Chapter 11

Conducting Polymers for Sodium-Ion Batteries

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Abstract

Sodium-ion batteries are regarded as the most promising substitute of lithium-ion batteries in the future, due to the low cost and sustainability. The appropriate electrode material is the key to achieve commercialization of sodium-ion batteries. Conducting polymers can be applied on both cathodes and anodes separately or as a component of the composite, because of the good conductivity and flexibility. In this chapter, the recent progress of conducting polymer applied in SIBs will be reviewed. There will be two main parts focused on cathodes and anodes, respectively.

Keywords

Conducting Polymers, Cathode, Anode, Sodium-Ion Batteries

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1. Introduction

After over twenty-year development, lithium-ion batteries (LIBs) have not only dominated the portable electronics market but also become the prime candidate of power supply for electric vehicles (EV) and plug-in hybrid electric vehicles (PHEV) [1, 2]. However, the dramatically increasing demand for higher energy density gradually exposes several limits of the first-generation lithium-ion batteries. On the one hand, the lack of appropriate electrode materials leads to difficulties for achieving high energy density. On the other hand, the limitation and high cost of global lithium resources also sound the alarm for the sustainability of Li-ion batteries. Up to now, a considerable number of novel electrode materials have been developed and countless efforts have been done to improve the electrochemical performance of these materials. In order to realize sustainable and low-cost energy storage systems, sodium-ion batteries (SIBs) started to return to the vision of academia in about 2010 [2]. The origin of SIBs can be dated back to the 1960s when high-temperature Na-based batteries were started to be studied [3]. In the following decade, high-temperature Na-S [4, 5] and Na metal chloride [6, 7] batteries could be regarded as the predecessors of sodium-ion batteries, which were developed and successfully commercialized in some specific applications. Since 1980, using ions (Li and Na ions) as charge carriers became a research hotspot to realize the normal-temperature electrochemical energy storages [8]. At that time, both LIBs and SIBs were expected to realize commercialization in the near future. The appearance of graphite-based anodes greatly promoted the development of lithium-ion batteries which were finally commercialized in 1992 by Sony [9-11]. In contrast, due to the relative-low energy density and lack of appropriate anode materials, studies of sodium-ion batteries gradually stepped into a frozen period [8]. After a 20-year dormancy, the destiny of sodium-ion batteries finally embraced a turning point in around 2010.

Sodium, an element belongs to the same family of lithium, has similar physical and chemical properties of lithium, which is the best candidate to substitute lithium for battery applications. Compared to lithium, the abundant reserve and low cost of sodium resource make it possible to establish low-cost and scale-up systems for energy storage
However, due to some natural characteristics of sodium, sodium-ion batteries are inferior to lithium-ion batteries in some aspects, such as the lower energy density derived from the higher equivalent weight of Na. In addition, because of the difference between ion radii, some electrode materials which exhibit good performance in LIBs cannot be applied on SIBs directly [13, 14]. As for sodium-ion batteries, developing appropriate and high-performance electrode materials is also the key point to promote SIBs to the market as soon as possible. Currently, normal electrode materials of SIBs include transition metal oxides, phosphates, fluorides, sulfides, phosphides as cathodes, and carbonaceous materials, semi-metals (Ge, Sb, Sn) as anodes [8, 15, 16]. Moreover, organic electrode materials for SIBs are gaining increasing research interest due to a number of intrinsic advantages, such as lighter weight, lower cost, more sensitive to sodium ions, better plasticity and flexibility than inorganic materials [17-19]. Generally, most of organic electrode materials (small organic molecules and polymers) suffer from their poor conductivity, leading to insufficient electrochemical performance [20, 21]. Fortunately, there is an exception which is the conducting polymers.

