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2022-11-28

Advances and Atomistic Insights of Electrolytes for Lithium-Ion Batteries and Beyond

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for Lithium-Ion Batteries and Beyo and Technology, Department of Chemical Engineering, Tsinghua University, Beijing 100084, China)

Abstract: Electrolytes and the associated electrode-electrolyte interfaces are crucial for the development and application of
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interministic modeling infinitent interface in this regard,

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for Lithium-Ion Batteries and Beyond

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5. Instit 2. Institute of Materials Research, Tsinghua Shenzhen International Graduate School, Tsinghua University, Shenzhen 518055, China; 3. School of Chemical Engineering and Advanced Materials, The University of Adelaide, Adelai **Extraine the system are these are particularly in the system and Advanced Materials, The University of Adeliade, Adelaide, SA 5005, Australia; 4. Beijing Key Laboratory of Green Chemical Reaction Engineering and Technolo** Surface and the probability and the children in the method in the state of the probability, the contexts of the method A deliade, SA 5005, Australia, 4. Beijing Key Laboratory of Green Chemical Reaction Engineering and Are the constrained in the second of Chemical Engineering (3.1900) and the constrained revertibles) and Technology of the Chemical Engineering Tsinghua University, Beijing 100084, China)
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 Key words: lithium-ion batteries; electrolytes; atomistic modeling; solid electrolyte interphase; solidgude for hance electrolyte design. Finally, we highlight the prospects of theoretical sectemny of novel electrolytes.
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ducesigned as well. This is particularly important to make a-formed electrode-electrologistic inter-
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because the as-formed electrode-electrivite inter-

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Tables 1 and 2. The high dielectric constant of cyclic
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Tables 1 and 2. The high dielectric constant of cyclic
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Li⁺ transport between (e.g., ethyl methyl carbonate (EMC), diethyl carbon-

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| | rinsic properties of the associated electrolyte. | | to mitigate the viscosity and lower the melting point | |
|--|--|--|---|-------------|
| ctrolytes of LIBs ctrolyte Interphase | Understanding Conventional Ele- Electrolyte Components and Solid Ele- After decades of intensive development, the skele- composition of the electrolyte for commercialized Bs has long been established ^[8] . Although differing ong manufacturers and scenarios, the majority of ctrolytes employ the carbonate electrolyte formula h cyclic carbonates (e.g., ethylene carbonate (EC), d propylene carbonate (PC)), linear carbonates | | of the electrolyte; the lithium salt is required to be dissociated in the electrolyte to some extent which enables the free Li ⁺ transport, and should be electro- chemically stable toward both electrodes. Moreover, the electrically insulating and ionically conductive solid electrolyte interphase (SEI) between anode and electrolyte is spontaneously formed by the reduction reactions of electrolytes in the initial cycles ^[9] . An optimal SEI layer passivates the anode surface against further side reactions while facilitating Li ⁺ transport ^[10-12] . When the electrolyte reductive stability limit is reached during the initial charging of a Li-ion | |
| | | | Table 1 Common carbonate solvents in LIB electrolytes. | |
| | Cyclic carbonate | | Linear carbonate | |
| EC | ${\rm P}{\bf C}$ | DMC | DEC | ${\rm EMC}$ |
| | Ω | | | |
| High dielectric constant | | | Low viscosity Lowmelting point | |
| | LiBF ₄ | Table 2 Common salts in LIB electrolytes. LiPF ₆ | LiTFSI | |
| | | F_{μ} | | |

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SEI co on the hattery anode surface^[13]. The initially formed species, which could introduce additional error and

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formation is to understand the solvation chemistry of eyclic earbonate, and linear carbonate in tailoring the solvention stract with the solvention structure, ionic conduction, and reduction stract expecially dependent on electrolyte species. When electrolytes react with the solvation structure, ionic conduction, and reduction
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a r Most recently, the internally referenced diffusion-or-
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i dered spectroscopy (IR-DOSY)^[32,33] has been utilized
to construct a solvating power series, which provides
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as-e to construct a solvating power series, which provides
a reliable quantitative measure of the lithium solvat-
ing power of common electrolyte solvents. Using the
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endent reactions—in-

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bond cleavage, and states of solvent moieties, the scaling of peak area in-

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products, and thermal decomposition, etc.) as well as

continuous electrolyte reduction^{[23, 1}R in as loos been

consinuous electrolyte reactions and the solution and consistent of the solution

observed that gases (part continuous electrolyte reduction^{121,214}. It has also been

onserved that gases (particularly H₃, C_{H4}, and CO) solvent in a binary or termacy clearely the system and their aging depend both on the specific components observed that gasses (particularly H₃, C₂H₁, and CO) solvent in a binary or termary electrolyte system. This
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sequenc sequence of reactions and their aging depend both on

the specific components and concentration, including

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additives and impurities, of the balk electrolyte, and the specific components and concentration, including

drolyte system and will be especially instructive in the

dditivies and impurities, of the bulk electrolyte, and selection of suitable solvent/co-solvent for lithium
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selection of suitable solvent/co-solvent for lithium

on the specific anode material and its surface chem-

batistics and alternative approach, computational methods on the specific anode material and its surface chem-

batteries.

Sity and structure¹¹³. Hence, in principle, every anode

As an alternative approach, computational methods

material requires a differently tialored elec istry and structure⁽¹⁹⁾. Hence, in principle, every anode

material requires a differently tailored electrolyte, to

material requires and interestigate determines and classical molecular dynamics (AMD)^{19:48}

anchieve material requires a differently tailored electrolyte, to

including ab initio molecular dynamics (AIMD)^{{15-30}

calibuse maximal compatibility—both electrochemi-

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eally as suredlar smoothing results in modeling the
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2.2 Solvation Structure

2.2 Solvation Structure

2.7 Solvation Structure

2.7 Solvation Structure

1. The first step to understand the mechanisms of SFI et al.^{ps}I everaled the different **2.2 Solvation Structure** solvation and transport behaviors. For example, Home first step to understand the solvation chemistry of equice arbon-
the first term (solven the convention of SEI et al. ^{pa} revealed the differ The first step to understand the mechanisms of SFI et al.¹⁸⁶ revealed the different roles of the Li salt,
formation is to understand the solvation chemistry of explic carboate, and linear carbonate in talioning the
decl anode surface to form the SEI, the reaction products

are theight dependent on the starting recestants as well

are thight dependent on the starting recestants as well

as their immediate solvation environment⁸⁴. There a reliable quantitative measure of the lithium solvating power of common electrolyte solvents. Using the as-established solvating power series, researchers can easily evaluate the solvation state of an individual solvent ing power of common electrolyte solvents. Using the as-established solvating power series, researchers can easily evaluate the solvation state of an individual solvent in a binary or ternary electrolyte system. This repor as-established solvating power series, researchers can
easily evaluate the solvation state of an individual
solvent in a binary or ternary electrolyte system. This
reported solvating power series will provide insight-
ful easily evaluate the solvation state of an individual
solvent in a binary or ternary electrolyte system. This
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ful guidance for the future design of functional elec-
tro solvent in a binary or ternary electrolyte system. This
reported solvating power series will provide insight-
ful guidance for the future design of functional elec-
trolyte system and will be especially instructive in the ternary electrolyte system. This
wer series will provide insight-
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be especially instructive in the
solvent/co-solvent for lithium
proach, computational methods
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system and will be especially instructive in the
on of suitable solvent/co-solvent for lithium
ess.
an alternative approach, c ful guidance for the future design of functional electrolyte system and will be especially instructive in the selection of suitable solvent/co-solvent for lithium batteries.
As an alternative approach, computational metho trolyte system and will be especially instructive in the selection of suitable solvent/co-solvent for lithium batteries.

As an alternative approach, computational methods including *ab initio* molecular dynamics (AIMD)[[] selection of suitable solvent/co-solvent for lithium
batteries.
As an alternative approach, computational methods
including *ab initio* molecular dynamics (AIMD)^{[34,36]</sub>
and classical molecular dynamics (MD)^[37,41] sim} batteries.

As an alternative approach, computational methods

including *ab* initio molecular dynamics (AIMD)<sup>[34.56]</sub>

and classical molecular dynamics (MD)^[37.41] simula-

tions have shown satisfying results in model</sup> As an alternative approach, computational methods
including *ab* initio molecular dynamics (AIMD)^{[34.36}]
and classical molecular dynamics (MD)^[37.41] simula-
tions have shown satisfying results in modeling the
solvati including *ab* initio molecular dynamics (AIMD)^{[34,36}]
and classical molecular dynamics (MD)^[37,41] simula-
tions have shown satisfying results in modeling the
solvation and transport behaviors. For example, Hou
et al and classical molecular dynamics $(MD)^{[3741]}$ simula-
tions have shown satisfying results in modeling the
solvation and transport behaviors. For example, Hou
et al. ^[26] revealed the different roles of the Li salt,
cycl tions have shown satisfying results in modeling the
solvation and transport behaviors. For example, Hou
et al. ^[26] revealed the different roles of the Li salt,
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cyclic carbonate, and linear carbonate in tailoring the
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r et al. ^[26] revealed the different roles of the Li salt,
cyclic carbonate, and linear carbonate in tailoring the
solvation structure, ionic conduction, and reduction
reactions of mixed-carbonate electrolytes (MCEs,
Figu cyclic carbonate, and linear carbonate in tailoring the
solvation structure, ionic conduction, and reduction
reactions of mixed-carbonate electrolytes (MCEs,
Figure 1a), the one of broad commercial interest. The
EC-base (solvation structure, ionic conduction, and reduction
reactions of mixed-carbonate electrolytes (MCEs,
Figure 1a), the one of broad commercial interest. The
EC-base (1.2 mol·L⁻¹ LiPF₆ in EC) and Gen2 (1.2
mol·L⁻¹ LiP reactions of mixed-carbonate electrolytes (MCEs,
Figure 1a), the one of broad commercial interest. The
EC-base (1.2 mol·L⁻¹ LiPF₆ in EC) and Gen2 (1.2
mol·L⁻¹ LiPF₆ in EC and EMC) electrolytes were in-
vestigated

