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Review

Conductive Polymers on the Cathode Side of Lithium-Sulfur Batteries

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Abstract: Lithium--sulfur batteries are regarded as highly promising next-generation energy storage devices due to their outstanding theoretical specific capacity (1675 mAh/g), high energy density (2600 Wh/kg), abundant raw material sources, environmental compatibility, and low cost. Nevertheless, their practical implementation remains hindered by rapid capacity fading and limited cycle life. Conductive polymers can not only enhance overall electrode conductivity but also provide physical or chemical barriers that suppress the shuttle effect of lithium polysulfides, thus alleviating capacity decay and improving electrochemical performance. This review focuses on the use of polypyrrole, polythiophene, polyaniline, and polyacrylonitrile in lithium--sulfur batteries. Representative advances in recent years are highlighted, and the future role of polymers in advancing the practical application of lithium--sulfur batteries is envisioned.

Keywords: Lithium-sulfur battery; Polymer; Cathode; Separator

1. Introduction

The booming energy demand and formidable environmental crisis are igniting interest in a variety of efficient storage and sustainable utilization of clean, renewable energy, such as solar, wind, wave, geothermy and so on. When it comes to energy storage, people will definitely think of batteries. As a new form of energy use, secondary batteries are gradually changing human production and life [1]. The lithium-ion battery is light in weight and compact in operation. It operates at a voltage of about 4 V and has an energy density of 150-250 Wh/kg [2]. Due to its high-capacity energy storage, lithium-ion batteries have led to an increase in the consumer electronics market [3]. However, as a representative of practical applications, lithium-ion batteries almost reached the limit of theoretical energy density [4-6]. Low energy density batteries can not meet the demand for energy storage, and it is necessary to develop energy storage devices with higher energy density. At this time, people turned their attention to lithium-sulfur batteries with a theoretical energy density of 2600 Wh/kg [7-10]. Moreover, lithium-sulfur batteries have the advantages of abundant sulfur reserves, low price and environmental friendliness, which makes the research of lithium-sulfur batteries rapidly popular. This technology of lithium-sulfur battery is by no means new, as it was filed as early as 1957 and patented in 1960 [11]. However, as we know, the lithium-sulfur battery has too low volumetric energy density and has safety hazards, so it has not been taken seriously. As the research on the failure mechanism of lithium-sulfur batteries becomes clearer, people realize that they can be "treated". Nowadays, lithium-sulfur batteries have been the scientific focus subject of research the world over [12]. Lithium-sulfur batteries can return to the hot trend of research, which is inseparable from the "healing agent", and many of the healing agents are polymers. The history of the use of natural polymers by humans was quite long, until 1839 when C. Goodyear proposed the vulcanization of natural rubber, which made substantial progress in polymer applications [13-15]. Today, polymers have become a

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necessity for people's daily life and production. As we know, lithium-sulfur batteries are rich in sulfur, low in price and environmentally friendly [16-20]. However, it also has poor conductivity of sulfur and its discharge products [21-24], the shuttle effect of polysulfide soluble in the electrolyte [25-29], the serious volume expansion of the final discharge product of sulfur [30-33], and the safety of the anode dendrites [34-37]. Polymers are still useful for serious problems such as volume expansion, lithium dendrites, and shuttle effects. The polymer, especially the conductive polymer, can improve the overall conductivity of the battery by doping or coating, and the polymer can also inhibit the shuttle of polysulfide by physical or chemical means. In addition, the polymer itself is more flexible [38-41], which is advantageous for buffering the volume expansion and dendrite control of the sulfur reaction process. Thus, the application of polymers to lithium-sulfur batteries may produce a synergistic effect of 1+1>2. This review mainly introduces polypyrrole, polythiophene, polyaniline and polyacrylonitrile, and also involves some other polymers(as illustrated in Figure 1), and prospects for the application of conductive polymers on the cathode side of lithium-sulfur battery.

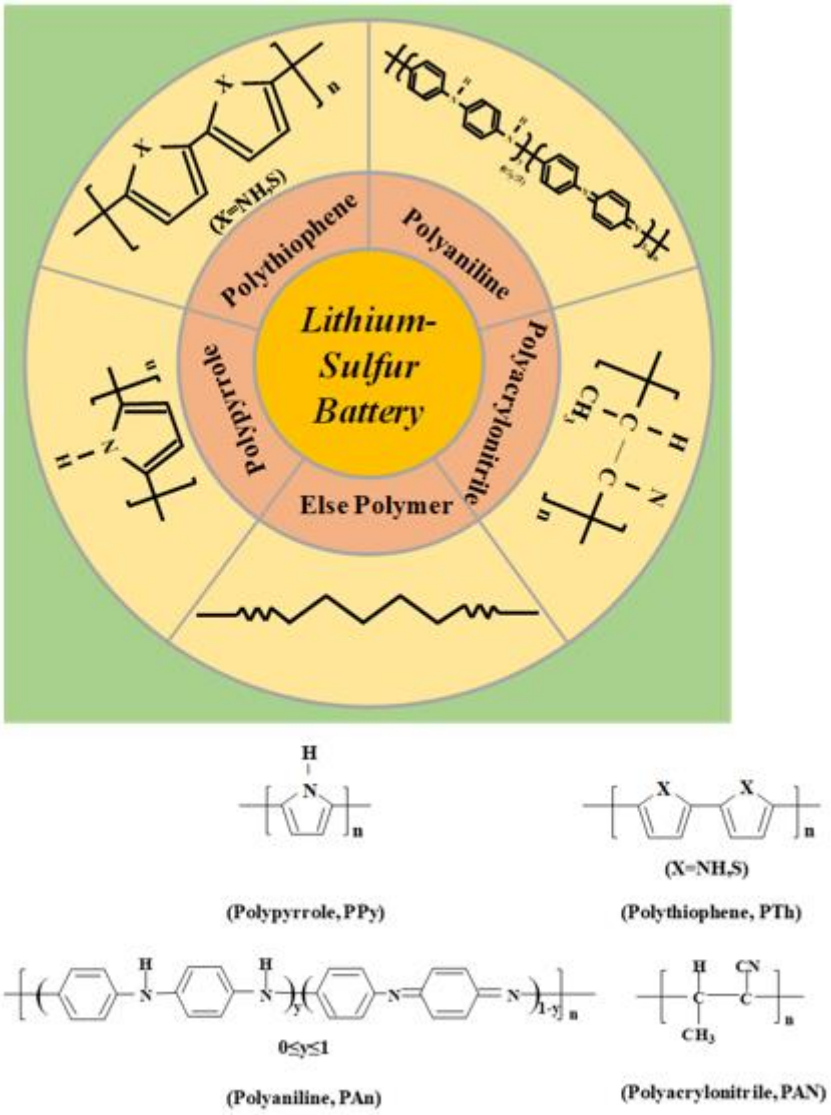

Figure 1. Schematic Diagram of the Structure of Four Conductive Polymers

2. Polymers in Lithium-Sulfur Batteries

Lithium-sulfur battery has an ultra-high theoretical energy density of 2600Wh/kg, which is ten times that of commercial lithium-ion batteries. Moreover, sulfur is abundant and sulfur itself has no environmental threats. However, lithium-sulfur batteries are far

