The progress and perspective of electrospun carbon nanofibers based anode materials for potassium ion storage: A mini review

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Potassium ion battery (PIB) is one of the promising substitutes for lithium ion battery. Developing new anode materials is still one of the key factors to promote the electrochemical performances of PIB. Electrospun carbon nanofibers based binder-free electrodes have received widely researches because multi-advantages of good conductivity, large surface areas and structural stability. In this review, the latest research progress in enhancing the potassium ion storage properties based on electrospun carbon nanofibers through introducing porous structure, doping heteroatoms and preparing composites have been summarized. The effects of pore structure, heteroatom types and non-metal/metal loading on specific surface area, conductivity, lattice distance and potassium storage performance of the electrospun carbon nanofiber electrode were emphatically discussed. In addition, several suggestions for industrial application of electrospun carbon nanofiber electrodes for potassium ion storage were proposed in this review.

KEYWORDS

potassium ion storage, anode electrode, electrospun carbon nanofibers, porous structure, heteroatom doping, composites

1 Introduction

As the main power source for portable electronic devices and electric vehicles (EVs), and energy storage systems for wind and solar energy, lithium-ion batteries have achieved great success in commercial applications (Hameer and van Niekerk, 2015; M. Gür, 2018; Zhong et al., 2022). It is supposed that the global demand for energy storage devices will be close to 900 Gwh in 2025. Nevertheless, the global reserves of lithium are scarce and unevenly distributed, resulting in difficulties in meeting the market demand. In addition, the failure to reduce the cost of lithium-ion batteries will seriously affect their long-term development (Gu et al., 2017; L. Kulova et al., 2020; Wang et al., 2022a). Therefore, it is imperative to investigate inexpensive battery systems to resolve the above problems.

Potassium, located in the alkali metals group, possesses analogous physical and chemical properties to lithium. Furthermore, the high crust abundance of potassium (K: 1.5 wt%; Li: 0.0017 wt%) and similar redox potential to lithium (K$^+$/K: −2.93 V vs. SHE; Li$^+$/Li: −3.04 V vs. SHE), making potassium ion battery being one of the potential substitutes for lithium ion battery (Hwang et al., 2018; Zhang et al., 2021; Li et al., 2022).

During the commercialization process of potassium ion battery, several problems still need to be resolved. For instance, the large radius of potassium ion (K$^+$: 1.38 Å; Li$^+$: 0.76 Å) may lead to pulverization of active materials during the repeated potassiation/depotassiation procedure, resulting in poor cycling stability (Zhang et al., 2018a; Deng et al., 2021; Sun et al., 2022a). In addition, the relatively sluggish diffusion kinetics of large K ions in the solid electrode leads to inferior rate performance (An et al., 2018; Liu et al., 2018; Cao et al., 2021a). Therefore, it is significant to select suitable anode material and optimize structure from the microscopic point of view to prompt the commercial application and development of potassium ion batteries.

Among numerous kinds of anode materials, carbon based materials are still considered to be the most promising electrode materials for commercial potassium ion batteries because of their advantages of good chemical stability, high conductivity and environmental friendliness (Bin et al., 2018; Cao et al., 2021b; Hong et al., 2021; Thakur et al., 2022). In particular, carbon nanofibers prepared via an electrospinning technique are regarded as a kind of potential anode materials for potassium ion storage based on the following advantages: 1) the one-dimensional structure of nanofibers can offer a rapid transfer channel for electrons; 2) the unique network architecture is good to infiltration of electrolyte, alleviating the concentration polarization; 3) electrospun carbon nanofibers can be directly used as self-supporting electrode for their high flexibility and excellent tensile strength. In conclusion, the above superiorities of electrospun carbon nanofibers can endow fast kinetics and robust structural integrity of electrodes, improving energy density, power density and cycling stability of battery (Chen et al., 2020; Zheng et al., 2021a; Yuan et al., 2021).

2 Application of electrospun carbon nanofibers in potassium ion battery

2.1 Electrospun carbon nanofibers anode

In recent years, electrospun carbon nanofibers electrode has been verified as a successful anode for potassium ion batteries (Zhang et al., 2020a; Touja et al., 2020). Zhao et al. applied free-standing porous carbon nanofiber paper as anode of potassium ion battery, and the results revealed enormous potential of carbon nanofibers as potassium ion anode electrode

(reversible capacity of 211 mAh g^{-1} after 1,200 cycles at current density of 0.2 A g^{-1} and remain 100 mAh g^{-1} at 7.7 A g^{-1}) (Zhao et al., 2017). Tian et al. (2020) prepared highly graphitized carbon nanofibers (HG-CNFs) through carbonization at 1,400°C after electrospinning and pre-oxidization procedures. When the free-standing electrode was served as the anode for potassium ion batteries, the HG-CNFs electrode exhibited a high reversible capacity (200 mAh g^{-1}) below 0.2 V (Figure 1), which is beneficial to acquiring high energy density. In addition, the electrode also obtained superior rate (226 mAh g^{-1} at 35 C (1 C = 279 mA g^{-1})) and stability performances (less than 0.008% per cycle during 400 cycles) resulted from the freestanding fibrous networks and highly graphitic structure.