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mantly "exit-entry" type with identified energetic and
electrostatic origins (Figure 1d). This mechanism ex-
framework provides a new and accurate tool for de-
plains the non-intuitive preferential enrousd of cyclic
errif mantly "exit-entry" type with identified energetic and

trolytes (Figure 2a). The application of the theoretical

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pl electrostatic origins (Figure 1d). This mechanism ex-

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planism the one-intuitive preferration results remov plains the non-intuitive preferential removal of cyclic scribing the ion transport that can successfully cap-
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(d) Sample trajectory of EC-PF, exchange in the Ge **35.6%**
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GG) species in the Gen2 electrolyte. The population separated ion pairs (SSIP), conta **Figure 1** (a) These representative solvation structures of solven separate (A(16) and aggregate (A(16) species in the tienz detective)te. The population of SSIP, CIP, and AGG species in (b) FC-base and (c) Gen2 electroly **Figure 1** (a) Three representative solvation structures of solvation separated ion pairs, (SSIP), contact ion pairs, (CIP), and aggregate (AGG) species in the Gen2 electrolyte. He population of SSIP, CIP, and AGG species (AOG) species in the Gm2 electrolyte. The population of SSIP, CIP, and AGG species in (b) EC-base and (c) Gm2 electrolytes.

(d) Sample trajectory of EC-PF₆ exchange in the Gm2 electrolyte. Reproduced with permission fr (d) Sample trajectory of EC-Pi^r exchange in the Gen2 electrolyte. Reproduced with permission from ref ^{[30}]. Copyright 2019 The
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nantly "exit-entry" type with identified energetic and

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electrostatic origins (Figure 1d). This mechanism ex-

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P, and AGG species in (b) EC-base and (c) Gen2 electrolytes.

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trolytes (Figure 2a). The appl and AGG species in (b) EC-base and (c) Gen2 electrolytes.

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2b, i framework provides a new and accurate tool for describing the ion transport that can successfully cap-
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2b, in contrast to the EC-base electrolyte, the Gen2
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2b, in contrast to the EC-base electrolyte, the Gen2
electrolyte exhibited greater self-diffusion coeffi-
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Fig. permission from ref¹⁹⁹. Copyright 2021 The Authors. (b) The calculated self-di permission from $\epsilon^{n/6}$. Copyright 2021 The Authors, (b) The calculated self-attifacion coefficients of electrolyte species (EG, InMA;

FEC, PF₆, and Li) in the EC-base. ECF, Con2, and Grare Beterolytes, (Copyright 20 EEC, Fr₅, and Li) in the EC-bass, ECF, Gen2, and Geni² electrolytes. (c) The experimental (Exp) and calculated (Cal) ionic con-
diactivity at 298 K. Reproduced with permission from ref^{por}. Copyright 2012 The Authors ductivity at 298 K. Reproduced with permission from ret²⁰. Copyright 2019 The Authors, (a) Mechanistic model of the SEI formation. Reproduced with permission from me^{rgin}. Copyright 2012 The Authors, (color on line)
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 3 Novel Electrochemistry in Li-Ion

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3.1 Anodes

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 3.1 Anodes

There is intense interest in developing new anode

materials that store highe **3 Novel Electrochemistry in Li-Ion**
Batteries and Associated Challen-
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3.1 Anodes
There is intense interest in developing new anode
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3.1 **Anodes**
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3.1 Anodes
There is intense interest in developing new anode
materials that store higher densities of lithium^[80]. One
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3.1 Anodes
There is intense interest in developing new anode
materials that store higher densities of lithium^[80]. One
of the most promising anode materials for future high
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for vecanters (CF and Rb⁻)²⁰¹, fluorocthylene carbonate (FEC)

for vecante, by utilizing modified salts and elec-

additives^{185,39}. Beyond the Li-Si convers for example, by utilizing modified salts and elec-

ventilives^{us, an}d high-concentration electrolytes^{is)}.

Reyend the Li-Si conversion chemistries, lithium
 Exact consideration of the electrolyte design has

metal i rolyte additives,^{185,195}.

Heygond the 11-55 conversion chemistries, lithium Less consideration of the electrolyte design has

Heygond the 11-55 conversion chemistries, lithium Less consideration of the electrolyte desig Beyond the Li-Si conversion chemistries, lithium

Less consideration of the electrolyte design has

metres bare and stability promising anole to further

increase the energy density of batteries because of its candode el metal is regarded as another promising anode to further
increase the energy density of harteries because of its cathode-electrotyle interplace (CFI) for conventional
inigh theoretical specific equestivy (3860 mAh rg') and increase the energy density of batteries because of its

intight theoretical specific capacity (3860 mAh $_2$) and

interaction potential (-3.04 V vs, standard hydrogen

is hower detection potential (-3.04 V vs, standard high theoretical specific capacity (3860 mAh $_2$ °) and LHBs. However, the application of high-voltage cather
low reduction potential (-3.04 V vs. standard hydrogen ode materials such as Ni-rich cathode materials¹⁹
det

ditives $(Cs^+$ and Rb^+ ^[59], fluoroethylene carbonate (FEC)

Example 18
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 eromated materials along with the cathode-electrolyte interprise (CEI) f Example 14
 Containerally and discharging process of Si anodes. (b) Illustration of the un-

approaches are employed to facilitate the *in situ* for-

mation of a stable SEI layer, such as metal cation ad-

ditives (C **Dendritic**
 Example 18 growth
 Example 20 growth
 Example 20 growth
 Example 20 growth and discharging process of Si anodes. (b) Illustration of the un-

approaches are employed to facilitate the *in situ* for-
 Dendritic
 Example 18 and discharging process of Si anodes. (b) Illustration of the un-

approaches are employed to facilitate the *in situ* for-

mation of a stable SEI layer, such as metal cation ad-

ditives (Cs⁺ **Dendritic**
 Example 10 growth
 Example 20 growth
 Example 20 and discharging process of Si anodes. (b) Illustration of the un-
 Approaches are employed to facilitate the *in situ* **for-

additives (Cs^{*} and Rb⁺) EXECT:** FUNCTIVE THE THE THE THE CONDED IN the paper of the same of the same approaches are employed to facilitate the *in situ* formation of a stable SEI layer, such as metal cation additives (Cs⁺ and Rb⁻¹⁹⁶⁹). fluo g and discharging process of Si anodes. (b) Illustration of the un-
approaches are employed to facilitate the *in situ* for-
mation of a stable SEI layer, such as metal cation ad-
ditives (Cs⁺ and Rb⁻)^{[50}], fluoroet gand discharging process or 5 anodes. (b) intestantion of the direct
approaches are employed to facilitate the *in situ* for-
mation of a stable SEI layer, such as metal cation ad-
ditives (Cs^{*} and Rb⁻)^[30], fluoroe approaches are employed to facilitate the *in situ* for-
mation of a stable SEI layer, such as metal cation ad-
ditives (Cs^{*} and Rb^{*})^[59], fluoroethylene carbonate (FEC)
additive^[60], and high-concentration electr approaches are employed to facilitate the *in situ* for-
mation of a stable SEI layer, such as metal cation ad-
ditives (Cs⁺ and Rb⁻)^{[50}], fluoroethylene carbonate (FEC)
additive^[60], and high-concentration electr approaches are employed to facilitate the *in situ* for-
mation of a stable SEI layer, such as metal cation ad-
ditives (Cs⁺ and Rb⁻¹^[69], fluoroethylene carbonate (FEC)
additive^[60], and high-concentration electr mation of a stable SEI layer, such as metal cation additives (Cs⁺ and Rb^{-1)[30}], fluoroethylene carbonate (FEC) additive^[60], and high-concentration electrolyte^[61].
3.2 Cathodes
Less consideration of the electr ditives (Cs⁺ and Rb^{-y[69]}, fluoroethylene carbonate (FEC)
additive^[60], and high-concentration electrolyte^[61].
3.2 Cathodes
Less consideration of the electrolyte design has
been given to the cathode materials a additive^[60], and high-concentration electrolyte^[61].
3.2 **Cathodes**
Less consideration of the electrolyte design has
been given to the cathode materials along with the
cathode-electrolyte interphase (CEI) for convent **3.2 Cathodes**
Less consideration of the electrolyte design has
been given to the cathode materials along with the
cathode-electrolyte interphase (CEI) for conventional
LIBs. However, the application of high-voltage cath-Less consideration of the electrolyte design has
been given to the cathode materials along with the
cathode-electrolyte interphase (CEI) for conventional
LIBs. However, the application of high-voltage cath-
ode materials been given to the cathode materials along with the cathode-electrolyte interphase (CEI) for conventional LIBs. However, the application of high-voltage cath-
ode materials such as Ni-rich cathode materials⁶³¹ and cationcathode-electrolyte interphase (CEI) for conventional
LIBs. However, the application of high-voltage cath-
ode materials such as Ni-rich cathode materials^[62] and
cation-disordered rocksalt-type cathode materials^[62]
h LIBs. However, the application of high-voltage cath-
ode materials such as Ni-rich cathode materials^[62] and
cation-disordered rocksalt-type cathode materials^[62] and
has raised new challenges for designing electrolyt

the *H*(*L Electrochem.*) 2022, 28(11), 2219007 (7 of 18)

and diffusion of soluble intermediates, polysulfides

(PS), and a series of concurrent side reactions at in-

efficiency, and uniform surface morphore

terface

sections.

