

Article

Facile Synthesis of Salt-Assisted Multiroom Carbon/Vanadium Sulfide Microspheres for Fast and Durable Potassium-Ion Storage

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Abstract

Potassium-ion batteries hold great promise for large-scale energy storage, but their commercialization is hindered by the large ionic radius of potassium, which causes sluggish kinetics and severe volume expansion in anode materials. To address this, we present a scalable spray-drying strategy coupled with NaCl salt-templating to synthesize hierarchical porous carbon/vanadium sulfide microspheres (p-V₃S₄/C MS). In this structure, V₃S₄ nanoparticles are uniformly encapsulated within a dextrin-derived amorphous carbon matrix, and pores are formed via selective NaCl etching. This unique architecture accommodates volume fluctuations while providing rapid ion diffusion pathways. As a result, the p-V₃S₄/C MS anode exhibits outstanding electrochemical performance, maintaining a reversible capacity of 107 mA h g^{−1} after 2000 cycles at 2.0 A g^{−1}, and achieves a high pseudocapacitive contribution of 93% at 2.0 mV s^{−1}. Furthermore, a full cell paired with a Prussian blue (PB) cathode demonstrates practical viability and robust reversibility. Our findings demonstrate that this structural engineering effectively mitigates internal resistance and structural degradation, offering a cost-effective route for mass-producing high-performance anodes for next-generation energy storage.

Keywords: potassium-ion batteries; porous microspheres; vanadium sulfide; composite; spray-drying

1. Introduction

Over the past few decades, lithium-ion batteries (LIBs) have been central to the modern energy industry, powering portable electronics, electric vehicles (EVs), and large-scale energy storage systems (ESSs) [1–3]. However, the uneven distribution, rising mining costs, and limited reserves of lithium hinder their sustainable expansion. Consequently, potassium-ion batteries (PIBs) have emerged as a promising alternative due to the high abundance and low cost of potassium [4,5]. Notably, the redox potential of potassium (−2.93 V vs. standard hydrogen electrode) is comparable to that of lithium. This feature enables high operating voltages and energy densities competitive with LIBs [6,7].

Despite its potential, the commercial viability of PIBs remains in a nascent stage. A critical challenge arises from the large ionic radius of potassium ions (1.38 Å) [8–10]. Repeated potassiation and depotassiation induce severe volumetric expansion within the electrode host. This mechanical strain leads to structural pulverization and electrical isolation, causing rapid capacity decay [11,12]. Furthermore, the sluggish kinetics inherent to



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the large ion size severely restricts high-rate capability. Consequently, designing advanced anode architectures that can accommodate volume fluctuations while preserving efficient ion and electron transport pathways is imperative [13,14].

To address the capacity limitations of commercial graphite, layered transition metal chalcogenides (LTMCs) with large interlayer spacing have been proposed as alternatives. LTMCs features a structure where transition metal (M) and chalcogen (X) layers are held together by weak van der Waals forces [15,16]. This configuration provides a favorable environment for potassium ion diffusion. Unlike typical LTMCs (e.g., MoS_2), which exhibit semiconducting behavior with low conductivity, vanadium sulfides (e.g., VS_2 , V_3S_4 , V_5S_8) possess metallic conductivity [17–19]. This property offers a distinct advantage in charge transfer efficiency. Among these, V_3S_4 strikes an optimal balance between structural stability and capacity. Interstitial vanadium atoms induce a “self-pillaring effect” that strongly reinforces the interlayer structure [20]. This mechanism suppresses particle exfoliation and preserves lattice integrity, thereby enabling simultaneous high capacity and long-term cyclability.

Despite these intrinsic merits, low electrical conductivity and substantial volume expansion remain critical hurdles for practical application. Thus, sophisticated structural design is imperative to ensure stable performance within the electrode environment [21]. In this approach, encapsulating active materials within a uniform carbon matrix serves a dual purpose. It acts as a buffer to physically accommodate volume expansion stress. Simultaneously, it reinforces inter-particle electrical connectivity and minimizes parasitic side reactions with the electrolyte [22–24]. Furthermore, introducing porous or hollow architectures increases the electrolyte–electrode contact area. This shortens ion diffusion paths and accelerates reaction kinetics [25,26]. Additionally, engineering spherical morphologies disperses mechanical stress concentrations. It also secures high tap density, which is favorable for electrode fabrication [27,28]. However, conventional synthesis strategies, such as hard-templating or hydrothermal methods, face severe limitations in scalability. Their complex procedures and low cost-effectiveness hinder mass production.

In this work, we propose a facile strategy to synthesize porous microspheres in which V_3S_4 nanoparticles are uniformly embedded within a conductive amorphous carbon matrix (denoted as p- $\text{V}_3\text{S}_4/\text{C MS}$). This is achieved by combining a cost-effective and eco-friendly NaCl template with a spray-drying process. NaCl serves as a pore-forming agent, enabling precise pore engineering. Dextrin, selected for its high water solubility and compatibility with aqueous processing, is utilized as the carbon source to construct a uniform and robust carbon network on the active material surface. Notably, during the subsequent one-step heat treatment, the carbon matrix promotes a carbothermal reduction effect. This facilitates the formation of a highly crystalline V_3S_4 phase. The resulting p- $\text{V}_3\text{S}_4/\text{C MS}$ demonstrates superior electrochemical performance, attributed to the mechanical buffering provided by the porous architecture and the enhanced electrical connectivity of the carbon scaffold. These results suggest that our proposed synthesis route offers an efficient and scalable pathway for the mass production of high-performance anode materials for PIBs.

