Supplementary Information for

Ultrafast High-Capacity NiZn Battery with NiAlCo Layered Double Hydroxide

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Experiment Details

Synthesis of few-walled carbon nanotubes (FWCNTs): FWCNTs were synthesized using a catalytic chemical vapor deposition method. Co/Mo catalyst supported on porous MgO powder was used as the catalyst for nanotube growth and ethanol was used as the carbon precursor. In a typical synthesis procedure, the as-prepared catalyst powder was placed in a 3-inch horizontal tubular furnace and heated to 950˚C under the flow of Ar. Then ethanol was injected to the growth chamber for 30 minutes. These FWCNTs are composed of highly graphitized side walls with low defect density and are demonstrated to have superior mechanical and electrical properties compare to SWCNTs and other more defective MWCNTs.

Synthesis of control groups of NiAlCo LDH nanoplates, β-$Ni(OH)_2$/CNT and NiAlCo LDH/CNT hybrid with thicker nanoplates: NiAlCo LDH nanoplates were synthesized by typical procedures of NiAlCo LDH/CNT synthesis without CNT precursors. β-$Ni(OH)_2$/CNT hybrid was synthesized by typical procedures of α-$Ni(OH)_2$/CNT synthesis with water as solvent in the solvothermal steps. NiAlCo LDH/CNT hybrid with thicker nanoplates was synthesized by typical procedures of NiAlCo LDH/CNT synthesis with water as solvent in the solvothermal steps.

Calculation of specific discharging capacity, average Ni ion redox charge, energy density and power density: Specific discharging capacity was calculated by $Cs = I\Delta t/m$, where I was the discharging current, Δt was the discharging time and m was the total mass of active electrode materials. Average Ni ion redox charge is calculated by $C_{redox} = I\Delta t/nF$, where I was the discharging current, Δt was the discharging time, n was the number of Ni atoms in mol, F is the Faraday constant, equal to 96485 C/mol. Energy density and power density were calculated by using equations $d_e = \int IVdt/m$ and $d_p = d_e/\Delta t$, where I was the discharging current, V was the discharging voltage, dt was the time differential, m was the total mass of the active electrode materials, Δt was the discharging time.

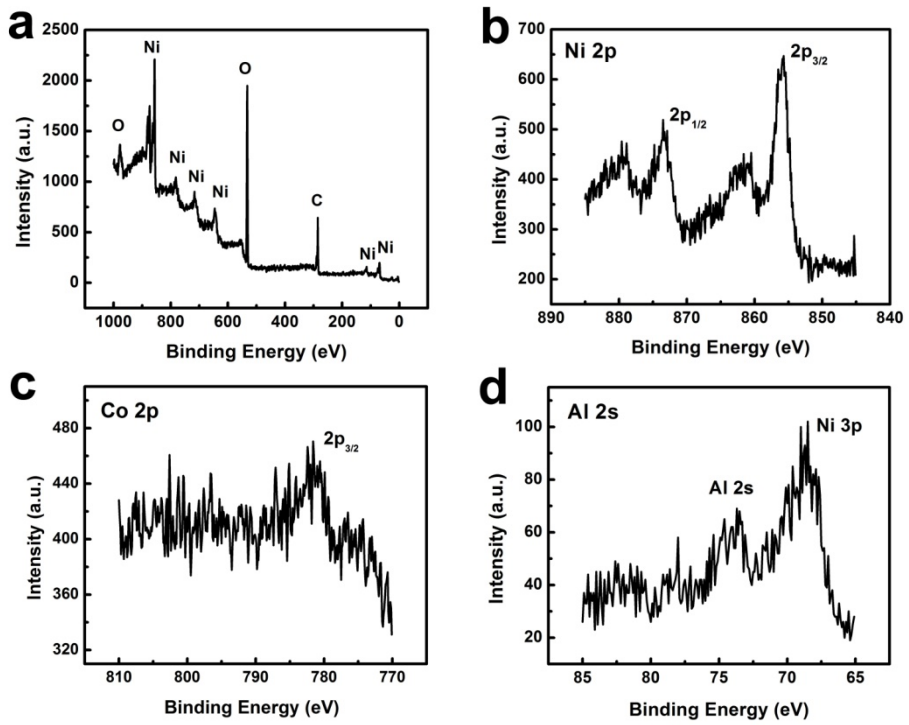

Figure S1. a) XPS survey spectrum, b) high-resolution Ni 2p (c) Co 2p and (d) Al 2s spectra of NiAlCo LDH/CNT hybrid material.

