Supporting Information

Ultra-Dense NiS_2/Reduced Graphene Oxide Composite Cathode for High-Volumetric/Gravimetric Energy Density Nickel-Zinc Batteries

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Measurement of tap density for electrode:

The tap density (ρ) of the electrodes was calculated by the following equation:

$$\rho = m/(S \times h) \qquad (1)$$

where m is mass of the electrode containing active material, electrical additives and binders, S and h are area and thickness of the active material on the electrode, respectively.

Calculations of capacity, energy and power densities:

Specific capacity (C_s, mAh g^{-1}) and volumetric capacity (C_v, mAh cm^{-3}) of the fabricated cathode material are calculated based on discharging curves using the following equations (1-2):

$$C_s = \frac{\int_0^t I\,dt}{m} \qquad (1)$$

$$C_v = \frac{\int_0^t I\,dt}{V} \qquad (2)$$

where I is the discharging current (mA), t is the discharging time (h), and m (g) is the mass of active material in cathode, V is the volume of the whole cathode material containing active material and additives.

The gravimetric energy density (E_s, Wh kg^{-1}) and power density (P_s, W kg^{-1}) are calculated by using the following equations (3-4):

$$E_s = \int_0^t \frac{\Delta V \times I}{m} dt \qquad (3)$$

$$P_s = \frac{E_s}{\Delta t} \qquad (4)$$

where ΔV is the discharging voltage range (V), I is the discharging current (mA), t is the discharging time (h), and m is the active mass of HD-NiS_2/rGO-5 cathode (g).

The volumetric energy density ($E_{V\text{ - cell}}$, mWh cm^{-3}) and power density ($P_{V\text{ - cell}}$, mW cm^{-3}) of the assembled Ni-Zn battery are calculated using the following equations (5-6):

$$E_{V\text{ - }cell} = \int_0^t \frac{\Delta V \times I}{V\text{ - }cell} dt \quad (5)$$

$$P_{V\text{ - }cell} = \frac{E_{V\text{ - }cell}}{t} \quad (6)$$

where ΔV is the discharging voltage range (V), I is the discharging current (mA), t is the discharging time (h), $V\text{ - cell}$ is the volume of the whole HD-NiS_2/rGO-5//Zn battery, including active materials, current collector, separator and Zn plate.

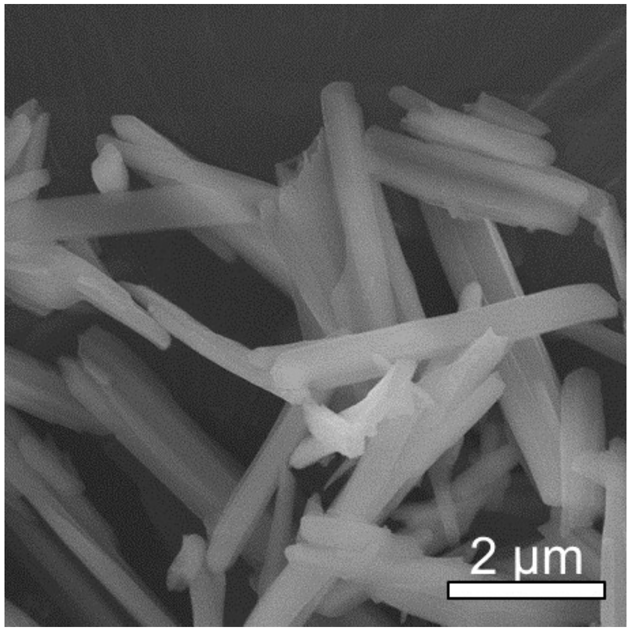

Figure S1. SEM image of Ni-MOF crystals.

