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Biomimetic *Setaria viridis*-Inspired $\text{Co}_3\text{O}_4/\text{CoS}$ Hierarchical Architecture on Carbon Cloth for Advanced Lithium Storage

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ABSTRACT

To overcome the severe volume variation and intrinsically low electronic conductivity associated with Co_3O_4 , a $\text{Co}_3\text{O}_4/\text{CoS}@CC$ composite was constructed via sequential hydrothermal synthesis and electroprecipitation. This material employs flexible carbon cloth as the substrate, on which arrays of cobalt tetroxide (Co_3O_4) nanowires are first grown, followed by the electrodeposition of cobalt sulphide (CoS) onto the nanowire arrays. When evaluated lithium-ion batteries, the $\text{Co}_3\text{O}_4/\text{CoS}@CC$ electrode delivered an initial discharge specific capacity of 1152.38 mA h g⁻¹ at 0.1 A g⁻¹. After 200 charge-discharge cycles, it retained a reversible capacity of 613.07 mA h g⁻¹ with a coulombic efficiency of 99.72%. Under a high loading current of 500 mA g⁻¹, the average discharge capacity reached 578.92 mA h g⁻¹. The biomimetic *Setaria viridis*-inspired hierarchical architecture endows the $\text{Co}_3\text{O}_4/\text{CoS}@CC$ electrode with excellent long-term cycling durability and outstanding rate performance, which provides an enlarged specific surface area, an efficient conductive network, and improved structural robustness, thereby significantly enhancing the electrochemical performance.

1. Introduction

The widespread adoption of consumer electronics together has greatly increased the demand for lithium-ion batteries (LIBs), thereby stimulating extensive research on high-performance electrode materials^[1-5]. Co_3O_4 is regarded as a promising

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representative of transition-metal oxides because it combines a high theoretical lithium-storage capacity with excellent redox activity, environmental benignity, and cost-effectiveness^[6]. Nevertheless, the practical application of Co₃O₄ as a standalone anode material is hindered by its intrinsically low electrical conductivity, insufficient utilization of active species, and the substantial gap between the experimentally obtained capacity and its theoretical storage capability, particularly under large current loads or fast scan conditions^[7, 8]. Integrating Co₃O₄ with other functional materials provides a practical route to overcoming these intrinsic limitations. In particular, transition metal sulfides (TMS), especially cobalt sulfide (CoS), have been extensively explored because they possess high theoretical capacity together with superior electrical conductivity^[9-11]. Despite these advantages, substantial volume fluctuations during repeated cycling gradually disrupt the structural integrity of the electrode, thereby compromising cycling durability and rate performance^[12-14]. The introduction of nanostructured CoS provides abundant interfacial exposure and abundant electrochemically accessible regions, thereby accelerating charge transfer and Li⁺ migration while effectively relieving stress-induced structural deterioration^[15, 16].

Carbon cloth (CC) is composed of an interconnected network of micron-sized carbon fibers, forming a stable conductive framework that provides continuous electron transport pathways for active materials while reducing interfacial resistance. In addition to its excellent electrical conductivity, CC exhibits remarkable flexibility and mechanical robustness, making it an attractive current collector for flexible energy storage devices^[17, 18]. Its three-dimensional porous architecture provides abundant space for accommodating electroactive components, including cobalt sulfide and silicon-based materials, etc., while simultaneously facilitating electrolyte infiltration and lithium-ion transport. Furthermore, the interconnected porous framework provides sufficient space to buffer the repeated expansion and contraction of the electrode during electrochemical cycling. These structural and electrochemical merits make carbon cloth an attractive platform for the construction of next-generation electrode architectures^[19, 20].

In this study, three-dimensional (3D) Co₃O₄/CoS array electrodes were fabricated on CC through integrating hydrothermal growth with a subsequent electrodeposition

process to develop high-performance lithium-storage electrodes. Initially, Co_3O_4 was directly formed as aligned nanowire arrays across the surface of the CC support through a single hydrothermal treatment. Subsequently, CoS nanoparticles were deposited onto the Co_3O_4 nanowire arrays by electrodeposition, yielding the $\text{Co}_3\text{O}_4/\text{CoS}$ composite array architecture. Benefiting from this integrated structure, the as-prepared $\text{Co}_3\text{O}_4/\text{CoS}$ composites supported on CC exhibited outstanding electrochemical performance.

2. Experiments

2.1 Materials

Commercially available hydrophilic CC was employed as the supporting substrate for the electrode. The synthesis was carried out using cobalt nitrate hexahydrate ($\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$), urea ($\text{CO}(\text{NH}_2)_2$), ammonium fluoride (NH_4F), cobalt chloride hexahydrate ($\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$), and thiourea (H_2NSNH_2), all of which were purchased from Suzhou Willett Environmental Technology Co. No purification or pretreatment was applied to any of the chemicals before use.