from industrialized, mainly due to the shuttle effect, poor conductivity of sulfur and its discharge products and volume expansion of up to 80%. Polymers are undoubtedly a good medicine for the treatment of these "conditions". In terms of structure, polymers usually have a carbon chain as the main part, and the chains are connected by intermolecular forces. This theoretically provides a possible "elemental effect" for its inhibition of the shuttle effect of polysulfides. Polymers are used in lithium-sulfur batteries, especially conductive polymers, which not only increase the overall electronic conductivity but also certain specific functional groups or structures can inhibit the shuttle effect of lithium polysulfides. This article will introduce and summarize the application of polypyrrole, polythiophene, polyaniline, polyacrylonitrile and other conductive polymers on the cathode side of lithium-sulfur batteries, as shown in Figure2.

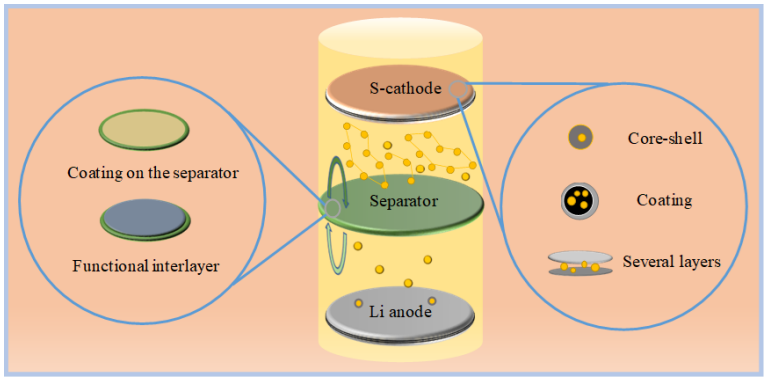

Figure 2. Schematic Diagram of Several Parts Outlined in This Review

2.1. Polypyrrole

Polypyrrole has a conjugated structure in which -C-C- and -C=C- are alternately connected. The π-electrons in the double bonds are similar to free electrons in the metal, and can conduct electricity in the presence of an electric field, and their conductivity can reach 10^{-2}-10^{-3}S/cm.

Polypyrrole can not only inhibit the shuttle effect of polysulfide due to its nitrogen content, but also because its electrochemical window is more consistent with that of lithium-sulfur batteries, which means that it can not only serve as a conductive agent but also bring about an increase in capacity.

2.1.1. Cathode

When it comes to the cathode of lithium-sulfur, the first thing that comes to mind is the poor conductivity of sulfur. In order to solve this problem, many researchers applied polypyrrole to improve the conductivity of sulfur.

Xin et al. made a sulfur-polypyrrole composite as a cathode material for lithium-sulfur battery by a facile ball-milling route [42]. This method is easy and efficient. Compared to the pure sulfur cathode, it has better special capacity, cycle performance and multiplier performance, which is mainly the polypyrrole not only acting as a conductive but as an inhibitor of shuttle effect of polysulfide. Zhang et al. also made a branched sulfur/polypyrrole nanocomposite by a simple one-step ballmilling without heat-treatment [43]. Due to the high conductivity and large area of polypyrrole, which shows the discharge special capacity of 1320mAh/g.

In order to further improve the electron conductivity on the surface of sulfur particles, many people are working on a coating of polypyrrole on the surface of sulfur particles, such as a core-shell structure. Fu et al. prepared a core-shell structure sulfur-polypyrrole composite cathode, which consists of spherical sulfur particles coated with polypyrrole(Figure.3a)[44]. Polypyrrole as a conductive coating increases the electronic conductivity on the surface of the sulfur particles. Ma et al. designed a sulfur cathode with three-dimensional (3D) cubic mesoporous carbon CMK-8 as the matrix of sulfur, and polypyrrole (PPy) as the wrapping layer (The microstructure of the polypyrrole-coated

CMK-8 / S composite is shown in Figure 3b.)[45]. The coating of polypyrrole not acts as the inhibitor of polysulfide but as a rapid lithium-ion channel. Wu et al [46]. reported a facile in-situ sulfur deposition and chemical oxidative polymerization to prepare acetylene black/sulfur@polypyrrole (AB/S@PPy) composites for lithium-sulfur battery. PPy covers onto the surface of the AB/S composite forming a core-shell structure.

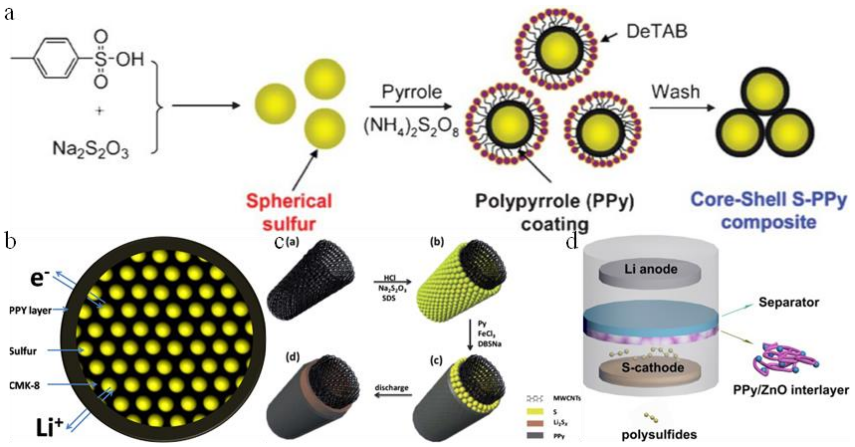

Figure 3. Schematic Diagram of Polypyrrole as Sulfur Coating and Interlayer: A. Synthetic Flow Chart of Sulfur-Polypyrrole Composites with Core-Shell Structure [44]; B. Schematic of CMK-8/S Composite Coated with Polypyrrole [45]; C. Schematic Illustration for the Synthesis and Discharge Process of the Dual Core--Shell Structured MWCNTs@S@PPy Composite [50]; D. Schematic of the Cell with PPy/ZnO Interlayer [51].

