[Journal of Electrochemistry](https://jelectrochem.xmu.edu.cn/journal)

[Volume 28](https://jelectrochem.xmu.edu.cn/journal/vol28) Issue 12 Special Issue: In Honor of Professor [Yu-Sheng Yang on the Occasion of His 90th](https://jelectrochem.xmu.edu.cn/journal/vol28/iss12) [Birthday \(](https://jelectrochem.xmu.edu.cn/journal/vol28/iss12)II)

2022-12-28

Advances on Composite Cathodes for Lithium-Sulfur Batteries

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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>

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J. Electrochem. 2022, 28(12), 2219013 (1 of 13)
DOI: 10.13208/j.electrochem.2219013 http://electrochem.xmu.edu.cn
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DOI: 10.13208/j.electrochem.2219013
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Xi-Yao Li¹, Chang-Xin Zhao¹, Bo-Quan Li²³⁺, Jia-Qi Huang^{2.3}, Qia 3. School of Materials Science, Beijing Institute of Technology, sherical Engineering during Material Science and Engineering, Beijing Institute of Technology, Beijing Institute of Chemical Engineering, Beijing 100084, Ch

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Xi-Yao Li¹, Chang-Xin Zhao¹, Bo-Quan Li²³, Jia-Qi Huang²³, Qiang Zhang²⁴

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Xi-Yao Li¹, Chang-Xin Zhao¹, Bo-Quan Li²²⁺, Jia-Qi Huang²⁻³, Qiang Zhang¹⁶

(*I. Beijing Key Laboratory of Green Chemical Reaction Engineering and Tec* **Advances on Composite Cathodes for Lithium-Sulfur Batteries**

Xi-Yao Li', Chang-Xin Zhao', Bo-Quan Li²³, Jia-Qi Huang^{2,3}, Qiang Zhang'

(*I. Beijing Key Laboratory of Green Chemical Reaction Engineering and Technolog* X_i-X_0 O. L¹₁, Chang-Xin Zhao¹, Bo-Quan Li²²², Jia-Qi Huang²³, Qiang Zhang¹²

(*I. Reijing Key Laboratory of Green Chemical Reaction Engineering and Technology, Department of

<i>Chemical Engineering, Tsinghu* Xi-Yao Li¹, Chang-Xin Zhao¹, Bo-Quan Li²³^r, Jia-Qi Huang²³, Qiang Zhang¹
(*I. Beijing Key Laboratory of Green Glemical Reaction Engineering and Technology, Department of
Chemical Engineering, Tsinghua Universi* (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 Multidiscriplinary Scienc (1. Beyong Key Laboratory of Green Chemical Necation by appearing and Technology, Uleyartami of
Chemical Kegineering, Thington University, Beijing 100084, China; 2. Advanced Research Institute of
Multidisciplinary Science Chemical Engineering, Tsinghua University, Reijing 100084, China; 2. Advanced Research Institute of

3. School of Materials Science and Engineering, Beijing Institute of Technology, Beijing 100081, China;

3. School of Mat Multidistriplinary 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-3. School of Materials Science and Engineering, Beijing Institute of Technology, Beijing 100081, China)
 Abstract: 1.ithium-sulfur (1i-S) hateries are dcemed as high-promising next-generation energy storage technique du uligh theoretical energy density, where the sulfar cathodes with high specific capacity guarantee the energy density advantage

directly descrimes the battery performances. A for decades of sephoration, the most promising and directly determine the battery performances. After decades of exploration, the most promising sulfar cathodes are sulfur-
carbons composite (SCC) cathodes and sulfurizated polyceryloonticle (SPAN) cathodes. In this ma composite (SC) cuthodes and sulfurized polyencyloniirle (SPAN) cuthodes. In this munoscript, recent udvances on SC and SPAN

activides in 1.i-S hatteries are comprehensively revived. The electrochemical reaction eirers
an eathodes in Li-S hatteries are comprehensively reviewed. The electrochemical reaction circumstances on S/C and SPAN cathodes
are firstly introduced and compared to revers the working mechanism of the two types of Li-S batt are finly introduced and compared to reveal the working mechanisms of the two types of Li-S batteries. The SC cathools mainly undergo solid-liquid-solid conversion processes with typical distributed in the sumparison, the undege solid-liquid-solid multi-plaste conversion processes with spinal donkbe-plasau change-discharge polarization curves. In
the comparison, the SPAN cultodes follow solid-solid conversion and exhibit single-plateau chan comparison, the PAN cultureles iolitow side-desired conversion and exhibit single-platear directions and conversion and exhibit single-platear directions, in the system content of the SC and SPAN cultures with SC cultures mg ant, are onalized a parameteris and a metallon and the significant parameters are respectively meson and a metallon entired intervals are proportional methods attention, efficient leachtroataly de-
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also, and redox comediation. For SPAN catholos, the main optimizing attaques are electrode structure modification, morphology
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high specific capacity guarantee the energy density advantage
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zxhibit single-plateau charge-di al double-plateau charge-discharge polarization curves. In
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Key words: lithium-sulfur buttery: sulfurcarbon composite culbode; sulfurized polysery bonixity of 2600 Wh · kg⁻¹ in theory,

The global mpid energy consumption drives mas-

which is 10 times higher than the current c egulation by co-polymerization, heterotom doping at moleculate level, and extrinsic reados uses of Li-S buteries with SC or SDAS cultubes are systematically umalgeed ubroads are specified buter possimulation. At last, curr S batteries with S/C or SPAN cathodes are systematically analyzed through the comparison of several battery paractives on challenges and opportunities of S/C and SPAN cathodes in Li-S batteries are presented to guide futur

1 Introduction ultrahigh energy density of $2600 \text{ Wh} \cdot \text{kg}^{-1}$ in theory, the electrode structure monitoation, einterni electroceataryst de-
theoretical species are electrode structure modification, morphology
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way, the performance determining factor of Li-S batteries based on the system of the s as high as 1672 mAh - g-1, which guarantees the ultra-

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ploration, the most p vanced sulfur cathodes is the key for constructing

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able events during the development of the above two

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breakthrough very recently. For instance, Cheng et.

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eclels hased on the SC cathodes¹⁰, Chen, Liu, and co-

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under –60 °C based on the SPAN cathodes^[19]. workers realized stable eycling of Li-S pouch cells

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odes in Li-S batteries is essentially required to sum-
odes are composed of nanosized sulfur and conduc-
marize the recent odes in Li-S batteries is essentially required to summarize the recent advances and provide insightful tive carbon after ball-milling and heat melting. War

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perspectives for future development.

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Herein, recent advances on S/C and SPAN cath-

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In 2009, Nazar's group made a br

The electro Herein, recent advances on S/C and SPAN cath-

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Redox co-mediators (coRMs)

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SPAN cathodes in Li-S batteries.

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SPAN cathodes in Li-S batteries.

