Experimental section

The preparation procedure of the ultrathin $Ni(OH)_2$ nanowall film is described as follows. All of the chemicals were of analytical grade and used without further purification. In a typical procedure, $Ni(NO_3)_2 \cdot 6H_2O$ (1.45 g, 5 mmol) and HMT (1.40 g, 10 mmol) were dissolved in 35~38 mL distilled water and stirred to form a clear solution. Nickel foam (about 3.3 cm x 1 cm) was carefully cleaned with concentrated HCl solution (37 wt.%) in an ultrasound bath for 5 min in order to remove the surface oxide 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 10 h, and then allowed to cool to room temperature within 15 minutes using cooling water. The thin film on the metal substrate was rinsed several times with distilled water and ethanol with the assistance of ultrasonication, and dried at 80 °C for 6 h.

X-ray powder diffraction (XRD) patterns were recorded on a X-ray diffractometer (Rigaku D/max 2500) at a scan rate of 10 °/min in the 2θ range from 15 to 90°. The size and morphology of the samples were characterized using a field-emission scanning electron microscope (SEM) (JEOL JSM6335) operating at 20 kV. High-resolution transmission electron microscopy (HRTEM) measurements were carried out using a JEOL JEM 2100 system operating at 200 kV.

The electrochemical measurements were carried out at 298 K in a three-electrode glass cell connected to an electrochemical workstation. The thin film on the metal substrate (1 cm x 1 cm) was used as a working electrode. A platinum foil (1.0 cm^2) and a saturated calomel electrode were used as a counter and reference electrode, respectively. Fresh 1 mol/L NaOH aqueous solution was used as the electrolyte after purging under N_2 for 15 min. The electrochemical performances of the samples were evaluated on an AUTOLAB workstation for cyclic voltammetry (CV) and chronopotentiometry (CP) tests.

Capacitance calculation

Theoretical capacitance calculation: The required amount of power used for electrolysis of 1 mol active material is 1 F which is equal to 96485 C. For $Ni(OH)_2$, its molar mass is 93 g/mol, so theoretical specific capacitance is calculated as follows:

$$F = \frac{Q}{U} = \frac{96485/93}{U} = \frac{1037.5}{U}$$

Where U is the voltage window, Q is electrical energy per 1 gram. When U is 0.44V, the theoretical pesudocapacitance is 2358 F/g for our sample.

Mass calculation: After hydrothermal synthesis of nickel hydroxide on a nickel foam, we got a mass increase of 2.7 mg and capacity increase of 7.85 F per square centimeter. In order to eliminate the influence of the background, we did a control experiment by hydrothermal treatment of nickel foam in the similar solution without nickel salts, and finally got a mass increase of 0.133 mg and capacity increase of 0.48 F per square centimeter. Then we calculated the quantity of nickel hydroxide resulting from the substrate oxidation or Ni^{2+} hydrothermal translation by the following equations:

$$34x + 93y = 2.7 * 10^{-3} \qquad (1)$$

$$34x = 0.133 * 10^{-3} \qquad (2)$$

Where x is the mole of nickel hydroxide obtained from the nickel foam substrate and y is the mole number of nickel hydroxide synthesized from the added nickel source. By solving the equations, the values of x and y are 0.0039 and 0.0276, respectively. So **the total mass of active materials is 2.93 mg**.

Specific capacitance calculation: We also did further calculation to verify whether all the nickel hydroxide was utilized or not. The following is an equation proposed.

$$0.48 + \theta * y * 93 + z * \frac{y*93*10^{-3}}{4150*10^{3}*6*10^{-9}} = 7.85 \qquad (3)$$

Where the three terms, represent the background capacitance, pseudocapacitance

and electrochemical double layered capacitance (EDLC), respectively. The total of them equals to the real capacitance obtained. These two independent variables (θ and z) are the specific pesudocapacitance, a constant value of capacitance versus square meter (F/cm^2), respectively. While x and y was got above.

Because equation 3 contains two independent variables (θ and z), it is insoluble. For solving the equation, the other equation is needed. We make three tangents at the beginning, middle (platform) and final sections in the discharge curve at 5 mA/cm^2 as seen in Fig. S4. The first and the third tangents are extended up to the abscissa, almost parallel. The junction points correspond to 120s and 697s, respectively. Therefore, the ratio of EDLC to pseudocapacitance is 120/577 and the other equation is proposed as follows:

$$\frac{z*\frac{y*93*10^{-3}}{4150*10^{3}*6*10^{-9}}}{0.48+\theta*y*93}=\frac{120}{577} \qquad (4)$$

Combination between Equation 3 and 4, the values of θ (2350) and z (6.08) are calculated. So we can draw the following conclusions:

(a) the specific capacitance is 2675 F/g;

(b) the contribution of EDLC is 325 F/g;

(c) the contribution of pseudocapacitance is 2350 F/g, close to the theoretical capacitance (2358 F/g).