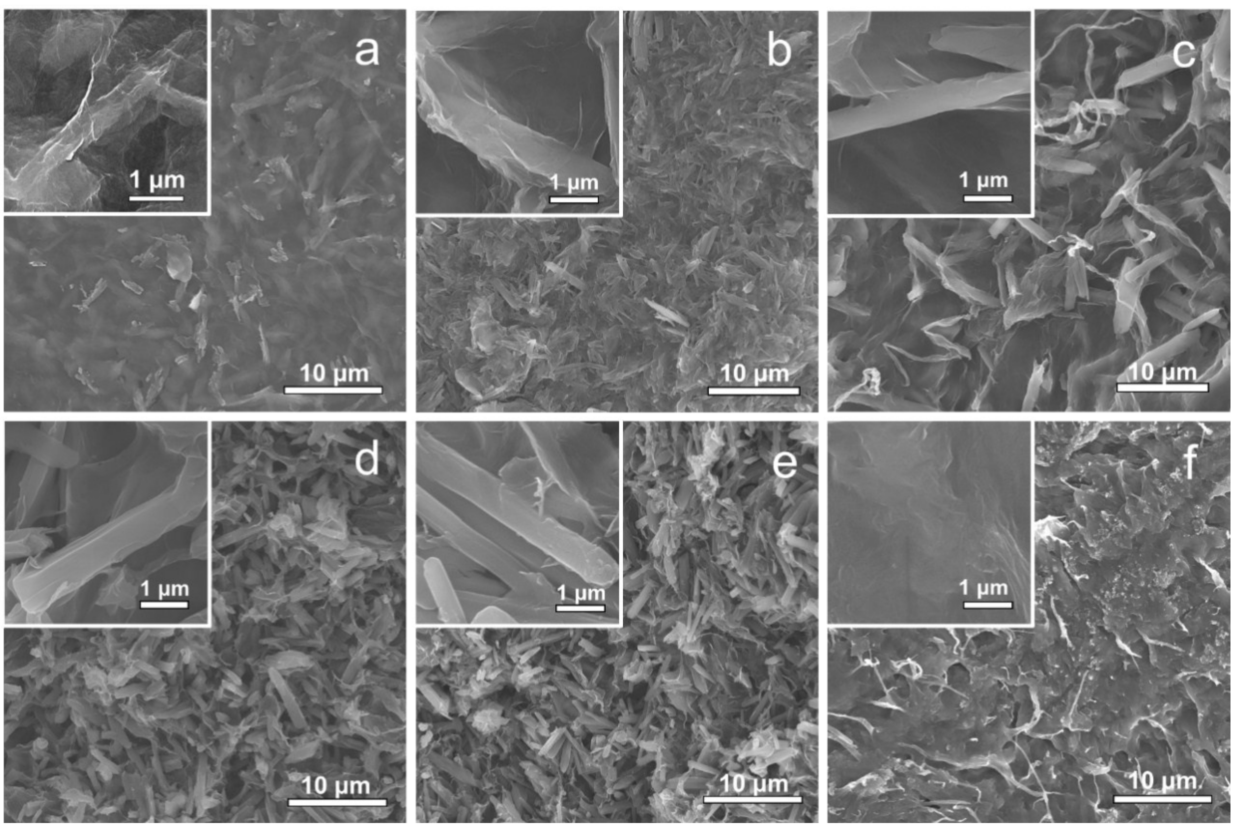

Figure S2. SEM images of HD-MOF/GO-n with various MOF to GO weight ratios (n) of (a) 1 : 1, (b) 3 : 1, (c) 5 : 1, (d) 7.5 : 1, and (e) 10 : 1. (f) SEM image of HD-GO.

