Journal of Electrochemistry

Volume 28
Issue 12 *Special Issue：In Honor of Professor Yu-Sheng Yang on the Occasion of His 90th Birthday (II)*

2022-12-28

Advances on Composite Cathodes for Lithium-Sulfur Batteries

Xi-Yao Li

Chang-Xin Zhao

Bo-Quan Li
2. Advanced Research Institute of Multidisciplinary Science, Beijing Institute of Technology, Beijing 100081, China;3. School of Materials Science and Engineering, Beijing Institute of Technology, Beijing 100081, China;, libq@bit.edu.cn

Jia-Qi Huang

Qiang Zhang
1. Beijing Key Laboratory of Green Chemical Reaction Engineering and Technology, Department of Chemical Engineering, Tsinghua University, Beijing 100084, China;, zhang-qiang@mails.tsinghua.edu.cn

Recommended Citation

Xi-Yao Li, Chang-Xin Zhao, Bo-Quan Li, Jia-Qi Huang, Qiang Zhang. Advances on Composite Cathodes for Lithium-Sulfur Batteries[J]. *Journal of Electrochemistry*, 2022 , 28(12): 2219013.
DOI: 10.13208/j.electrochem.2219013
Available at: https://jelectrochem.xmu.edu.cn/journal/vol28/iss12/2

电 化 学
J. Electrochem. 2022, 28(12), 2219013 (1 of 13)

[Review] DOI: 10.13208/j.electrochem.2219013 http://electrochem.xmu.edu.cn

Advances on Composite Cathodes for Lithium-Sulfur Batteries

Xi-Yao Li[1], Chang-Xin Zhao[1], Bo-Quan Li[2,3*], Jia-Qi Huang[2,3], Qiang Zhang[1*]
(*1. Beijing Key Laboratory of Green Chemical Reaction Engineering and Technology, Department of Chemical Engineering, Tsinghua University, Beijing 100084, China; 2. Advanced Research Institute of Multidisciplinary Science, Beijing Institute of Technology, Beijing 100081, China; 3. School of Materials Science and Engineering, Beijing Institute of Technology, Beijing 100081, China*)

Abstract: Lithium-sulfur (Li-S) batteries are deemed as high-promising next-generation energy storage technique due to their ultrahigh theoretical energy density, where the sulfur cathodes with high specific capacity guarantee the energy density advantage and directly determine the battery performances. After decades of exploration, the most promising sulfur cathodes are sulfur/carbon composite (S/C) cathodes and sulfurized polyacrylonitrile (SPAN) cathodes. In this manuscript, recent advances on S/C and SPAN cathodes in Li-S batteries are comprehensively reviewed. The electrochemical reaction circumstances on S/C and SPAN cathodes are firstly introduced and compared to reveal the working mechanisms of the two types of Li-S batteries. The S/C cathodes mainly undergo solid-liquid-solid multi-phase conversion processes with typical double-plateau charge-discharge polarization curves. In comparison, the SPAN cathodes follow solid-solid conversion and exhibit single-plateau charge-discharge characteristics. Following that, key challenges and targeted optimizing strategies of the S/C and SPAN cathodes are respectively presented and discussed. For Li-S batteries with S/C cathodes, the main optimizing strategies are electrode structure modification, efficient electrocatalyst design, and redox comediation. For SPAN cathodes, the main optimizing strategies are electrode structure modification, morphology regulation by co-polymerization, heteroatom doping at molecular level, and extrinsic redox mediation. At last, current research status of Li-S batteries with S/C or SPAN cathodes are systematically analyzed through the comparison of several battery parameters, and perspectives on challenges and opportunities of S/C and SPAN cathodes in Li-S batteries are presented to guide future researches.

Key words: lithium-sulfur battery; sulfur/carbon composite cathode; sulfurized polyacrylonitrile cathode; lithium polysulfide

1 Introduction

The global rapid energy consumption drives massive application of clean energy and the development of energy storage techniques[1-3]. Electrochemical energy storage devices, represented by rechargeable batteries, are widely applied in personal electronics, electric vehicles, and smart grids[4,5]. Energy density of rechargeable batteries is regarded as one of the most significant parameters[6,7]. Among the various battery systems, lithium-sulfur (Li-S) batteries possess ultrahigh energy density of 2600 $\mathrm{Wh \cdot kg^{-1}}$ in theory, which is 10 times higher than the current commercial lithium-ion batteries[8,9]. Additionally, sulfur is abundant in resources and cheap in price. The above advantages endow Li-S batteries as one of the most promising next-generation energy storage techniques[10].

Typical Li-S batteries are composed of lithium metal anodes, ether-based or carbonate-based electrolyte, and sulfur composite cathodes[11]. Notably, the theoretical specific capacity of the sulfur cathode is

Cite as: Li X Y, Zhao C X, Li B Q, Huang J Q, Zhang Q. Advances on composite cathodes for lithium-sulfur batteries. *J. Electrochem.*, 2022, 28(12): 2219013

Received: 2022-12-08, Revised: 2022-12-02. *Corresponding author, Tel: (86-10) 62789041, E-mail: zhang-qiang@mails.tsinghua.edu.cn (Zhang Q.); Tel: (86-10)68911612, libq@bit.edu.cn (Li B.-Q.)

as high as 1672 mAh·g^{-1}, which guarantees the ultra-high energy densities of Li-S batteries[12,13]. In this way, the performance determining factor of Li-S batteries mainly lies in the cathodic performance of the sulfur active materials[14]. Therefore, developing advanced sulfur cathodes is the key for constructing high-performance Li-S batteries. After decades of exploration, the most promising sulfur cathodes are sulfur/carbon composite (S/C) cathodes and sulfurized polyacrylonitrile (SPAN) cathodes[15], and the markable events during the development of the above two composite cathodes are depicted in Figure 1. Researches on these two cathodes have made significant breakthrough very recently. For instance, Cheng et. al. constructed 700 Wh·kg^{-1} rechargeable Li-S pouch cells based on the S/C cathodes[16]. Chen, Liu, and coworkers realized stable cycling of Li-S pouch cells under −60 ℃ based on the SPAN cathodes[17]. To this end, a systematical review on S/C and SPAN cathodes in Li-S batteries is essentially required to summarize the recent advances and provide insightful perspectives for future development.

Herein, recent advances on S/C and SPAN cathodes in Li-S batteries are comprehensively reviewed. The electrochemical reaction circumstances on S/C and SPAN cathodes are firstly introduced and compared to reveal the working mechanism of the two types of Li-S batteries. Key challenges and corresponding optimizing strategies of the current S/C and SPAN cathodes are then respectively presented and discussed. At last, current research status of Li-S batteries with S/C or SPAN cathodes are systematically analyzed through the comparisons of several battery parameters, and perspectives on challenges and opportunities of S/C and SPAN cathodes in Li-S batteries are presented to guide future researches.

2 Working Mechanism of S/C and SPAN Cathodes in Li-S Batteries

2.1 Working Mechanism of S/C Cathodes

Since elemental sulfur (S_8) and its reduction solid products (i.e., lithium sulfide, Li_2S) are electronically insulating in nature, the electrochemical reactions of sulfur are highly dependent on the substrate conductive carbon materials[18]. Therefore, typical S/C cathodes are composed of nanosized sulfur and conductive carbon after ball-milling and heat melting. Wang et. al. firstly reported the S/C composited material in 2002 and applied it as the cathode in Li-S batteries[19]. In 2009, Nazar's group made a breakthrough by developing highly ordered nanostructured carbon to accommodate S_8[20]. Since then, S/C cathodes started their booming development in aspects of carbon optimizations, electrode structural modifications, polar interface regulations, etc.

