

Article

Hydrothermally Synthesized SrS/Polyaniline Nanocomposite Electrodes for Asymmetric Supercapacitor Devices with Enhanced Charge-Storage Performance

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Abstract

The growing demand for efficient and sustainable energy-storage systems has intensified efforts to develop materials capable of delivering both high power output and reliable capacity retention. Conventional supercapacitors excel in rapid charge–discharge processes and offer outstanding cycling durability; however, their inherently low energy density limits large-scale use. In contrast, batteries provide high energy densities but typically display slower power response and poorer rate capability. Consequently, hybrid storage systems that merge capacitive and faradaic mechanisms have emerged as a compelling strategy to overcome these shortcomings. In this study, a SrS/polyaniline (SrS/PANI) nanocomposite was fabricated via hydrothermal synthesis and evaluated as an electrode material for hybrid supercapacitor architectures. Structural and morphological characterisation confirmed the formation of a nanoscale composite with well-integrated phases. Electrochemical performance was first evaluated in a three-electrode half-cell configuration, where the optimized SrS/PANI (50/50 wt%) electrode delivered a GCD-derived specific capacity of 580 C g^{−1} at 0.4 A g^{−1}. The electrode was then assembled into an asymmetric two-electrode device, which achieved an energy density of 18 Wh kg^{−1}, a power density of 2980 W kg^{−1}, and 75% capacity retention after 1000 cycles. Overall, the findings indicate that the SrS/PANI composite exhibits improved charge-storage behaviour arising from the combined contribution of redox-active SrS and the conducting-polymer component PANI, underscoring its promise for hybrid energy-storage applications.



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1. Introduction

Energy security has become a central issue for both scientific and industrial communities, driven by accelerating climate change and global economic growth [1]. Rising concerns over environmental degradation, increasing extraction costs, and the progressive depletion of fossil resources further intensify the urgency for alternative solutions [2]. This situation

underscores the need for energy technologies that are efficient, clean, and sustainable. Although extensive research has focused on improving conventional systems, many practical applications remain constrained by the limitations of Li-ion batteries and fuel cells [3]. Recently, interest in supercapacitors has grown dramatically [4]. Many researchers and industries are interested in these devices due to their rapid charge–discharge rates, lengthy life cycles (up to one million cycles for certain types), and outstanding power performance compared to conventional batteries.

Supercapacitors, also known as ultracapacitors or electrochemical capacitors, store electricity through two main mechanisms [5]. In electric double-layer capacitors (EDLCs), capacitance arises from reversible ion adsorption/desorption at the electrode/electrolyte interface without redox reactions, enabling extremely fast ion transport and, therefore, very high charge–discharge rates and power density. In pseudocapacitors, charge storage predominantly occurs through fast surface redox reactions of electrode materials such as conducting polymers or transition-metal oxides/sulfides [6]. Although conducting polymers and metal oxides offer high theoretical capacitance, they generally do not conduct electricity as well as carbon-based materials, and their performance degrades relatively quickly after many cycles [7]. In contrast, pure EDLCs can be charged and discharged up to a million times before losing significant capacity [8].

The energy density of commercial EDLCs is currently limited to 3–5 Wh/kg, which is substantially lower than that of industrial lithium-ion batteries (100–275 Wh/kg) [9]. Therefore, the greatest challenge in the field of supercapacitor research is to increase specific energy while sacrificing as little power density and cyclic stability as possible. Three of the most common commercial energy storage methods in use today are conventional capacitors, rechargeable batteries, and fuel cells [10]. The rapid development of new technologies has dramatically increased global energy demand and raised serious concerns about the rapid depletion of fossil fuels, which are predicted to run out in the near future [11]. More consideration must therefore be given to sustainable and renewable energy sources. Still, these resources require dependable and efficient energy storage devices (ESDs) to store and deliver the harvested energy effectively [12].

In the past, various types of rechargeable batteries and supercapacitors have been widely employed to construct such ESDs [13]. Different nanomaterials, including metal oxides, sulfides, phosphates, phosphides, and hydroxides, are commonly used to fabricate high-energy rechargeable batteries [14]. These materials provide exceptional energy density and are promising for high-energy applications [15]. However, supercapacitors still lag in delivering high specific energy despite their exceptional specific power and long-term cyclic stability [16]. Regarding charge storage mechanisms, supercapacitors are divided into electric double-layer capacitors (EDLCs) and pseudocapacitors (PCs) [17]. Carbonaceous compounds such as activated carbon, carbon black, graphene, carbon nanotubes, and carbon-derived carbides are the main electrode materials for EDLCs [18]. In the meantime, conducting polymers, transition-metal oxides (RuO_2 , MnO_2 , V_2O_5 , Co_3O_4 , IrO_2), and metal sulphides constitute the most common pseudocapacitor electrode materials.

Increasing the specific energy of these capacitors without compromising power density or cyclic stability remains a significant experimental challenge [19]. In recent years, numerous hybrid supercapacitors with enhanced energy density have been reported using battery-grade and carbonaceous electrode combinations, including $\text{TiO}_2//\text{CDC}$, $\text{Co}_{0.5}\text{Mn}_{0.5}\text{S}//\text{AC}$, $\text{VN@C}//\text{AC}$, $\text{TiO}_2//\text{rGO}$, and $\text{Co}_3(\text{PO}_4)_2//\text{AC}$. Transition metal oxides, metal–organic frameworks (MOFs), phosphides, and nitrides remain the most widely studied active materials in supercapacitor research. Although these materials exhibit notable electrochemical performance, their practical application is often limited by relatively low electrical conductivity and poor long-term stability.

Transition metal sulfides (TMS), on the other hand, possess a range of redox-active sites, nanocrystalline structures with various morphologies, lower electronegativity of sulfur compared to oxygen, a more flexible crystal structure, higher electrical conductivity, better mechanical and thermal stability, and generally higher charge-storage capacity than corresponding oxides [20]. Because sulfur has lower electronegativity than oxygen, TMS materials show improved ion transport and overall electrochemical performance [21].

Among various metal sulfide electrode materials, strontium-based sulfide materials have recently shown particularly promising results. Adinaveen et al. previously fabricated SrS-activated carbon composite electrodes and demonstrated outstanding device resilience, excellent cyclic stability, and a high specific capacitance of 400 F/g at 3 mV/s. Iqbal et al. reported graphene oxide-assisted SrS electrodes that achieved an extraordinarily high specific capacitance of 1831.14 F/g in a three-electrode configuration. These remarkable values indicate that strontium sulfide and related alkali-earth metal sulfides may exhibit exceptional conductivity, a large effective surface area, and very high efficiency, making them highly suitable for both battery-type and supercapacitor applications [22].

