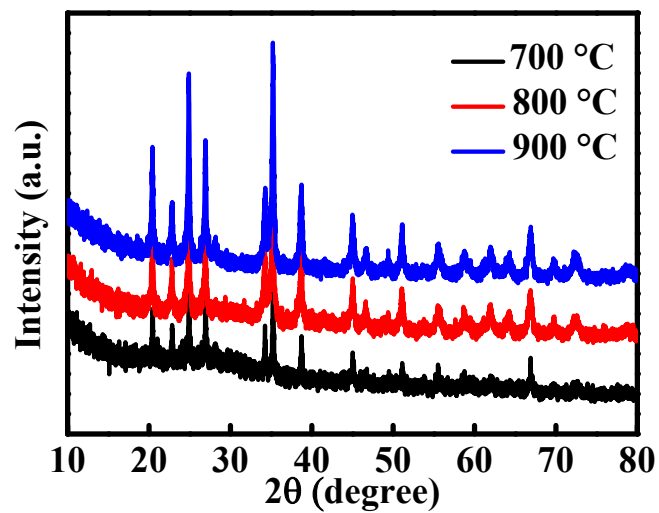
**Fig. S2.** Digital photographs of hybrid hydrogel obtained after hydrothermal treatment for different  $C_6H_8O_7 \cdot H_2O$  contents of (a) 2, (b) 4 and (c) 6 mmol.



**Fig. S3.** FT-IR spectras of C<sub>6</sub>H<sub>8</sub>O<sub>7</sub>·H<sub>2</sub>O, hydrogel and 3DHP-VPO<sub>4</sub>@C.

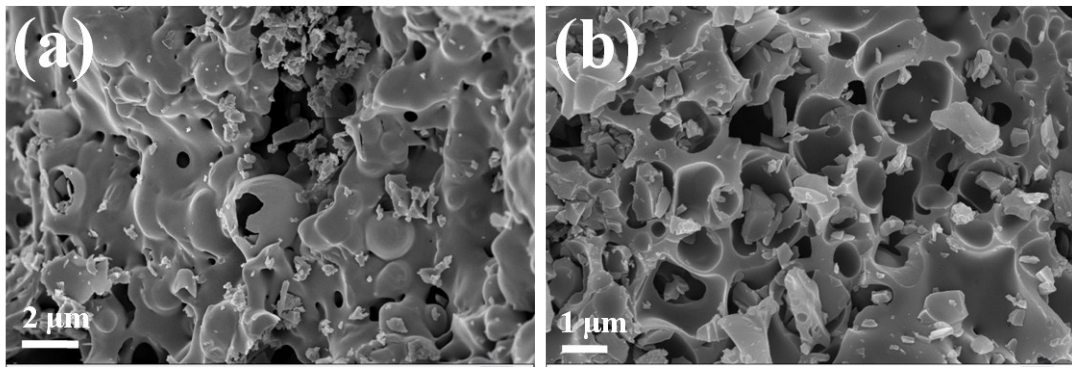


**Fig. S4.** N<sub>2</sub> adsorption-desorption isotherm (a) and pore size distribution (b) of 3DHP-VPO<sub>4</sub>@C according to the NLDFT model.

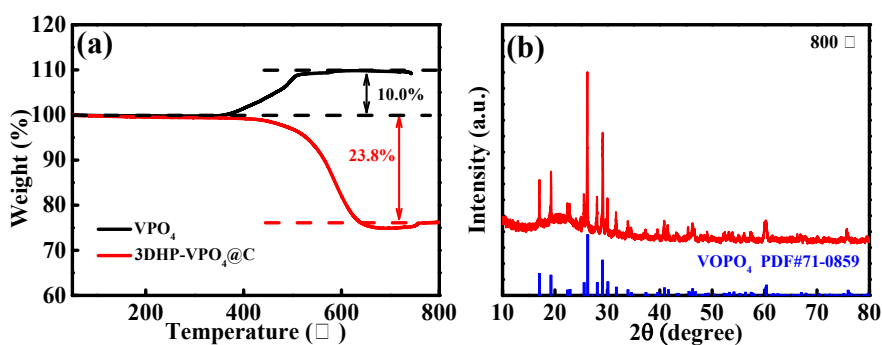


**Fig. S5.** XRD patterns of VPO<sub>4</sub>@C calcined at 700, 800 and 900 °C.

All XRD peaks of 3DHP-VPO<sub>4</sub>@C, VPO<sub>4</sub>@C-700 and VPO<sub>4</sub>@C-900 can be assigned to the orthorhombic VPO<sub>4</sub> phase (PDF#076-2023).