2. Results and Discussion

2.1. Synthesis Process and SEM Analysis

Figure 1 illustrates the overall synthesis process of porous p- $\text{V}_3\text{S}_4/\text{C MS}$. This synthesis strategy employs a facile salt-templating method combined with spray-drying, involving three precisely controlled stages. First, spherical precursors comprising ammonium metavanadate (NH_4VO_3), dextrin, and NaCl are prepared via spray-drying. Subsequently, a single-step heat treatment induces simultaneous carbonization and sulfidation. During this process, dextrin transforms into conductive carbon matrix, while the vanadium precursor

crystallizes into the V_3S_4 phase. Finally, a highly developed porous structure is realized by removing the sacrificial NaCl template through selective etching.

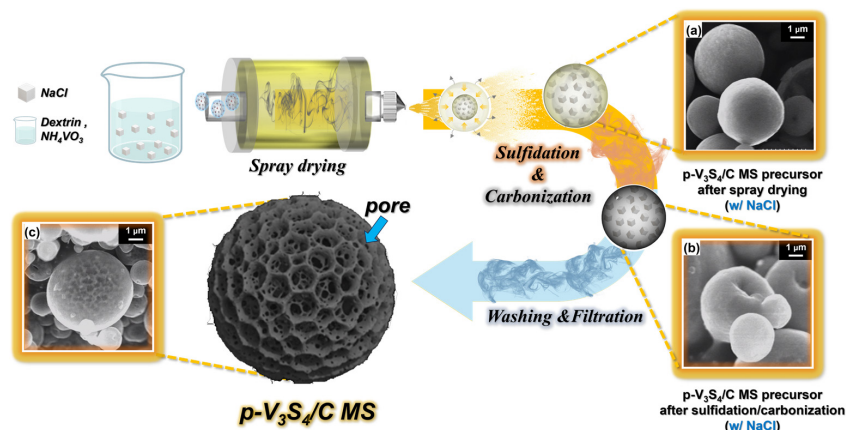


Figure 1. Morphological evolution of the $p\text{-V}_3\text{S}_4/\text{C}$ MS composite at different synthesis stages: SEM images obtained after (a) spray drying, (b) simultaneous sulfidation and carbonization, and (c) NaCl washing.

The morphological evolution and structural integrity of the samples at each synthesis stage were investigated using scanning electron microscopy (SEM). As shown in Figure 1a, the spray-dried precursors exhibit a smooth surface and a well-defined spherical geometry. Following sulfidation and carbonization (Figure 1b), the sacrificial NaCl templates were selectively removed by washing with deionized (DI) water. The complete dissolution of both internal and external NaCl crystals resulted in the formation of a highly porous spherical structure (Figure 1c). Notably, despite the formation of extensive voids, the microspheres retained their perfect spherical shape without any collapse or fracture. This structural stability is attributed to the three-dimensionally interconnected carbon network, which effectively reinforces the composite [29]. Consequently, these results validate that the proposed spray-drying assisted salt-templating strategy is a robust and facile approach for fabricating porous spherical particles. Specifically, the control sample synthesized without NaCl retains a spherical geometry but exhibits a dimpled morphology (Figure S1a,b). In contrast, the NaCl-incorporated sample maintains a smooth surface texture. This observation confirms the dual role of NaCl as both a pore-forming agent and a structural scaffold that mitigates shrinkage stress during spray-drying process. Energy-dispersive X-ray spectroscopy (EDX) mapping images shown in Figure S1c exhibit a uniform distribution of V_3S_4 nanoparticles.

2.2. TEM, SEM, and XPS Analysis

Figure 2a,b presents transmission electron microscopy (TEM) images of the $p\text{-V}_3\text{S}_4/\text{C}$ MS. These images verify the successful formation of a highly porous internal structure following the removal of the sacrificial NaCl template, consistent with the intended design. Interestingly, the microspheres display a unique core-shell-like structure rather than a simple porous structure. Specifically, carbon shell encapsulates the exterior, while the V_3S_4 is predominantly concentrated within the core. In contrast, the V_3S_4/C MS exhibits a dense interior devoid of pores (Figure 2c). The X-ray diffraction (XRD) pattern (Figure 2d) confirms the formation of a crystalline V_3S_4 phase. As shown in Figure 2e, the high-resolution (HR) TEM image displays a distinct lattice spacing of 0.25 nm, which corresponds to the (111) plane of V_3S_4 [30]. Furthermore, EDX mapping (Figure 2f) demonstrates a uniform distribution of V and S elements without agglomeration, indicating that the carbon matrix homogeneously encapsulates the V_3S_4 . These results suggest that the uniform