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 Example 1 A brief timeline about the development of S/C and
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es are composed of nanosized sulfur and conduc-

carbon after ball-mil **Figure 1** A brief timeline about the development of S/C and SPAN cathodes in Li-S batteries.
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pared to reveal the w and SPAN cathodes are firstly introduced and com-

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types of Li-S hatteries. Key challenges and corresponding optimizing strategies and corresponding optimizing strategies of the current SC and

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In 2009, Nazar's group made a breakthrough by developing highly ordered nanostructured carbon to accommod 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 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, ele veloping highly ordered nanostructured carbon to
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interface regulations, etc.
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2.4 V to 2.1 V represents the conversion process **ELEVALUATE 12.1**
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(Figure 2b). Concretely, the first plateau from

al irreversible reactions taking place in the

2.4 V to 2.1 V represents the conversion process

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SPAN stru **ing mechanism of** S/C cathodes obeys the solid-liq-
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19 **EVALUATION EXAMONDER (Figure 2b).** Oncretely, the first plateau from all irreversible reactions taking place in the first dis-
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al irreversible reactions taking place in the first dis-
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SPAN structure is reformed and the SPAN cathode is
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charge cycle of the SPAN cathode, during which the
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24 V to 2.1 V represents the conversion process

26 O cou **EVALUAT ENTERT (FOCT 2000)** (16 (Figure 2b). Concretely, the first plateau from all irreversible reactions taking place in the first dis-

2.4 V to 2.1 V represents the conversion process charge cycle of the SPAN cathode pathway (Figure 2b). Concretely, the first plateau from

2.4 V to 2.1 V represents the conversion process

2.4 V to 2.1 V represents the conversion process

charge cycle of the SPAN cathode, during which the

from solid S pathway (Figure 2b). Concretely, the first plateau from al irreversible reactions taking place in the first dis-

2.4 V to 2.1 V represents the conversion process charge cycle of the SPAN cathode, during which the

from so 2.4 V to 2.1 V represents the conversion process

charge cycle of the SPAN cathode, during which the

from solid S₄ to soluble LiPSs, and the second

SPAN stratetre is reformed and the SPAN cathode is

plateau around 2. from solid S_R to soluble LiPSs, and the second

SPAN structure is reformed and the SPAN cathode is

plateau around 2.1 V represents the conversion pro-

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ing me plateau around 2.1 V represents the conversion pro-
entivated⁵⁰. Once the above activation process is fin-
eign concharing these polyces and the specifical digits²⁹⁷. In bring, the valid-liq ing enchanism of SC cathods cess from LiPSs to solid Li₅⁽²⁴²⁾. In brief, the work-ished, the SPAN cathode can reversibly cycle during
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possible implacements of SC cahoos obcysts to solid-line long-l ing mechanism of S/C cathodes obeys the solid-liq-

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Worthy to be noted, the working mechanism of

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al irreversible reactions taking place in the first dis-
charge cycle of the SPAN cathode, during which the
SPAN structure is reformed and the SPAN cathode is
activated^[30]. Once the above 28(12), 2219013 (3 of 13)

al irreversible reactions taking place in the first dis-

charge cycle of the SPAN cathode, during which the

SPAN structure is reformed and the SPAN cathode is

activated^[30]. Once the above 28(12), 2219013 (3 of 13)

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SPAN structure is reformed and the SPAN cathode is
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SPAN structure is reformed and the SPAN cathode is
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charge cycle of the SPAN cathode, during which the
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SPAN structure is reformed and the SPAN cathode is
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long-las SPAN structure is reformed and the SPAN cathode is
activated^[30]. Once the above activation process is fin-
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Worthy to be noted activated¹³⁰. Once the above activation process is fin-
ished, the SPAN cathode can reversibly cycle during
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SPAN cathodes obeys the s ished, the SPAN cathode can reversibly cycle during
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(Figure 2c), which is quite dif by cycle during
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Worthy to be noted, the working mechanism of

SPAN cathodes obeys the solid-solid reaction pathway

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id-liquid-solid reaction p Worthy to be noted, the working mechanism of
SPAN cathodes obeys the solid-solid reaction pathway
(Figure 2c), which is quite different from the sol-
id-liquid-solid reaction pathway for S/C cathodes^[31].
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 $\#E\#(J. Electron) \ge 222, 28(12), 2219013 \text{ (4 of 13)}$
it, it can be assured that the capacity for SPAN cath-
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it, it can be assured that the capacity for SPAN cath-

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Previous literatures have reported that introducing

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it, it can be assured that the capacity for SPAN cath-

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previous literatures have re $\frac{dE}{dt}$ $\frac{dE}{dt}$

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specific capacity were greatly enhanced, and an ultra-
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and content, lean electrolyte, etc.)^{[35,10}]. The sluggish fur loading of 10 mg_s · em⁻² could be act
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 $\# \ell \Hrightarrow{\mathcal{H}}(L \to Electrochem.)$ 2022, 28(12), 2219013 (4 of 13)

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3.1, it can be assured that the capacity for SPAN cath-

3.1 **Electrode Structure Modification**

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can be assured that the capacity for SPAN cath-

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from a for S-S bonds¹⁸¹. In brief, the working mechanical respec the main challenge for S/C cathodes is the slug-

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3.1. **Electrode Structure Modification**

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 3.1 Electrode Structure Modification

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3.1 Electrode Structure Modification

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Previous literatures have reported that introducing

mains of SPAN catho it, it can be assured that the capacity for SAN can-
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experiments are report ones is provided by the reversion between tracking and gen-

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mism of SPAN cuthodes obeys the solid soli eration of S—N in prior. It is mell, the working mech-

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time of SPAN eathedes obeys the solid-solid rea-

ti ann of bYAN caliboses onesys the sould scattering and profilence are actions into a particular reductions, and modifical the LiPSs and this diffusion properties^{38,28}. Combining the above two of SPAN itself constitute to to particle and the tracted structure of the particle structure and the structure and the structure and functions $\int_{1/2}^{1/2} f_{1/2}^{1/2} f_{2/2}^{1/2} f_{2/2}^{1/2} f_{2/2}^{1/2} f_{2/2}^{1/2} f_{2/2}^{1/2} f_{2/2}^{1/2} f_{2/2}^{1/2} f_{2/2}$ LENEVAN), and the
FOREX) and the method with the surface are alternational constrained to discussed of SPAN itself constitute to be the research focus.
 Constrained area, the main challenges and Optimizing Strate-
 Exa of SPAN itself constitute to be the research locus. methods together, Zhang et. al. adopted an ant-nest-
 Collenges and Optimizing Strate- like hierarchical protous carbot with high specific
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The main **3 Challenges and Optimizing Strate-** like hierarchical porous carbon with high specific
 rigies for S/C Cathodes for some and regulated pore structure in S/C

The main challenge for S/C cathodes is the slug-

adhodes⁽ **gies for S/C Cathodes**

The main challnge for S/C antoodes is the slug-

informinalization and the The main challnge for S/C cathodes is the slug-

specific capacity were greatly enhanced, and an ultra-

tions (e.g., lar The main challenge for S/C cathodes is the slug-
staboles¹⁴⁴¹. As a result, both the polarization and the
sight kintices especially under harsh working condi-
specific equacity over geneally whenced, and an ultra-
and c $28(12)$, 2219013 (4 of 13)
 3.1 Electrode Structure Modification

Previous literatures have reported that introducing

high specific area materials can provide abundant re-

action sites for the sulfur redox reactions, 12), 2219013 (4 of 13)
 1 Electrode Structure Modification

Previous literatures have reported that introducing

gh specific area materials can provide abundant re-

tion sites for the sulfur redox reactions, and modi-
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 3.1 Electrode Structure Modification

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action sites for the sulfur redox reactions, a 28(12), 2219013 (4 of 13)
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action sites for the sulfur redox reactions, a 28(12), 2219013 (4 of 13)
 3.1 Electrode Structure Modification

Previous literatures have reported that introducing

high specific area materials can provide abundant re-

action sites for the sulfur redox reactions, a Li⁺diffusion properties^[42,43]. Combining the above two 2), 2219013 (4 of 13)
 Electrode Structure Modification