A range of synthesis strategies, including hydrothermal, solvothermal, chemical precipitation, and sonochemical techniques, are commonly used to produce these nano-materials [23]. Among them, the sonochemical route exploits acoustic cavitation to trigger and accelerate reaction processes [24]. The collapse of cavitation bubbles produces intense local pressure and high-velocity inter-particle collisions, enabling effective control over particle size, morphology, and dispersion [25]. This approach is attractive because it is straightforward, rapid, and economical, often yielding uniformly distributed nanoparticles with minimal agglomeration [26].

In the present work, strontium sulfide (SrS) and polyaniline (PANI)-incorporated SrS composite electrode materials were successfully synthesized via a facile hydrothermal approach. The crystal structure, elemental composition, and surface morphology of the prepared samples were systematically investigated using X-ray diffraction (XRD), scanning electron microscopy (SEM), and EDX. Electrochemical performance and charge-storage mechanisms were first studied in three-electrode cells to identify the best-performing material. The optimized electrode was then combined with a carbonaceous counter electrode to assemble asymmetric two-electrode supercapacitor devices. The electrochemical charge-storage behaviour of the fabricated electrodes was systematically characterised using cyclic voltammetry, galvanostatic charge–discharge analysis, cycling stability testing, and energy/power density evaluation in both three-electrode and two-electrode configurations. Finally, specific energy, specific power, and cycling performance of the fabricated devices were compared with recently reported values to highlight the advantages of the hydrothermally prepared SrS-based electrodes for practical energy storage applications.

2. Materials and Methods

2.1. Materials

Strontium nitrate ($\text{Sr}(\text{NO}_3)_2$) and sodium sulfide (Na_2S) were purchased from Merck (Darmstadt, Germany). Aniline ($\text{C}_6\text{H}_5\text{NH}_2$), ammonium persulfate (APS, $(\text{NH}_4)_2\text{S}_2\text{O}_8$), carbon black, polyvinylidene fluoride (PVDF), and N-methyl-2-pyrrolidone (NMP, 99.5%) were supplied by Sigma-Aldrich (St. Louis, MO, USA). Nickel foam was obtained from Urich New Tech Sdn. Bhd., Sungai Petani, Kedah, Malaysia. All chemicals were of analytical grade and were used as received without further purification. Deionized water (DI water, $18.2 \text{ M}\Omega\cdot\text{cm}$ at 25°C) was used throughout the experiments.

2.2. Preparation of Strontium Sulfide (SrS)

Strontium sulfide (SrS) was synthesized via a hydrothermal method, as illustrated in Figure 1. In a typical procedure, strontium nitrate ($\text{Sr}(\text{NO}_3)_2$) was dissolved in 50 mL of deionized (DI) water under constant magnetic stirring. Separately, sodium sulfide (Na_2S) was dissolved in another 50 mL of DI water and added dropwise to the $\text{Sr}(\text{NO}_3)_2$ solution at 30 °C while stirring continuously. The resulting mixture was transferred to a Teflon-lined stainless-steel autoclave, sealed, and maintained at 180 °C for 16 h. After natural cooling to room temperature, the white precipitate was collected by centrifugation at 6000 rpm for 15 min, washed several times with DI water and absolute ethanol to remove residual ions and impurities, and dried in an oven at 100 °C for 12 h.

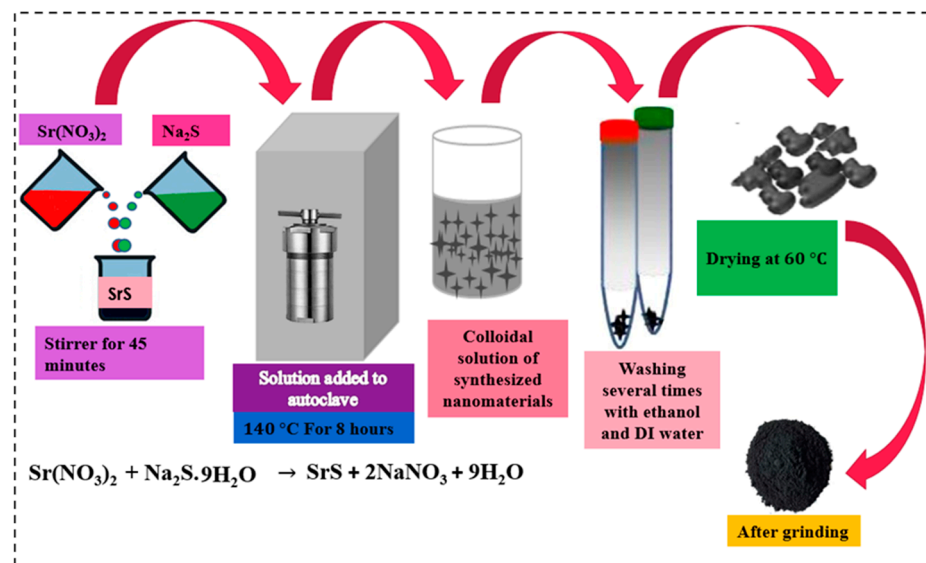


Figure 1. Schematic illustration of the hydrothermal synthesis of SrS nanomaterials. The red arrows indicate the sequential synthesis steps, including reagent mixing, stirring, hydrothermal treatment, washing, drying, and grinding.

2.3. Preparation of Polyaniline (PANI) and SrS/PANI Composite Synthesis

Polyaniline (PANI) was prepared by chemical oxidative polymerization of aniline [27]. Two solvents were used: hydrochloric acid and a 30 mL solution containing 0.2 M APS. The APS solution was added dropwise in small portions to the aniline mixture while maintaining a constant low temperature. Continuous stirring was applied during the preparation of the aniline solution. A dark-green precipitate appeared as the reaction proceeded. The deep-green product was repeatedly washed with distilled deionized water to remove any impurities and pollutants. Afterward, the SrS/PANI composite was synthesized using the same polymerization procedure in the presence of SrS.