encapsulation of V_3S_4 by the carbon matrix not only enhances electrical conductivity but also effectively mitigates structural degradation during the charge/discharge process. To quantitatively characterize the porous structure observed in the SEM and TEM images, N_2 adsorption–desorption isotherms were analyzed (Figure 2g,h). As shown in Figure 2g, the synthesized p- V_3S_4 /C MS exhibit a distinct hysteresis loop in the relative pressure (P/P_0) range of 0.4–0.9, indicating typical mesoporous characteristics. The corresponding pore size distribution curves (Figure 2h) reveal that the p- V_3S_4 /C MS possess a broader pore distribution in the 1–100 nm range compared to the V_3S_4 /C MS. This implies a hierarchical pore structure, likely resulting from the selective etching of the NaCl template. However, the V_3S_4 /C MS exhibited a negligible specific surface area due to the lack of pore development. Specifically, the surface area of the p- V_3S_4 /C MS was determined to be $85.1 \text{ m}^2 \text{ g}^{-1}$, which is substantially higher than that of the V_3S_4 /C MS ($0.8753 \text{ m}^2 \text{ g}^{-1}$). This well-developed hierarchical pore structure is expected to enhance electrode stability by serving a dual function: (1) providing rapid diffusion pathways for potassium ions, and (2) acting as a structural buffer to effectively accommodate the volume expansion of V_3S_4 during cycling. To determine the carbon content, TGA was conducted in an air atmosphere. Based on the TGA result, the carbon content in the p- V_3S_4 /C MS composite is calculated to be 46.7% (Figure S2). The observed weight loss corresponds to carbon combustion and the oxidation of V_3S_4 into V_2O_5 . Typical amorphous carbon provides a limited direct capacity contribution to K-ion storage [31]. However, this uniform carbon matrix effectively buffers volume expansion and provides highly conductive pathways, which collectively lead to the superior electrochemical performance of the composite. Furthermore, X-ray photoelectron spectroscopy (XPS) was employed to elucidate the chemical bonding states of the constituent elements within the p- V_3S_4 /C MS (Figure 2i–k). The survey spectrum (Figure S3) confirms the presence of V, S, C, and O. The HR V 2p spectrum can be deconvoluted into peak assigned to V^{2+} , V^{3+} , and V^{4+} . This distribution indicates that vanadium in V_3S_4 exists in an average oxidation state of approximately +2.7, demonstrating that the V^{5+} in the precursor was successfully reduced to form a mixed-valence state [32]. Notably, the direct sulfidation of NH_4VO_3 is thermodynamically sluggish due to the high dissociation energy of its strong V–O bonds. In this respect, the intimate and homogeneous mixture of NH_4VO_3 and dextrin achieved via the spray-drying process creates an ideal localized environment for carbothermal reduction during subsequent calcination [33]. The uniformly distributed carbon acts as an effective reducing agent to cleave the robust V–O bonds, thereby accelerating the complete phase transformation into highly crystalline V_3S_4 . To experimentally verify this, a control sample synthesized without dextrin was subjected to the identical heat treatment. As shown in Figure S4, this sample exhibited prominent peaks corresponding to V_2O_3 , confirming that the sulfidation reaction was severely incomplete. However, a relatively high intensity of the V^{4+} peak was detected. This result is attributed to inevitable surface oxidation, given the high surface sensitivity of the XPS technique. This interpretation is further corroborated by the presence of SO_x observed near 168 eV in the S 2p spectrum (Figure 2j) [34]. In the S 2p spectrum, the distinct doublet observed in the range of 161–163 eV corresponds to the characteristic $2p_{3/2}$ and $2p_{1/2}$ orbitals of metal-sulfide bonds, confirming the successful formation of V–S bonds within the V_3S_4 phase [35]. Notably, an additional peak is identified in the binding energy region of 164–165 eV, which is typically attributed to C–S bonds [36]. This suggests that sulfur atoms were doped into the carbon lattice during the thermal conversion of the dextrin into amorphous carbon [37]. The presence of these C–S bonds is further corroborated by the C 1s spectrum (Figure 2k), where a corresponding peak is observed at approximately 285.7 eV. Considering the surface-sensitive nature of the XPS technique, the presence of the C–S bond indicates that sulfur is successfully incorporated, at least into the surface and

near-surface regions of the carbon matrix. This surface-level heteroatom doping is highly beneficial, as it can induce extrinsic defects and provide abundant active sites, facilitating surface-driven electrochemical reactions. In the C 1s spectrum, the prominent peak centered at approximately 284.6 eV is assigned to sp^2 -hybridized C–C bonds, indicating the partially graphitic nature of the carbon matrix [38]. This graphitic network plays a crucial role in enhancing the overall electrical conductivity of the electrode. The peak observed near 288.5 eV is ascribed to oxygen-containing functional groups, such as C=O bonds [38]. This likely originates from residual groups that survive the carbonization of the dextrin or species formed during the surface activation process. In summary, these results substantiate the successful synthesis of porous microspheres, featuring V_3S_4 nanoparticles homogeneously embedded within a conductive carbon framework via the spray-drying process.

