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Facilely Synthesized NiCo₂O₄/NiCo₂O₄ Nanofile Arrays Supported on Nickel Foam by a Hydrothermal Method and Their Excellent Performance for High-Rate Supercapacitance

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Abstract: NiCo₂O₄ nanoleaf arrays (NCO NLAs) and NiCo₂O₄/NiCO₂O₄ nanofile arrays (NCO/NCO NFAs) material was fabricated on flexible nickel foam (NF) using a facile hydrothermal approach. The electrochemical performance, including the specific capacitance, charge/discharge cycles, and lifecycle of the material after the hydrothermal treatment, was assessed. The morphological and structural behaviors of the NF@NCO NLAs and NF@NCO/NCO NFAs electrodes were analyzed using a range of analysis techniques. The as-obtained nanocomposite of the NF@NCO/NCO NFAs material delivered outstanding electrochemical performance, including an ultrahigh specific capacitance (Cs) of 2312 F g^{-1} at a current density of 2 mA cm⁻², along with excellent cycling stability (98.7% capacitance retention after 5000 cycles at 5 mA cm⁻²). These values were higher than those of NF@NCO NLAs (Cs of 1950 F g^{-1} and 96.3% retention). The enhanced specific capacitance was attributed to the large electrochemical surface area, which allows for higher electrical conductivity and rapid transport between the electrons and ions as well as a much lower charge-transfer resistance and superior rate capability. These results clearly show that a combination of two types of binary metal oxides could be favorable for improving electrochemical performance and is expected to play a major role in the future development of nanofile-like composites (NF@NCO/NCO NFAs) for supercapacitor applications.

Keywords: nickel foam (NF); NiCo₂O₄ nanoleaf; NiCo₂O₄ / NiCo₂O₄ nanofile; Superior electrochemical performance; hydrothermal approach; supercapacitor

1. Introduction

One of the greatest scientific and technological societal challenges of the present time is achieving secure, sustainable, and highly effective energy conversion and storage equipment system [1–3]. Over the last few decades, a range of energy storage devices, such as solar cells, batteries, lithium-ion batteries, electrochemical capacitors, fuel cells, and supercapacitors (SCs), have been reported [4]. Among the above energy storage devices, SCs have considerable potential and are used widely in storage devices owing to their rapid charge-discharge cycle performance, outstanding cyclability, eco-friendly nature, long cycling life span, and high rate abilities [5–7]. Various materials for faradic redox reactions (pseudocapacitors (PCs), such as Co_3O_4 , NiO, MnO₂, and RuO₂, have been used as electrodes to achieve high energy capability for SCs [8,9]. Nevertheless, the lower electrical conductivity of traditional PCs limits their utility because they cannot provide high performance at high current densities. Recently, new types of bimetallic oxides (BMOs), such as $CuCo_2O_4$, $CoMoO_4$, NiMoO₄, MnMoO₄, and NiCo₂O₄, have attracted considerable attention in the field of SCs because of their

high energy storage performance, including high electrical conductivity and superior energy density compared to common PC oxides [10–12]. Therefore, it is important to develop various bimetallic oxide materials for higher performance SCs.

Among the BMOs available, NiCo2O4 has attracted attention for use in high performance electrochemical SCs applications owing to its outstanding electrochemical conductivity, high theoretical capacitance, and rapid redox activities [13-15]. Different types of nanostructures (NiCo₂O₄), including nanoplates/sheets, nanoparticles/wires and needles, nanospheres/tubes, and nanorods, have been fabricated for SC applications [16–18]. For example, Zou et al. fabricated chain-like NiCo₂O₄ nanowires using a facile hydrothermal and thermal decomposition approach and reported a specific capacitance (Cs) of 1284 F g^{-1} at 2 A g^{-1} [19]. Zhang et al. synthesized hierarchical mesoporous spinal NiCo₂O₄ via a simple hydrothermal and post annealing process and reported a Cs of 1619.1 F g^{-1} at a 2.0 A g^{-1} [20]. Lei et al. synthesized three-dimensional (3D) hierarchical flower-shaped NiCo₂O₄ microspheres through a microwave-assisted heating reflux method and a delivered a Cs of 1006 F g^{-1} at 1 A g^{-1} [21]. Moreover, Yedluri et al. fabricated NiCo2O4 nanoplates by a simple chemical bath deposition approach and reported a Cs of 1018 F g^{-1} at 20 mA g^{-2} [22]. Recently, Yuan et al. synthesized ultrathin mesoporous NiCo2O4 nanosheets through a co-electrodeposition approach and reported a Cs of 1450 F g⁻¹ at 20 A g⁻¹ [23]. Zhu et al. fabricated a spinal NiCo₂O₄ electrode by facile hydrothermal synthesis and achieved a high Cs of 1254 F g^{-1} at 2 A g^{-1} [24]. Zou et al. synthesized NiCo₂O₄ nanowires through a hydrothermal approach and reported a high Cs of 1284 F g^{-1} at 2 A g^{-1} [25]. Despite these studies highlighting the advantages of NiCo2O4 electrodes, their poor specific capacitance still limits their wider commercialization; the very low energy and power densities are challenges that remain to be solved.