2.2 Preparation of $\text{Co}_3\text{O}_4/\text{CC}$ nanowire arrays

A hydrothermal process was employed to obtain the $\text{Co}_3\text{O}_4/\text{CC}$ composite. To formulate the reaction precursor, 2 mmol of $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, 12 mmol of NH_4F , and 20 mmol of $\text{CO}(\text{NH}_2)_2$ were dissolved in 40 mL of DI and maintained under continuous stirring at ambient temperature for 30 min. A piece of carbon cloth immersed in the resulting precursor solution was loaded into a 50 mL Teflon-lined hydrothermal reactor. Crystal growth was induced by heating the sealed reactor at 110 °C for 5 h. Upon completion of the hydrothermal treatment, the reactor was left to return to ambient temperature naturally prior to opening. The carbon cloth carrying the as-prepared precursor was then removed, thoroughly cleaned with deionized water until no residual reactants, and placed in an oven at 60 °C for 3 h. Finally, thermal conversion of the precursor into Co_3O_4 was accomplished by heating the precursor-coated carbon cloth to 350 °C under a continuous N_2 at a ramping rate of 5 °C min^{-1} , followed by a 2 h dwell at the target temperature to obtain the $\text{Co}_3\text{O}_4/\text{CC}$ composite.

2.3 Preparation of $\text{Co}_3\text{O}_4/\text{CoS}@ \text{CC}$ nanowire arrays

The $\text{Co}_3\text{O}_4/\text{CoS}@ \text{CC}$ sample was fabricated by electrodeposition using a conventional three-electrode configuration. An electrolyte containing $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$

(0.05 M) and H_2NSNH_2 (1.0 M) was first prepared for the deposition process. The $\text{Co}_3\text{O}_4/\text{CC}$ sample was connected as the working electrode, while a platinum plate and Ag/AgCl electrode were used to complete the electrochemical cell. CoS growth was achieved by CV deposition over a potential window ranging from -1.2 V and 0.2 V with 10 mV S^{-1} , and the deposition procedure was repeated for eight consecutive cycles. Upon completion of electrodeposition, the coated CC was sequentially washed using ethanol followed by DI to remove residual electrolyte, after which it was placed in a vacuum oven maintained at 60°C for 3 h.

2.4 Instrumentation

Structural information was obtained from X-ray diffraction measurements performed on a Rigaku Dmax-2000 diffractometer employing $\text{CuK}\alpha$ radiation. The microscopy of the synthesized materials and the spatial distribution of the constituent elements were examined using a JEOL JMS-7800F field-emission scanning electron microscope equipped with an energy-dispersive X-ray spectroscopy detector. X-ray photoelectron spectroscopy (ESCALAB250) was employed to probe the chemical environment and electronic states of the surface elements.

2.5 Electrochemical evaluation

The electrochemical properties of the prepared electrodes were evaluated in CR2032 coin cells. All assembly procedures were completed inside an argon atmosphere under ultra-low oxygen and moisture conditions (O_2 and $\text{H}_2\text{O} < 0.01 \text{ ppm}$). The cell configuration consisted of the prepared electrode, lithium metal foil, and a Celgard 2400 separator. Galvanostatic charge/discharge (GCD) experiments were carried out using a NEWARE multichannel battery analyzer between 0.01 and 3.0 V (vs. Li/Li^+), and CV was acquired on a CHI660e instrument at 0.1 mV s^{-1} over the same voltage window. The impedance response of the cells was subsequently analyzed by electrochemical impedance spectroscopy within the frequency range from 10^{-2} to 10^6 Hz .

3. Results and discussion

The $\text{Co}_3\text{O}_4/\text{CoS}@ \text{CC}$ composite was constructed through sequential hydrothermal growth and electrodeposition, as illustrated in Figure 1. Initially, ammonium fluoride was introduced as a structure-directing agent to enable the in situ hydrothermal growth of the Co_3O_4 precursor on the CC substrate. After subsequent heat treatment, well-

defined Co_3O_4 nanowire arrays were obtained. The $\text{Co}_3\text{O}_4/\text{CoS}@/\text{CC}$ composite was then fabricated by depositing CoS on the Co_3O_4 nanowire arrays through an electrodeposition process. For convenience, the $\text{Co}_3\text{O}_4/\text{CC}$ and $\text{Co}_3\text{O}_4/\text{CoS}@/\text{CC}$ electrodes are referred to as Sample A and Sample B, in the following figure captions.

