3D Highly Conductive Silver Nanowire@PEDOT:PSS Composite Sponges for Flexible Conductors and Their All-Solid-State Supercapacitor Applications

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Although increasing attention has been paid to wearable electronic devices in recent years, flexible supercapacitors with high performance remain not readily available because of the limitations of flexible electrode types. A highly conductive 3D macroporous sponge is fabricated by coating poly(3,4-ethyl enedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS)/silver nanowires (AgNWs) on a commercial sponge using a simple and low-cost “immersion method.” The fabricated flexible 3D sponge conductor shows a high electrical conductivity of 3.94 × 10$^{-4}$ S cm$^{-1}$ with good stability in various environments and under bending deformation. To exploit the potential of the flexible 3D PEDOT:PSS/AgNW coating on the sponge as a current collector for energy-related applications, urchin-like Co(OH)$_2$F arrays are directly grown on PEDOT:PSS/AgNW-on-sponge conductors for all-solid-state supercapacitors. The resulting symmetric all-solid-state supercapacitor exhibits a mass-specific capacitance of 103.7 F g$^{-1}$ at a current density of 1 A g$^{-1}$, retaining 85.8% of the initial capacitance after 3000 cycles, and mechanical flexibility during bending. This fabrication of the 3D lightweight conductor can be easily scaled up for mass production and introduces new opportunities for flexible electronic applications.

1. Introduction

With the advent of flexible electronics, flexible and lightweight supercapacitors have attracted great attention as promising electrochemical energy storage devices for high power delivery or energy-harvesting applications in the emerging field of flexible and/or wearable devices such as internal energy recovery systems, memory backup, biomedical monitoring, and flexible consumer electronics.[1–3] As a new research trend for flexible supercapacitors, advanced nanostructured materials, for example, polyurethane or melamine sponges) with 3D open-cell structure analogue metal foams as a template has attracted intensive research interest for solving energy and environmental issues because of the low cost, ease of fabrication, 3D porous structures, light weight, and high flexibility. Thus, plastic sponges have drawn significant attention as a support and/or template to construct 3D active materials. However, these polymer foams are electrical insulators. Consequently, tremendous efforts have been devoted to synthesize 3D network conductors on commercial sponges by simply dip-coating (reduced) graphene oxides and/or carbon-nanotube dispersion and/or pyrolized carbon sponges.[5–14] However, it is difficult to produce carbonaceous foams/sponges in a large scale because of their relatively high-cost and harsh processing conditions (high-temperature carbonization: ≈800–1000 °C).[15] Furthermore, because of the high resistance of carbon-based materials, for example, 0.34 S cm$^{-1}$ carbonized polyurethane foam (0.34 S cm$^{-1}$).[16] reduced graphene oxide coated on polyurethane foam (0.0009–0.0025 S cm$^{-1}$),[17] and carbon nanotube coated on polyurethane (1 S cm$^{-1}$),[18] these supercapacitor sponges face issues in high internal resistance.[19,20] Their performance in terms of electrical conductivity remains inferior to that of metal foams, which results in a reduced power density. Lately, the ramifications of 3D networks with high conductivity as substrate templates have been a notably fascinating topic for energy and/or sensing from both scientific and technological viewpoints.[7,8,21–30] Therefore, there is an urgent need for a new class of flexible 3D open-porous substrates to develop advanced energy-related storages with enhanced energy density without sacrificing the power delivery and cycle stability to satisfy the future flexible energy-related demands. Nonetheless,
because of the restrictions of the types of flexible current collectors with high electrical conductivity, there are limitations in manufacturing various flexible electronic devices. Among various conductive materials, silver nanowires (AgNWs) are easily corroded, but percolation networks of AgNWs have attracted increasing attention for developing flexible conductive films because of their excellent conductivity (conductivity of silver \( \approx 6.3 \times 10^7 \, \text{S cm}^{-1} \)) compared to graphene and carbon nanotubes.

