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Architecting a Robust Yarn-Like CNT Framework for Structurally Resilient ZnSe Anodes in Potassium–Ion Batteries

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ABSTRACT

The development of structurally stable anodes with high capacities and long cycle lives is essential for the development of potassium–ion batteries. Conversion-type anodes have attracted considerable attention in potassium–ion batteries (PIBs) due to their high theoretical capacities. However, their practical application is hindered by severe volume variation, sluggish reaction kinetics, and structural degradation during repeated potassiation and depotassiation. Herein, we report a novel yarn-like microsphere (Y-CNT/ZnSe@NC) architecture that integrates a nanoconfined active material with a conductive flexible framework. This structure was fabricated through a scalable spray-drying process followed by carbonization and selenization, enabling the uniform encapsulation of N-doped carbon-coated ZnSe nanoparticles within a three-dimensional network of carbon nanotubes (CNTs). The resulting microspheres exhibited a compact yet porous architecture that enhanced ion transport and provided a continuous electron-conductive network. The conformal CNT framework effectively mitigated the volume changes of ZnSe during cycling, suppressed nanocrystal overgrowth during heat treatment, and maintained structural integrity. Consequently, the Y-CNT/ZnSe@NC anode exhibited superior electrochemical performance, delivering a reversible capacity of 220 mA h g^{−1} with a Coulombic efficiency of ~99% over 2000 cycles and excellent rate capability. In contrast, CNT-free electrodes suffer from capacity fading owing to structural degradation. In-depth analyses further confirmed the mechanical robustness and stable charge-transfer characteristics of the CNT-embedded structure. This study highlights the advantages of uniformly encapsulating active materials within a CNT network and provides a versatile platform for designing next-generation high-energy-density long-life anodes based on transition-metal chalcogenides.

1 | Introduction

Transition-metal selenides (TMSes) are considered promising anodes for potassium–ion batteries (PIBs) owing to their high theoretical capacity, cost-effectiveness, [1–6] and superior electronic conductivity compared with those of oxides and sulfides [7]. These features, along with the higher ionic conductivity of

their discharge products, contribute to enhanced reaction kinetics [1]. Despite these attractive features, TMSes still face significant challenges, including low intrinsic conductivity, substantial volume expansion, and particle agglomeration during repeated charge/discharge cycles, which result in compromised cycling stability and rate performance [8]. Accordingly, structural regulation has emerged as a key strategy to suppress

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framework degradation and alleviate volume fluctuation during cycling. Pre-intercalation approaches have demonstrated the effectiveness of lattice modulation in enhancing stability in PIB [9]. However, conversion-type TMSes undergo repeated bond-breaking and reconstruction reactions, requiring alternative stabilization strategies.

Recently, metal-organic frameworks (MOFs) have attracted considerable attention as precursors for TMSes [10, 11]. Under controlled pyrolysis in an inert atmosphere, the metal ions and organic ligands within MOFs can be converted into various transition-metal compounds and carbon matrices, respectively [12–15]. In this context, integrating TMSes with carbon matrices through MOF templates can enhance the electrical conductivity and mitigate volume changes, thereby improving the cycling stability. Moreover, compared with pure carbon, carbon derived from imidazolate ligands benefits from inherent nitrogen doping, which introduces defects and active sites, which further boost the electrochemical performance [8]. However, MOF-derived TMSes often suffer from low interparticle charge transport [16] and insufficient structural stability to accommodate the large ionic radius of K^+ [17].

To address these challenges, various conductive matrices have been employed to form composites with MOF-derived transition-metal compounds [18–21]. Two-dimensional (2D) materials, including MXene and graphene, have attracted considerable attention as composite matrices because of their superior conductivity and large surface area [22, 23]. However, these 2D materials tend to be restacked without proper surface modification, which significantly reduces ion accessibility [24, 25]. Furthermore, although their layered structures are potentially favorable for ion transport, insufficient interlayer spacing can limit ion diffusion and negatively affect their electrochemical performance [26]. In contrast, carbon nanotubes (CNTs) with their one-dimensional and high-aspect-ratio morphology naturally form entangled networks that create abundant pores and interstitial spaces. This facilitates ion transport, accommodates volume expansion [27], and promotes the formation of a highly interconnected conductive network that enables efficient electron transport [28]. However, traditional approaches that simply mix CNTs with active materials often result in severe aggregation and low bulk density, preventing the formation of a uniform conductive network and thereby limiting the energy density [29, 30]. These limitations necessitate the development of a novel strategy for the homogeneous integration of CNTs with active materials. By assembling CNTs into a three-dimensional (3D) interconnected architecture that envelops the active material, not only can a uniform dispersion of CNTs be achieved, but adverse physical changes, such as drastic volume changes, of the active material can also be significantly mitigated. In such a configuration, the enveloping CNT network serves as a flexible yet robust framework that buffers mechanical stress during repeated cycling, ensuring that the active material retains its structural robustness [31, 32]. Consequently, this innovative architecture not only enhances the electron transport efficiency but also stabilizes the electrode structure, leading to improved energy density and long-term cycling stability [33, 34].

In this work, we propose a novel strategy for fabricating spherical yarn-like CNT-based microspheres through a spray-drying

process for use as high-performance anodes for PIBs. Specifically, our approach involves spray drying, followed by sequential carbonization and selenization, resulting in a 3D architecture in which ultrathin CNTs uniformly envelop MOF-derived ZnSe nanoparticles coated with N-doped carbon (Y-CNT/ZnSe@NC), forming a compact, yarn-like spherical structure. This well-wrapped configuration ensures the homogeneous dispersion of both CNTs and active particles while providing a mechanically robust framework that effectively buffers the substantial volume changes of ZnSe during repeated charge/discharge cycles. The CNT network facilitates efficient electron transport and relieves mechanical stress, thereby preserving the structural integrity of the active material. Electrochemical analyses revealed that the Y-CNT/ZnSe@NC anode achieved long-term stability, delivering 225 mA h g^{-1} after 2000 cycles at 0.5 A g^{-1} with nearly 100% Coulombic efficiency. Even at 3.0 A g^{-1} , the anode retained a capacity of 148 mA h g^{-1} , indicating an excellent rate performance. Based on these findings, this paper presents an integrated design approach that combines 3D structural engineering, uniform CNT encapsulation, and scalable spray-drying fabrication to overcome the key limitations of ZnSe-based PIB anodes. The proposed design strategy provides a versatile and scalable platform that can be applied to various MOF-derived transition-metal chalcogenides for advanced potassium-ion energy storage.

2 | Experimental

2.1 | Preparation of Acid-Treated Multiwall CNT

240 mL of H_2SO_4 (Samchun, 95%) and 80 mL of HNO_3 (Samchun, 60%) were mixed and heated to 85°C . Then, 6 g of multiwall CNT was dispersed into the solution and mildly stirred for 6 h. After acid treatment, the mixture was washed several times with DI water.

2.2 | Preparation of Y-CNT/ZnSe@NC

First, ZIF-8 nanoparticles were synthesized following the procedure reported in the literature [35]. Then, ZIF-8 particles (0.9 g) and acid-treated CNTs (0.9 g) were dispersed in deionized water, and the suspension was diluted to a final volume of 300 mL. Subsequently, the resulting suspension was spray-dried at an inlet temperature of 200°C and an outlet temperature of 100°C , using a flow rate of 5.4 mL min^{-1} . The resulting Y-CNT/ZIF-8 was then heated at 700°C (5°C min^{-1}) for 3 h in an Ar atmosphere to carbonize ZIF-8 particles. Subsequently, the Y-CNT/ZnSe@NC was obtained by heating the carbonized product with selenium powder at 300°C (5°C min^{-1}) for 12 h in an Ar/H_2 (95:5 v/v%).