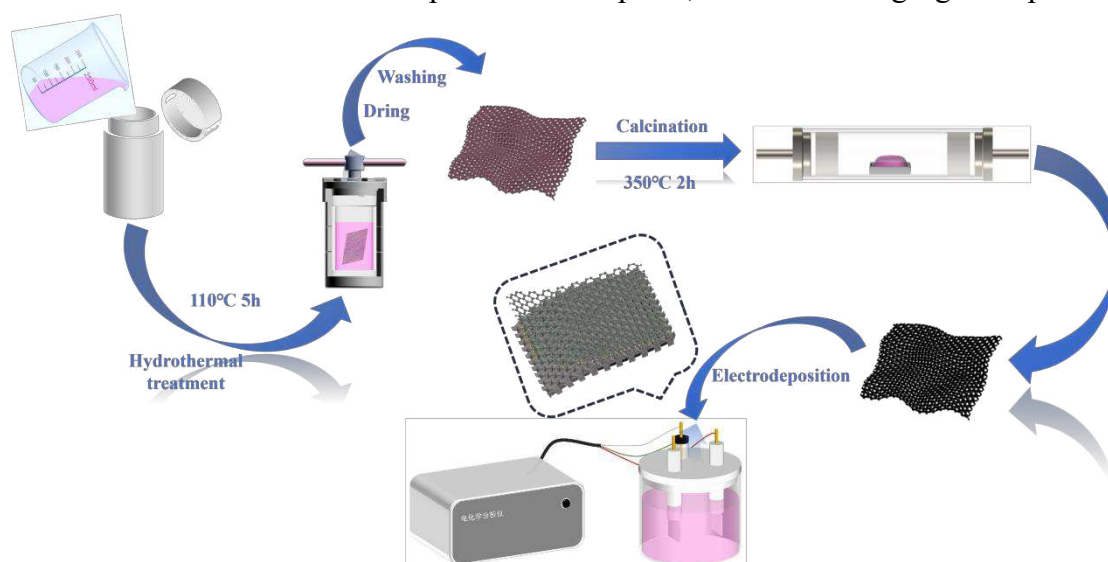


Figure 1 Sequential preparation procedure for constructing Sample A and Sample B.

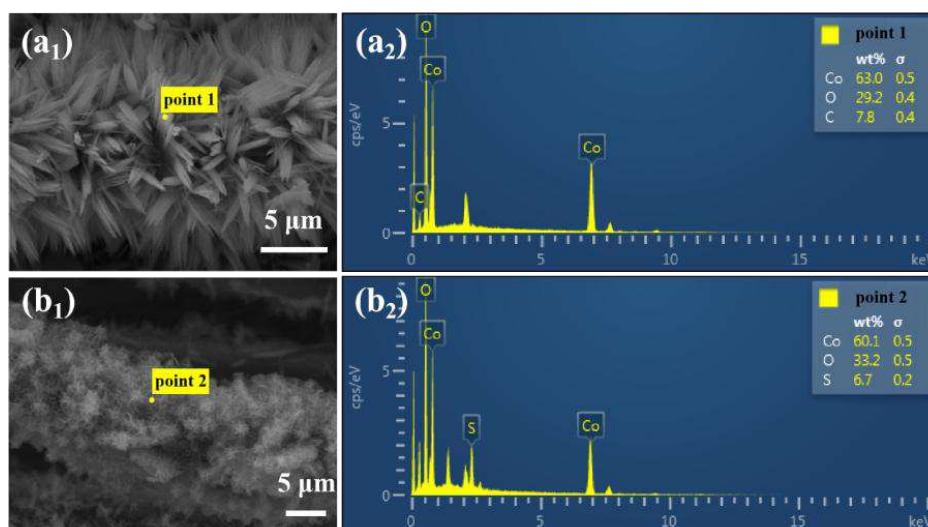


Figure 2. FE-SEM images and point EDS spectra of the two samples: (a₁, a₂) Sample A; (b₁, b₂) Sample B.

The morphological characteristics and elemental composition of Samples A and B were investigated by FE-SEM coupled with point EDS analysis, as illustrated in Figure 2. As shown in Figures 2a₁ and 2a₂, the $\text{Co}_3\text{O}_4/\text{CC}$ electrode is composed of densely distributed nanowire arrays with uniform dimensions and homogeneous morphology.

Statistical measurements indicate that the nanowires are approximately 3.3 μm in length, while the average tip diameter is about 0.5 μm . The corresponding point EDS spectrum confirms Co, O, and C as the major elemental constituents of Sample A. Following electrodeposition, the morphology of the nanowire arrays is retained, while the point EDS spectrum of Sample B, shown in Figures 2b₁ and 2b₂, reveals the coexistence of Co, O, and S, indicating the successful introduction of the sulfide phase.

Representative FE-SEM images together with EDS elemental mapping of the Co₃O₄/CoS@CC composite after electrodeposition are displayed in Figure 3. As presented in Figures 3a and 3b, the surfaces of the Co₃O₄ nanowire arrays are densely covered with nanoscale flower-like structures, resulting in a distinctive biomimetic *Setaria viridis*-inspired hierarchical architecture. This hierarchical morphology is constructed from numerous interconnected nanosheets with a typical size of around 20 nm. Such a structure design effectively enlarges the specific surface area, thereby providing abundant electrochemical reaction interfaces while reducing the migration distance for both lithium ions and electrons. Moreover, the integration of two-dimensional nanosheets with one-dimensional nanowire arrays enhances the mechanical robustness of the electrode, mitigates active-material pulverization under repeated cycling, and consequently improves cycling stability [21, 22].

