Supplementary Material

S1.1. Characterization

The sample degree of crystallinity was assessed using Cu K_α 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. By combining energy dispersive X-ray spectroscopy (EDS) with a field emission scanning electron microscope (FE-SEM), the elemental makeup of the active materials was determined. FTIR analysis is measured using the Perkin Elmer (Spectrum 100). A Thermo Scientific X-ray photoelectron spectroscopy (XPS) setup using Al K_α radiation was used to investigate the elemental composition and valence states.

S1.2. Pre-treatment of Ni-foam

First, the nickel foam substrate, which had a thickness of 2 mm, was divided into rectangles of 15×10 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.

S1.3. 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) with the active material, carbon black, and polyvinylidene difluoride (PVDF) (8:1:1). 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). The weight of each electrode was determined with the microbalance prior to and after the preparation. An electrochemical workstation called the Biologic SP-200 was used for all of the electrochemical studies.

$$Cs\ (C/g) = I\Delta t / m \quad \text{(S1)}$$

$$Qs\ (mAh/g) = I\Delta t / (3.6m) \quad \text{(S2)}$$

where: I = discharge current in amperes (A), Δt = discharge time in seconds (s), and
m = mass of active material in grams (g)

As per the power law, scan rate dependence on CV current can be written as:

$$i = av^b \quad \text{(S3)}$$

where ‘b’ and ‘a’ are adjustable parameters. The ‘b’ value can be estimated from the slope of the graph between log(i) vs log(v), plotted at a fixed potential. The value of ‘b’ also specifies that the current is the combination of surface-controlled (capacitive) and diffusion-controlled currents:

$$i\,(V) = k_1(v) + k_2(v^{1/2}) \quad \text{(S4)}$$

where $k_1(v)$ and $k_2(v^{1/2})$ represent the capacitive and adsorption/desorption currents, respectively. The values of $k_1$and k_2 can be obtained by plotting i(V)/ $v^{1/2}$ along the y-axis and $v^{1/2}$ along the x-axis. The slope and intercept of this linear fit corresponds to the value of $k_1$and k_2, respectively, and can be used to explain the contribution of the capacitive and

diffusion-controlled processes to the total charge storage.

S1.4. 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.

S1.5. Cu/Ni-O//AC hybrid device fabrication

A hybrid coin-cell-type supercapacitor (HS) device was developed with Cu/Ni-O material on Ni foam as the positive electrode, and activated carbon as the negative electrode, separated by filter paper soaked in PVA/KOH gel electrolyte. The positive electrode was prepared by coating slurry of Cu/Ni-O, carbon black, and PVDF in a mass ratio of 90: 5:5 in NMP, on nickel foam, dried at 60 °C. The prepared negative electrode, the aqueous slurry of the activated carbon (AC) was directly coated onto Ni foam and dried at 60 °C. The Cu/Ni-O//AC supercapacitor device was prepared based on masses of the active materials 1.1 mg for Cu/Ni-O positive electrode and 2.2 mg for AC negative electrode. Thus, the mass ratio m^+/m^- of the experimentally obtained supercapacitor equals 0.50 (1:2) with a total mass of the active material 3.3 mg. Calculation of the charge balanced mass ratio (S5):

$$m^+/m^- = (C^- \times \Delta V^-) / Q^+ \quad \text{(S5)}$$

where Q^+ (C/g) is the specific capacity of the Cu/Ni-O positive electrode, C^- is the specific capacitance of the AC negative electrode, and ΔV^- (V) is the potential window of the AC electrode, all obtained from three-electrode GCD measurements at 1 A/g.

Based on the specific capacity of Cu/Ni-O electrodes equal to 467.1 C/g at current density 0.5 A/g and the specific capacitance of AC equal to 110 F/g at current density 1 A/g with 1.0 V potential window, the mass ratio is:

$m^+/m^- = (110 \times 1.0)/467.1 = 0.236$

This amounts to a positive-to-negative mass ratio of about 1:4.25. This figure denotes a benchmark estimation from measurement results obtained at various current densities and not the optimum mass ratio that can be determined. In this regard, the mass ratio used in the experiment of 1:2 denoted a larger charge storage capacity at the positive electrode as compared to the negative electrode under the experimental conditions. The total active material mass of both electrodes amounted to 3.3 mg.

In addition, the specific capacitance (F/g), energy density (E_d) (Wh/kg) and power density (P_d) (W/kg) of the device were estimated.

$$Cs = 2I\int V(t)\,dt / m\,(\Delta v)2 \qquad \text{(S6)}$$

$$E_d = I\int V\,dt / (3.6\,m) \qquad \text{(S7)}$$

$$Pd = 3600E / \Delta t \qquad \text{(S8)}$$

Where I is the discharge current (A), $\int Vdt$ is the voltage-time integral (V·s), m is the total active mass (g), ΔV is the discharge voltage window (V), E is the energy density (Wh/kg), P is the power density (W/kg), and Δt is the discharge time (s).

Table S1. Electrochemical performance of the fabricated supercapacitor device compared with recently reported Cu-based materials.

Device/material	**Window /Type**	**Specific Capacitance (F/g@A/g)**	**Energy density (Wh/kg)**	**Power density (W/kg)**	**References**
PANI/NiO–CuO symmetric device	1.6 V	157.5 @ 3.5	35.0	1326.31	[45]
CDs/CuO–NiO@Ni foam // AC	1.5 V	635 @ 5 mV/s	20.3	151.1	[46]
C/Ru@NiO–CuO // rGO/α-Co3O4	Solid-state ASC	152.5 @ 0.5	21.18	1800	[47]
RGO@Cu–NiO symmetric device	SSD	261.25 @ 1.5	43.54	750	[48]
CuO–NiO/PANI@Ni foam // AC	ASC	1806.3	37.3	400	[49]
CuO–NiO // AC	ASC	752.72 @ 1	43.39	750	[50]
CuO–NiO@rGO/PANI // activated-carbon fabric	ASC	Device tested at high current density	32.7	4745.6 max PD	[51]
CuO–NiO composite asymmetric device	1.6 V	112.8 @ 0.3	40.1	212.6	This work