In this work, we develop a simple, strong-durability and low-cost process with high electrical conductivity to fabricate a composite of a poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS)/AgNW hybrid coated on the commercially available polyurethane sponge (PUS), which has a 3D macroporous structure, as a support for conductive 3D networks, as shown in Figure 1. PEDOT:PSS was selected to ensure both good conductivity and corrosion protection of the AgNW networks. The obtained 3D open macroporous PEDOT:PSS/AgNW/PUS conductor showed superior electrical conductivity \((3.94 \times 10^{-4} \, \text{S cm}^{-1})\) by the volume based on the total porous foam and flexibility for constructing flexible energy storage devices. To demonstrate its potential energy-related applications, hierarchical sea-urchin-like cobalt hydroxide (Co(OH)F) arrays were grown on the as-prepared PEDOT:PSS/AgNW/PUS conductor through a hydrothermal reaction and directly used as a supercapacitor electrode. The electrochemical performance of the as-prepared 3D Co(OH)F/PEDOT:PSS/AgNW/PUS was evaluated by cyclic voltammetry (CV) and constant-current charge-discharge cycling techniques. The specific capacitance (SC) of Co(OH)F/PEDOT:PSS/AgNW/PUS can reach 103.7 F g\(^{-1}\) at a current density of 1 A g\(^{-1}\) because of the fast electron and ion transport, large electroactive surface area, and excellent structural stability. The electrode also exhibits an excellent charge/discharge rate and good cycling stability, where it retains over 85.8% of its initial charge after 3000 cycles. Hence, this 3D flexible open-porous conductor design may provide a direction to develop flexible high-performance electrochemical electrodes for future wearable electronics and other functional nanoscale devices.

2. Results and Discussion

The Co(OH)F/PEDOT:PSS/AgNW/PUS flexible electrode for electrochemical capacitors is shown in Figure 1a,b. The pre-cleaned PUS with 3D-interconnected micro-skeleton networks was immersed into a solution of AgNW dispersion. After the solvent was evaporated, the color of the sponge changed from yellow to gray (inset of Figure 1a, and Figure S1 in the Supporting Information), and the skeleton frameworks in PUS were uniformly coated with a percolated network of AgNWs. Finally, the PEDOT:PSS/AgNW/PUS samples were prepared by immersion into the PEDOT:PSS solution as the adhesion layer between the AgNW networks and PUS skeletons and the conducting layer of empty space between the AgNW networks to prepare anticorrosive AgNW networks. The resulting products were uniformly wrapped on AgNW/PUS skeletons to form the desired PEDOT:PSS/AgNW/PUS flexible conductors, which changed the color from gray to dark gray (Figure 1a, inset). Figure 2a–d shows the scanning electron microscope (SEM) images at different magnifications of the AgNW and PEDOT:PSS/AgNW on the PUS skeletons before (Figure 2a,b) and after (Figure 2c,d) the PEDOT:PSS coating. A high-density AgNW network coating was formed on the surface of
PUS skeletons, where a dense network of randomly oriented AgNWs is observed. PEDOT:PSS on the AgNW/PUS skeletons penetrated the AgNW networks and became embedded in AgNWs. The AgNW coated on the PUS showed an electric conductivity of $=3.61 \times 10^{-4}$ S cm$^{-1}$. After the PEDOT:PSS coating on the AgNW/PUS, the conductivity increased by 9.09% ($=3.94 \times 10^{-4}$ S cm$^{-1}$) at room temperature. As shown in the SEM images of Figure 2c,d, the rough surface of the AgNW networks was smoothened and tightly welded with PEDOT:PSS to bridge the charge transport across adjacent AgNWs and increase the conductivity of the AgNW networks. Furthermore, the PEDOT:PSS could be tightly adhered between the AgNW networks and the PUS via the capillary force during the solvent evaporation process of the PEDOT:PSS solution, which is a strong adhesion force, as inferred from Figure 2d.

To test the durability of the PEDOT:PSS/AgNW/PUS composite conductor under stress, we measured their resistance change at different bending angles. As shown in Figure 3a, the mean resistance varied from 11.2 to only 10.8 $\Omega$ when the sample was subjected to a bending angle of up to 135°. This change in resistance with bending angle may occur because of the change in pressure at the AgNW junctions in a given area. The stable electrical and electrical-mechanical fatigue properties of PEDOT:PSS/AgNW/PUS and AgNW/PUS were investigated through repeated bending-relaxation cycles at an angle of $=120^\circ$, and a representative result is illustrated in Figure 3b.