The corresponding EDS elemental mapping images (Figures 3e-h) demonstrate that the presence of Co, O, C, and S throughout the composite. All four elements are uniformly distributed throughout the electrode, while the S element is mainly concentrated on the surface of the nanowire arrays, providing direct evidence that CoS has been successfully deposited onto the Co₃O₄ nanowire arrays. To examine the morphological integrity of the composite during long-term electrochemical operation, the cell was disassembled after 200 cycles, and the resulting microstructural evolution is presented in Figure S1.

The morphology of the Co₃O₄/CoS@CC sample following 200 galvanostatic cycles at 0.1 A g⁻¹ in Figure S1, that the original morphology undergoes noticeable degradation during prolonged cycling. Nevertheless, portions of the porous architecture remain intact, allowing the electrode to preserve a certain level of capacity after 200 cycles. The biomimetic *Setaria viridis*-inspired hierarchical structure effectively

buffers the mechanical strain associated with Li^+ insertion, thereby contributing to the structural stability of the electrode. However, the observed aggregation and collapse of the nanosheets suggest that repeated lithiation/delithiation gradually obscures the boundaries between adjacent nanosheets, leading to a reduction in the available reaction interface and fewer electrochemically accessible sites remain. This structural evolution represents responsible for the progressive loss in reversible capacity during cycling.

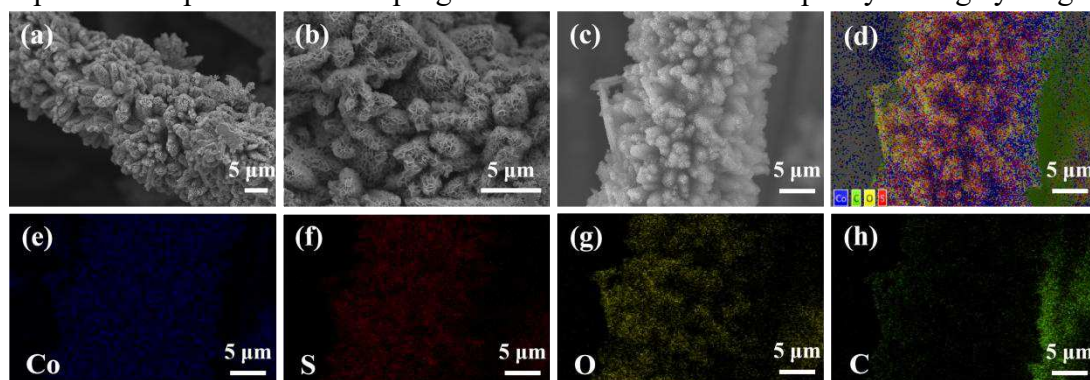


Figure 3 FE-SEM characterization of the $\text{Co}_3\text{O}_4/\text{CoS}@CC$ composite: (a, b) SEM micrographs; (c-h) Corresponding elemental maps of Co, O, S, and C.

Figure 4a compares the XRD profiles of Samples A and B. For the $\text{Co}_3\text{O}_4/CC$ sample, reflections located at $2\theta = 31.3^\circ$, 36.8° , 44.8° , 59.4° , and 65.2° are assigned to the (220), (311), (400), (511), and (440) lattice planes of Co_3O_4 (PDF#43-1003), respectively [23]. Following the electrodeposition of CoS onto the $\text{Co}_3\text{O}_4/CC$ electrode, these diffraction peaks exhibit a systematic shift. This behavior is mainly associated with the incorporation of cobalt sulfide, which promotes interfacial ion interdiffusion and/or solid-solution formation at the $\text{Co}_3\text{O}_4/\text{CoS}$ interface. In particular, sulfide ions (S^{2-}) may interact with the Co_3O_4 lattice in the interfacial region, while changes in the valence state or coordination environment of cobalt ions can generate tensile stress within the lattice. Since S^{2-} possesses a larger ionic radius than O^{2-} , lattice expansion occurs, leading to an increase in the lattice constant [24]. Collectively, these structural features confirm the presence of the $\text{Co}_3\text{O}_4/\text{CoS}$ heterostructure with a well-developed heterointerface.

XPS characterization was carried out to determine the elemental species and chemical environments in the $\text{Co}_3\text{O}_4/\text{CoS}@CC$ composite. The survey spectrum (Fig.

