

Supplementary Information

O-vacancy Enriched NiO Hexagonal Platelets Fabricated on Ni Foam as Self-supported Electrode for Extraordinary Pseudocapacitance

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Donor density calculation^{1,2}

The slopes determined from the Mott–Schottky plots could be used to estimate the donor densities using the following equation:^{1,2}

$$Nd = \left(\frac{2}{e_0 \varepsilon \varepsilon_0} \right) \left[\frac{d\left(\frac{1}{C^2}\right)}{dV} \right]^{-1}$$

where N_d is the donor density, e_0 the electron charge, ε the dielectric constant of NiO (30),³ ε_0 the permittivity of vacuum, C is the capacitance, and V the potential applied at the electrode. It should be pointed out that the above Mott–Schottky equation was derived based on a planar electrode model while in our work the electrode materials are stacked hexagonal platelets, which could cause errors in determining the carrier densities as the geometric area of the electrode for the calculation is much smaller than the electrode effective surface area. Nevertheless, a qualitative comparison of carrier densities between the NiO-N₂-1h and NiO-air-1h samples can be made because the two electrodes have similar morphology and surface area.

Mass measurement and calculation

The weight of a sample was measured using a Mettler Toledo XS205DU microbalance (0.01 mg sensitivity). Before weighing, all samples were thoroughly dried in a vacuum oven until the mass displayed no variation. The loading mass of the NiO active materials can be obtained by measuring the weight change of the samples before and after fabricating process and calculating according to the following equation:⁴

$$m = C^*(m_2 - m_1)$$

where m is the loading mass of the NiO active materials, m_1 is the mass of the Ni foam before fabricating process and m_2 is the mass of the sample after fabricating process, C is a constant which represents the molar mass ratio of $\frac{M_{NiO}}{M_{NiO} - M_{Ni}}$. The

mass loading of the active material NiO in the work is ~ 5.00 mg/cm².

Electrochemical specific capacitance calculation

Calculation of specific capacitance (C_s , F g⁻¹) in this work is based upon subtraction of the capacitance which belongs to the Ni foam current collector from the total capacitance of the as-prepared NiO (NiO-N₂-1h or NiO-air-1h) electrode according to following equations:

(1) based upon the CV curves:⁵

$$C_s = \frac{Q_{NiO} - Q_{Ni\ foam}}{\Delta V \times m_{NiO}} = \frac{1}{\nu \times m_{NiO} \times (V_c - V_a)} \left[\int_{V_a}^{V_c} i_{NiO}(V) dV - \int_{V_a}^{V_c} i_{Ni\ foam}(V) dV \right]$$

where m_{NiO} is the mass of the NiO electroactive material in the electrode (g), ν is the potential scan rate (V s⁻¹), V_a is the anodic potential (V), V_c is the cathodic potential (V), $i(V)$ is the response current density (A) and V is the potential (V).

(2) based upon the galvanostatic charge-discharge curves:^{5,6}

$$C_s = \frac{I \times \Delta t_{NiO} - I \times \Delta t_{Ni\ foam}}{m_{NiO} \times \Delta V}$$

where I is the discharge current (A), Δt is the discharge time (s), m_{NiO} is the mass of the NiO electroactive material in the electrode (g) and ΔV is the potential change during discharge (V).

Contribution of the Ni foam collector

In order to evaluate the contribution of Ni foam collector in the electrochemical measurement, CV curves and galvanostatic charge-discharge curves of a blank Ni foam collector with the same working area were measured under identical conditions, which are compared with those of the NiO-N₂-1h electrode in Fig. S6 and Fig. S7. Obviously, the contribution of the blank Ni foam collector in the electrochemical measurement is very small (~1.7% in CV measurements and ~1.1% in galvanostatic charge-discharge tests).