The change in resistance of the conductor can be expressed as $\Delta R = (R - R_0)/R_0$, where $R_0$ is the initially measured resistance, and $R$ is the value measured after the substrate bend. The PEDOT:PSS/AgNW/PUS showed almost no change in resistance during 200 cycles of bending, but the resistance slightly increased. This increase is attributed to the strong bonding between the AgNW networks and the PEDOT:PSS matrix, which could prevent sliding at the interface in bending cycles. In contrast, the AgNW/PUS without PEDOT:PSS significantly degraded because of slipping and/or delamination in two crossed AgNWs and easy oxidation of the AgNWs in air. To explore the compressive uniaxial strain-dependent conductivity of the PEDOT:PSS/AgNW/PUS during the compression process, PEDOT:PSS/AgNW/PUS was imbedded into two parallel copper foil electrodes. It was connected to an electrical loop to detect the electrical resistance changes, when it was uniaxially compressed. Figure S2 in the Supporting Information shows that the electrical resistance gradually decreases with the uniaxial compression strain: it changed by 0.57% when it was 68.19% compressed. Upon releasing, the reverse occurred, as reflected in the change in resistance. The gradual decrease in electrical resistance of the uniaxial compressed PEDOT:PSS/AgNW/PUS can be explained by the more contacting points of the silver nanowire network, which results from the compression. Thus, flexible PEDOT:PSS/AgNW/PUS can be used in various technologies as supercapacitors, batteries, sensors, and conductors.

To exploit the potential of the flexible 3D PEDOT:PSS/AgNW/PUS conductor as a current collector for energy-related applications, we designed and prepared an all-solid-state supercapacitor device. Nanostructured transition metal hydroxides such as Ni(OH)$_2$, Co(OH)$_2$, and nickel cobalt hydroxide have been widely studied for use as electrodes for supercapacitors. Among the transition metal hydroxide materials, we selected nanostructured cobalt-based hydroxide to grow on PEDOT:PSS/AgNW/PUS because of its high specific capacity. Figure 1a schematically illustrates the synthesis procedure of the 3D Co(OH)$_2$ sea-urchin-like arrays, which were directly grown on the PEDOT:PSS/AgNW/PUS conductor via a facile hydrothermal process.

Figure 4a–d shows a typical low-resolution SEM image of Co(OH)$_2$/PEDOT:PSS/AgNW/PUS. Apparently, Co(OH)$_2$ uniformly covers the 3D PEDOT:PSS/AgNW/PUS structure and maintains the original skeleton structure of the PUS. Evidently, Co(OH)$_2$ displays a type of uniform urchin-like structure with numerous dense 1D individually aligned nanoneedles, and many urchin-like clusters are stacked together and tightly bundled perpendicularly to a central core. This unique nanostructure enables better electrolyte access within the electrode materials and more efficient exposure of active sites to the electrolyte. Furthermore, the elemental mapping in Figure S3 in the Supporting Information shows that Co and O are well distributed throughout the nanostructure, which confirms the formation of Co(OH)$_2$. Figure 4e–g shows representative transmission electron microscope (TEM) images and a selected area electron diffraction (SAED) pattern of a Co(OH)$_2$ nanoneedle. To observe more deeply into the lattice structure, high-resolution TEM (HRTEM) was used. Figure 4f shows an HRTEM image of the Co(OH)$_2$ nanoneedle, where two classes of lattice
fringes are present and all corresponding inter-plane distances are \( \approx 2.52 \text{ Å} \) (including the (111) and (1\bar{1}1) crystal planes). The SAED pattern (inset in Figure 3d) reveals a good single crystal-like nature of the Co(OH)F nanowires.\(^{[16]}\)