Previous literatures have reported that introducing

the specific area materials can provide abundant re-

ion sites for the sulfur redox reactions, and modi-

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 3.1 Electrode Structure Modification

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action sites for the sulfur redox reactions, a 28(12), 2219013 (4 of 13)
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action sites for the sulfur redox reactions, and 28(12), 2219013 (4 of 13)
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3.1 Electrode Structure Modification

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 3.1 Electrode Structure Modification

Previous literatures have reported that introducing

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action sites for the sulfur redox reactions, a **re Modification**
reported that introducing
can provide abundant re-
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3.1 Electrode Structure Modification

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action sites for the sulfur redox reactions, and 28(12), 2219013 (4 of 13)

3.1 Electrode Structure Modification

Previous literatures have reported that introducing

high specific area materials can provide abundant re-

action sites for the sulfur redox reactions, and **3.1 Electrode Structure Modification**
Previous literatures have reported that introducing
high specific area materials can provide abundant re-
action sites for the sulfur redox reactions, and modi-
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Previous literatures have reported that introducing
high specific area materials can provide abundant re-
action sites for the sulfur redox reactions, and modi-
fying the pore structur Previous literatures have reported that introducing
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Li⁻ di high specific area materials can provide abundant re-
action sites for the sulfur redox reactions, and modi-
fying the pore structure can regulate the LiPSs and
Li⁺ diffusion properties^(2,2,3). Combining the above two action sites for the sulfur redox reactions, and modi-
fying the pore structure can regulate the LiPSs and
Li⁺ diffusion properties^(2,43). Combining the above two
methods together, Zhang et. al. adopted an ant-nest-
l fying 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 surfa 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*
catho 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
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surface area and regulated pore structure in S/C
cathodes^[44]. As a result, both the polarization and the
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high a 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 ultra-
high area capacity of 11.6 mAh · cm⁻² at a high sul-
fu cathodes^[44]. As a result, both the polarization and the specific capacity were greatly enhanced, and an ultra-
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fur loading of 10 mg·cm⁻² could be achieved. Never-
 specific capacity were greatly enhanced, and an ultra-
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fur loading of 10 mg_s · cm⁻² could be achieved. Never-
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 电化学(*J. Electrochem.*) 2022, 28(12), 2219013 (4 of 13)
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 $# \&L \# (J. Electron) 2022, 28(12), 2219013 (5 of 13) = 4 μL·mg⁻¹). Other electrode structure modification
approaches such as designing S/C shell structures
and network structures also obtained favorable ef-
fects^[46,47]. In brief, the porosity, specific surface area,
and tortuosity are three significant parameters to be
behaviors inevitably$ $\#E\#(J. Electronchem.)$ 2022, 28(12), 2219013 (5 of 13)
 $\#L \cdot mg^{-1}$). Other electrode structure modification

approaches such as designing S/C shell structures

and network structures also obtained favorable ef-

fects^[46,47]. I $\pm 4 \mu L \cdot mg^{-1}$). Other electrode structure modification
 $\pm 4 \mu L \cdot mg^{-1}$). Other electrode structure modification

the Mo₆S₈ surface got lithiation into Li_nMo₆S₈ during

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approaches such as designing S/C shell structures the discharge pr

= 4 μ L-mg⁻¹). Other electrods state and stream-

= 4 μ L-mg⁻¹). Other electrods state and direction the Mos₅, surface got lithiation into Li,N₀₅, during

approaches such as designing S/C shell structures th =4 μ L -mg⁻¹). Other electrode structure modification the Mo₆S₁ surface got lithiation into Li,Mo₆S₁ during
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and network structures sube obsined favorable ef-

covered by a gel layer due to the solvent simulaneous

fictis^{46.47}. In brief, the prossity, specific surface area,

po and network structures also obtained favorable ef-
covered by a gel layer due to the solvent simultaneous
fects⁴⁶⁶⁷, In brief, the porosity, specific surface area, polymerization¹⁹³³. The above surface reconstruction
 fcets^(wa,7). In brief, the porosity, specific surface area, polymerization¹⁵³. The above surface reconstruction and ortinuality are three significant parameters to be behaviors inevidably affect the LiF's electroceath and tortuosity are three significant parameters to be behaviors inevitably affect the LiPS electrocatalysis
considered during SC cathode design, and the SC mechanisms and cffects. In brief, electrocatalysts are
cathodes ca considered during S/C cathode design, and the S/C mochanisms and cffcets. In brief, clectrocatalysts are carbitative assumestives are realized to overcome the sluggish charge transferse the above three parameters are well eathodes can exhibit superior performances only when

throbuced to overcome the sluggish charge transfer

the above three parameters are vell balanced (Table 1). kincties, and their design should fully consider the in-

3 the above three parameters are well balanced (Table 1). kinetics, and their design should fully consider the in-

TE purpose of introducing clettrocalizy sto soft refacial issues as well as the solidiquid-solid reaction
 3.2 **Efficient Electrocatalyst Design** terfacial issues as well as the solidiquid-solid reaction
The purpose of introducing electrocatalysts to S/C pathways in Li-S batteries.
can be strengthen LiPS adsorption and facil-
 The purpose of introducing electrocatalysts to S/C pathways in Li-S batteries.

cathodes is to strengthen LiPS adsorption and facili-

2.33 Redox Comediation on LiPSs

tate interfacial charge transfer¹⁶⁸¹. Previous rese cathodes is to strengthen LiPS adsorption and facili-

atate interfacial charge transfer^{tan}. Previous research-

and the electrode structural modifica-

tas have proved that electrodealy sorted as transis-

tion metall tate interfacial charge transfer^{t68}. Previous research-

is have proved that electrocale stractural modifica-

tion strategy overcoming the LiFSs diffusion issues

tion metal components, herero-doped curbon, and

their es have proved that electrocatalysts such as transi-
tion metal compounds, hetero-doped carbon, and
and the electrocatalysis
their composites can efficiently reduce the reaction
reactivativy, the reactivation energy and p

 $#E\# (J. Electrochem.) 2022, 28(12), 2219013 (5 of 13)$ (b). Other electrode structure modification the Mo₆S₈ surface got lithiation into Li_sMo₆S₈ during tuch as designing S/C shell structures the discharge process^[54], an ¹⁴ (*L Electrochem.*) 2022, 28(12), 2219013 (5 of 13)
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approaches such as designing S/C shell structures

the Mo_iS_s surface got lithiation into Li,Mo_iS_s during

and network stru $\pm 4\mu L \cdot mg^{-4}$). Other electrode structure modification

= 4 $\mu L \cdot mg^{-4}$). Other electrode structure modification the Mo₈S_s surface got lithiation into Li,Mo₈S_s during

approaches such as designing S/C shell stru $\mathbb{E}/\mathbb{E}^2(L_E(\epsilon_{trrochem}) \geq 23(12), 2219013$ (5 of 13)
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 EXAMPLE 11 (The relation and the MoS₅ surface got lithiation into LiMoS₅ during

approaches such as designing S/C shell structures the discharge process¹⁹¹, and the MoS₅ the $\ell^2\ell^2$ (ℓ $\ell^2\ell^-$ (ℓ $\ell^2\ell^-$ (ℓ $\ell^2\ell^-$) (Other electrodes structure modification the MoS_b surface got linitation into LiMoS_b, during approaches such as designing S/C shell structures the discha \pm (*Emg*⁻¹). Other electrode structure modification the Mo_S₆ surface got lithiation into Li,Mo_S₆ during
approachos such as designing S/C shell structures the discharge process^[54], and the MoS₅ surface wa $+ \frac{1}{2}$ ($\frac{1}{2}$ $+ \frac{1}{2}$ $t_1/2 + t_2/2$. Deterocheme, 2022, 28(12), 2210013 (5 of 13)

approaches such as designing S/C shell structures the discharge process¹⁸¹, and the MoS₅ during

approaches such as designing S/C shell structures the disch $28(12)$, 2219013 (5 of 13)
the Mo₆S₈ surface got lithiation into Li_xMo₆S₈ during
the discharge process^[54], and the MoS₂ surface was
covered by a gel layer due to the solvent simultaneous
polymerization[[] $28(12)$, 2219013 (5 of 13)
the Mo₆S₈ surface got lithiation into Li_xMo₆S₈ during
the discharge process^[54], and the MoS₂ surface was
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the discharge process^[54], and the MoS₂ surface was