 $\#E\#(J. Electron)$ 2022, 28(11), 2219007 (7 of 18)
and diffusion of soluble intermediates, polysulfides electrochemical performance, improved Coulombic
(PS), and a series of concurrent side reactions at in-
terfaces or in solut $\#l\&\#(J. \text{Electrochem.})$ 2022, 28(11), 2219007 (7 of 18)
and diffusion of soluble intermediates, polysulfides
(PS), and a series of concurrent side reactions at in-
terfaces or in solution^[64]. The shuttle effect not only a the $\{E\}^{\omega_k}(I. Electronem.)$ 2022, 28(11), 2219007 (7 of 18)
and diffusion of soluble intermediates, polysulfides
(PS), and a series of concurrent side reactions at in-
efficiency, and uniform surface morphology of the
terfaces **EVALUATE 1999**
 Example 12: Electrochem.) 2022, 28(11), 2219007 (7 of 18)

and diffusion of soluble intermediates, polysulfides electrochemical performance, improved Coulombic

(PS), and a series of concurrent side reactions at in-

ef $\frac{d}{dt}\mathcal{L}^{\omega}(I, Bleertochem.)$ 2022, 28(11), 2219007 (7 of 18)

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(PS), and a series of concurrent side reactions at in-

efficiency, and uniform surface morphology of the

t \pm *KE+(L Electrolem.*) 2022, 28(11), 2219007 (7 of 18)
and diffusion of soluble intermediates, polysulfides
(PS), and a series of concurrent side reactions at in-
efficiency, and uniform surface morphology of the
terfac **in controlling the deleterious shuttle reactions and ship controlling the section of Soluble intermediates, polysulfides electrochemical performance, improved Coulombic (PS), and a series of concurrent side reactions at EVALUATION Example 12 EXALUATION EXALUATION EVALUATION EVALUATION EXALUATION EXALUATION EXALUATION** in the diffusion of soluble intermediates, polysulfides

in the diffusion of soluble intermediates, polysulfides

(PS), and a series of concurrent side reactions at in-

effeciency, and uniform surface morphology of the \pm (*E.*) \pm (*E.*) \pm (*E.*) \pm (*E.*) \pm (*E.*) \pm (*E.*) \pm (*E.*) \pm (*E.*) \pm (*E.*) \pm (*E.*) \pm (*E.*) \pm (*E.*) \pm (*E.*) \pm (*E.*) \pm (*E.*) \pm (*E.*) \pm (*E.*) \pm (*E.*) \pm ($\pm 1/2+2/4$. *Electrochem*, 2022, 28(11), 2219007 (7 of 18)

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(PS), and a series of concurrent side reactions at in-

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 EVALUATION (PS), and a series of concurrent side reactions at in-

electric electric part $\pm \frac{1}{2}E^2$ (*L. Electrochem.*) 2022, 28(11), 2219007 (7 of 18)
termediates, polysulfides electrochemical performance, improved Coulombic
rent side reactions at in-
efficiency, and uniform surface morphology of the
res th (*k*²/²(*L Electrochem.*) 2022, 28(11), 2219007 (7 of 18)

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(PS), and a series of concurrent side reactions at in-

efficiency, and uniform surface morpholog and diffusion of soluble intermediates, polysulfides electrolegation, and a minima surface morphology of the

(PS), and a series of consurrent side reactions at in-

efficiency, and uniform surface morphology of the

terf and diffusion of soluble intermediates, polysulfides

detrochemical performance, improved Coulombic

(FS), and a series of concurrent side reactions at in-

efficiency, and uniform surface morphology of the

terfaces or i (PS), and a series of concurrent side reactions at in-

efficiency, and uniform surface morphology of the

terfaces or in solution¹⁶⁴¹. The shuttel effect not only anode (Figure 4b). To understand the composition and

g terfaces or in solution¹⁰⁴¹. The shuttle effect not only
anode (Figure 4b). To understand the composition and
gives rise to a severe loss of active materials from the
memphology of the Si SEI formed with FEC, exten-
cat es rise to a severe loss of active materials from the morphology of the Si SEI formed with FEC, exten-

hold expling life. Rational selection ror columbic efficiency sive experiments using scanning transmission electrolyt cathode, but also renders poor Coulombic efficiency
and cyincomiats using scanning transmission clear-
and edetrolytes with reduced PS solubility is critical
to resocpy (FEIEM)^{ne)}, clearbor and
in encorrelation and elec and cycling life. Rational selection or design of opti-

tron microscopy (STEM)⁹⁹, electron energy loss spec-

and cleartiolytical selections shuttle reactions and

troscopy (F.TIR)¹⁵⁸, Pourie-transform infrared specmal electrolytes with reduced PS solubility is critical

in contorling the detectroius shuttle reactions and troscopy (FFILS)⁽¹⁰⁷⁾, Fourier-transform infrared species

protecting the detectroius shuttle, reactions and t in controlling the deletrious shuttle reactions and

protecting the celetricous shuttle reactions and

protecting in claristic strateg⁶⁶. In fact, Li alsts used

in traditional LHBs (e.g., LiPF₆ LiBF₆) and solvents
 protecting the electrode surface⁸⁸. In fact, Li salts used

or py (XPS)^{86,71}, atomic force microscopy (AFM)^{gr,71},

in traditional LIEs (e.g., LiFF_p, LiFF_p) and solvents

(e.g., ester, carbonates, phosphates) are in traditional LIBs (e.g., LiPF₆, LiBF₁) and solvents

focal, extraptional concentration for nuclear magnetic resonance (NMR)^{is, 78}, differential

(L.S, ester, carbonats, phosphartes) are unsuitable for nuclear magn (e.g., ester, carbonates, phosphates) are unsuitable for

nuclear magnetic resonance (NMR)^{(6, 31}), differential

Li-S battery applications due to their parasitic reac-

electrochemical mass spectrometry (DEMS)^[79], an Li-S buttery applications due to their parasitic reac-

electrochemical mass spectrometry (DEMS)¹⁷⁶, and

troly explores with 1Pss⁶⁶. Therefore, so far, an chercal elec-

troly explorement in the of the secondary on t tions with PSs⁽ⁱⁱ⁾. Therefore, so far, an etheraal electronic time-of-flight secondary ion mass spectrometry

trolyte system with 1 mol - L¹-LITES1 in a binary sol-

co-workers^{(sav}) exgested that a 10% contracted de vent mixture of 1,3-dioxolane and 1,2-dimethoxyeth-

aco-workers¹⁵³] suggested that a 10% content of

However, although clttes content of the continue of the popularity in the However, although clttes are non-reactive w ane (DOL/DME) is considered default for Li-S cells.

can provide the optimal improvement

However, although ethers are non-reactive with poly-

surfleds, they dissolver polysulfides at high-concentra-

point of the standa wever, although ethers are non-reactive with poly

carbonate electrolyte in terms of impedance and ca-

fides, they dissolve polysulfides at high-concentra-

pared with a standard *ECDMC* electrolyte, the use of

In summa sulfides, they dissolve polysulfides at high-conentra-

incom an denote encourage parastic is hutting^{one}.

In summary, issues including but not limited to introduce the parad with a standard EC/DMC cletrolyte, the use o tion and hence encourage parasitic shuttling⁵⁶¹. In summary, issues including but not limited to interest and the stability modified the organic SEI componentiality and parameterizal constrained the stability and includ In summary, issues including but not limited to in-

termediate dissolution, dendritic lithium deposition, ments derived from LEDC and soluble linear

termediate dissolution, dendritic lithium deposition, ments derived fr termediate dissolution, dendritic lithium deposition, nents derived from LEDC and soluble linear
and pulverization are found to be deriminand for the oligomers to soluble and insoluble encslinked poly
equacity and cycling and pulverization are found to be detrimental for the oligomers to soluble and insoluble crosslinked poly
equacity and cycling performance of notel carbotal (erhylene oxideal polymers (such as lithium poly
and/or anode ma equacity and cycling performance of novel cathode

and cheriophose ond polymers (such as lithium poly

cards material sate above in Figure 3b.

(windence above a and/or anode materials, as shown in Figure 3b.