2.3 | Preparation of ZnSe@NC

For the synthesis of ZnSe@NC, the as-prepared ZIF-8 nanoparticles were subjected to the same carbonization and subsequent selenization processes as described for Y-CNT/ZnSe@NC, without the incorporation of CNT. Specifically, the ZIF-8 particles were first carbonized at 700°C (5°C min^{-1}) for 3 h under

an Ar atmosphere, and then selenized with selenium powder at 300°C (5°C min⁻¹) for 3 h in a H₂/Ar (95:5 v/v%) mixed atmosphere to yield ZnSe@NC.

3 | Results and Discussion

Figure 1A shows a schematic of the synthesis process. First, polyhedral ZIF-8 precursors with an average diameter of ~100 nm were prepared using a simple room-temperature precipitation method [35], and their X-ray diffraction (XRD)

patterns matched well with the simulated patterns (Supporting Information S1: Figure S1). Subsequently, Y-CNT/ZIF-8 microspheres were synthesized through spray drying of a well-dispersed CNT/ZIF-8 suspension.

To clarify the role of the CNT/ZIF-8 mass ratio in determining the morphology and structural characteristics of Y-CNT/ZIF-8 microspheres, samples with various ratios were synthesized through spray drying (Figure 2). Even without external additives, the CNTs were naturally interconnected in a 3D manner, forming nearly spherical microspheres (Figure 2A). This is because the droplets containing CNTs rapidly evaporated upon

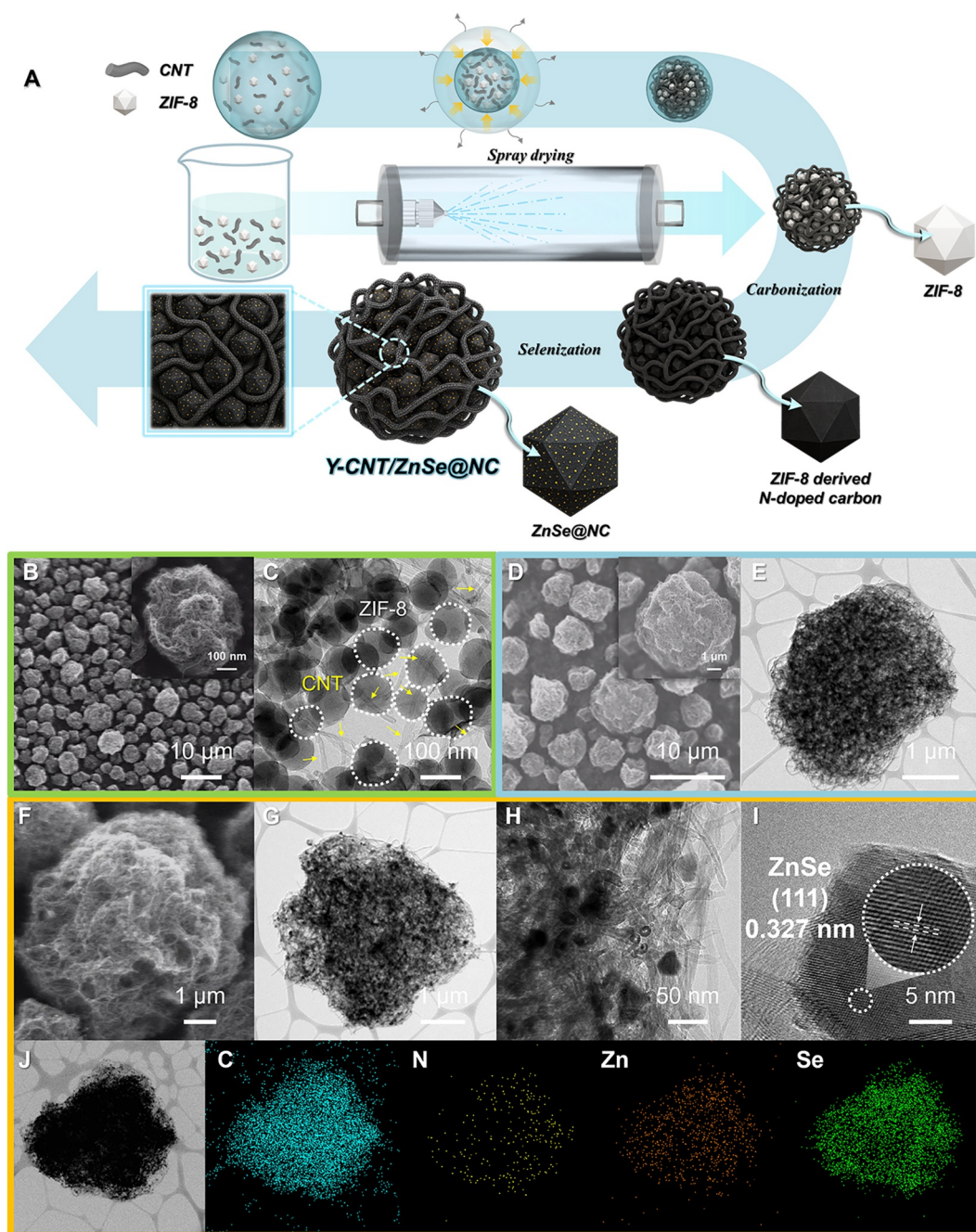


FIGURE 1 | Synthesis and structural characterization of Y-CNT/ZnSe@NC microspheres. (A) Schematic illustration of the preparation for Y-CNT/ZnSe@NC. SEM and TEM images of Y-CNT/ZIF-8 (B, C) before and (D, E) after carbonization. (F) SEM image, (G, H) TEM images, (I) HR-TEM image, and (J) elemental mapping of Y-CNT/ZnSe@NC.

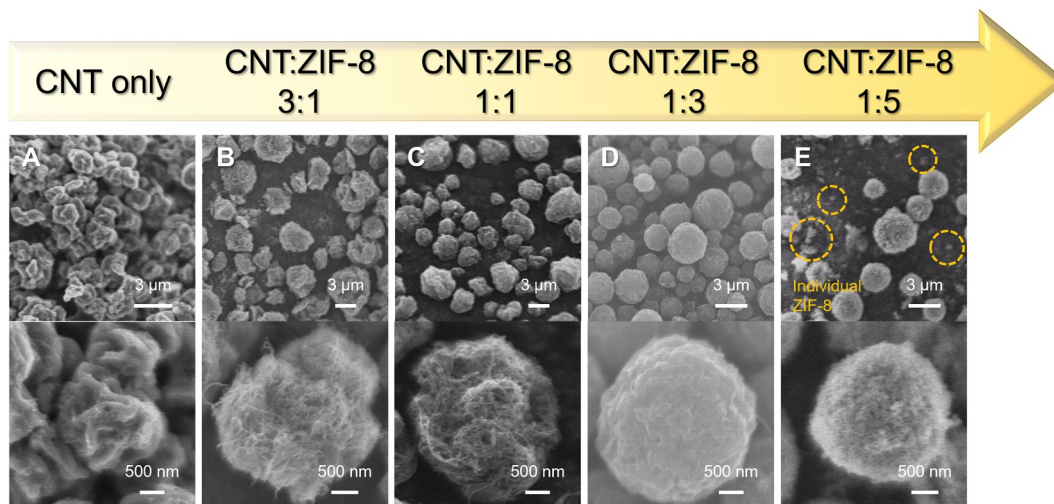


FIGURE 2 | SEM images of CNT/ZIF-8 composites with different mass ratios. (A) CNT only, (B) 3:1, (C) 1:1, (D) 1:3, and (E) 1:5.