2.4. Electrode Manufacturing

Nickel foam was used as the current collector and substrate for electrochemical characterization. The electrode slurry was prepared by mixing 75 wt% active material (SrS/PANI), 15 wt% acetylene black (conductive additive), and 10 wt% polyvinylidene fluoride (PVDF) binder in N-methyl-2-pyrrolidone (NMP). The mixture was stirred continuously for 9 h to obtain a homogeneous slurry. This slurry was then coated onto 1 cm² pieces of pre-treated nickel foam. The coated electrodes were dried overnight at 90 °C in a vacuum oven. The active material mass loading on the nickel foam was approximately 5.5 mg cm^{−2}. For the asymmetric supercapacitor device, SrS/PANI-coated nickel foam was used as the positive electrode, and activated carbon (AC)-coated nickel foam as the negative elec-

trode. A cellulose separator soaked in 3 M KOH aqueous electrolyte was placed between the two electrodes to prevent short-circuiting. The full device was finally assembled in a CR2032 coin-cell configuration.

2.5. Electrochemical Performance Evolution

Electrochemical testing was conducted in a three-electrode configuration, employing Hg/HgO as the reference electrode and a platinum wire as the counter electrode [28]. The working electrode consisted of SrS or SrS/PANI coated on pretreated nickel foam, as described in Section 2.4. A 3 M KOH aqueous solution served as the electrolyte for all three-electrode measurements.

Multiple characterization techniques were applied to assess device performance. Cyclic voltammetry (CV) was used to probe the redox behavior of the electrodes. At the same time, the rate capability and charge-storage mechanism were assessed from the CV and GCD profiles across multiple current densities. The energy-storage capability of the fabricated electrodes was further evaluated through galvanostatic charge–discharge (GCD) measurements.

2.6. Device Testing

An asymmetric supercapacitor (ASSC) was assembled using SrS/PANI as the positive electrode and activated carbon (AC) as the negative electrode, both deposited on nickel foam [29]. The electrodes were immersed in a 3.0 M KOH aqueous electrolyte. Cyclic voltammetry (CV) measurements were performed over the potential range of 0–1.6 V, with scan rates of 5–105 mV s^{−1}. Galvanostatic charge–discharge (GCD) tests were carried out over the same voltage range (0–1.6 V) at current densities between 1.5 and 6 A g^{−1} to assess charge–discharge behaviour with scan rates of 5–40 mV s^{−1}. Specific capacitance (C) for both the device and the three-electrode configuration was calculated using standard relationships derived from CV and GCD data [30]:

$$C = \frac{I}{mv} \int_{V_i}^{V_f} I \times V dV$$

In the present situation, C denotes the assigned capacity and CV area, while v and m denote the scan rate and active mass, respectively. The initial value (V_i = 0 V) and the final value (V_f = 0.7 V) indicate the potential window. This equation may be used to get the GCD of the specific capacity.

$$C = \frac{I \times \Delta t}{m \Delta V}$$

Here, I is the applied current (A), Δt is the discharge time (s), m is the total active mass of both electrodes (g), and ΔV is the potential window excluding the IR drop. For the three-electrode configuration, the specific capacitance was calculated using the same equations. Still, m corresponds to the mass of the working electrode only, and ΔV is the operating potential window of the working electrode. The energy density (E, Wh kg^{−1}) and power density (P, W kg^{−1}) of the ASSC device were determined using the following relationships:

$$E = \frac{Cs(\Delta V)}{2 \times 3.6}$$

$$P = \frac{Ed \times 3600}{t}$$

where ΔV is the operating potential window (V), and Δt is the discharge time (s).

3. Results

3.1. Structural and Morphological Study

Figure 2 presents the X-ray diffraction (XRD) patterns of pristine SrS and the SrS/PANI composite. XRD analysis was used to examine the crystalline structure and phase characteristics of the synthesized materials. The average crystallite size (D) was calculated using the Scherrer equation:

$$D = K\lambda / \beta \cos \theta$$

where K is the shape factor (typically 0.9), λ is the X-ray wavelength (Cu $K\alpha = 0.15406$ nm), β is the full width at half maximum (FWHM) of the selected diffraction peak in radians, and θ is the Bragg angle. The crystallite-size calculation was performed using the most intense and well-resolved SrS diffraction peak corresponding to the (200) plane at approximately $2\theta = 30.7^\circ$. The peak-position, FWHM, and calculated crystallite-size values are summarized in Table 1. The calculated crystallite sizes were 17.2 nm for pristine SrS and 16.5 nm for the SrS/PANI composite.

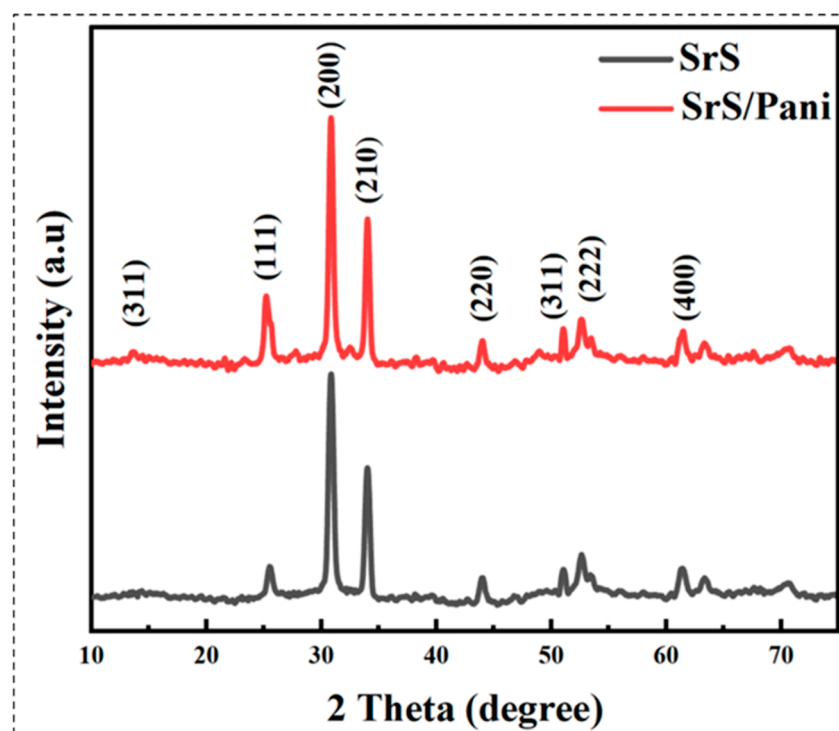


Figure 2. XRD pattern of SrS and SrS/PANI.