The powder X-ray diffraction (XRD) pattern of Co(OH)F scratched off from the PEDOT:PSS/AgNW/PUS is shown in Figure 5a. The diffraction peaks are at 2\( \theta \) values of 20.9\( ^\circ \), 32.3\( ^\circ \), 33.5\( ^\circ \), 34.7\( ^\circ \), 35.6\( ^\circ \), 38.8\( ^\circ \), 39.9\( ^\circ \), 43.6\( ^\circ \), 51.9\( ^\circ \), 52.8\( ^\circ \), 56.9\( ^\circ \), 59.1\( ^\circ \), and 61.6\( ^\circ \), which demonstrates the crystalline planes \((110), (310), (201), (400), (111), (211), (410), (311), (221), (420), (511), (002)\), and \((601)\) of orthorhombic Co(OH)F with lattice constants \(a = 10.305\ \text{Å}, b = 4.677\ \text{Å}, \) and \(c = 3.126\ \text{Å}\) (JCPDS card no. 50–0827). The strong and sharp diffraction peaks suggest that the synthesized Co(OH)F is well crystallized.\(^{[32,37]}\) Micro-Raman spectroscopy was used to study the bonded interface of the Co(OH)F/PEDOT:PSS/AgNW/PUS. Figure 5b displays the Raman spectrum of the trampled-Co(OH)F/PEDOT:PSS/AgNW/PUS sample with an excitation wavelength of 532 nm. The main bands in Figure 5b are at 247.4 cm\(^{-1}\) for Ag–O; 657.5, 606.6, 443.1, and 368 cm\(^{-1}\) for Co(OH)F; and 1370.4, 1429.4, 1515.2, and 1560.7 cm\(^{-1}\) for PEDOT:PSS.\(^{[38–40]}\) After the hydrothermal reaction, we confirmed that each component on the PUS substrate preserved its character. Furthermore, X-ray photoelectron spectroscopy (XPS) was used to characterize the chemical state of the Co(OH)F/PEDOT:PSS/AgNW/PUS composite. The full XPS spectrum (Figure S4, Supporting Information) was consistent with all expected elements in Co(OH)F/PEDOT:PSS/AgNW/PUS, which is consistent with the SAED, XRD, and Raman results. The broader spin-orbit doublet with binding energies of Co 2\( \text{P}_{1/2} \) (781.7 eV) and Co 2\( \text{P}_{3/2} \) (797.9 eV) are shown in Figure 5c. In addition, there are two satellite peaks at 785.4 and 802.3 eV. The spin-orbit splitting of \(\Delta E_s \approx 16.2\ \text{eV}\) indicates that Co existed as Co(OH)F.\(^{[41,42]}\) The high-resolution O1s spectrum (Figure 5d) shows three oxygen species: the Co–O group (binding energy (BE) = 530.7–531.1 eV), Co–OH group (BE = 531.6–532.9 eV), and chemisorbed oxygen (BE = 534.5–535.7 eV).\(^{[41,42]}\)

The obtained hierarchical Co(OH)F urchin-like arrays on PEDOT:PSS/AgNW/PUS are examined for their applications in supercapacitors. First, we examined in a three-electrode cell configuration in a 2.0 m KOH aqueous solution. Figure S5 in the Supporting Information shows the CV curves of Co(OH)F/PEDOT:PSS/AgNW/PUS at different scan rates of 5–50 mV s\(^{-1}\). The appearance of distinct redox peaks between –0.2 and 0.4 V versus Ag/AgCl confirms the pseudocapacitive behavior of the materials and implies a good electrochemical reversibility. The current density obviously increases with the increase in scan rates in the CV curves, which suggests the presence of surface redox reactions and the pseudocapacitance nature. To further evaluate the practical performance of the Co(OH)F/PEDOT:PSS/AgNW/PUS composite as an electrode and a current collector with flexibility and good electrical conductivity, we performed more tests with an all-solid-state symmetric supercapacitor. Electrochemical experiments were conducted in a two-electrode configuration in a PVA/KOH gel electrolyte. Figure 6a shows the typical CV curves of the Co(OH)F/PEDOT:PSS/AgNW/PUS device at 0–1.0 V at various scan rates of 1–100 mV s\(^{-1}\). The CV curves almost retain a quasi-rectangular shape, which suggests an ideal capacitive behavior of the symmetric supercapacitor. Even at a scan rate of 100 mV s\(^{-1}\), the CV curves display a quasi-rectangular shape. However, the well-protected samples from 1 to 100 mV s\(^{-1}\) suggest the high rate capability of Co(OH)F/PEDOT:PSS/AgNW/PUS for power delivery.\(^{[43]}\) The area of the CV curves increases with increasing scan rate, which can be attributed to the efficient ion diffusion in the polymer electrolyte at higher scan rates.\(^{[44]}\) Consistently, the profile of the galvanostatic charge/discharge curves with various current densities in the voltage range of 0–1 V and their symmetric near-triangular shape indicate nearly ideal capacitive characteristics and a fast Faraday redox reaction (Figure 6b). The linear profile of the charge and discharge curves reveals the good capacitive characteristics of the solid-state supercapacitor device. The SCs at various discharge current densities for Co(OH)F/PEDOT:PSS/AgNW/PUS were calculated from the charge/discharge curves, as shown in Figure 6c. The highest SC of the Co(OH)F/PEDOT:PSS/AgNW/PUS composite based on the Co(OH)F mass was 103.7 F g\(^{-1}\) at a current density of 1 A g\(^{-1}\). The SC decreases with the increase in current because...
a high current makes more active material insufficient in the redox reaction when the scan rate increases. The Ragone plots of the symmetric supercapacitor derived from the discharge curve are shown in Figure S6 in the Supporting Information. This device showed the maximum energy density of 3.6 Wh kg\(^{-1}\) at a power density of 122.26 W kg\(^{-1}\) and a current density of 1 A g\(^{-1}\). At 20 A g\(^{-1}\), the power increases to 1242 W kg\(^{-1}\) while maintains the energy of 2 Wh kg\(^{-1}\).