Conducting polymers refer to those polymers which can present electric conductivity derived from the conjugated structures in their molecule chains [22-24]. Alan MacDiarmid, Hideki Shirakawa, and Alan J. Heeger discovered the unique characteristic of conducting polymers in 1976 and they were awarded the Noble Prize in Chemistry in 2002 for this [22, 25, 26]. Since the 1980s, due to the unique property, conducting polymers have been applied in various electronic devices, including transistors, sensors, memories, actuators/artificial muscles, supercapacitors, and lithium-ion batteries [27-32]. In recent years, the renaissance of sodium-ion batteries provided opportunities for conducting polymers to explore more applications. Inversely, conducting polymers could be used to construct appropriate electrode materials to achieving high energy density sodium-ion batteries. Conducting polymers can not only be applied as cathodes or anodes directly due to the redox-doping mechanism but also be used to fabricate composite electrodes with conventional materials [20, 21]. As a part of the electrode, conducting polymer is able to enhance the conductivity of the electrode, and alleviate the volume changes of those materials which suffered from this problem[33, 34]. Normally, the conducting polymers applied on sodium-ion batteries include polypyrrole (PPy), polyaniline (PANI), polythiophene (PTH), poly(3,4-ethylene dioxythiophene) (PEDOT), and so on. Molecular structures of these conducting polymers are presented in Fig. 1. At present, conducting polymers have been regarded as an important branch of SIB electrode materials and additives. In this chapter, we will review the progress of the studies about applications of conducting polymers in sodium-ion batteries. Hereinafter, there will be two main parts which focus on the studies of cathodes and anodes,
respectively. In each part, different applying types of conducting polymers will be discussed separately.

![Molecular structures of typical conducting polymers.](image)

2. Applications on cathode materials

2.1 Doped and pure conducting polymer cathodes

Due to the redox-doping reaction mechanism, the electrochemical activity of conducting polymers have been demonstrated and they have been widely applied as electrode materials for lithium-ion batteries since about 2000 [35-38]. Up to the 2010s, conducting polymers started to be considered to act as cathodes for sodium-ion batteries. However, when conducting polymers were directly used as cathodes in SIBs, they would react with large electrolyte anions via a p-doping/de-doping mechanism, leading to a poor capacity utilization of the polymer chains [39]. In order to solve this problem, one strategy appeared recently which is doping some ionizable groups in the conducting polymers to change the redox reaction mechanism from conventional p-doping/de-doping processes of large electrolyte anions to the insertion-extraction reactions of small cations (Na ions) [39-47].

Cao and Yang synthesized redox-active Fe(CN)$_6^{4-}$-doped polypyrrole (PPy-FC) which exhibited good Na-storage performance as a cathode for SIBs [41]. The PPy-FC cathode could keep a reversible capacity of 110 mAh g$^{-1}$ after 100 cycles at a rate of 50 mA g$^{-1}$, corresponding to a capacity retention of 85%. In addition, it could also deliver a specific capacity of ca. 70 mAh g$^{-1}$ at a high rate of 1600 mA g$^{-1}$, and recover mostly its initial capacity of 125 mAh g$^{-1}$ when the current density was changed back to 50 mA g$^{-1}$. Based on the analysis of Fourier-transform infrared spectroscopy (FTIR), the reaction
mechanism of PPy-FC cathode at the first discharge can be presented as followed equation (Eq. 1):

$$\text{PPy}_{17.1}^{4+}\cdot\text{Fe(CN)}_6^{4-} + 4\text{Na}^+ + 4e^- \leftrightarrow \text{PPy}_{17.1}^{4+}\cdot\text{Fe(CN)}_6^{3-}\cdot\text{Na}^+$$  \hspace{1cm} (1)

When the charging continued, the oxidation of the Fe(CN)$_6^{4-}$ dopant and electrochemical redox of PPy chains would happen, as presented by Eq. 2 and Eq. 3:

$$\text{PPy}_{17.1}^{4+}\cdot\text{Fe(CN)}_6^{4-} + \text{PF}_6^- - e^- \leftrightarrow \text{PPy}_{17.1}^{4+}\cdot\text{Fe(CN)}_6^{3-}\cdot\text{PF}_6^-$$  \hspace{1cm} (2)

$$\text{PPy}_{17.1}^{4+}\cdot\text{Fe(CN)}_6^{3-}\cdot\text{PF}_6^- + x\text{PF}_6^- - xe^- \leftrightarrow \text{PPy}_{17.1}^{(4+x)^+}\cdot\text{Fe(CN)}_6^{3-}(1+x)\text{PF}_6^-$$  \hspace{1cm} (3)

According to these equations, the theory capacity of PPy-FC cathode could be regarded as a combination of Na-insertion/extraction (80 mAh g$^{-1}$), Fe(CN)$_6^{3-/4-}$ redox (20 mAh g$^{-1}$), and p-doping reaction (35 mAh g$^{-1}$), which was close agreement with the experiment value. Besides, the chemical structure and redox mechanism of PPy-FC conducting polymer are displayed by Fig. 2(a).

