

## Supporting Information

# **In-situ Coupled Nickel-based Layered Double Hydroxides with MXene to Enhance the Supercapacitor Performance**

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## Results and Discussion

### 1. Composition, Structures and Morphologies Characterization

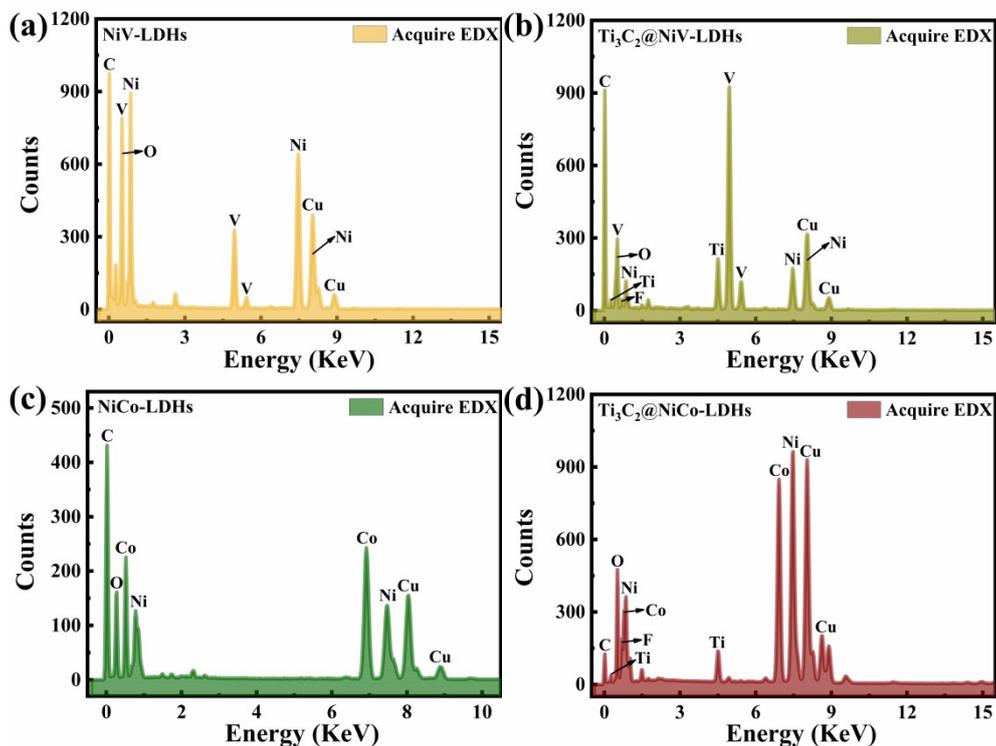


Fig. S1. The EDX spectrum of (a) NiV-LDHs, (b)  $\text{Ti}_3\text{C}_2@\text{NiV-LDHs}$ , (c) NiCo-LDHs and (d)  $\text{Ti}_3\text{C}_2@\text{NiCo-LDHs}$ .