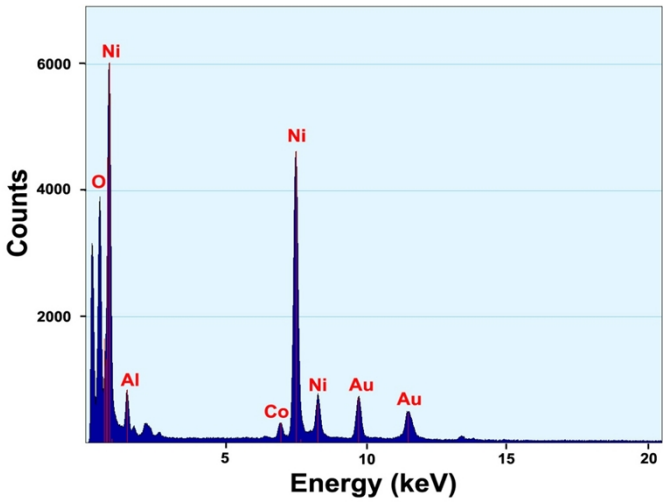

Figure S2. Energy dispersive spectrum (EDS) of the NiAlCo LDH/CNT hybrid material on a gold grid. The Ni/Al/Co atomic ratio was estimated to be 87.5/7.9/4.6.

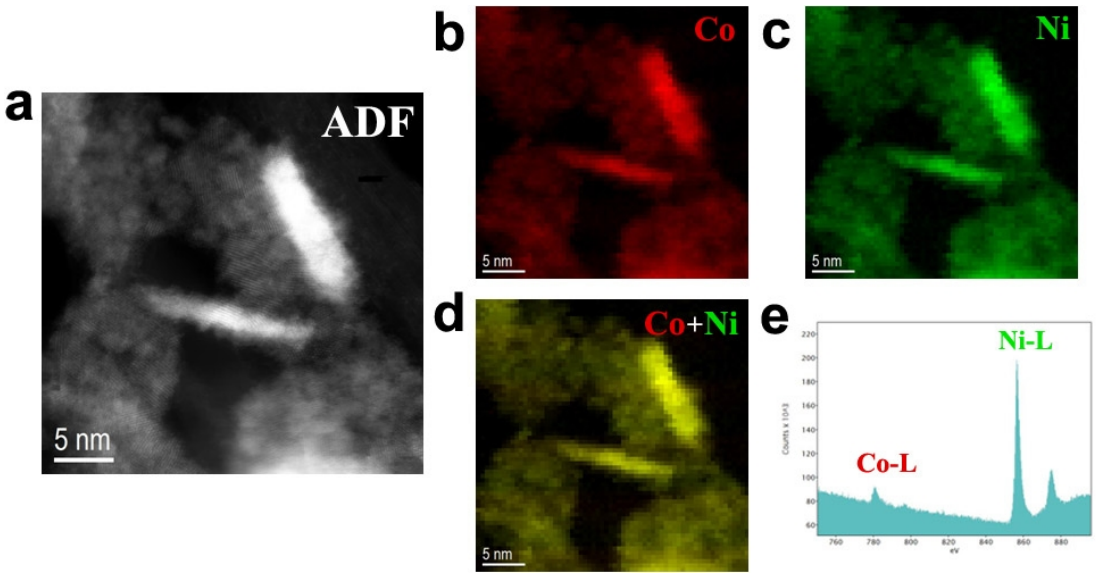

Figure S3. a) Simultaneously acquired ADF image during EELS spectrum imaging for chemical mapping. The separated nanoparticle structure in individual nanoplate was due to destruction of hydroxide into oxide under electron beam. (b-c) Chemical maps for the spatial distribution of Co and Ni. (d) Reconstructed chemical map with Co in red and Ni in green, showing that uniform distribution of Co and Ni in NiAlCo LDH. (e) EELS spectrum of NiAlCo LDH/CNT. Segregated structure in individual plate is due to the fact that hydroxide is generally sensitive to electron beam, which decomposes hydroxide into oxide.

