

Supporting Information for

**Unleashing the Ultra-fast Sodium Ion Storage Mechanisms in
Interface-engineered Monolayer MoS₂/C Interoverlapped
Superstructure with Robust Charge Transfer Networks**

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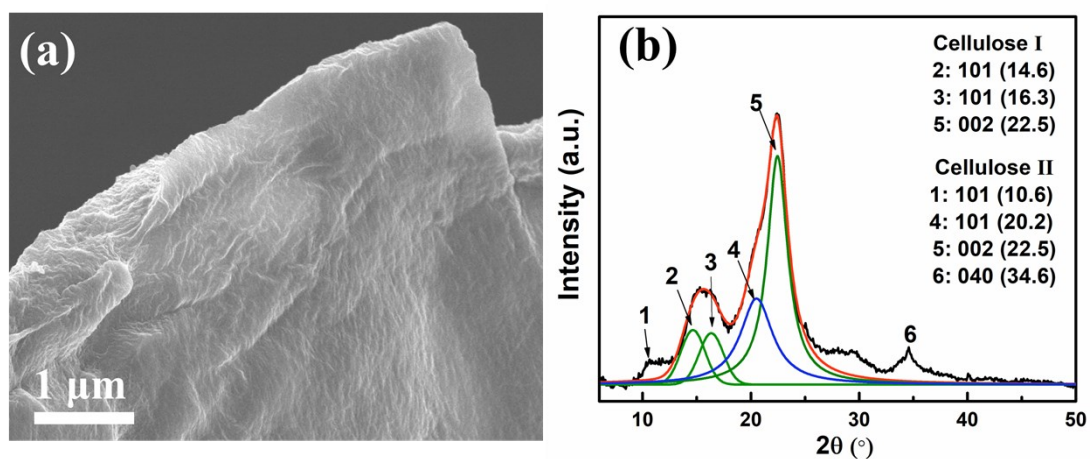


Fig. S1. (a) SEM image and (b) XRD patterns of colloidal bagasse cellulose sheets.

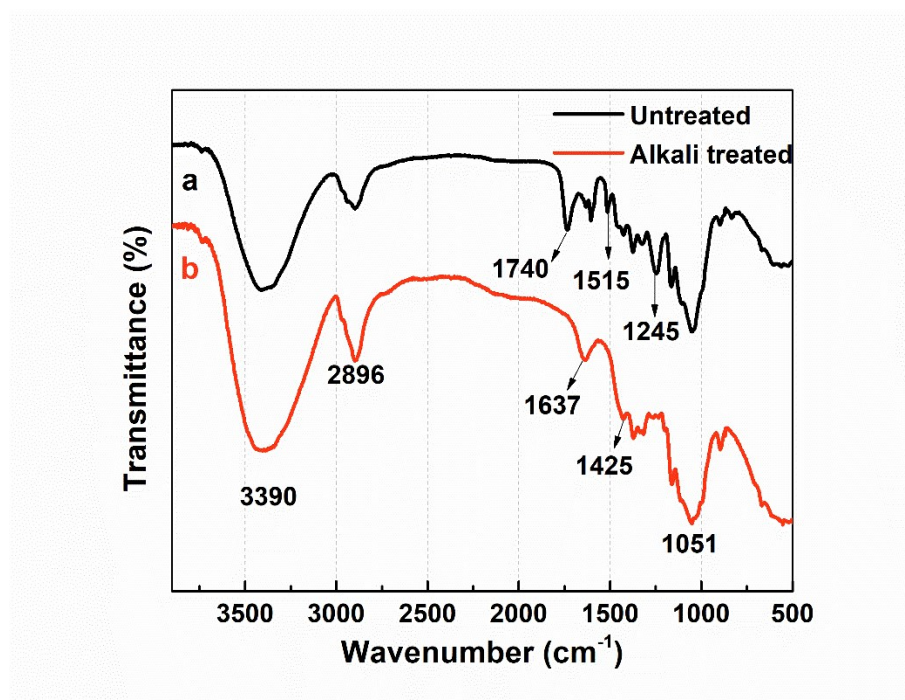


Fig. S2. FTIR spectra of colloidal bagasse cellulose sheets.

