Supplementary Information (SI) for Journal of Materials Chemistry C.

Supporting Information

1. Experimental details

1.1 Chemicals and reagents

Nickel (II) nitrate hexahydrate and Cobalt(II) nitrate hexahydrate, Polyvinylpyrrolidone (PVP), from DAEJUNG; Trimesic acid (TA) from Sigma Aldrich, and N, N-Dimethylformamide (DMF) from Alfa Aesar were utilized in their original states

1.2 Characterization

The sample degree of crystallinity was assessed using Cu Kα radiation at a wavelength of 1.5406 Å, which was used in an X-ray diffraction system fitted with a Shimadzu XRD-6100 diffractometer. Using a field emission scanning electron microscope (FESEM), more precisely a Hitachi S-4800 model, the morphology and particle sizes of the precursors and final products were investigated. A 200 kV accelerating voltage transmission electron microscope (HRTEM), specifically the Tecnai G2 F20 S-Twin model, was used to further study the products' microscopic structures. A Thermo Scientific X-ray photoelectron spectroscopy (XPS) setup using Al Kα radiation was used to investigate the elemental composition and valence states.

1.3 Pre-treatment of Ni-foam

First, the nickel foam substrate, which had a thickness of 2 mm, was divided into rectangles of 15 × 5 mm^2. This was followed by immersing the nickel foam in a solution made up of a 3:1 mixture of DI and 20% hydrochloric acid. After that, the surface oxide was removed by ultrasonic cleaning for fifteen minutes. After that, the nickel foam was immersed in DI water and put through one more ultrasonic cleaning cycle lasting 15 minutes, with fresh DI water added at the end of each cycle. Six iterations of this method were conducted. Afterward, ethanol was added instead of DI water, and the surface was cleaned once more to remove any last traces of acid and organic material. Last but not least, the cleaned nickel foam had to be dried at 90 °C for 12 hours in a vacuum oven.

1.4 Electrochemical tests

A typical three-electrode setup was used to assess the electrodes' electrochemical performance. The Hg/HgO reference electrode and the platinum mesh counter electrode made up this configuration. The working electrode was created by mixing N-methyl-2-pyrrolidone (NMP) in a specified mass ratio with the active material, carbon black, and polyvinylidene difluoride (PVDF). After applying the resultant slurry to nickel foam by the drop casting technique, it was allowed to dry for 12 hours at 90 °C in the oven. To evaluate the electrode's electrochemical activity, some electrochemical methods were used, such as cyclic voltammetry (CV), galvanostatic charge-discharge (GCD), and electrochemical impedance spectroscopy (EIS). GCD testing was run across a specified voltage range at various current densities, while CV tests were run at various scan rates in an aqueous solution of 1 M KOH. At the open-circuit potential, EIS measurements were performed at various frequencies. An electrochemical workstation called the Biologic SP-200 was used for all of the electrochemical studies. Explains how the specific capacitance (Csp) of a three-electrode cell was calculated by integrating the discharge curves.

$$C_{sp}\left(\frac{f}{g}\right) = \frac{I\,\Delta t}{\Delta V\, m} \tag{1}$$

where I is current, Δt *is discharge time, and* ΔV is potential.

1.5 Preparation of gel electrolyte

First, we dissolved 5.6 grams of PVA in 50 milliliters of pure DI water at 90 °C, stirring well to create a clear solution. This was the first step in creating the alkaline polyvinyl alcohol/potassium hydroxide (PVA/KOH) gel electrolyte. A clear, viscous solution was formed after about an hour. Then, a gel-like solution was created by gradually adding 6 grams of KOH, which had been completely dissolved in 10 milliliters of DI, and stirring constantly to the cleared PVA solution. Before being used any further, the PVA/KOH gel electrolyte was finally allowed to cool to room temperature.

1.6 MD-Co/Ni-4//AC device fabrication

A hybrid coin-cell-type asymmetric supercapacitor (HS) device was developed with MD-Co/Ni material on Ni foam as the positive electrode, and active carbon as the negative electrode, is separated with filter paper as separator. The positive electrode was prepared by coating slurry of MD-Co/Ni-4, carbon black, and PVDF in a mass ratio of 90: 5:5 in NMP, on nickel foam, dried at 60 °C. The prepare the negative electrode, the aqueous slurry of the activated carbon (AC) was directly coated onto Ni foam and dried at 60 °C. The mass of the electrode was measured both before and after the coating process.

For an ASC, the device typically consists of two electrodes with different charge storage mechanisms, such as a battery-type electrode (faradaic redox reactions) and a capacitive electrode (electric double-layer capacitance). Ensuring an optimal mass ratio between the two electrodes is crucial to maintain charge balance and maximize the performance of the device. This is often based on the principle that the charge stored on the positive electrode (Q+) should equal the charge stored on the negative electrode (Q-), expressed as:

$$Q^{+}=Q^{-} \tag{2}$$

Where $Q=C\times V$, C is the specific capacitance, and V is the potential window.

Given that the specific capacitance of the electrodes may differ, the mass loading (m) of the active materials must be adjusted to balance the charges. This can be expressed by:

$$m^+/m^-=(C^-\times V^-)/(C^+\times V^+) \quad (3)$$

Where m+ and m− are the mass loadings of the positive and negative electrodes, and C^+, C^-, V^+, and V^- are the specific capacitance and potential windows of the positive and negative electrodes, respectively. The estimated mass of the electrode is 1.02 mg/cm^2.