In order to further boost the potassium ion storage performances of electrospun carbon nanofibers, several kinds of modification methods on electrospun carbon nanofibers were studied. Particularly, designing porous structure in electrospun carbon nanofibers is an efficient way, which has been demonstrated to provide abundant active sites for the adsorption of K$^+$, and further promoted the capacitive contribution to the potassium-storage capacity beyond the intercalation mechanism (Li et al., 2018; Zheng et al., 2021b; Zhou et al., 2021; Sun et al., 2022b). Furthermore, porous structure can provide sufficient spaces to mitigate large volume change, achieving long cycling stability (Xie et al., 2017; Chen et al., 2018). Zhang et al. (2019) fabricated a free-standing carbon nanofibers anode with hierarchical porous structure (Figures 2Ai–iv) by using polymethyl methacrylate (PMMA) as foaming agent, which was thermal decomposed under 240–280°C. The electrode exhibited great potential as binding-free anode in potassium-based dual-ion batteries, which showed a high reversible capacity of 197 mAh g^{-1} (50 mA g^{-1}) and excellent cycling stability (65 mAh g^{-1} after 346 cycles at a specific current of 100 mA g^{-1}). Chen et al. (2022) further investigated the influence of PMMA content on pore structure and battery performances, demonstrating the porous richness and surface roughness of nanofibers were enhanced with the increase of PMMA and obtaining enhanced potassium ion storage performance (Figure 2B). Wu et al. (2021) synthesized macroporous honeycomb-like carbon nanofibers (MHCNFs) by using poly(vinyl alcohol) (PVA) and poly(tetrafluoroethylene) (PTFE) as precursors, where PTFE mainly played the role in forming pore substance. The binder-free electrode exhibited ultrahigh potassium storage ability and competitive rate capability, maintaining 252.6 mAh g^{-1} over repeating 2,500 cycles at 1,000 mA g^{-1} (Figure 2C). Besides these easily decomposed organic matter as pore-making agent, metal based substance containing ZnCl$_2$, ZIF-8, Sb$_2$S$_3$ and so on were applied as well, and the obtained electrode exhibited good potassium ion storage performances (Kim et al., 2013; Hu et al., 2020; Liu et al., 2022).
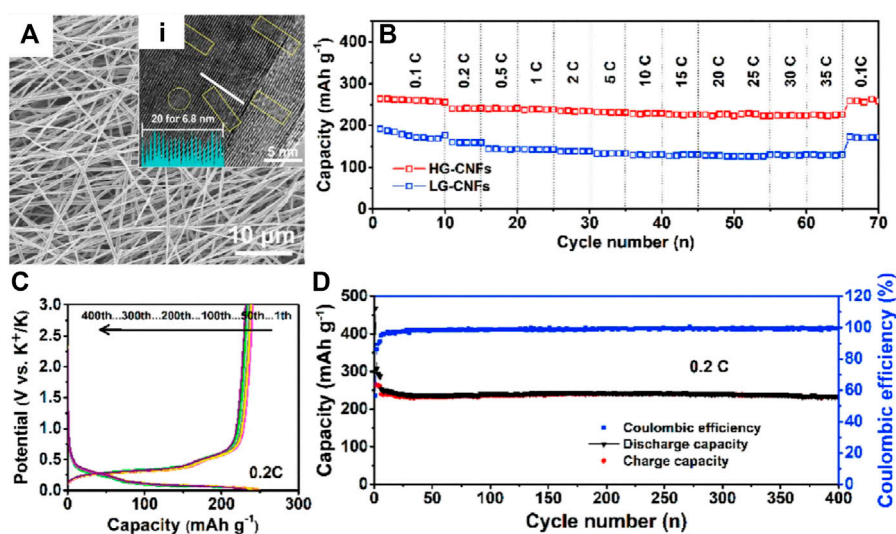
FIGURE 1
(A) SEM and **(i)** HRTEM images of HG-CNFs. **(B)** Comparison of the rate capability from 0.1 to 35 C of lowly and highly graphitized carbon nanofibers (LG- CNFs and HG-CNFs). **(C)** The charge-discharge curves after different cycles at 0.2 C **(D)** Long-term cycle stability and its coulombic efficiency of HG-CNFs at 0.2 C (Tian et al., 2020).

Doping heteroatom (N, O, S, P etc.) in carbon nanofibers is also verified an as effective strategy to boost the potassium ion storage property on the basis of the following reasons: 1) heteroatom doping can enhance the electronic conductivity of carbon nanofibers through tuning the local electronic structure to store charge on the surface of the carbon lattice (Ma et al., 2019); 2) heteroatom doping can provide large numbers of active sites for adsorbing K$^+$ (Tao et al., 2020); 3) heteroatom doping can increase the interlayer distance of carbon nanofibers for their large covalent radius (Wang et al., 2022b).

In particular, the N atom is the most common doped element in electrospun carbon nanofibers because the carbon resource is always derived from PAN, which contains N element. Therefore, the *in-situ* the N doped carbon nanofibers can be easily synthesized during the carbonation process. N-doping has been verified to enhance reactivity by generating more defects and increase conductivity by ameliorating the locally electronic configuration (Xu et al., 2018; Yang et al., 2019; Pei et al., 2020). N-doped carbon nanofibers prepared by Chen et al. (2022) showed higher specific capacity and better rate performance than the non-atom doped carbon nanofibers used PVA as precursor.