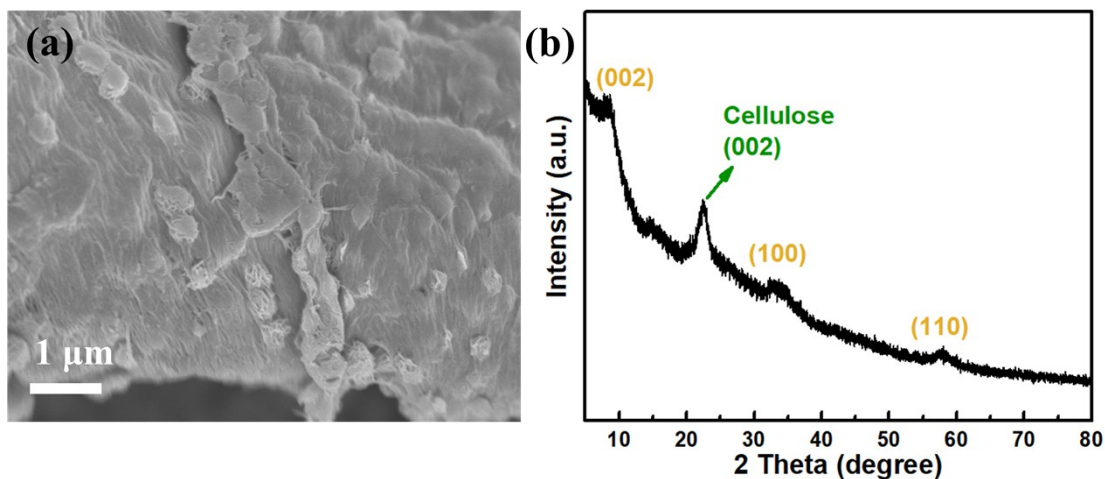


Fig. S3. (a) SEM image and (b) XRD patterns of MoS_2 /carbohydrate nanoflowers embedded in the colloidal bagasse sheets (MoS_2 /carbohydrate-CBS).

As shown in Fig. S3b, the diffraction peaks at 22.6° is ascribed to (002) planes of cellulose. The diffraction peaks at 32.6° and 57.7° can be indexed to (100) and (110) planes of 2H- MoS_2 (JCPDS card No. 06-0097), respectively. The new peaks at 8.6° ($d = 1.02 \text{ nm}$) is ascribed to (002) plane. Compared to pristine 2H- MoS_2 , the expanded interlayer is probably caused by the insertion of carbohydrate molecules into the adjacent MoS_2 monolayers.

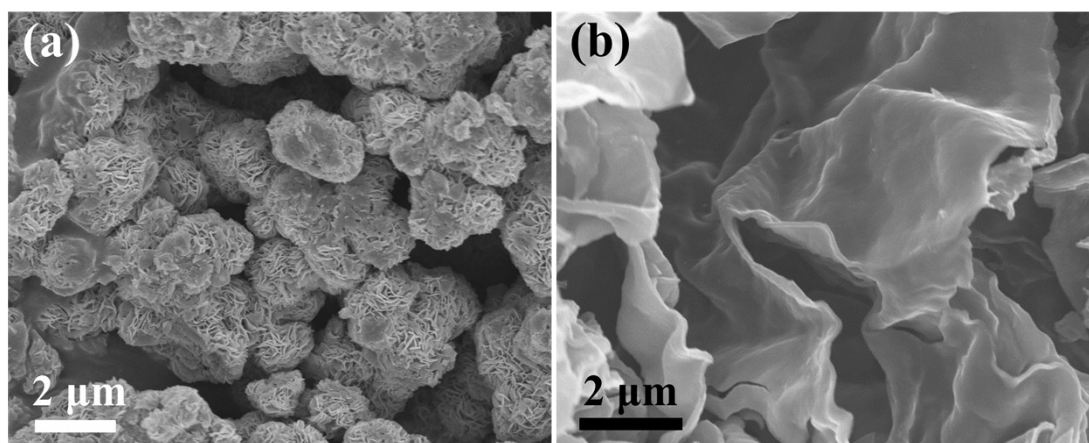


Fig. S4. SEM images of (a) the pristine MoS_2 and (b) the pristine carbon.