Structures can be concerned by the anode surface and resist voltime expansions

Structures and socialized high-concernention electrolytes have Meanwhile, FEC incurs increase Strategies including high-concentration electrolytes

and ocalized and resist volume expansion^{8, 75, 70}

and localized high-concentration electrolytes have Meanwhile, FEC incurs increased formation of Li-F,

been consid and localized high-concentration electrolytes have

Meanwhile, FEC incurs increased formation of Lif.

because a value approaches to mitigate the and less formation of Li₅Co, and organic earborate

issues. A more detail been considered as viable approaches to mitigate the and less formation of LicO, and organic carbonate issues. A more clatiled discussion on these noed species, easiling in an overall lower interfacial electrolyte designs $28(11)$, 2219007 (7 of 18)
electrochemical performance, improved Coulombic
efficiency, and uniform surface morphology of the
anode (Figure 4b). To understand the composition and
morphology of the Si SEI formed with FEC 28(11), 2219007 (7 of 18)
electrochemical performance, improved Coulombic
efficiency, and uniform surface morphology of the
anode (Figure 4b). To understand the composition and
morphology of the Si SEI formed with FEC, ex 28(11), 2219007 (7 of 18)
electrochemical performance, improved Coulombic
efficiency, and uniform surface morphology of the
anode (Figure 4b). To understand the composition and
morphology of the Si SEI formed with FEC, ex 28(11), 2219007 (7 of 18)
electrochemical performance, improved Coulombic
efficiency, and uniform surface morphology of the
anode (Figure 4b). To understand the composition and
morphology of the Si SEI formed with FEC, ex 28(11), 2219007 (7 of 18)

electrochemical performance, improved Coulombic

efficiency, and uniform surface morphology of the

anode (Figure 4b). To understand the composition and

morphology of the Si SEI formed with FEC 28(11), 2219007 (7 of 18)
electrochemical performance, improved Coulombic
efficiency, and uniform surface morphology of the
anode (Figure 4b). To understand the composition and
morphology of the Si SEI formed with FEC, ex 28(11), 2219007 (7 of 18)
electrochemical performance, improved Coulombic
efficiency, and uniform surface morphology of the
anode (Figure 4b). To understand the composition and
morphology of the Si SEI formed with FEC, ex 28(11), 2219007 (7 of 18)
electrochemical performance, improved Coulombic
efficiency, and uniform surface morphology of the
anode (Figure 4b). To understand the composition and
morphology of the Si SEI formed with FEC, ex 28(11), 2219007 (7 of 18)
electrochemical performance, improved Coulombic
efficiency, and uniform surface morphology of the
anode (Figure 4b). To understand the composition and
morphology of the Si SEI formed with FEC, ex 28(11), 2219007 (7 of 18)
electrochemical performance, improved Coulombic
efficiency, and uniform surface morphology of the
anode (Figure 4b). To understand the composition and
morphology of the Si SEI formed with FEC, ex 28(11), 2219007 (7 of 18)
electrochemical performance, improved Coulombic
efficiency, and uniform surface morphology of the
anode (Figure 4b). To understand the composition and
morphology of the Si SEI formed with FEC, ex 28(11), 2219007 (7 of 18)
electrochemical performance, improved Coulombic
efficiency, and uniform surface morphology of the
anode (Figure 4b). To understand the composition and
morphology of the Si SEI formed with FEC, ex 28(11), 2219007 (7 of 18)
electrochemical performance, improved Coulombic
efficiency, and uniform surface morphology of the
anode (Figure 4b). To understand the composition and
morphology of the Si SEI formed with FEC, ex 28(11), 2219007 (7 of 18)
electrochemical performance, improved Coulombic
efficiency, and uniform surface morphology of the
anode (Figure 4b). To understand the composition and
morphology of the Si SEI formed with FEC, ex 28(11), 2219007 (7 of 18)
electrochemical performance, improved Coulombic
efficiency, and uniform surface morphology of the
anode (Figure 4b). To understand the composition and
morphology of the Si SEI formed with FEC, ex $\frac{1}{2(11)}$, $\frac{1}{22}$, and uniform surface morphology of the anode (Figure 4b). To understand the composition and morphology of the Si SEI formed with FEC, extensive experiments using scanning transmission electron mic electrochemical performance, improved Coulombic
efficiency, and uniform surface morphology of the
anode (Figure 4b). To understand the composition and
morphology of the Si SEI formed with FEC, exten-
sive experiments usin efficiency, and uniform surface morphology of the
anode (Figure 4b). To understand the composition and
morphology of the Si SEI formed with FEC, exten-
sive experiments using scanning transmission elec-
tron microscopy (S anode (Figure 4b). To understand the composition and
morphology of the Si SEI formed with FEC, exten-
sive experiments using scanning transmission elec-
tron microscopy (STEM)^[69], electron energy loss spec-
troscopy (E morphology of the Si SEI formed with FEC, exten-
sive experiments using scanning transmission elec-
tron microscopy (STEM)^[69], electron energy loss spec-
troscopy (EELS)^[69], Fourier-transform infrared spec-
troscopy sive experiments using scanning transmission electron microscopy (STEM)^[67], electron energy loss spectroscopy (EELS)^[68], Fourier-transform infrared spectroscopy (XPS)^[6,71], atomic force microscopy (AFM)^{[72,73}], tron microscopy (STEM)^[67], electron energy loss spec-
troscopy (EELS)^[69], Fourier-transform infrared spec-
troscopy (FTIR)^[55, 70], X-ray photoelectron spectros-
copy (XPS)^[6, 71], atomic force microscopy (AFM) troscopy (EELS)^[69], Fourier-transform infrared spec-
troscopy (TTIR)^[55, 70], X-ray photoelectron spectros-
copy (XPS)^[6, 71], atomic force microscopy (AFM)^{[72, 73}],
hard X-ray photoelectron spectroscopy (HAXPES) troscopy (FTIR)^{155, 70}], X-ray photoelectron spectros-
copy (XPS)^{6, 71}], atomic force microscopy (AFM)^{172, 73}],
hard X-ray photoelectron spectroscopy (HAXPES)^{68, 74}],
nuclear magnetic resonance (NMR)^{16, 75}], dif copy (XPS)^{6,71}, atomic force microscopy (AFM)^{[72, 73},
hard X-ray photoelectron spectroscopy (HAXPES)^{[68, 74}],
nuclear magnetic resonance (NMR)^[6, 75], differential
electrochemical mass spectrometry (DEMS)^{[76}, an hard X-ray photoelectron spectroscopy (HAXPES)^{68, 74},
nuclear magnetic resonance (NMR)^{16, 75}, differential
electrochemical mass spectrometry (DEMS)^{[76}, and
time-of-flight secondary ion mass spectrometry
(TOF-SIMS)[[] nuclear magnetic resonance (NMR)^[6, 75], differential
electrochemical mass spectrometry (DEMS)^[76], and
time-of-flight secondary ion mass spectrometry
(TOF-SIMS)^[77], etc., have been undertaken. Lucht and
co-workers electrochemical mass spectrometry (DEMS)^[76], and
time-of-flight secondary ion mass spectrometry
(TOF-SIMS)^[77], etc., have been undertaken. Lucht and
co-workers^[58] suggested that a 10% content of FEC
can provide t time-of-flight secondary ion mass spectrometry
(TOF-SIMS)^{*rm*}, etc., have been undertaken. Lucht and
co-workers^[55] suggested that a 10% content of FEC
can provide the optimal improvement on the base
carbonate electro (TOF-SIMS)⁷⁷⁷, etc., have been undertaken. Lucht and co-workers^[53] suggested that a 10% content of FEC can provide the optimal improvement on the base carbonate electrolyte in terms of impedance and capacity retentio co-workers^[53] suggested that a 10% content of FEC
can provide the optimal improvement on the base
carbonate electrolyte in terms of impedance and ca-
pacity retention. Subsequent studies found that, com-
pared with a s can provide the optimal improvement on the base
carbonate electrolyte in terms of impedance and ca-
pacity retention. Subsequent studies found that, com-
pared with a standard EC/DMC electrolyte, the use of
10% FEC additi carbonate electrolyte in terms of impedance and ca-
pacity retention. Subsequent studies found that, com-
pared with a standard EC/DMC electrolyte, the use of
10% FEC additive modified the organic SEI compo-
nents derived pacity retention. Subsequent studies found that, com-
pared with a standard EC/DMC electrolyte, the use of
10% FEC additive modified the organic SEI compo-
nents derived from LEDC and soluble linear
oligomers to soluble a pared with a standard EC/DMC electrolyte, the use of 10% FEC additive modified the organic SEI components derived from LEDC and soluble linear oligomers to soluble and insoluble crosslinked poly (ethylene carbonate)), whi 10% FEC additive modified the organic SEI components derived from LEDC and soluble linear oligomers to soluble and insoluble crosslinked poly (ethylene carbonate)), which could better passivate the anode surface and resis nents derived from LEDC and soluble linear
oligomers to soluble and insoluble crosslinked poly
(ethylene oxide)-based polymers (such as lithium poly
(vinylene carbonate)), which could better passivate
the anode surface an oligomers to soluble and insoluble crosslinked poly
(ethylene oxide)-based polymers (such as lithium poly
(vinylene carbonate)), which could better passivate
the anode surface and resist volume expansion^{[6,75,78}].
Meanw (ethylene oxide)-based polymers (such as lithium poly
(vinylene carbonate)), which could better passivate
the anode surface and resist volume expansion^{[6,75,78}].
Meanwhile, FEC incurs increased formation of LiF,
and les ymers (such as lithium poly
ich could better passivate
st volume expansion^[6, 75, 78].
ncreased formation of LiF,
CO₃ and organic carbonate
overall lower interfacial
 $e^{[79]}$. Moreover, there is ev-
ss the LiPF₆ dec (vinylene carbonate)), which could better passivate
the anode surface and resist volume expansion^{[6,75,78}].
Meanwhile, FEC incurs increased formation of LiF,
and less formation of Li_ECO₃ and organic carbonate
specie the anode surface and resist volume expansion^{[6, 75, 78}].
Meanwhile, FEC incurs increased formation of LiF,
and less formation of Li_CCo₃ and organic carbonate
species, resulting in an overall lower interfacial
imped Meanwhile, FEC incurs increased formation of LiF,
and less formation of Li_CCo₃ and organic carbonate
species, resulting in an overall lower interfacial
impedance of the Si anode^[79]. Moreover, there is ev-
idence th and less formation of Li₂CO₃ and organic carbonate
species, resulting in an overall lower interfacial
impedance of the Si anode^[79]. Moreover, there is ev-
idence that FEC influences the LiPF₆ decomposition
reacti on of Li₂CO₃ and organic carbonate
g in an overall lower interfacial
e Si anode^[79]. Moreover, there is ev-
influences the LiPF₆ decomposition
y suppress further salt degradation
yeles^[68]. Hou et al.^[12] inve 电化学(*J. Electrochem.*) 2022, 28(11), 2219007 (7 of 18)