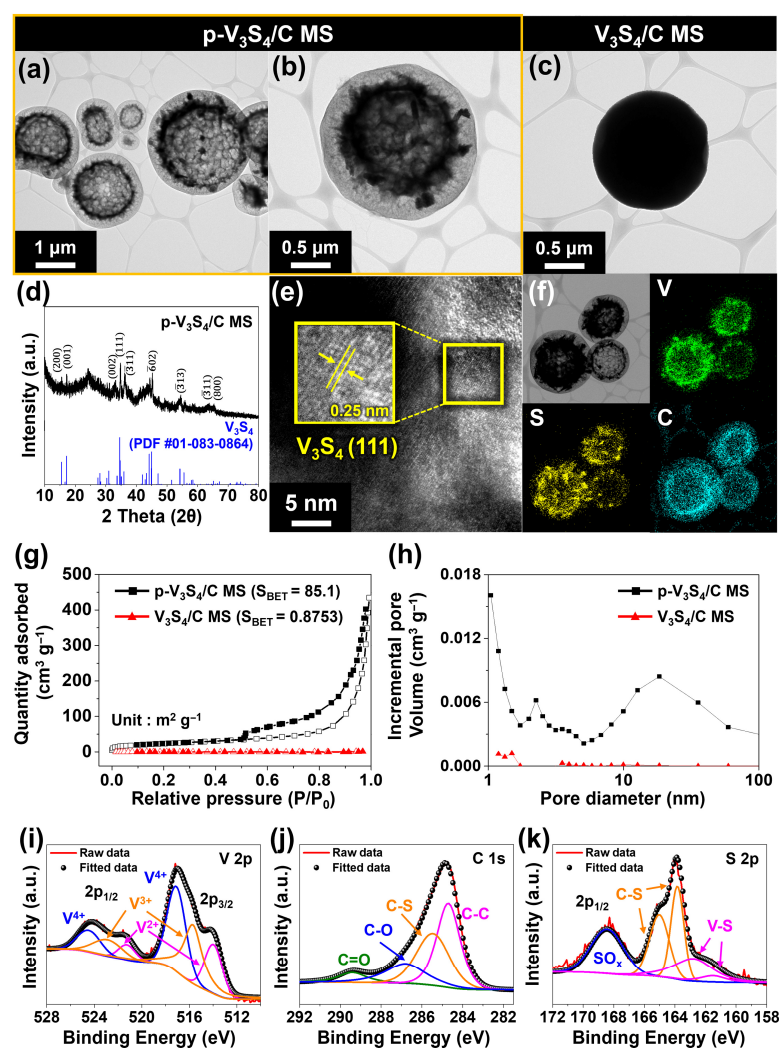


Figure 2. Morphological and structural characterization. (a,b) TEM images of p- V_3S_4 /C MS and (c) V_3S_4 /C MS. (d) XRD pattern, (e) HR TEM image, and (f) EDX mapping of p- V_3S_4 /C MS. (g) N_2 adsorption/desorption isotherm and (h) corresponding pore size distribution. HR XPS spectra of p- V_3S_4 /C MS for (i) V 2p, (j) C 1s, and (k) S 2p.

2.3. Electrochemical Performance and Resistance Evolution Analysis

To investigate the electrochemical potassium storage behavior of the p- V_3S_4 /C MS electrode, cyclic voltammetry (CV) was conducted (Figure 3a). Especially, unlike in lithium- and sodium-ion batteries, distinct redox peaks were observed in KIBs [39]. This behavior is attributed to the large ionic radius of K^+ , which renders the intercalation/deintercalation

mechanism kinetically and thermodynamically more favorable than the conversion reaction. Furthermore, the hierarchical porous structure and abundant defects in the carbon matrix induce a highly pseudocapacitive charge storage behavior. This specific reaction mechanism, combined with the dominant surface-driven pseudocapacitance, aligns perfectly with the sloping voltage profiles (i.e., the absence of distinct plateaus) observed in the galvanostatic charge–discharge (GCD) curves (Figure 3b) [39]. Additionally, the broad peak observed between 1.5 and 0.01 V in the initial cathodic scan disappears in subsequent cycles, which is ascribed to irreversible reactions such as the formation of a solid electrolyte interphase (SEI) layer [40]. Analogous electrochemical behavior was observed for the control V_3S_4/C MS electrode (Figure S5). For both electrodes, the CV curves in subsequent cycles are nearly identical and well overlapped. These results indicate the formation of a stable SEI layer, which effectively suppresses further parasitic side reactions and ensures highly reversible electrochemical reactions during cycling. To substantiate the electrochemical potassium storage property observed in the CV analysis, the rate capability was evaluated at various current densities (Figure 3c). The V_3S_4/C MS electrode exhibited rapid capacity decay as the current density increased. In particular, at a high current density of 10 A g^{-1} , negligible capacity was delivered, which is attributed to sluggish K^+ diffusion kinetics resulting from its dense structure. In contrast, the p- V_3S_4/C MS demonstrated superior capacity retention compared to the V_3S_4/C MS across all current densities. This improvement is ascribed to the porous microsphere structure, which facilitates electrolyte infiltration and significantly shortens the K^+ diffusion distance. Simultaneously, the conductive carbon scaffold enhances electrical connectivity between active materials, enabling rapid charge transfer [41]. When the current density returned to 0.5 A g^{-1} , the capacities of both electrodes recovered to their initial levels. This indicates that both electrodes operate via the aforementioned intercalation/deintercalation mechanism and possess structural stability. To further verify electrode stability, cycling performance was evaluated (Figure 3d). Both electrodes underwent an initial stabilization phase at a low current density of 0.1 A g^{-1} for 5 cycles, followed by cycling at 0.5 A g^{-1} . A gradual increase in capacity was observed for both electrodes as cycling progressed. This phenomenon is attributed to the electrochemical activation process commonly reported for transition metal sulfide anodes [42]. After 600 cycles, the p- V_3S_4/C MS delivered a high capacity of 230 mA h g^{-1} , whereas the V_3S_4/C MS showed a much lower capacity of 141 mA h g^{-1} . This pronounced capacity discrepancy results from the enhanced K^+ kinetics enabled by the porous structure of the p- V_3S_4/C MS. To rigorously validate the superior kinetics and structural stability of the p- V_3S_4/C MS electrode, long-term cycling performance was assessed at a high current density of 2.0 A g^{-1} (Figure 3e). The p- V_3S_4/C MS electrode delivered an initial capacity of 148 mA h g^{-1} and retained 107 mA h g^{-1} even after 2000 cycles. In sharp contrast, the V_3S_4/C MS electrode exhibited rapid capacity decay, retaining only 38 mA h g^{-1} after 1000 cycles. The inferior performance of the V_3S_4/C MS is attributed to its dense structure, which hinders K^+ diffusion and consequently hampers the reversible intercalation/deintercalation process [39]. The electrochemical performance of the p- V_3S_4/C MS electrode was systematically compared with previously reported V_3S_4 -based and other representative metal sulfide KIB anodes (Table S1). The comparison clearly indicates that the p- V_3S_4/C MS composite delivers a highly competitive capacity and stable long-term cycle life, demonstrating its solid potential as a promising anode material for KIBs.