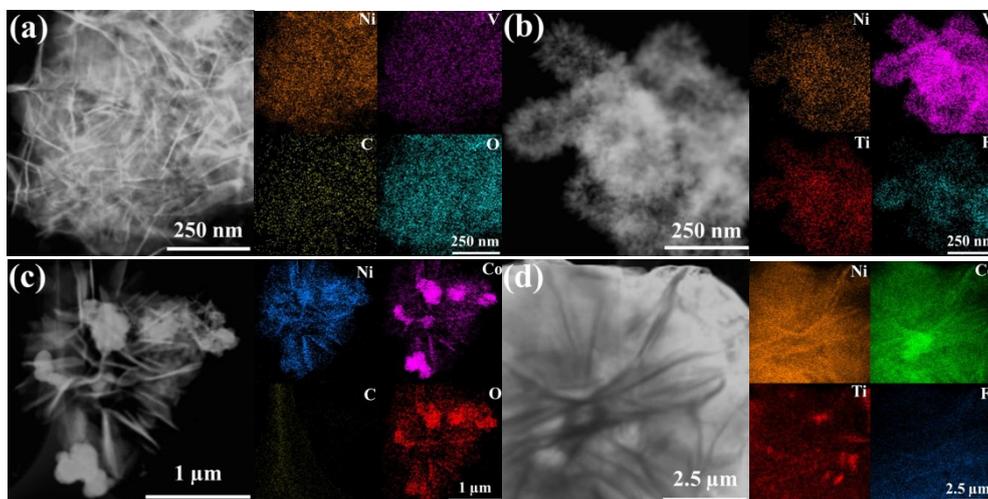
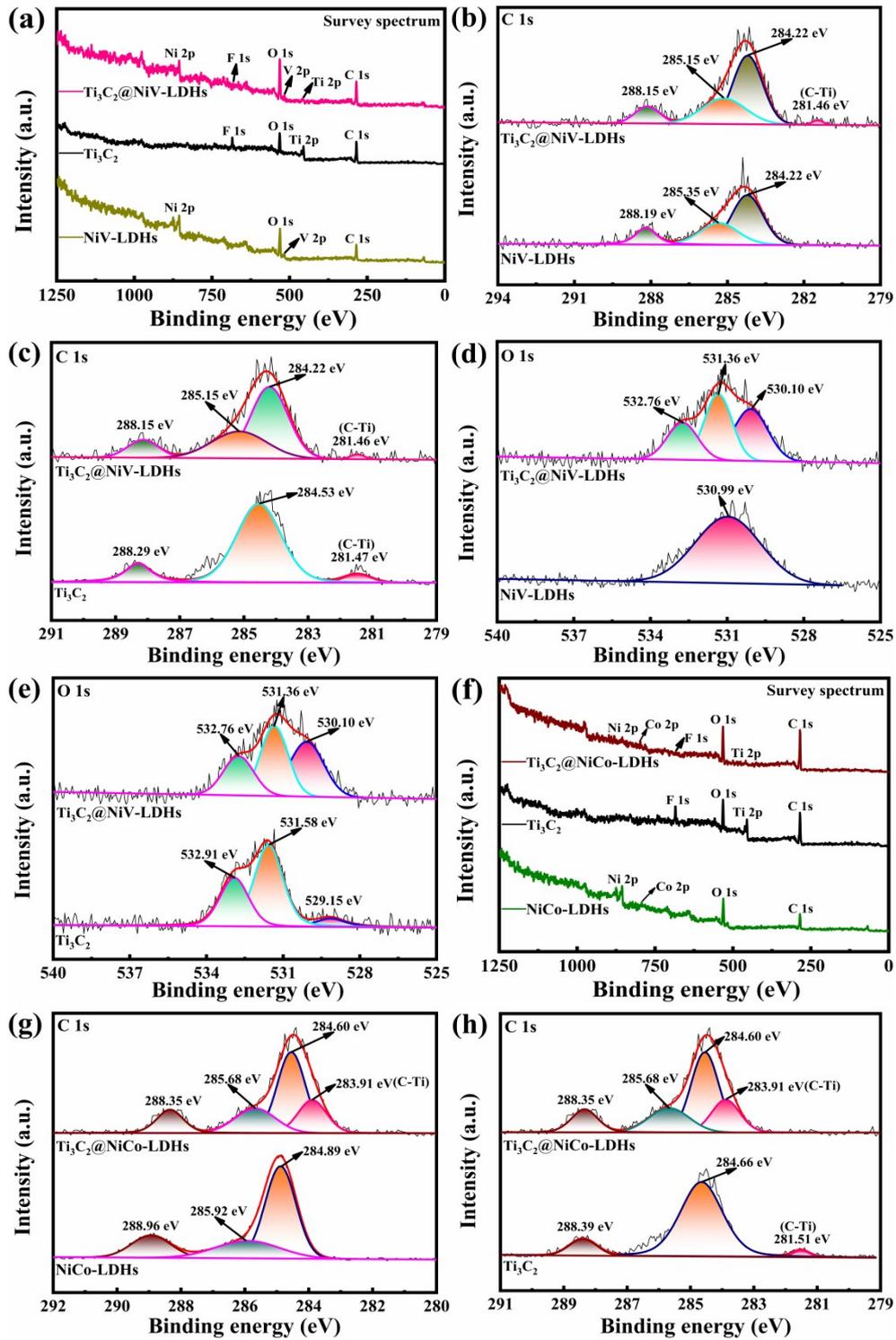
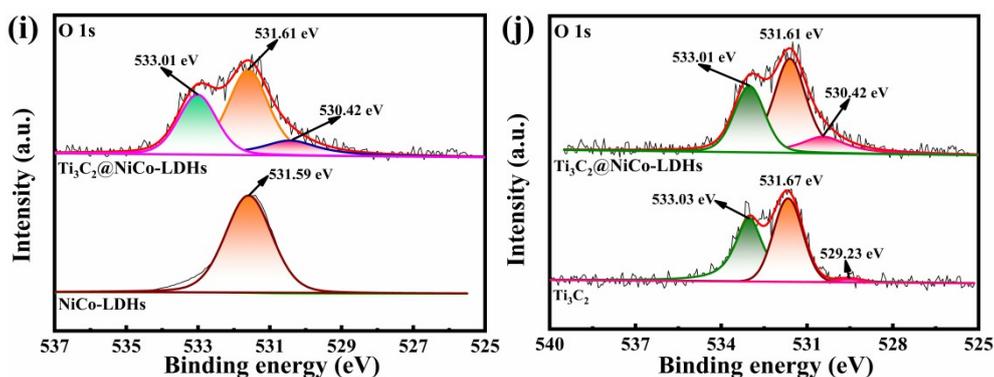