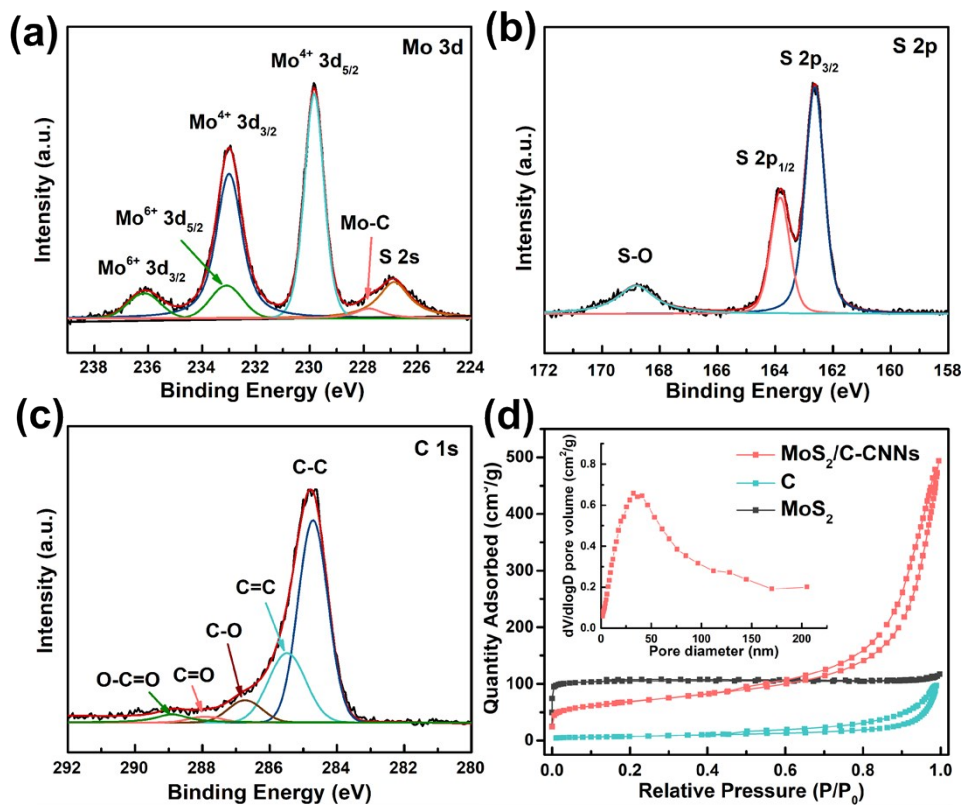


Fig. S5. (a-c) High-resolution XPS spectra of Mo 3d, S 2p and C 1s core level of MoS₂/C-CNNs; (d) N₂ adsorption-desorption isotherms of the MoS₂/C-CNNs, bulk MoS₂ and pure C samples, and inset is the corresponding pore size distribution of MoS₂/C-CNNs.

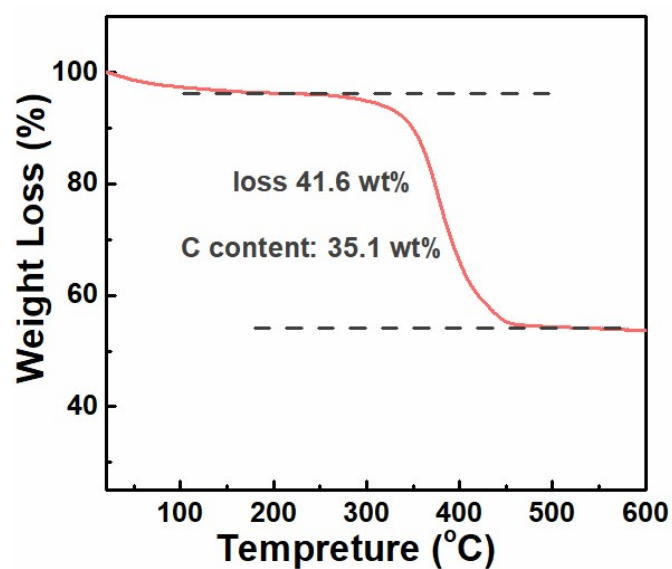


Fig. S6. TGA measurement of MoS₂/C-CNNs.