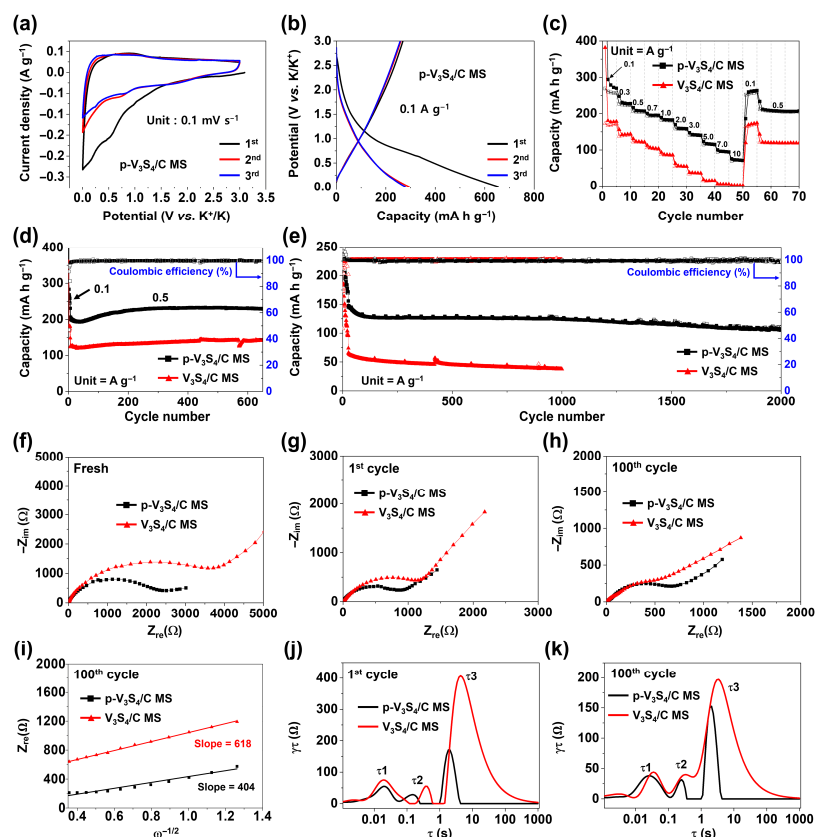


Figure 3. (a) CV curves at 0.1 mV s⁻¹ and (b) GCD profiles at 0.1 A g⁻¹. (c) Rate capability at various current densities from 0.1 to 10 A g⁻¹. (d) Cycling stability at 0.5 A g⁻¹ and (e) long-term stability at 2.0 A g⁻¹. Nyquist plots at (f) fresh, (g) 1st cycle, and (h) 100th cycle. (i) angular frequency calculation at 100 cycles. DRT analysis after (j) 1st cycle and (k) 100th cycle.

To investigate the evolution of internal electrode resistance during cycling, electrochemical impedance spectroscopy (EIS) measurements were performed. The specific resistance values obtained from the EIS measurements were extracted by fitting the data to a Randles equivalent circuit model (Figure S6), and these values are summarized in Table S2. Figure 3f–h displays the Nyquist plots obtained at the pristine state (before cycling) and after the 1st and 100th cycles, respectively. The Nyquist plots typically consist of semicircles in the high-to-medium frequency regions and a sloping line in the low-frequency region, which correspond to the solid electrolyte interphase (SEI) layer resistance (R_{SEI}), charge transfer resistance (R_{ct}), and Warburg impedance related to ion diffusion, respectively. For all electrodes, the diameter of the semicircles progressively decreases upon cycling, indicating a continuous reduction in both R_{SEI} and R_{ct} [43]. Notably, a drastic impedance change is observed between the pristine state and the first cycle, which is primarily attributed to the formation of the SEI layer. Although the formation of this SEI layer consumes initial irreversible capacity, it facilitates enhanced ionic conductivity at the electrode–electrolyte interface during subsequent cycles [44]. This progressive decrease in interfacial resistance minimizes the overpotential during charge/discharge processes, thereby enabling excellent high-rate capability [45]. Furthermore, the ion diffusion kinetics corresponding to the Warburg impedance were evaluated through angular frequency analysis (Figure 3i). The results reveal that the p-V₃S₄/C MS exhibits a lower slope compared to the V₃S₄/C MS, indicating superior ion diffusion kinetics. This suggests that the highly porous architecture facilitates deep electrolyte penetration, effectively reducing the ion diffusion resistance at the electrode–electrolyte interface [46]. To elucidate the origins of this resistance reduction, the distribution of relaxation times (DRT) analysis was conducted