The use of active materials in SCs should be improved to increase the specific capacitance, cycling stability, and energy storage. Combining two types of BMOs to form unique hierarchical nanostructures is an efficient way to improve the electrochemical (EC) performance. For example, Cui et al. prepared NiCo₂O₄@MnMoO₄ nanocolumn array structures via a two-step hydrothermal route, which delivered a Cs of 1705.3 F g^{-1} at 5 mA cm⁻² [26]. Mai et al. synthesized MnMoO₄@CoMoO₄ nanowires via a facile hydrothermal method and achieved a high Cs of 187.1 F g^{-1} at 1 A g^{-1} [27]. In addition, Yuan et al. used a facile two-step hydrothermal route to develop a NiCo2O4@MnMoO4 core-shell nanoarray skeleton, exhibiting a high Cs of 1169 F g^{-1} at 2.5 mA cm⁻² [28]. Furthermore, Cheng et al. reported a core/shell NiCo₂O₄@NiCo₂O₄ nanocactus for SC applications, showing a super high Cs of 1264 F g^{-1} at 2 A g^{-1} [29]. Recently, Gu et al. used a facile hydrothermal method to synthesize core-shell NiCo₂O₄@MnMoO₄ nanomaterials over a nickel foam substrate and reported a high Cs of 1118 F g⁻¹ at 1 A g⁻¹ and high cycling stability [30]. Zhang et al. presented hierarchical nanospheres of a NiMoO₄@CoMoO₄ electrode material via a hydrothermal method for SCs applications, showing an excellent Cs of 1601.6 F g^{-1} at 2 A g^{-1} with superior cycling stability [31]. Overall, these studies suggested that a combination of two types of BMOs material is crucial for improving the high EC performance. To the best of our knowledge, the performance of an NF@NCO nanoleaf arrays (NLAs) and NF@NCO/NCO nanofile arrays (NFAs) composite is not satisfactory, and it is a big challenge to further improve the energy storage performance of SCs using NF@NCO NLAs and NF@NCO/NCO NFAs composite materials. Herein, we presented the synthesis of NF@NCO NLAs and NF@NCO/NCO NFAs composite nanostructures using a facile hydrothermal approach. Owing to the high surface area and structural features, the NF@NCO NLAs and NF@NCO/NCO NFAs composite electrodes delivered high specific capacitances with good cycling stabilities when employed as an electrode material for SCs. Compared to the previously reported composite metal oxides, the excellent electrochemical results suggest that the NF@NCO/NCO NFAs composite could be a promising candidate for future research work on SCs applications.

Inspired by the above ideas, this study fabricated a hierarchical NCO/NCO NFAs material on a nickel foam (NF) substrate via a facile hydrothermal technique for high-performance SC applications. Compared to the above electrochemical (EC) performances, the hierarchical NF@NCO/NCO NFAs

electrode material provided a facile synthetic process to simultaneously enhance the specific capacitance (Cs), cycling stability, and electrical conductivity toward flexible high-performance EC applications. The NF@NCO/NCO NFAs scaffold architecture, which grows on another network SC of an NF@NCO NLAs material, provides a large surface area with extensive electrochemical active sites. The NF@NCO/NCO NFAs electrode delivered a high Cs of 2312 F g⁻¹ at 2 mA cm⁻² and excellent cycling stability (98.7% capacitance retention after 5000 cycles) compared to SCs of NF@NCO NLAs (Cs of 1950 F g⁻¹ at 2 mA cm⁻² and 96.3% capacitance retention after 5000 cycles). This type of combination can be expanded to a range of BMOs for various integrated devices to enhance the Cs, cycling life span, and energy storage performance for improved applications.