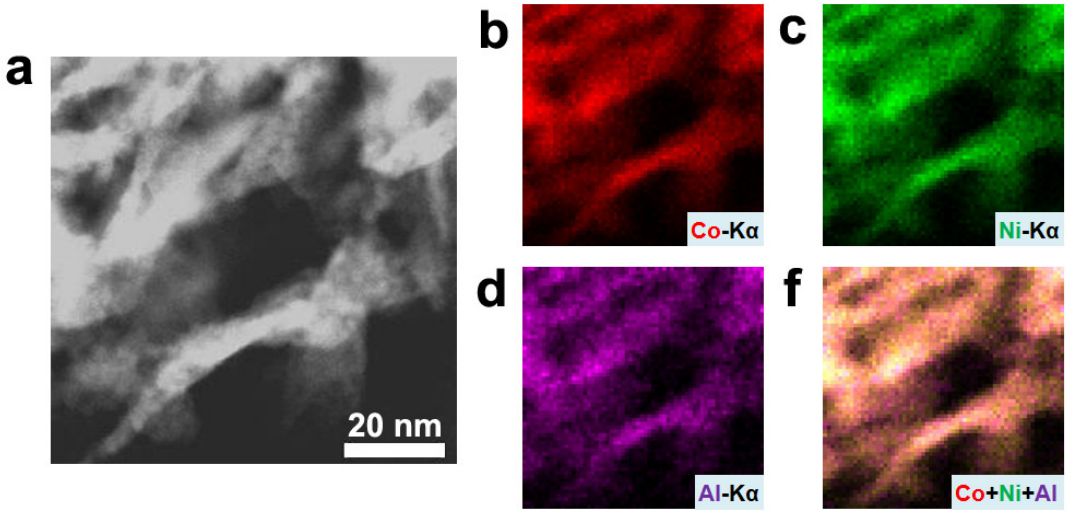

Figure S4. a) Simultaneously acquired ADF image during EDS spectrum imaging for chemical mapping. (b-c) Chemical maps for the spatial distribution of Co, Ni and Al. (d) Reconstructed chemical map with Co in red and Ni in green and Al in purple, showing that uniform distribution of Co, Al and Ni in NiAlCo LDH.

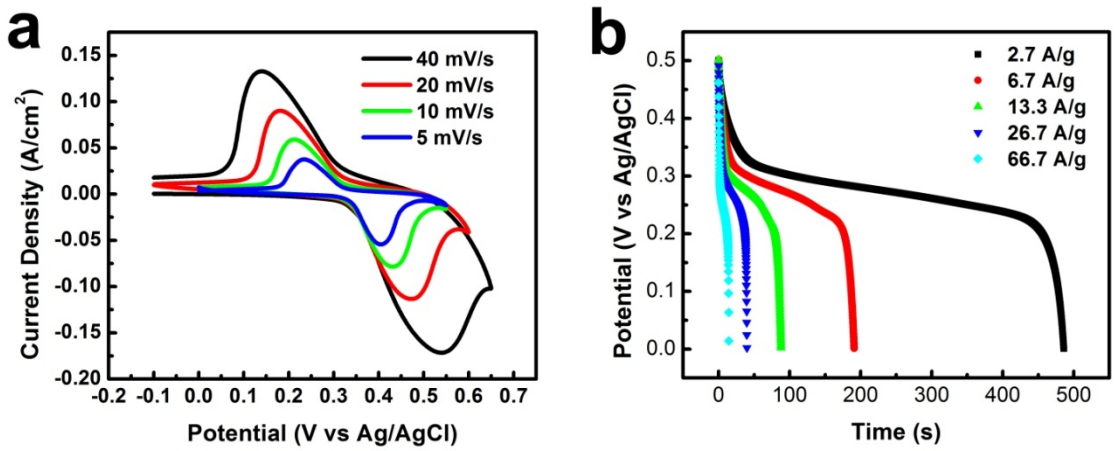

Figure S5. a) CV curves of NiAlCo LDH/CNT under different scan rates in 1 M KOH. B) Galvanostatic discharge curves of NiAlCo LDH/CNT at different discharge current density in 1 M KOH.