based on the Nyquist plots (Figure 3j,k). In the DRT profiles, the distinct peaks denoted as τ_1 , τ_2 , and τ_3 correspond to the R_{SEI} , R_{ct} , and ion diffusion resistance, respectively [47]. Figure 3j,k present the DRT profiles of the p- V_3S_4 /C MS and V_3S_4 /C MS electrodes after the 1st and 100th cycles, respectively. After the initial cycle, the p- V_3S_4 /C MS electrode exhibits lower intensities for all τ peaks compared to the control V_3S_4 /C MS (Figure 3j). Notably, the τ_3 peak exhibits a pronounced difference after cycling. This demonstrates that the large specific surface area and hierarchical pore network of the p- V_3S_4 /C MS facilitate rapid electrolyte infiltration, thereby accelerating the ion diffusion kinetics at the electrode–electrolyte interface [39]. This finding is highly consistent with the angular frequency analysis presented in Figure 3i. A similar trend in resistance reduction is also observed in the DRT profiles after 100 cycles (Figure 3k). In conclusion, the DRT analysis corroborates that the hierarchical porous architecture of the p- V_3S_4 /C MS effectively mitigates all internal resistances upon cycling. In particular, the substantially reduced ion diffusion resistance, as opposed to the dense V_3S_4 /C MS, underpins its excellent structural stability and superior electrochemical performance even at high current densities.

2.4. Electrochemical Kinetics and Pseudocapacitive Contribution Analysis

To gain deeper insights into the enhanced charge transfer and kinetics enabled by the hierarchical porous architecture of the p- V_3S_4 /C MS, CV-based capacitive contribution analysis was performed (Figure 4). First, CV measurements were conducted at various scan rates ranging from 0.1 to 2.0 mV s^{-1} to distinguish between pseudocapacitive and diffusion-controlled processes (Figure 4a,d). According to the power-law relationship, $i = av^b$, a b-value approaching 1 indicates a pseudocapacitive-dominated behavior, whereas a value close to 0.5 signifies diffusion-controlled kinetics [48]. The b-values for the anodic and cathodic peaks of the p- V_3S_4 /C MS electrode were determined to be 0.97 and 0.98, respectively (Figure 4b), suggesting a predominantly pseudocapacitive mechanism. Furthermore, the control V_3S_4 /C MS electrode also exhibited b-values of 0.82 and 0.90 (Figure 4e), demonstrating that pseudocapacitance governs the charge storage in both electrodes. The capacitive contribution was quantified using the equation $i = k_1v + k_2v^{1/2}$, where k_1v and $k_2v^{1/2}$ represent the pseudocapacitive and diffusion-controlled current contributions, respectively [49]. The p- V_3S_4 /C MS electrode exhibited a substantially higher capacitive contribution than the V_3S_4 /C MS electrode across all scan rates, reaching an overwhelming 93% at a high scan rate of 2.0 mV s^{-1} (Figure 4c). This significantly surpasses the value of the control sample (87%, Figure 4f), demonstrating that the p- V_3S_4 /C MS facilitates rapid, resistance-free charge transfer even under high-rate conditions. This superior pseudocapacitive behavior is attributed to the unique structural features engineered via the spray-drying process and NaCl salt templating. First, the hierarchical porous architecture plays a pivotal role in maximizing the electrolyte-accessible electroactive surface area, thereby promoting surface Faradaic reactions. Second, as previously reported, the abundant extrinsic defects and disordered structures introduced within the S-doped carbon matrix act as additional active sites that facilitate the rapid adsorption and desorption of K^+ ions, further accelerating the surface reaction kinetics [50].

2.5. Potassium Ion Full Cell Configuration

The viability of the p- V_3S_4 /C MS anode for practical applications was investigated by constructing a potassium-ion full cell coupled with a pre-activated Prussian blue (PB) cathode (denoted as p- V_3S_4 /C MS//PB) (Figure 5). By analyzing the galvanostatic charge/discharge behaviors of both individual electrodes, the working voltage range was established at 1.5–4.0 V (Figure 5a). Moreover, the negative-to-positive (N/P) ratio was precisely adjusted to 1.2 based on the equation $N/P = (C_{anode} \times m_{anode}) / (C_{cathode} \times m_{cathode})$