On this basis, bi-heteroatom co-doped in carbon nanofibers were also received extensively research and expected to acquire high-performance carbon nanofibers anodes through synergistic effect between two different heteroatoms. Adams et al. (2017) has obtained binder-free N- and O- rich carbon nanofiber anodes (CNF-O) through plasma oxidation treatment with electrospun carbon nanofibers derived from PAN. They discovered that the surface area of CNF-O has been increased from 139 to 153 m^2 g^{-1} after plasma oxidation treatment, which resulted in a higher coulombic efficiency (99.5%) than CNF (97.5%) and increased pseudo-capacitance behavior (Figure 3A). Li et al. (2020) also obtained N and O codoped electrospun carbon nanofibers via the precursor of PAN and preasphaltene (PA), and the electrode delivered an impressive potassium ion storage rate (103 mA h g^{-1} at 1 A g^{-1}) and discernible cycling performance with a capacity retention of 98% even after 320 cycles. Recently, Zheng et al. (2022) still optimized the interlayer spacing through N, O codoping in carbon nanofibers, and assosiated with the active sites induced by N, O heteroatom-work together to provide an intercalation-adsorption mechanism for storing K$^+$ in carbonaceous materials.

Hu et al. (2020) obtained phosphorus/nitrogen co-doped hierarchical porous carbon nanofibers (PN-HPCNF) by using triphenylphosphine (TPP), urea and polyacrylonitrile (PAN) as P, N and C resources. Characterization results displayed that P/N