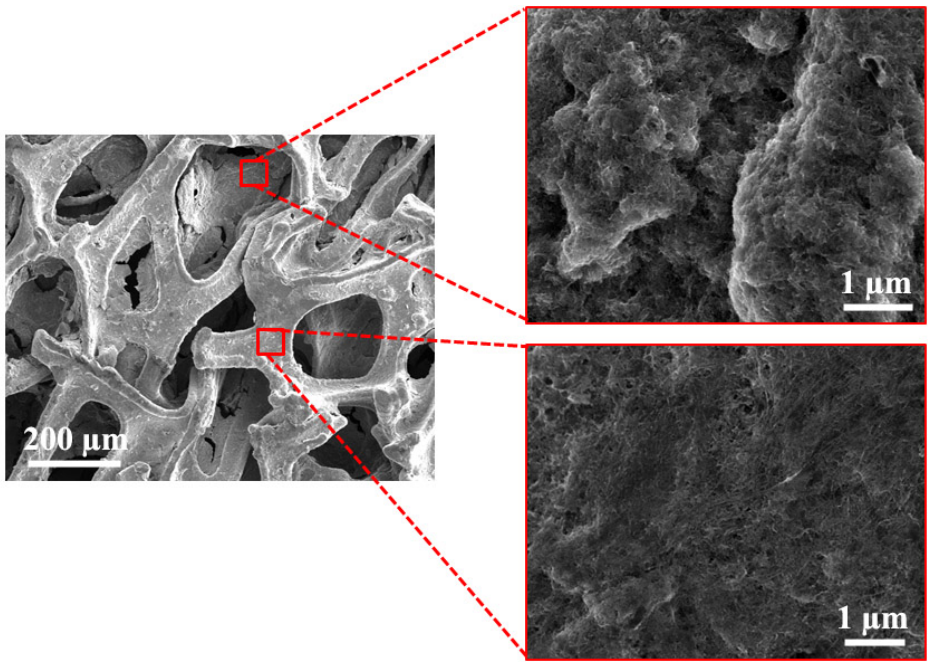

Figure S6. SEM image of full electrode (NiAlCo LDH/CNT on Ni foam at a loading of 20 mg/cm^2)

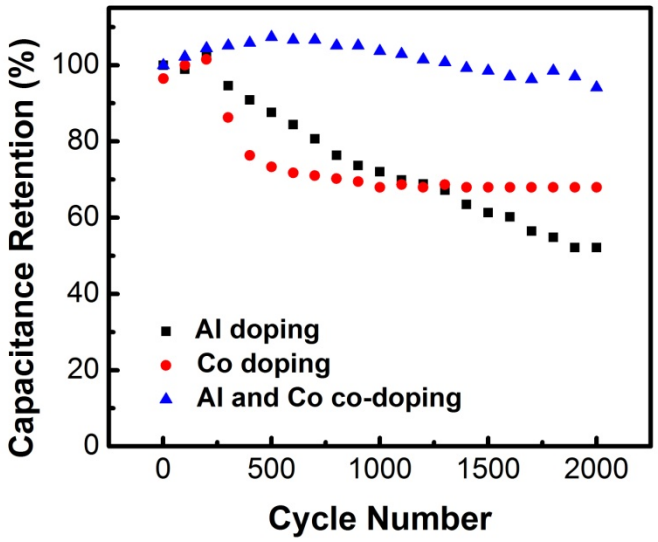

Figure S7. Capacity retention vs cycle number of the hybrids with Al doping, Co doping and Al, Co co-doping at charge/discharge current density of 66.7 A/g in 1 M KOH