exposure to hot air during the spray drying process, causing the CNTs to entangle with one another. However, in the absence of ZIF-8 nanoparticles, the CNT microspheres exhibited a crumpled morphology, which can be attributed to the uneven shrinkage caused by the internal voids resulting from entanglement. Interestingly, as the weight ratio of the ZIF-8 nanoparticles to CNTs increased, the resulting microspheres gradually transformed into a more perfect spherical morphology (Figure 2B–D). With increasing incorporation of ZIF-8 nanoparticles, more particles occupied the internal pores formed within the CNT-based microspheres, thereby preventing significant deformation during the rapid drying process. When the CNT-to-ZIF-8 mass ratio reached 1:5, individual ZIF-8 nanoparticles were observed because of excess ZIF-8 that was not encapsulated in the CNT microspheres (Figure 2E). As discussed later, the CNT-to-ZIF-8 mass ratio of 1:1 demonstrated the best electrochemical performance; therefore, subsequent analyses were conducted on samples with a ratio of 1:1.

Figure 1B shows the scanning electron microscopy (SEM) and transmission electron microscopy (TEM) images of the Y-CNT/ZIF-8 (1:1) microspheres. The microspheres exhibit a spherical morphology with a diameter of approximately 3 μm , and the CNTs are strongly entangled. Unlike the original one-dimensional CNT structure, this spherical morphology facilitates uniform slurry casting and high packing density in the electrode [36, 37]. TEM image (Figure 1C) reveals that the ZIF-8 nanoparticles were interconnected through the CNT network. Notably, the ZIF-8 structure remained unchanged after spray drying. The conductive CNT network not only connects the nanoparticles to provide favorable electron transport pathways but also creates internal voids between the nanoparticles [38], effectively accommodating volume changes during charge/discharge cycles. Even after carbonization, which converts the N-containing organic linkers within ZIF-8 into N-doped carbon, the overall particle morphology remained largely unchanged and well preserved (Figure 1D,E).

The Y-CNT/ZnSe@NC microspheres were obtained following the selenization process. The Y-CNT/ZnSe@NC microspheres retained their spherical shape with no significant overall changes;

however, small ZnSe particles formed during selenization were observed both on the surfaces and within the microsphere (Figure 1F,G). Detailed TEM analysis (Figure 1H) confirmed that ZnSe nanocrystals are uniformly distributed within the 3D CNT framework, with an average size of approximately 30 nm. Despite undergoing two high-temperature treatments (carbonization and selenization), ZnSe remained in the form of small nanocrystals, indicating that the crystal growth was effectively suppressed. This limited crystal growth was attributed to the spatial confinement of the Zn-based nanoparticles within the N-doped carbon matrix, which originated from organic linkers during the reaction. Additionally, the XRD analysis of the Y-CNT/ZnSe@NC sample revealed cubic-phase ZnSe and graphitic carbon peaks, confirming the coexistence of ZnSe nanocrystals and CNT in the composite (Supporting Information S1: Figure S2). As observed in the high-resolution transmission electron microscopy (HR-TEM) image (Figure 1I), lattice fringes with a spacing of 0.18 nm are assigned to the (311) plane of the cubic ZnSe phase. Elemental mapping of C, N, Zn, and Se (Figure 1J) confirmed the uniform distribution of CNTs, N-doped carbon, and ZnSe nanocrystals.

For comparison, ZnSe@N-doped carbon (ZnSe@NC) nanoparticles were obtained by employing only ZIF-8 as the precursor, followed by carbonization and selenization. After the two-step heat treatment, the characteristic polyhedral shape of ZIF-8 was retained, but the particles shrank to a size of approximately 70 nm due to the decomposition of the organic ligands (Supporting Information S1: Figure S3A). Notably, the XRD pattern of the ZnSe@NC sample displayed significantly sharper peaks corresponding to cubic ZnSe than those of Y-CNT/ZnSe@NC, indicating that the ZnSe crystals in ZnSe@NC underwent more pronounced growth (Supporting Information S1: Figure S2) and this excessive crystal growth is further corroborated by the TEM observations (Supporting Information S1: Figure S3B). This suggests that, in addition to the N-doped carbon matrix, the introduction of CNTs was crucial for inhibiting the overgrowth of ZnSe crystals during selenization by providing spatial confinement and structural restrictions. Energy-dispersive X-ray spectroscopy (EDX) mapping further confirmed the presence of C, N, Zn, and Se in the ZnSe@NC nanoparticles (Supporting Information S1: Figure S3C).

Figure 3A shows the N_2 adsorption–desorption isotherms and pore size distributions of the Y-CNT/ZnSe@NC microspheres and ZnSe@NC. Although pristine ZIF-8 usually has a specific surface area of more than $1000 \text{ m}^2 \text{ g}^{-1}$ because of its unique microporous structure, ZIF-8-derived ZnSe@NC has a relatively small specific surface area of $74 \text{ m}^2 \text{ g}^{-1}$ because of the presence of ZnSe nanocrystals with a very small surface area, which occupy some pores of ZIF-8-derived carbon, thereby partially blocking the pores [39, 40]. Despite the presence of ZnSe nanoparticles in both samples, the specific surface area of Y-CNT/ZnSe@NC microspheres was 45% larger than that of ZnSe@NC, reaching $107 \text{ m}^2 \text{ g}^{-1}$. This increase can be attributed to the inter pore structure formed by the interconnected CNTs as well as the smaller ZnSe particle size within the Y-CNT/ZnSe@NC microspheres, as confirmed by XRD and TEM analyses.

The carbon structures of Y-CNT/ZnSe@NC microspheres and ZnSe@NC nanoparticles were analyzed using Raman spectroscopy (Figure 3B). Two distinct peaks appeared at ~ 1320 and $\sim 1570 \text{ cm}^{-1}$, which can be assigned to the defect-related D band and the crystalline graphite G band, respectively. Intensity ratio (I_D/I_G) between the D and G band was employed to evaluate the defect density and degree of crystallinity. Notably, the I_D/I_G value for Y-CNT/ZnSe@NC (1.05) was lower than that for ZnSe@NC (1.31), indicating that the CNT network exhibited higher crystallinity, which facilitated efficient electron transfer.