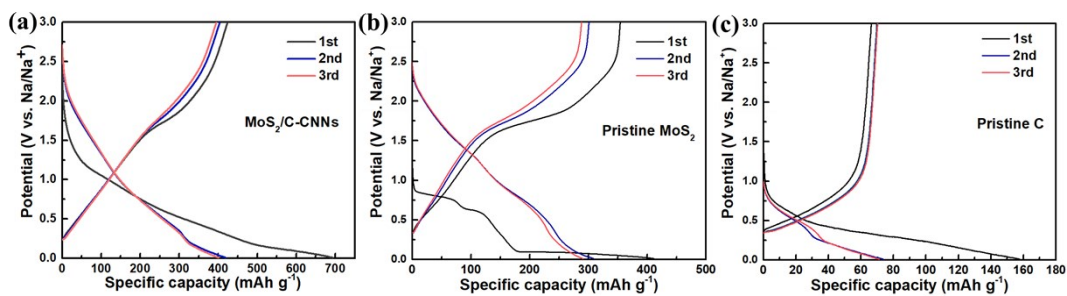


Fig. S7. The first three charge/discharge voltage profiles of (a) the MoS₂/C-CNNs, (b) the pristine MoS₂ and (c) the pristine C electrode at 200 mA g⁻¹ between 0.01 and 3.0 V.

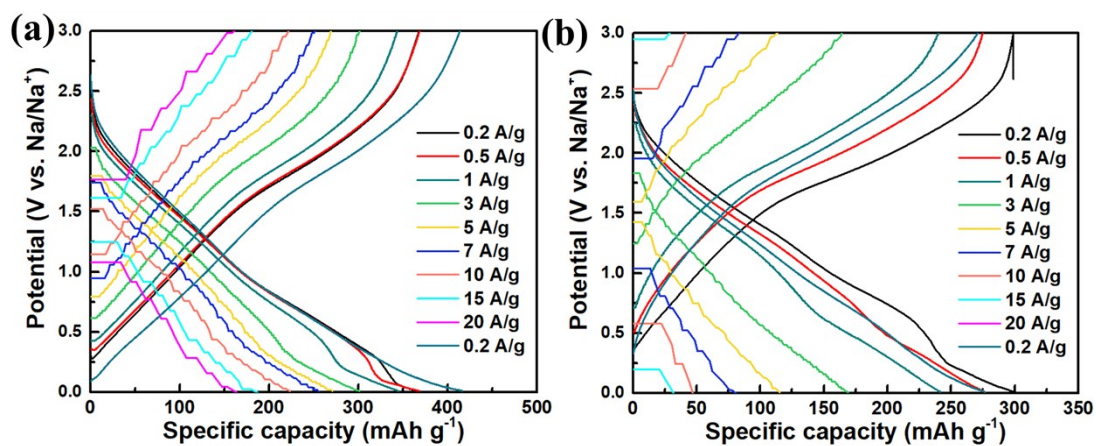


Fig. S8. The charge/discharge voltage profiles of (a) the MoS₂/C-CNNs electrode and (b) the pristine MoS₂ electrode at different current densities.

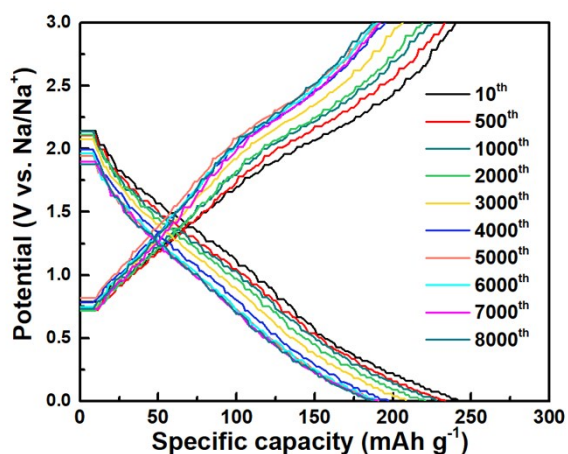


Fig. S9. The charge/discharge voltage profiles of the MoS₂/C-CNNs electrode at different cycles under 5 A g⁻¹.