Figures

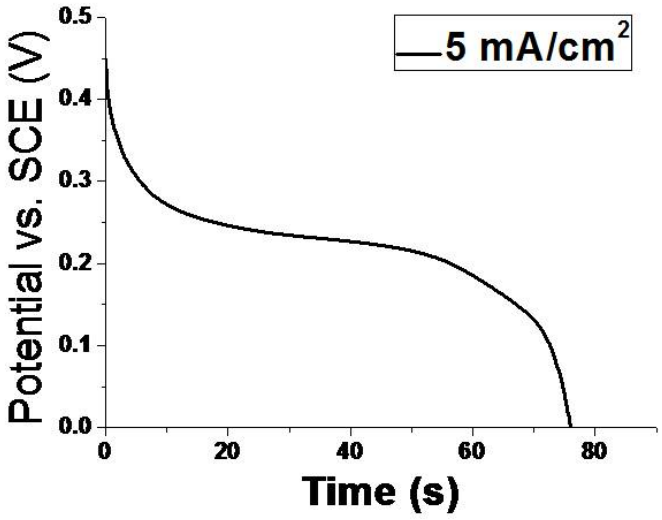

Fig. S1 Galvanostatic discharge curve of the pure Nickel foam at 5 mA/cm^2 after oxidation.

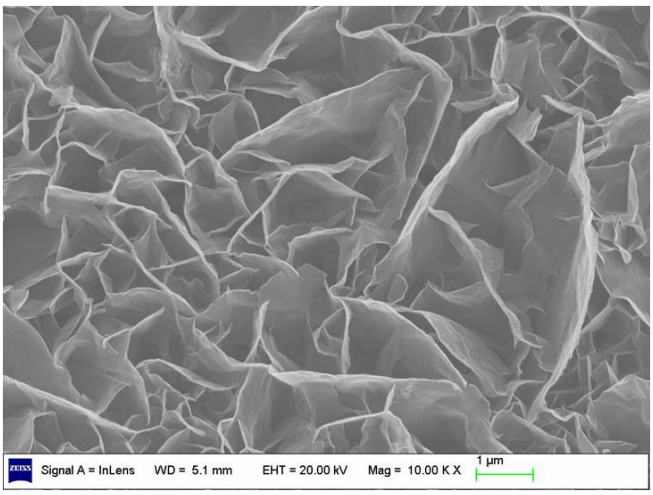

Fig. S2 Typical SEM image of the $Ni(OH)_2$ nanowall film after 500 cycles.

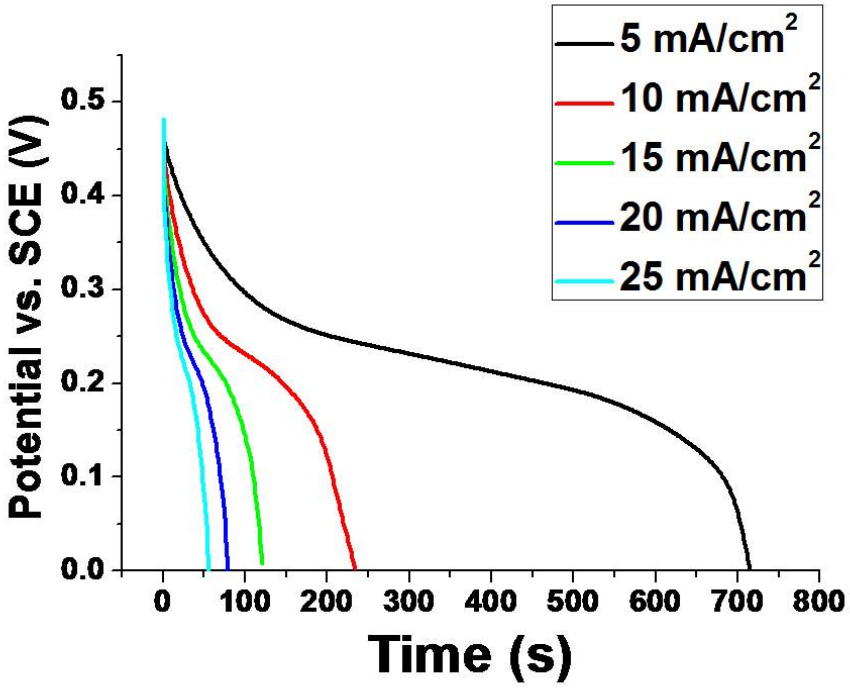

Fig. S3 Discharge curves at various current densities of the $Ni(OH)_2$ nanowall film after 500 cycles.

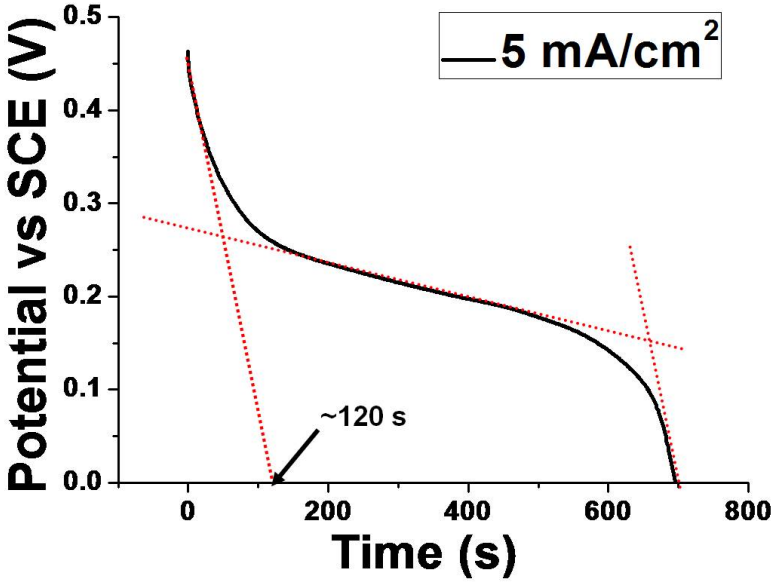

Fig. S4 Mathematical analysis of the discharge curve at 5 mA/cm^2.