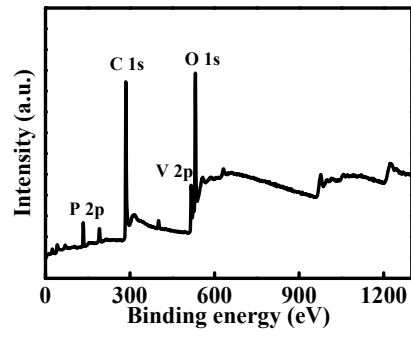
**Fig. S6.** FESEM images of VPO<sub>4</sub>@C calcined at 700 (a) and 900 °C (b).



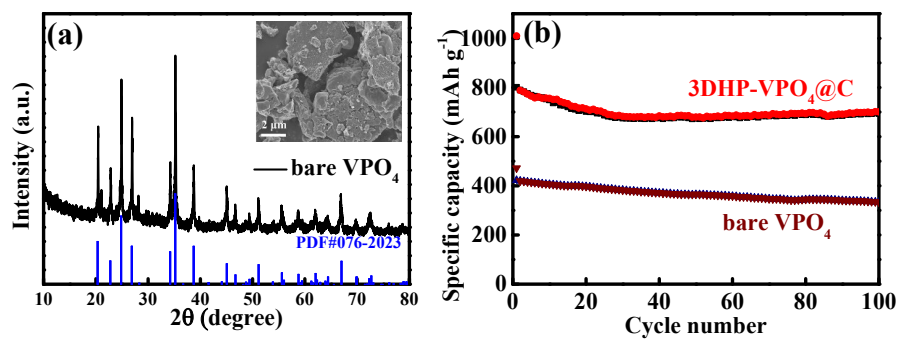
**Fig. S7.** (a) TGA curves of 3DHP-VPO<sub>4</sub>@C and VPO<sub>4</sub> in the temperature range of 30-800 °C in the flowing of air atmosphere; (b) XRD pattern for final product of VPO<sub>4</sub> sintered at 800 °C under air atmosphere.

The TGA test is operated in air flow to calculate the carbon content of 3DHP-VPO<sub>4</sub>@C. The apparent increasing mass of VPO<sub>4</sub> is corresponded to the oxidation of VPO<sub>4</sub> to VOPO<sub>4</sub> (**Fig. S6b**). In contrast, the rapid mass loss for 3DHP-VPO<sub>4</sub>@C is related to the removal of carbonous materials. The carbon content in 3DHP-VPO<sub>4</sub>@C is determined to be 33.8 wt%.





**Fig. S8.** XPS curves of survey spectrum of 3DHP-VPO<sub>4</sub>@C.



**Fig. S9.** (a) XRD pattern of bare VPO<sub>4</sub> and the inset is its FESEM image; (b) The cycling performances of 3DHP-VPO<sub>4</sub>@C and bare VPO<sub>4</sub> at 100 mA g<sup>-1</sup>.

**Table S1.** Comparison of electrochemical performance between the current electrode and state-of-the-art VPO<sub>4</sub>-based electrodes.

VPO <sub>4</sub> -based electrodes	Current density (mA g <sup>-1</sup> )/Rate	Initial specific capacity (mA h g <sup>-1</sup> )	Cycle number	Specific capacity (mA h g <sup>-1</sup> )	Ref.
VPO <sub>4</sub> /C/Ag	0.2 C	857.8	100	324.2	[S1]
VPO <sub>4</sub> /C/3DG	0.2 C	976.8	30	632	[S2]
	5 C	369	100	338.8	
a-VPO <sub>4</sub> /C	200	1094.6	50	804.5	[S3]
Nano-sheets-VPO <sub>4</sub>	0.05 C	788.7			[S4]
Core-shell VPO <sub>4</sub> /C	20	887.3	30	343	[S5]
MVHP-VPO <sub>4</sub> @C NSs	100	943	100	630	[S6]
VPO <sub>4</sub> @C/rGO	100	1074	100	395.3	[S7]
VPO <sub>4</sub> /rGO	100	567	100	475	[S8]
Current work	100	1009.4	100	700.5	
(3DHP-VPO <sub>4</sub> @C)	2000	542.1	2000	288.5	

**Table S2.** Impedance parameters of 3DHP-VPO<sub>4</sub>@C, VPO<sub>4</sub>@C-700 and VPO<sub>4</sub>@C-900 electrodes fitted with the circuit model of R(QR)(QR)W.

Electrodes	R <sub>s</sub> (Ω)	R <sub>ct</sub> (Ω)	Z <sub>w</sub> (Ω)
3DHP-VPO <sub>4</sub> @C	3.5	20.4	1.5
VPO <sub>4</sub> @C-700	7.4	46.7	5.4
VPO <sub>4</sub> @C-900	5.2	27.6	2.8

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