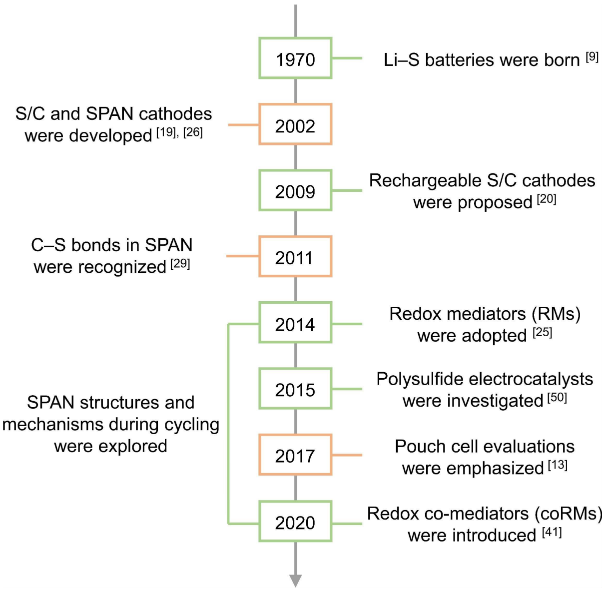

Figure 1 A brief timeline about the development of S/C and SPAN cathodes in Li-S batteries.

Notably, the working mechanism of typical S/C cathodes obeys the solid-liquid-solid reaction pathway (Figure 2a). Concretely, taking the discharge process as an example, solid S_8 is reduced into soluble lithium polysulfides (LiPSs, Li_2S_n, $n = 3 \sim 8$) dissolving in electrolyte in the first place, and the LiPSs then gain electrons and convert into solid Li_2S[21]. Based on the solid-liquid-solid reaction pathway, the sulfur species are not fixed on the S/C cathodes but "mobile" in electrolyte[22]. In this way, their diffusion and reaction behaviors in bulk electrolyte and in S/C cathodes require equally attention during electrode and electrolyte design[23]. The double-plateau discharge profiles further reflect the solid-liquid-solid reaction pathway (Figure 2b). Concretely, the first plateau from 2.4 V to 2.1 V represents the conversion process from solid S_8 to soluble LiPSs, and the second plateau around 2.1 V represents the conversion process from LiPSs to solid Li_2S[24,25]. In brief, the working mechanism of S/C cathodes obeys the solid-liquid-solid reaction pathway, thus not only the cathode itself but also solvated LiPSs in electrolyte play key roles in the cathodic redox reactions and determining the battery performances.

2.2 Working Mechanism of SPAN Cathodes

The SPAN cathode is firstly reported by Wang et. al. in 2002[26]. Concretely, the SPAN powder is synthesized by mixing sulfur and PAN under a weight ratio of 4:1 with subsequent ball-milling and furnace heating[27]. It is widely deemed that polycyclic hexatomic rings containing carbon and nitrogen atoms are generated in SPAN, and sulfur atoms serve as side groups and connect these polycyclic hexatomic rings[28]. During the first discharge process, S—S bonds and some C—S bonds break, and the conjugated structure in SPAN can partially storage Li^+ [29]. There are several irreversible reactions taking place in the first discharge cycle of the SPAN cathode, during which the SPAN structure is reformed and the SPAN cathode is activated[30]. Once the above activation process is finished, the SPAN cathode can reversibly cycle during long-lasting charge-discharge processes.

Worthy to be noted, the working mechanism of SPAN cathodes obeys the solid-solid reaction pathway (Figure 2c), which is quite different from the solid-liquid-solid reaction pathway for S/C cathodes [31]. Some previous researches deemed that a robust cathode-electrolyte-interphase (CEI) layer is formed through reactions between the sulfur species and the solvents, which can inhibit subsequent dissolution of sulfur[32]. As a result, soluble LiPSs will not be generated during the charge-discharge processes. The charge-discharge profiles of Li-S batteries with SPAN cathodes are also quite different from the ones with S/C cathodes (Figure 2d). A typical single plateau around 1.9 V is exhibited during the charge and discharge processes, suggesting that Li^+ intercalation reactions rather than phase conversion reactions occur[33]. Though the actual structure of SPAN is still inexplic-

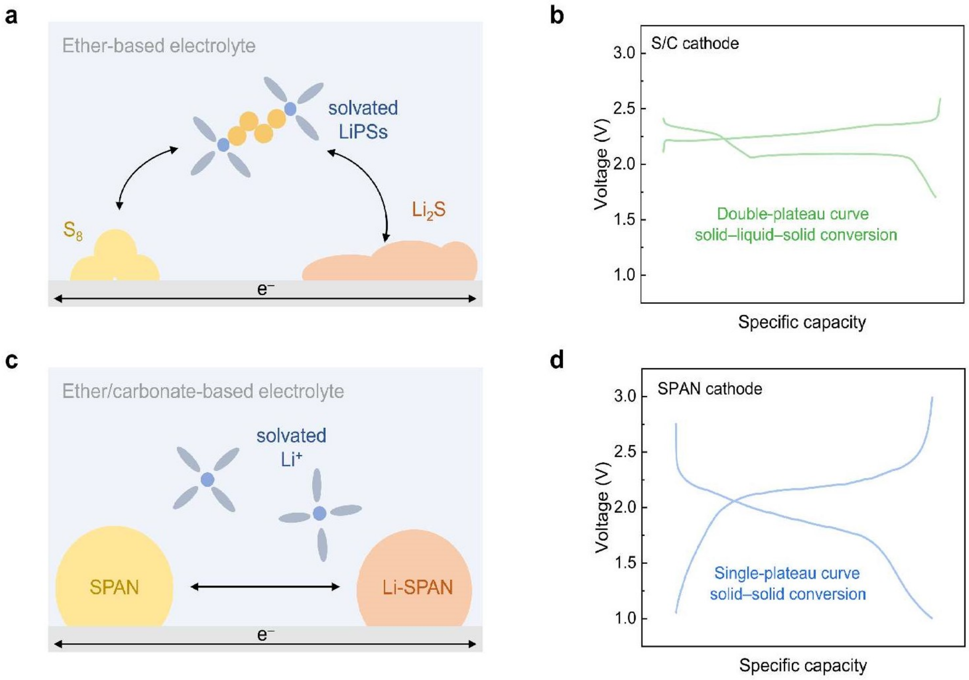

Figure 2 Schematic illustration of cathodic reactions and charge-discharge profiles in Li-S batteries with (a)-(b) S/C cathodes or (c)-(d) SPAN cathodes.

it, it can be assured that the capacity for SPAN cathodes is provided by the reversible breaking and generation of S—S bonds[34]. In brief, the working mechanism of SPAN cathodes obeys the solid-solid reaction pathway between SPAN and lithiated SPAN (Li-SPAN), and therefore the structure and functions of SPAN itself constitute to be the research focus.

3 Challenges and Optimizing Strategies for S/C Cathodes

The main challenge for S/C cathodes is the sluggish kinetics especially under harsh working conditions (e.g., large current densities, high sulfur loading and content, lean electrolyte, etc.)[35,36]. The sluggish cathodic kinetics further render unbearable polarization and low specific capacity in Li-S batteries [37]. Since LiPSs serve as the key intermediates in regulating the performances of S/C cathodes, both the LiPSs diffusivity and reactivity at the cathodic interface are taken into consideration to tackle the kinetic challenges[38]. In detail, the LiPSs diffusion pathway can be tuned by optimizing the electrode structure (e.g., porosity, specific surface area, tortuosity, tap density, etc.), and the LiPSs reactivity can be regulated by introducing polar hosts and electrocatalysts. Additionally, redox mediation and comediation strategies can improve the reactivity and diffusivity at the same time. Detailed discussion on these three strategies is presented in Figure 3.