Table 1. Scherrer crystallite-size calculation parameters derived from the dominant SrS (200) diffraction peak.

Sample	Selected Peak	2θ ($^\circ$)	θ ($^\circ$)	FWHM β ($^\circ$)	FWHM β (Rad)	Crystallite Size D (nm)
Pristine SrS	(200)	30.70	15.35	0.50	0.00873	17.2
SrS/PANI	(200)	30.81	15.41	0.52	0.00908	16.5

The XRD pattern of pristine SrS exhibits characteristic diffraction peaks at 2θ values of approximately 25.0° , 30.7° , 34.0° , 44.1° , 50.9° , 52.7° , and 61.5° , corresponding to the indexed SrS planes shown in Figure 2. After the formation of the SrS/PANI composite, the main SrS diffraction peaks are retained, indicating that the crystalline structure of SrS is largely preserved during the in situ polymerization process. Compared with pristine SrS,

the SrS/PANI composite shows slight changes in peak intensity and a modest increase in background intensity, which can be associated with the presence of the PANI phase. No distinct crystalline peak of PANI is observed, suggesting that PANI is likely amorphous or weakly crystalline, or that its contribution is masked by the stronger SrS diffraction peaks.

The surface morphology was further examined by scanning electron microscopy (SEM). Figure 3a shows that pristine SrS consists of well-defined, faceted nanosized particles with sharp edges and polyhedral morphology, with particle/agglomerate sizes mainly in the range of 80–150 nm. In comparison, the SEM image of the SrS/PANI composite shown in Figure 3b exhibits a rougher and more heterogeneous surface morphology. The original faceted morphology of SrS appears partially covered by granular surface features, which can be attributed to the formation of PANI during in situ polymerization. This morphological change supports the successful preparation of the SrS/PANI composite.

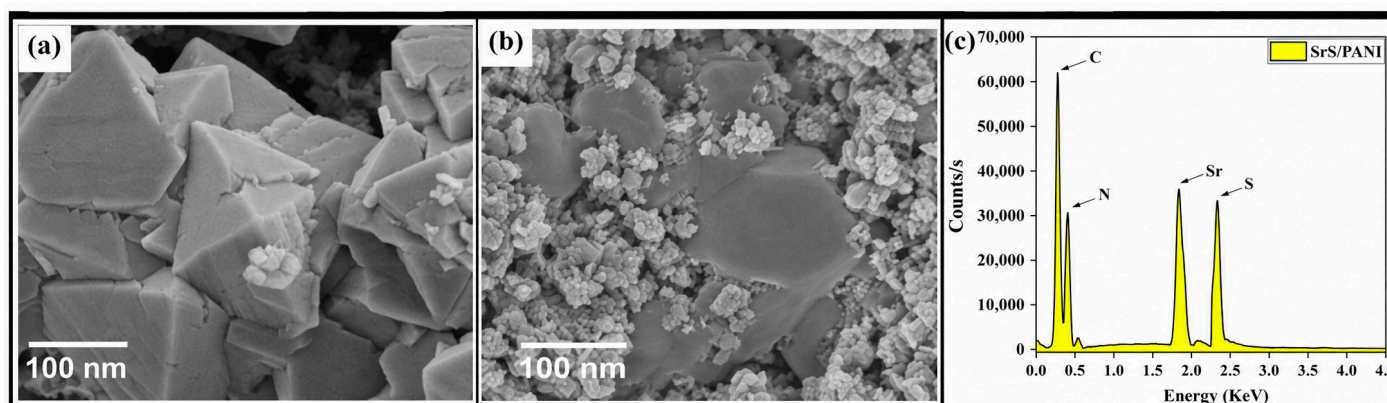


Figure 3. (a,b): SEM images of SrS and SrS/PANI. (c) The EDX graph of the SrS/PANI composite.

The elemental composition and chemical purity of the synthesized SrS/PANI composite were investigated using energy-dispersive X-ray spectroscopy (EDX). As presented in Figure 3c, the EDX spectrum exhibits characteristic signals corresponding to C, N, Sr, and S elements, confirming the successful formation of the SrS/PANI hybrid structure. The C and N peaks originate from the polyaniline (PANI) polymer chain, whereas the Sr and S signals are attributed to the SrS inorganic phase. The Sr and S elements are identified through their characteristic X-ray emission lines, where the Sr peaks mainly arise from the Sr L-series transitions, while sulfur is typically detected through its S K α emission line. The observed elemental signals confirm the coexistence of the inorganic SrS phase and the organic conducting polymer matrix. No additional peaks associated with foreign elements such as Cl, Na, Ca, Si, or other metallic impurities were detected. It should be noted that hydrogen (H) is not observed in the EDX spectrum because EDX detectors are generally unable to detect light elements with very low atomic numbers. The presence of both Sr and S peaks with an approximately 1:1 atomic ratio (Sr:12.00 at.% and S:12.00 at.%) confirms the formation of the SrS phase with the expected stoichiometric composition. Meanwhile, the dominant carbon contribution (65.00 at.%) and nitrogen content (11.00 at.%) originate from the PANI structure, indicating successful incorporation of the conducting polymer component. The quantitative elemental analysis obtained from EDX is summarized in Table 2. The composite consists of 44.35 wt.% Sr, 16.23 wt.% S, 32.93 wt.% C, and 6.49 wt.% N, corresponding to 12.00, 12.00, 65.00, and 11.00 at.%, respectively. The obtained elemental distribution confirms the successful formation of the SrS/PANI hybrid composite and demonstrates the coexistence of the inorganic SrS phase with the organic conducting polymer network.

Table 2. Elemental composition of SrS/PANI composite obtained from EDX analysis.

Element	Emission Line	wt. %	at. %
Strontium (Sr)	L α	44.35	12.00
Sulfur (S)	K α	16.23	12.00
Carbon (C)	K α	32.93	65.00
Nitrogen (N)	K α	6.49	11.00
Total	—	100.00	100.00

3.2. Three-Electrode Assembly Measurement

3.2.1. Cyclic Voltammetry (CV)

The electrochemical behaviour of pristine SrS and SrS/PANI composite electrodes was investigated using a three-electrode configuration. Cyclic voltammetry was performed in 3 M KOH aqueous electrolyte within a 0.6 V potential window for both pristine SrS and SrS/PANI composite electrodes. The CV curves of the prepared electrodes are shown in Figure 4 at scan rates ranging from 3 to 50 mV s^{−1}.