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the discharge process^[54], and the MoS₂ surface was
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the discharge process^[54], and the MoS₂ surface was
covered by a gel layer due to the solvent simultaneous
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the Mo₆S_s surface got lithiation into Li_sMo₆S_s during
the discharge process^[54], and the MoS₂ surface was
covered by a gel layer due to the solvent simultaneous
polymerization[[] $28(12)$, 2219013 (5 of 13)
the Mo₆S₈ surface got lithiation into Li_sMo₆S₈ during
the discharge process^[54], and the MoS₂ surface was
covered by a gel layer due to the solvent simultaneous
polymerization[[] $28(12)$, 2219013 (5 of 13)
the Mo₆S_s surface got lithiation into Li,Mo₆S_s during
the discharge process^[54], and the MoS₂ surface was
covered by a gel layer due to the solvent simultaneous
polymerization^{[53} (12), 2219013 (5 of 13)

Mo_oS_s surface got lithiation into Li,Mo_oS_s during

discharge process^[54], and the MoS₂ surface was

vered by a gel layer due to the solvent simultaneous

lymerization^[55]. The above $28(12)$, 2219013 (5 of 13)
the Mo₆S₈ surface got lithiation into Li_MO₆S₈ during
the discharge process^[34], and the MoS₂ surface was
covered by a gel layer due to the solvent simultaneous
polymerization^{[5} $28(12)$, 2219013 (5 of 13)
the Mo_oS₈ surface got lithiation into Li_xMo_oS₈ during
the discharge process^[34], and the MoS₂ surface was
covered by a gel layer due to the solvent simultaneous
polymerization[[] $28(12)$, 2219013 (5 of 13)
the Mo₆S₈ surface got lithiation into Li,Mo₆S₈ during
the discharge process^[54], and the MoS₂ surface was
covered by a gel layer due to the solvent simultaneous
polymerization^{[53} **Example 12**
the Mo₆S₈ surface got lithiation into Li,Mo₆S₈ during
the discharge process^[34], and the MoS₂ surface was
covered by a gel layer due to the solvent simultaneous
polymerization^[35]. The above sur the Mo_sS_s surface got lithiation into $LiMo₆S₈$ during
the discharge process^[34], and the MoS₂ surface was
covered by a gel layer due to the solvent simultaneous
polymerization^[35]. The above surface re $\frac{\text{H} \& \frac{2}{3} \times (J. \text{ *Electrochem.*}) 2022, 28(12), 2219013 (5 of 13)}{\text{structure modification}}$ the Mo₆S₈ surface got lithiation into Li_sMo₆S₈ during

S/C shell structures the discharge process^[34], and the MoS₂ surface was

btai

ed that electrocatalysts such as transi-

tion strategy overcoming the LiPSs diffusion issues

tens can efficiently reduce the reaction

tens can efficiently reduce the reaction

tergetical reduces are effect the reaction tion metal compounds, hetero-doped carbon, and und the electrocatalysis strategy promoting the LiPSs
their composites can efficiently reduce the reaction
functions rategy can
derivation onergy and promote the SCC cathode their composites can efficiently reduce the reaction
redox reactivity, the redox connectiation carreggy and promote the SC callode per-
accelerate that the SC callodes per-
formances^{169,30}. Since the solid-liquid-solid activation energy and promote the S/C cathode per-

accelerate the reactivity and diffusivity of LiPSs on

formances(^{as,78}). Since the solid-liquid-solid reaction

galardikas and disclendes at are time. In detail, organ formances^{(*n.sn*}). Since the solid-liquid-solid reaction S/C cathodes at the same time. In detail, organic
partway incruishly resulted solids LiFSs for disulfides and disclenides can serve as redox concid-
navay from th pathway inevitably results partial soluble LiPSs far disulfides and diselenides can serve as redox comedi-
away from the canholic interhece and difficult to par-
ators (co-RMs) to spontancously react with LiPSs in
trigine away from the cathodic interface and difficult to par-
actors (co-RMs) to spontaneously react with LiPSs in
ticipate in electrode reactions, expanding the interac-
Li-S batterics, afford an additional electrodenical partticipate in electrode reactions, expanding the interac-
Li-S batteries, afford an additional chemical reaction
have aspect into considerations, Zhao et. al. designed
way besides the original electrochemical park-
also abs tion range of electrocatalysts is essential. Taking the pathway besides the original electrochemical path-
above aspect into considerations. Zhoo et al. designed way, and promote the overall sulfur redox kinetics^{86.91},
 above aspect into considerations, Zhao et al. designed

as way, and promote the overall sulfur redox kinetics^{(86,9}),

at security hurds concernally by parting the ac-

Trive interved di-tertiary butyl

tive sites (porphy a semi-immobilized electrocatalyst by grafting the ac-

For instance, Zhao et. al. introduced di-tertiary butyl

divise disc (prophyrin) onto soft segments (polypyrrole)

disculfie (DDDS) into Li-S batteries as a co-RM

f tive sites (porphyrin) onto soft segments (polypyrrole) distifide (DtbDS) into Li-S batteries as a co-RM

(Figure 36)¹⁶⁰. Consequently, the semi-immobilizing (Figure 36)¹⁶¹¹, Concertely, the S—S bond in DbDS

(Figure (Figure 3b)¹⁶⁰). Consequently, the semiimmobilizing (Figure 3c)⁴⁰¹. Concretely, the S—S bond in DtbDS
strategy in practical Li-S batterics realized superior breaks spontaneously and the sulfur chains in LiPS
rate perf strategy in practical Li-S batterics realized superior

threaks spontaneously and the sulfur chains in LiPs

rate performances as well as long lifespan, and it also

endowed pouch cells with high energy density up to

Dtb rate performances as well as long lifespan, and it also molecules are subsequently intervalated into the endoved pouch cells with high energy density up to DhbNS molecule to form modified LiPS molecules and 343 Wh ·kg⁻¹ was sulfurized into \cos_x phases during initial cycles^[53], density of 384 Wh · kg⁻¹ was achieved in working Li-S the discharge process^[54], and the MoS₂ surface was
covered by a gel layer due to the solvent simultaneous
polymerization^[58]. The above surface reconstruction
behaviors inevitably affect the LiPS electrocatalysis
m covered by a gel layer due to the solvent simultaneous
polymerization^[53]. The above surface reconstruction
behaviors inevitably affect the LiPS electrocatalysis
mechanisms and effects. In brief, electrocatalysts are
int 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
kineti 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 in-
terf mechanisms and effects. In brief, electrocatalysts are
introduced to overcome the sluggish charge transfer
kinetics, and their design should fully consider the in-
terfacial issues as well as the solidliquid-solid reaction introduced to overcome the sluggish charge transfer
kinetics, and their design should fully consider the in-
terfacial issues as well as the solidliquid-solid reaction
pathways in Li-S batteries.
3.3 Redox Comediation on kinetics, and their design should fully consider the in-
terfacial issues as well as the solidiquid-solid reaction
pathways in Li-S batteries.
3.3 **Redox Comediation on LiPSs**
Different from the electrode structural modif terfacial issues as well as the solidliquid-solid reaction
pathways in Li-S batteries.
3.3 **Redox Comediation on LiPSs**
Different from the electrode structural modifica-
tion strategy overcoming the LiPSs diffusion issues pathways in Li-S batteries.
 3.3 Redox Comediation on LiPSs