In addition to this simple coating, some people have also explored the use of multi-layered polypyrrole. Liang et al. made a novel three-layer-3D structure split-half-tubular polypyrrole@sulfur@polypyrrole composite cathode that prepared by means of chemical oxidation polymerization and chemical deposition [47]. Although its discharge capacity is not very high, this novel design breaks through the limitations of traditional cladding, and realizes the application of both inner and outer coating with conductive polypyrrole.

In fact, in my view, if we want to use polypyrrole, we should use it as a polymer for functions that the inorganic polymer can't achieve, such as strong inhibition of polysulfide shuttle effect. Because polypyrrole, even if it conducts electricity well, is not comparable to things like graphene and carbon nanotubes.

Zhang et al. made porous sulfur/polypyrrole/multiwalled carbon nanotube composite (S/PPy/MWCNT) by using an in-situ polymerization method for the aqueous suspension of polypyrrole monomer, nano-sulfur, and multi-walled carbon nanotubes [48]. Wei et al. synthesized a novel polypyrrole coated sulfur/multi-walled carbon nanotube (PPy@S/MWCNT) composite cathode for lithium-sulfur battery [49]. Wang et al. introduce a novel dual core--shell structured sulfur composite with multi-walled carbon nanotubes (MWCNTs) and polypyrrole (PPy), MWCNTs@S@PPy (Schematic diagram of the synthesis and discharge process of the dual-core shell structure MWCNTs @ S @ PPy composite is shown in Figure 3c.), as a cathode material for Li--S batteries via a facile one-pot method. In fact, they all use the polypyrrole to inhibit the shuttle of polysulfide, and the true electrical conductivity is based on better electrical conductivity of inorganic materials[50]. The realization of such double synergies is more advantageous to the realization of more usable lithium sulfur battery.

2.1.2. Separator

When it comes to the separator, it is easy to think of Celgard membrances. In most cases, the common diaphragm is semi-crystalline polyolefin, such as polyethylene or polypropylene. Although the price of these polymers themselves is only about $1-2/kg, the price of the commercially available membranes is $120-240/kg. This is mainly due to the complicated process required to carefully machine the pore structure on the

membrane. Although the membrane with micro-nano pore size is an inactive component in the battery, its importance is crucial. It is well known that the main function of the separator is to separate the anode and cathode, to ensure the passage of ions, and to block electrons. Modifications to the membrane range from modification of the membrane itself to individual functional second barrier layers, and it appears that the latter is more popular. It is not only simple, but also has a good improvement effect. In this review, I've included other functional interlayers in the diaphragm. Therefore, the application of polypyrrole in the lithium-sulfur cell diaphragm will be divided into two parts: the modification on the separator and the non-diaphragm functional layer.

The polypyrrole coating on the separator can not only inhibit the passage of polysulfide, but also cushion the expansion of sulfur volume in the discharge process, and polypyrrole as a conductive coating layer can also reduce electrode polarization.

Ma et al. used polypyrrole nanotubes and nanowires, reduced graphene oxide(rGO), as the diaphragm coating layer of the lithium-sulfur battery [52]. The result was that the polypyrrole modified diaphragm was more effective in inhibiting the migration of polysulfide than reduced graphene oxide. Moreover, ppy-modified diaphragm is also more permeable to electrolyte, and the battery corresponding to the membrane modified with polypyrrole nanotubes has an average coulomb efficiency of 90.6% in the electrolyte without $LiNO_3$. This indicates that the application of polypyrrole on the diaphragm side is conducive to the inhibition of polysulfide shuttle effect of lithium-sulfur batteries, which will be of great help to the improvement of cycle life in the application of lithium-sulfur batteries. Yin et al.prepared a new interlayer by coating the polypyrrole/ZnO composite slurry on the diaphragm (Figure 3d. is a schematic view of a battery with a PPy / ZnO interlayer) [51]. It makes use of PPy/ZnO composite coating as the diffusion absorber of polysulfide so as to achieve the inhibitory effect on the shuttle of polysulfide. Li et al.obtained a flexible free-standing lithium-sulfur battery by directly attaching the sulfur coated with the polypyrrole nano-fiber on the polypyrrole film [53]. The free-standing structure, which has no AL foil, is lighter than a flexible battery normally built with layering. This design also utilizes the polypyrrole to realize the inhibition of polysulfide shuttle and the relatively strong synergistic effect of polypyrrole on the adhesion of active substances, which provides the possibility to achieve a larger proportion of active substances.

Polypyrrole can also be used to modify the non-membrane functional layer, which can actually achieve the same effect with directly dealing with separator, and may be easier to implement. Wu et al. added a layer of polypyrrole treated carbon paper(CP) between the sulfur cathode and diaphragm to improve capacity retention and cycle life of the lithium-sulfur battery [54]. Due to the porous structure and conductivity of the treated polypyrrole coating, the battery with PPy-coated CP exhibits excellent cycling performance and better multiplier performance than the pure CP battery. In addition, they also quantitatively expressed the impact of carbon paper thickness on battery capacity and multiplier performance, which is of great value for the production and application of lithium sulfur batteries. Ma et al. self-assembled polypyrrole nanotube film(PNTF)using the simple method from PPy nanotubes [55]. The PNTF is sandwiched between the sulfur cathode and the separator, acting as a functional interlayer for the Li-S battery. Due to the adsorption effect between PPy and polysulfide lithium and the conductivity of PPy film, the polymer intermediate layer can not only significantly reduce the polarization of the sulfur cathode, but also effectively inhibit the shuttle effect and the redistribution of the active material during charging and discharging. Although these applications of polypyrrole in the lithium sulfur battery separator cannot eliminate the shuttle effect, they do a feasibility exploration for the practical application of the lithium sulfur battery, which is of guiding significance in promoting the long cycle under the high specific capacity of the lithium sulfur battery.