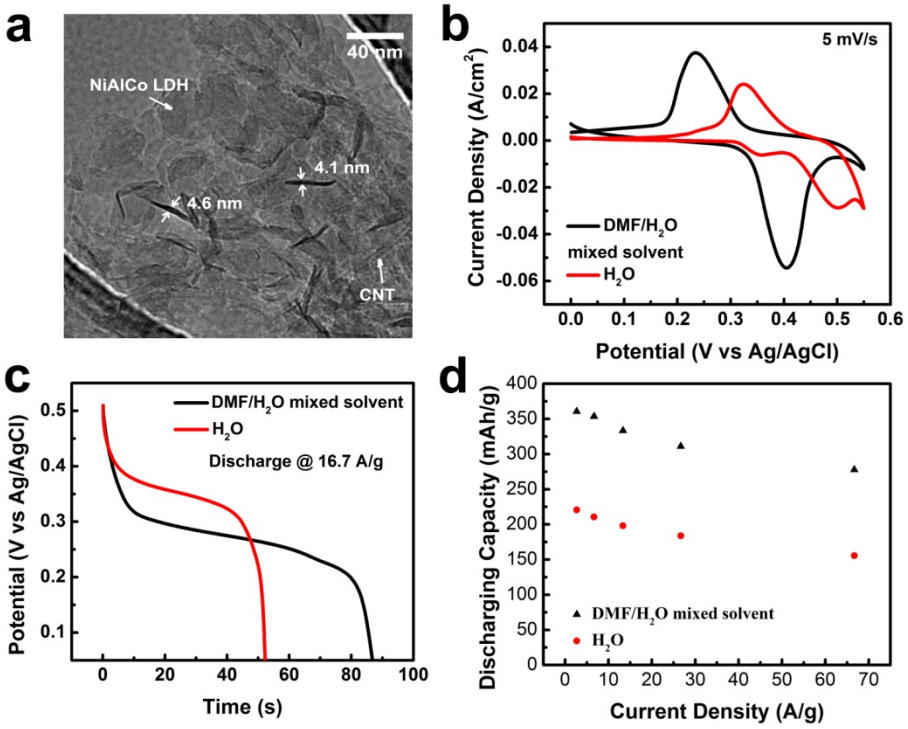

Figure S8. a) TEM images of NiAlCo LDH/CNT synthesized by replacing DMF with H_2O in the solvothermal step of synthesis. b) CV curves of NiAlCo LDH/CNT by DMF method (black) and H_2O method (red) under 5 mV/s in 1 M KOH. c) Galvanostatic discharge curves of NiAlCo LDH/CNT at discharging current density of 16.7 A/g. d) Calculated specific discharging capacity relative to the mass of active material under different current density.

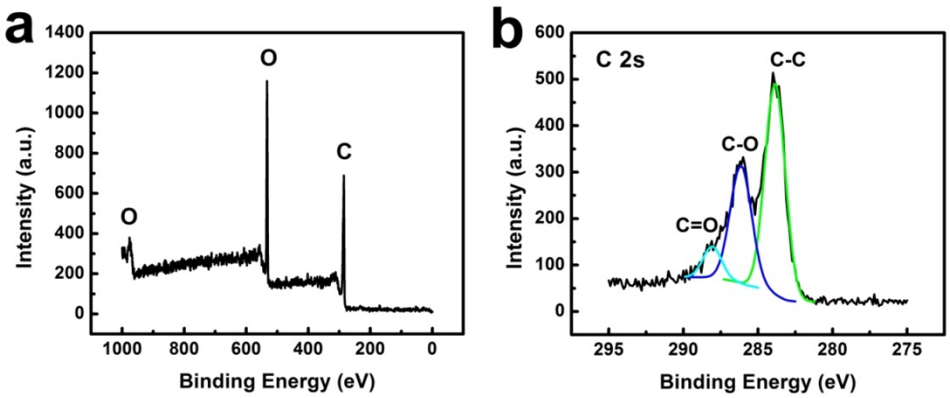

Figure S9. a) XPS survey spectrum, b) high-resolution C 2s spectra of few-walled CNT

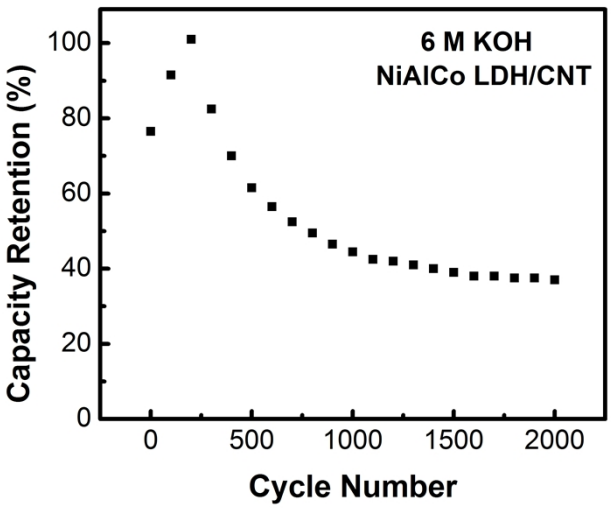

Figure S10. Capacity retention vs cycle number of the NiAlCo LDH/CNT hybrid at charge/discharge current density of 66.7 A/g in 6 M KOH.