to ensure optimal electrochemical synergy. Accordingly, the active material mass loadings were strictly controlled at 1.2 mg cm^{-2} for the anode and 3.3 mg cm^{-2} for the cathode. The assembled full cell also displayed exceptional rate performance, delivering reversible capacities of 56, 51, 44, 35, 26, 22, and 11 mA h g^{-1} under varying current densities of 0.05, 0.1, 0.2, 0.4, 0.8, 1, and 2 A g^{-1} , respectively (Figure 5b). Excluding the initial irreversibility, the p-V₃S₄/C MS//PB full cell exhibited nearly identical galvanostatic charge–discharge (GCD) profiles over the initial three cycles, demonstrating its high electrochemical reversibility (Figure 5c). Under extended cycling conditions, the p-V₃S₄/C MS//PB demonstrated robust reversibility (Figure 5d), a capacity of 40 mA h g^{-1} was maintained after 100 cycles at 0.2 A g^{-1} , accompanied by a Coulombic efficiency of nearly 100% based on the anode loading mass. These results reflect highly compatible redox kinetics between the anode and cathode. Ultimately, this full-cell evaluation serves as a promising proof-of-concept, demonstrating the preliminary validation of the p-V₃S₄/C MS anode's compatibility with the PB cathode for potential future applications.

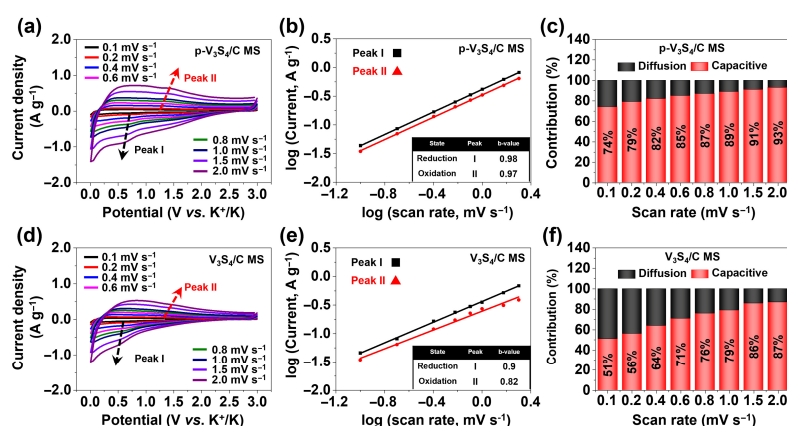


Figure 4. CV curves at various scan rates of (a) p-V₃S₄/C MS and (d) V₃S₄/C MS. b-value determination for redox peak of (b) p-V₃S₄/C MS and (e) V₃S₄/C MS. Capacitive contribution of (c) p-V₃S₄/C MS and (f) V₃S₄/C MS.

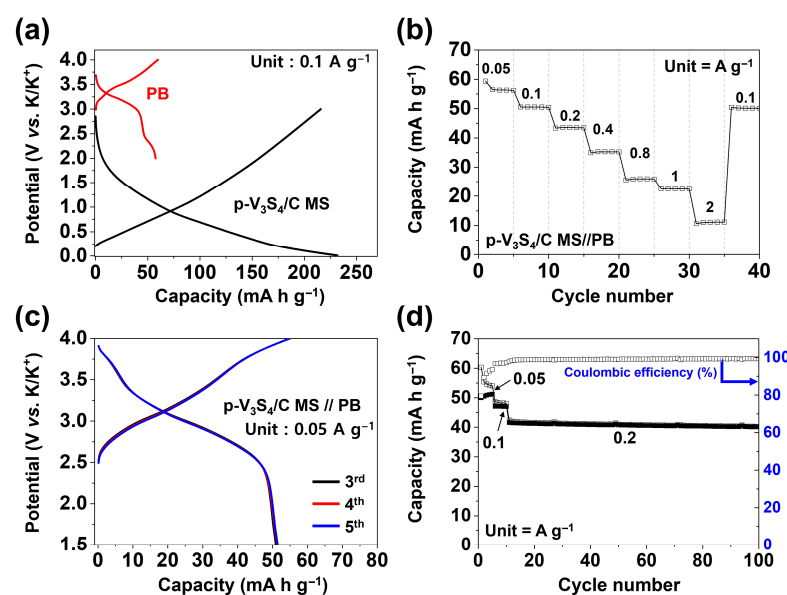


Figure 5. (a) GCD profiles of p-V₃S₄/C MS and PB cathode in half cells at 0.1 A g^{-1} . (b) Rate performance of p-V₃S₄/C MS//PB full cell. (c) GCD profiles of p-V₃S₄/C MS//PB full cell at 0.05 A g^{-1} . (d) Cycling performance of p-V₃S₄/C MS//PB full cell at 0.2 A g^{-1} .

3. Materials and Methods

3.1. Synthesis of *p*-V₃S₄/C MS

The porous V₃S₄/C microspheres (*p*-V₃S₄/C MS) were fabricated using a facile salt-templating strategy combined with a spray-drying technique. The precursor solution was prepared by dissolving 0.1 M NH₄VO₃ (ThermoFisher Scientific, Waltham, MA, USA) as the vanadium source, 20 g L^{−1} dextrin ((C₆H₁₀O₅)_n, Samchun, Republic of Korea) as the carbon source, and 26.5 g L^{−1} sodium chloride (NaCl, Junsei, Tokyo, Japan) as the sacrificial template in deionized water. The homogeneous solution was then atomized using a spray dryer. During this process, the fluid nozzle pressure was maintained at 2.0 bar, with an outlet temperature controlled at 100 °C. The obtained precursor powders were subjected to a synchronous carbonization and sulfidation process at 700 °C for 3 h with a heating rate of 5 °C min^{−1} under an Ar atmosphere. To develop the porous architecture, the calcined microspheres were stirred in deionized water for 1 h to completely dissolve the NaCl template, followed by filtration and drying. For comparison, solid V₃S₄/C microspheres (V₃S₄/C MS) were synthesized via an identical procedure, excluding the addition of NaCl and the subsequent selective etching step.