3.1 Electrode Structure Modification

Previous literatures have reported that introducing high specific area materials can provide abundant reaction sites for the sulfur redox reactions, and modifying the pore structure can regulate the LiPSs and Li^+ diffusion properties[42,43]. Combining the above two methods together, Zhang et. al. adopted an ant-nest-like hierarchical porous carbon with high specific surface area and regulated pore structure in S/C cathodes[44]. As a result, both the polarization and the specific capacity were greatly enhanced, and an ultrahigh area capacity of 11.6 $mAh\cdot cm^{-2}$ at a high sulfur loading of 10 $mg_S\cdot cm^{-2}$ could be achieved. Nevertheless, carbon materials with high specific area and porosity inevitably make the S/C cathode taking up much electrolytes, which is unbeneficial especially under lean electrolyte conditions[45]. Recently, Lu and co-workers balanced the porosity, specific area, and tortuosity of S/C cathodes, and demonstrated a dense single-particle-layer cathode (Figure 3a)[39]. Concretely, compared with the small S/C particles, relatively large S/C particles on cathode can promote electrolyte penetration and restrain soluble LiPSs out of the cathodic pores, which efficiently reduces the capacity loss during cycling. As a result, Li-S cells with the dense single-particle-layer cathode exhibited superior capacity over 1000 $mAh\cdot g^{-1}$ even under lean electrolyte conditions (electrolyte to sulfur (E/S) ratio

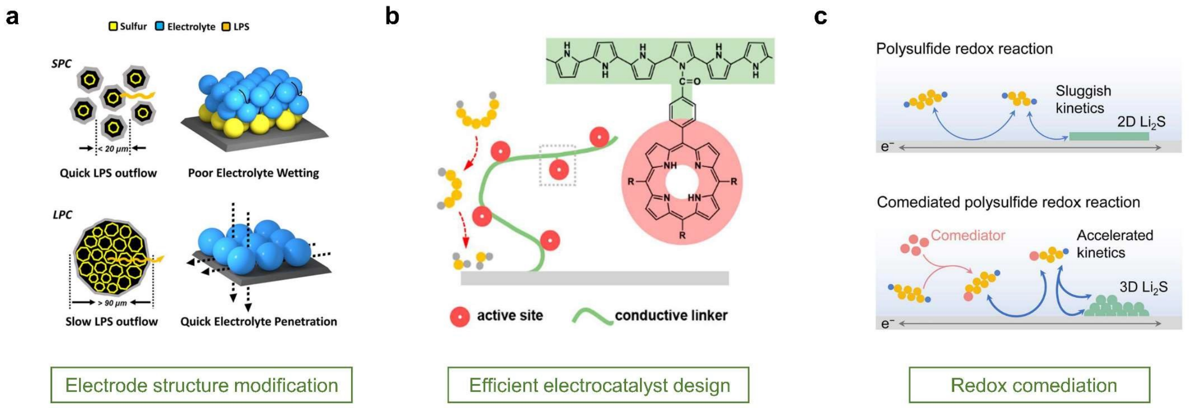

Figure 3 Optimizing strategies for S/C cathodes. (a) Electrode structure modification[39]. Reproduced with permission from Ref [39]. Copyright 2022 Royal Society of Chemistry. (b) Efficient electrocatalyst design[40]. Reproduced with permission from Ref [40]. Copy right 2021 American Chemical Society. (c) Redox comediation[41]. Reproduced with permission from Ref [41]. Copyright 2020 Elsevier.

= 4 μL·mg^{-1}). Other electrode structure modification approaches such as designing S/C shell structures and network structures also obtained favorable effects[46,47]. In brief, the porosity, specific surface area, and tortuosity are three significant parameters to be considered during S/C cathode design, and the S/C cathodes can exhibit superior performances only when the above three parameters are well balanced (Table 1).

3.2 Efficient Electrocatalyst Design

The purpose of introducing electrocatalysts to S/C cathodes is to strengthen LiPS adsorption and facilitate interfacial charge transfer[48]. Previous researches have proved that electrocatalysts such as transition metal compounds, hetero-doped carbon, and their composites can efficiently reduce the reaction activation energy and promote the S/C cathode performances[49,50]. Since the solid-liquid-solid reaction pathway inevitably results partial soluble LiPSs far away from the cathodic interface and difficult to participate in electrode reactions, expanding the interaction range of electrocatalysts is essential. Taking the above aspect into considerations, Zhao et. al. designed a semi-immobilized electrocatalyst by grafting the active sites (porphyrin) onto soft segments (polypyrrole) (Figure 3b)[40]. Consequently, the semi immobilizing strategy in practical Li-S batteries realized superior rate performances as well as long lifespan, and it also endowed pouch cells with high energy density up to 343 Wh·kg^{-1}. Another efficient design is to combining electrocatalysts with different functions together. For example, Yang and co-workers demonstrated that metal oxides (e.g., TiO_2) are effective in adsorbing LiPSs onto the cathodic interface, while metal nitrides and sulfides (e.g., TiN and Ni_3S_2) are expert in converting LiPSs[51,52]. Based on the above discovery, they composited the adsorption-type and conversion-type electrocatalysts together, and endowed Li-S batteries with superior cycling stability for 3000 cycles. Additionally, since the LiPS electrocatalysis procedure happens on the electrocatalyst surface, the surface variation of electrocatalysts should be specially acknowledged. For instance, the Co_4N surface was sulfurized into CoS_x phases during initial cycles[53], the Mo_6S_8 surface got lithiation into $Li_xMo_6S_8$ during the discharge process[54], and the MoS_2 surface was covered by a gel layer due to the solvent simultaneous polymerization[55]. The above surface reconstruction behaviors inevitably affect the LiPS electrocatalysis mechanisms and effects. In brief, electrocatalysts are introduced to overcome the sluggish charge transfer kinetics, and their design should fully consider the interfacial issues as well as the solidliquid-solid reaction pathways in Li-S batteries.

3.3 Redox Comediation on LiPSs

Different from the electrode structural modification strategy overcoming the LiPSs diffusion issues and the electrocatalysis strategy promoting the LiPSs redox reactivity, the redox comediation strategy can accelerate the reactivity and diffusivity of LiPSs on S/C cathodes at the same time. In detail, organic disulfides and diselenides can serve as redox comediators (co-RMs) to spontaneously react with LiPSs in Li-S batteries, afford an additional chemical reaction pathway besides the original electrochemical pathway, and promote the overall sulfur redox kinetics[56,57]. For instance, Zhao et. al. introduced di-tertiary butyl disulfide (DtbDS) into Li-S batteries as a co-RM (Figure 3c)[41]. Concretely, the S—S bond in DtbDS breaks spontaneously and the sulfur chains in LiPS molecules are subsequently intercalated into the DtbDS molecule to form modified LiPS molecules grafted with organic groups (*i.e.*, tb-S_{n+1}Li). Both the diffusivity and the reactivity of the modified LiPSs are higher than those of the pristine LiPSs, which endow the S/C cathode with reduced polarization, promoted discharge capacity, and superior rate performances even under lean electrolyte (E/S ratio = 5 μL·mg^{-1}) and high sulfur loading (5.0 mg_S·cm^{-2}) conditions. Compared with sulfur, selenium usually possesses higher redox reactivity. As a result, diselenides are also considered as promising co-RMs[58]. By adopting the mixture of diphenyl diselenide (Ph-Se) and dimethyl diselenide (Me-Se), the cathodic kinetics were greatly enhanced considering polarization and discharge capacity, and an actual energy density of 384 Wh·kg^{-1} was achieved in working Li-S pouch cells[59]. In brief, the redox comediation strategy is impressively effective in accelerating both the diffusivity and the reactivity of LiPSs, and it can promote full-range sulfur redox kinetics on S/C cathodes.