co-doped can introduce numerous mesopores and micropores in the carbon nanofibers (PN-CNF) for the reason of released gases (such as PH$_3$, NH$_3$, etc.) during annealing procedures (Figure 3B). In addition, an expanded interlayer spacing (0.41 nm) of PN-HPCNF has been obtained, which is much wider than that of N-HPCNF (0.37 nm) and HPCNF (0.35 nm) (Figure 3C). The unique structure rooted in P/N doping is contributing to accelerating K-ion intercalation/deintercalation, offering sufficient space to buffer volume variation, and possessing superior potassium ion storage performances (Figure 3D).
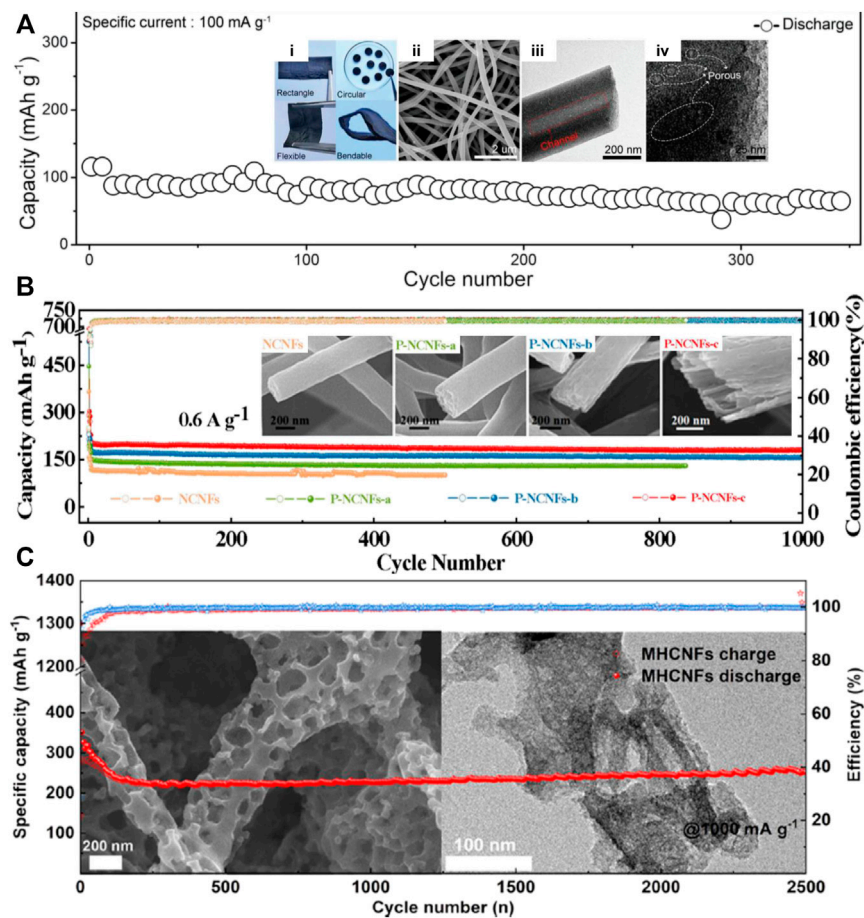
FIGURE 2
(A) Cycling stability of the potassium-based dual-ion batteries based on the hierarchically porous carbon fibers anode at a specific current of 100 mA g^{-1}. **(i)** Optical images of the hierarchically porous carbon fibers tailored into different shapes and bended to different states. **(ii)** SEM image of the hierarchically porous carbon fibers electrode. **(iii)** and **(iv)** Normal and enlarged TEM images of a single fiber in the electrode (Zhang et al., 2019). **(B)** Cycling potassium ion storage performances of electrodes with different PMMA contents (a: 0.4 g; b 0.8 g; c 1.2 g) at a current density of 0.6 A g^{-1} (Chen et al., 2022). **(C)** Cycling behavior of MHCNFs at 1,000 mA g^{-1} (Wu et al., 2021).
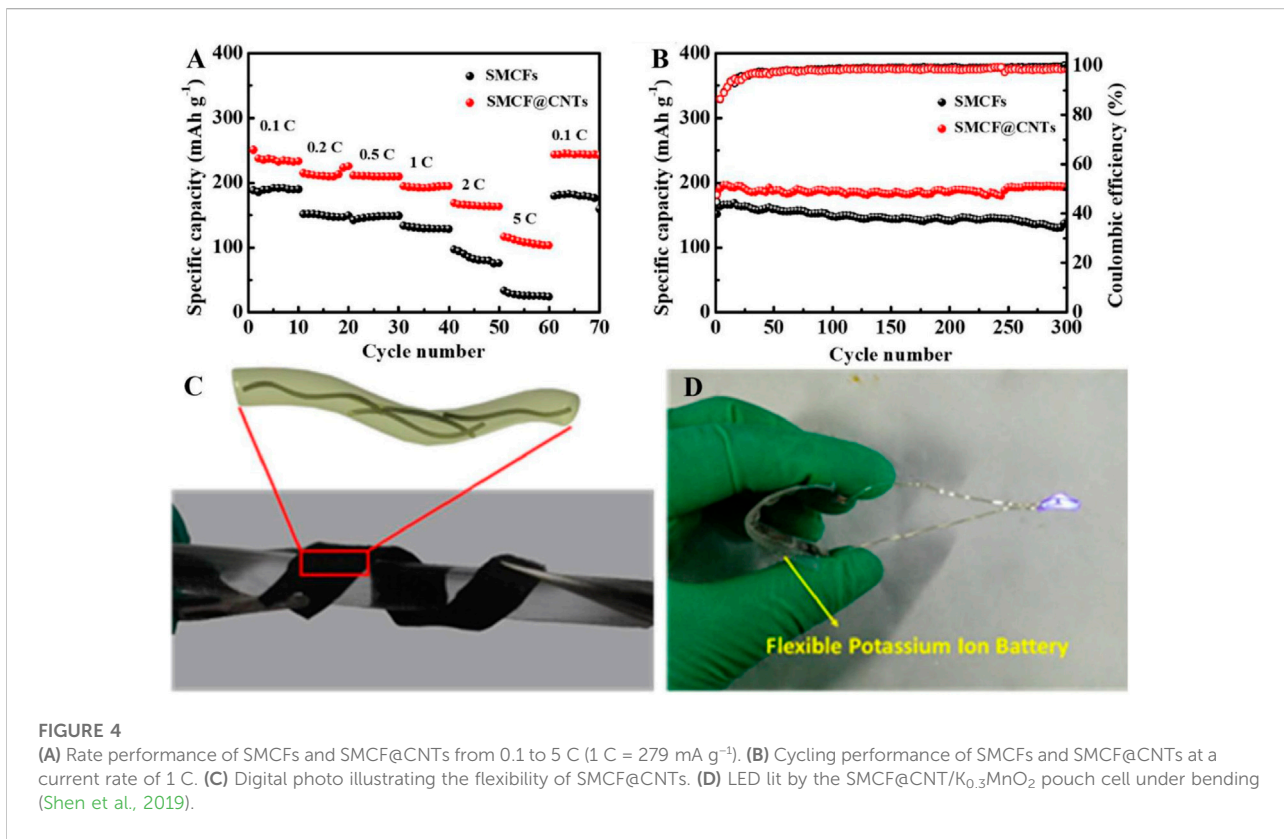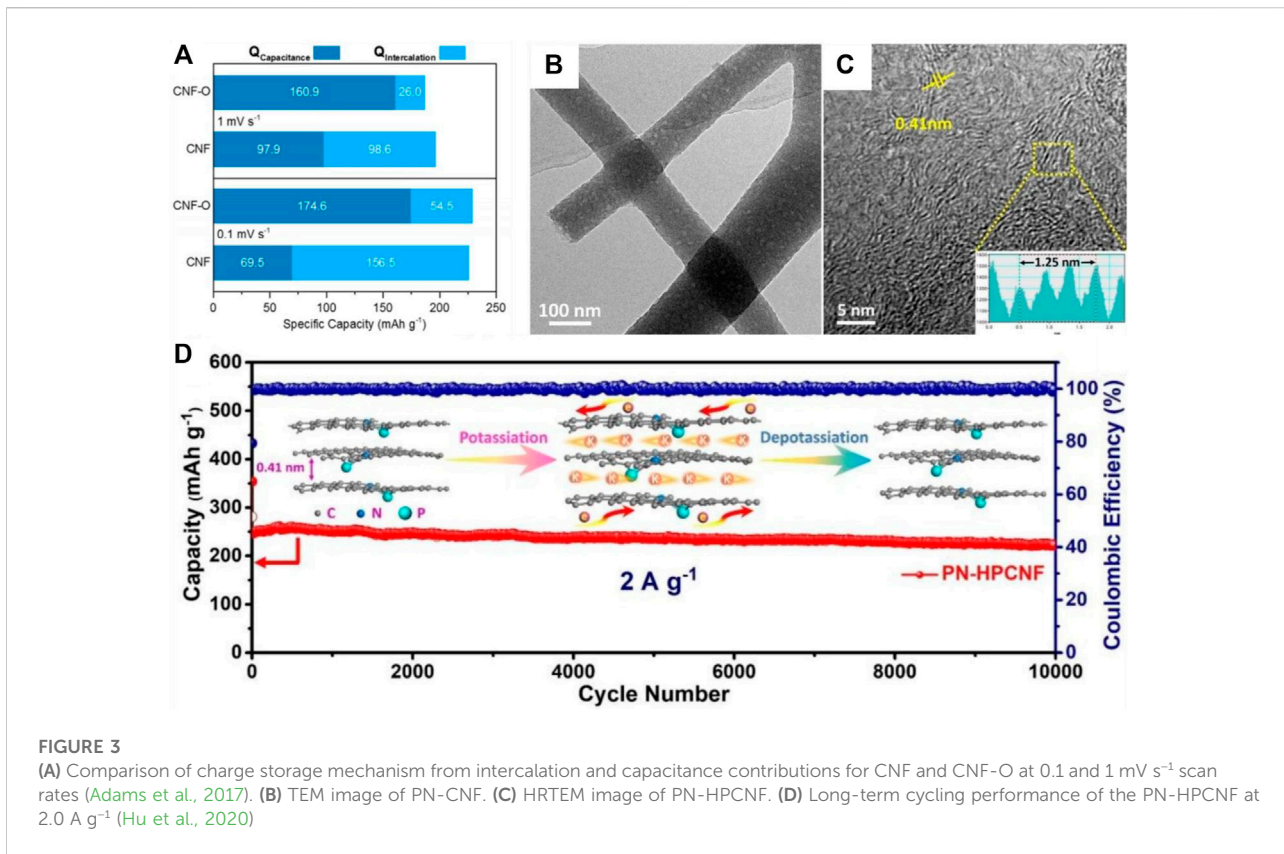
2.2 Electrospun carbon nanofibers based composites anode