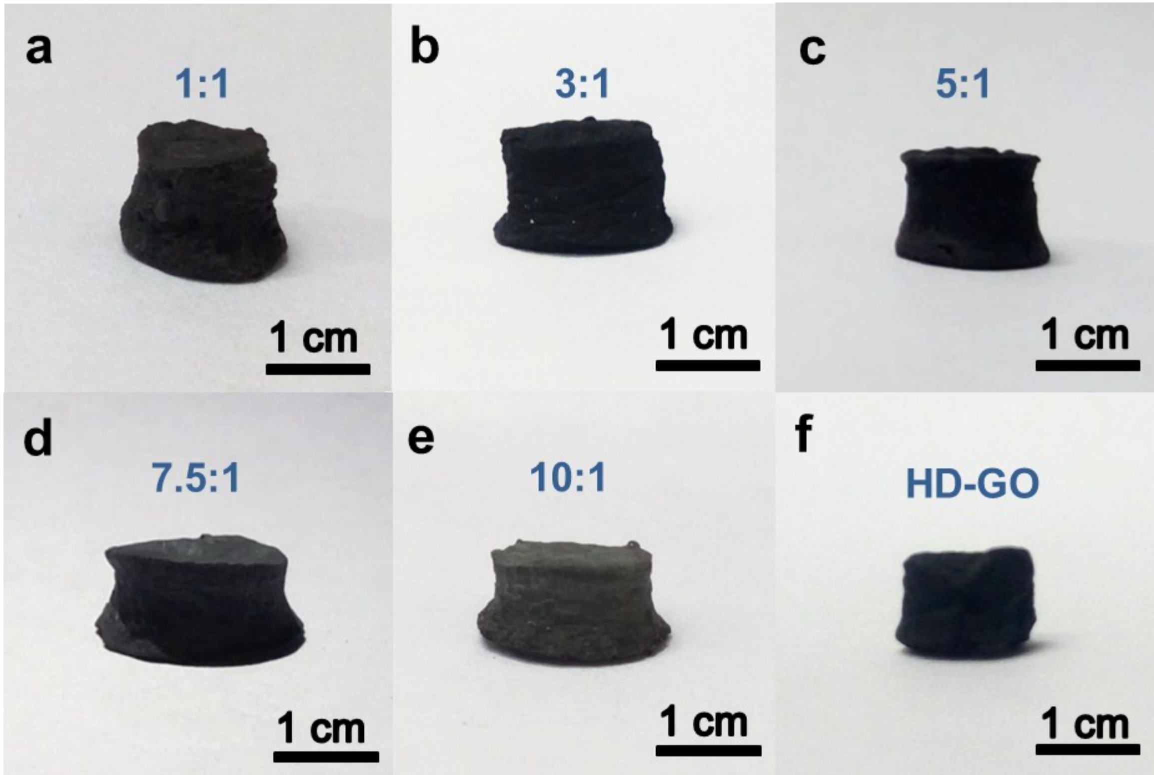

Figure S3. Photographs of (a-e) HD-MOF/GO-n with various MOF to GO weight ratios, and (f) HD-GO.

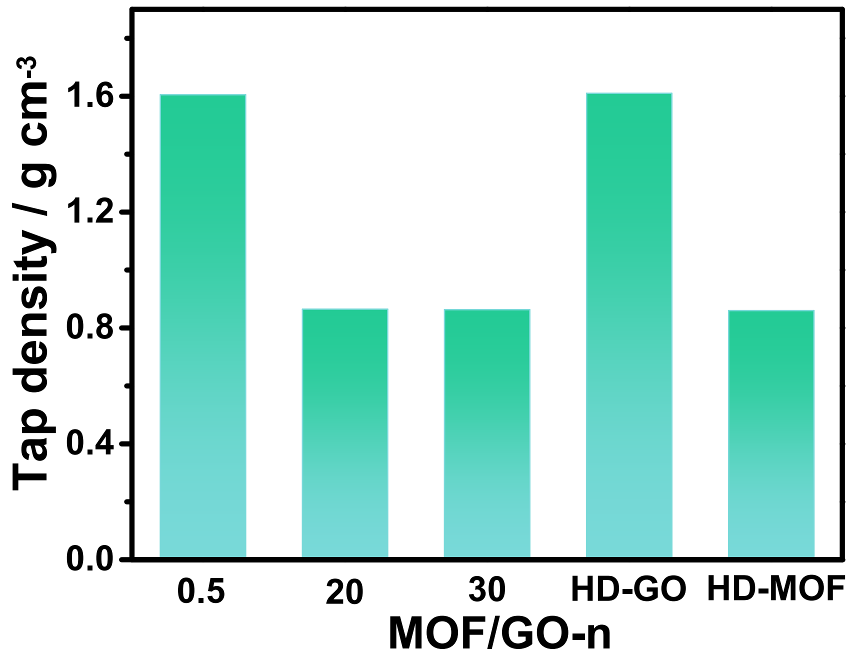

Figure S4. Tap densities of HD-MOF/GO-n (n indicates the weight ratio of MOF to GO), HD-GO and HD-MOF.