4 Challenges and Optimizing Strategies for SPAN Cathodes

Main challenges for SPAN cathodes are kinetic issues as well. However, different from the S/C cathodes, the optimizing strategies do not aim at soluble LiPSs but rather SPAN itself. Concretely, the kinetic issues are originated from the poor electronic conductivity of micrometer-sized SPAN powders (around 10^{-9} S·cm^{-1})[66]. Meanwhile, volume variation during cycling destroys the structural stability of SPAN cathodes, which may further aggravate the cathodic kinetics issues. Additionally, in ether-based electrolyte, part of the sulfur species may be dissolved into the electrolyte, impair the SPAN kinetics, and reduce the cathodic cycling stability[67]. Aiming at the above challenges, major optimizing strategies are proposed, including electrode structure modification, morphology regulation by co-polymerization, heteroatom doping at molecular level, and extrinsic redox mediation (Figure 4).

4.1 Electrode Structure Modification

Since the electronic conductivity of SPAN is relatively low, it is promising to modify electrode structures by introducing high-conductive carbon materials and constructing efficient ionic and electronic pathways. For instance, Yin et.al. introduced oxidized graphene (GO) into the SPAN cathodes to make the SPAN powders evenly dispersed on the conductive network (Figure 4a)[68]. As a result, Li-S cells with the GO composited SPAN cathodes could cycle stably and exhibit a specific capacity of nearly 900 mAh·g^{-1} even under a high rate of 6.0 C (1 C = 1672 mA·g^{-1}). However, GO with high specific area always takes up much electrolyte and is incompatible with lean electrolyte conditions. To this end, electrospinning processing is reported to compensate the electronic conductivity and tolerant low electrolyte volume at the same time. For example, Wang et al. mixed carbon nanotubes (CNTs) and PAN together through the electrospinning method, and then sulfurized them under high temperatures[64]. Consequently, the SPAN exhibited a nanosized slice morphology on the CNTs, which endowed the Li-S cells with a specific capacity of 1180 mAh·g^{-1} without capacity fading over 1000 cycles. In brief, electrode structure

Table 1 Summary of advanced Li-S battery performances with S/C or SPAN cathodes.

	Component	Sulfur loading (mg$_S$·cm^{-2})	Specific capacity (mAh·g^{-1})	Cycling lifespan	Reference
S/C cathode	G@ppy-por	5.0	940 @ 0.2 C	70 @ 0.2 C	Zhao et al.[40]
	MoS$_2$ with TEA	4.0	988 @ 0.3 C	100 @ 0.3 C	Li et al.[37]
	DPDSe	5.0	924 @ 0.1 C	55 @ 0.1 C	Zhao et al.[58]
	LPC	4.0	1001 @ 0.1 C	100 @ 0.1 C	Feng et al.[39]
	7TiN:3TiO$_2$-G	1.2	800 @ 1.0 C	2000 @ 1.0 C	Zhou et al.[51]
	Co$_4$N/NG	4.1	1109 @ 0.5 C	150 @ 0.5 C	Zhao et al.[53]
SPAN cathode	Se$_{0.06}$SPAN	1 ~ 3	1240 @ 0.26 C	800 @ 0.26 C	Chen et al.[60]
	BEAQ	1.5	1109 @ 1.0 C	160 @ 1.0 C	Zhao et al.[61]
	Fibrous SPAN	0.672	600 @ 4.0 C	1000 @ 4.0 C	Frey et al.[62]
	SPAN-CNT20	0.9 ~ 1.1	1106 @ 1.0 C	500 @ 1.0 C	Razzaq et al.[63]
	SPAN/CNT-12	2.0	1180 @ 0.48 C	1000 @ 0.48 C	Wang et al.[64]
	Te$_{0.04}$S$_{0.96}$@pPAN	3.11	870 @ 0.12 C	100 @ 0.12 C	Li et al.[65]

modification mainly concentrates on the SPAN conductivity enhancement. Compatibility with high sulfur content and lean electrolyte shall be taken into consideration.

4.2 Morphology Regulation by Co-Polymerization

One reason for low conductivity of SPAN cathodes is that the SPAN particles are rather large in size (in several micrometers)[69]. Therefore, reducing the SPAN size is promising. Co-polymerization between PAN and other polymers is expected to regulate the SPAN morphology. Based on the above thoughts, Frey et al. chose the poly(methyl methacrylate)/PAN (PMMA/PAN) fiber as the precursor to synthesize SPAN (Figure 4b) [62]. Microscopic images suggested that the as-obtained SPAN exhibited the fibrous morphology with around 200 nm in diameter, much smaller than the SPAN particle. Additionally, the fibrous morphology with cylindrical macropores formed eletronic conduction networks and provided directed Li^+ diffusion pathways. As a result, Li-S cells showed high rate capability up to 8.0 C and excellent cycling stability over 1200 cycles. In brief, regulating the SPAN morphology and reducing their size through altering the precursor composition by co-polymerization is effective to compensate conductivity and tackle kinetic issues.

4.3 Heteroatom Doping at Molecular Level

Elemental doping, especially selenium (Se) and tellurium (Te) doping, is effective in regulating the chemical and electrochemical properties of SPAN cathode materials[70]. On the one hand, Se and Te doping can enhance the electronic conductivity of SPAN. For instance, Li et al. doped 4at% Te into SPAN and manifested that the electronic conductivity increased from 2.0×10^{-9} to 3.5×10^{-9} $S\cdot cm^{-1}$ after Te doping (Figure 4c)[65]. As a result, the Te-doped SPAN cathode was compatible with both ether and carbonate