2. Experimental Methods

2.1. Chemicals and Materials

Analytical grade nickel nitrate hexahydrate (Ni(NO₃)₂·6H₂O), cobalt chloride hexahydrate (CoCl₂·6H₂O), thiourea (CH₄N₂S), hexamethylenetetramine (C₆H₁₂N₄), and potassium hydroxide (KOH) were obtained from Sigma-Aldrich Co. manufacturer, Seoul city, South Korea and used directly in the fabrication of NF@NCO/NCO NFAs.

2.2. Fabrication of the NF@NCO NLAs and NF@NCO/NCO NFAs Materials

The NF@NCO/NCO NFAs and NF@NCO NLAs electrode materials used a simple hydrothermal technique. Before the growth process, the NF (approximately $1.5 \times 2 \text{ cm}^2$) was cleaned sequentially with acetone, 2 M HCl solution, ethanol, and deionized water (DI) with ultrasonication for 20 min each and then dried at 60 °C for 8 h. In a typical hydrothermal procedure, the NF@NiCo₂O₄ NLAs were prepared in a round bottom flask containing a 100 mL of solution of 0.1 M of Ni(NO₃)₂·6H₂O and 0.1 M of Co(NO₃)₂·6H₂O in DI water with vigorous stirring for 1 h using a magnetic stirrer. Subsequently, 0.8 M of CH₄N₂S and 0.08 M of C₆H₁₂N₄ were added slowly to the previous solution with stirring for 2 h. Finally, a clear pink solution was formed under vigorous stirring. The prepared recipe was then transferred to a 100 mL Teflon-lined stainless-steel (TLSS) autoclave with the prepared NF as growth solution and heated at 160 °C for 18 h. After the reaction, the NF@NCO NLAs product electrodes were removed from the TLSS autoclave and cooled naturally to room temperature. The resulting product was then washed with DI water and ethanol several times. Finally, the obtained product was dried at 100 °C for 10 h to obtain the NF@NCO NLAs.

Finally, the NF@NCO/NCO NFAs composite was fabricated using the following process. A NF@NCO/NCO NFAs electrode was also prepared using the above relevant chemicals for the NF@NCO NLAs, including 0.1 M of Ni(NO₃)₂·6H₂O, 0.1 M of Co(NO₃)₂·6H₂O, 0.8 M of CH₄N₂S, and 0.08 M of C₆H₁₂N₄. The as-prepared solution was vigorously stirred, and then NF@NCO NFAs foams were placed vertically in the prepared solution, kept at 150 °C for 20 h in an electric oven, then cooled naturally to room temperature. After the reaction, the obtained Ni foams were removed from the oven and washed sequentially with DI water and ethanol and then dried at overnight at 90 °C in order to obtain the final composite (NF@NCO/NCO NFAs) material. The mass-loading of the active material in NF@NCO/NCO NFAs and NF@NCO NLAs were calculated to be 6 mg cm⁻² and 5 mg cm⁻², respectively.

2.3. Material Characterizations

The fabricated NF@NCO NLAs and NF@NCO/NCO NFAs samples were analyzed by field emission scanning electron microscopy (HR-SEM, Hitachi S4800) to study their microstructure and surface morphology. High-resolution transmission electron microscopy (HR-TEM, CJ111) was performed to investigate the crystalline structure and morphology. X-ray photoelectron spectroscopy (XPS, ESCALAB-250Xi) was conducted to examine the chemical composition and surface states. X-ray diffraction (XRD, D/max-2400, Rigaku at 30 kV and 30 mA) was performed to study the crystal structure.