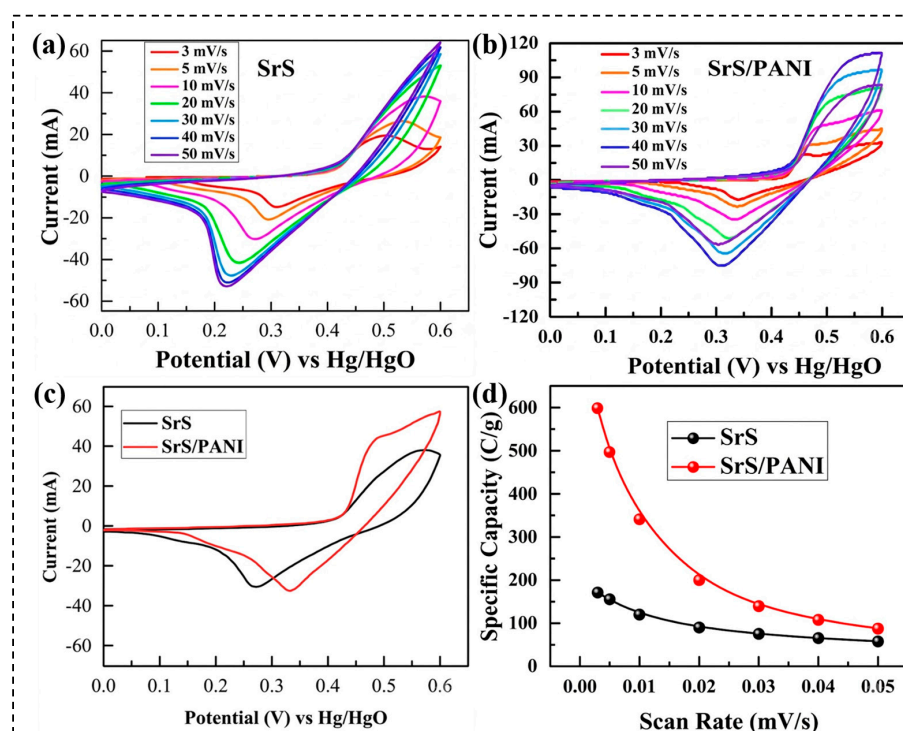
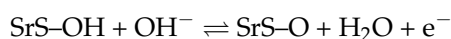
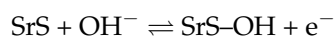


Figure 4. (a) CV curves of pristine SrS at scan rates of 3–50 mV s^{−1}; (b) CV curves of SrS/PANI composite at scan rates of 3–50 mV s^{−1}; (c) comparative CV curves of SrS and SrS/PANI at 3 mV s^{−1}; (d) specific capacity as a function of scan rate for all electrode samples.

Distinct redox peaks are observed for the tested electrodes, indicating Faradaic charge-storage behaviour associated with reversible redox reactions at the electrode–electrolyte interface in the KOH electrolyte. The possible surface redox reactions of SrS in alkaline KOH electrolyte can be expressed as follows:



Overall:



These reactions suggest that OH^- ions from the alkaline electrolyte participate in reversible surface hydroxylation/oxidation of SrS during the charge–discharge process. Therefore, the redox peaks observed in the CV curves can be associated with Faradaic reactions occurring at the SrS/electrolyte interface.

With increasing scan rate, the redox peaks become more intense, suggesting a rapid electrochemical response of the electrode materials. The SrS/PANI (50/50 wt%) composite outperformed the other tested electrodes, delivering a CV-derived specific capacity of 192 C g^{-1} at 3 mV s^{-1} . For comparison, pristine SrS delivered a CV-derived specific capacity of 127 C g^{-1} at the same scan rate.

The charge-storage performance of the electrode is influenced by ion diffusion, electrolyte accessibility, electrode–electrolyte contact, and charge-transfer behaviour. At higher scan rates, the specific capacity decreases because the shorter interaction time limits ion diffusion and complete utilization of the active material. The incorporation of PANI affects the charge-storage response of SrS, and the optimized SrS/PANI (50/50 wt%) composite shows improved rate capability, maintaining a comparatively higher capacity even at 50 mV s^{-1} , as shown in Figure 4d. The CV curves of the SrS/PANI composite retain their overall shape at higher scan rates, indicating acceptable rate capability and reversible electrochemical behaviour.

The specific capacity of the electrode material was calculated using the following equation:

$$Q_s = \frac{1}{2vm \int_{v_i}^{v_f} I \times V dv}$$

This equation represents the required capacity as Q , with the area under the curve, the scan rate, and the active mass on nickel foam denoted in Figure 4d, which shows the CV-derived specific capacity of each electrode as a function of scan rate. Pristine SrS and the optimized SrS/PANI (50/50 wt%) composite exhibited CV-derived specific capacities of 127 and 192 C g^{-1} , respectively, at 3 mV s^{-1} .

The rate-capability comparison in Figure 4d shows that the SrS/PANI composite retains a higher proportion of its capacity at elevated scan rates relative to pristine SrS, indicating improved charge-storage performance.

3.2.2. Galvanostatic Charging and Discharging (GCD)

Galvanostatic charge–discharge measurements were performed to further evaluate the charge-storage behaviour, rate performance, and specific capacity of pristine SrS and SrS/PANI composite electrodes. The measurements were carried out in a three-electrode configuration using 3 M KOH aqueous electrolyte, with Hg/HgO as the reference electrode. The electrodes were tested within a constant potential window of 0.6 V at current densities ranging from 0.4 to 2.0 A g^{-1} .

The GCD curves of pristine SrS and the SrS/PANI composite are shown in Figure 5. The charge–discharge profiles exhibit non-linear behaviour, indicating the presence of Faradaic redox reactions during the charge-storage process. This behaviour is consistent with the redox peaks observed in the CV curves. Compared with pristine SrS, the SrS/PANI (50/50 wt%) composite shows a longer discharge duration, suggesting improved charge-storage capability. A noticeable IR drop is observed at the beginning of the discharge process, particularly at higher current densities. This instantaneous voltage drop reflects the internal resistance and polarization of the electrode system, including the electrolyte resistance, intrinsic resistance of the electrode material, and contact resistance between the active material and the nickel foam current collector. In general, a smaller IR drop indicates

lower internal resistance and better charge-transfer behaviour, which is beneficial for rate capability and power performance. The GCD profiles suggest that the SrS/PANI composite exhibits acceptable polarization behaviour during rapid charge–discharge operation, supporting its improved rate performance compared with pristine SrS.