X-ray photoelectron spectroscopy (XPS) was carried out to explore the surface characteristics of the Y-CNT/ZnSe@NC microspheres. The high-resolution C 1s spectrum (Figure 3C) was fitted into three components assigned to C=C bonds (284.03 eV), C–C bonds (284.53 eV), and C–N bonds (285.48 eV) [41, 42]. The pronounced C=C peak corresponds to sp^2 bonding. As the carbon derived from the ZIF precursor is formed at a

relatively moderate carbonization temperature, it predominantly consists of disordered carbon. Therefore, the observed C=C feature mainly originates from the intrinsically graphitic CNT framework, which retains its ordered sp^2 structure within the composite. In the high-resolution N 1s spectrum (Figure 3D), the three peaks at 398.2 , 399.9 , and 401.6 eV are assigned to pyridinic N, pyrrolic N, and graphitic N, respectively [43, 44]. These nitrogen species serve as active sites for K^+ ion interactions and enhance conductivity, thereby contributing to increased capacity and rapid charge/discharge performance [45–47]. The high-resolution Zn 2p spectrum (Figure 3E) displayed two characteristic peaks at 1021.6 eV (Zn $2p_{3/2}$) and 1044.6 eV (Zn $2p_{1/2}$), showing an energy difference of 23 eV . This confirms that Zn exists in the Zn^{2+} oxidation state [48, 49], which is consistent with the previously reported values for Zn in ZnSe compounds [50]. Additionally, the high-resolution Se 3d spectrum (Figure 3F) revealed two peaks at 55.9 eV (Se $3d_{5/2}$) and 55.7 eV (Se $3d_{3/2}$), indicating the presence of Se^{2-} in ZnSe, along with an additional peak at 58.2 eV ascribed to SeO_2 from partial oxidation at the surface.

To better understand the K^+ storage behavior of the Y-CNT/ZnSe@NC electrode, its electrochemical kinetics and K^+ diffusivity were systematically investigated (Figure 4). In Figure 4A, the cyclic voltammetry (CV) curves of the Y-CNT/ZnSe@NC microspheres at 0.1 mV s^{-1} were obtained, which presented the characteristic peaks of both ZnSe@NC and CNT, as displayed in Supporting Information S1: Figure S4. During the initial discharge, a broad peak was observed within the potential range of 1.3 – 0.5 V , corresponding to the reduction of ZnSe and the irreversible formation of the solid-electrolyte interphase (SEI). [51]. Additionally, considering CV curve of the CNT electrode shown in Supporting Information S1: Figure S4B, the appearance of a sharp peak positioned around 0.01 V and the following anodic peak corresponds to K^+ ion insertion/extraction into the CNT,

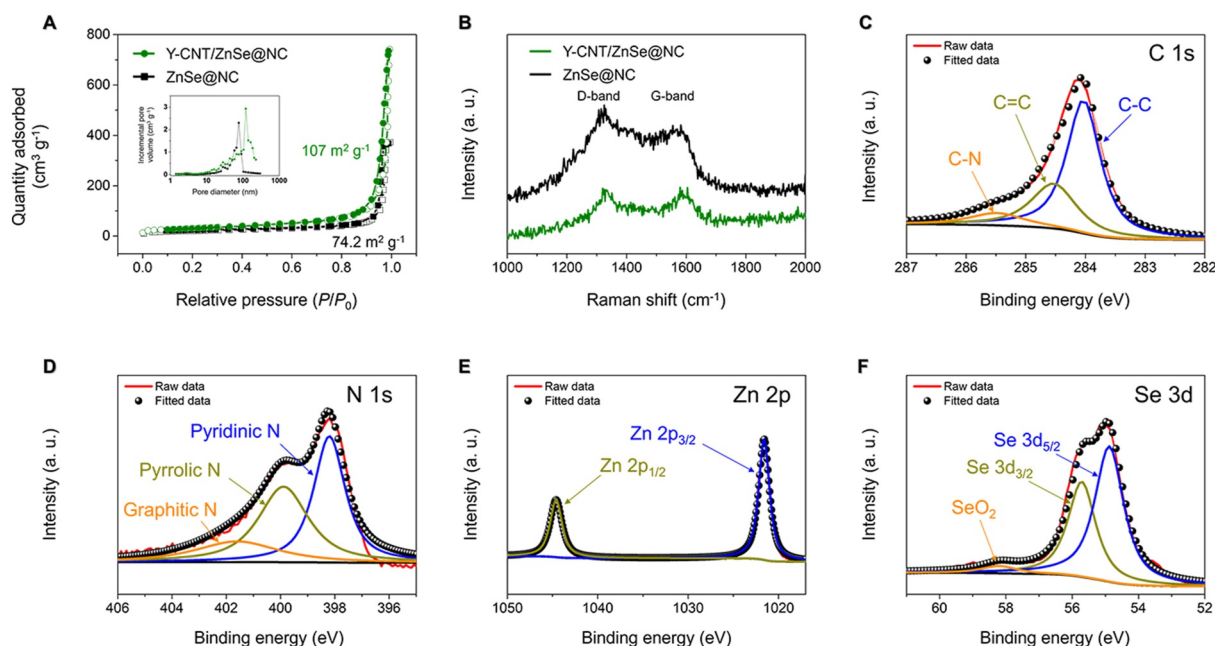


FIGURE 3 | Structural and surface chemical characterization of Y-CNT/ZnSe@NC and ZnSe@NC. (A) N_2 adsorption/desorption isotherms & BJH pore size distributions and (B) Raman spectra of Y-CNT/ZnSe@NC and ZnSe@NC. High resolution XPS spectra of Y-CNT/ZnSe@NC: (C) C 1s, (D) N 1s, (E) Zn 2p, and (F) Se 3d.