Table S1. Electrochemical performances of previously reported MoS₂-based SIB anodes in comparison with our MoS₂/C-CNNs

Materials	Cycling data	Rate capability	reference
MoS ₂ nanosheets	267/125th@1A/g	226@5A/g	ACS Nano, 13 (2019), 5533-5540.
MoS ₂ nanosheets	324/200th@1A/g	253@2A/g	Nano Energy 61 (2019) 361-369.
MoS ₂ /C hollow nanospheres	262/600th@2A/g	211@10A/g	Nano Energy 51 (2018) 546-555.
MoS ₂ @carbon nanospheres	150.2/300th@5A/g	204@5A/g	Nano Energy 62 (2019) 299-309.
MoS ₂ @carbon nanowall	265/1000th@1A/g	235@2A/g	Adv. Funct. Mater. 27 (2017) 1702116.
MoS ₂ /carbon framework	360/100th@0.1A/g	171@5A/g	Nano Energy 48 (2018) 526-535.
MoS ₂ /N-Graphene	245/1300th@1A/g	153@20A/g	Adv. Energy Mater. 2018, 8, 1703300
PCNF@MoS ₂ @PEDOT	410/1000th@2A/g	313@20A/g	Energy Storage Mater. 2020, 25, 114
MoS ₂ /carbon framework	183.5/3000th@2A/g	~195@15A/g	J. Mater. Chem. A, 2018, 6, 14742
MoS₂/C-CNNs	193.5/8000th@5A/g	223@10A/g 159@20A/g	Our work

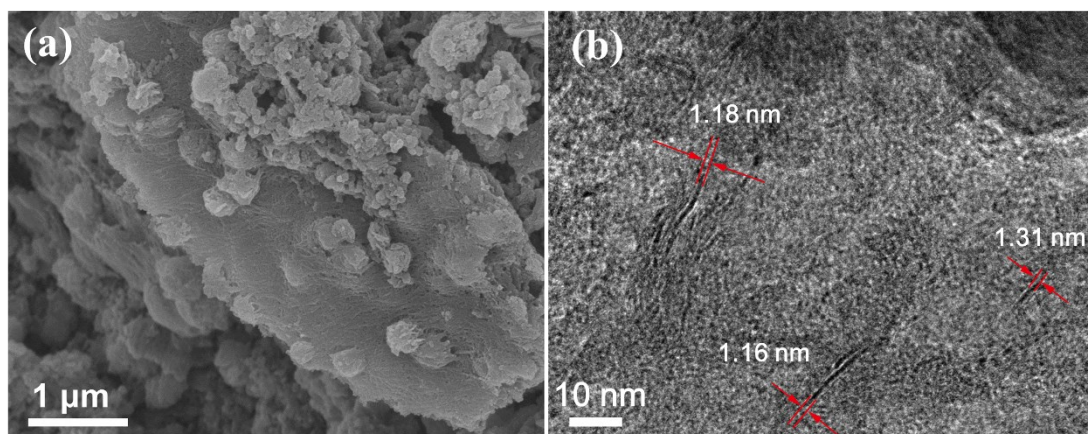


Fig. S10. (a) SEM image of MoS₂/C-CNNs electrode after 1000 cycles; (b) HRTEM image of MoS₂/C-CNNs electrode after 5 cycles.

Table S2. EIS fitting results of pristine MoS₂ and MoS₂/C-CNNs.

	R_s/Ω	R_f/Ω	R_{ct}/Ω	Z_w-R/Ω
Pristine MoS ₂ after 10 th	6.01	6.13	256.8	454
MoS ₂ /C-CNNs after 10 th	5.39	15.42	50.70	88.3
MoS ₂ /C-CNNs after 1000 th	6.71	12.01	50.46	90.0
MoS ₂ /C-CNNs after 2000 th	6.91	15.29	43.23	129.3
MoS ₂ /C-CNNs after 4000 th	6.52	22.71	37.92	133.4
MoS ₂ /C-CNNs after 6000 th	5.97	26.77	35.24	111.7