2.2. Polythiophene

Similar to polypyrrole, polythiophene is also a conductive polymer, and its conductivity is better than polypyrrole. Although the energy gap of polythiophene is small, its oxidation doped potential barrier is high, so its oxidation state is unstable in air and easy to be reduced to its intrinsic state. Therefore, polythiophene is easy to be reduced doped, especially 3 bits are easy to be introduced into side chains. Furthermore, the electrochemical properties of polythiophene vary greatly according to the side chains introduced. One of its derivatives, poly (3, 4-ethylenedioxythiophene) (PEDOT), is an important hole transport material.

Cathode: As for the cathode of li-sulfur battery, there is still a problem of poor conductivity of sulfur, and multiple sulfide is easily dissolved in electrolyte to form shuttle effect. The application of polythiophene in the cathode of li-sulfur battery also focuses on solving these two problems. Wu et al [56]. used in situ chemical oxidative polymerization to synthesize a core-shell sulfur/polythiophene composite cathode of lithium-sulfur battery with chloroform as solvent, thiophene as reaction reagent and ferric chloride as oxidant at 0 C. As a conductive additive and porous adsorbent, polythiophene is evenly coated on the surface of sulfur particles (Figure 4b. shows a schematic of the S-PTh composite.), which effectively guarantees the electrochemical performance and cycle life of lithium sulfur battery. This method requires a large number of solvents, the reaction conditions are not easy to achieve, and it is not easy to realize for practical application. However, as a new idea development, this method provides a choice for the cathode of li-sulfur battery, and the synthesized material structure is relatively stable, which is of guiding significance for the control of actual production conditions. Before this, wu et al. prepared the cathode of sulfur-polythiophene composite by a similar method [57]. Its synthesis conditions are more operable than those described above, and polythiophene is also used as conductive additive and porous adsorbent, which effectively improves the discharge capacity and cyclability of the battery.

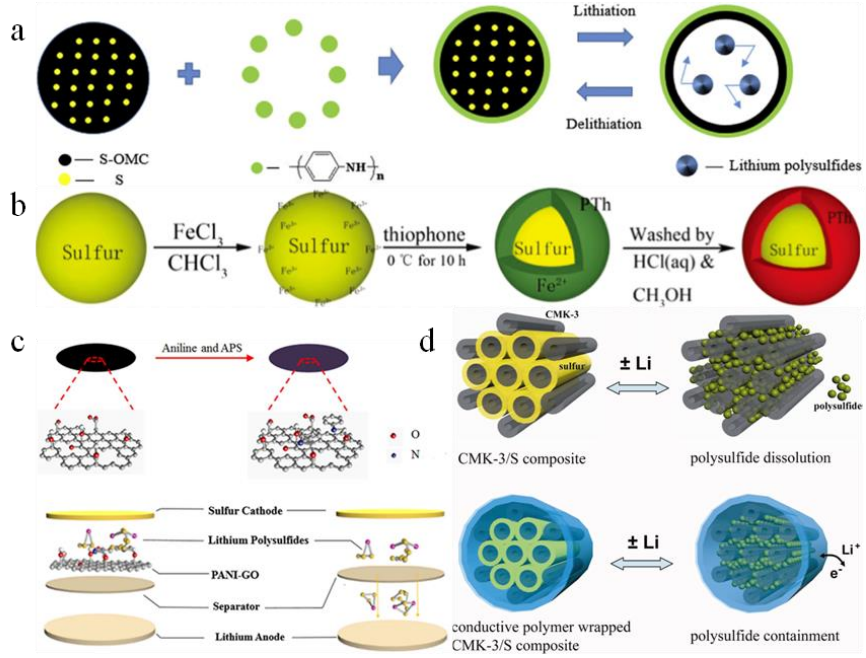

Figure 4. Schematic Diagram of the Structure: A. Schematic Illustration of the Fabrication Process of Polyaniline-coated Spherical Ordered Mesoporous carbon/Sulfur [65]. B. Synthesis of the S-PTh Composites [56]. C. Schematic of a Pani-go Interlayer Inserted between Cathode and Separator in Li-S Batteries [68]. D. Scheme of PEDOT:PSS-coated CMK-3/sulfur Composite for Improving the Cathode Performance [60].

From the point of view of chemical bond, all above mentioned should be a simple mixing of polythiophene and sulfur particles. Of course, the way in which polythiophene and sulfur particles are combined is not just a simple mix, but a way to bond them together.

Oschmann et al. first reported the co-polymerization of Grignard translocation derivatives of sulfur and allyl terminated poly(3-hexylthiophen-2, 5-dibasic) (P3HT) [58]. Moreover, the formation of C-S bond in copolymers was characterized by many methods such as NMR spectrum and X-ray fine absorption spectrum. This bond of S-P3HT may make the structure more reliable than a simple mixture of sulfur and P3HT. In my opinion, the most important thing is that it gives us a new way of thinking. If there is a framework that can implement the sulfur component without free movement, will there be shuttle effect? This will be of great benefit to the practical application of lithium-sulfur batteries. Xiao et al. designed a sandwich structure with vacuum filtration, in which nano-sulfur is evenly coated by graphene and poly(3,4-ethylene-dioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) [59]. This unique hierarchical design not only provides a highly conductive network but also the compact design itself enhances the close connection of the components and, more importantly, facilitates the physical and chemical synergies of the shuttle inhibition of polysulfide. This design achieves a high specific capacity of 701mAh/g at a large rate of 4C, and the flexibility of the structure design helps to promote its application in flexible energy storage devices. In order to limit polysulfide more effectively, the surface coating should be rigid and stable (As shown in Figure.4d, poly (3, 4-ethylenedioxythiophene) - poly (styrene sulfonate) (PEDOT: PSS) is a good choice based on these standards, as it is stable and rigid in an electrochemical environment.), but it should not be too hard to crack due to the expansion of sulfur volume during circulation [60]. And it should have both ionic and electron conductivity. Poly (3, 4-ethoxythiophene) - poly (styrene sulfonate) (PEDOT: PSS) is a good choice based on these standards, as it is stable and has moderate rigidity in an electrochemical environment. Chen et al [61]. reported the synthesis of ultrafine S nanoparticles with a diameter of 10-20 nm by membrane-assisted precipitation technique. The S nanoparticles are then coated with conductive poly(3,4-ethylenedioxythiophene) (PEDOT) to form S/PEDOT core/shell nanoparticles. The encapsulation of the conductive PEDOT shell limits the diffusion of polysulfides, reducing the self-discharge and shuttle effects, thereby improving cycle stability. The obtained S/PEDOT core/shell nanoparticles showed an initial discharge capacity of 1117 mAh/g and a stable capacity of 930 mAh/g after 50 cycles. This work is simple to operate, and the idea is clear, that is, the ultra-small size of the S nanoparticle is conducive to electrical conduction and improve sulfur utilization. This provides guidance for the practical application of lithium-sulfur batteries.