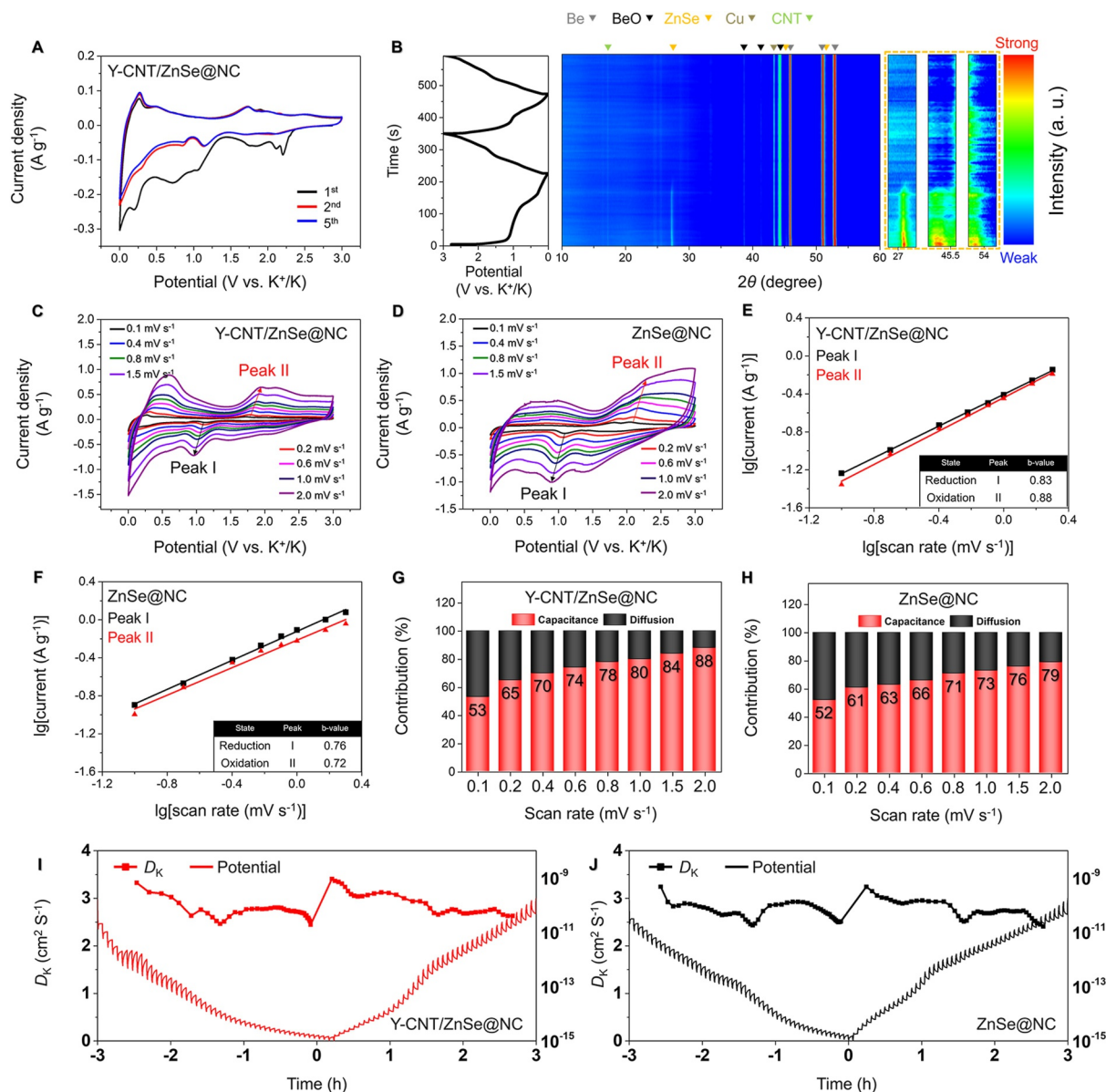


FIGURE 4 | Electrochemical kinetics and K^+ storage behavior. (A) Cyclic voltammetry curves and (B) GCD curve (left) with the corresponding in situ XRD contour map (right) of Y-CNT/ZnSe@NC. (C, D) Cyclic voltammetry curves at various scan rates. (E, F) Linear relationships between logarithm current and logarithm scan rate of Peak I and II. (G, H) Capacitive contribution under various scan rates. (I, J) GITT profiles and calculated K^+ diffusion coefficients.

which is consistent with a previous report [52]. In the subsequent anodic process, the broad peak observed at 1.5–2.0 V is related to the formation of ZnSe [12]. It is worth noting that CV curves from subsequent cycles overlapped well, indicating the excellent reversibility of the Y-CNT/ZnSe@NC electrode.

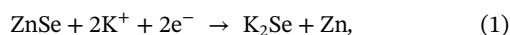
In addition, in situ XRD was performed to monitor the structural evolution of ZnSe during potassiation and depotassiation. As shown in Figure 4B, although the diffraction peak of CNT remains visible throughout the consecutive charge–discharge cycles, multiple characteristic diffraction planes of cubic ZnSe, including the (111), (220), and (311) planes, progressively weaken and eventually disappear during the discharge process. Notably, these diffraction peaks do not fully recover upon the subsequent charge process, suggesting that the re-formed ZnSe phase does not return to its original crystallographic state. This

behavior may be attributed to a combination of factors, including the formation of ultrafine ZnSe domains below the XRD detection limit (consistent with HR-TEM observation in Supporting Information S1: Figure S5A), as well as increased structural disorder, such as reduced long-range crystallinity, potential amorphization, or incomplete phase reconversion during cycling. Moreover, no discernible peak shift or continuous lattice expansion of the ZnSe planes is observed prior to their disappearance, suggesting that ZnSe does not undergo a K^+ intercalation process accompanied by lattice parameter changes, but instead evolves through a conversion reaction. During the discharge process, additional diffraction features corresponding to K_2Se are clearly observed in the in situ XRD patterns (Supporting Information S1: Figure S6), providing direct evidence for the electrochemical reduction of ZnSe and confirming the occurrence of a conversion reaction. This

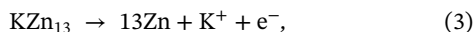
observation is further supported by HR-TEM analysis (Supporting Information S1: Figure S5B), which directly confirms the presence of the K_2Se phase after discharge. Although no distinct diffraction planes of KZn_{13} can be resolved in the in situ XRD patterns due to overlap with strong Cu-related peaks from the current collector (Supporting Information S1: Figure S6), the presence of the KZn_{13} phase after deep discharge is confirmed by HR-TEM analysis (Supporting Information S1: Figure S5B). Collectively, these results demonstrate that ZnSe undergoes a reversible conversion reaction accompanied by nanoscale structural reconstruction during cycling. The formation of ultrafine ZnSe nanocrystals, together with the CNT network, increases the electrochemically active surface area and alleviates the mechanical stress associated with repeated conversion reactions, thereby contributing to the enhanced structural stability and prolonged cycling performance of the Y-CNT/ZnSe@NC electrode.

Based on the above in situ XRD and ex situ TEM analysis at representative discharge and charge states, and the relevant previous studies [53–55], the K^+ storage behavior of the Y-CNT/ZnSe@NC electrode can be described as follows:

Discharge reaction:



Charge reaction:



To probe the reaction kinetics, CV measurements were conducted on Y-CNT/ZnSe@NC and ZnSe@NC electrodes using scan rates in the range of 0.1–2.0 mV s^{−1} (Figure 4C,D). As the scan rate increased, both electrodes exhibited peak shifts due to polarization [56]; however, the Y-CNT/ZnSe@NC electrode showed more moderate changes in cathodic and anodic peak potentials compared with ZnSe@NC (Supporting Information S1: Figure S7A,B). Consistently, the Y-CNT/ZnSe@NC electrode exhibits a smaller interpeak voltage difference over the entire scan-rate range (Supporting Information S1: Figure S7C), quantitatively confirming reduced polarization and faster reaction kinetics. This improved kinetic behavior can be attributed to the structural features of the Y-CNT/ZnSe@NC microspheres. The interconnected CNT framework bridges ZnSe nanocrystals to form continuous electron transport pathways, enhancing electronic conductivity and facilitating efficient charge transfer during the conversion reaction. In addition, the CNT-induced suppression of ZnSe crystal growth produces smaller nanocrystals, further contributing to reduced polarization and improved electrochemical kinetics [57].

To further elucidate the intrinsic reaction kinetics of the electrodes, the relationship between the peak current (i) and scan rate (v) was analyzed using the following equation: $i = av^b$, where the b -value reflects the relative contribution of capacitive-controlled and diffusion-controlled processes [58]. As shown in Figure 4E,F,

the Y-CNT/ZnSe@NC electrode exhibits b -values of 0.83 for the cathodic peak and 0.88 for the anodic peak, which are consistently higher than those obtained for the ZnSe@NC electrode (0.76 and 0.72, respectively). In general, a b -value approaching 0.5 corresponds to a diffusion-limited process, whereas a value close to 1.0 is characteristic of capacitive-controlled charge storage [59]. Accordingly, the higher b -values observed for the Y-CNT/ZnSe@NC electrode indicate that its electrochemical reactions are more strongly governed by capacitive-controlled processes compared with the ZnSe@NC electrode. Such enhanced capacitive characteristics can be attributed to the interconnected three-dimensional CNT network, which facilitates rapid electron transport, improves charge-transfer kinetics, and provides abundant electrochemically accessible interfaces.