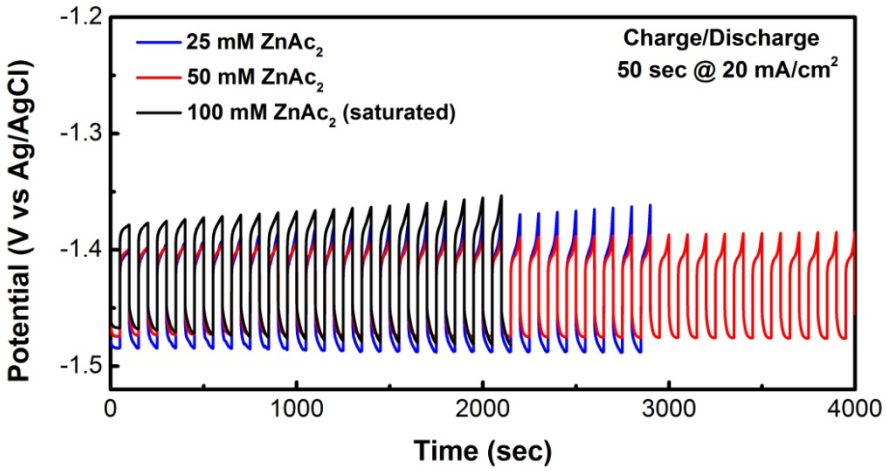

Figure S11. Charge-Discharge cycling curves of electro-deposited Zn on Cu foil with an area of 1 cm^2 at the current density of 20 mA/cm^2 in 1 M KOH containing $ZnAc_2$ with different concentration. The charge/discharge time was fixed at 50 second matching the discharge time of NiAlCo LDH/CNT. Zinc was pre-deposited cathodically for 100 sec at 20 mA/cm^2 prior to measurement to obtain good cycling stability.

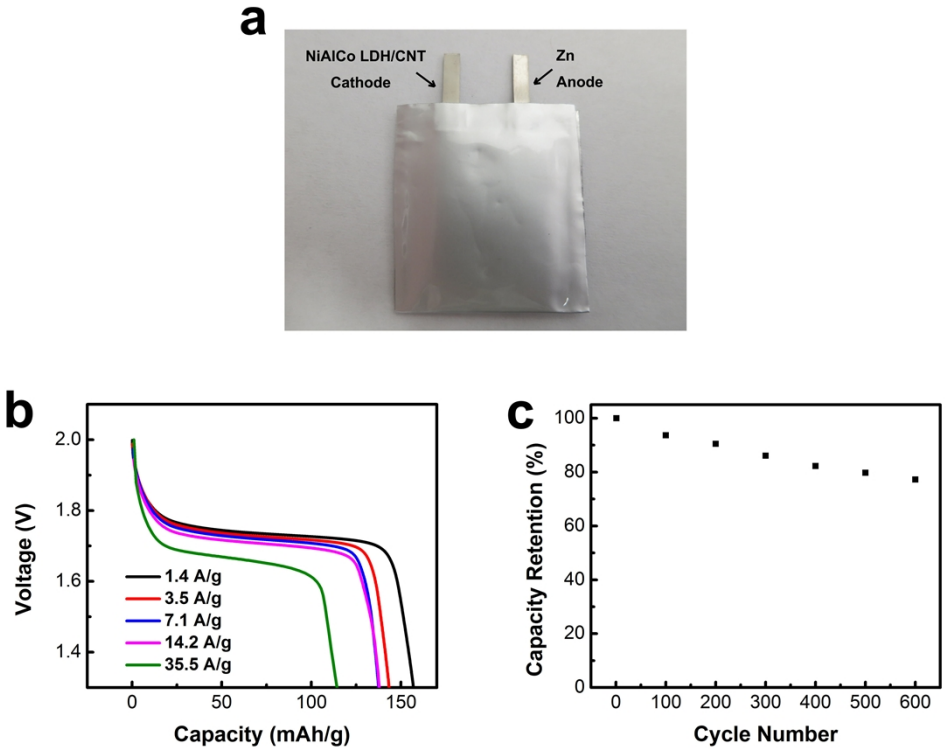

Figure S12. a) Illustration of NiZn pouch cell (~1.0 mg NiAlCo LDH/CNT). b) Galvanostatic curves of NiZn pouch cell at various current density. c) Discharge capacity stability curves (capacity retention vs cycle number) of the NiZn pouch cell at the current density of 14.2 A/g.