Numerous attentions have been focused on the electrospun carbon nanofibers based composites anode to further boost the potassium ion storage properties. On one hand, many kinds of active materials were embedded in the electrospun carbon nanofibers, which played a significant role in protecting active materials and obtained high specific capacity, good rate and long cycling performances (Zhang et al., 2016; Wang et al., 2022c). On the other hand, electrospun carbon nanofibers were also applied as the carrier, where the active materials can grow on the surface to prevent agglomeration and further promote the potassium ion storage properties (Xue et al., 2019; YaoLin et al., 2022). At present, active materials, such as non-metal, metallide, metal

and alloy and so on, were associated with electrospun carbon nanofibers to acquire composite electrodes.

2.2.1 Non metal

Niu et al. (2021) obtained a high performance potassium ion storage material consisting of porous nitrogen-doped carbon nanofibers with graphene (G-PCNFs). The electrochemical performance manifested that graphene can improve the electrical conductivity of the composite materials (the electrical conductivity of 5.5 S m^{-1} for G-PCNFs and 1.6 S m^{-1} for PCNFs. The electrode displayed a high reversible capacity (358 mA h g^{-1} at 0.1 A g^{-1} after 200 cycles), excellent rate performance (101 mAh g^{-1} at 5 A g^{-1}) and ultralong cycling stability (276 mAh g^{-1} at 2 A g^{-1} after 2000 cycles). Shen et al.

FIGURE 3
(A) Comparison of charge storage mechanism from intercalation and capacitance contributions for CNF and CNF-O at 0.1 and 1 mV s^{-1} scan rates (Adams et al., 2017). **(B)** TEM image of PN-CNF. **(C)** HRTEM image of PN-HPCNF. **(D)** Long-term cycling performance of the PN-HPCNF at 2.0 A g^{-1} (Hu et al., 2020)

FIGURE 4
(A) Rate performance of SMCFs and SMCF@CNTs from 0.1 to 5 C (1 C = 279 mA g^{-1}). **(B)** Cycling performance of SMCFs and SMCF@CNTs at a current rate of 1 C. **(C)** Digital photo illustrating the flexibility of SMCF@CNTs. **(D)** LED lit by the SMCF@CNT/K$_{0.3}$MnO$_2$ pouch cell under bending (Shen et al., 2019).