Polyaniline (PANI) is also a promising electrode material for sodium-ion batteries. In 2015, Jiang and Wang reported their work about sulfonated polyaniline, poly(aniline-co-amino benzene sulfonic sodium) (PANS), as the cathode for sodium-ion batteries [45]. They selected the sulfonate as doping group which has stronger electron-withdrawing ability onto the redox-active polyaniline chains, in order to improve the drawbacks derived from the large doping group (benzene sulfonic sodium) as they reported previously [44]. The PANS electrode was fabricated by a chemical oxidative copolymerization. The chemical structure of PANS and its reaction mechanism during the charge/discharge process are presented in Fig. 2(b). Based on the reversible Na-insertion/extraction reaction, this PANS cathode exhibited a reversible capacity of 133 mAh g$^{-1}$ after 200 cycles (100 mA g$^{-1}$), corresponding to high capacity retention over 96%.
Except for modifying active groups on polymer chains, another strategy to enhance the Na-storage performance of CPs is constructing nanostructures in order to facilitate electrolyte penetration and ionic transfer [48-51]. In 2015, Wang and Su synthesized PPy hollow nanospheres using poly(methyl methacrylate) (PMMA) nanospheres as templates, as shown in Fig. 3(a). The average values of the inner and outer diameters of the as-prepared PPy hollow nanospheres were 136.5 nm and 242 nm, respectively. Due to such hollow structure, this PPy cathode exhibited stable cyclability as well as good rate performance. In 2016, Feng and Mai fabricated 2D mesoporous polypyrrole (mPPy) nanosheets with controlled pore sizes via synergic and demonstrated its superior electrochemical performance as SIB cathode [49]. In this work, amphiphilic aliphatic amine (octadecylamine, OTA) and polystyrene-block-poly(ethylene oxide) (PS-b-PEO) block copolymers were selected as templates. Because of the self-assembly property, OTA and PS-b-PEO firstly formed sandwich-like assemblies by monolayer arrangement of spherical micelles (PS-b-PEO) on both surfaces of the 2D bilayers (OTA). Then pyrrole monomers were polymerized on the surface of this 2D templates due to the adsorption of poly(ethylene oxide) domains. After removal the templates, 2D mPPy nanosheets were obtained with pore sizes of 6.8 – 13.6 nm, thickness of 25 – 30 nm, and surface area of 96 m² g⁻¹. The pore size of as-prepared mPPy nanosheets could be
controlled by varying PS block lengths of PS-\(b\)-PEO deblocks. As for the electrochemical performance, this PPy cathode maintained its initial capacity of 83 mAh g\(^{-1}\) after 120 cycles at a high rate of 300 mA g\(^{-1}\). Moreover, polyaniline hollow nanofibers (Fig. 3(b)) and normal nanofibers were synthesized at most recently, and they exhibited competitive sodium storage performance [50, 51].

![Figure 3](image.png)

**Figure 3.** a) Schematic diagram of the synthesis of PPy hollow nanospheres, ref.[48], copyright the Royal Society of Chemistry 2015. b) Scheme for the fabrication of the PANI hollow nanofibers, ref.[50], copyright 2019 Elsevier Ltd..

