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Supporting Information

Hierarchical Architecture composite of N-doped Hollow Polyhedrons

Anchored on Ti3C2T^X Nanosheets for Advanced Lithium-Ion and

Sodium-Ion Capacitors

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1.1 Materials

Cobalt nitrate hexahydrate $(Co(NO₃)₂·6H₂O, \geq 99.0%)$, zinc nitrate hexahydrate $(Zn(NO₃)₂·6H₂O, \geq 99.0%)$, Dimethylimidazole $(C₄H₆N₂, \geq 99.8%)$, Hydrochloric acid (HCl, 12 M, 37 wt.%), Sulphuric acid (H₂SO₄, 95.0%~98%) and Methanol (CH₃OH, ≥99.8%) were purchased from Sinopharm Chemical Reagent. Titanium-aluminium carbon (Ti₃AlC₂, \geq 99.9%) was purchased from Yiyi Science and Technology, Jilin Province, and Lithium fluoride (LiF, ≥98.0%) was purchased from Shanghai Aladdin. The materials and reagents used in this work can be used without secondary purification and are of analytical grade.

Fig. S1 Destroyed Co@NC images, (a) SEM, (b) TEM

As can be seen from the figure, without the protection of the outermost Zn atoms, the hollow polyhedral structure is damaged by the haphazardly growing CNTs and the shell ruptures.

Fig. S2 Aperture distribution of Co@NC and Co@NC/Ti₃C₂T_x

Fig. S3 Co@NC and Co@NC/Ti₃C₂T_x electrode materials Amplified EIS curve for the high frequency region

Fig. S4 EIS plots of LIB and SIB of $Co@NC/Ti_3C_2T_x$ **electrode materials**

In Fig. S4, the 45 degrees in the low-frequency region of the EIS curves of $Co@NC/Ti_3C_2T_x$ LIB and SIB indicate the battery-like characteristics of the materials. The specific fitted values are shown in Table S1.

Fig. S5 Specific capacity plots of AC electrode materials in LIB and SIB

In Fig. S5, the discharge specific capacity values are 132 mAh/g for LIB(AC) and 101 mAh/g for SIB(AC).

Fig. S6 CV matching plot for $Co@NC/Ti_3C_2T_x//AC$ LIHC and SIHC devices

Fig. S7 Specific capacitance value plots for $\text{Co}(\mathbb{Q})\text{NC}/\text{Ti}_3\text{C}_2\text{T}_3$ //AC LIHC and SIHC devices

Fig. S7 shows the specific capacitance values of the $Co@NC/Ti_3C_2T_x//AC$ devices. Specifically, the specific capacitance values in the LIHC at current densities of 0.08, 0.12, 0.2, 0.4, 0.8, 2, and 5 A/g have 93, 63, 58, 56, 53, 41, and 28 F/g. The corresponding specific capacitance values for the SICH are 64, 53, 51, 50, 45, 25, and 13 F/g.

Fig. S8 EIS diagrams for Co@NC/Ti3C2Tx//AC LIHC and SIHC devices

In Fig. S8, the 45 degrees in the low frequency region of the EIS curves for $Co@NC/Ti_3C_2T_x//AC LHC$ and SIHC indicate the battery-like characteristics of the materials. The fitted values are shown in Table S1.

Electrode materials	$Co@NC/Ti_3C_2T_x$	Co@NC	LIB	SIB	LIC	SIC
R_S (ohm-cm ²)	0.78	0.94	4.9	3.1	21.7	17.0
R_{CT} (ohm-cm ²)	0.0073	0.01	45.6	122.1	25.0	91.4
$C1$ (F/cm ²)	0.020	0.045	$2.3E-4$	$2.4E - 4$	3.8E-5	8.6E-5
$C2$ (F/cm ²)	0.39	0.22	0.012	8.5E-4	0.011	$1.5E-3$
Warburg. $Yo(S\text{-}sec^{0.5})$	0.76	0.29	0.05	0.08	0.041	5.6E-3
R_1 (ohm-cm ²)	277	425	283	723	290	288
Error	1.21%	1.12%	$<1.35\%$	$<1.17\%$	$< 0.86\%$	$<1.31\%$

Table S1. EIS data. (Three repeated experiments were conducted.)

Error estimates evaluation

The error estimates are a description of the experimental effectiveness of the corresponding data. Suppose we have done n repeated experiments and get the datas Xi (i = 1,2, ..., n), and the error bars are calculated as follows:

$$
a = \frac{1}{n} \sum_{1}^{n} Xi
$$
\n
$$
error bar = \pm \sqrt{\sum_{1}^{n} (Xi - a)^{2}}
$$
\n
$$
n - 1
$$
\n(1)

Electric charge matching:

According to the principle of charge balance, the mass ratio between the cathode and the anode is 5:1. The formula and details are shown below:

$$
Q^+ = Q^- \mathit{=} It
$$

$$
\Delta E \cdot m \cdot C_{electrode} = \Delta E + m + C_{electrode} +
$$

Where, Q^+ , Q^- for the cathode and anode electrode charge (C), I is current (A), t for the time (s), ΔE_+ , ΔE_- for the positive and negative electrodes voltage window (V), m_{+} , m_r for the positive and negative electrode active material loading (g), $C_{electrodef}$ and Celectrode- for the positive and negative electrodes of the mass ratio of capacitance (mAh/g) .