To investigate the electrochemical behavior at the electrode/electrolyte interface in detail, electrochemical impedance spectroscopy (EIS) was measured at open circuit potentials in the frequency range from 0.01 Hz to 10 kHz. The corresponding Nyquist impedance plot consists of a semicircle in the high-frequency region and a straight line in the low-frequency one. The high-frequency intercept at the real axis is the equivalent series resistance (ESR), the diameter of the semicircle corresponds to the charge transfer resistance of the electrode, and the straight line in the low-frequency region represents the Warburg resistance. The straight-line part leans more toward the imaginary axis in the low-frequency regions, which indicates a good capacitive behavior of the material. The ESR and charge transfer resistance are \(\approx 1.25\) and \(1.85\) \(\Omega\), respectively. The low ESR of the Co(OH)F/PEDOT:PSS/AgNW/PUS electrode can be attributed to two critical factors: (i) the Co(OH)F nanowire arrays formed a highly conductive network directly coated on the PEDOT:PSS/AgNW as a current collector and provided the electrode with low internal and interfacial resistances; (ii) the open space among the Co(OH)F nanowires provides a large electrolyte/electrode contact area and effective electrolyte-accessible channels for ion transportation and shortens the distance for ion diffusion.

The long-term cycling performance was also investigated to evaluate the electrode stability. As shown in Figure 6e, \(\approx 85.8\)% of the initial capacitance was retained after 3000 cycles of charging and discharging at a current density of 5 A g\(^{-1}\). Together, these two aspects produce excellent electrochemical results. To test the feasibility for flexible capacitance applications, a simple solid-state supercapacitor based on the Co(OH)F/PEDOT:PSS/AgNW/PUS was also prepared for a demonstration.

For application consideration, flexible and portable electronics may require highly flexible power sources. Therefore,
to investigate the mechanical flexibility of the device, we performed the CV measurement of the symmetric device at a scan rate of 50 mV s\(^{-1}\) at different operation voltages and powers. We measured the CV curves of the supercapacitor at various bending angles (0\(^\circ\), 30\(^\circ\), 60\(^\circ\), 90\(^\circ\), and 120\(^\circ\)), as shown in the inset of Figure 6f. The CV curves maintain a nearly constant capacitive behavior, as shown in Figure 6f. Only 12.8% decay in SC was observed at a bending angle of 120\(^\circ\), which may enable the collapse of the Co(OH)F urchin-like arrays on PEDOT:PSS/AgNW/PUS at the core during the bending process. Thus, the increase in bending performance of the electrode is attributed to the strong bonding between the Co(OH)F nanowire arrays and the PEDOT:PSS/AgNW matrix. This performance can be attributed to the flexibility character because of the PEDOT:PSS/AgNW/PUS.

3. Conclusion

In conclusion, a facile chemical method that can be scaled up was developed to prepare a flexible 3D macroporous conductor with high electrical conductivity, which consists of a PEDOT:PSS/AgNW composite coated on a commercial sponge. The key novelty is the use of a flexible/wearable 3D PEDOT:PSS/AgNW with chemical stability as a conductive collecting substrate. The fabricated flexible 3D sponge conductor exhibited a high electrical conductivity of 3.94 \(\times 10^{-4}\) S cm\(^{-1}\) with good stability under bending deformation. Co(OH)F urchin-like arrays were directly grown on the PEDOT:PSS/AgNW/PUS 3D flexible conductor as a current collector, and its practical application was explored in a flexible all-solid-state symmetric supercapacitor. The resulting symmetric all-solid-state supercapacitor exhibited a capacitance of 103.7 F g\(^{-1}\) at a current density of 1 A g\(^{-1}\), retained 85.8% of the initial capacitance after 3000 cycles, and was mechanically flexible during bending. Because of the simple fabrication procedures and enhanced electrochemical performance, such 3D structures have high potential for use in flexible conductors for the wide accessibility and high scalability of practical applications.