Different from the electrode structural modifica-

tion strategy overcoming the LiPSs diffusion issues

and the electrocatalysis strategy promoting the LiPSs
 3.3 Redox Comediation on LiPSs
Different from the electrode structural modifica-
tion strategy overcoming the LiPSs diffusion issues
and the electrocatalysis strategy promoting the LiPSs
redox reactivity, the redox come Different from the electrode structural modifica-
tion strategy overcoming the LiPSs diffusion issues
and the electrocatalysis strategy promoting the LiPSs
redox reactivity, the redox comediation strategy can
accelerate t tion 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 cath 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 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 comedi-
ators (coaccelerate the reactivity and diffusivity of LiPSs on
S/C cathodes at the same time. In detail, organic
disulfides and diselenides can serve as redox comedi-
ators (co-RMs) to spontaneously react with LiPSs in
Li-S batter S/C cathodes at the same time. In detail, organic
disulfides and disclenides can serve as redox comedi-
ators (co-RMs) to spontaneously react with LiPSs in
Li-S batteries, afford an additional chemical reaction
pathway be 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 prom $\mu L \cdot mg^{-1}$ and high sulfur loading (5.0 mg_s \cdot cm⁻²) RMs) to spontaneously react with LiPSs in
ries, afford an additional chemical reaction
besides the original electrochemical path-
promote the overall sulfur redox kinetics^{[66,67}].
coe, Zhao et. al. introduced di-tertiar $\left(\frac{1}{2} \right)$ Li-S batteries, afford an additional chemical reaction
pathway besides the original electrochemical path-
way, and promote the overall sulfur redox kinetics^{[96,87}].
For instance, Zhao et. al. introduced di-tertiary buty pathway besides the original electrochemical path-
way, and promote the overall sulfur redox kinetics^{[96,67}].
For instance, Zhao et. al. introduced di-tertiary butyl
disulfide (DtbDS) into Li-S batteries as a co-RM
(Fig way, and promote the overall sulfur redox kinetics^{[96,97}].
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 br 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 ar 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 f (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 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_{*m*-I}Li). Both the
diffusivity 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 t

 $#E\#(J. Electrochem.)$ 2022, 28(12), 2219013 (6 of 13)
pouch cells^[59]. In brief, the redox comediation strat-
egy is impressively effective in accelerating both the
diffusivity and the reactivity of LiPSs, and it can pro-
tivel the \mathcal{C}_{rel} is impressively effective in accelerating both the Since the electronic conductivity of SPAN is rela-
eifflisivity and the reactivity of LiPSs, and it can pro-
time the electronic conductivity of SPAN is odes.

ders (around 10 -⁹ ^S窑cm-¹ ders (around 10^{-9} S \cdot cm⁻¹)^[66]. Meanwhile, volume issues as well. However, different from the S/C conductive network (Figure 4a)⁶⁸⁸. As a result, Lie cathodes, the optimizing strategies do not aim at cells with the GO composited SPAN cathodes consolide LiFSs but rather

 $\mathbb{E}\left(\mathbb{R}^2\right)$. In brief, the redox comediation strat-
pouch cells^[99]. In brief, the redox comediation strat-
eigy is impressively effective in accelerating both the Since the electronic conductivity of SPAN is r $\frac{dE}{E}$ (*Leterochem.*) 2022, 28(12), 2219013 (6 of 13)

pouch cells^[59]. In brief, the redox comediation strat-

egy is impressively effective in accelerating both the

since the electronic conductivity of SPAN is r $#R \cong (J. Electrochem.) 2022, 28(12), 2219013 (6 of 13)$

pouch cells⁽³⁹⁾. In brief, the redoc comediation strat-

egy is impressively effective in acceleration strat-

diffusivity and the reactivity of LiPSs, and it can pro-

invest $\text{#E}^{\text{g}}(J. \text{ *Electrochem.*}) 2022, 28(12), 2219013 (6 of 13) \\\\ \text{the cells}^{\text{(59)}}. \text{ In brief, the redox compediation strat-} \textbf{4.1} \text{ *Electrode Structure Modification* is impressively effective in accelerating both the
\n5. Since the electronic conductivity of SPAN
\n2. If the probability of CPAN is the activity of LIPSs, and it can pro-} \text{ is increasing to modify electro-} \\\ \text{the full-range sulfur redox kinetics on S/C cath-} \text{tures by introducing high-conductive carbon
\nas and constructing efficient ionic and e
\n**Challenges and Optimizing Structure** ($ **EVALUATION EXAMORE 1999** (50 m) and etherwise are conjuncted for the simple simple simple simplessively effective in accelerating both the Since the electronic conductivity of SPAN is relativity of LiPSs, and it can prov **Example 19**
 Example 1998. In brief, the redox comediation strat-**4.1 Electrode Structure Modification**

egy is impressively effective in accelerating both the

Since the electronic conductivity of SPAN is rela-

diffu **EVALUATION THE SET ALTERT CONTIFICATE CONTIFICATE SET ALTERT AND THE SET ART AND THE SET AND THE SET** soluble LiPSs but rather SPAN itself. Concretely, 4×4 *Electrochem.*) 2022, 28(12), 2219013 (6 of 13)
pouch cells¹⁵⁹¹. In brief, the redox comediation strat-
egy is impressively effective in accelerating both the
diffusivity of SPAN is rela-
diffusivity and the rea (0.42×1) Electrochem, 2022, 28(12), 2219013 (6 of 13)
pouch cells⁵⁹⁾. In brief, the redox connediation strat-
equision strate-
equision in the size definition of the size of the size of the size of definition of FAN $#_3(E\#^2(L \text{ *Electrochem*), 2022, 28(12), 2219013 (6 of 13)$

redox comediation strat-

in accelerating both the

since the electronic conductivity of SPAN is rela-

of LiPSs, and it can pro-

tively low, it is promising to modi th $\ell E^{\phi}(L \text{ Rlectmeden})$ 2022, 28(12), 2219013 (6 of 13)
pouch cells¹⁶⁹. In brief, the redox comediation strat-
4.1 Electrode Structure Modification
egy is impressively effective in accelerating both the
since the clectr pouch cells¹⁹⁹¹. In brief, the redox comediation strate-

egy is impressively effective in accelerating both the

diffusivity and the reactivity of LiPSs, and it can pro-

diffusivity and the reactivity of LiPSs, and it pouch cells¹⁹⁹. In brief, the redox connediation strat-

egy is impressively directive in accelerating both the since the lectronic conductivity of PRAN is rela-

difflisivity and the reativity of LiPSs, and it can pro-
 egy is impressively effective in accelerating both the

diffusivity of due tractivity of SPAN is rela-

diffusivity and the reactivity of SPAN is the more ively low, it is promising to modify electrode struction

one full diffusivity and the reactivity of LiPSs, and it can pro-

interestign in modify obcerted struc-

odes.