To quantitatively evaluate the capacitive contribution inferred from the b -value analysis, the proportion of charge storage originating from capacitive-controlled processes was calculated based on the CV curves using a well-known power series equation [60, 61]. As illustrated in Figure 4G,H, the Y-CNT/ZnSe@NC electrode consistently delivers a higher capacitive contribution than the ZnSe@NC electrode over the entire range of scan rates. This behavior can be rationalized by the synergistic structural characteristics of the composite. The integration of ZIF-derived carbon with the interconnected CNT framework provides a relatively high specific surface area and abundant electrochemically accessible interfaces. Meanwhile, the CNT network effectively suppresses excessive ZnSe growth, resulting in ultrafine ZnSe nanocrystals that are uniformly distributed within the conductive matrix. The combined effects of enlarged interfacial area and reduced particle size increase the number of surface-accessible redox sites and significantly shorten ion diffusion pathways. Consequently, the increased capacitive contribution can be understood as reflecting enhanced surface-controlled kinetic features arising from the enlarged electrochemically accessible interface and shortened ion diffusion pathways in the Y-CNT/ZnSe@NC electrode. Such a pseudocapacitive-dominated kinetic behavior underlies the excellent rate capability of the Y-CNT/ZnSe@NC electrode and highlights the critical role of the three-dimensional CNT network in interconnecting ZnSe nanocrystals, thereby enabling rapid charge/discharge kinetics without altering the intrinsic bulk storage mechanism.

The galvanostatic intermittent titration technique (GITT) was applied to determine the diffusivity of K^+ , and the calculated diffusion coefficient of K^+ (D_{K^+}) is shown in Figure 4I,J. The detailed equation for the calculation is given in Supporting Information S1: Figure S8. During the potassiation process, the ion diffusion coefficient gradually decreased with an increasing degree of potassiation, which can be attributed to the continuous accumulation of K^+ ions on the exposed active surface sites, leading to elongated diffusion pathways for the subsequently inserted ions [62]. However, a slight increase in D_{K^+} was observed at ~1.3 V during the potassiation process, which corresponds to the onset of the ZnSe conversion reaction, as shown in the CV curve. This increase is attributed to the phase transition of ZnSe into Zn and K_2Se , which generated new diffusion pathways [55]. The diffusion coefficients in Y-ZnSe@NC were calculated to be approximately 4.01×10^{-11} to 9.26×10^{-10} cm² s^{−1}, which are higher than those of ZnSe@NC (1.64×10^{-11} to 4.73×10^{-10} cm² s^{−1}). This is because the abundant inner space

within the CNT network facilitates rapid ion transport, whereas the ultrafine ZnSe nanocrystals offer an enlarged contact area with the electrolyte and reduce the ion diffusion pathway, thereby enhancing the ion insertion efficiency.

To evaluate K^+ storage performance, half-cells of Y-ZnSe@NC, ZnSe@NC, and CNT were assembled with potassium metal as the counter electrode. The initial Coulombic efficiencies of the Y-CNT/ZnSe@NC, ZnSe@NC, and CNT electrodes were measured to be 71.97%, 65.39%, and 25.87%, respectively (Supporting Information S1: Figure S9). Interestingly, this superior ICE was achieved despite the larger BET specific surface area of Y-CNT/ZnSe@NC ($107 \text{ m}^2 \text{ g}^{-1}$) relative to ZnSe@NC ($74.2 \text{ m}^2 \text{ g}^{-1}$). Generally, a higher surface area is expected to lower the ICE due to increased contact with the electrolyte and subsequent decomposition. The reversed trend observed here suggests that the 3D interconnected CNT framework plays a critical role in regulating the SEI formation. By providing a highly conductive network, the Y-CNT architecture facilitates a uniform electric field and K-ion flux at the interface, which prevents localized overpotentials and suppresses excessive parasitic reactions. This results in the formation of a more efficient and stable SEI layer even with an increased electrochemically active surface. Although direct chemical characterization of the SEI remains a subject for future investigation, these electrochemical and structural correlations underscore the beneficial role of the yarn-like CNT architecture in maintaining interfacial integrity. After performing five activation cycles at 0.1 A g^{-1} , the galvanostatic charge–discharge (GCD) curves of all electrodes were recorded (Figure 5A). Compared with ZnSe@NC, the Y-ZnSe@NC electrode exhibited more pronounced voltage plateaus, indicating a more efficient conversion reaction of ZnSe owing to the CNT network. In contrast, the CNT electrode exhibited no distinct voltage plateaus, indicating the absence of conversion reactions with K^+ ions.

The cycling stabilities of the three electrodes were tested, and the corresponding results are depicted in Figure 5B. The Y-CNT/ZnSe@NC electrode exhibited superior long-term cycling stability and reversibility, delivering a discharge capacity of approximately 220 mA h g^{-1} with a Coulombic efficiency close to 99% after 2000 cycles without noticeable performance degradation. In contrast, the ZnSe@NC electrode showed a very high discharge capacity of 364 mA h g^{-1} , but the capacity decayed rapidly after 40 cycles and decreased to 88 mA h g^{-1} after 2000 cycles. This pronounced difference in cycling behavior clearly highlights the superior structural stability of the Y-CNT/ZnSe@NC electrode during prolonged cycling.

Such differences in cycling performance could also be observed from the GCD curves of selected cycles (Figure 5C,D). Although the Y-CNT/ZnSe@NC electrode maintained stable GCD curves in both capacity and voltage plateaus even after 10, 50, 100, 200, 300, 400, 500, and 1000 cycles, the ZnSe@NC electrode suffered from severe capacity fading, increasingly indistinct plateaus, and significant hysteresis with continued cycling. This result can be attributed to severe structural degradation triggered by successive K^+ insertion and extraction, where substantial volume expansion during cycling results in the pulverization of the active material. Additionally, the CNT electrode showed a low discharge capacity of 163 mA h g^{-1} owing to the relatively low

theoretical capacity of carbon materials, but a very stable performance even after 2000 cycles.

To further investigate the structural stability of the electrode during the charge–discharge process, the surface of the electrode was examined in the fresh state and after 1500 cycles using SEM (Supporting Information S1: Figure S10). The fresh ZnSe@NC electrode exhibited a nonuniform surface with noticeable particle agglomeration, which could be attributed to the absence of CNTs. Without the support of the CNT network, the ZnSe@NC particles tend to aggregate more readily, leading to poor dispersion and surface roughness during the slurry coating process. In contrast, the Y-CNT/ZnSe@NC electrode displayed a much smoother and more uniform surface owing to the well-dispersed CNT network, which prevented particle settling and ensured homogeneous distribution. After 1500 cycles, multiple cracks and partial detachment of the active materials were observed in the ZnSe@NC electrode. The high-magnification SEM images further reveal that distinct particle features are difficult to identify after cycling, likely due to the accumulation of binder and SEI layers covering the electrode surface. These structural degradations suggest that the electrode cannot effectively withstand the mechanical stress arising from repeated volume changes during charge–discharge processes. As a result, active material loss and disruption of conductive pathways are likely to contribute to rapid capacity decay. In contrast, the Y-CNT/ZnSe@NC electrode maintained a relatively smooth and intact morphology even after prolonged cycling. The high-magnification SEM images confirm that the spherical morphology of the composite particles is largely preserved after 1500 cycles, without severe cracking or structural collapse. This observation indicates that the interconnected CNT network and the internal porous structure effectively buffer volume expansion and maintain structural coherence. Moreover, the mechanical flexibility and robustness of the CNT framework help sustain continuous conductive pathways throughout cycling. Consequently, the remarkable structural stability of the Y-CNT/ZnSe@NC electrode enables a reversible charge–discharge process and contributes to its outstanding long-term cycling performance.