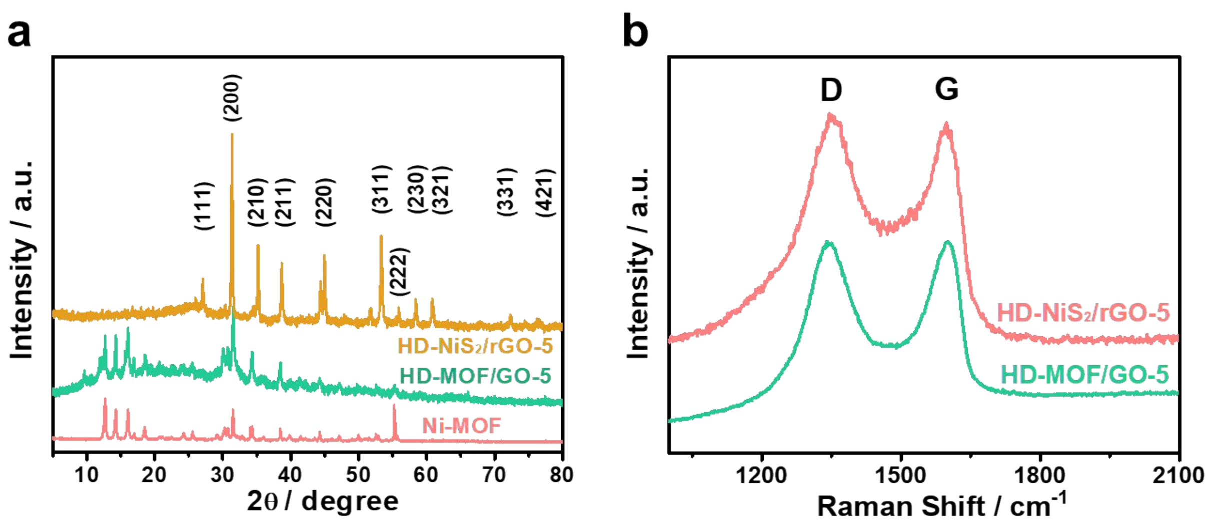

Figure S5. (a) XRD patterns of Ni-MOF crystals, HD-MOF/GO-5 and HD-NiS_2/rGO-5. (b) Raman spectra of HD-MOF/GO-5 and HD-NiS_2/rGO-5. The intensity ratios of D to G band in HD-MOF/GO-5 and HD-NiS_2/rGO-5 are 1.0 and 1.03.