modify although high-conductive archor materi-

odes.
 4 Challenges and Optimizing Strate- and sonstructing efficie mote full-range sulfur redox kinetics on S/C cath-

are by introducing high-conductive carbon materi-

ds. Challenges and Optimizing Strate-

pathways. For instance, Yin et.al. introduced oxi-

gies for SPAN Cathodes

Main als and constructing efficient ionic and electronic
 gies for SPAN Cathodes
 dizero challenges and Optimizing Strate- pathways. For instance, Yin etal. introduced oxi-

Main challenges for SPAN enhodes are kincic make **4 Challenges and Optimizing Strate-** pathways. For instance, Vin et.al. introduced oxigies for SPAN cathodes

including three case of CO) into the SPAN carbodes are kinetic

make the SPAN powders evenly dispersed on the
 gies for SPAN Cathodes

Main challenges for SPAN cathodes are kinetic make the SPAN carbodes to the

issues as well. However, different from the SC conductive network (Figure 4a)¹⁶⁸), as a result, Li-S

issues as well Main challenges for SPAN cathodes are kinetic

inske the SPAN powders evenly dispersed on the

issues as well. However, different from the S/C conductive network (Figure 4a)⁵⁰¹. As a result, Li-S

cells with the GO comp $28(12)$, 2219013 (6 of 13)
 4.1 Electrode Structure Modification

Since the electronic conductivity of SPAN is rela-

tively low, it is promising to modify electrode struc-

tures by introducing high-conductive carbon 12), 2219013 (6 of 13)
 I Electrode Structure Modification

Since the electronic conductivity of SPAN is rela-

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 4.1 Electrode Structure Modification

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Since the electronic conductivity of SPAN is rela-

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Since the electronic conductivity of SPAN is rela-

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Since the electronic conductivity of SPAN is rela-

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Since the electronic conductivity of SPAN is rela-

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lectronic conductivity of SPAN is rela-

is promising to modify electrode struc-

ducing high-conductive carbon materi-

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rustance, Yin et.al. 28(12), 2219013 (6 of 13)
 4.1 Electrode Structure Modification

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electronic conductivity of SPAN is rela-

is promising to modify electrode struc-

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 4.1 Electrode Structure Modification

Since the electronic conductivity of SPAN is rela-

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tures by introducing high-conductive carbon ma 28(12), 2219013 (6 of 13)
 4.1 Electrode Structure Modification

Since the electronic conductivity of SPAN is rela-

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tures by introducing high-conductive carbon ma **4.1 Electrode Structure Modification**

Since the electronic conductivity of SPAN is rela-

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als and constructing **4.1 Electrode Structure Modification**
Since the electronic conductivity of SPAN is rela-
tively low, it is promising to modify electrode struc-
tures by introducing high-conductive carbon materi-
als and constructing eff Since the electronic conductivity of SPAN is rela-
tively low, it is promising to modify electrode struc-
tures by introducing high-conductive carbon materi-
als and constructing efficient ionic and electronic
pathways. F tively low, it is promising to modify electrode struc-
tures by introducing high-conductive carbon materi-
als and constructing efficient ionic and electronic
pathways. For instance, Yin et.al. introduced oxi-
dized graphe tures 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 eve als and constructing efficient ionic and electronic
pathways. For instance, Yin et.al. introduced oxi-
dized graphene (GO) into the SPAN cathodes to
make the SPAN powders evenly dispersed on the
conductive network (Figure 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 compos dized 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 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 e 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· micrometer-sized SPAN pow-

I672 mA·g⁻¹). However, GO with high specific area

com⁻¹)^{(66]}. Meanwhile, volume

always takes up much electrolyte conditions. To this incompatible

issues. Additionally, in ether-

signi 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, electro-
spinning processing is reported to compensate the
electron g f). However, OO with migh spectra accass up much electrolyte and is incompatible electrolyte conditions. To this end, electro-
rocessing is reported to compensate the conductivity and tolerant low electrolyte the same t Fable 1 Summary of advanced Li-S battery performances with S/C or SPAN cathodes.

Table 1 Summary of advanced Li-S battery performances with S/C or SPAN cannot and incorporate of the sulfur performance and the same time. 电化学(*J. Electrochem.*) 2022, 28(12), 2219013 (6 of 13)
dox comediation strat-
a correlating both the Since the electronic conductivity of SPAN is rela-
EiPSs, and it can pro-
tively low, it is promising to modify electro

| variation during cycling destroys the structural stabil- ity 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 kinet- ics, and reduce the cathodic cycling stability ^[67] . Aim- ing at the above challenges, major optimizing strate- gies are proposed, including electrode structure mod- ification, morphology regulation by co-polymeriza- tion, heteroatom doping at molecular level, and ex- trinsic redox mediation (Figure 4). Table 1 Summary of advanced Li-S battery performances with S/C or SPAN cathodes. | | | with lean electrolyte conditions. To this end, electro- spinning 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 sulfur- ized them under high temperatures ^[64] . Consequently, the SPAN exhibited a nanosized slice morphology on the CNTs, which endowed the Li-S cells with a spe- cific capacity of 1180 mAh \cdot g ⁻¹ without capacity fad- ing over 1000 cycles. In brief, electrode structure | | |
|---|-------------------------|--|---|-------------------------------------|-----------------------------|
| | Component | Sulfur loading $(mg_s \cdot cm^{-2})$ | Specific capacity $(mAh·g-1)$ | Cycling lifespan | Reference |
| | G@ppy-por | 5.0 | 940 @ 0.2 C | 70 @ 0.2 C | Zhao et al.[40] |
| | $MoS2$ with TEA | 4.0 | 988 @ 0.3 C | 100 @ 0.3 C | Li et al. [37] |
| | | | 924 @ 0.1 C | 55 @ 0.1 C | Zhao et al. ^[58] |
| | DPDSe | 5.0 | | | |
| S/C cathode | LPC | 4.0 | 1001 @ 0.1 C | $100 \ (a) 0.1 C$ | Feng et al.[39] |
| | $7TiN:3TiO2-G$ | 1.2 | 800 @ 1.0 C | 2000 @ 1.0 C | Zhou et al. ^[51] |
| | Co_4N/NG | 4.1 | 1109 @ 0.5 C | 150 @ 0.5 C | Zhao et al. ^[53] |
| | Se _{0.06} SPAN | $1 \sim 3$ | 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 | $1000 \; (\omega, 4.0 \; \text{C})$ | Frey et al. ^[62] |
| SPAN cathode | SPAN-CNT20 | $0.9 \sim 1.1$ | 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] |

eration.

