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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, \geq 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, \geq 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/Ti_3C_2T_x



Fig. S3 Co@NC and Co@NC/Ti $_3C_2T_x$ electrode materials Amplified EIS curve for the high frequency region



Fig. S4 EIS plots of LIB and SIB of Co@NC/Ti₃C₂T_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 Co@NC/Ti₃C₂T_x//AC LIHC and SIHC devices

Fig. S7 shows the specific capacitance values of the Co@NC/Ti₃C₂T_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/Ti₃C₂T_x//AC LIHC and SIHC devices

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

Electrode materials	Co@NC/Ti ₃ C ₂ T _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-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:

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

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:

$$Q^+ = Q^- = It$$

$$\Delta E_{\cdot} m_{\cdot} C_{electrode} = \Delta E_{+} m_{+} C_{electrode+}$$

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_- for the positive and negative electrode active material loading (g), $C_{electrode+}$ and $C_{electrode-}$ for the positive and negative electrodes of the mass ratio of capacitance (mAh/g).