4b) reveals the coexistence of Co, S, O, and C, indicating that all expected elements are incorporated into the fabricated material. Particular attention was first directed to the cobalt and sulfur regions because they provide direct evidence for the formation of the $\text{Co}_3\text{O}_4/\text{CoS}$ heterostructure. As displayed in Fig. 4e, the Co 2p spectrum contains two spin-orbit doublets located at 779.79/781.18 eV and 795.65/797.04 eV, together with satellite features centered at 785.26 and 802.06 eV. The spin-orbit splitting energy is approximately 15.9 eV, which is characteristic of the coexistence of Co^{2+} and Co^{3+} oxidation states^[25,26]. Correspondingly, the S 2p spectrum (Fig. 4f) can be resolved into three pairs of peaks. The doublet at 163.29 and 164.92 eV is assigned to C-S bonding, while the remaining doublets at 166.54/167.44 eV and 168.50/169.85 eV originate from SO_3^{2-} and SO_4^{2-} species, respectively^[27,28]. These spectral features collectively verify the successful incorporation of the sulfide phase into the composite. In addition, three O 1s components (Fig. 4d) are resolved at 530.36, 531.29, and 532.69 eV, representing O_L , O_V , and O_ads , respectively. The C 1s spectrum (Fig. 4c) exhibits three characteristic contributions at 284.80, 286.05, and 289.19 eV, which are attributed to C-C, C-O, and C=O bonds, respectively^[29]. Taken together, the XPS results provide convincing evidence for the coexistence of cobalt oxide and cobalt sulfide in the $\text{Co}_3\text{O}_4/\text{CoS}@CC$ composite.

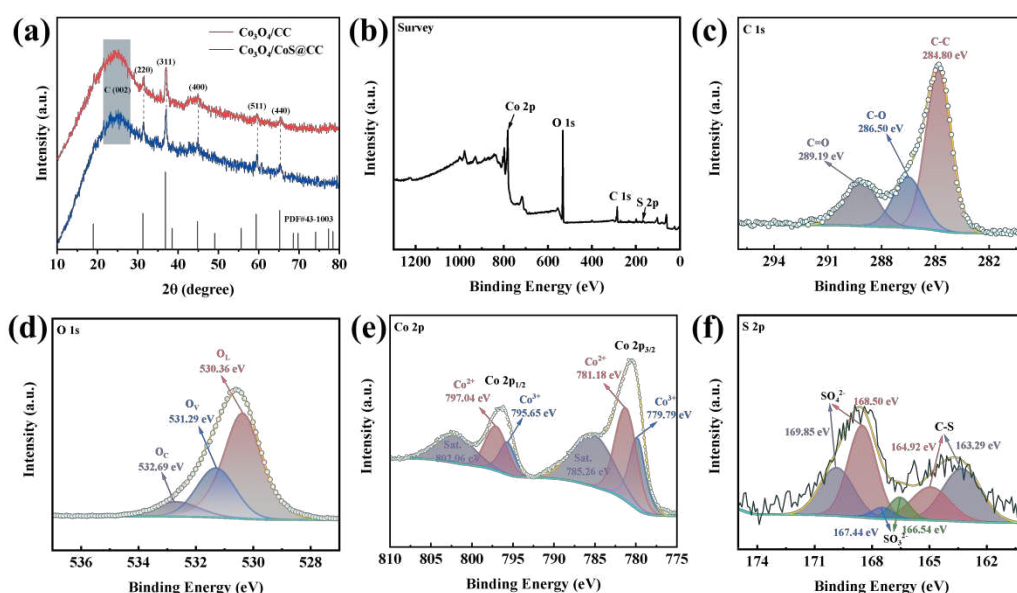
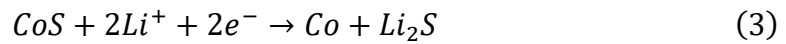
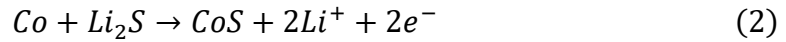
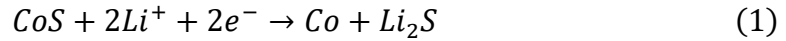


Figure 4 Structure and XPS analysis of the prepared samples.

(a) XRD results for Samples A and B; (b) Full-scan XPS spectrum of Sample B; (c-f) Deconvoluted core-level spectra corresponding to C 1s, O 1s, Co 2p, and S 2p.

The electrochemical behavior of the Co₃O₄/CoS@CC electrode was investigated using CV, where the potential was swept between 0.01 and 3.00 V at 0.1 mV s⁻¹ (Figure 5a). The excellent overlap between the subsequent CV profiles indicates that stable and reversible redox behavior is established once the first-cycle activation is completed. In contrast, the first cycle exhibits obvious differences from the subsequent scans. Two cathodic peaks located at 0.82 and 0.43 V are observed during the initial lithiation process. The former is associated with the conversion of CoS into metallic Co accompanied by Li₂S formation, whereas the latter mainly originates from electrolyte decomposition together with the establishment of the SEI^[1,30]. After the first cycle, the anodic peak is detected at 1.12 V, while the cathodic peaks appear at 0.58 and 1.72 V. The displacement of these characteristic peaks reflects the gradual activation of the electrode and the improved kinetics of the electrochemical reactions. The reduction (Equation (1)) and oxidation (Equation (2)) processes observed during cycling follow the electrochemical equations below. Accordingly, the reversible lithium-storage mechanism of Sample B is summarized in the corresponding reaction equations (Equation (3))^[13,31].