EXAMPLE 12
 EXAMPLE 12 HEPACE Electrochem.) 2022. 28(12), 2219013 (7 of 13)
 modification mainly concentrates on the SPAN con-

conduction networks and provided directed Li⁺ diffu-

ductivity enhancement. Compatibility with high sulfur

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modification mainly concentrates on the SPAN concentrates and provided directed Li² diffu-

ductivity enhancement. Computable with high sulfire sinth co 40. (*k*</sup>/ε *Blectochem*) 2022, 28((2), 2219013 (7 of 13)

modification mainly concentrates on the SPAN concentrates suggested that the directed Li⁺ diffu-

ductivity enhancement. Compatibility with high sulfare

con modification mainly concentrates on the SPAN concentrates on conduction networks and provided directed Li⁺ diffu-
ductivity enhancement. Compatibility with high sulfar sion pathways. As a result, Li-S cells showed high
 modification mainly concentrates on the SPAN con-

conduction networks and provided directed Li⁴ diffu-

ductrivity enhancement. Compatiblity with high sulfrier sion pathways. As a result, Li-S- cells showed high

conte ductivity enhancement. Compatibility with high sulfur

exontant and can clearly by the fit expanding to the consideration.

exoncent and lean clearly by the spatial be taken into consideration of the spatial behavior of t content and lean electrolyte shall be taken into considential mate capability up to 8.0 C and excellent cycling sta-
 ation.
 412 Morphology Regulation by Co-Poly- implyed years. In brief, regulating the SPAN
 412 Mo

 $\frac{\text{d} \mathcal{H}^{\text{H}}(J. \text{Electrochem.}) 2022, 28(12), 2219013 (7 \text{ of } 13)}{\text{modification mainly concentrates on the SPAN con-}}$ conduction networks and provided directed Li⁴ diffu-

ductivity enhancement. Compatibility with high sulfur

conduction networks and provided $\#E\#(J. Electron, 2022, 28(12), 2219013 (7 of 13)$

modification mainly concentrates on the SPAN concentrators of conduction networks and provided directed Li⁺ diffu-

ductivity enhancement. Compatibility with high sulfur

conten **Example 19** Example 1992, 2012), 2219013 (7 of 13)

Englisheductivity enhancement. Compatibility with high sulfur

ductivity enhancement. Compatibility with high sulfur

content and lean electrolyte shall be taken into ^{ti} det²² (*L* Electrochem.) 2022, 28(12), 2219013 (7 of 13)

modification mainly concentrates on the SPAN concentration networks and provided directed Li⁺ diffu-

ductivity enhancement. Compatibility with high sulf **merization** the precursor composition by co-polymerization is ef-**EVALUATION**
 EVALUATION
 EVALUATION in the SPAN particles are rather than the SPAN particles are related to the SPAN con-
 in the SPAN con- conduction networks and provided directed Li⁺ diffu-

ductivity enhancement. Compatibility with high sulfur sio **Example 120**
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sion pathways. As a result, Li-S cells showed high
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bility over 1200 cycles. In br conduction networks and provided directed Li⁺ diffu- $28(12)$, 2219013 (7 of 13)
conduction networks and provided directed Li⁺ diffu-
sion pathways. As a result, Li-S cells showed high
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28(12), 2219013 (7 of 13)

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sion pathways. As a result, Li-S cells showed high
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As a result, Li-S cells showed high
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Electrolytes, and delivered high capacities of 1507 5 Conclusions and

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

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 Example 1980 (*L Electrochem*, 2022, 28(12), 2219013 (8 of 13)
 Electrolytes, and delivered high capacities of 1507 5 Conclusions and Outlooks

and 861 mAh $_2$ ⁻⁴ at 0.1 and 10 A $_2$ ⁻⁴, respectiv **EVALUATION FOR THE 12000 EXALUATION THE SECT AT ALL AT AL Example 19**
 Example 19 the $2^x(L$ *Recurreduen,* 2022, 28(12), 2219013 (8 of 13)

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and 861 mAh $_2$ ⁻¹ at 0.1 and 10 A $_2$ ⁻¹, respectively.

On the other hand, Se and Te doping in SPAN can $m_1/k^2\%$ (*L Electrochem*ical 2022, 28(12), 2219013 (8 of 13)

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On the other hand, Se and Te d electrolyte, and delivered mgn capacites of 1507

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on the other hand, Se and Te doping in SPAN can

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depende In the unit of small is the material, se all the unit of small is the state is the state in the compatibility of the state is with set can be the state of the shemical reactivity⁶⁷⁰. Chen et al. introduced 6at⁹⁶ batter givental several of the interests with EN cathodes a particles with SC cathods are leadingly advanta-
Sc into SPAN cathods and found that the irreversible
several moderate with SCC cathods are relatively advanta-
Sc into increation in Section and Theoretic and American Caroline and American conducts in SEAN calibods are relatively awaitable since SPAN calibods and predominant in sulfur content. However, Li-Selectrolyte is genatly relieved se under a searly distance and point of the interesting and the ending sulfir besteen the mergy density is greatly relieved and predominant in sulfir content. However, Li-S electrolyte is greatly relieved^{fora}. The above

[2-methoxyethoxy]ethoxy)anthra-9,10-quinon
metal anode is originated from the soluble LiPSs gen-In morphology of SPAN usaully exminds builty with Li metal anode as well as electrolyte. The morphology of SPAN usaully exsults in long electronic conduction
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in the institute issues are due to the intrinsic sol-

distance and large contact resistance¹⁰⁸. From this point

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of view, extrinsic redox mediation is expert in centrated soluble LiPSs with limited charge transfer
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the proof electronic conductivity by coupling

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and the redox mediations. Therefore, both mechanism in-

a chemical pathway naming the poor electroche conductivity is volupling
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(DEAQ) as an extrinsic redox mediator, and t parmay. Znao et al. aaoptea the 1,5-bis(\angle -(\angle)))))), murthis \angle , (10, hunom

at 0.1 and 10 A窑g-¹ *Electrochem.*) 2022, 28(12), 2219013 (8 of 13)

cities of 1507 **5 Conclusions and Outlooks**

, respectively. The Li-S batteries with S/C or SPAN cathodes ex-

in SPAN can

hibit distinct characteristics (Figure 5). In de 28(12), 2219013 (8 of 13)

5 **Conclusions and Outlooks**

The Li-S batteries with S/C or SPAN cathodes ex-

hibit distinct characteristics (Figure 5). In detail, com-

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hibit distinct characteristics (Figure 5). In detail, com-
pared with Li-S batteries with SPAN cathodes, Li-S
batteries with S/C cathodes are re **Conclusions and Outlooks**
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pared with Li-S batteries with SPAN cathodes, Li-S
batteries with S/C cathodes are relatively advanta-
geous in ou hibit distinct characteristics (Figure 5). In detail, com-
pared with Li-S batteries with SPAN cathodes, Li-S
batteries with S/C cathodes are relatively advanta-
geous in output voltage, superior in energy density,
and pre pared with Li-S batteries with SPAN cathodes, Li-S
batteries with S/C cathodes are relatively advanta-
geous in output voltage, superior in energy density,
and predominant in sulfur content. However, Li-S
batteries with SP batteries with S/C cathodes are relatively advanta-
geous in output voltage, superior in energy density,
and predominant in sulfur content. However, Li-S
batteries with SPAN cathodes exhibit fantastic
lean-electrolyte tole geous 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 电化学(*J. Electrochem.*) 2022, 28(12), 2219013 (8 of 13)
gh capacities of 1507 **5 Conclusions and Outlooks**
10 A · g⁻¹, respectively. The Li-S batteries with S/C or SPAN cathodes ex-
e doping in SPAN can hibit distinct c