FIGURE 5
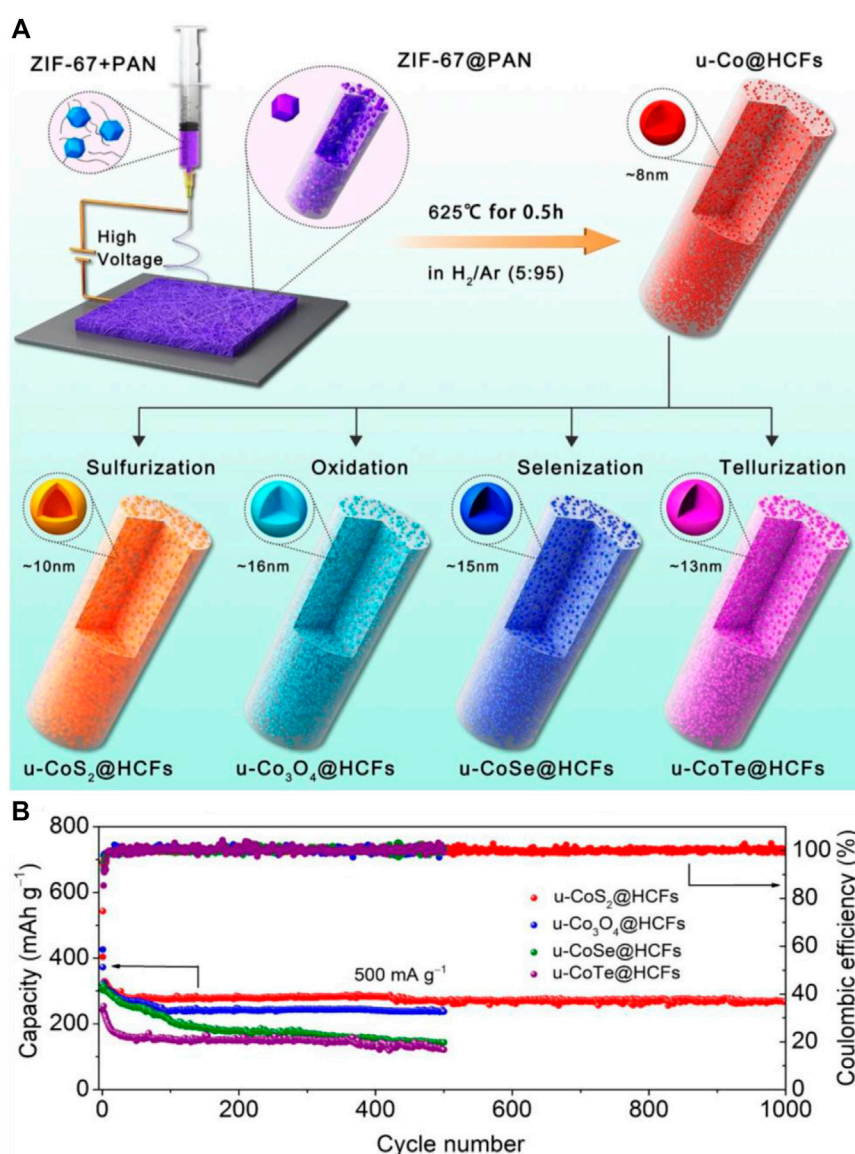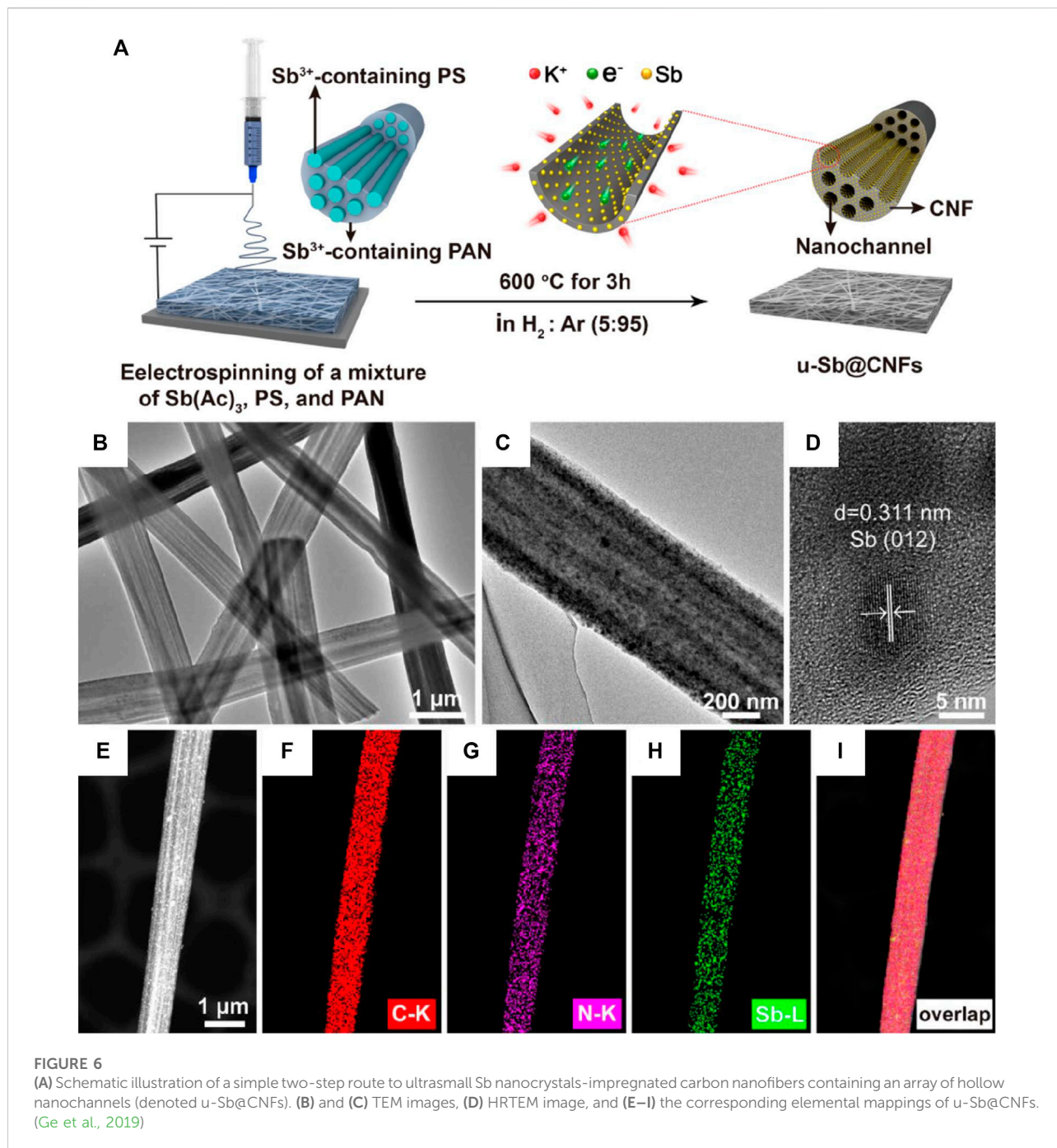**(A)** Schematic illustration of fabricating hierarchical porous nanofibers with closely integrated ultrasmall CoM_x (M = S, O, Se, and Te) nanoparticles and carbon matrix. **(B)** Long-term cycling stability under 500 mA g^{-1} of u-CoM_x@HCFs (Lai et al., 2021).