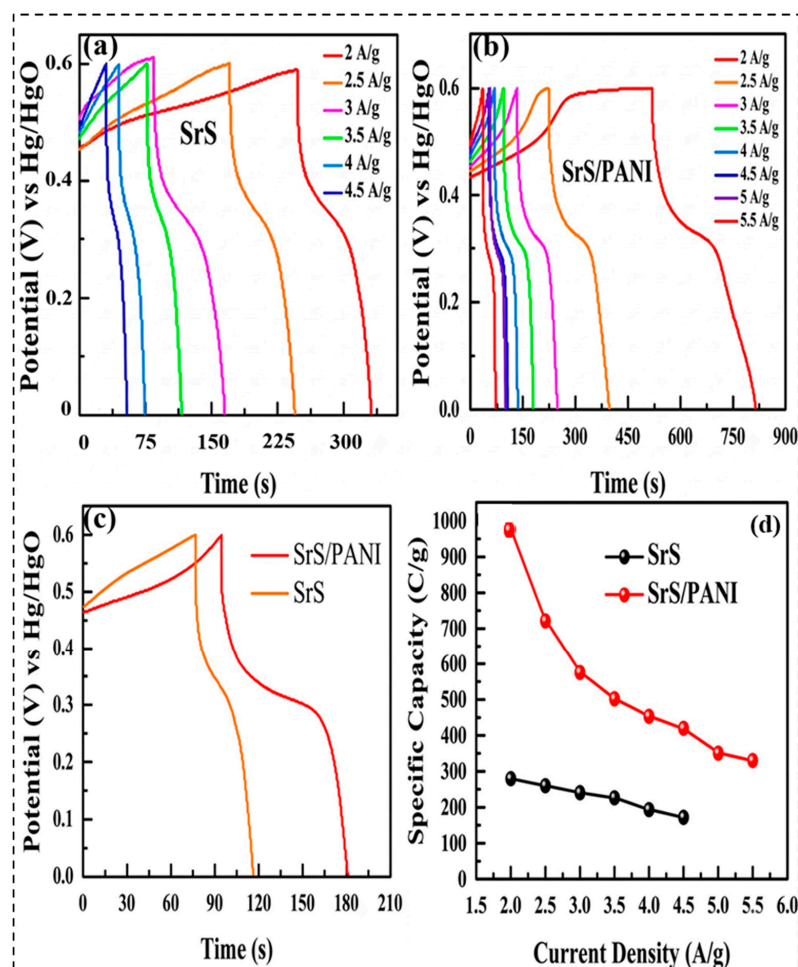


Figure 5. (a,b) GCD of SrS and SrS/PANI. (c) Comparison of GCD Curves. (d) GCD-specific capacity comparison.

The discharge time is longer than the charge time in the GCD profiles, which can be associated with the Faradaic/battery-type charge-storage behaviour of the SrS/PANI electrode and the relatively slow ion-diffusion process during discharge. In addition, electrode polarization and internal resistance, as reflected by the IR drop, can contribute to the asymmetry between the charge and discharge branches. Although the 0–0.6 V potential window was used consistently for the present GCD analysis, narrowing the potential window to 0–0.5 V may reduce polarization and improve charge–discharge balance.

The specific capacity was also calculated from the discharge curves using the following formula:

$$Q_s = \frac{I \times \Delta t}{m}$$

where Q_s is the specific capacity, I is the discharge current, Δt is the discharge time, and m is the active mass of the electrode material. The SrS/PANI (50/50 wt%) composite delivered the highest GCD-derived specific capacity of 580 C g^{-1} at 0.4 A g^{-1} , compared with 240 C g^{-1} for pristine SrS under the same current density. As shown in Figure 5d, the specific capacity of all electrodes decreases with increasing current density. This decrease is mainly caused by the shorter discharge time at higher current density, which limits

OH^- ion diffusion into the active electrode material and reduces the complete utilization of available redox-active sites. In addition, the increased polarization and IR drop at higher current density can further restrict charge transfer and lower the measured capacity. Therefore, although the SrS/PANI composite maintains a higher specific capacity than pristine SrS over the tested current-density range, the relatively rapid capacity decrease indicates that further optimization of electrode conductivity, active-material dispersion, and electrode/electrolyte interfacial contact is required to improve high-rate performance.

The improved electrochemical performance of the SrS/PANI composite is due to the combined contribution of both components. SrS acts as the main redox-active component and contributes to Faradaic charge storage through reversible reactions with OH^- ions in the alkaline electrolyte. PANI, as a conducting-polymer component, may assist electron transport and help maintain electrode integrity during repeated charge–discharge processes.

3.2.3. Charge-Storage Kinetics (b-Value Analysis)

To quantitatively evaluate the charge-storage kinetics, the relationship between the peak current (i_p) and scan rate (v) was analysed using the power law:

$$i_p = a \cdot v^b$$

which in logarithmic form becomes:

$$\log(i_p) = b \cdot \log(v) + \log(a)$$

The b-value, obtained from the slope of $\log(i_p)$ versus $\log(v)$, indicates the dominant charge-storage mechanism: a value near 0.5 corresponds to a diffusion-controlled (battery-type) process, whereas a value near 1.0 indicates a surface-controlled (capacitive) process.

Figure 6 shows the $\log(i_p)$ – $\log(v)$ plots for the cathodic peaks of the SrS and SrS/PANI electrodes. Linear fitting yielded b-values of approximately 0.68 for both electrodes ($R^2 > 0.99$), indicating that charge storage proceeds through a combination of diffusion-controlled and capacitive contributions rather than a single mechanism. These b-values close to 0.7 confirm the mixed (hybrid) charge-storage behaviour, consistent with the non-linear GCD profiles and the redox peaks observed in the CV curves. The similar b-values for SrS and SrS/PANI indicate that incorporating PANI does not fundamentally change the charge-storage mechanism but rather enhances the overall specific capacity and charge transport, in agreement with the higher capacity measured for the composite.