In addition, the E_d (Wh/kg) and P_d (W/ kg) of the device were assessed on the entire mass of the energetic materials, as per Eqs. S2 and S3:

$$E_d = \frac{1}{2}\left[\frac{C_s\,(V_f - V_i)^2}{3.6}\right] \quad (4)$$

$$P_d = \frac{3600\;X\;E_d}{\Delta t} \quad (5)$$

where Δt and $(V_f - V_i)$ are discharge time (s) and potential window for discharge process (V), respectively.

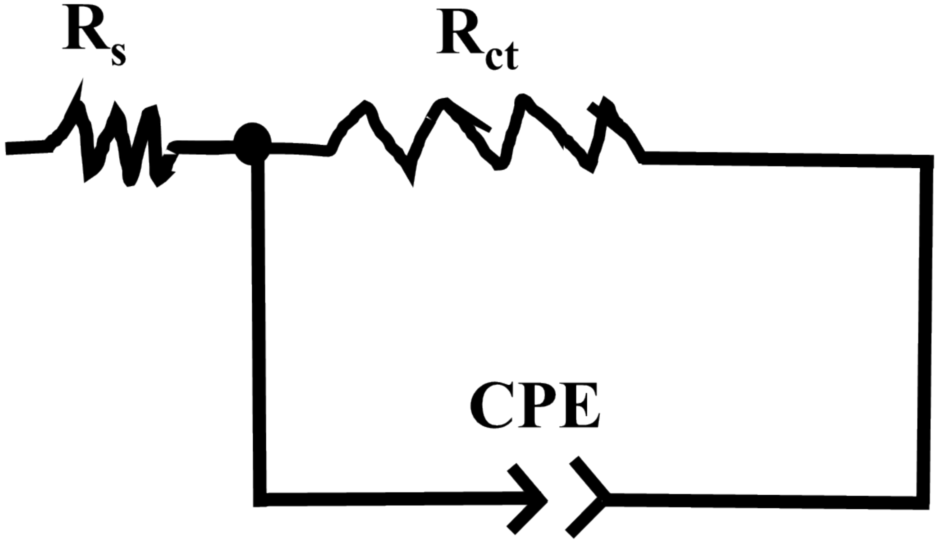

Fig. S1 Equivalent circuit diagram for MD-Co/Ni-4 nanostructure.

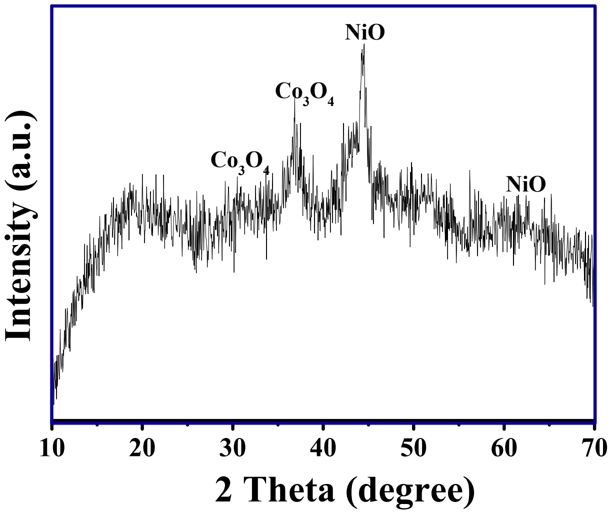

Fig. S2 XRD pattern of the MD-Co/Ni-4 nanostructure post-cycling test.

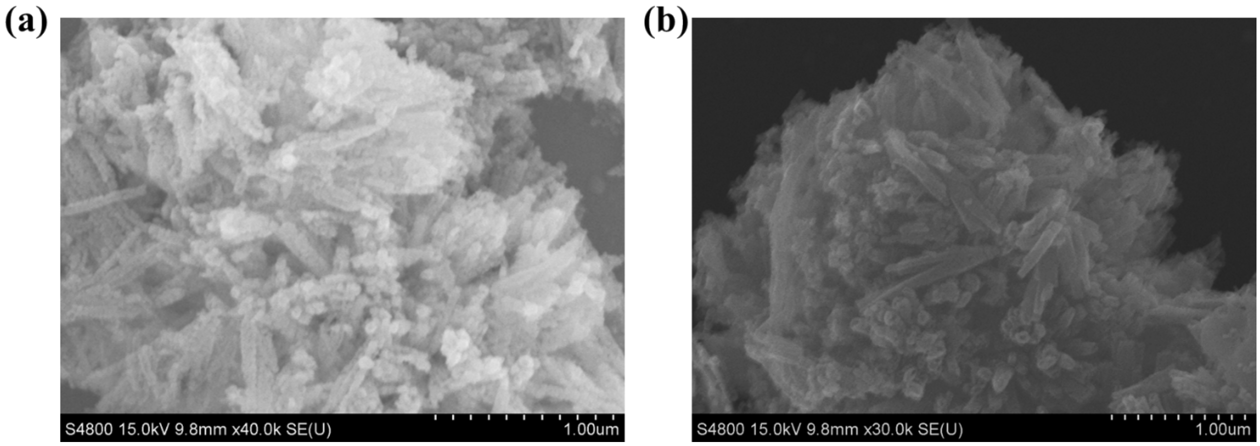

Fig. S3 FESEM images: (a) prior to the cycling test and (b) post-cycling test.