(2019) also investigated the potassium ion storage properties of self-supporting carbon nanotubes encapsulated in sub-micro carbon fiber electrode (SMCF@CNTs). As displayed in Figures 4A,B, the SMCF@CNT anode exhibited high specific capacity, good rate performance, and cycling stability compared with sub-micro carbon fiber (SMCFs). Furthermore, the SMCF@CNT anode was coupled with $K_{0.3}MnO_2$ cathode, and the pouch cell delivered a capacity of 74.0 mAh g^{-1} at 20 mA g^{-1} with good flexibility (Figures 4C,D). Wu et al. (2019) designed a unique structure of embedding red phosphorus into free-standing nitrogen-doped porous hollow carbon nanofibers to

circumvent the problems of pulverization and aggregation of P particles simultaneously. In this work, the electrode realized unprecedented long cycle life with high reversible capacity (465 mAh g^{-1} at 2 A g^{-1} after 800 cycles) as well as outstanding rate capability (342 mAh g^{-1} at 5 A g^{-1}).

2.2.2 Metallide
Metal oxides showed high specific capacity in lithium storage performance, so the potassium storage performance based on the metal oxides has been widely studied. Huang et al. (2018) synthesized SnO_2-graphene-carbon nanofibers electrode for

FIGURE 6
(A) Schematic illustration of a simple two-step route to ultrasmall Sb nanocrystals-impregnated carbon nanofibers containing an array of hollow nanochannels (denoted u-Sb@CNFs). **(B)** and **(C)** TEM images, **(D)** HRTEM image, and **(E−I)** the corresponding elemental mappings of u-Sb@CNFs. (Ge et al., 2019)

anode of PIBs, where carbon nanofibers and the encapsuled graphene protected the SnO_2 from destruction. The electrode exhibited great cycling stability (202.06 mAh g^{-1} at 100 mA g^{-1} after 100 cycles) and high rate capacity (114.81 mAh g^{-1} at 1 A g^{-1}). Jin et al. (2018) fabricated a flexible and self-standing electrode that consisted of V_2O_3 nanoparticles embedded in porous N-doped carbon nanofibers. The electrode mainly followed the mechanism of intercalation pseudocapacitance and displayed long cycle life.

Large numbers of studies showed that metal sulfide also has good potassium storage performance. Hence, metal sulfides were also used with electrospun carbon fibers to improve potassium storage. Chen et al. (2019) encapsuled FeS_2 and graphene in the electrospun carbon nanofibers, resulting in long-term life of K$^+$ storage (120 mAh g^{-1} at 1 A g^{-1} after 680 cycles). Lai et al. (2021) closely coupled Co-based ultrafine nanoparticles (CoS_2, Co_3O_4, $CoSe$, $CoTe$) and hierarchical porous carbon nanofibers through