some to share a simula three-times the prediction in the prediction and predicted incores and the expert in the state of the state of the state in the state of the capacity decept in the capacity decept is predicted and t electrony is peatry increased and better and and conduction and the solution and the poor electronic state of the angle of the solution and the conductivity and eclerated and the capacity decay is inhibited. Consequently l meare that the vealure of state greate particles with the reader of the chemical value of the original electrochemical pathway of the original electrochemical pathway on the set of the set original electrochemical space of eterative and the equality vector is equality the effect and the equality. We be diped spectral and the along the standard of the area and the along the standard solo cycles with nearly 100% Coulombic efficiency literatur on vycies with insing throws containing emisative and the entropy and the state of the state and numeral over the species in the species of Section and the electronic stars and the distribution of $\sinh x = 0$ and $\sinh x$ ore, neterotomic oppuging can impower in execution. The section and exergence is the same time, resulting in greatly and opportunities of SC and SPAN cathodes in Li-S
cathodes at the same time, resulting in greatly and opp conductively and extended redox to the median method of the same time, resulting in greatly end batteries are respectively presented as follows.
 4.4 Extrinsic Redox Mediation
 4.4 Extrinsic Redox Mediation
 4.4 Extr eature sature the state and state and the space of the state state and battery performances. To rS/C cathodes, there are mainly three aspects of **4.4 Extrinsic Redox Mediation** challenges: kinetics issues under high rates, The Hauthous kinetics assumed valuation and the station of LiFSs in the conduct and the compatition of PAN by the morphology of SPAN usually exhibits bulk issues under low E/S ratios, and eathodic incompatities with severa **4.4 EXIFINE REQUA YOUGHDATION**
 EXIFINE REQUA YOUGHDATION
 EXIFINE REQUA YOUGHDATION
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generally longer stability. Note that above arguments
are based on the batteries with SPAN cathodes exhibit fantastic
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generally longer stability. Note that above arguments
are based on the average level according to reported
literatures. lean-electrolyte tolerance, higher rate capability, and
generally longer stability. Note that above arguments
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literatures. Therefore, Li-S batteries with S/C or SPAN
cat 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 re-
sear 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 re-
search directions shall be different. Key challenges
and o literatures. Therefore, Li-S batteries with S/C or SPAN
cathodes face different challenges, and the future re-
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batteri cathodes face different challenges, and the future re-
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and opportunities of S/C and SPAN cathodes in Li-S
batteries are respectively presented as follows.
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challenges: kinet 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 lo batteries are respectively presented as follows.
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bility with Li metal anode as well as electrolyte. The first t challenges: kinetics issues under high rates, kinetics issues under low E/S ratios, and cathodic incompati-
bility with Li metal anode as well as electrolyte. The
first two kinetic issues are due to the intrinsic sol-
id-l issues under low E/S ratios, and cathodic incompati-
bility with Li metal anode as well as electrolyte. The
first two kinetic issues are due to the intrinsic sol-
id-liquid-solid reaction pathway, which renders con-
centra bility with Li metal anode as well as electrolyte. The
first two kinetic issues are due to the intrinsic sol-
id-liquid-solid reaction pathway, which renders con-
centrated soluble LiPSs with limited charge transfer
rates first two kinetic issues are due to the intrinsic sol-
id-liquid-solid reaction pathway, which renders con-
centrated soluble LiPSs with limited charge transfer
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interfa id-liquid-solid reaction pathway, which renders con-
centrated soluble LiPSs with limited charge transfer
rates and low diffusion rates dominating the cathodic
interfacial kinetics. Therefore, both mechanism in-
vestigatio ntrated soluble LiPSs with limited charge transfer
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to address this is rates and low diffusion rates dominating the cathodic
interfacial kinetics. Therefore, both mechanism in-
vestigation and kinetic promoters are specially need-
ed to address this issue. The incompatibility with Li
metal an 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 cat vestigation 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, corrodi ed to address this issue. The incompatibility with Li
metal anode is originated from the soluble LiPSs gen-
erating at cathode, diffusing to the anodic compart-
ment, corroding the Li metal, and rendering rapid an-
ode fai

cathodes.

This work was supported by Natural Scientific

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Foundation of China (22100007), Beijing hatitute of

Technology Research Fund Program for Young Schol-

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 Euronometric carbon and the subset content in turn. Due to the unclear chemical struc-**Example 19**

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Dr. Xue-Qiang Zhang for the helpful discussion.

Figure 5 (comparison of Li-**Surface of the structure and the Tsinghua University Initiative Scientific

Figure 5 Comparison of Li-S batteries with SC or SPAN

EP Research Program. We thank Dr. Wei-Jing Chen and

EP Research Program. We thank Dr. We Example 19** Research Program. We thank Dr. Wei-Jing Chen and
 Equire 5 Computison of Li-S batteries with SC or SPAN
 Equire 5 Computison of Li-S batteries with SC or SPAN
 Example 2 (1) Share 1, Song 8 V, Wang 1, Z **Bonds and the set in t Figure 5** Compatison of Li-S batteries with SC or SPAN
 References:

[1] Shen L, Song Y W, Wang J, Zhao C X, Bi C X, Sun S Y,

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alteriors or exponentially ethodes.
 References:

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1) Shen, 1, Nong 2, Nhang Q. Synergistic cathosis on da-

al-atom sites for high-performance lithium-sulfor batteries

the precursor composition (1) Shen L, Song Y W, Wang J, Zhao C X, Bi C X, Sin S Y,

2) 24 al-atom sites for high-performance lithium-sulfite burstly is a change of the B (2) 2022 (2022) (2022) (2022) (2022) (2023)

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al-atom sites for high-performance limin-sultir state-

the precursor composition and structure may alter the

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PAN erystallinity, and tion are two effective approaches. In detail, tuning

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PAN ery the precursor composition and structure may alter the

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reacting with the PAN precursor. Optimizing the subtrized spinel cultos all-solid-

manufacturing IN crystallinity, and make more clemental sulfur
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manufacturing process (e.g., introducing electrosyin-

ming technique), on the other hand, may reduce it as W_N D, X_Li, Li, L_i, Wang S, X_N P, Li, Li, Now S, Li, Li, manufacturing process (e.g., introducing electrospin-

in Wu F, Liu L, Wang S, Xu J R, Liu P S, Yan W L, Peng J,

migreching techniques the sulfure in the columer based on the rand, may reduce the sulfure simulations is s ming technique), on the other hand, may reduce the

wo D X, Li H. Soild state ionies -selected topics and new

account in turn. Due to the unclear chemical struc-

content in turn. Due to the unclear chemical struc-
 $\frac{$ amount of conductive carbon and increase the sulfur

originations of the integral structure distantions and the tender chemical structure (separations on SPAN cathodes may be confused

ture, researches on SPAN cathodes ma content in turn. Due to the unclear chemical struc-

ture, researches an PAN calmbolic intention and machine-learning molecular dynamics simulature, researches on PAN canbodis may be confused into the machine-learning mol ture, researches on SPAN cathodes may be confused

into its one the liquid electrolyte for ecchargeable batteries [J].

Spectroscopics are cxpected to probe the specific

spectroscopics are cxpected to probe the specific

 $28(12)$, 2219013 (9 of 13)
with SPAN cathodes are promising for large-scale
energy storage due to their advantages of low costs
and long cycling lifespan. Witnessing the recent pro-
gresses of S/C and SPAN cathodes for $28(12)$, 2219013 (9 of 13)
with SPAN cathodes are promising for large-scale
energy storage due to their advantages of low costs
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gresses of S/C and SPAN cathodes for $28(12)$, 2219013 (9 of 13)
with SPAN cathodes are promising for large-scale
energy storage due to their advantages of low costs
and long cycling lifespan. Witnessing the recent pro-
gresses of S/C and SPAN cathodes for 28(12), 2219013 (9 of 13)
with SPAN cathodes are promising for large-scale
energy storage due to their advantages of low costs
and long cycling lifespan. Witnessing the recent pro-
gresses of S/C and SPAN cathodes for Li-S 28(12), 2219013 (9 of 13)

with SPAN cathodes are promising for large-scale

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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