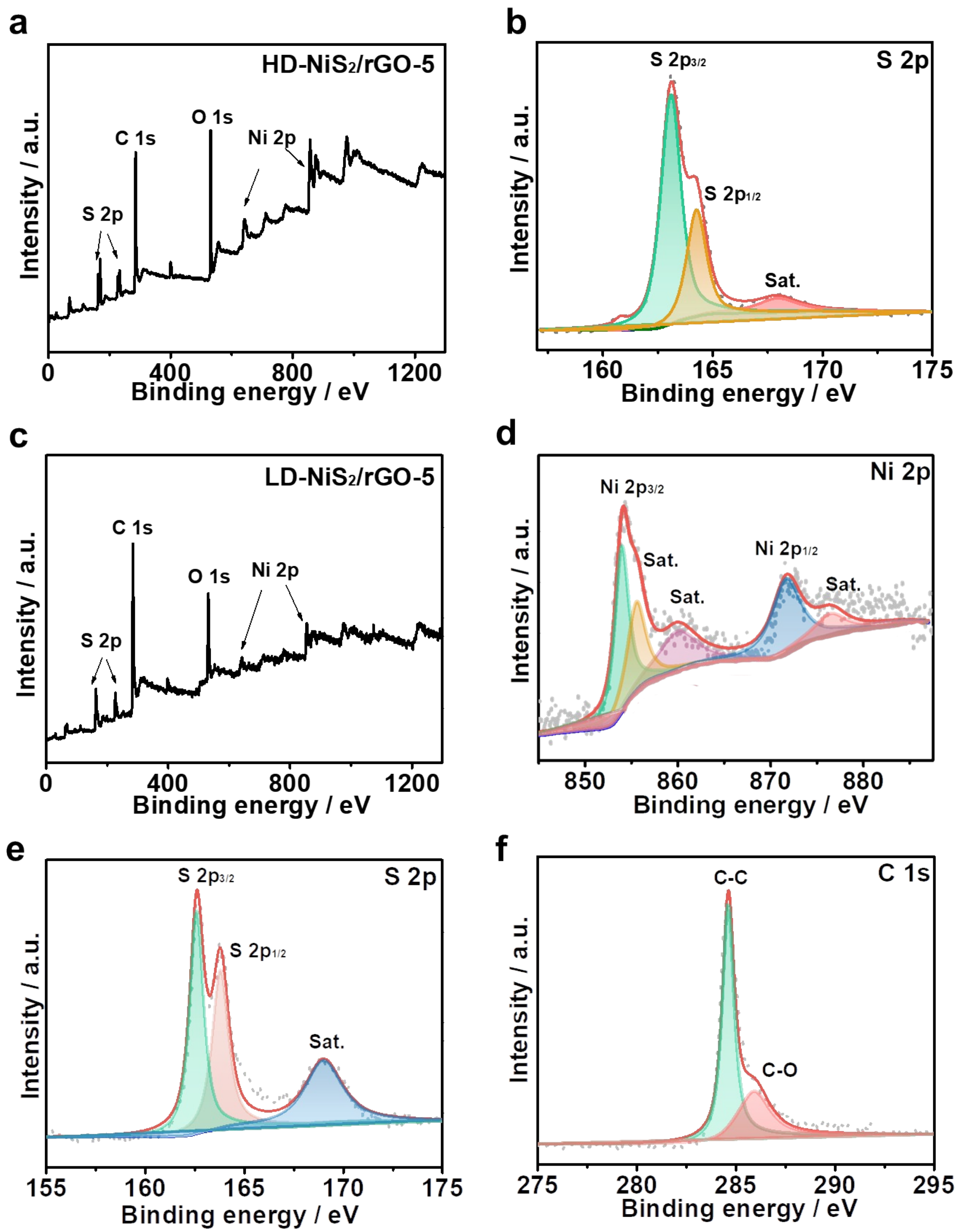

Figure S6. (a-b) XPS survey scan spectrum and high-resolution spectrum of S 2p of the HD-NiS_2/rGO-5. (c-f) XPS survey scan of the LD-NiS_2/rGO-5 and corresponding high-resolution spectra of Ni 2p, S 2p and C 1s.