The influence of CNT/ZIF-8 proportion on the electrochemical behavior of Y-CNT/ZnSe@NC microspheres was investigated by preparing two additional samples with weight ratios of CNT-to-ZIF-8 set at 1:3 and 3:1. Supporting Information S1: Figure S11A compares the electrochemical performances of these electrodes. The results reveal distinct discharge capacities that correlate with varying ZnSe contents, as shown in Supporting Information S1: Figure S11B. Notably, the sample with the CNT:ZIF-8 ratio of 1:3 exhibited the highest discharge capacity. However, as shown in Figure 2E, excessive ZIF-8 resulted in individual ZnSe@NC particles that were not interconnected by the CNT network. These particles undergo uncontrolled volume expansion during cycling, ultimately compromising their structural integrity and resulting in poor cycling stability. In contrast, both the 1:1 and 3:1 (CNT:ZIF-8) samples demonstrated highly stable cycling performances over 2000 cycles, although the 1:1 sample showed a slightly lower discharge capacity owing to its reduced ZnSe content. Overall, these findings indicate that careful tuning of the CNT-to-ZIF-8 ratio is critical for optimizing the electrochemical performance, as it directly affects both the discharge capacity and cycling stability of the electrode.

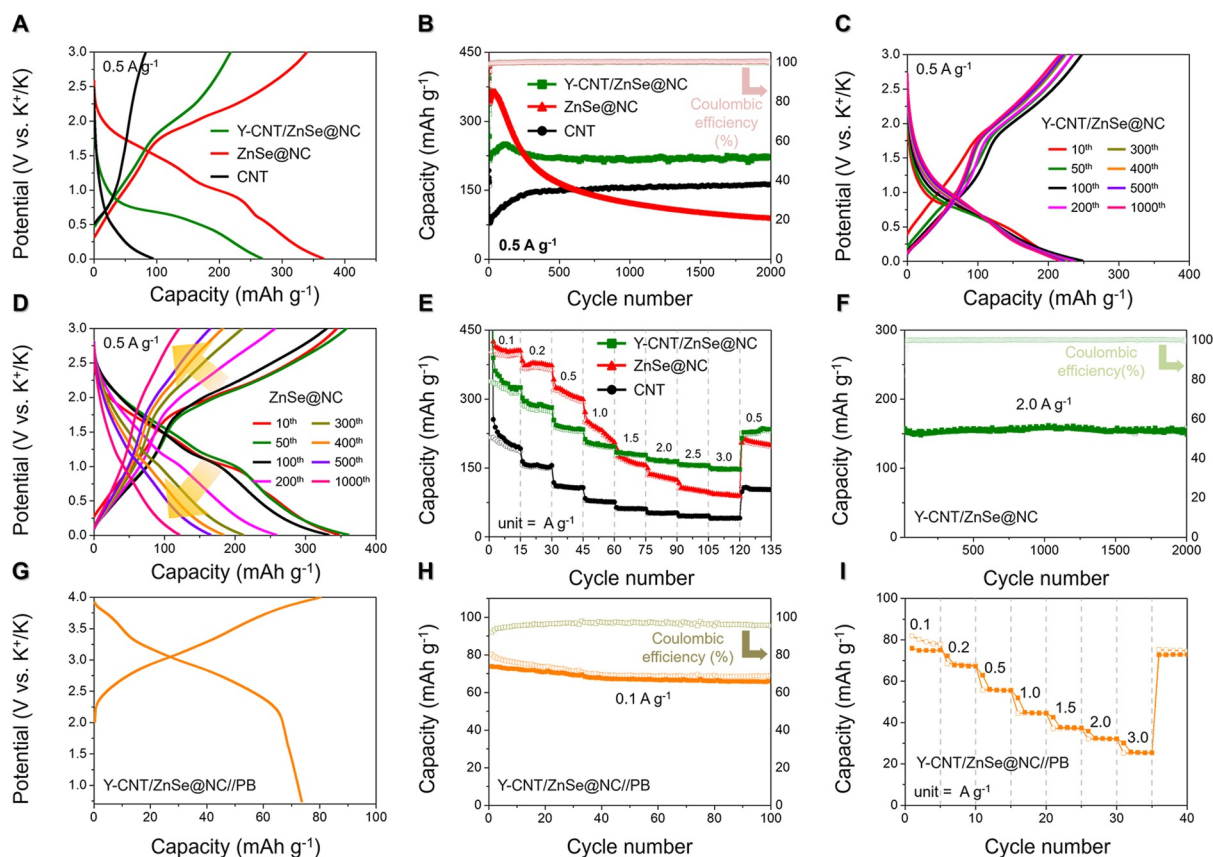


FIGURE 5 | Electrochemical performance of Y-CNT/ZnSe@NC in half-cell and full-cell. (A) GCD curves of Y-CNT/ZnSe@NC, ZnSe@NC, and CNT electrodes at 0.5 A g^{-1} for the 6th cycle. (B) Long-term cycling performance. GCD curves of (C) Y-CNT/ZnSe@NC and (D) ZnSe@NC at the 10th, 50th, 100th, 200th, 300th, 400th, 500th, and 1000th cycles. (E) Rate performance. (F) Long-term cycling performance at 2.0 A g^{-1} of Y-CNT/ZnSe@NC. (G) GCD curve, (H) cycling performance, and (I) rate performance of Y-CNT/ZnSe@NC//Fe-PBA full-cell.

In Figure 5E, the Y-CNT/ZnSe electrode demonstrates a superior rate performance with the high discharge capacities of 325, 289, 234, 197, 178, 163, 155, and 148 mA h g^{-1} at 0.1, 0.2, 0.5, 1.0, 1.5, 2.0, 2.5, and 3.0 A g^{-1} , respectively. Notably, once the current density returned to 0.5 A g^{-1} , the Y-CNT/ZnSe@NC electrode showed an almost fully recovered discharge capacity of 229 A g^{-1} , indicating excellent reversibility even under high-rate conditions. On the contrary, the ZnSe@NC electrode exhibited a delivered discharge capacity under low-rate conditions because of the higher ZnSe content but suffered a pronounced capacity decline as the current density increased, ultimately achieving a substantially reduced discharge capacity of 90 mA h g^{-1} at 3.0 A g^{-1} . The outstanding rate performance of the Y-CNT/ZnSe@NC electrode was mainly ascribed to the desirable conductive pathway of the interconnected CNT network and fast ion transport owing to the internal pores, which was confirmed by the GITT. Inspired by the outstanding cycling stability and rate capability, we further evaluated the long-term cycling performance at 2.0 A g^{-1} (Figure 5F). At a 2.0 A g^{-1} , the Y-CNT/ZnSe@NC electrode delivered a stable discharge capacity of 155 mA h g^{-1} , demonstrating its structural robustness under harsh cycling conditions. These findings highlight the critical role of the structured CNT in the design of high-performance conversion-based PIB anode materials. The Y-CNT/ZnSe@NC electrode exhibited overall electrochemical performance superior to that of Zn-based composite previously reported for PIBs. (Supporting Information S1: Table S1).