Figures 5b and 5c present the constant-current cycling curves of Samples A and B measured at 100 mA g⁻¹. Compared with the Co₃O₄/CC electrode, the Co₃O₄/CoS@CC electrode exhibits substantially enhanced lithium-storage capability throughout the cycling process. In the initial cycle, the discharge/charge capacities reach 1152.36/1146.67 mA h g⁻¹, whereas the corresponding values for Co₃O₄/CC are only 930.89/926.53 mA h g⁻¹. Part of the lithium inventory is irreversibly consumed during the first cycle because of SEI generation and concurrent parasitic reactions, leading to the discrepancy during the initial lithiation process^[32,33]. With continued cycling, the constant-current cycling curves of the Co₃O₄/CoS@CC electrode become highly reproducible and nearly coincide, demonstrating its excellent electrochemical reversibility as a LIB anode. After 200 cycles, the Co₃O₄/CoS@CC electrode retained reversible capacities as high as 613.09/611.38 mA h g⁻¹ (discharge/charge), whereas the

Co₃O₄/CC electrode delivered only 213.20/211.71 mA h g⁻¹. Such electrochemical behaviors reflect the excellent redox reversibility and long-term cycling durability of the Co₃O₄/CoS@CC composite electrode.

The cycling performance of Samples A and B measured at 100 mA g⁻¹ is illustrated in Figure 5d. The Co₃O₄/CoS@CC electrode delivered 1152.38 mA h g⁻¹ in its first discharge process. During prolonged cycling, the capacity initially decreased, subsequently increased, and finally exhibited a gradual decline. Despite this evolution, a high discharge capacity of 613.07 mA h g⁻¹ was retained 200 cycles, accompanied by a coulombic efficiency of 99.72 %. In contrast, the Co₃O₄/CC electrode exhibited 930.90 mA h g⁻¹ in the first discharge process, with the capacity showing an initial increase followed by continuous decay, ultimately retaining only 213.20 mA h g⁻¹ by the 200th cycle while maintaining the coulombic efficiency of 99.77 %. The characteristic cycling behavior of the Co₃O₄/CoS@CC electrode reflects a typical “negative activation” or “activation-failure” process. At the early stage of cycling, interfacial side reactions consume part of the active lithium and generate a SEI, giving rise to the initial capacity decrease. This process consumes both electrolyte and lithium ions, leading to irreversible capacity loss during the early cycles [34]. In addition, residual impurities or metastable phases remaining on the electrode surface may participate in irreversible side reactions, further contributing to the initial decline. For the Co₃O₄/CoS@CC electrode, the gradual recovery in capacity is mainly ascribed to the progressive wetting of its hierarchical porous architecture by the electrolyte, allowing both Co₃O₄ and CoS to become increasingly involved in the electrochemical reactions [35]. Furthermore, CoS may experience lattice reconstruction or phase transformation during the initial cycling process, exposing additional electrochemically active sites and thereby gradually enhancing the reversible capacity [36]. The final capacity fading observed after extended cycling is primarily attributed to structural degradation, including weakened interfacial adhesion, pulverization or partial detachment of the CoS layer, and a substantial increase in contact resistance between the carbon cloth substrate and the active material, all of which accelerate the deterioration of electrochemical performance [7]. Notably, the onset of the capacity recovery stage for the Co₃O₄/CoS@CC electrode occurs later than that of the Co₃O₄/CC

electrode. This delayed activation behavior is also consistent with the existence of the deposited CoS layer on the Co_3O_4 framework.

Figure 5e compares the rate capabilities of Samples A and B under stepwise current loading. For the $\text{Co}_3\text{O}_4/\text{CC}$ electrode, the average discharge specific capacities reached 1105.88, 873.68, 738.88, and 578.92 mA h g^{-1} when the applied current was sequentially increased from 0.1, 0.2, 0.3, and 0.5 A g^{-1} . Under the same testing conditions, the $\text{Co}_3\text{O}_4/\text{CC}$ electrode delivered lower average discharge specific capacities of 874.88, 662.81, 563.75, and 456 mA h g^{-1} . When the applied current was restored to 0.1 A g^{-1} , the $\text{Co}_3\text{O}_4/\text{CoS}@CC$ electrode still recovered 871.38 mA h g^{-1} , demonstrating its excellent reversibility under varying current densities. The enhanced rate response is closely related to the hierarchical $\text{Co}_3\text{O}_4/\text{CoS}$ architecture. The CoS nanosheets expose additional reaction interfaces and establish more efficient electron/ion transport pathways, while the porous heterostructure facilitates electrolyte penetration and alleviates polarization during high-rate cycling, thereby maintaining rapid reaction kinetics even under increased current loads [37].