### 2.2 Conducting polymer-based composite cathode

Up to now, a verity of inorganic cathode materials have been developed and almost dominated the cathode field of sodium-ion batteries, including transition metal oxides [52], phosphates [53], phosphorus-based polyanion materials [54-56], Prussian blue (PB) and its analogs sodium manganese hexacyanoferrates (NMHFC) [57-59], and so on. Although these materials display high theoretical capacities and potential plateaus, most of them suffer from low conductivity, leading to poor electrochemical performance. Conducting polymers are ideal partners for these conventional cathode materials, which can not only enhance the electric conductivity of the electrode but also decrease the dosage of additional binders and conductive agents. In recent years, inorganic/organic (CPs) composite SIB cathodes have attracted much attention and gained good effects [33, 60-67].

In 2015, Chou et al. synthesized ClO\(_4\)-doped polypyrrole coated Na\(_{1+x}\)MnFe(CN)\(_6\) composite (NMHFC@PPy) via a facile self-polymerization approach, which was successfully applied as high-performance cathode for SIBs [60]. In this composite, PPy played multiple important roles, including conductive coating layer to enhance the conductivity, protective layer to reduce the dissolution of Mn in the electrolyte, and
active materials to increase the capacity. The electrochemical reaction mechanism of NMHFC@PPy composite is displayed in Fig. 4(a). Due to these advantages above, the NMHFC@PPy composite cathode exhibited improved electrochemical performance than that of bare NMHFC electrode, as shown in Fig. 4(a). This composite cathode could maintain capacity retention of 67% after 200 cycles (at 0.1 C, 1 C = 120 mA g⁻¹), and keep 46% of initial capacity under 40 C rate. In 2017, Wang et al. reported a novel nanostructured composite cathode where a string of nickel hexacyanoferrate (NiCHF) nanocubes coaxially anchored on bipolar conducting polymer (BCP) coated CNTs (CNTs@BCP) [63], as presented in Fig 4(b). The bipolar conducting polymer was derived from intrinsic PANI (p-doping BCP) and Fe(CN)₆³⁻-doped PANI (n-doping BCP). The synthesis of CNTs@NiHCF-BCP cathode was realized via an electrochemical route. At first, a CNT modified electrode was obtained by distributing the CNTs (solution) on a stainless-steel sheet and undergoing a drying process. Then, the NiHCF and BCP were coaxially grown on it directly in a three-electrode system. The final hybrid cathode was obtained after rinsing and drying, which could be used in batteries without extra addition of binder and conductive agent. This CNT@NiHCF-BCP cathode exhibited a good cycling stability, which retained a reversible capacity of about 122 mAh g⁻¹ after 500 cycles at a rate of 0.5 C. Moreover, it also remained at 97 mAh g⁻¹ at a high rate of 15 C, corresponding to 77% retention of its reversible capacity at 1 C. Such good electrochemical performance of CNT@NiHCF-BCP cathode could be attributed to the synergistic effect of the three active components of CNTs, NiHCF nanocubes, and BCP. In terms of structure, the network built by these three components benefited the electronic and ionic transportations. In addition, all these three components could provide capacity via different reaction mechanisms. The NiHCF can react with sodium ions electrochemically, as described by Eq. 4:

\[
\text{NaNiFe(CN)_6}^+ + \text{Na}^+ + e^- \leftrightarrow \text{Na}_2\text{NiFe(CN)_6} \quad (4)
\]

In case of PANI, the electrochemical reactions are realized by p-doping/de-doping of the ClO₄⁻ anions and p-doping/de-doping of the Fe(CN)₆³⁻ anions, which presented by Eq. 5 and Eq. 6:

\[
[(\text{C}_6\text{H}_6\text{N}^+)(\text{ClO}_4^-)] + \text{Na}^+ + e^- \leftrightarrow \text{C}_6\text{H}_6\text{N} + (\text{Na}^+)(\text{ClO}_4^-) \quad (5)
\]

\[
[(\text{C}_6\text{H}_6\text{N}^+)\text{Fe(CN)_6}^{3-}] + 4\text{Na}^+ + 4e^- \leftrightarrow [(\text{C}_6\text{H}_6\text{N}^+)\text{Fe(CN)_6}^{4-}(\text{Na}^+)_4] \quad (6)
\]
3. Applications on anode materials