mediates, polysulfides electrochemical performance, improved Coulombic

t side reactions at in-

shuttle effect not only and e (Figure 4b). To understand the composi

 $# \&L \# (J. \nElectrochem.) 2022, 28(11), 2219007 (8 of 18)$ 4c, the Li⁺-coordinated FEC was found to reduce pri-

or to EC and uncoordinated FEC, thus passivating the >EC ≈ FEC ≈ VC, which

anode surface at an early onset (ca. 0.3

 $\# \# \# (L \text{ } Electrochem.) 2022, 28(11), 2219007 (8 of 18)$

-coordinated FEC was found to reduce pri-

ing ability of the additives follows the order of VEC

md uncoordinated FEC, thus passivating the
 \geq EC \approx FEC \approx VC, whic $\frac{4}{5}$ (*L Electrochem.*) 2022, 28(11), 2219007 (8 of 18)

4c, the Li⁺-coordinated FEC was found to reduce pri-

or to EC and uncoordinated FEC, thus passivating the
 \geq EC \approx FEC \approx VC, which indicates tha $\#E\#(J. Electron)$ 2022, 28(11), 2219007 (8 of 18)
4c, the Li⁺-coordinated FEC was found to reduce pri-
or to EC and uncoordinated FEC, thus passivating the
anode surface at an early onset (ca. 0.3 V higher than
anode surfac ELET UNITEL THE CONDUCT ALTER THE UNITELY CONDUCT AND A polyees determined the coordinated FEC was found to reduce prior imp ability of the additives follows the order of VEC or to EC and uncoordinated FEC, thus passivat **EART (Example 1998)**
 EART (Example 1992), 28(11), 2219007 (8 of 18)

4c, the Li⁺-coordinated FEC was found to reduce prior ing ability of the additives follows the order of VI

or to EC and uncoordinated FEC, thus $\frac{36}{2}$ (*J. Electrochem.*) 2022, 28(11), 2219007 (8 of 18)

uund to reduce pri-

ing ability of the additives follows the order of VEC

hus passivating the \geq EC \approx FEC \approx VC, which indicates that VEC has the

0. **EVALUATION 1989**
 EVALUATION ASSAUTE ASSAUT ASSAUT ASSAUT ASSAUTE ASSAUTE AND ASSAUTE ADDED AND THE CONDUCT OF C and uncoordinated FEC was passivating the \geq **FC** \approx **FC will mathed strate at an early onset (ca. 0.3 V** \pm *m* (*E in the mechanistic insight that will aid in the rational design of a lower relation phenomenon and the ration of phenomenon and the ration of <i>PEC* or to *EC* and uncoordinated *FEC*, thus passivating the \rightarrow **EVALUATION EVALUATION**
 EVALUATION EVALUATION EVALUATION $\text{ft}(E^{\#}(I, Electmchehem.))$ 2022, 28(11), 2219007 (8 of 18)

the Li-coordinated FEC was found to reduce pri-

to EC and uncoordinated FEC, thus passivating the
 \geq EC \approx FEC \approx VC, which indicates that VEC has the

dede **EVALUATIVE FOLUTE CONDET ALTERT CONDENSISE (VERTUAL SET ALTERT CONDUCTS (VERTUAL SUPPLY) (CONDENDING A SET ALTERT CONDENDING SUPPLY) (CONDENDING SUPPLY) (CONDENDING SUPPLY) (CONDENDING THE CONDENDING SUPPLY) (CONDENDING in the figure 1.1** (a) the solution of the solution reaction of the solution of the solution of the solution reaction reaction of the solution reaction reac **EVALUATION THE THE SERIES (EXECT)** THE SERIEV (THE SERIEV (THE SERIEV (THE SERIEV THE SERIEV THE SERIEV THE SERIEV THE SERIEV THE SERIEV OF A SERIEV TH **in** the $E^{\omega}(L)E(eta) = 222,28(11), 2219007$ (8 or 18)
 4c, the Li²-coordinated FFC was found to reduce pri-

ing ability of the additives follows the order of VFC

or to EC and uncoordinated FEC, thus passivaling the **(d)**
 (d)
 (d) $\frac{1}{2}$ (*ki*) $\frac{1}{2}$ (*ki* **E.** Exection of the EUC in the showed by the solution of the Europham simulation of the solution of the European of the solution of the sol **Example 19** Contributed FEC with a sheather the first solven in the distinct of the distincts follows the order of VEC
or to FC and uncoordinated FFC, thus passivating the \rightarrow FC \rightarrow FC \rightarrow FC \rightarrow VC, which indicates the interaction of the additives follows the order of VEC

sisivating the
 $>EC \approx FEC \approx VC$, which indicates that VEC has the

strongest solvating ability. With higher reduction po-

higher than

strongest solvating ability. W 28(11), 2219007 (8 of 18)
ing ability of the additives follows the order of VEC
>EC \approx FEC \approx VC, which indicates that VEC has the
strongest solvating ability. With higher reduction po-
tentials of FEC and VEC over PC 28(11), 2219007 (8 of 18)
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strongest solvating ability. With higher reduction po-
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strongest solvating ability. With higher reduction po-

tentials of FEC and VEC 28(11), 2219007 (8 of 18)
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tentials of FEC and VEC over PC 28(11), 2219007 (8 of 18)
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strongest solvating ability. With higher reduction po-
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>EC \approx FEC \approx VC, which indicates that VEC has the
strongest solvating ability. With higher reduction po-
tentials of FEC and VEC over PC 28(11), 2219007 (8 of 18)

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strongest solvating ability. With higher reduction po-

tentials of FEC and VEC 28(11), 2219007 (8 of 18)

ing ability of the additives follows the order of VEC
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strongest solvating ability. With higher reduction po-

tentials of FEC and VEC over PC, the products. 28(11), 2219007 (8 of 18)

ing ability of the additives follows the order of VEC

>EC \approx FEC \approx VC, which indicates that VEC has the

strongest solvating ability. With higher reduction po-

tentials of FEC and VEC ove 11), 2219007 (8 of 18)
ability of the additives follows the order of VEC
ability of the additives follows the order of VEC
 \approx FEC \approx VC, which indicates that VEC has the
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ing ability of the additives follows the order of VEC

>EC ≈ FEC ≈ VC, which indicates that VEC has the

strongest solvating ability. With higher reduction po-

tentials of FEC and VEC over PC, high-concentration electrolytes (LHCE) have been $\exists E(E \neq U, \text{Electrochem.}) \quad 2022, 28(11), 2219007 \, (8 \text{ of } 18)$

as found to reduce pri-

ing ability of the additives follows the order of VEC

C, thus passivating the >EC ≈ FEC ≈ VC, which indicates that VEC has the

(ca. 0.3

Coulombic efficiencies of the Si/Li half-cells cycled between 0.12 and 0.9 V at 500 mA · g⁻¹ (Si) using FEC/LP40 (black) and LP40

Example 10
 Example 10 the reduction of the selectrostee of the simulation (Figure 3b) and experiments (Figure 5c), access the oxidation stability of the electrolyte at the simulation (Figure 3b) and experiments (Figu \pm defined to tune the SEI bear inorganic-rich chemical signatures
strengthe diatomy of the signature of diatomy of the simulation (Figure 5b) and experiments (Figure 5c),
example is desc. Specifically, HCFs has been re th($k^{\#}(L\ Kerenolem, \Sigma)$ 22, 28(11), 2219007 (9 of 18)
designed to tune the SFI chemistry as well as in-in Li²-solvation sheath was observed in both MD
crease the oxidation stability of the electrolyte at the simulation (Fi the $\frac{dE}{dE}$ (*E* E (*E*) (*E* $\frac{1}{2}E\# (L\ Electrocheen, 2022, 28(11), 2219007 (9 of 18)$
designed to tune the SEI chemistry as well as in-
in Li-solvation sheath was observed in both MD
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crease the oxidation stability of the electrolyte at the simulation (Figure 5b) and experiments (Figure 5c),
cathode side. S designed to tune the SEI chemistry as well as in-

in Li⁺-solvation shachl was observed in both

crease the oxidation stability of the electrolyte at the simulation (Figure 5b) and experiments (Figure

cathode side. Spe erease the oxidation stability of the clectrolyte at the

simulation (Figure 5b) and experiments (Figure 5c),

archodes ide. Specifically, if CF: has been reported to

leaving a high portion (40%) of Li² becalisively su cathode side. Specifically, HCE has been reported to leaving a high portion (40%) of Li¹ exprovide increased oxidative stability, absence of ex-