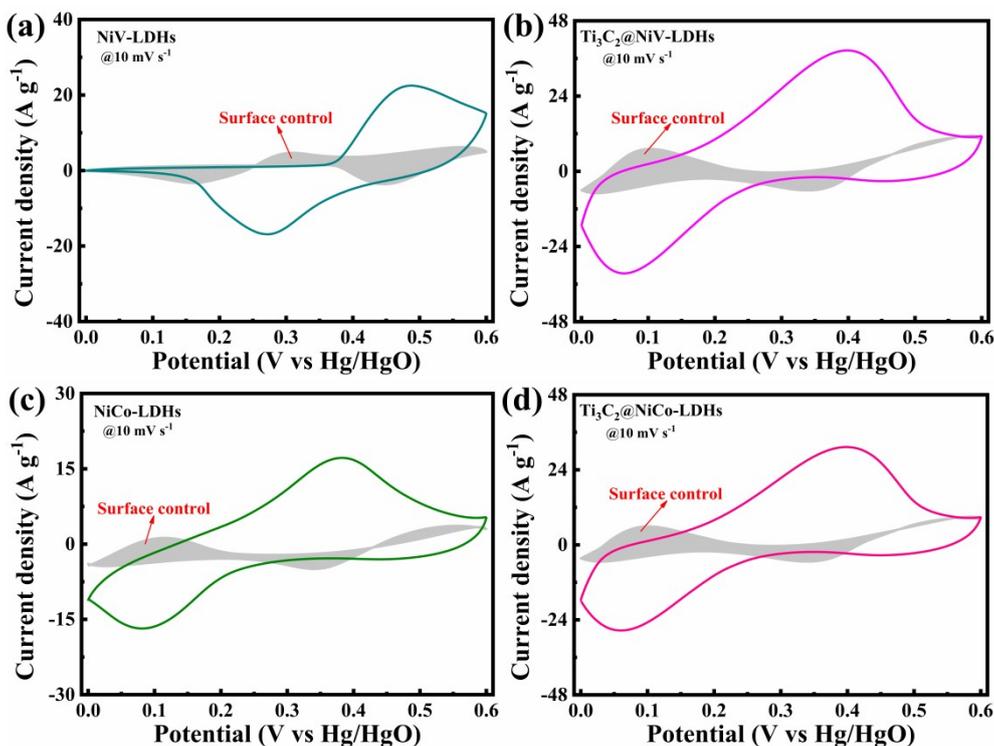
Fig. S2. Elements mapping of (a) NiV-LDHs, (b)  $\text{Ti}_3\text{C}_2@\text{NiV-LDHs}$ , (c) NiCo-LDHs and (d)  $\text{Ti}_3\text{C}_2@\text{NiCo-LDHs}$ .