3.1 Doped and pure conducting polymer anodes

Similar to the applications on cathodes, conducting polymers can also be used as anode materials for sodium-ion batteries based on the redox-doping mechanism [68-71]. In 2016, Liu and Wang applied polypyrrole to act as anode for SIBs for the first time and compared different electrochemical performance between submicron polypyrrole (S-PPy)
and bulk polypyrrole (B-PPy) [68]. In result, the S-PPy electrode exhibited better Na-
storage property which could be ascribed to the enhanced electrical contact between PPy
particles due to the smaller size. Besides, the smaller particles size also benefited the
penetration of the electrolyte into active material, leading to improved electrochemical
performance. Except for the typical conducting polymers, like PPy and PANI, several
novel conductive polymers were synthesized recently and their potential applications as
SIB anodes have been demonstrated. Armand et al. synthesized a family of
polySchiff/oligoether terpolymers which exhibited electrochemical activity with Na⁺ ions
and gained good performance as SIB anodes [70]. In addition, Jiang et al. prepared
cyclized polyacrylonitrile nanofibers (cPAN-NFs) as anode for SIBs, through a scalable
electrospinning technique followed by thermal stabilization [71]. cPAN was a typical
poly(N-heteroacene) with the substitution of one nitrogen atom in each aromatic ring,
which displayed enhanced n-dopable reactivity. The cPAN-NFs anode exhibited a high
specific capacity of 527 mAh g⁻¹ at 0.05 A g⁻¹. Moreover, it could maintain a reversible
capacity of ca. 220 mAh g⁻¹ after 3500 cycles at 5 A g⁻¹, corresponding to a capacity
retention of 99.4 %.

3.2 Conducting polymer-based composite anode

Although some conducting polymers exhibited stable cyclability as SIB anodes, the
specific capacities were still insufficient compared to those inorganic anodes. However,
the popular inorganic anodes suffer from serve volume expansion upon sodiation, such as
tin, antimony materials and their oxides [2]. Conducting polymers, as ideal buffer matrix,
have been widely used to constructing composite anodes for sodium-ion batteries [34, 72-
85].

Antimony (Sb) is one of the favorite anode materials for sodium-ion batteries because of
its high theoretical capacity of 660 mAh g⁻¹ [86]. In 2015, Nam and Kwon combined
conducting polymer with Sb-based material, in order to improve its electrochemical
performance [34]. Firstly, the PPy nanowire network was prepared through an
electrochemical approach. PPy nanowires grew on an oxidizable metal substrate via a
cathodic electro-polymerization method, as shown in Fig. 5(a) [87]. Then, Sb and Sb₂O₃
nanocrystals were electrodeposited on the PPy network via Eq. 7 and Eq. 8, forming
Sb/Sb₂O₃-PPy composite anode material.

\[
Sb_2(C_4H_2O_6)_2^{2-} + 4H^+ + 6e^- \leftrightarrow 2Sb + 2(C_4H_2O_6)^{2-} \tag{7}
\]

\[
Sb_2(C_4H_2O_6)_2^{2-} + 2OH^- + H_2O \leftrightarrow Sb_2O_3 + 2(C_4H_2O_6)^{2-} \tag{8}
\]
The porous network generated by crosslinked PPy nanowires could not only alleviate volumetric changes of Sb and Sb$_2$O$_3$ during cycling processes but also enhance the electric and ionic conductivity of the electrode. As a result, this composite anode presented good stability within 100 cycles (at 60 mA g$^{-1}$), and retained a reversible capacity of 512 mAh g$^{-1}$ with a retention of 98%. The existence of PPy network also endowed a good rate performance of this composite electrode, which maintained a capacity of 299 mAh g$^{-1}$ at a high current density of 3300 mA g$^{-1}$.