Supplementary Figures and Tables

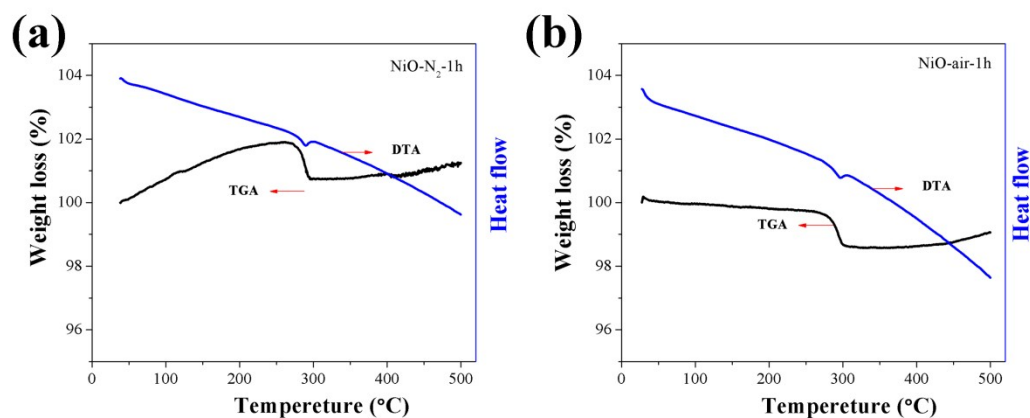


Fig. S1 TGA and DTA curves of the Ni(OH)₂ precursor under different atmospheres:

(a) N₂ and (b) air.

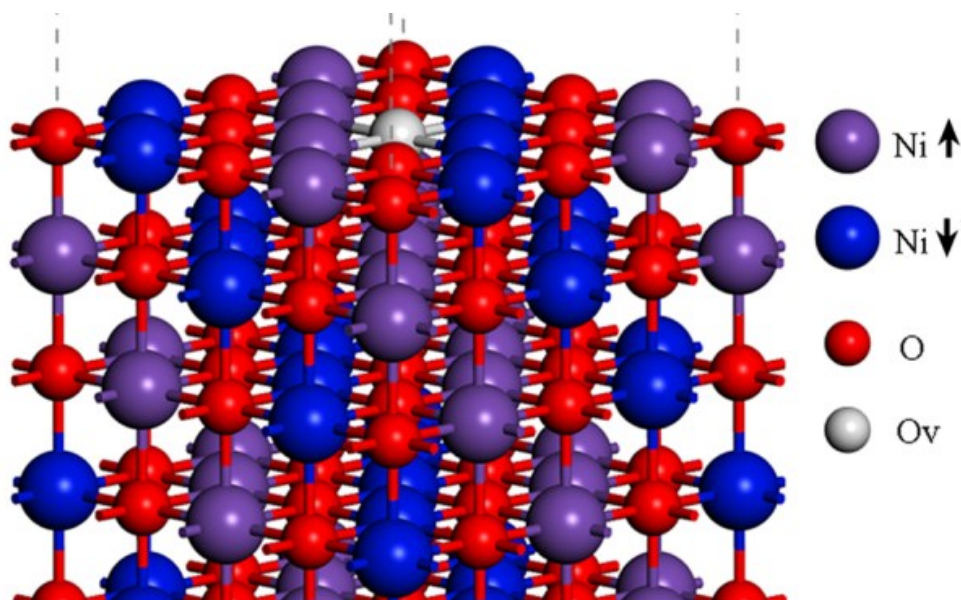


Fig. S2 Illustration of supercell unit in the first five atomic layers below the surface.

Violet and blue spheres refer to Ni atoms with different spins, and the red (gray) spheres refer to O (vacancy) atoms.

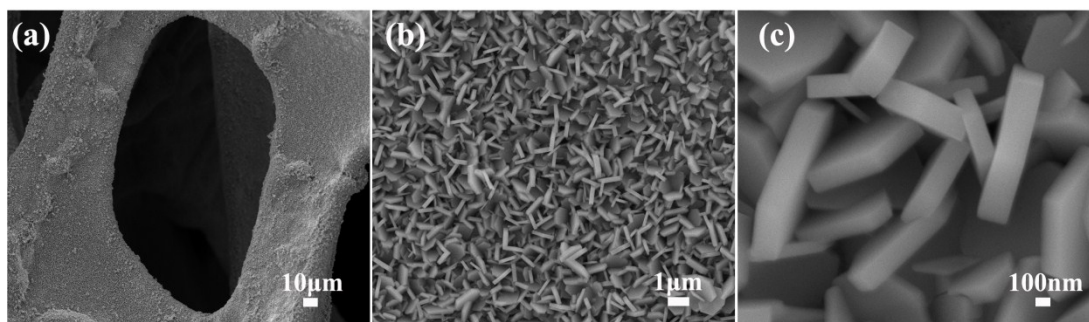


Fig. S3 SEM images of the Ni(OH)₂ precursor.

