Supporting Information

Experimental Section:

CoAl-LDH nanosheet array on Ni foam were synthesized by a simple hydrothermal method. In a typical procedure, $Co(NO_3)_2 \cdot 6H_2O$ (2 mmol), $Al(NO_3)_3 \cdot 6H_2O$ (2 mmol), NH_4F (8 mmol) and $CO(NH_2)_2$ (10 mmol) were dissolved in 36 mL of distilled water and stirred to form a clear solution. Nickel foam (about 3 cm × 2 cm) was carefully cleaned with concentrated HCl solution (37 wt%) in an ultrasound bath for 5 min in order to remove the surface NiO layer, and then deionized water and absolute ethanol were used for 5 min each to ensure the surface of the Ni foam was well cleaned. The aqueous solution and the Ni foam were transferred to a 40 mL Teflon-lined stainless-steel autoclave, which was sealed, maintained at 100 °C for 24 h, and then allowed to cool to room temperature within 15 min using cooling water. The pink thin film on the metal substrate was immersed in 5 $molL^{-1}$ NaOH for 1-3 min and subsequently rinsed with distilled water, ethanol each for 5 minutes with the assistance of ultrasonication, and dried at 80 °C for 6 h. The weight of CoAl-LDH was accurately measured by weighing the Ni foam before and after hydrothermal process, a loading mass of ~10.5 $mg \cdot cm^{-2}$ is achieved. MPCCH thin nanosheet array was obtained by immersing the CoAl-LDH in 5 $mol \cdot L^{-1}$ NaOH solution with vigorous agitation overnight, and then the sample was rinsed several times by distilled water, dried at 80 °C for 6 h, the mass-loading of MPCCH thin nanosheet array was about 8 $mg \cdot cm^{-2}$.

X-ray powder diffraction patterns were recorded on an X-ray diffractometer (Rigaku D/max 2500) at a scan rate of 10 (°)/min in the range from 5 to 90°. The size and morphology of the samples were characterized using a field-emission SEM (JEOL JSM6335) operating at 20 kV and a TEM system (H800) operating at 200 kV. FT-IR spectrum was carried out by using a model of Nicolet 8700.

The electrochemical measurements were carried out at room temperature in a three-electrode glass cell connected to an electrochemical workstation (CHI 660D, chenghua, shanghai.). Fresh film (CoAl-LDH nanosheet array and MPCCH array) on

the metal substrate (1 cm × 1 cm) was used as the working electrodes. A platinum electrode and a saturated calomel electrode were used as counter and reference electrodes, respectively. Freshly prepared 2 mol/L NaOH aqueous solution was used as the electrolyte.

The specific capacitances C were calculated from the galvanostatic discharge curves using Equation 1, where 'I' is the current applied, 'ΔV/Δt' is the slope of the discharge curve, and 'm' is the mass of the sample on one electrode.

$$C = \frac{I * \Delta t}{\Delta V * m} \quad \text{(Equation 1)}$$

Figures:

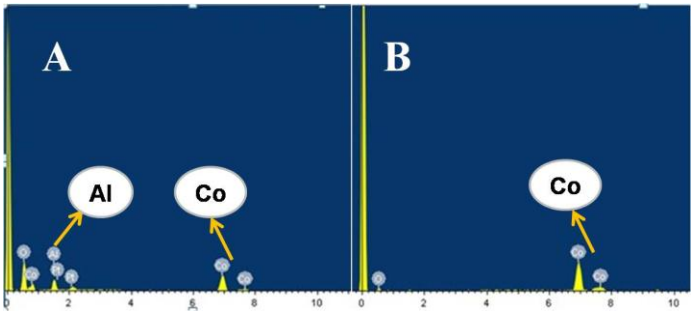

Figure S1: EDS spectrum of CoAl-LDH nanosheet array (A) and MPCCH thin nanosheet array (B).

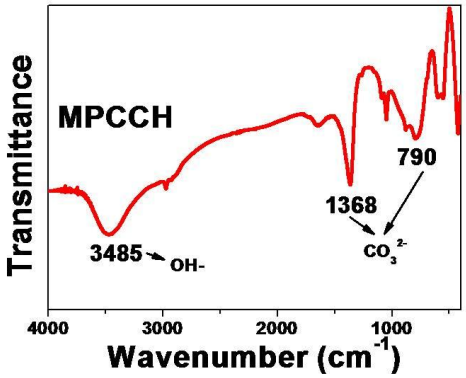

Figure S2: FT-IR spectrum of MPCCH.