Figure 5. a) Schematic diagram showing the effects of the reactivity of radical cations on the formation of polypyrrole nanostructures in cathodic electropolymerization, ref.[87], copyright the Royal Society of Chemistry 2013. b) Scheme illustration of synthesis for the CNT@SnO$_2$@PPy nanocomposites, ref.[82], copyright 2018 Elsevier B.V.. c) Schematic illustration of the preparation of the amorphous Bi$_2$S$_3$–polypyrrole hollow spheres (Bi$_2$S$_3$–PPy HSs) electrode, ref.[85], copyright the Royal Society of Chemistry 2013.
Tin oxides are another kind of promising anode material for Na-ion batteries, and their applications need effective methods to improve the cycling stability. Wang et al. fabricated SnO$_2$-PPy composite anode by an in-situ hydrothermal synthesis, where SnO$_2$ nanoparticles are homogeneously distributed on the surfaces of the PPy nanotubes [75]. Li et al. prepared sandwich-like CNT@SnO$_2$@PPy nanocomposites, in which SnO$_2$ nanoparticles were deposited on the surface of CNTs via a liquid-phase deposition (LPD) approach and PPy were further coated on the surface of CNT@SnO$_2$ hybrids by a chemical-polymerization route (Fig. 5(b)) [82]. Due to the assistance of PPy, the electrochemical performance of this anodes was effectively improved. Besides, Tang et al. synthesized amorphous Bi$_2$S$_3$ hollow spheres (Bi$_2$S$_3$-PPy HSs) which could be used as anode for SIBs after reduction as well as sulfur host of Li-S batteries after sulfuration [85]. As illustrated in Fig. 5(c), the BiG spheres were prepared by a one-pot hydrothermal method, in which Bi nanoparticles were encapsulated in large glycol spheres. Then, PPy was coated on the surface of BiG spheres to form BiG-PPy spheres. Driven by the Kirkendall Effect, the hollow structured Bi$_2$S$_3$-PPy spheres were gained. After an electro-reduction process, the as-obtained Bi-PPy hollow sphere could be used as anode for sodium-ion batteries. Due to the hollow structure and PPy shell, this anode exhibited good rate performance with a reversible capacity of 278 mAh g$^{-1}$ at 3000 mA g$^{-1}$ and a long lifespan upon 5000 cycles when cycled at 1000 mA g$^{-1}$. Because of the outstanding plasticity and conductivity, polypyrrole almost became the first choice for constructing CPs-based anodes for sodium-ion batteries. There were more studies about high-performance SIB anodes assisted by PPy, including graphene oxide-PPy composites[76], CoFe$_2$O$_4$-PPy nanotubes [77], ZnS@PPy spheres [78], TsONa/PPy/TiO$_2$/Ti composite [79], Sb$_2$Se$_3$@PPy microclips [81], CoP@PPy nanowires/carbon paper composite [83], and flowerlike Sb$_2$S$_3$/PPy microspheres.

**Conclusions & Outlooks**

Since the discovery of conducting polymers in 1976, they have been widely applied in various energy storage devices due to their unique properties. Sodium-ion batteries, as one of the most promising techniques for future energy storages, embraced an explosive development and gained great progress since 2010. Constructing high-performance electrode materials has been regarded as the requirement to promote SIBs to practical applications. Conducting polymers have been demonstrated that they could play significant roles in both cathodes and anodes of SIBs. One the one hand, doped or pure conducting polymers could be directly used as cathodes or anodes due to the redox-doping mechanism. Moreover, assisted with nanostructures, CPs electrodes could boost better electrochemical performance. On the other hand, conducting polymers could
effective improve the electrochemical performance of inorganic electrodes via forming composite materials. Particularly, conducting polymers could enhance the electric conductivity of conventional inorganic cathodes, and also act as buffer matrix for those anodes which suffer from the volumetric expansion during the cycling process. Meanwhile, conducting polymers could also act as binder and conductive agent, which is beneficial to increase the energy density of batteries. After several-year development, various conducting polymers have been applied in sodium-ion batteries, including PPy, PANI, PTh, and PEDOT. Among them, PPy attracted most attention especially in the field of composite anodes.

With the increasing demand for low-cost and sustainable energy storage devices, sodium-ion batteries have become the premier candidate for the large-scale energy storage system in the future. At present, one bottleneck of SIB development is the lack of appropriate electrode materials. The emerge of conducting polymers can be considered as a good complement for the electrode material resource library of SIBs. It is convinced that sodium-ion batteries will realize commercial applications in the near future with the assistant of conducting polymers.

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