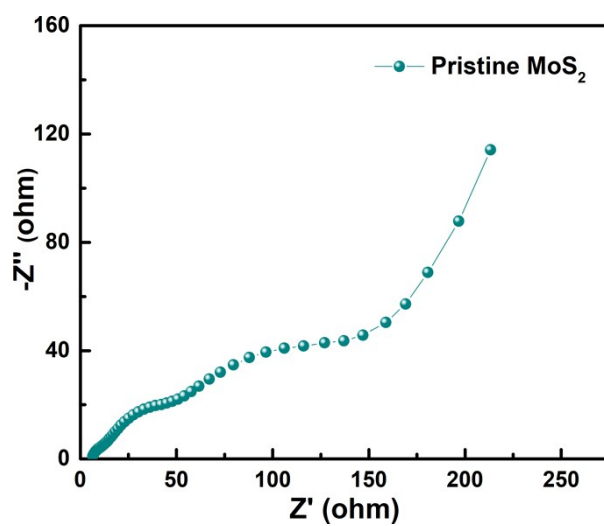


Fig. S11. Nyquist plots of pristine MoS₂ electrode after the 10th circles.

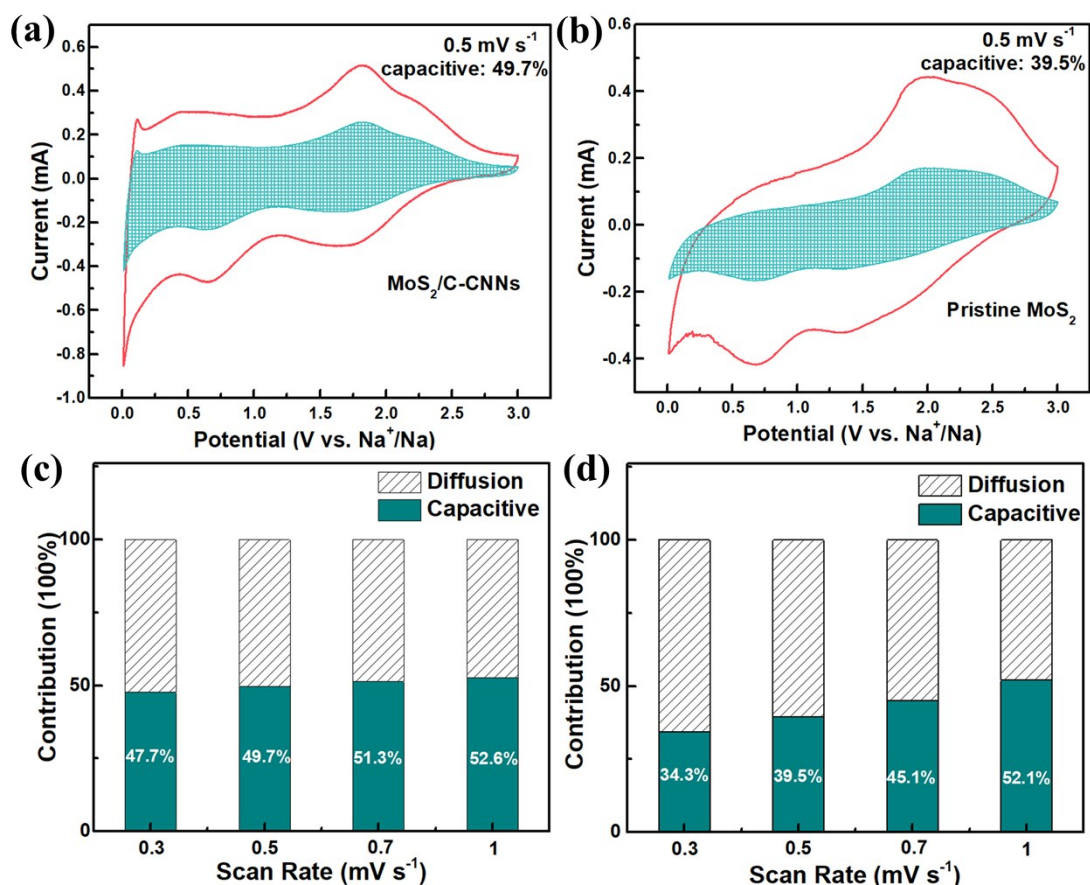


Fig. S12. Separation of the capacitive and diffusion currents in the MoS₂/C-CNNs (a) and the MoS₂ (b) at a scan rate of 0.5 mV s⁻¹, respectively; contribution ratio of the capacitive charge versus scan rate of the MoS₂/C-CNNs (c) and the MoS₂ (d).