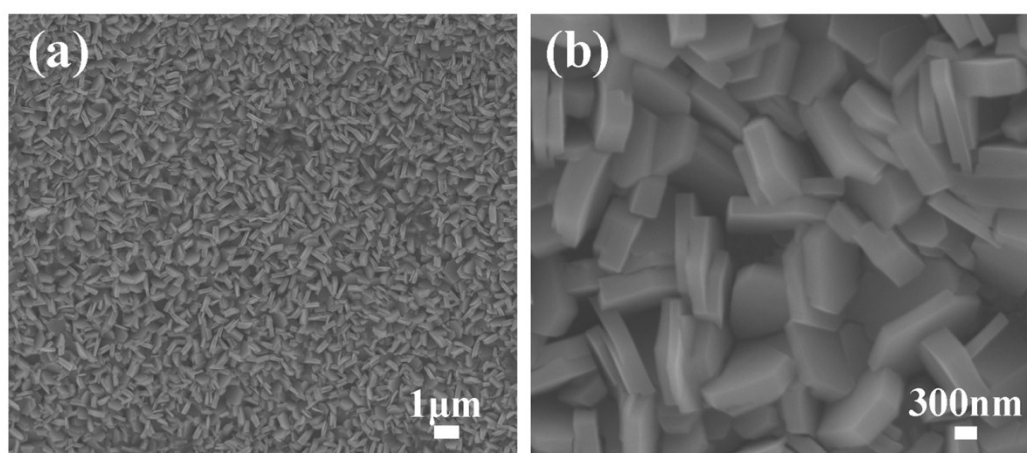


Fig. S4 SEM images of the NiO-N₂-1h sample after sustained ultrasonication in a 2.0 M KOH aqueous solution for 2 h.

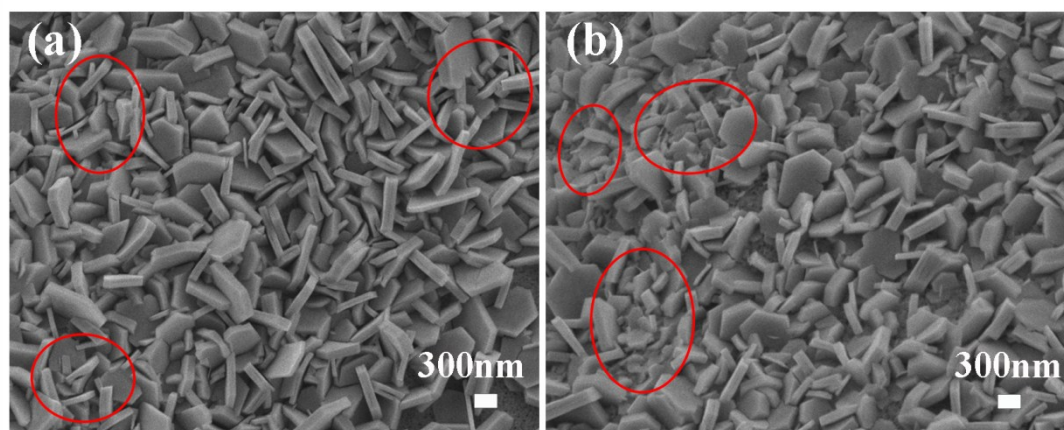


Fig. S5 SEM images of different samples: (a) NiO-N₂-3h, and (b) NiO-N₂-10h.

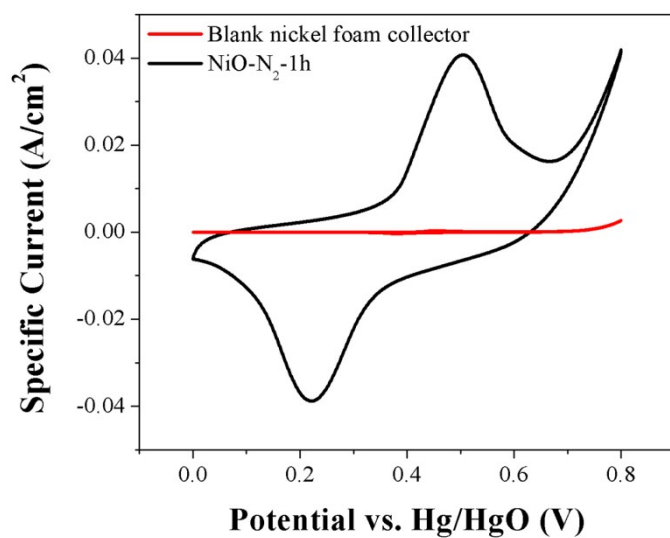


Fig. S6 Comparison of CV curves of the blank Ni foam collector and the as-prepared NiO-N₂-1h electrode at a scan rate of 10 mV s⁻¹.