For the application of polythiophene in lithium-sulfur battery separators, more researchers are required to invest in it. This review will not introduce this part.

2.3. Polyaniline

The electrical activity of polyaniline is derived from the P electron conjugate structure in the molecular chain: with the expansion of the P electron system in the molecular chain, P bonding and P* antibonding forms valence band and conduction band respectively. This non-localized P electron conjugate structure can form p-type and n-type conducting state through doping, so that polyaniline presents high conductivity. Moreover, it is feasible to apply it to the cathode of lithium sulfur battery because of its structure is easy to produce proton acid substitution reaction.

2.3.1. Cathode

Polyaniline, as a kind of polymer with good conductivity, can greatly improve the poor conductivity of the sulfur cathode of li-sulfur batteries, which is conducive to obtaining better electrochemical performance.

Zhang et al. prepared polyaniline polysulfide (SPAn) as the cathode of the lithium-sulfur battery [62]. They first synthesized polyaniline by classical chemical oxidation method, replaced the hydrogen element on the six-membered ring with chlorine element of hydrochloric acid to prepare polyaniline chloride (CPAn), and finally replaced it with sulfur element in sodium sulfide and elemental sulfur. The chlorine on CPAn is removed to produce SPAn. This is a new way of thinking, which gives the control of polysulfide directly to polyaniline. However, the polysulfide shuttle effect comes from its solubility

in the electrolyte, so what is the solubility of this material in the supporting electrolyte? Further experimental results are needed. In comparison, polyaniline, which exists as a cladding layer, is more operable in practical application. Li et al. reported a novel composite, sulfur (S) encapsulated in porous carbon nanospheres (PCNS) and coated with conductive polyaniline (PANI) (PCNS-S@PANI), as cathode of lithium--sulfur battery [63]. Li et al. reported a sulfur coated with a conductive polyaniline under the control of ascorbic acid, where the sulfur is loaded on the curved graphene(CG) that is stripped from the multi-walled carbon nanotubes [64]. The CG-S@PANi composite was prepared to be used as cathode of lithium sulfur battery. Ding et al. designed a polyaniline@spherical ordered mesoporous carbon/sulfur nanocomposite (PANI@S-OMC/S), and S-OMC/S was wrapped by PANI via in-situ polymerization [65]. A schematic diagram of a process for preparing a polyaniline-coated spherical ordered mesoporous carbon/sulfur is shown in Figure 4a. The conductive polyaniline coating acts as a barrier layer, enhancing the electrical conductivity of the electrode while inhibiting the passage of polysulfide. The initial discharge capacity of the cathode at 0.1C is as high as 1626mAh/g, and the capacity is still 1338mAh/g after 100 cycles, which is higher than the initial capacity of many works. This shows that the coating is not only operational, performance may also be more uniform. Taking into account the different solubility of sulfur in liquid phase and at appropriate temperature, zou et al. prepared a kind of nonfilling polyaniline coating of sulfur/acetylene black composite by simple and environmentally friendly method [66]. Acetylene black particles with the particle size of 50 nm are partially infiltrated with sulfur, just the outer surface are coated with polyaniline shell. When used for lithium battery cathode material, it shows a significantly increase rate capability (initial capacity of 810 mAh/g at 2C , 486 mAh/g at 5C), and rate stability after 500 cycles(> 351 mAh/g at 5C). The excellent multiplier performance is due to the fact that the conductive polyaniline not only enhances the overall conductivity, but also physically blocks the "shuttle effect" between sulfur and electrolyte, which will hopefully make the lithium sulfur battery a real candidate for practical application.

2.3.2. Separator

As one of the core components in lithium-sulfur batteries, the separator not only can block the role of electrons, but also can suppress the rapid decay of lithium-sulfur battery capacity by modification, improve the utilization of active sulfur, and make lithium-sulfur batteries more practical.

Shi et al. designed a functional multi-walled carbon nanotube/sulfonated polyaniline (MWCNT / SPANI) modified separator to improve the electrochemical performance of Li-S batteries with high sulfur loading [67]. The MWCNT / SPANI coated membrane is highly selective for Li^+ but rejects the transport of polysulfide anions. In addition, the MWCNT / SPANI coating can also be used as an upper current collector and provides a conduit for electron and ion transport to increase sulfur utilization and ensure reactivation of the captured active material. With the ion-selective MWCNT / SPANI coated separator, the electrochemical performance of Li-S batteries based on cathodes with high sulfur loading (5 mg cm^{-2}) and high sulfur content (72 wt%) has been significantly improved. After 100 cycles, it provided a reversible capacity of 913 mAh/g at a current density of 100 mA/g, much higher than the cell using the original Celgard separator. This ion-selective MWCNT/SPANI modified diaphragm simultaneously meets the high sulfur loading and active material utilization ratio, which has a great promotion effect on the progress of lithium sulfur battery commercial diaphragm. Of course, a interlayer structure can also be made for diaphragm applications. Yin et al. designed a novel Li-S battery with a polyaniline-graphene oxide (PANI-GO) film sandwiched between a sulfur cathode and a separator (Figure.4c), which exhibits high cycle stability and improved rate performance [68]. The initial specific capacity was 1261 mAh/g at 0.5 C, and the capacity retention after 150 cycles was 73.0%. Excellent electrochemical performance thanks to the PANI-GO film not only can effectively reduce the diffusion of lithium polysulfide to the negative electrode through the chemical interaction between the anthracene group (-N =) of the

anthracene ring and polysulfide, but also It can increase the utilization of sulfur due to its conductive network. This design makes reasonable use of the synergy between the two components, but it is also difficult to guarantee the perfect contact between the interlayer itself and the diaphragm in the actual operation. Therefore, in my opinion, its practicability is not as good as directly modifying the diaphragm.