3.2. Characterizations

The morphology and microstructure of the prepared samples were examined employing field-emission scanning electron microscopy (FE-SEM, Hitachi S-5000, Hitachi Ltd., Tokyo, Japan) and field-emission transmission electron microscopy (FE-TEM, JEM-2100F, Korea Basic Institute Busan Center, Busan, Republic of Korea). To identify the crystalline phases, X-ray diffraction (XRD) measurements were conducted on an Rigaku SmartLab (Tokyo, Japan) utilizing Cu-K α radiation (wavelength = 1.5418 Å). Surface chemical compositions and electronic states were analyzed via X-ray photoelectron spectroscopy (XPS, K-Alpha+, ThermoFisher Scientific, Waltham, MA, USA, Center for Research Facilities, Chung-Ang University, Seoul, Republic of Korea) equipped with a monochromated Al K α source. Furthermore, nitrogen adsorption–desorption isotherms were obtained to calculate the specific surface area and pore size distribution based on the Brunauer–Emmett–Teller (BET) theory.

3.3. Electrochemical Measurements

To investigate the electrochemical properties of the synthesized *p*-V₃S₄/C MS, CR2032-type coin cells were assembled. The working electrodes were fabricated by mixing the active material, Super P (conductive agent), and carboxymethyl cellulose (CMC, binder) in deionized water at a mass ratio of 7:2:1. The resulting slurry was uniformly cast onto a copper foil current collector. The mass loading of the active material on the current collector was strictly controlled at approximately 1.2 mg cm^{−2}. To ensure the reliability and statistical reproducibility of the electrochemical data, at least 3 independent cells were assembled and tested for each condition. The variation in cycling capacity and rate capability among different cells was within approximately 3–5%, confirming good reproducibility of the electrochemical performance. Coin cell fabrication was conducted within an argon-filled glove box, employing potassium metal as the counter/reference electrode and a glass fiber filter as the separator. The electrolyte utilized was 3 M potassium bis(fluorosulfonyl)imide (KFSI) dissolved in a mixture of ethylene carbonate (EC) and diethyl carbonate (DEC). Electrochemical data, including galvanostatic charge–discharge profiles and cyclic voltammetry (CV) curves, were recorded using a battery testing system (WBCS3000L, WonATech, Seoul, Republic of Korea) within a potential window of 0.01–3.0 V (vs. K/K⁺). Furthermore, impedance spectra were acquired via electrochemical impedance spectroscopy (EIS) in the frequency region between 100 kHz and 0.1 Hz.

4. Conclusions

In summary, hierarchical porous p-V₃S₄/C microspheres (p-V₃S₄/C MS) were successfully designed and synthesized via a facile, scalable spray-drying strategy coupled with NaCl salt-templating. The structural design effectively addresses the inherent challenges of transition metal sulfide anodes for KIBs, namely, substantial volume expansion and sluggish K⁺ kinetics. The selective removal of the NaCl template creates a highly developed porous architecture that facilitates rapid electrolyte penetration and shortens the ion diffusion pathways. Concurrently, the robust and highly conductive amorphous carbon matrix serves as a mechanical buffer, preserving the structural integrity of the V₃S₄ nanoparticles during repeated potassiation/depotassiation processes. Detailed kinetic investigations, including DRT and capacitive contribution analyses, corroborated that this unique architecture significantly mitigates internal ion diffusion resistance and promotes surface-driven pseudocapacitive charge storage (reaching 93% at 2.0 mV s^{−1}). Consequently, the p-V₃S₄/C MS electrode exhibited overwhelming long-term cycling stability (retaining 107 mA h g^{−1} after 2000 cycles at 2.0 A g^{−1}) and superior rate capabilities compared to the dense control sample. Furthermore, the preliminary practical viability of the composite was validated through the successful operation of a p-V₃S₄/C MS//PB full cell, which demonstrated robust reversibility and promising energy storage performance as a proof-of-concept for future applications. Collectively, this work not only presents a highly competitive anode material for KIBs but also provides a cost-effective and scalable structural engineering pathway for advanced energy storage systems.

Supplementary Materials: The following supporting information can be downloaded at <https://www.mdpi.com/article/10.3390/batteries12030096/s1>. Figure S1: (a,b) SEM images, (c) EDX mapping images of V₃S₄/C MS; Figure S2: TGA curves of p-V₃S₄/C MS in air condition; Figure S3: XPS survey spectrum of p-V₃S₄/C MS; Figure S4: XRD patterns of NH₄VO₃ after the sulfidation process without dextrin; Figure S5: GCD profiles of V₃S₄/C MS at 0.1 A g^{−1}; Figure S6: Typical Randles-type equivalent circuit model used for the EIS fitting.; Table S1: Comparison of electrochemical performance for V_xS_y-based anode materials in KIBs; Table S2: Calculated resistance values of the electrodes after 1st and 100 cycles. References [51,52] are cited in the Supplementary Materials.

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