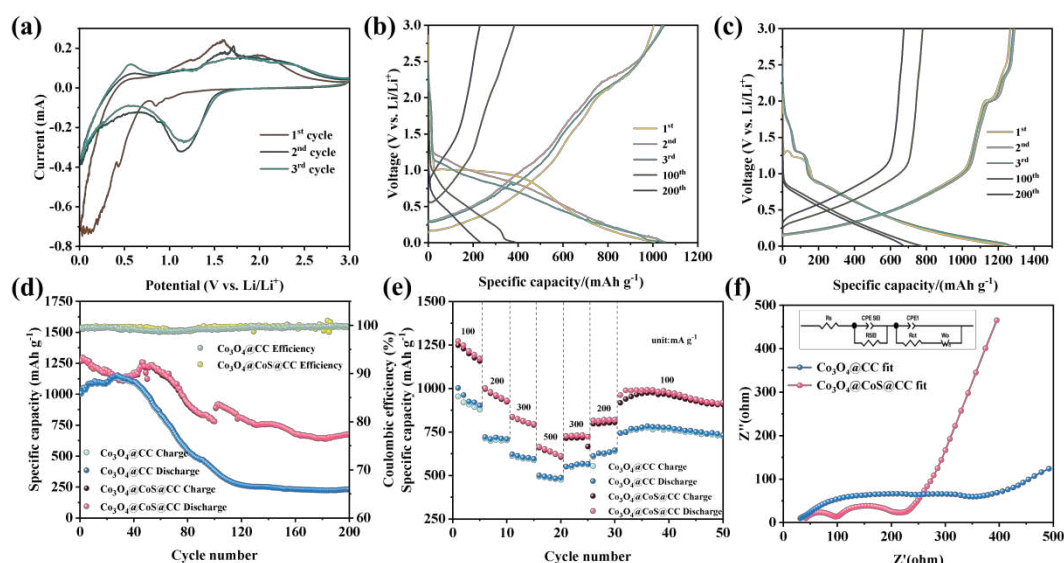


Figure 5 Electrochemical performance of Samples A and B: (a) Cyclic voltammograms of Sample B; (b, c) GCD profiles at 100 mA g^{-1} ; (d) Cycling stability at 100 mA g^{-1} ; (e) Rate capability under different current loads; (f) Nyquist spectra acquired after 100 cycles.

To clarify the kinetic differences between the two electrodes, EIS measurements were performed, and the corresponding Nyquist plots are displayed in Figure 5f. The impedance profiles were analyzed using the equivalent electrical circuit shown in the inset, from which the ohmic resistance (R_s), interfacial film resistance (R_{SEI}), and

charge-transfer resistance (R_{CT}) were determined. Compared with the $\text{Co}_3\text{O}_4/\text{CC}$ electrode, the $\text{Co}_3\text{O}_4/\text{CoS}@CC$ electrode exhibits a markedly lower R_{SEI} (56.68 Ω versus 259.8 Ω) and a dramatically reduced R_{CT} (104 Ω versus 2250 Ω), indicating substantially accelerated interfacial charge transport after CoS deposition. The reduced interfacial resistance can be ascribed to the conductive CoS shell and the intimate $\text{Co}_3\text{O}_4/\text{CoS}$ heterointerface, which together facilitate electron transport, accelerate Li^+ migration across the electrode/electrolyte interface, and suppress electrochemical polarization during cycling^[38]. These results demonstrate that the incorporation of CoS effectively improves the charge transport pathways within Sample B. Moreover, the low-frequency region of Sample B exhibits a steeper and more nearly vertical line than that of Sample A, suggesting faster lithium-ion diffusion in the electrolyte and enhanced diffusion kinetics within the composite electrode^[39]. Taken together, the impedance analysis confirms that introducing CoS effectively optimizes the electrochemical reaction environment of the electrode.

Figure 6a presents a series of photographs recorded during the dynamic contact-angle measurement. Immediately after the electrolyte droplet contacted the electrode surface, continuous spreading was observed. By 19.4 s, the droplet had completely infiltrated the electrode, and the final contact angle was below 90° , indicating complete wetting of the electrode surface. These observations demonstrate the excellent hydrophilicity of the $\text{Co}_3\text{O}_4/\text{CoS}@CC$ composite. The superior wettability can originate from the combination of the interconnected porous carbon cloth framework and the in situ constructed hierarchical $\text{Co}_3\text{O}_4/\text{CoS}$ heterostructure which together form continuous pore channels that markedly enhance electrolyte infiltration and retention. Improved wettability enables Li^+ to access the active material more readily, strengthens electrolyte–electrode interfacial contact, minimizes resistance associated with interfacial charge transfer, and thereby accelerates the overall electrochemical process. Consequently, the above structural and interfacial features provide a reasonable explanation for the remarkable electrochemical performance of Sample B under both prolonged cycling and high-current operation.