foliation, low volation by volation specificant ion-

foliation, low vola

 $\exists \ell \in \mathbb{Z}$ $\exists \ell \in \mathbb{Z}$ $\exists \ell \in \mathbb{Z}$ $\ell \in \mathbb{Z}$ as well as in-
designed to tune the SEI chemistry as well as in-
in Li^{*}-solvation sheath was observed in both MD
crease the oxidation stability of the electrolyt $\#l\&\#(J. \nElectrochem.)$ 2022, 28(11), 2219007 (9 of 18)

designed to tune the SEI chemistry as well as in-

crease the oxidation stability of the electrolyte at the

simulation (Figure 5b) and experiments (Figure 5c),

cathod $\pm \frac{1}{2}$ (*L Electrochem.*) 2022, 28(11), 2219007 (9 of 18)
designed to tune the SEI chemistry as well as in-
in Li⁺-solvation sheath was observed in both MD
crease the oxidation stability of the electrolyte at the **EVAPPLE 12000** (9 of 18)
 EVAPPLE 12000 (9 of 18)
 EVAPPLE 12000 (1900) (1900) (9 of 18)
 EVAPPLE 12000 (1900) (1900) (9 of 18)
 EVAPPLE 12000 (1900) (Figure 5b) and experiments (Figure 5c),

cathode side. Specif $\pm \frac{\text{d}E}{\text{d}E}$ (*L Electrochem.*) 2022, 28(11), 2219007 (9 of 18)

designed to tune the SEI chemitsty as well as in-

in Li⁻solvation shealth was observed in both MD

crease the oxidation stability of the electr **Example 10**
 Example 10 tune the SEI chemistry as well as in-

in Li⁻solvation shealt was observed in both MD

crease the oxidation stability of the electrolyte at the simulation (Figure 5b) and experiments (Figure 5 **EVACUTE 12000** (9 of 18)
 EVACUTE 12000 (10 of 18)
 EVACUTE 12000 (Figure 5b) and experiments (Figure 5c),

cathode side. Specifically therein are largely to the extractor of the simulation (Figure 1) and designed to tune the SEI chemistry as well as in-
in Li⁻solvation shealth was observed in both MD
crease the oxidation stability of the electrolyte a **in the state of the molecules, such the simulation (Figure 5b) and experiments (Figure 5c), eathode side. Specifically, HCE has b** $28(11)$, 2219007 (9 of 18)
in Li⁺-solvation sheath was observed in both MD
simulation (Figure 5b) and experiments (Figure 5c),
leaving a high portion (40%) of Li⁺ exclusively sur-
rounded by water molecules only, w 2219007 (9 of 18)

-solvation sheath was observed in both MD

tion (Figure 5b) and experiments (Figure 5c),

g a high portion (40%) of Li⁺ exclusively sur-

ed by water molecules only, while the rest are

y surrounded by $28(11)$, 2219007 (9 of 18)
in Li⁺-solvation sheath was observed in both MD
simulation (Figure 5b) and experiments (Figure 5c),
leaving a high portion (40%) of Li⁺ exclusively sur-
rounded by water molecules only, w 28(11), 2219007 (9 of 18)

in Li⁺-solvation sheath was observed in both MD

simulation (Figure 5b) and experiments (Figure 5c),

leaving a high portion (40%) of Li⁺ exclusively sur-

rounded by water molecules only, w ed in both MD
ents (Figure 5c),
exclusively sur-
hile the rest are
in longer length
le (1 nm) with
One benefit of 28(11), 2219007 (9 of 18)
in Li⁺-solvation sheath was observed in both MD
simulation (Figure 5b) and experiments (Figure 5c),
leaving a high portion (40%) of Li⁺ exclusively sur-
rounded by water molecules only, while 28(11), 2219007 (9 of 18)

in Li⁺-solvation sheath was observed in both MD

simulation (Figure 5b) and experiments (Figure 5c),

leaving a high portion (40%) of Li⁺ exclusively sur-

rounded by water molecules only, w $28(11)$, 2219007 (9 of 18)
in Li⁺-solvation sheath was observed in both MD
simulation (Figure 5b) and experiments (Figure 5c),
leaving a high portion (40%) of Li⁺ exclusively sur-
rounded by water molecules only, w 28(11), 2219007 (9 of 18)
in Li⁺-solvation sheath was observed in both MD
simulation (Figure 5b) and experiments (Figure 5c),
leaving a high portion (40%) of Li⁺ exclusively sur-
rounded by water molecules only, while $28(11)$, $2219007 (9 of 18)$
in Li⁺-solvation sheath was observed in both MD
simulation (Figure 5b) and experiments (Figure 5c),
leaving a high portion (40%) of Li⁺ exclusively sur-
rounded by water molecules only, whil $28(11)$, $2219007 (9 \text{ of } 18)$
in Li⁺-solvation sheath was observed in both MD
simulation (Figure 5b) and experiments (Figure 5c),
leaving a high portion (40%) of Li⁺ exclusively sur-
rounded by water molecules only, 219007 (9 of 18)
olvation sheath was observed in both MD
on (Figure 5b) and experiments (Figure 5c),
a high portion (40%) of Li⁺ exclusively sur-
by water molecules only, while the rest are
surrounded by TFSI, which in 28(11), 2219007 (9 of 18)
in Li⁺-solvation sheath was observed in both MD
simulation (Figure 5b) and experiments (Figure 5c),
leaving a high portion (40%) of Li⁺ exclusively sur-
rounded by water molecules only, while (9 of 18)

1 sheath was observed in both MD

ure 5b) and experiments (Figure 5c),

portion (40%) of Li⁺ exclusively sur-

ter molecules only, while the rest are

ded by TFSI, which in longer length

rogeneity on nano-sc 28(11), 2219007 (9 of 18)
in Li⁺-solvation sheath was observed in both MD
simulation (Figure 5b) and experiments (Figure 5c),
leaving a high portion (40%) of Li⁺ exclusively sur-
rounded by water molecules only, while 28(11), 2219007 (9 of 18)
in Li'-solvation sheath was observed in both MD
simulation (Figure 5b) and experiments (Figure 5c),
leaving a high portion (40%) of Li' exclusively sur-
rounded by water molecules only, while the 28(11), 2219007 (9 of 18)
in Li⁺-solvation sheath was observed in both MD
simulation (Figure 5b) and experiments (Figure 5c),
leaving a high portion (40%) of Li⁺ exclusively sur-
rounded by water molecules only, while (11), 2219007 (9 of 18)
Li^{*}-solvation sheath was observed in both MD
Li^{*}-solvation sheath was observed in both MD
ulation (Figure 5b) and experiments (Figure 5c),
ving a high portion (40%) of Li^{*} exclusively sur-
an $28(11)$, 2219007 (9 of 18)
in Li⁺-solvation sheath was observed in both MD
simulation (Figure 5b) and experiments (Figure 5c),
leaving a high portion (40%) of Li⁺ exclusively sur-
rounded by water molecules only, w $2x(x)$, $2x(x)$, $6x(x)$ in the increase in both MD simulation (Figure 5b) and experiments (Figure 5c), leaving a high portion (40%) of Li⁺ exclusively surrounded by water molecules only, while the rest are mainly surroun in Li⁺-solvation sheath was observed in both MD
simulation (Figure 5b) and experiments (Figure 5c),
leaving a high portion (40%) of Li⁺ exclusively sur-
rounded by water molecules only, while the rest are
mainly surrou simulation (Figure 5b) and experiments (Figure 5c),
leaving a high portion (40%) of Li⁺ exclusively sur-
rounded by water molecules only, while the rest are
mainly surrounded by TFSI, which in longer length
leads to a h leaving a high portion (40%) of Li⁺ exclusively sur-
rounded by water molecules only, while the rest are
mainly surrounded by TFSI, which in longer length
leads to a heterogeneity on nano-scale (1 nm) with
water-rich an 电化学(*J. Electrochem.*) 2022, 28(11), 2219007 (9 of 18)
mistry as well as in-
f the electrolyte at the simulation (Figure 5b) and experiments (Figure 5c),
E has been reported to leaving a high portion (40%) of Li⁺ exclu

 $#E#(J. Electron) \xrightarrow{\text{Re}} (J. Perform) \xrightarrow{\text{Re}} (J. Perform) \xrightarrow{\text{Re}} (J. Common) \xrightarrow$ $\# \&\# \langle I. \text{ \&Letrochem.} \rangle$ 2022, 28(11), 2219007 (10 of 18)

dinating co-solvent)^[88]. Polyfluorinated ethers, such trolytes, predominant types of materials include gar-

as bis(2,2,2-trifluoroethyl) ether (BTFE)^[87], $#E^{\omega}(L \t{Electrochem.})$ 2022, 28(11), 2219007 (10 of 18)

dinating co-solvent)^[80]. Polyfluorinated ethers, such trolytes, predominant types of materials include gar-

as bis(2,2,2-trifluoroethyl) ether (BTFE)^[87], are u $\mathbb{E}E\ddot{\cong} (L\text{ }Electrochem.)$
 $\mathbb{E}E\ddot{\cong} (L\text{ }Electrochem.)$ 2022, 28(11), 2219007 (10 of 18)

dinating co-solvent)^[86]. Polyfluorinated ethers, such trolytes, predominant types of materials include gar-

as bis(2,2,2-trifl \oplus \oplus bility. The addition of diluents separates the bulk and \pm *Retingalem,* 2022, 28(11), 2219007 (10 of 18)

dinating co-solvent)^{[186}. Polyfluorinated cthers, such trolytes, predominant types of materials include gar-