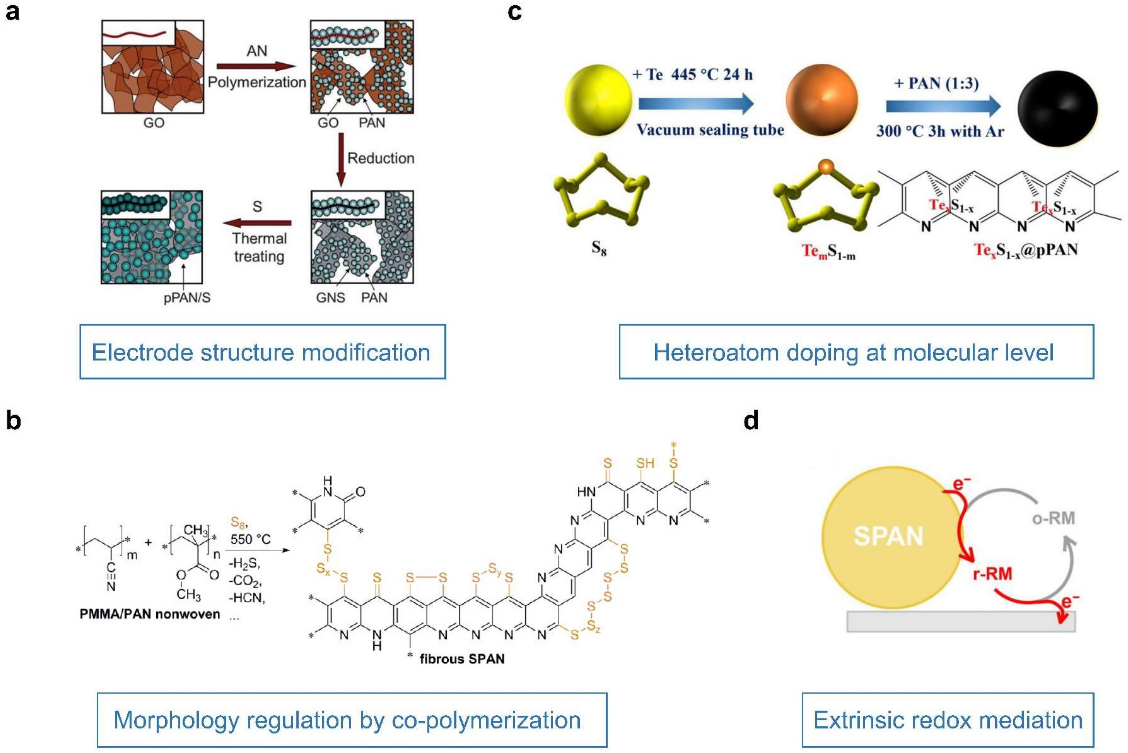

Figure 4 Optimizing strategies for SPAN cathodes. (a) Electrode structure modification[68]. Reproduced with permission from Ref [68]. Copyright 2012 Royal Society of Chemistry. (b) Morphology regulation by co-polymerization[62]. Reproduced with permission from Ref [62]. Copyright 2017 American Chemical Society. (c) Heteroatom doping at molecular level[65]. Reproduced with permission from Ref [65]. Copyright 2019 Elsevier. (d) Extrinsic redox mediation[61]. Reproduced with permission from Ref [61]. Copyright 2020 Wiley.

electrolytes, and delivered high capacities of 1507 and 861 mAh·g^{-1} at 0.1 and 10 A·g^{-1}, respectively. On the other hand, Se and Te doping in SPAN can generate several Se/Te-S bonds with enhanced electrochemical reactivity[71]. Chen et al. introduced 6at% Se into SPAN cathode and found that the irreversible sulfur loss due to sulfur dissolution in ether-based electrolyte is greatly relieved[60]. The above results indicate that the cathodic reaction rates are greatly accelerated and the capacity decay is inhibited. Consequently, the Se-doped SPAN cathodes can undergo 800 cycles with nearly 100% Coulombic efficiency and an ultralow 0.029% capacity decay per cycle. In brief, heteroatom doping can improve the electronic conductivity and electrochemical reactivity of SPAN cathodes at the same time, resulting in greatly enhanced cathodic kinetics and battery performances.

4.4 Extrinsic Redox Mediation

The morphology of SPAN usually exhibits bulk particles with several micrometers in diameter, which inevitably results in long electronic conduction distance and large contact resistance[63]. From this point of view, extrinsic redox mediation is expert in handling the poor electronic conductivity by coupling a chemical pathway on the original electrochemical pathway. Zhao et al. adopted the 1,5-bis(2-(2-[2-methoxyethoxy]ethoxy)ethoxy)anthra-9,10-quinon (BEAQ) as an extrinsic redox mediator, and the redox potential of BEAQ is quite compatible with that of SPAN (Figure 4d)[61]. Concretely, the BEAQ oxidized redox peak (2.46 V) is slightly higher than the SPAN oxidization peak (2.42 V), illustrating that the delithiation of Li-SPAN by the oxidation of BEAQ is thermodynamically feasible. Consequently, the redox-mediation-assisted SPAN cathodes exhibited reduced polarization, higher specific capacity, improved rate performance, and longer cycling lifespan in both ether-based and carbonate-based electrolytes. In brief, the redox mediation strategy promises to overcome the poor conductivity issue and enhance the cathodic kinetics, while researches in this field require to pay more attention.

5 Conclusions and Outlooks

The Li-S batteries with S/C or SPAN cathodes exhibit distinct characteristics (Figure 5). In detail, compared with Li-S batteries with SPAN cathodes, Li-S batteries with S/C cathodes are relatively advantageous in output voltage, superior in energy density, and predominant in sulfur content. However, Li-S batteries with SPAN cathodes exhibit fantastic lean-electrolyte tolerance, higher rate capability, and generally longer stability. Note that above arguments are based on the average level according to reported literatures. Therefore, Li-S batteries with S/C or SPAN cathodes face different challenges, and the future research directions shall be different. Key challenges and opportunities of S/C and SPAN cathodes in Li-S batteries are respectively presented as follows.

For S/C cathodes, there are mainly three aspects of challenges: kinetics issues under high rates, kinetics issues under low E/S ratios, and cathodic incompatibility with Li metal anode as well as electrolyte. The first two kinetic issues are due to the intrinsic solid-liquid-solid reaction pathway, which renders concentrated soluble LiPSs with limited charge transfer rates and low diffusion rates dominating the cathodic interfacial kinetics. Therefore, both mechanism investigation and kinetic promoters are specially needed to address this issue. The incompatibility with Li metal anode is originated from the soluble LiPSs generating at cathode, diffusing to the anodic compartment, corroding the Li metal, and rendering rapid anode failure. Meantime, the incompatibility with electrolyte refers to limited selection of the electrolyte component due to the requirement of dissolving LiPSs. Therefore, exploring the solvating properties especially the solvation structure of LiPSs in electrolyte is essential to balance the cathodic and anodic performances, and overcome the cathodic incompatibility with Li metal anode as well as electrolyte.

For SPAN cathodes, key challenges mainly exist in the following three aspects: low sulfur content, unclear chemical structure, and sulfur dissolution in ether-based electrolyte. To increase the sulfur content, precursor modification and manufacturing optimiza

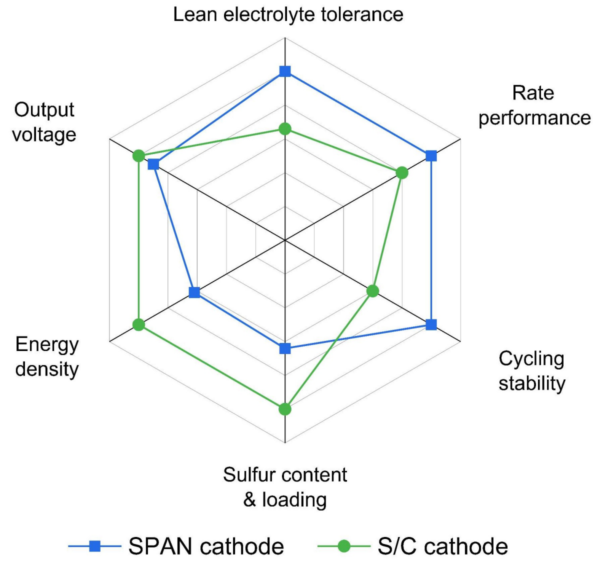

Figure 5 Comparison of Li-S batteries with S/C or SPAN cathodes.

tion are two effective approaches. In detail, tuning the precursor composition and structure may alter the PAN crystallinity, and make more elemental sulfur reacting with the PAN precursor. Optimizing the manufacturing process (e.g., introducing electrospinning technique), on the other hand, may reduce the amount of conductive carbon and increase the sulfur content in turn. Due to the unclear chemical structure, researches on SPAN cathodes may be confused at molecular-level structure design. Therefore, *in situ* spectroscopies are expected to probe the specific bonds and their dynamic variation during charge and discharge processes. To suppress sulfur dissolution in ether-based electrolyte, explorations on cathodic interfacial changes during cycles should be firstly conducted. Optimizing the electrolyte components by introducing some additives to generate robust CEI with high ionic conductivity is also promising.