Figure 6b presents a photograph of Sample B under a bending condition. Even when the electrode was bent to a large angle using tweezers, neither detachment of the

active material nor fracture of the carbon cloth substrate was observed, demonstrating that the composite effectively inherits the outstanding mechanical flexibility of the carbon cloth support. This excellent mechanical durability is attributed to the interconnected carbon fiber network, which provides high structural integrity and exceptional tolerance to mechanical deformation. Meanwhile, the $\text{Co}_3\text{O}_4/\text{CoS}$ nanoarchitecture formed directly on the carbon cloth remains mechanically integrated with the substrate, minimizing the detachment of electrochemically active species arising from repeated expansion and contraction. Ultimately, the $\text{Co}_3\text{O}_4/\text{CoS}@CC$ composite exhibits the mechanical robustness required for application in flexible energy storage devices.

As illustrated in Figure 6c, a CR2032 coin cell assembled with the $\text{Co}_3\text{O}_4/\text{CoS}@CC$ electrode is capable of continuously powering multiple LED indicators for up to 2 h, demonstrating its reliable electrochemical output and excellent energy-storage capability under practical operating conditions. Combined with the superior wettability and outstanding mechanical flexibility discussed above, this device further confirms direct evidence for the promising electrochemical behavior of Sample B. These results collectively highlight the considerable potential of Sample B for practical and industrial applications in lithium-ion energy storage systems.

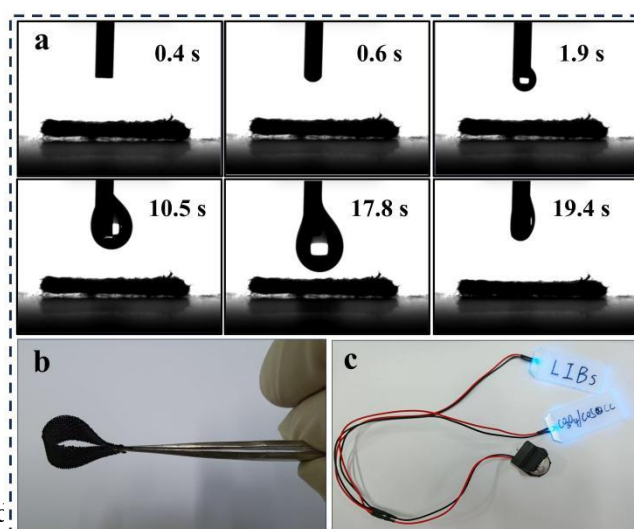


Figure 6 Physical characterization of the $\text{Co}_3\text{O}_4/\text{CoS}@CC$ composite. (a) Photograph showing the flexibility of the electrode under bending. (b) Photograph showing the flexibility of the electrode under bending. (c) Demonstration of the assembled CR2032 coin cell powering multiple LEDs.

4. Conclusions

In this work, a $\text{Co}_3\text{O}_4/\text{CoS}@CC$ composite was successfully fabricated through a

straightforward combination of hydrothermal synthesis and electrodeposition. The introduction of the CoS nanoflower architecture effectively increases the specific surface area, improves electrical conductivity, promotes Li^+ transport, and buffers the volume variation associated with repeated charge-discharge processes. Owing to these structural advantages, the $\text{Co}_3\text{O}_4/\text{CoS}@CC$ composite exhibits outstanding lithium-storage behavior. Even after 200 cycles at 0.1 A g^{-1} , the electrode maintained a capacity of $613.07 \text{ mA h g}^{-1}$, far exceeding the value of $213.20 \text{ mA h g}^{-1}$ obtained for the $\text{Co}_3\text{O}_4/CC$ electrode. The favorable lithium-storage performance primarily arises from the abundant redox-active sites and superior electronic conductivity provided by CoS. In the composite architecture, $\text{Co}_3\text{O}_4/CC$ serves as the supporting framework, while electrodeposited CoS as nanosheets are uniformly distributed over the Co_3O_4 nanowire arrays. This hierarchical configuration facilitates electrolyte infiltration and charge transport, while simultaneously buffering the dimensional variation generated during repeated cycling. Furthermore, the present study offers a facile strategy for the preparation of ultrafine CoS nanosheets. Benefiting from its flexibility, sustained electrochemical output, and long-term operational durability, the resulting $\text{Co}_3\text{O}_4/\text{CoS}@CC$ composite demonstrates considerable potential for practical lithium-ion battery applications.

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