2.4. Electrochemical Measurements

Cyclic voltammetry (CV), galvanostatic charge-discharge (GCD) analysis, and electrochemical impedance spectroscopy (EIS) of the as-developed electrodes (NF@NCO NLAs and NF@NCO/NCO NFAs) were performed using a Biologic SP-150 electrochemical workstation in a three-electrode setup with a 2.0 M KOH aqueous electrolyte. In the three-electrode setup, the as-developed samples, platinum wire, and Ag/AgCl were used as the working, counter, and reference electrodes, respectively. All electrodes were tested in a 2.0 M KOH electrolyte with CV recorded at the potential window of -0.2 to 0.5 V (V versus Ag/AgCl). GCD was recorded at 0 to 0.4 V versus Ag/AgCl. For the three-electrode setup, the specific capacitance (*Cs*), energy density (*E*, W h kg⁻¹), and power density (*P*, W kg⁻¹) were calculated from the GCD curves using following equations:

$$C_s = \frac{I \times t}{m \times V} \tag{1}$$

$$E = \frac{C_s \times (\Delta V^2)}{2} \tag{2}$$

$$P = \frac{E}{t} \tag{3}$$

where *I* (A) represents current density; *m* (g) is the mass of the electrode; ΔV (V) is the voltage window; and Δt (s) is the discharge time difference [32].

3. Results and Discussion

Figure S1 in the Supplementary material presents schematic illustration of NF@NCO/NCO NFAs. Figure 1a-f presents FESEM images of the NF@NCO/NCO NFAs and NF@NCO NLAs samples. Figure 1a-c depicts the SEM images of the NF@NCO/NCO NFAs on NF substrate at various magnifications. Leaf-like NF@NCO NLAs were grown compactly over the surface of NF@NCO/NCO NFAs. The low-magnification SEM images of NF@NCO/NCO NFAs (Figure 1a-c) showed that the NF was covered uniformly and densely with the as-deposited material. As shown in Figure 1b, the nanofiles were interconnected with each other to form a stable and uniform structure with a network structure and no voids on the NF surface area. The high-magnification image in Figure 1c revealed highly ordered nanofiles grown homogeneously over the surface to form appropriate diffusion channels for electrolyte penetration, which provides a large specific surface area that boosts the intercalation/deintercalation rate, undoubtedly enhances the electrochemical performance, and enables a high transportation efficiency of ions and electrons. FESEM of the as-fabricated leaf-like structure of NF@NCO NLAs at various magnifications revealed a different structure (Figure 1d–f). NF@NCO possesses a type of wire/leaf-like nanostructure morphology with various leaf sizes, a large number of voids, and loosely connected networks surrounded by the surface of the nanoleaf (Figure 1d,e). This type of network delivered weak intercalation between the NCO and NF surface area and resulted in poorer electrochemical performance with lower electrical conductivity, as shown in Figure 1f.

Figure 2 presents HR-TEM, TEM, and HAADF-STEM with EDX images of NF@NCO/NCO NFAs and NF@NCO NLAs. The structure and morphology of the active NF@NCO/NCO NFAs electrodes were analyzed, and are shown in Figure 2a,b. The nanofile morphology was observed over the entire surface of NF@NCO NLAs and the results concur with the FE-SEM images (Figure 1a–c). The HR-TEM image (Figure 2b) showed that the lattice fringe spacing of the files were 0.247 and 0.205 nm, which were assigned to the (311) and (400) planes of the file-like nanostructured NiCO₂O₄, respectively. As shown in Figure 2c,d, the HR-TEM and TEM of the NF@NCO NLAs sample taken

from the NF substrate clearly showed a flake-like nanostructure similar to FESEM (Figure 1d–f). In addition, the HAADF-STEM with EDS mapping analysis revealed a homogeneous distribution and the coexistence of Co, Ni, and O in the NF@NCO/NCO NFAs electrode (Figure 2e,f).



Figure 1. Morphological characterization of NF@NCO/NCO nanofile arrays (NFAs) and NF@NCO nanoleaf arrays (NLAs). FESEM images of the prepared NF@NCO/NCO NFAs (**a**–**c**) and NF@NCO NLAs (**d**–**f**) and their corresponding high magnification images and EDX data (**g**) and (**h**).



Figure 2. TEM and HR-TEM images of the NF@NCO/NCO NFAs (**a**) and (**b**) and NF@NCO NLAs (**c**) and (**d**) grown on nickel foam at different magnifications and STEM-EDS color elemental mapping images of the NF@NCO/NCO NFAs electrode (**e**–**g**).

The fabricated NF@NCO/NCO NFAs and NF@NCO NLAs samples were examined further by powder XRD and XPS to determine the crystal nature and phase purity. Figure 3a shows the XRD patterns of the NF@NCO/NCO NFAs, NF@NCO NLAs, and NF substrate samples, respectively. As shown in Figure 3a, XRD peaks were observed at (111), (220), (222), (422), and (511), and were attributed to the spinal cubic structure of NiCo₂O₄ and well-matched with the JCPDS card number (JCPDS card No. 73-1702) [33]. Other XRD peaks at (311), (400), and (440) were also observed due to the NF substrate and were well-matched with the JCPDS card number (JCPDS card No-04-0850). No other impurity or other peaks were observed, which confirmed the purity of the as-fabricated material.