In summary, Li-S batteries with S/C cathodes and SPAN cathodes are very promising for energy storage based on the great progresses made in the recent years. For future application of the two cathode techniques, Li-S batteries with S/C cathodes are expected to address high-energy-density energy storage circumstances such as spacecrafts, while Li-S batteries with SPAN cathodes are promising for large-scale energy storage due to their advantages of low costs and long cycling lifespan. Witnessing the recent progresses of S/C and SPAN cathodes for Li-S batteries, researchers are full of confidence on the development of advanced Li-S batteries.

Acknowledgements:

This work was supported by Natural Scientific Foundation of China (22109007), Beijing Natural Science Foundation (JQ20004), Beijing Institute of Technology Research Fund Program for Young Scholars, and the Tsinghua University Initiative Scientific Research Program. We thank Dr. Wei-Jing Chen and Dr. Xue-Qiang Zhang for the helpful discussion.

References:

[1] Shen L, Song Y W, Wang J, Zhao C X, Bi C X, Sun S Y, Zhang X Q, Li B Q, Zhang Q. Synergistic catalysis on dual-atom sites for high-performance lithium-sulfur batteries [J]. Small Struct., 2022: 2200205.

[2] Wang Y, Lv Y, Su Y B, Chen L Q, Li H, Wu F. 5V-class sulfurized spinel cathode stable in sulfide all-solid-state batteries[J]. Nano Energy, 2021, 90: 106589.

[3] Wu F, Liu L L, Wang S, Xu J R, Lu P S, Yan W L, Peng J, Wu D X, Li H. Solid state ionics -selected topics and new directions[J]. Prog. Mater. Sci., 2022, 126: 100921.

[4] Yao N, Chen X, Fu Z H, Zhang Q. Applying classical, *ab initio*, and machine-learning molecular dynamics simulations to the liquid electrolyte for rechargeable batteries[J]. Chem. Rev., 2022, 122(12): 10970-11021.

[5] Wang Y, Wang Z X, Wu D X, Niu Q H, Lu P S, Ma T H, Su Y B, Chen L Q, Li H, Wu F. Stable Ni-rich layered oxide cathode for sulfide-based all-solid-state lithium battery [J]. eScience, 2022, 2(5): 537-545.

[6] Ye G, Zhao M, Hou L P, Chen W J, Zhang X Q, Li B Q, Huang J Q. Evaluation on a 400 Wh·kg^{-1} lithium-sulfur pouch cell[J]. J. Energy Chem., 2022, 66: 24-29.

[7] Wang J C, Zhang Z Y, Han J F, Wang X F, Chen L Q, Li H, Wu F. Interfacial and cycle stability of sulfide all-solid-state batteries with Ni-rich layered oxide cathodes [J]. Nano Energy, 2022, 100: 107528.

[8] Peng H J, Huang J Q, Zhang Q. A review of flexible lithium-sulfur and analogous alkali metal-chalcogen rechargeable batteries[J]. Chem. Soc. Rev., 2017, 46(17): 5237-5288.

[9] Yao N P, Heredy L A, Saunders R C. Secondary lithium-sulfur battery[J]. J. Electrochem. Soc., 1970, 117: C247.

[10] Xue L X, Li Y Y, Hu A J, Zhou M J, Chen W, Lei T Y, Yan Y C, Huang J W, Yang C T, Wang X F, Hu Y, Xiong J. *In-situ/operando* Raman techniques in lithium-sulfur batteries[J]. Small Struct., 2022, 3(3): 2100170.

[11] Hou L P, Yao L Y, Bi C X, Xie J, Li B Q, Huang J Q, Zhang X Q. High-valence sulfur-containing species in solid electrolyte interphase stabilizes lithium metal anodes in lithium-sulfur batteries[J]. J. Energy Chem., 2022, 68: 300-305.

[12] Song Y W, Peng Y Q, Zhao M, Lu Y, Liu J N, Li B Q, Zhang Q. Understanding the impedance response of lithium polysulfide symmetric cells[J]. Small Sci., 2021, 1(11): 2100042.

[13] Cheng X B, Yan C, Huang J Q, Li P, Zhu L, Zhao L, Zhang Y, Zhu W, Yang S T, Zhang Q. The gap between long lifespan Li-S coin and pouch cells: The importance of lithium metal anode protection[J]. Energy Storage Mater., 2017, 6: 18-25.

[14] Kong L, Jin Q, Zhang X T, Li B Q, Chen J X, Zhu W C, Huang J Q, Zhang Q. Towards full demonstration of high areal loading sulfur cathode in lithium-sulfur batteries[J]. J. Energy Chem., 2019, 39: 17-22.

[15] Chen W J, Li B Q, Zhao C X, Zhao M, Yuan T Q, Sun R C, Huang J Q, Zhang Q. Electrolyte regulation towards stable lithium-metal anodes in lithium-sulfur batteries with sulfurized polyacrylonitrile cathodes[J]. Angew. Chem. Int. Ed., 2020, 59(27): 10732-10745.

[16] Cheng Q, Chen Z X, Li X Y, Hou L P, Bi C X, Zhang X Q, Huang J Q, Li B Q. Constructing a 700 Wh·kg^{-1}-level rechargeable lithium-sulfur pouch cell[J]. J. Energy Chem., 2023, 76: 181-186.

[17] Holoubek J, Liu H D, Wu Z H, Yin Y J, Xing X, Cai G R, Yu S C, Zhou H Y, Pascal T A, Chen Z, Liu P. Tailoring electrolyte solvation for Li metal batteries cycled at ultra-low temperature[J]. Nat. Energy, 2021, 6(3): 303-313.

[18] Zhao M, Peng Y Q, Li B Q, Zhang X Q, Huang J Q. Regulation of carbon distribution to construct high-sulfur-content cathode in lithium-sulfur batteries[J]. J. Energy Chem., 2021, 56: 203-208.

[19] Wang J L, Yang J, Xie J Y, Xu N X, Li Y. Sulfur-carbon nano-composite as cathode for rechargeable lithium battery based on gel electrolyte[J]. Electrochem. Commun., 2002, 4(6): 499-502.

[20] Ji X, Lee K T, Nazar L F. A highly ordered nanostructured carbon-sulphur cathode for lithium-sulphur batteries[J]. Nat. Mater., 2009, 8(6): 500-506.

[21] Manthiram A, Fu Y, Chung S H, Zu C, Su Y S. Rechargeable lithium-sulfur batteries[J]. Chem. Rev., 2014, 114(23): 11751-11787.

[22] Zak J J, Kim S S, Laskowski F A L, See K A. An exploration of sulfur redox in lithium battery cathodes[J]. J. Am. Chem. Soc., 2022, 144(23): 10119-10132.

[23] Li G R, Wang S, Zhang Y N, Li M, Chen Z W, Lu J. Revisiting the role of polysulfides in lithium-sulfur batteries [J]. Adv. Mater., 2018, 30(22): 1705590.

[24] Lei J, Liu T, Chen J J, Zheng M S, Zhang Q, Mao B W, Dong Q F. Exploring and understanding the roles of Li_2S_n and the strategies to beyond present Li-S batteries [J]. Chem, 2020, 6(10): 2533-2557.

[25] Meini S, Elazari R, Rosenman A, Garsuch A, Aurbach D. The use of redox mediators for enhancing utilization of Li_2S cathodes for advanced Li-S battery systems[J]. J. Phys. Chem. Lett., 2014, 5(5): 915-918.