4. Experimental Section

Preparation of the PEDOT:PSS/AgNW/PUS Conductor: Precleaned PUS pieces were immersed in a 0.5 wt% dispersion of AgNWs (NANOPYXIS) in isopropyl alcohol for 10 min; then, they were removed and dried at 110\(^\circ\)C for 20 min. The mass loading of AgNW was \(\approx 0.4\) mg cm\(^{-2}\). The obtained AgNW/PUS piece was immersed into a dilute with a volume ratio of 1:5 of (PEDOT:PSS solution (2.0–4.0% in H\(_2\)O, Aldrich)):(H\(_2\)O) as the adhesion layer between the AgNW networks and PUS skeletons and the conducting layer of empty space between the AgNW networks to prepare the anticorrosive AgNW networks. After 10 min, the PEDOT:PSS-coated AgNW/PUS pieces were dried at 110\(^\circ\)C for \(\approx 40\) min. The PEDOT:PSS mass loading onto AgNW/PUS was \(\approx 0.2\) mg cm\(^{-2}\).

Synthesis of Co(OH)F on PEDOT:PSS/AgNW/PUS Electrode: Co(OH)F sea-urchin-like arrays were synthesized on PEDOT:PSS/AgNW/PUS in a simple hydrothermal method. In the typical procedure, Co(NO\(_3\))\(_2\)-6H\(_2\)O (1.16 g, 4 mmol), NH\(_4\)F (0.6 g, 16 mmol), and urea (1.8 g, 20 mmol) were dissolved in 36 mL of deionized water and stirred for 30 min to form a clear solution. The solution was transferred into a 40 mL Teflon-lined stainless-steel autoclave. Then, a PEDOT:PSS/AgNW/PUS piece was vertically immersed into the above solution. Hydrothermal synthesis was performed at 110\(^\circ\)C for 20 h, following which the piece...
was allowed to naturally cool to room temperature. The Co(OH)F/PEDOT:PSS/AgNW/PUS piece was carefully washed several times with deionized water and freeze-dried. The mass loading of Co(OH)F was \( \approx 25.5 \text{ mg cm}^{-2} \).

**Characterization:** The XRD pattern was collected using an X-ray diffractometer (SMARTLAB, Rigaku) equipped with a conventional Cu K\( \alpha \) radiation source (\( \lambda = 1.54 \) Å). The Raman spectrum was measured using a LabRam Aramis Raman spectrometer from Horiba Jobin Yvon. The instrument was fitted with a 532 nm laser. The XPS measurement was performed using a Thermo Fisher Scientific system (K-ALPHA ESCA SYSTEM) with a monochromatic Al K\( \alpha \) X-ray source. The microstructure was observed using field-emission SEM (JSM-6701F/INCA Energy, JEOL). The TEM and HRTEM images were obtained using a JEOL-ARM200F with a spherical aberration corrector. The PEDOT:PSS/AgNW/PUS was imbedded into two parallel copper foil electrodes, and two copper wires were connected to the electrodes. The resistance of AgNW/PUS and PEDOT:PSS/AgNW/PUS was measured using a Digital Multimeter (Fluke 87V multimeter).

**Fabrication of All-Solid-State Supercapacitor:** The polyvinyl alcohol (PVA)/KOH gel electrolyte was prepared as follows: 4.5 g of KOH was added into 60 mL of deionized water; then, 6 g of PVA powder was added. The entire mixture was heated to \( \approx 85 \) °C under stirring until the solution became clear. Two Co(OH)F/PEDOT:PSS/AgNW/PUS electrode pieces were immersed into the PVA/KOH solution for 1 h; then, they were removed, assembled with a separator in between, and left overnight until the electrolyte solidified. Finally, a 1.3 × 3 × 0.2 cm\(^3\) area Co(OH)F/PEDOT:PSS/AgNW/PUS sample was pressed at 8 MPa and had a geometric surface area of \( \approx 1.3 \times 3 \) cm\(^2\).

**Electrochemical Measurements of All-Solid-State Supercapacitor:** A two-electrode cell configuration was used to measure the performance of Co(OH)F/PEDOT:PSS/AgNW/PUS as symmetric supercapacitor electrodes using a CHI660E electrochemical workstation. All electrodes were used without any binder or conduct additive. The electrochemical performances were characterized by cyclic voltammetry, galvanostatic charge–discharge tests, and EIS. The gravimetric SC was obtained from the discharge process according to the following equation:
\( C = \Delta \tau / m \Delta V \), where \( I \) (A) is the discharge current, \( \Delta \tau \) is the time for full discharge, \( m \) is the total Co(OH)F mass of the two electrodes (g), and \( \Delta V \) is the potential change after the full discharge.

Supporting Information
Supporting Information is available from the Wiley Online Library or the author.

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Conflict of Interest
The authors declare no conflict of interest.

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