Figure 3. (a) XRD pattern of the as-synthesized NF@NCO/NCO NFAs, NF@NCO NLAs, and Nickel foam material, (b) XPS survey spectrum of NF@NCO NLAs and NF@NCO/NCO NFAs, and (**c–e**) high-resolution XPS spectrum of Ni 2p (c), Co 2p, and O1s (e) for NF@NCO/NCO NFAs and NF@NCO NLAs.

The oxidation states and elemental identification of NF@NCO/NCO NFAs and NF@NCO NLAs electrodes were examined by XPS, as shown in Figure 3b. Figure 3b shows the survey spectrum of both samples. Ni 2p, Co 2p, and O1s signals with no other peaks were detected, which confirmed that the samples were pure. Figure 3c shows the convoluted Ni 2p spectrum, which exhibits two major peaks and two broad satellites (Sat.), such as Ni $2p_{3/2}$ peaks at approximately 854.0 and 872.0 eV indicating Ni²⁺ and Ni³⁺, respectively, and Ni $2p_{1/2}$ peaks at 855.7 and 873.5 eV for Ni²⁺ and Ni³⁺, respectively [34]. The high-resolution Co 2p spectrum (Figure 3d) revealed peaks at 779.4 and 794.5 eV, which were assigned to Co²⁺, and the Co 2p peaks at 782.5 and 796.1 eV were attributed to Co³⁺, which are in agreement with previous studies. Figure 3e [35] presents the O 1s spectrum, in which there are three main peaks that appear at 529.3, 531.0, and 532.6 eV, corresponding to metal-oxygen bonds and the multiplicity of chemisorbed oxygen-metal bonds within the surface of the samples (NF@NCO/NCO NFAs and NF@NCO NLAs) [36–38].

The electrochemical properties of the NF@NCO NLAs and NF@NCO/NCO NFAs samples were investigated in a three-electrode system with an aqueous 2.0 M KOH electrolyte using CV and GCD techniques. Figure 4a shows the CV curves of the as-prepared electrodes at a scan rate of 50 mV s⁻¹ over the voltage window from -0.2 to 0.5 V, respectively. The CV integrated area and the oxidation and reduction currents of the NF@NCO/NCO NFAs electrode were much larger than that of the NF@NCO NLAs electrode. In addition, the electrocapacitance of both as-prepared electrodes was examined by GCD measurements at 2 mA cm⁻² in a voltage window of 0 to 0.4 V, as shown in Figure 4b. The GCD plots revealed significant differences between the two as-prepared electrodes, which showed a larger enclosed area for the NF@NCO/NCO NFAs than that of the NF@NCO NLAs electrode.



Figure 4. Comparison of (**a**) cyclic voltammetry (CV) and (**b**) galvanostatic charge-discharge (GCD) plots of the NF@NCO NLAs and NF@NCO/NCO NFAs electrodes measured at a constant scan rate and current density. (**c**) CV and (**d**) GCD behaviors of the NF@NCO/NCO NFAs electrodes at various scan rates (2, 25, and 50 mV s⁻¹) and current densities (2–20 mA cm⁻²). (**e**) CV and (**f**) GCD behaviors of the NF@NCO NLAs electrodes at various scan rates (2, 25, and 50 mV s⁻¹), and current densities (2–20 mA cm⁻²).

Figure 4c,e present the CV curves of the as-prepared electrodes (NF@NCO/NCO NFAs and NF@NCO NLAs) analyzed over the voltage window, -0.2 to 0.5 V, at scan rates of 2, 25, and 50 mV s⁻¹. Typical CV curves (Figure 4c,e) of the NF@NCO/NCO NFAs composite electrode material showed a much larger integrated area than that of the NF@NCO NLAs electrode, indicating good ion diffusion, good reversibility of active materials, and high specific capacitance. In addition, the anodic and cathodic peak position shifted to lower and higher potentials, respectively, and exhibited a similar structure, which is an indication of the kinetic reversibility and pseudocapacitance behavior of the as-prepared electrodes. The GCD curves of the NF@NCO/NCO NFAs and NF@NCO NLAs electrodes were analyzed at current densities ranging from 2 to 20 mA cm⁻², as shown in Figure 4d,f. The GCD curves of the two as-obtained electrodes were similar, and a pair of charge-discharge plots that were similar to the reflection process of oxidation and reduction were obtained. Moreover, the flat voltage plateaus in the GCD results indicate typical pseudocapacitive behavior, which is in agreement with the electrochemical characteristics of the CV curves.