To evaluate the practical applicability of the Y-CNT/ZnSe@NC anode, potassium-ion full cells were assembled using a representative Fe-based Prussian blue analog (Fe-PBA) cathode. Prior to full-cell assembly, the electrochemical performance of the Fe-PBA cathode was assessed in a half-cell configuration. As shown in Supporting Information S1: Figure S12, the Fe-PBA cathode exhibits high reversibility, delivering a stable specific capacity of 68 mA h g^{-1} at 0.1 A g^{-1} , confirming its reliable K^+ storage behavior. Based on this stable cathode performance, full cells were constructed by pairing the Fe-PBA cathode with the Y-CNT/ZnSe@NC anode. The full cell delivers an initial discharge capacity of approximately 73.8 mA h g^{-1} and maintains a reversible capacity of 66 mA h g^{-1} with a Coulombic efficiency approaching $\sim 96\%$ during prolonged cycling (Figure 5G,H). The rate capability test further demonstrates that the full cell retains 25.3 mA h g^{-1} at elevated current densities, and the capacity substantially recovers to 72.9 mA h g^{-1} upon returning to the initial current density (Figure 5I). These results indicate that the fast reaction kinetics and structural stability of the Y-CNT/ZnSe@NC anode are preserved in the full-cell configuration, confirming its applicability in practical PIB systems.

To evaluate the change in electrode resistance, ex situ electrochemical impedance spectroscopy (EIS) analysis was performed (Figure 6A–C). Nyquist plots comprising a depressed semicircle and a subsequent sloping line were obtained for different states. The fresh Y-CNT/ZnSe electrode presented a lower charge

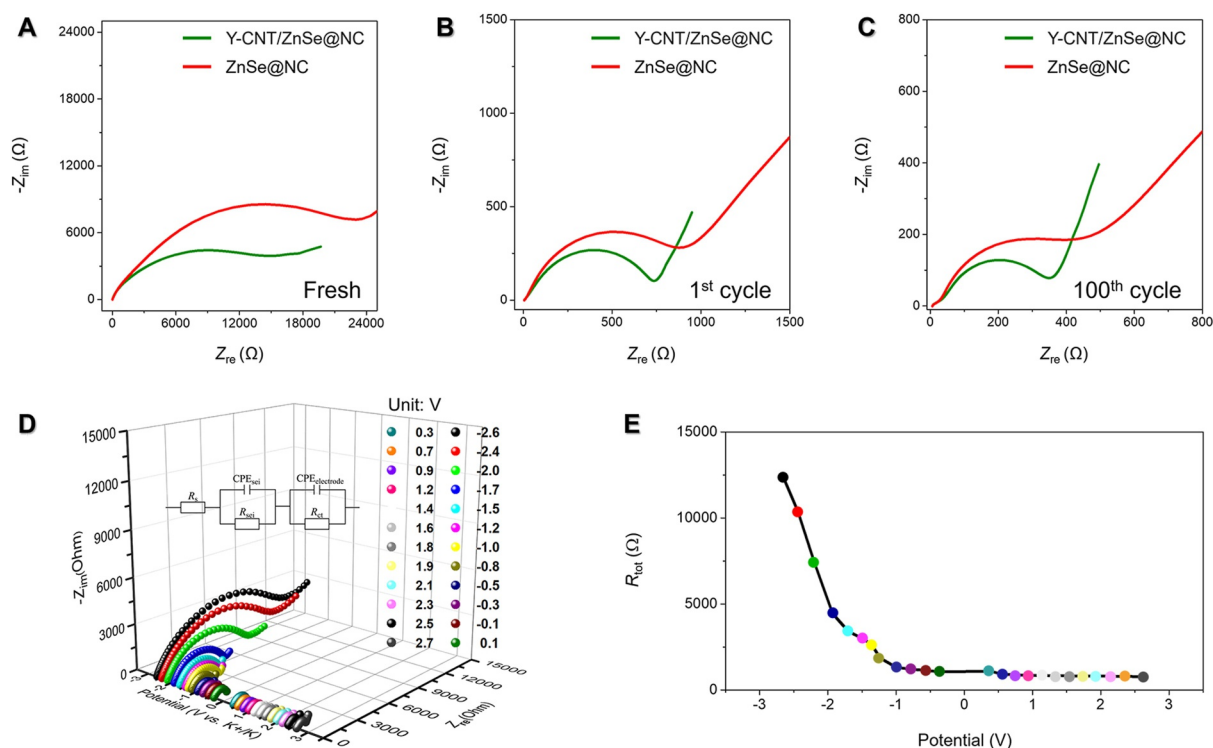


FIGURE 6 | Ex situ and in situ EIS analysis. Nyquist plots of Y-CNT/ZnSe@NC and ZnSe@NC: (A) fresh state, (B) after 1st cycle, and (C) 100th cycle. In situ EIS analysis of Y-CNT/ZnSe electrode: (D) Nyquist plots (inset: Randle-type equivalent circuit model) and (E) variation of total resistance with cell potential.

transfer resistance (R_{ct}) than the ZnSe@NC electrode, suggesting that the CNT network effectively interconnects the ZnSe nanocrystals, forming favorable electron transport pathways. Following the first cycle, the R_{ct} values of both electrodes exhibited a notable decline, with the Y-CNT/ZnSe electrode maintaining a lower R_{ct} value. The significant reduction in the electrode resistance after the first cycle was further investigated using in situ EIS. The measurements were initially performed from a potential of -2.6 V (the minus sign corresponds to the discharge step) within the frequency range of 0.01 – 100 kHz, and corresponding Nyquist plots were subsequently obtained at the prespecified potentials (Figure 6D), which were then fitted by a Randle-type equivalent circuit. In this model, R_s , R_{SEI} , and R_{ct} represent the series resistance, interfacial resistance associated with the SEI layer, and charge-transfer resistance, respectively. Figure 6E depicts the variation in the R_{tot} value, representing the sum of R_s , R_{SEI} , and R_{ct} during the initial cycling process. A sharp decrease in R_{tot} was observed during the discharge process, which can be ascribed to the formation of a stable SEI layer and electrode activation [12, 63]. Y-CNT/ZnSe still showed a lower R_{ct} value than ZnSe@NC even after 100 cycles, which is due to the porous structure of the CNT, which alleviates electrode volume expansion and maintains structural stability.

4 | Conclusions

In summary, we presented a novel strategy for constructing yarn-like CNT/ZnSe@NC microspheres through a scalable spray-drying process, enabling the integration of a robust CNT

network with MOF-derived ZnSe nanoparticles. This unique 3D architecture effectively buffered volume changes, suppressed excessive crystal growth, and enhanced electron/ion transport by providing a continuous conductive network and porous framework. Compared with conventional ZnSe@NC or CNT composites, the yarn-like structure offers superior structural resilience and stable electrochemical behavior, even under extended cycling. The well-interconnected CNT framework plays a critical role in preserving the integrity of the active material, maintaining the conductive pathways, and ensuring long-term performance. This study highlights a promising and scalable design platform for high-performance anodes in potassium-ion batteries that can be extended to other metal chalcogenide systems for advanced energy-storage applications.

Author Contributions

Jeong Ho Na: conceptualization, data curation, formal analysis, investigation, methodology, visualization, writing – original draft. **Hyun Jin Kim:** formal analysis, investigation, methodology, software, visualization, writing – original draft, validation. **Daiha Shin:** formal analysis, investigation, resources, software, visualization, writing – review and editing. **Hong Geun Oh:** investigation, validation, writing – review and editing, conceptualization, methodology, software, data curation. **Yun Jae Lee:** formal analysis, investigation, methodology, visualization, writing – review and editing, validation. **Haeseong Jang:** formal analysis, investigation, resources, visualization, writing – review and editing. **Jiung Cho:** investigation, project administration, resources, supervision, writing – review and editing. **Seung-Keun Park:** conceptualization, funding acquisition, project administration, resources, software, supervision, writing – review and editing.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Supporting Information

Additional supporting information can be found online in the Supporting Information section.

Supporting Information S1: rar270361-sup-0001-suppl-data.docx.