2.4. Polyacrylonitrile

Polyacrylonitrile (PAN) has a strong dipole interaction with lithium ions due to its highly polar(-C≡N) group on its molecular chain. Therefore, in the presence of polyacrylonitrile, the diffusion coefficient of lithium ions will increase, that is, the ionic conductivity is higher.

2.4.1. Cathode

As we all know, the cathode of lithium-sulfur battery has poor conductivity. However, the conductivity of polyacrylonitrile itself is not very good, so it is easy to think of a solution to surface coating with conductive carbon.

Peng et al [69]. prepared a carbon-coated sulfur/polypyrrole (C@S/PAN) core-shell structured composite via a novel solution treatment method. The design utilizes polypyrrole to inhibit the migration of polysulfide, and uses surface carbon layers to act as secondary electronic conductors and buffer the expansion of sulfur during charging and discharging. This method is simple and operability is strong. Similarly, K et al [70]. recently prepared a sulfur/polyacrylonitrile-PAN/acetylene black-AB composite by encapsulating polypyrrole and sulfur into the pores of acetylene black using a solution treatment technique. In the S/PAN/AB composite, AB acts as a conductive matrix while acting as a conductive matrix, and PAN acts as a flexible layer to buffer stress caused by volume changes during charge and discharge. This design can be said to be a synergistic application of the coating material and polyacrylonitrile, but the solution method has the drawbacks of solvent utilization.

In addition to solution processing techniques, many researchers have adopted pyrolysis methods. Compared to the solution method, pyrolysis plays a greater role in promoting industrialization.

Wang et al. prepared a conductive polymer-sulfur composite by heating a mixture of polyacrylonitrile powder and sublimed sulfur at 280-300 C under argon for 6 hours. In this design, sulfur and polyacrylonitrile are fused at the molecular level at 300 C, and sulfur acts as a dehydrogenating agent for polyacrylonitrile, resulting in a backbone similar to polyacetylene [71]. At the same time, the polyacrylonitrile crystals allow the sulfur to be immobilized due to the highly polar -CN functional group. The design itself is very good, it achieves the reaction to fix sulfur. According to this idea, Yin et al. designed a novel pyrolyzed polyacrylonitrile-sulfur@MWCNT composite via in situ polymerization of acrylonitrile on the surface of MWCNT, mixing with sulfur and final pyrolysis (A schematic diagram of in-situ polymerization of pPAN-S@MWCNT composite is shown in Figure 5b.) [72]. This design ensures that the sulfur atoms are embedded in the pyrolyzed polyacrylonitrile (PAN) matrix. In other words, the shuttle effect of sulfur is greatly reduced, greatly improving the utilization of sulfur. As a sublimation of the above work, Jin et al. recently proposed a conjugate double-bond lithium-ion storage mechanism [73]. Through solid-state NMR and XPS analysis, the authors found that in the discharge process, in addition to the reaction between sulfur and lithium, the C=N and C=C groups can also react with lithium to form Li-C-N-Li and Li-C-C-Li also provides capacity to produce a higher actual discharge capacity than the theoretical specific capacity. The specific discharge capacities after 2 and 100 cycles were 1729 and 1702 mAh/g, respectively, which were greater than the theoretical specific capacity of elemental sulfur (1675 mAh/g). The above results indicate that other mechanisms of lithium storage exist in addition to the reaction between sulfur and lithium during charging/discharging. The conjugate double-bond lithium-ion storage mechanism not only provides a very stable cathode material, but more importantly, it provides a new idea for the practical application of high-capacity electrode materials.

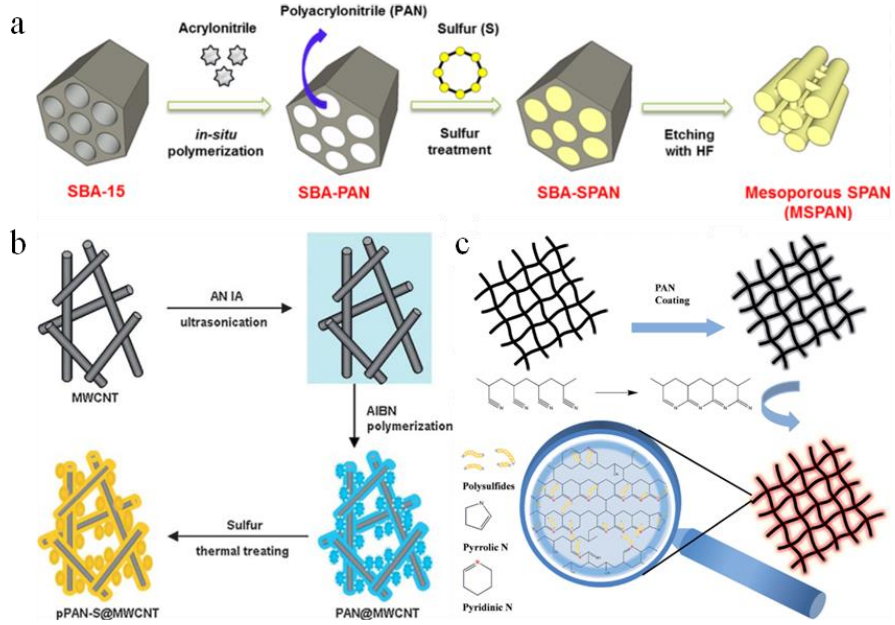

Figure 5. Schematic Diagram of Structure: A. An Illustration of the Sequential Manufacturing Steps of an Mspan Composite [77]. B. Schematic Representation of In-Situ Polymerization and Systematization of pPAN-S@MWCNT Composites [72]. C. Schematic Diagram of the Synthetic Chemical Structure of CPAN [81].

In addition to the use of pyrolysis methods, researchers have also developed, for example, vapor pressure in high pressure reactors to synthesize sulfurized polyacrylonitrile. Liu et al. synthesized a sulfurized polyacrylonitrile (S@pPAN) composite in a high pressure reactor under a certain vapor pressure [74]. They found that the hydrogen content and degree of graphitization in the S@pPAN composite decreased with increasing vapor pressure of the synthesis, and the appropriate vapor pressure helped to promote the reaction between polyacrylonitrile and sulfur so that the conductive molecular structure of sulfurized polyacrylonitrile was more complete. The sulfurized polyacrylonitrile composite (S@pPAN-5) prepared by them under the steam pressure of 5MPa has a high initial discharge capacity of 1821mAh/g, and the capacity remains at 1357mAh/g after 100 cycles. This study has noted the influential factors of pressure, which is instructive for the actual industrial production conditions of lithium-sulfur batteries, and reports on the similar discharge specific capacity of 1821 mAh/g are extremely rare.