The specific capacitance of the NF@NCO NLAs and NF@NCO/NCO NFAs samples at current densities of 2, 5, 10, and 20 mA cm⁻² were 2312, 2172, 2030, and 1782 F g⁻¹, respectively. In contrast, the corresponding specific capacitance of the NF@NCO NLAs electrodes were 1950, 1595, 1328, and 715 F g^{-1} , respectively. These results confirm that the NF@NCO/NCO NFAs electrode had a much higher specific capacitance than the NF@NCO NLAs electrode. The remarkable electrochemical performance of the NF@NCO/NCO NFAs electrode could be attributed to the excellent adhesion to the NF substrate with a large surface area and electrical connection of the active material to the current collector to ensure effective accessibility of the electrolyte ions and electrons. Moreover, EIS was conducted to examine the ion diffusion and electrical conductivity of the as-obtained samples, as depicted in Figure 5b. The measurements were performed over the frequency range 0.01 Hz to 100 kHz using a three-electrode system. The NF@NCO/NCO NFAs composite electrode exhibited a semicircle part in the high frequency region and a vertical line in the low frequency region, which represents the charge transfer resistance (R_{ct}) and equivalent series resistance (R_{s}), respectively. The nanofile-like NF@NCO/NCO NFAs electrode exhibited a lower R_{ct} (0.25 Ω) than the NF@NCO NLAs ($R_{ct} = 0.5 \Omega$) electrode. Moreover, the NF@NCO/NCO NFAs electrode delivered a more vertical line than that of the NF@NCO NLAs electrode, indicating a higher efficient electro-active surface area that results in faster electrolyte ion diffusion during the redox reaction kinetics with excellent conductivity. In addition, the NF@NCO NLAs and NF@NCO/NCO NFAs supercapacitor devices exhibited an excellent stable high capacitance retention of 98.7% and 96.3%, respectively, after 5000 cycles at a current density of 5 mA cm⁻² in a 2.0 M KOH aqueous electrolyte solution. Nevertheless, the NF@NCO/NCO NFAs supercapacitor device has more active sites and numerous pathways for electrolyte ion penetration, which promotes a rapid electrochemical reaction and improves the energy storage performance.



Figure 5. (**a**) Specific capacitance at various current densities, (**b**) Nyquist plots, and (**c**) Cycling stability of the as-prepared NF@NCO/NCO NFAs and NF@NCO NLAs samples.

4. Conclusions

A novel composite material with a NF@NCO/NCO NFAs-like structure was fabricated on an NF substrate using a facile hydrothermal process. The excellent supercapacitance performance of the NF@NCO/NCO NFAs was found to depend mainly on easy pathways between the transportation of electrons and ions, a large electrochemical surface area, more accessible electroactive sites, and great adhesion to the NF substrate to enhance energy storage and exhibit higher electrochemical performance than the NF@NCO NLAs electrode. The nanofile-like NF@NCO/NCO NFAs covered the NF@NCO NLAs surface area fully. As a result, the synthesized hierarchical nanofile-like NF@NCO/NCO NFAs composite electrode exhibited superior energy storage performance, including a high specific capacitance of 2312 F g⁻¹ at a current density of 2 mA cm⁻², as well as a higher cycling stability at a current density of 5 mA cm⁻² (98.7% retention after 5000 cycles) than the NF@NCO NLAs (96.3% retention after 5000 cycles). These results demonstrate the outstanding cycling stability, excellent electrochemical performance, and higher electrical conductivity of the hierarchical nanofile-like NF@NCO/NCO NFAs composite compared to the NF@NCO NLAs composite, highlighting its potential for flexible energy storage systems.

Supplementary Materials: The following are available online at http://www.mdpi.com/1996-1073/12/7/1308/s1, Figure S1: Schematic illustration of NF@NCO/NCO NFAs.

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