Analysis: The broad peak centered at 3485 cm^{-1} corresponds to the O-H stretching vibration of water molecules in the interlayer and H-bonded OH group, companied with the bending mode at 1630 cm^{-1}. The intense peaks at 1368 and 790 cm^{-1} are ascribed to the v3 vibration and bending modes of CO_3^{2-}, respectively.

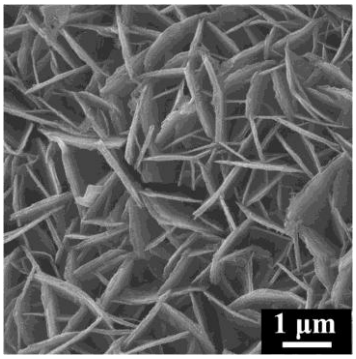

Figure S3: Typical SEM image of MPCCH thin nanosheet array after 2000 cycles.

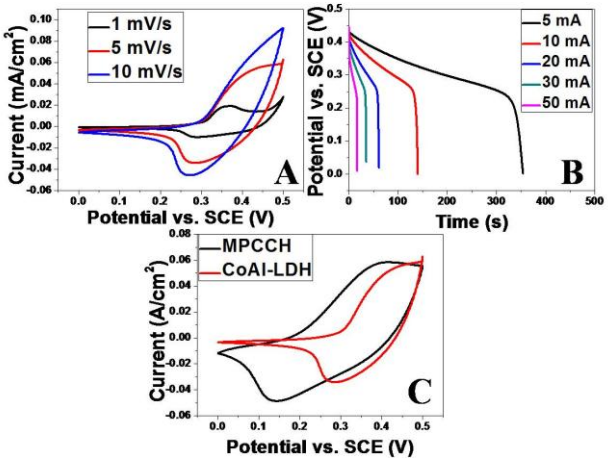

Figure S4: Electrochemical characterization of the CoAl-LDH nanosheet array: (A) CV curves of the CoAl-LDH nanosheet array at different scan rates(1-10 mV/s); (B) Galvanostatic discharge curves of the MPCCH at various discharge current densities (5-50 mA/cm^2); (C) Comparison of CV curves obtained from the same scan rate (5mV/s) between MPCCH and CoAl-LDH.

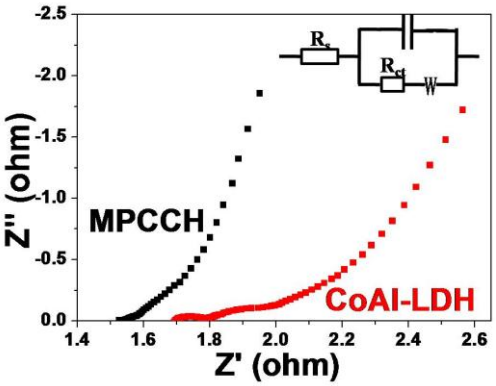

Figure S5: Electrochemical impedance spectra of MPCCH thin nanosheet array and CoAl-LDH nanosheet array, the inset is the electrical equivalent circuit.

Analysis: The Nyquist plot of the EIS tests in 2 M NaOH solution with a frequency loop from 105 Hz to 0.01 Hz using a perturbation amplitude of 5 mV at 0.3 V versus SCE. The intersection of the plots at the x-axis represents the solution resistance (Rs), which includes the following three terms: the resistance of the NaOH aqueous solution, the intrinsic resistance of the electroactive materials themselves and the contact resistance at the interface between electroactive materials and the current collector. And at the high-medium frequency region, a semicircle can be found and its

diameter stands for the charge transfer resistance (R_{ct}) in the electrochemical process. As clearly shown in the figure, the MPCCH thin nanosheet electrode shows better electronic conductivity and lower charge transfer resistance than CoAl-LDH nanosheet array. Therefore, the as-synthesized MPCCH thin nanosheet array electrode can obtain a large specific capacitance and electrochemical utilization even at high charge–discharge rates. In addition, in the low frequency region, the impedance plot of MPCCH is more vertical, indicating the better electrochemical capacitance of the MPCCH in the NaOH aqueous solution than CoAl-LDH.