[26] Wang J L, Yang J, Xie J Y, Xu N X. A novel conductive polymer-sulfur composite cathode material for recharge able lithium batteries[J]. Adv. Mater., 2002, 14(13-14): 963-965.

[27] Chen Z Y, Zhou J J, Guo Y S, Liang C D, Yang J, Wang J L, Nuli Y N. A compatible carbonate electrolyte with lithium anode for high performance lithium sulfur battery [J]. Electrochim. Acta, 2018, 282: 555-562.

[28] Wang W X, Cao Z, Elia G A, Wu Y Q, Wahyudi W, Abou-Hamad E, Emwas A H, Cavallo L, Li L J, Ming J. Recognizing the mechanism of sulfurized polyacrylonitrile cathode materials for Li-S batteries and beyond in Al-S batteries[J]. ACS Energy Lett., 2018, 3(12): 2899-2907.

[29] Fanous J, Wegner M, Grimminger J, Andresen Ä, Buch meiser M R. Structure-related electrochemistry of sulfur-poly(acrylonitrile) composite cathode materials for rechargeable lithium batteries[J]. Chem. Mater., 2011, 23(22): 5024-5028.

[30] Zhang S S. Understanding of sulfurized polyacrylonitrile for superior performance lithium/sulfur battery[J]. Energies, 2014, 7(7): 4588-4600.

[31] Warneke S, Hintennach A, Buchmeiser M R. Communication—influence of carbonate-based electrolyte composition on cell performance of span-based lithium-sulfur batteries[J]. J. Electrochem. Soc., 2018, 165(10): A2093.

[32] Yang H J, Naveed A, Li Q Y, Guo C, Chen J H, Lei J Y, Yang J, Nuli Y N, Wang J L. Lithium sulfur batteries with compatible electrolyte both for stable cathode and dendrite-free anode[J]. Energy Storage Mater., 2018, 15: 299-307.

[33] Li Q Y, Yang H J, Xie L S, Yang J, Nuli Y N, Wang J L. Guar gum as a novel binder for sulfur composite cathodes

in rechargeable lithium batteries[J]. Chem. Commun., 2016, 52(92): 13479-13482.

[34] Wei S, Ma L, Hendrickson K E, Tu Z, Archer L A. Metal-sulfur battery cathodes based on PAN-sulfur composites[J]. J. Am. Chem. Soc., 2015, 137(37): 12143-12152.

[35] Zhao M, Li B Q, Peng H J, Yuan H, Wei J Y, Huang J Q. Lithium-sulfur batteries under lean electrolyte conditions: Challenges and opportunities[J]. Angew. Chem. Int. Ed., 2020, 59(31): 12636-12652.

[36] Peng H J, Huang J Q, Cheng X B, Zhang Q. Review on high-loading and high-energy lithium-sulfur batteries[J]. Adv. Energy Mater., 2017, 7(24): 1700260.

[37] Li X Y, Feng S, Zhao M, Zhao C X, Chen X, Li B Q, Huang J Q, Zhang Q. Surface gelation on disulfide electrocatalysts in lithium-sulfur batteries[J]. Angew. Chem. Int. Ed., 2022, 61(7): e202114671.

[38] Wang Z K, Li Y, Ji H Q, Zhou J Q, Qian T, Yan C L. Unity of opposites between soluble and insoluble lithium polysulfides in lithium-sulfur batteries[J]. Adv. Mater., 2022, 34(47).

[39] Feng S, Singh R K, Fu Y C, Li Z, Wang Y L, Bao J, Xu Z J, Li G S, Anderson C, Shi L L, Lin Y H, Khalifah P G, Wang W, Liu J, Xiao J, Lu D P. Low-tortuous and dense single-particle-layer electrode for high-energy lithium-sulfur batteries[J]. Energy Environ. Sci., 2022, 15(9): 3842-3853.

[40] Zhao C X, Li X Y, Zhao M, Chen Z X, Song Y W, Chen W J, Liu J N, Wang B, Zhang X Q, Chen C M, Li B Q, Huang J Q, Zhang Q. Semi-immobilized molecular electrocatalysts for high-performance lithium-sulfur batteries [J]. J. Am. Chem. Soc., 2021, 143(47): 19865-19872.

[41] Zhao M, Li B Q, Chen X, Xie J, Yuan H, Huang J Q. Redox comediation with organopolysulfides in working lithium-sulfur batteries[J]. Chem, 2020, 6(12): 3297-3311.

[42] Zhao M Q, Peng H J, Tian G L, Zhang Q, Huang J Q, Cheng X B, Tang C, Wei F. Hierarchical vine-tree-like carbon nanotube architectures: *In-situ* CVD self-assembly and their use as robust scaffolds for lithium-sulfur batteries[J]. Adv. Mater., 2014, 26(41): 7051-7058.

[43] Lv D, Zheng J, Li Q, Xie X, Ferrara S, Nie Z, Mehdi L B, Browning N D, Zhang J G, Graff G L, Liu J, Xiao J. High energy density lithium-sulfur batteries: Challenges of thick sulfur cathodes[J]. Adv. Energy Mater., 2015, 5 (16): 1402290.

[44] Zhang S J, Zhang Y S, Shao G S, Zhang P. Bio-inspired construction of electrocatalyst decorated hierarchical porous carbon nanoreactors with enhanced mass transfer ability towards rapid polysulfide redox reactions[J]. Nano Res., 2021, 14(11): 3942-3951.

[45] Kang N, Lin Y X, Yang L, Lu D P, Xiao J, Qi Y, Cai M. Cathode porosity is a missing key parameter to optimize lithium-sulfur battery energy density[J]. Nat. Commun., 2019, 10: 4597.

[46] Wang F, Zuo Z C, Li L, He F, Li Y L. Graphdiyne nanostructure for high-performance lithium-sulfur batteries[J]. Nano Energy, 2020, 68: 104307.

[47] Li B Q, Zhang S Y, Kong L, Peng H J, Zhang Q. Porphyrin organic framework hollow spheres and their applications in lithium-sulfur batteries[J]. Adv. Mater., 2018, 30 (23): 1707483.

[48] Liu X, Huang J Q, Zhang Q, Mai L. Nanostructured metal oxides and sulfides for lithium-sulfur batteries[J]. Adv. Mater., 2017, 29(20): 1601759.

[49] Geng C N, Hua W X, Wang D W, Ling G W, Zhang C, Yang Q H. Demystifying the catalysis in lithium-sulfur batteries: Characterization methods and techniques [J]. SusMat, 2021, 1(1): 51-65.

[50] Al Salem H, Babu G, V. Rao C, Arava L M R. Electrocatalytic polysulfide traps for controlling redox shuttle process of Li-S batteries[J]. J. Am. Chem. Soc., 2015, 137 (36): 11542-11545.

[51] Zhou T H, Lv W, Li J, Zhou G M, Zhao Y, Fan S X, Liu B L, Li B H, Kang F Y, Yang Q H. Twinborn TiO_2-tin heterostructures enabling smooth trapping-diffusion-conversion of polysulfides towards ultralong life lithium-sulfur batteries[J]. Energy Environ. Sci., 2017, 10(7): 1694-1703.

[52] Wang R C, Luo C, Wang T S, Zhou G M, Deng Y Q, He Y B, Zhang Q F, Kang F Y, Lü W, Yang Q H. Bidirectional catalysts for liquid-solid redox conversion in lithium-sulfur batteries[J]. Adv. Mater., 2020, 32(32): 2000315.