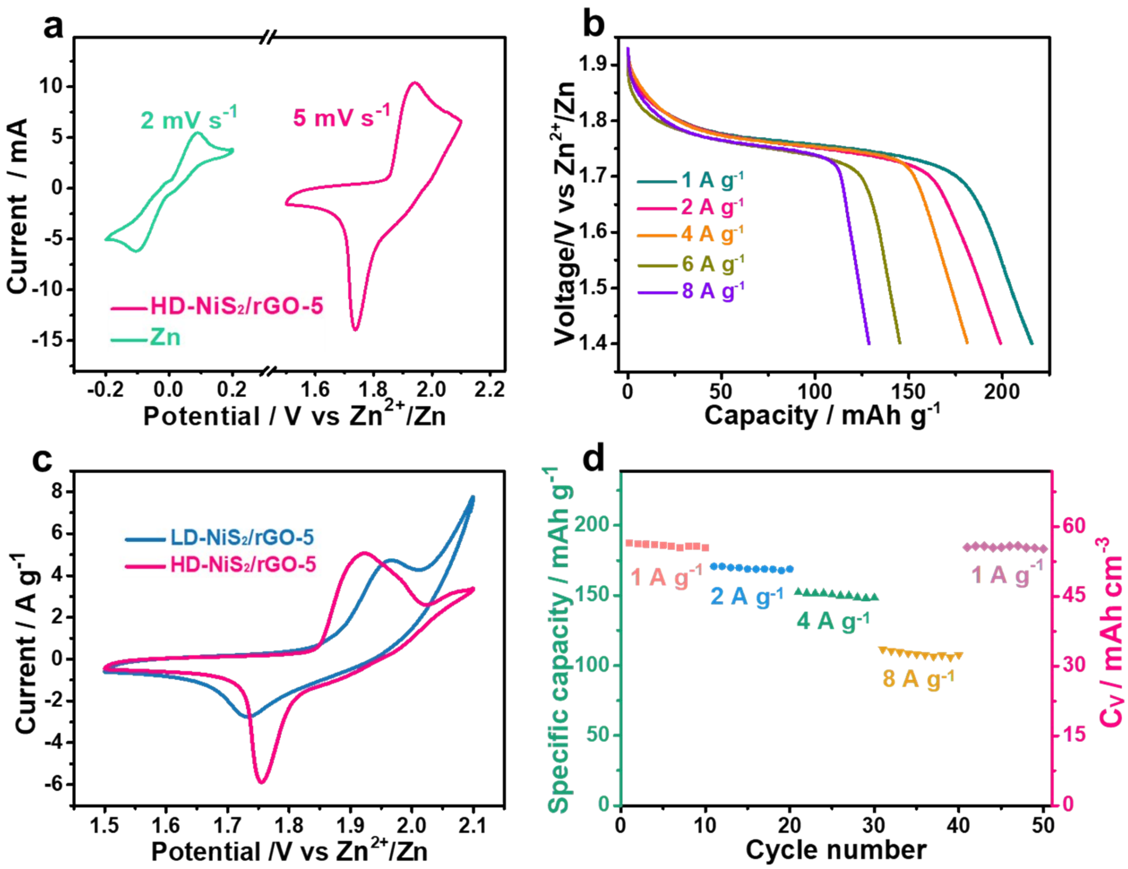

Figure S7. Electrochemical performance of HD-NiS_2/rGO-5//Zn battery, where Zn plate was used as both counter and reference electrode in 1 M KOH and 20 mM $Zn(CH_3COO)_2$. (a) CV curves of the Zn plate, and HD-NiS_2/rGO-5 electrode. (b) Discharging curves of HD-NiS_2/rGO-5//Zn battery at different current densities. (c) CV curves of LD- and HD-NiS_2/rGO-5//Zn battery at a scan rate of 5 mV s^{-1}. (d) Rate performance of the LD-NiS_2/rGO-5//Zn battery.