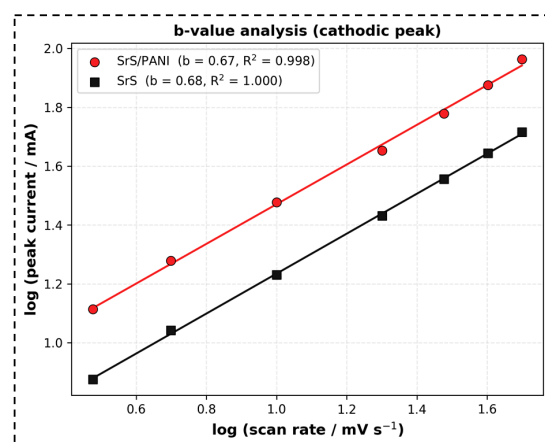


Figure 6. B-value analysis showing $\log$ (peak current) versus $\log$ (scan rate) for the cathodic peaks of the SrS and SrS/PANI electrodes, with the corresponding b-values and R^2 obtained from linear fitting.

3.3. Supercapattery Assembly

To demonstrate the potential applications of the produced material, an asymmetric supercapacitor device was fabricated using a two-electrode configuration [31,32]. SrS/PANI was used as the positive electrode material, while AC served as the negative electrode. A permeable membrane separator was placed between the two electrodes [33]. A schematic depiction of the device is shown in Figure 7a.

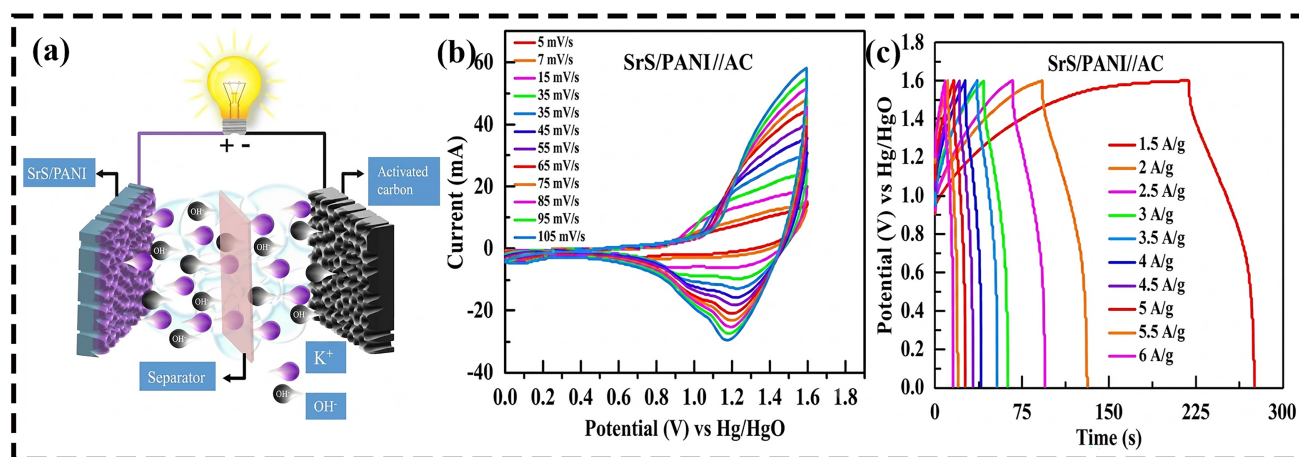


Figure 7. (a) Schematic illustration of the asymmetric supercapattery device. (b) CV of activated carbon and SrS/PANI at 5–105 mV/s. (c) Galvanic curves of the device.

The cyclic profile reveals how the device stores charge; in other words, AC contributes to non-faradaic processes, while SrS provides faradaic reactions [34]. Furthermore, the results confirm that the potential window of 0–1.6 V is stable and reliable for measuring asymmetric SCs. As shown in Figure 7b, the cyclic voltammogram of the device was recorded in a two-electrode cell at different scan rates (5–105 mV s^{−1}). The family of cyclic curves exhibits quasi-rectangular behavior, with a slight hump indicating the effect of the battery-grade electrode material (Faradaic reactions) and the rectangular portion highlighting the capacitive nature of AC (non-Faradaic reactions). These curves for the SrS/PANI-based device similarly retain their shape up to 105 mV s^{−1}, indicating a respectable rate capability of the hybrid device.

To further explore the electrochemical performance of the produced device, galvanostatic investigations were carried out in a two-electrode configuration at various current densities (Jd, 1.5–6 A g^{−1}), as seen in Figure 7c. A 3 M KOH aqueous electrolyte was used, and the device was operated within a constant potential window of 0–1.6 V. The device's GCD profile is nonlinear. As shown in the highlighted area, the triangular behavior indicates a capacitive nature, while the tiny spikes in the galvanostatic profile indicate redox activity. These findings demonstrate the existence of capacitive characteristics and reversible faradaic reactions. The galvanostatic curves clearly show that the electric double layer and redox processes contributed equally to the overall performance of the supercapacitor device.

To evaluate the cycling behaviour of the SrS/PANI//AC asymmetric device, 1000 consecutive charge–discharge cycles were performed, as shown in Figure 8a,b. The device retained 75% of its initial specific capacity after 1000 cycles, while the Coulombic efficiency remained approximately 90% during the cycling test. These results indicate acceptable electrochemical reversibility within the tested cycling range. However, because the cycling test was limited to 1000 cycles, an extended cycling test of 5000 cycles or more is required to more rigorously evaluate the long-term durability of the SrS/PANI-based asymmetric device.