[53] Zhao M, Peng H J, Li B Q, Chen X, Xie J, Liu X, Zhang Q, Huang J Q. Electrochemical phase evolution of metal-based pre-catalysts for high-rate polysulfide conversion [J]. Angew. Chem. Int. Ed., 2020, 59(23): 9011-9017.

[54] Xue W J, Shi Z, Suo L M, Wang C, Wang Z A, Wang H Z, So K P, Maurano A, Yu D W, Chen Y M, Qie L, Zhu Z, Xu G Y, Kong J, Li J. Intercalation-conversion hybrid cathodes enabling Li-S full-cell architectures with jointly superior gravimetric and volumetric energy densities [J]. Nat. Energy, 2019, 4(5): 374-382.

[55] Li X Y, Feng S, Zhao C X, Cheng Q, Chen Z X, Sun S Y, Chen X, Zhang X Q, Li B Q, Huang J Q, Zhang Q. Regulating lithium salt to inhibit surface gelation on an electrocatalyst for high-energy-density lithium-sulfur batteries[J]. J. Am. Chem. Soc., 2022, 144(32): 14638-14646.

[56] Xie J, Song Y W, Li B Q, Peng H J, Huang J Q, Zhang Q. Direct intermediate regulation enabled by sulfur containers in working lithium-sulfur batteries[J]. Angew. Chem. Int. Ed., 2020, 59(49): 22150-22155.

[57] Li X Y, Zhang Q. One stone two birds: Dual-effect kinetic regulation strategy for practical lithium-sulfur batteries [J]. J. Energy Chem., 2022, 65: 302-303.

[58] Zhao M, Chen X, Li X Y, Li B Q, Huang J Q. An organodiselenide comediator to facilitate sulfur redox kinetics in lithium-sulfur batteries[J]. Adv. Mater., 2021, 33(13): 2007298.

[59] Zhao M, Li X Y, Chen X, Li B Q, Kaskel S, Zhang Q, Huang J Q. Promoting the sulfur redox kinetics by mixed organodiselenides in high-energy-density lithium-sulfur batteries[J]. eScience, 2021, 1(1): 44-52.

[60] Chen X, Peng L F, Wang L H, Yang J Q, Hao Z X, Xiang J W, Yuan K, Huang Y H, Shan B, Yuan L X, Xie J. Ether-compatible sulfurized polyacrylonitrile cathode with excellent performance enabled by fast kinetics via selenium doping[J]. Nat. Commun., 2019, 10: 1021.

[61] Zhao C X, Chen W J, Zhao M, Song Y W, Liu J N, Li B Q, Yuan T, Chen C M, Zhang Q, Huang J Q. Redox mediator assists electron transfer in lithium-sulfur batteries with sulfurized polyacrylonitrile cathodes[J]. EcoMat, 2021, 3(1): e12066.

[62] Frey M, Zenn R K, Warneke S, Müller K, Hintennach A, Dinnebier R E, Buchmeiser M R. Easily accessible, textile fiber-based sulfurized poly(acrylonitrile) as Li/S cathode material: Correlating electrochemical performance with morphology and structure[J]. ACS Energy Lett., 2017, 2(3): 595-604.

[63] Razzaq A A, Yao Y Z, Shah R, Qi P W, Miao L X, Chen M Z, Zhao X H, Peng Y, Deng Z. High-performance lithium sulfur batteries enabled by a synergy between sulfur and carbon nanotubes[J]. Energy Storage Mater., 2019, 16: 194-202.

[64] Wang X F, Qian Y M, Wang L N, Yang H, Li H L, Zhao Y, Liu T X. Sulfurized polyacrylonitrile cathodes with high compatibility in both ether and carbonate electrolytes for ultrastable lithium-sulfur batteries[J]. Adv. Funct. Mater., 2019, 29(39): 1902929.

[65] Li S P, Han Z L, Hu W, Peng L F, Yang J Q, Wang L H, Zhang Y Y, Shan B, Xie J. Manipulating kinetics of sulfurized polyacrylonitrile with tellurium as eutectic accelerator to prevent polysulfide dissolution in lithium-sulfur battery under dissolution-deposition mechanism[J]. Nano Energy, 2019, 60: 153-161.

[66] Lebherz T, Frey M, Hintennach A, Buchmeiser M R. Influence of morphology of monolithic sulfur-poly(acrylonitrile) composites used as cathode materials in lithium-sulfur batteries on electrochemical performance [J]. RSC Adv., 2019, 9(13): 7181-7188.

[67] Zhou J J, Guo Y S, Liang C D, Cao L J, Pan H, Yang J, Wang J L. A new ether-based electrolyte for lithium sulfur batteries using a S@pPAN cathode[J]. Chem. Commun., 2018, 54(43): 5478-5481.

[68] Yin L C, Wang J L, Lin F J, Yang J, Nuli Y. Polyacrylonitrile/graphene composite as a precursor to a sulfur-based cathode material for high-rate rechargeable Li-S batteries[J]. Energy Environ. Sci., 2012, 5(5): 6966-6972.

[69] Yang H J, Chen J H, Yang J, Wang J L. Prospect of sulfurized pyrolyzed poly(acrylonitrile) (S@pPAN) cathode materials for rechargeable lithium batteries[J]. Angew. Chem. Int. Ed., 2020, 59(19): 7306-7318.

[70] Li Z, Zhang J T, Lu Y, Lou X W. A pyrolyzed polyacrylonitrile/selenium disulfide composite cathode with remarkable lithium and sodium storage performances [J]. Sci. Adv., 2018, 4(6): eaat1687.

[71] Wang L H, Chen X, Li S P, Yang J Q, Sun Y L, Peng L F, Shan B, Xie J. Effect of eutectic accelerator in selenium-doped sulfurized polyacrylonitrile for high performance room temperature sodium-sulfur batteries[J]. J. Mater. Chem. A, 2019, 7(20): 12732-12739.

锂硫电池复合正极研究进展

李西尧[1],赵长欣[1],李博权[2,3*],黄佳琦[2,3],张 强[1*]

(1. 清华大学化学工程系, 北京 100084, 中华人民共和国; 2. 北京理工大学前沿交叉科学研究院, 北京 100081, 中华人民共和国; 3. 北京理工大学材料学院,北京 100081, 中华人民共和国)

摘要: 锂硫电池因其超高的理论能量密度被视为极具前景的下一代电化学储能体系,其中高比容量的硫正极提供了锂硫电池的能量密度优势并直接决定了电池的实际性能。经过数十年的发展,最具前景的硫正极体系分别是硫碳复合(S/C)正极和硫化聚丙烯腈(SPAN)正极。本文系统综述了 S/C 正极和 SPAN 正极的最新研究进展。首先,简要介绍了两种正极的工作原理并进行了比较。S/C 正极发生固-液-固多相转化反应,充放电表现为双平台特征。与之相比,SPAN 正极发生固-固反应,充放电曲线为单平台。然后,对两种正极所面临的挑战和目前报道的优化策略进行了系统的分析与讨论。对于 S/C 正极,主要调控策略包括电极结构修饰、电催化剂设计与辅助氧化还原介体调控;对于 SPAN 正极,主要调控策略包括电极结构设计、电极形貌调控、杂原子掺杂和外源性氧化还原介体调控。最后,在电池尺度上对 S/C 正极和 SPAN 正极进行了综合比较,并对基于 S/C 正极和 SPAN 正极的锂硫电池在未来所面对的机遇与挑战进行了展望。

关键词: 锂硫电池; 硫碳复合正极; 硫化聚丙烯腈正极; 多硫化锂