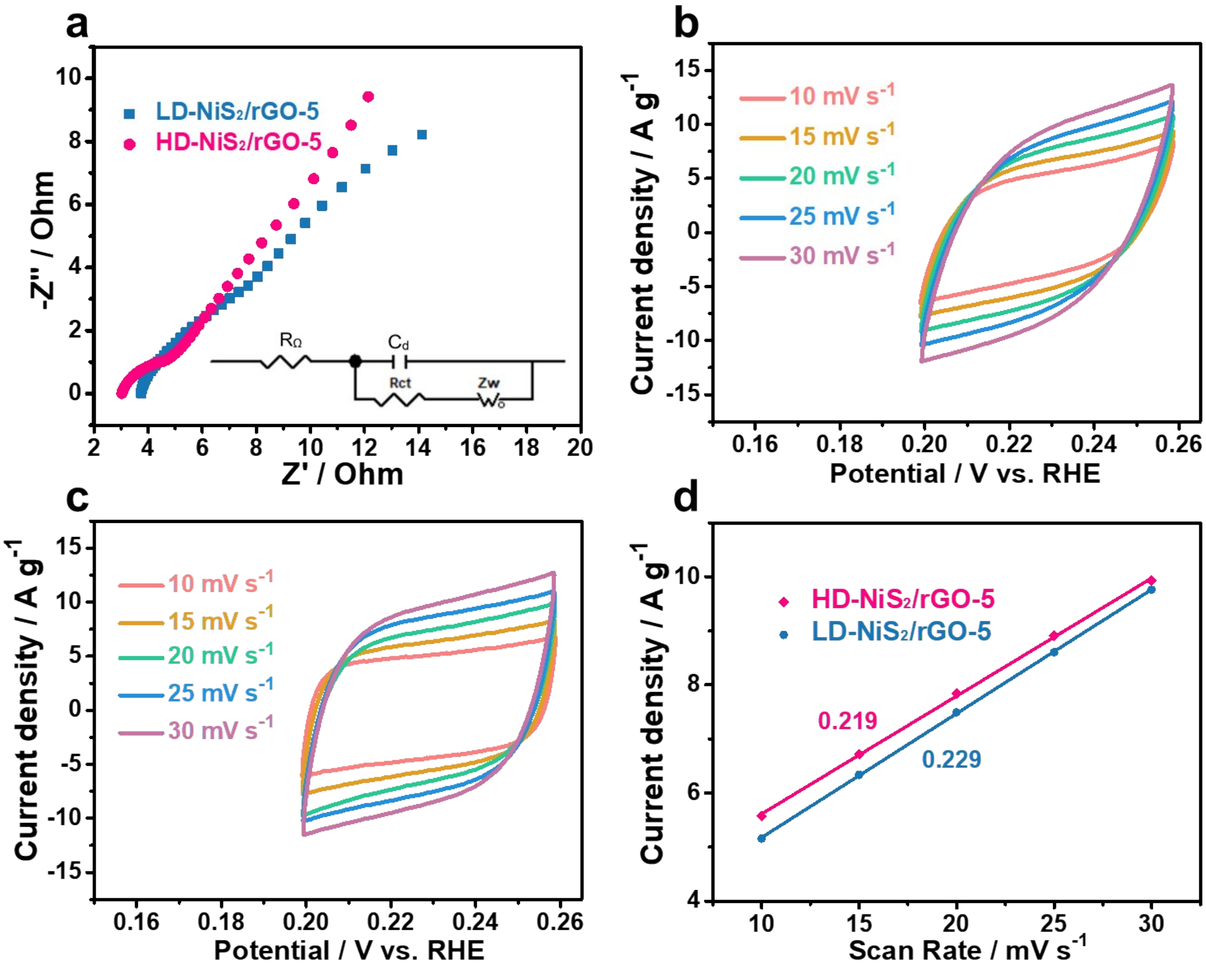

Figure S8. (a) Nyquist plots of LD- and HD-NiS_2/rGO-5//Zn battery. CV curves of (b) HD-NiS_2/rGO-5 and (c) LD-NiS_2/rGO-5. (d) The plots of current densities against various scan rates at 0.23 V.

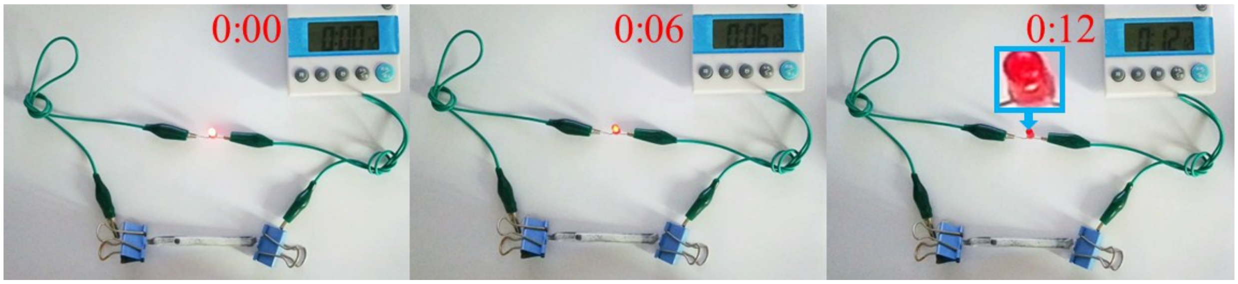

Figure S9. Photographs of a LED powered by the HD-NiS_2/rGO-5//Zn battery, which can keep lighting on for over 12 minutes.

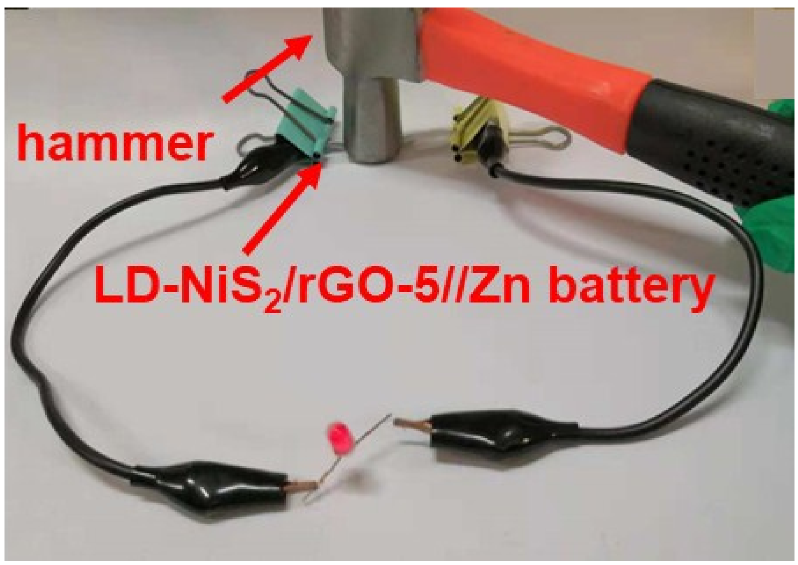

Figure S10. Photograph of a hammered LD-NiS_2/rGO-5//Zn battery showing interrupted power supply.

Movie S1. Movie showing a HD-NiS_2/rGO-5//Zn battery that is constantly being hammered still maintained a stable power output and continued to illuminate a LED.