The capacitive effect and the diffusion-controlled Na⁺ intercalation effects to the sodium storage can be expressed by the equation (Energy Storage Mater., 2018, 14, 129-135): $i(V) = k_1v + k_2v^{1/2}$.

$i(V)$ corresponds to the current value at the voltages, v refers to the scan rate, k_1v refers to the contribution of the capacitive effect provided by the surface-adsorbing charge and $k_2v^{1/2}$ refers to the solid diffusion control with intercalated/de-intercalated charges.

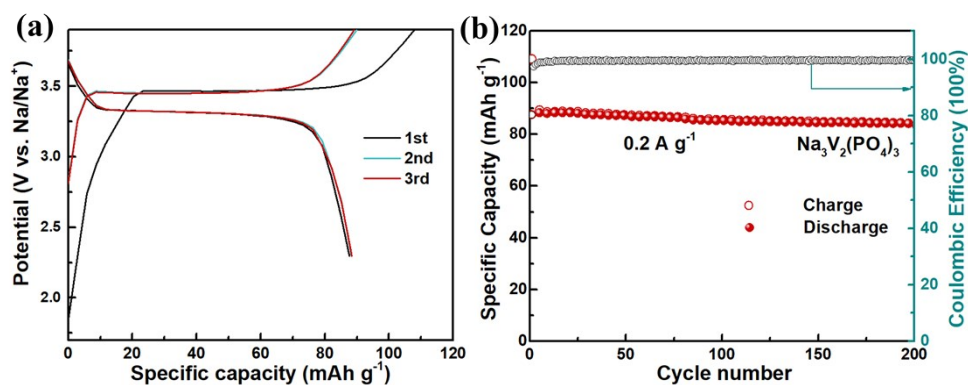


Fig. S13. (a) The first three charge/discharge voltage profiles of the $\text{Na}_3\text{V}_2(\text{PO}_4)_3$ at 0.2 A g^{-1} between 2.3 and 3.9 V; (b) Cycling performance/Coulombic efficiency of the $\text{Na}_3\text{V}_2(\text{PO}_4)_3$ at 0.2 A g^{-1} .

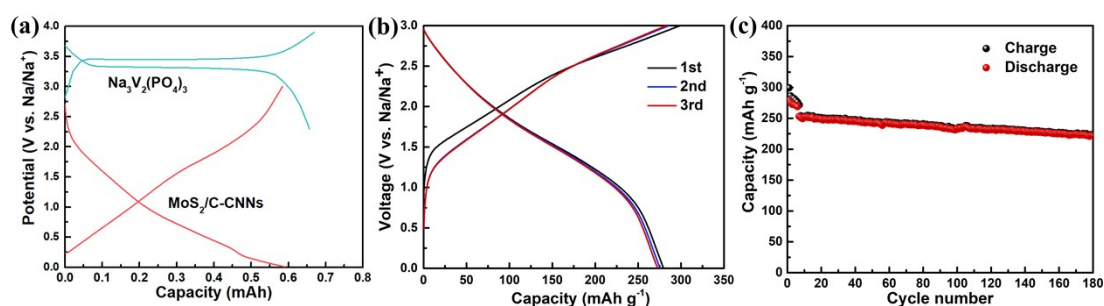


Fig. S14. (a) Charge-discharge profiles of $\text{MoS}_2/\text{C-CNNs}$ and $\text{Na}_3\text{V}_2(\text{PO}_4)_3$ half cells at a current density of 0.2 A g^{-1} ; (b) The charge and discharge profiles of a full-cell at 0.2 A g^{-1} according to the mass of anode materials; (c) Cycling performance of one full-cell at 1 A g^{-1} after first 7 cycles of low current density (0.2 A g^{-1}).