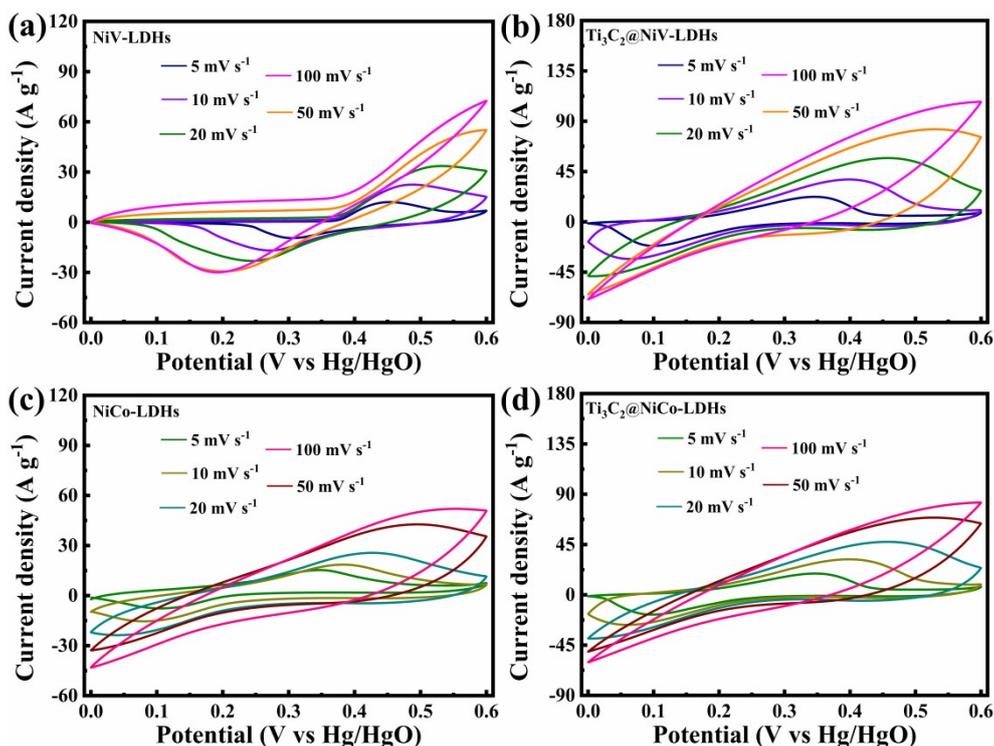


**Fig. S3.** The XPS spectra of the NiV-LDHs,  $\text{Ti}_3\text{C}_2$  and the  $\text{Ti}_3\text{C}_2@\text{NiV-LDHs}$ : (a) Survey spectrum, (b) C 1s, (c) C 1s, (d) O 1s, (e) O 1s, respectively. (f) Survey spectrum of NiCo-LDHs,  $\text{Ti}_3\text{C}_2$  and  $\text{Ti}_3\text{C}_2@\text{NiCo-LDHs}$ . (g) C 1s XPS spectrum of NiCo-LDHs and  $\text{Ti}_3\text{C}_2@\text{NiCo-LDHs}$ . (h) C 1s XPS spectrum of  $\text{Ti}_3\text{C}_2$  and  $\text{Ti}_3\text{C}_2@\text{NiCo-LDHs}$ . (i) O 1s XPS spectrum of NiCo-LDHs and  $\text{Ti}_3\text{C}_2@\text{NiCo-LDHs}$ . (j) O 1s XPS spectrum of  $\text{Ti}_3\text{C}_2$  and  $\text{Ti}_3\text{C}_2@\text{NiCo-LDHs}$ .