The synthesis process of polymer-containing electrode materials can also be used in situ polymerization.

Zhang et al. prepared sulfur/polyacrylonitrile (S/pPAN) composites by using in-situ polymerization of acrylonitrile and nano-sulfur particles as cathode for lithium sulfur battery [75]. Thanks to the uniform distribution of sulfur and polyacrylonitrile at the nanoscale level, the structural stability of S/pPAN is guaranteed and space is provided for buffer volume expansion. Liu et al. synthesized a series of nitrogen-doped mesoporous carbon(CPANs) by in-situ polymerization of polyacrylonitrile in SBA-15 template and then carbonizing at different temperatures. In this study, it was found that the carbon matrix CPAN-800 synthesized by carbonization at 800 C has many desirable properties such as high specific surface area and pore volume, medium nitrogen content, and highly ordered mesoporous structure [76]. Therefore, it was used to prepare S/CPAN-800 composite material as a cathode of lithium-sulfur battery. In this design, the formation of a suitable temperature carbonized ladder structure is utilized to stabilize the structure and the nitrogen-doped carbon material enhances the ion-electron conductivity and electrolyte wetting ability of the lithium-sulfur electrode material. Later, Liu et al. prepared a highly ordered mesoporous sulfurized polyacrylonitrile (MSPAN) composite

by in-situ polymerization of polyacrylonitrile using the same SBA-15 template method. The sequential manufacturing steps of the MSPAN composite are shown in Figure 5a [77]. The capacity of MSPAN cell remains at 755mAh/g after 200 cycles at 1C and 610mAh/g after 900 cycles at 2C. This is attributed to the highly porous structure of MSPAN composite material, which can effectively improve the wettability, accessibility and absorbability of electrolytes and promote rapid ion transfer in li-s batteries. This highly ordered mesoporous MSPAN composite is a promising cathode active material for advanced Li-S batteries, and is of great significance for the application of high-rate lithium-sulfur batteries.

Apart from the poor conductivity of sulfur, there are also some disadvantages of low utilization of sulfur in the cathode. Zhang et al.prepared a sulfur/polyacrylonitrile/$Mg_{0.6}Ni_{0.4}O$ composite material via wet ballmilling as a cathode for a lithium sulfur battery. By adding $Mg_{0.6}Ni_{0.4}O$, from smooth S/PAN bulk particles to rough nanostructure aggregates, the morphology of the complex changes significantly and the specific surface area increases twice, which is conducive to the uniform reactivity of the composite components [78]. In other words, the additive increases the specific surface area of the reaction, which greatly increases the utilization rate of the active sulfur. Similar ideas can be developed gradually. Inorganic materials also have high dimensional stability, good heat resistance and easy dispersion, etc. If the cooperative application of polymer in lithium sulfur battery can be realized, this is of far-reaching significance for the practical application of lithium sulfur battery.

2.4.2. Separator

The separator mainly functions to isolate electrons, but there is a serious shuttling effect on the lithium-sulfur battery. Therefore, it is necessary to consider the function of blocking the polysulfide for the diaphragm. In particular, a highly polar material such as polyacrylonitrile can have a more pronounced effect on the modification of the membrane.

Li et al. prepared a porous carbon nanofiber (PCNF) coating on a glass fiber (GF) separator to block the diffusion of polysulfide using an immiscible blend of polyacrylonitrile (PAN) and poly(methyl methacrylate) (PMMA), which was a PCNF coated GF (PCNF@GF) separator for lithium-sulfur cell [79]. Compared to the cell with the pure GF membrane, discharge capacities of the battery with PCNF@GF-7:3 at the initial and 200th cycle can be increased by 165% and 196%. The PCNF coating serves as a good polysulfide reservoir here, thereby increasing the utilization of the active material and ensuring good cyclability. This again confirms that surface active groups are effective for inhibition of lithium polysulfide shuttling.

It is well known that the surface of graphene oxide is rich in organic functional groups. If it is used together with polyacrylonitrile for the membrane modification of lithium-sulfur batteries, will it be unexpected?

Zhu et al. reported a sustainable and highly porous polyacrylonitrile/graphene oxide (PAN/GO) nanofiber membrane separator, while providing large capacity and excellent anti-self-discharge capability for lithium-sulfur batteries [80]. In addition to benefiting from the highly porous structure of the nanofibrous membrane and excellent electrolyte wettability, the improved performance can also be attributed to the superior barrier effect caused by the relatively high energy binding between -C≡N and Li_2S/ polysulfide, as well as the electrostatic interaction between GO and negatively charged substances (Sn^{2-}). The design of this diaphragm structure is very helpful for the future practical application of lithium-sulfur batteries. Because the development of a large piece of lithium-sulfur battery depends on the improvement of the performance of the diaphragm, which is also beneficial for the protection of cycle life and safety.

Of course, polyacrylonitrile itself has a strongly polar -C≡N which can be converted into direct functional groups of lithium polysulfide under special circumstances. Li et al. reported a cyclized polyacrylonitrile-cast carbon nanofiber (CP@CNF) film as an interlayer in a Li-S battery [81]. By developing the CP@CNF interlayer (Figure.5c), the battery assembled with the bare sulfur cathode has excellent rate performance and cycle

stability. After 200 cycles at 0.3C, the reversible capacity can be maintained at 710 mAh/g, and a capacity of 560 mAh/g can be obtained even at a 2C rate. The improved performance is attributed to the abundant pyridine group in the cyclized polyacrylonitrile matrix, which can capture polysulfide by strong interatomic attraction, and a three-dimensional porous conductive network composed of a carbon nanofiber skeleton and a conjugated polymer matrix, resulting in efficient transfer of electron and ion pathways. Although this work seems simple, it also uses chemical bonds to enhance the adsorption of polysulfides, which theoretically guides the tendency of the bond species to adsorb polysulfides.