as bis(2,2,2-trifluoroethyl) ether (BTFE)^[87], are usu \pm *Refunctional* (\pm *Retrochem.*) 2022, 28(11), 2219007 (10 of 18)

dinating co-solvent)^[86]. Polyfluorinated ethers, such trolytes, predominant types of materials include gar-

as bis(2,2,2-trifluoroethyl) ether **HECFC** *C*. *Electrochem.* $)$ 2022, 28(11), 2219007 (10 of 18)
dinating co-solvent)⁸⁸¹. Polyfluorinated ethers, such trolytes, predominant types of materials include gar-
as bis(2,2,2-trifluorocthyl) ether (BTFE)⁸⁸¹ $\text{th/2}^{\text{th}}(L)$ Electrochem, 2022, 28(11), 2219007 (10 of 18)

dinating co-solvent)^[86]. Polyfluorinated cthers, such trolytes, predominant types of materials include gar-

as bis(2,2,2-trifluorocthyl) cther (BTFE)[[] **THEP (EXECT)** The metrical conductions and anions still maintains are the theory and anions and include parameterisms are differently) efter (BTFF)³⁷³, are usual-
as bis(2,2,2-trifluoroethyl) ether (BTFF)³⁷³, are usu $\mathbb{E}(E \ncong CLE)$
 $\mathbb{E}(E \ncong CLE)$ $\mathbb{E}(E \ncong CLE)$

solvent)¹⁹⁶¹. Polyfluorinated ethers, such trolytes, predominant types of materials include gar-

as bis(2,2,2-trifluorechly)) ether (BTFE)¹⁹⁷¹, are usual

be they the $\#(\pm \pi)$ and $\#(\pm \pi)$ and $\#(\pm \pi)$ and $\pm \pi$ and **HET (FEFT)**⁸⁶. *Electrochem.*) 2022, 28(11), 2219007 (10 of 18)

dinating co-solvent)¹⁸⁸. Polyfluorinated ethers, such trolytes, predominant types of materials include gar-

as bis(2,2,2-trifluorochly)) ether (BTFE) **HERE OF** E **(Figure 5d)**, which is other and set all decreases in the physical conduction of the physical conduction of the physical conduction of the physical conduction of the physical capacity and their materials incl dinating co-solvent)¹⁶⁶. Polythorianal cher, such search search is a proposition of the highly fluorinated clusters, such a rolety, such a rolety, such a rolety is attributed derivatived in your site of the highest prop their low viscosity, sufficient electrochemical stabili-

their low viscosity, sufficient electrochemical stabili-

ty, appropriate miscibility, and poor solvating capa-

interfacial properties, and bility. The addition o ty, appropriate miscibility, and poor solvating capa⁻ mechanical properties, and excellent eithilig. The addition of diluents separates the bulk and stability against Li metal. Notably, since is interfacted responsibili

cathodes is attributed to the highly fluorinated CEI Polymer solid electrolytes based on lithium salls in
Meanwhile, the average bulk properties are improved
with Li metal and low cost¹⁹¹¹. However, the perfor-
by the p and SEI as decomposition products of the anions. polymer matrices exhibit favorable interfacial contact

Meanwhile, the werget bulk properties are improved with Linetal and low cost¹⁹⁸. However, the preformance of prope Meanwhile, the average bulk properties are improved

with Li metal and low cost¹⁹¹. However, the performation

by the preferred ion transport, viscosity, or wettabili-mane of polymer soid electrolytes is limited by

by by the preferred ion transport, viscosity, or wettabili-

annee of polymer solid electrolytes is limited by low

ionic conductivity¹, low lithium ion transferrence nu-

4.3 Solid State Electrolyte

mber^{ian}, and unsatis sty of the diluent.

4.3 Solid State Electrolyte

4.3 Solid State Electrolyte
 \sim Most and massifeared and
 \sim Most and massifeared and massifeared and
 \sim Most and the highest theoretical energy
 \sim Most and the **4.3 Solid State Electrolyte** mbcr²⁴⁴, and unsatisfactory clastic modulus⁸⁹⁴ com-

As mentioned in the provises section, the Li metal

approach is operated by definition has the highest theoretical energy uous efforts As mentioned in the previous section, the Li metal
and pared with inorganic crystalline clectrolytes. Contin-
anode by definition has the highest theoretical energy uous efforts have been made to further improve the
densi anode by definition has the highest theoretical energy uous efforts have been made to further improve their
However, the cleetronic and omic conductive Li den-

mechanical-stella-state of the mechanical-stellated
creductio density among any other anode materials of LIBs. mechanical/electrochemical stability and ionic con-
However, the electronic and ionic conductive Li don-
dorite Lien-
dorite Lien-dorite Lien-dorite and periodic into the d However, the electronic and ionic conductive Li den-

dirtiction policiality, e.g., introducing mechanical reinforcement

drive can penerate the porous separator and make \hat{a}^{\dagger} - blooks⁽¹⁹⁾, incorporating single-i drite can penetrate the porous separator and make di-
blocks¹⁹⁵³, incorporating single-ion conductors teret contact with the cathode, leading to the internal
show rise is phase iteration. Since the proformance metricals t contact with the cathode, leading to the internal place lithium salts¹⁶⁶, and double layer designs that relative of the batteries Moreover, uneven pre-balance the performance metricals⁶⁷⁶⁸, tatentials of active mate

dinating co-solvent)¹⁸⁹¹. Polyfluorinated ethers, such

and big(2,2-trifluorenehyl) ether (BTFFE)⁷⁹⁷, are usual-

and big and \sim net-t-type SSF (i.4i,a,7F_C/F, 1/ZO)²⁰^m, (1,4i,a,7F₂), (1,AS_P^T, 1,2(2)⁻¹, as bis(2,2,2-trifluorochly)) cther (BTFE)³⁸⁷, are usual-
net-type SSE (Li,La,Zr₁O₁₀, LLZO)³⁸³, UC_N
or oncidened as idelicated as a diluent due to $\lim_{\alpha \to 0} \frac{1}{\alpha \cdot 0}$; LCD₀) (LATP)⁸⁹, and their analogues by considered as ideal candidates as a diluent due to $Li_aA_{ac}Ti_a(PO_a) (LATP)⁹⁸$, and their analogues. In-
their low viscosity, sofficient leet
redovehimical stabili- organic SSEs exhibit satisfactory ionic conductivity,
t ity. The addition of diluents separates the bulk and

stability against Li metal. Notably, some of them

erfacial responsibilities of an electrolyte and as-

have ionic conductivity comparable to or sern-

sing these clie interfineial responsibilities of an electrolyte and as-
have ionic conductivity comparable to or even sur-
signs these rols to varying phass that are micro-
massing that of fugiod electrolytes ($1-10$ mS em/¹⁹⁷¹,
seopic signs these roles to varying phases that are micro-

secondary separated¹⁶⁴. Recease the dilutent daes not However, a trade-off exists where high classical

ecoordinate with the cations, the immediate local cn-

luse ty soopically separated^{na}. Because the dilucnt does not

However, a trade-off exists where high elastic modu-

coordinate with the calinos, the immediate local error hare the propriative den-

vironment of caions and anions coordinate with the cations, the immediate local en-

viryincantly leads to poor surface adhesion and high

virtuoment of cations and anions sill maintains the interfacial resistance. To circumvent the challenge,

solvati vironment of cations and anions still maintains the interfacial resistance. To circumvent the challenge,
esty
enside to the internal contact with soften remedy strategies have been capiter directing
means the strategies solvation structure of HCE (Figure 5d), which is often

remedy strategies have been explored including engi-

responsible for the interpharail chemistrics at elec-

redring an interfactal theyre between lithium metal

trod responsible for the interphasial ehemistries at clear-
neering an interfacial layer between lithium metal
to the active condition of am SSEs ⁹⁷⁶. The simulation
action of ambigation of ambigation of ambigation of ambiga trode surfaces⁽¹⁶⁰. The simultaneous stabilization of and SSEs^[30], and surface treatment of SSEs to reduce interded centrologies interfacial resistance and increase lithiophilicity^[92].