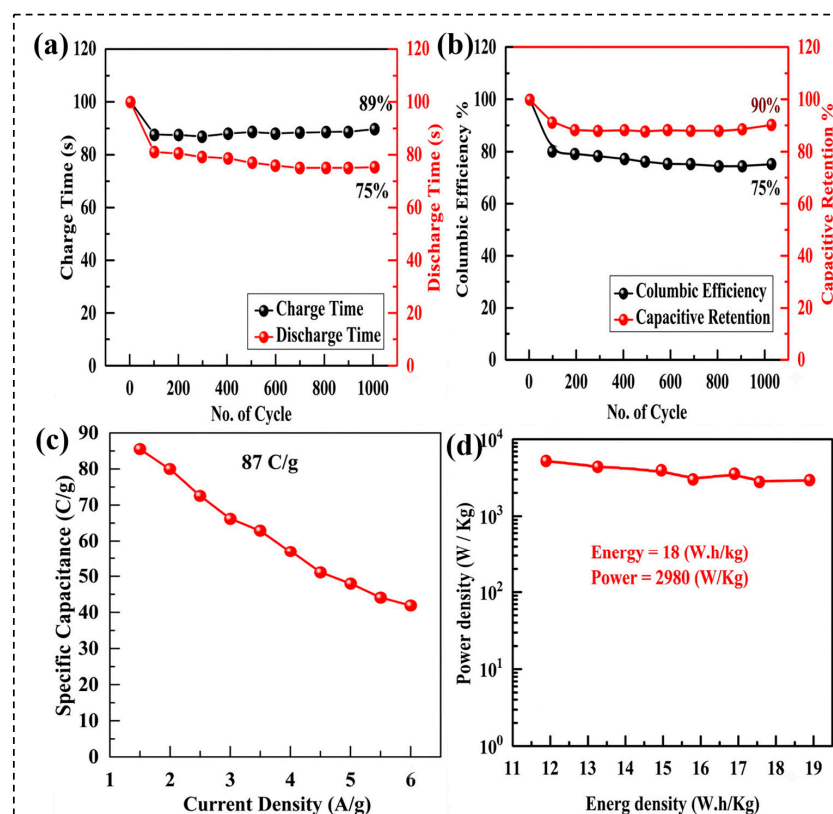


Figure 8. (a) Charge–discharge time measured for the device observed for 1000 cycles. (b) Stability measurement of the device observed for 1000 cyclic charge–discharges. (c) Specific capacity of SrS/PANI device calculated from GCD curve. (d) Plot exploring the energy density as a function of power density.

The following relations have been used to evaluate specific energy and power:

$$E = \frac{Q_s(\Delta V)}{2 \times 3.6} \quad (1)$$

$$P = \frac{Ed \times 3600}{\Delta t} \quad (2)$$

Here, Δt is the discharging time, and ΔV represents the potential window. Figure 8b shows that the device achieved a Q_s of 87 C/g at 6 A/g. Figure 8c shows the specific capacity of the device as a function of current density, reaching 87 C/g at the highest applied current density of 6 A g^{−1}, with higher values obtained at lower current densities. As shown in Figure 8d, the device achieved a power density of 2980 W/kg and an energy density of 18 Wh/kg.

To contextualise the performance of the fabricated SrS/PANI//AC asymmetric device within the current literature, Table 3 presents a direct comparison with recently reported PANI-based composite supercapattery systems. The SrS/PANI device delivers a competitive energy density of 18 Wh kg^{−1} and a high power density of 2980 W kg^{−1} using a simple two-component, hydrothermally prepared electrode. While ternary and binder-free systems such as CuCoS–PANI and MnNbS–PANI achieve higher specific capacities due to additional redox-active components and optimised morphologies, the present SrS/PANI composite offers a simpler synthesis route, lower material cost, and a wide operating voltage of 1.6 V. These results suggest that the SrS/PANI electrode is a promising low-cost candidate for hybrid energy-storage applications, although extended cycling tests are still required to fully assess its long-term durability.

Table 3. Comparison of Electrochemical Performance of SrS/PANI and Related Metal Sulfide/PANI Composite Asymmetric Supercapacitor Devices.

Electrode (Positive//Negative)	Synthesis Method	Electrolyte	Specific Capacity Electrode (C g ⁻¹)	Specific Capacity Device (C g ⁻¹)	Energy Density (Wh kg ⁻¹)	Power Density (W kg ⁻¹)	Cycling Stability	Ref.
SrS/PANI//AC	Hydrothermal + oxidative polymerization	3 M KOH	580 (GCD, 0.4 A g ⁻¹)	87 (6 A g ⁻¹)	18	2980	75%/ 1000 cycles	This work
CuCoS-PANI//AC	Hydrothermal (binder-free, Ni foam)	3 M KOH	1659 (1.0 A g ⁻¹)	-	54	1150	-	[35]
NiMnS-PANI//AC	Hydrothermal + chemical polymerization	1 M KOH	976 (three- electrode)	160 (7 A g ⁻¹)	35.73	850	98%/ 10,000 cycles	[36]
MnNbS/PANI//AC	Hydrothermal + blending	Not reported	1366 (2 A g ⁻¹)	118	26.2	2072	79%/ 1000 cycles	[37]
PANI@NbS//PANI@AC	Hydrothermal + polymerization	Not reported	1050	1207	-	-	85%/ 1000 cycles	[38]

4. Conclusions

This study reports the successful synthesis of strontium sulfide (SrS) and SrS/polyaniline (SrS/PANI) nanocomposites via a straightforward hydrothermal approach, followed by a comprehensive electrochemical assessment for hybrid energy-storage applications. Structural, morphological, and elemental analyses verified the proper formation of the composite and confirmed the effective incorporation of polyaniline with SrS particles. Electrochemical evaluation demonstrated a clear performance advantage for the SrS/PANI composite over pristine SrS. The optimized SrS/PANI composite electrode exhibited pronounced redox peaks, a CV-derived specific capacity of 192 C g⁻¹ at 3 mV s⁻¹, and a GCD-derived specific capacity of 580 C g⁻¹ at 0.4 A g⁻¹. The assembled SrS/PANI//AC asymmetric device delivered a device-level specific capacity of 87 C g⁻¹ at 6 A g⁻¹ and retained 75% of its initial capacity after 1000 cycles. These enhancements are mainly associated with the redox-active nature of SrS and the incorporation of PANI as a conducting-polymer component, which may support charge transport and structural stability during repeated cycling. When configured as an asymmetric device using SrS/PANI as the positive electrode and activated carbon as the negative electrode, the full cell delivered a wide operating voltage of 1.6 V, a specific capacity of 87 C g⁻¹, an energy density of 18 Wh kg⁻¹, and a power density of 2980 W kg⁻¹, together with acceptable cycling durability over 1000 cycles. Overall, the SrS/PANI nanocomposite emerges as a low-cost, high-performance electrode candidate for next-generation hybrid supercapacitor (supercapattery) systems. These findings provide a foundation for further optimization, scale-up studies, long-term operation of environmental monitoring equipment, and durability investigations to advance the material toward practical energy-storage deployment. Future studies should incorporate elemental mapping and advanced spectroscopic characterization to verify the elemental distribution, composition, and possible interfacial interactions within the SrS/PANI composite. In addition, extended cycling tests of 5000 cycles or more, together with post-cycling structural and morphological analyses, are required to more rigorously evaluate the long-term durability of the SrS/PANI-based asymmetric device.

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R.C., visualization; Y.P. (Yinlong Pan), funding acquisition, project administration; H.Z., resources, visualization. All authors have read and agreed to the published version of the manuscript.

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