## 2. Three-Electrode System

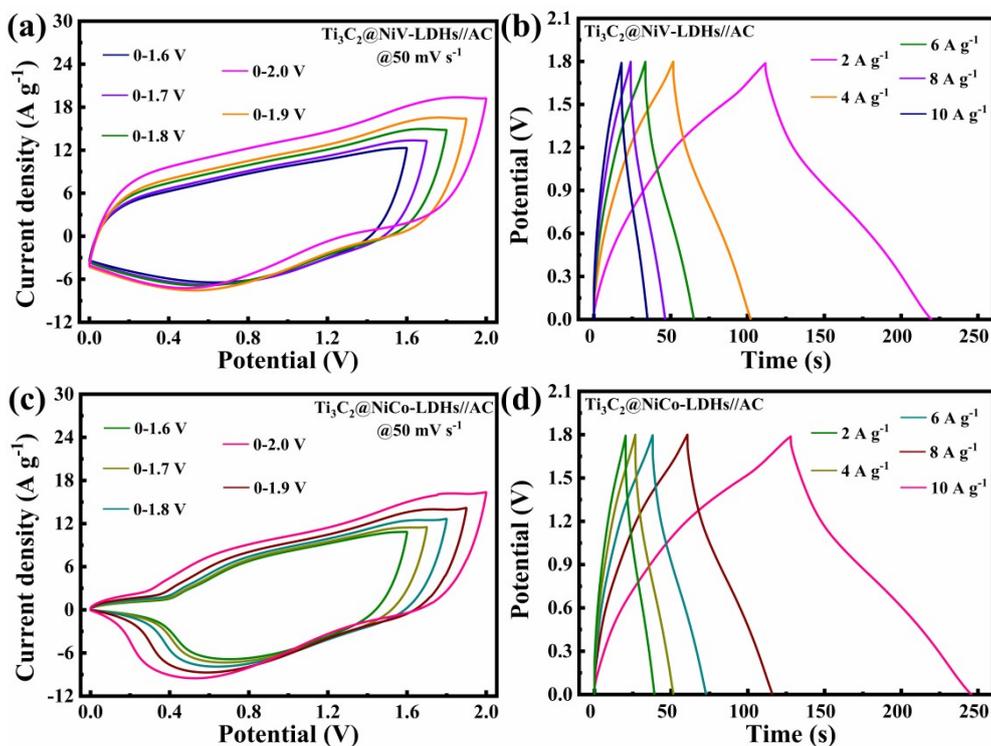


**Fig. S4.** (a) The shaded area represents the capacitance contribution of NiV-LDHs at  $10 \text{ mV s}^{-1}$ . (b) The shaded area indicates the capacitive contribution of  $\text{Ti}_3\text{C}_2@\text{NiV-LDHs}$  at  $10 \text{ mV s}^{-1}$ . (c) The gray area represents the portion of the NiCo-LDHs surface control contribution. (d) The gray area represents the portion of the  $\text{Ti}_3\text{C}_2@\text{NiCo-LDHs}$  surface control contribution.



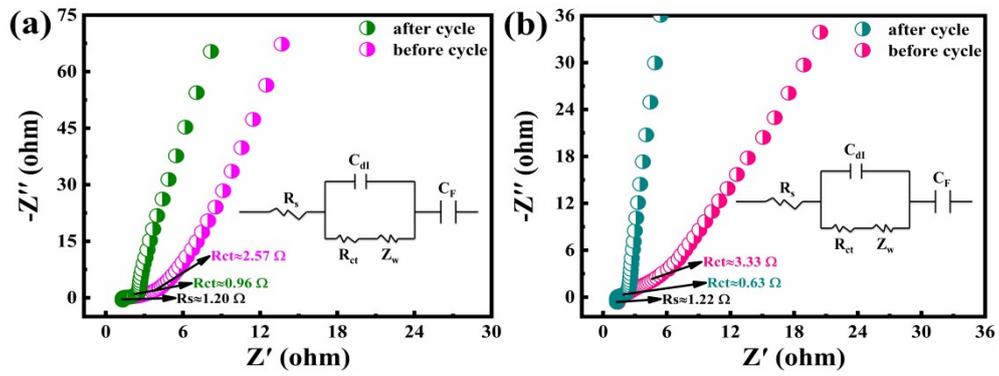
**Fig. S5.** (a) The cyclic voltammetry characteristic curve of NiV-LDHs at 5 to 100 mV s<sup>-1</sup>. (b) The CV curve of Ti<sub>3</sub>C<sub>2</sub>@NiV-LDHs at 5 to 100 mV s<sup>-1</sup>. (c) Cyclic characteristics curve of the NiCo-LDHs tested. (d) Cyclic characteristics curve of the Ti<sub>3</sub>C<sub>2</sub>@NiCo-LDHs tested.

### 3. Two-Electrode System



**Fig. S6.** (a) Cyclic voltammetry characteristic curves of Ti<sub>3</sub>C<sub>2</sub>@NiV-LDHs//AC ASC at 50 mV s<sup>-1</sup> of different voltage windows. (b) Constant current charge-discharge curves of Ti<sub>3</sub>C<sub>2</sub>@NiV-LDHs//AC ASC at 2 A g<sup>-1</sup> to 10 A

$g^{-1}$ . (c) Cyclic voltammetry curves of self-assembled  $Ti_3C_2@NiCo-LDHs//AC$  soft package devices under different voltage windows. (d) Constant current charge-discharge curve of the  $Ti_3C_2@NiCo-LDHs//AC$  device.



**Fig. S7.** (a) Charge transfer internal resistance curve of  $Ti_3C_2@NiV-LDHs//AC$  ASC. (b) EIS curves of the  $Ti_3C_2@NiCo-LDHs//AC$  device before and after long-term charge-discharge cycle test.