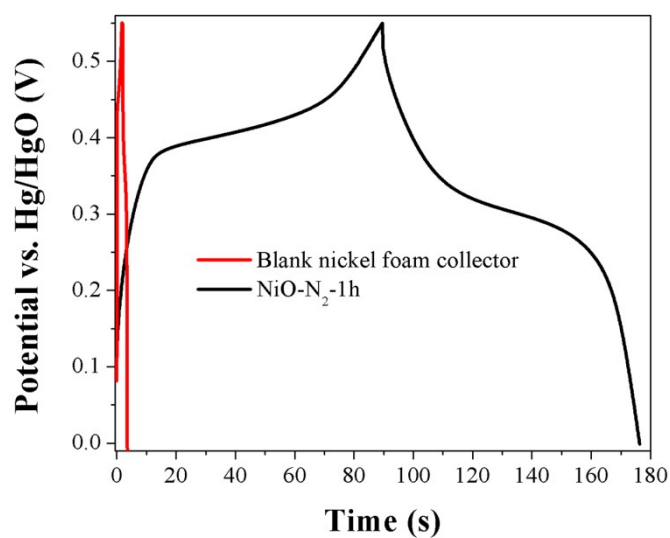


Fig. S7 Comparison of galvanostatic charge-discharge curves of the blank Ni foam collector and the as-prepared NiO-N₂-1h electrode at current density of 5 A g⁻¹.

Table S1. Comparison of the electrochemical performance of the as-prepared NiO-N₂-1h material in this work with other NiO-based materials reported in previous literatures.

Ref.	Active material	Maximum specific capacitance (F g ⁻¹)	Rate capability
This work	NiO platelets	2495 F g ⁻¹ at 1 mV s ⁻¹ 899 F g ⁻¹ at 0.5 A g ⁻¹	80% from 0.5 to 10 A g ⁻¹
7	NiO nanospheres	550 F g ⁻¹ at 1 A g ⁻¹	55% from 1 to 10 A g ⁻¹
8	NiO nanoplatelets	500 F g ⁻¹ at 0.4 mA cm ⁻²	–
9	NiO nanoflowers	480 F g ⁻¹ at 0.5 A g ⁻¹	53% from 0.5 to 5 A g ⁻¹
10	NiO/Ni nanoparticles	910 F g ⁻¹ at 5 mV s ⁻¹	–
11	NiO nanostructures	783 F g ⁻¹ at 2 mV s ⁻¹	14% from 2 to 100 mV s ⁻¹
12	NiO nanofibers	336 F g ⁻¹ at 5 mA cm ⁻²	54% from 5 to 20 mA cm ⁻²
13	NiO nanospheres	612.5 F g ⁻¹ at 0.5 A g ⁻¹	83% from 0.5 to 3 A g ⁻¹
14	NiO nanofibers	884 F g ⁻¹ at 0.5 A g ⁻¹	51% from 0.5 to 20 A g ⁻¹
15	NiO nanocolumns	686 F g ⁻¹ at 1 A g ⁻¹	57% from 1 to 5 A g ⁻¹
16	NiO nanosheets	674.2 F g ⁻¹ at 1 A g ⁻¹	64% from 1 to 20 A g ⁻¹
17	NiO nanoflakes	309 F g ⁻¹ at 1 A g ⁻¹	72% from 1 to 40 A g ⁻¹
18	NiO nanospheres	770 F g ⁻¹ at 2 A g ⁻¹	42% from 2 to 10 A g ⁻¹
19	NiO nanospheres	555 F g ⁻¹ at 1 A g ⁻¹	73% from 1 to 5 A g ⁻¹
20	NiO nanoparticles	1088.44 F g ⁻¹ at 5 mV s ⁻¹	15% from 5 to 200 mV s ⁻¹
21	NiO nanosheets	548 F g ⁻¹ at 2 mV s ⁻¹	68% from 0.5 to 10 A g ⁻¹

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