using precursors of ZIF-67 and PAN (Figure 5). Particularly, the ultrasmall CoS_2@hierarchical porous carbon nanofibers (u-CoS_2@HCFs) electrode showed high reversible capacity, excellent rate property, and ultralong cycling life (268 mAh g^{-1} at 500 mA g^{-1} after 1,000 cycles) when evaluated as anode for PIBs. He et al., (2022) proposed a facile approach to confine SnS nanocrystals in the walls of hollow multichannel carbon nanofibers to tackle the poor electronic conductivity and huge volume expansion of SnS. The electrode showed 415.5 mAh g^{-1} at 0.1 A g^{-1} after 300 cycles and 245.5 mAh g^{-1} at 1 A g^{-1} after 1,000 cycles. Yang et al. (2021) explored the potassium ion storage performances of In_2S_3/carbon nanofibers electrode. Owing to the presence of carbon nanofibers and the nano-scale of In_2S_3 particles, the volume effect of In_2S_3 was restrained and the conductivity of electrode was improved. Hence, the electrode delivered reversible capacities of 212.2 mAh g^{-1} even at 1 A g^{-1}. Mao et al. (2018) anchored ReS_2 nanosheets onto nitrogen doped carbon nanofibers (ReS_2/N-CNFs). The doped nitrogen can absorb sulfur and polysulfide generated by the conversion reaction. As a result, the ReS_2/N-CNFs composite anode maintains a reversible capacity of 253 mAh g^{-1} after 100 cycles at 50 mA g^{-1} in PIBs. Geng et al. (2021) designed a freestanding PIBs anode through supporting carbon-coated WS_2 nanosheets on carbon nanofibers, which possessed fast K^+ diffusion kinetics and high K-adsorption ability that verified by first-principles calculations and displayed good potassium ion storage properties. In addition, V_5S_8 (Liu et al., 2021), $NiCo_2S_4$ (Zhang et al., 2020b) etc. were also investigated as anode for PIBs with electrospun carbon nanofibers and all exhibited enhanced potassium ion storage properties than pure metal sulfides.

Metal selenides have similar properties to metal sulfides, so their potassium storage properties have been studied extensively as well Liao et al., (2019). Xu et al. (2021) embedded V_3Se_4 nanoparticles in nitrogen/phosphorus co-doped carbon nanofibers, which exhibited an excellent electrochemical performance for PIBs (450 mAh g^{-1} after 100 cycles at 50 mA g^{-1} and 207 mAh g^{-1} after 800 cycles at 2 A g^{-1}). Atangana Etogo et al. (2020) fabricated $Co_{0.85}$Se@carbon nanoboxes embedded in carbon nanofibers films with flexible and freestanding features. As a result, the unique electrode demonstrated excellent cycling stability (reversible capacity of 299 mA h g^{-1} at 1 A g^{-1} after 400 cycles) and remarkable rate performance (166 mA h g^{-1} at 5 A g^{-1}) as an anode for PIBs. Ho Na et al. (2021) also prepared ultrafine ZnSe nanocrystals encapsuled in N-doped porous carbon nanofibers electrode and applied as anode for PIBs. Consequently, the electrode exhibited excellent long-term cycling stability with a reversible capacity of 270 mAh g^{-1} at 0.5 A g^{-1} for 1,000 cycles and a high-rate capacity of 139 mA h g^{-1} at 2.0 A g^{-1}.

Besides, metallic carbide (MoC_2(Min et al., 2021)), metal phosphide (CoP (Miao et al., 2019), FeP (Wang et al., 2020), MoP (Yi et al., 2020)), et al. were applied as anode materials of potassium ion battery with electrospun carbon nanofibers. All electrodes exhibited remarkable potassium ion storage performances.

2.2.3 Metal and alloy

Metal and alloys always displayed high specific capacities for electrochemical storage Zhang et al. (2018b). Therefore, Metal and alloys were also applied as anode for PIBs. Ge et al. (2019) presented a simple strategy that ultrasmall Sb nanocrystals uniformly impregnated in carbon nanofibers containing an array of hollow nanochannels (Figure 6), and the electrode yielded high-performance PIBs with a reversible capacity of 225 mAh g^{-1} after 2000 cycles at 1 A g^{-1}. In addition, Huang et al. (2021) also fabricated porous Sb-graphene-carbon nanofibers electrode, presenting a great cycle life (204.95 mAh g^{-1} at 100 mA g^{-1} after 100 cycles) and excellent rate capability (120.83 mAh g^{-1} at 1 A g^{-1}). Han et al. (2019) embedded CoSb nanoparticles in carbon nanofibers to facilitate the pseudocapacitive behaviors, and the capacity of electrode maintained 250 mA h g^{-1} after 500 cycles at 1 A g^{-1}.

3 Conclusion and perspectives

In this review, the potassium ion anode electrodes based on electrospun carbon nanofibers have been concluded. The modification method for electrospun carbon nanofibers including porous structure designation, heteroatoms doping and composites synthesizing were discussed in detail. The effects of modification on electrospun carbon nanofibers specific surface area, conductivity, lattice distance and potassium storage performance of the electrospun carbon nanofiber electrode were emphatically summarized. The treatment of porous structure designation and heteroatoms doping can boost the cycling stability of electrospun carbon nanofibers. The composite preparation always bring high specific capacity. Therefore, combining the advantages of the three modification methods to prepare high performance potassium storage electrode is one of the future development direction.

Although electrospun carbon nanofibers based anodes have exhibited remarkable potassium ion storage performances, the potassium-based full cells and dual-ion batteries should be further researched, including their properties of energy density, power density and cycling life. In addition, Electrospun carbon nanofibers based electrode has been realized the application of PIBs anode directly and possess flexibility feature at some extent. Nevertheless, with the deeply

development of flexible batteries, electrospun carbon nanofibers with higher strength will be requested.

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MX, Writing manuscript; RL, Consulting and collecting literatures; TY, Writing manuscript; YD, Checking manuscript.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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