2.5. Else

Other polymers can also be applied to the cathode side of lithium-sulfur batteries. They may have a special surface functional group structure or a conductive network with excellent conductivity. Several other polymers will also be briefly described here.

Polydopamine, as a biorenewable polymer (structure containing nitrogen), is also of great interest to researchers. Deng et al. reported a simple method for preparing S@PDA (PDA=polydopamine) composites with core-shell structure and good electrochemical properties, as well as first-principles calculations for PDA and polysulfide interactions [82]. This structure makes full use of the advantages of the core shell structure with porous sulfur core. The high mechanical flexibility of PDA can adapt to the volume change during discharge/charging and good lithiumion conductivity. The enhanced interactions between nitrogen/oxygen atoms and lone electron pairs in PDA structures slow down the dissolution of polysulfide. And polydopamine is renewable, so the S@PDA electrode has great potential as a low-cost cathode of high-energy Li-S batteries.

The conductivity of polydopamine is inferior to the conductivity of carbon black, which is further sublimated as the above Deng work. Zhang et al. reported a sulfur/carbon black (S/C) cathode modified by self-polymerizing polydopamine (pDA) with the aid of a polymerization treatment [83]. As a novel and effective shell on the S / C cathode, pDA can block the shuttle effect of polysulfides. By enhancing the synergistic effect of conductivity and multiple blocking effects on polysulfides, the S/C @ pDA electrode exhibits improved electrochemical performance, including large specific capacity (1135 mAh/g at 0.2 C), high rate performance (533 mAh/g at 5 C) and long cycle life (965 mAh/g after 200 cycles). This design strategy may promote the development of high performance lithium-sulfur batteries.

In addition to polydopamine, polyurethanes that people often touch every day are also a good choice. Their structural controllability, elasticity and wear resistance are superior to other polymers, and they are also favored by researchers.

Xiao et al. reported a simple method for preparing nitrogen-doped porous carbon (NPC) from polyurethane foam (PUF) waste, which was impregnated with sulfur for use in lithium-sulfur batteries [84]. The NPC obtained has a unique interconnected sheet-like porous morphology with a large surface area of 1315 m^2/g. The NPC-S composite has a large capacity reversible capacity of 1118 mAh/g, with good cycle performance and excellent high rate performance. After 100 cycles at 5C (8.35 A/g), a reversible capacity of 460 mAh/g can be maintained. This work not only shows good electrochemical performance, but also it is also the reuse of waste resources, which gives us a good direction, adding a new possibility to reduce the application cost of lithium-sulfur batteries.

There are more and more other polymers involved in the application of the polymer in the lithium-sulfur cell membrane. In fact, as long as we understand the effect of introducing this modification, it is no longer very important to introduce what kind of polymer.

Zhang et al. used a polydopamine coated membrane as a separator to enhance the electrochemical performance of a lithium-sulfur battery [85]. The lithium-sulfur battery using the polydopamine coated separator showed a high specific capacity of 670 mAh g^{-1} after 200 cycles at 0.5 C, which is higher than the specific capacity of a battery having a

conventional separator. In this design, polydopamine is simply used as a physical barrier to inhibit the transfer of polysulfide.

Song et al. demonstrated a strategy for grafting poly (acrylic acid) (PAA) on the surface of a polypropylene (PP) separator to improve the electrochemical performance of Li-S cells by preventing the "shuttle effect" of polysulfide [86]. PP grafted with PAA (PP-g-PAA) separator allows Li^+ to migrate, while polysulfide anions are rejected by electrostatic repulsion. Therefore, the selective permeation separator can inhibit the polysulfide anions on the cathode side. In addition, the total reaction process of PP-g-PAA separator is easy and low cost. Due to the strong structural stability between PP and the grafted PAA, the multi-sulfide shuttles can be blocked uniformly and uniformly in the charge-discharge process. The battery using this diaphragm realized the ultra-low attenuation value of only 0.074% in each of the first 600 cycles. Song's work reported a simple and inexpensive diaphragm material that provided an opportunity to further control the cost of mass production of lithium-sulfur batteries.

Zhang et al. reported that the natural polymer Gum Arabic (GA) with precise oxygen-containing functional groups has a strong binding interaction with lithium polysulfide, which is deposited as a polysulfide shielding interlayer in carbon nanofibers (CNF) [87]. The obtained CNF-GA composite intermediate layer can achieve a high specific capacity of 880 mA h g^{-1} and an excellent performance of 827 mAh/g after 250 cycles under a sulfur load of 1.1 mg cm^{-2}. More importantly, a high reversible area capacity of 4.77 and 10.8 mAh cm^{-2} was obtained at high sulfur loads of 6 and 12 mg cm^{-2}. This result provides a simple and promising method for developing viable lithium-sulfur batteries with high sulfur loading and high reversible capacity.

3. Summary and Outlook

Lithium-sulfur batteries, as batteries with a mass energy density of 2,600 Wh/kg, are candidates for the most promising next-generation high-energy density batteries. Conductive polymers have a strong say in the case that lithium-sulfur batteries have poor conductivity of active substances and their discharge products, and the shuttle effect of polysulfide soluble in electrolytes leads to rapid attenuation of capacity. The problem of conductive polymers for poor conductivity and shuttle effect, in my opinion, depends mainly on the modification of the cathode side active material carrier and the membrane. Firstly, the conductive polymer has better conductivity. As the carrier material of the active material, whether it is coating, multi-layer, core shell, etc., the conductivity of the whole battery is significantly improved to some extent. Secondly, the conductive polymer acts as a sulfur carrier. While improving the overall conductivity, we should not ignore the advantages of its own structure. The controllability of its synthetic structure is better than the imposition of chemical bonds on other non-polymers. Therefore, it is known to contain such as oxygen-rich, Nitrogen-like functional groups help to inhibit polysulfide shuttles. Is it possible to solve this problem fundamentally? For example, SPAN is a good start; and then there is a diaphragm, which acts as an inactive component in the battery. On the basis of its own porous structural properties and its ability to be modified, we can artificially reprocess it, such as directly coating the membrane itself or as a separate intercalation. In summary, the application of conductive polymers on the cathode side of lithium-sulfur batteries is a systematic project. We can expect that lithium-sulfur batteries will achieve high conductivity and no shuttle in the future, which is crucial for solving human survival problems and energy dilemmas.

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