contact and SEI as decompos Ithium metal, and high capacity and high voltage interfacial resistance and increase lithiophilicity^(se),
eathodos is attributed to the highly floorinated CEI Polymer solid electrolytes based on lithium sals in
and SEI 28(11), 2219007 (10 of 18)
trolytes, predominant types of materials include gar-
net-type SSE (Li₇La₃Zr₂O₁₂, LLZO)^[88], Li₇P₃S₁₁ (LPS)^[89],
Li₁₄A₀₄Ti_{1.6}(PO₄)₃ (LATP)^[90], and their analogues. 28(11), 2219007 (10 of 18)
trolytes, predominant types of materials include gar-
net-type SSE (Li₇La₃Zr₂O₁₂, LLZO)^[88], Li₇P₃S₁₁ (LPS)^[89],
Li₁₄A₁₀₄Ti_{1.6}(PO₄)₃ (LATP)^[90], and their analogues $\frac{1}{2}$, 28(11), 2219007 (10 of 18)
trolytes, predominant types of materials include gar-
net-type SSE (Li₇La₃Zr₂O₁₂, LLZO)^[88], Li₇P₃S₁₁ (LPS)^[89],
Li₁₄A₁₀₄Ti_{1.6}(PO₄)₃ (LATP)^[90], and their analogues 28(11), 2219007 (10 of 18)
trolytes, predominant types of materials include gar-
net-type SSE (Li₇La₃Zr₂O₁₂, LLZO)^[88], Li₇P₃S₁₁ (LPS)^[89],
Li₁₄A₁₀₄Ti_{1.6}(PO₄)₃ (LATP)^[90], and their analogues 28(11), 2219007 (10 of 18)
trolytes, predominant types of materials include gar-
net-type SSE (Li₇La₃Zr₂O₁₂, LLZO)^[88], Li₁₇P₃S₁₁ (LPS)^[89],
Li₁₄A₁₀₄Ti₁₆(PO₄)₃ (LATP)^[90], and their analogues 28(11), 2219007 (10 of 18)
trolytes, predominant types of materials include gar-
net-type SSE (Li₇La₃Zr₂O₁₂, LLZO)^[88], Li₁P₃S₁₁ (LPS)^[89],
Li₁₄A_{l04}Ti₁₆(PO₄)₃ (LATP)^[90], and their analogues. 28(11), 2219007 (10 of 18)
trolytes, predominant types of materials include gar-
net-type SSE (Li₇La₃Zr₂O₁₂, LLZO)^[88], Li₁P₃S₁₁ (LPS)^[89],
L_{I4}A₁₀₄Ti₁₆(PO₄)₃ (LATP)^[90], and their analogues. 28(11), 2219007 (10 of 18)
trolytes, predominant types of materials include gar-
net-type SSE (Li₇La₃Zr₂O₁₂, LLZO)^[88], Li₁P₁S₁₁ (LPS)^[89],
Li₁₄A_{10.4}Ti_{1.6}(PO₄)₃ (LATP)^[90], and their analogue passing that of liquid electrolytes $(1 \sim 10 \text{ mS} \cdot \text{cm}^{-1})^{[7]}$. [7] 28(11), 2219007 (10 of 18)
trolytes, predominant types of materials include gar-
net-type SSE (Li₇La₃Zr₂O₁₂, LLZO)^[88], Li₁P₁S₁₁ (LPS)^[89],
Li₁₄A₈₀₄Ti_{1.6}(PO₄)₃ (LATP)^[90], and their analogues 28(11), 2219007 (10 of 18)
trolytes, predominant types of materials include gar-
net-type SSE (Li₁La₁Zr₂O₁₂, LLZO)^[88], Li₁P₂S₁₁ (LPS)^[89],
Li₁₄A₀₄Ti₁₆(PO₄), (LATP)^[69], and their analogues. In 28(11), 2219007 (10 of 18)
trolytes, predominant types of materials include gar-
net-type SSE (Li₁La₃Zr₂O₁₂, LLZO)^[88], Li₁P₂S₁₁ (LPS)^[89],
Li₁₄A_{B4}Ti₁₄(PO₄)₃ (LATP)^[69], and their analogues. 28(11), 2219007 (10 of 18)
trolytes, predominant types of materials include gar-
net-type SSE (Li₇La₃Zr₃O_{L3}, LLZO)^[88], Li₇P₃S₁₁ (LPS)^[89],
Li₁₄A_{l04}Ti₁₆(PO₄)₃ (LATP)^[89], and their analogues. 28(11), 2219007 (10 of 18)
trolytes, predominant types of materials include gar-
net-type SSE (Li₇La₃Zr₂O_{L3}, LLZO)^[88], Li₇P_S₁₁ (LPS)^[89],
Li₁₄A_{l04}Ti₁₆(PO_d)₃ (LATP)^[90], and their analogues. 28(11), 2219007 (10 of 18)
trolytes, predominant types of materials include gar-
net-type SSE (Li₇La₃Zr₂O_{L3}, LLZO)^[88], Li₇P_S₁₁ (LPS)^[89],
Li₁₄A_{l04}Ti₁₆(PO_a)₃ (LATP)^[90], and their analogues. $28(11)$, $2219007 (10 \text{ of } 18)$
trolytes, predominant types of materials include gar-
net-type SSE (Li₇La₃Zr₂O₁₃, LLZO)^[88], Li₇P₃S₁₁ (LPS)^[89],
Li₁₄A_{l04}Ti₁₄(PO₄)₃ (LATP)⁽⁹⁰⁾, and their analo $\text{Lev}(x, y, \Sigma, \Sigma, \text{O}_1)$. (1941.6)

relatives based comminant types of materials include gar-

net-type SSE (Li₇La₃Zr₂O₁₂, LLZO)^[88], Li₇P_{S11} (LPS)^[89],

Li₁₄A_{m4}Ti₁₆(PO₄)₃ (LATP)^[90], and thei trolytes, predominant types of materials include gar-
net-type SSE (Li₇La₃Zr₂O₁₂, LLZO)^[88], Li₇P_S₁₁ (LPS)^[89],
Li₁₄A_{m4}Ti₁₆(PO₄)₃ (LATP)^[90], and their analogues. In-
organic SSEs exhibit sat net-type SSE (Li₇La₃Zr₃O₁₂, LLZO)^[88], Li₇P_{S11} (LPS)^[89], Li₄A₀₄Ti₁₆(PO₄)₃ (LATP)^[90], and their analogues. Inorganic SSEs exhibit satisfactory ionic conductivity, mechanical properties, and ex Li₁₄A₀₄Ti₁₆(PO₄)₃ (LATP)^[90], and their analogues. In-
organic SSEs exhibit satisfactory ionic conductivity,
mechanical properties, and excellent electrochemical
stability against Li metal. Notably, some of th organic SSEs exhibit satisfactory ionic conductivity,
mechanical properties, and excellent electrochemical
stability against Li metal. Notably, some of them
have ionic conductivity comparable to or even sur-
passing that mechanical properties, and excellent electrochemical
stability against Li metal. Notably, some of them
have ionic conductivity comparable to or even sur-
passing that of liquid electrolytes $(1 \sim 10 \text{ mS} \cdot \text{cm}^{-1})^{[7]}$.
 stability against Li metal. Notably, some of them
have ionic conductivity comparable to or even sur-
passing that of liquid electrolytes $(1 \sim 10 \text{ mS} \cdot \text{cm}^{-1})^{[T]}$.
However, a trade-off exists where high elastic modu-
 have ionic conductivity comparable to or even sur-
passing that of liquid electrolytes $(1 \sim 10 \text{ mS} \cdot \text{cm}^{-1})^{[T]}$.
However, a trade-off exists where high elastic modu-
lus typically leads to poor surface adhesion and h passing that of liquid electrolytes $(1 \sim 10 \text{ mS} \cdot \text{cm}^{-1})^{[7]}$.

However, a trade-off exists where high elastic modulus typically leads to poor surface adhesion and high interfacial resistance. To circumvent the challe However, a trade-off exists where high elastic modulus typically leads to poor surface adhesion and high interfacial resistance. To circumvent the challenge, remedy strategies have been explored including engineering an i lus typically leads to poor surface adhesion and high
interfacial resistance. To circumvent the challenge,
remedy strategies have been explored including engi-
neering an interfacial layer between lithium metal
and SSEs[[] interfacial resistance. To circumvent the challenge,
remedy strategies have been explored including engi-
neering an interfacial layer between lithium metal
and SSEs^[90], and surface treatment of SSEs to reduce
interfac remedy strategies have been explored including engi-
neering an interfacial layer between lithium metal
and SSEs^{[91}, and surface treatment of SSEs to reduce
interfacial resistance and increase lithiophilicity¹⁷².
Polym Tring an interfacial layer between lithium metal

ISSEs^{[911}], and surface treatment of SSEs to reduce

refacial resistance and increase lithiophilicity^[92].

lymer solid electrolytes based on lithium salts in

ymer so and SSEs^{[911}], and surface treatment of SSEs to reduce
interfacial resistance and increase lithiophilicity^[92].
Polymer solid electrolytes based on lithium salts in
polymer matrices exhibit favorable interfacial conta interfacial resistance and increase lithiophilicity^[92].
Polymer solid electrolytes based on lithium salts in
polymer matrices exhibit favorable interfacial contact
with Li metal and low cost^[99]. However, the perfor-Polymer solid electrolytes based on lithium salts in
polymer matrices exhibit favorable interfacial contact
with Li metal and low cost¹⁹³¹. However, the perfor-
mance of polymer solid electrolytes is limited by low
ioni polymer matrices exhibit favorable interfacial contact
with Li metal and low cost¹⁹³¹. However, the perfor-
mance of polymer solid electrolytes is limited by low
ionic conductivity^[7], low lithium ion transference nuwith Li metal and low cost¹⁹³¹. However, the perfor-
mance of polymer solid electrolytes is limited by low
ionic conductivity^[7], low lithium ion transference nu-
mber¹⁴⁴¹, and unsatisfactory elastic modulus^{[941}] 电化学(*J. Electrochem.*) 2022, 28(11), 2219007 (10 of 18)

uorinated ethers, such trolytes, predominant types of materials include gar-

r (BTFE)^[87], are usual-

net-type SSE (Li₁La₃Zr₂O₁₂, LLZO)^[88], Li₁P₃

mance of polymer solid electrolytes is limited by low
ionic conductivity^[7], low lithium ion transference nu-
mber^[44], and unsatisfactory elastic modulus^[94] com-
pared with inorganic crystalline electrolytes. Cont ionic conductivity^[7], low lithium ion transference number^[44], and unsatisfactory elastic modulus^[94] com-
pared with inorganic crystalline electrolytes. Contin-
uous efforts have been made to further improve their mber¹⁴⁴¹, and unsatisfactory elastic modulus^{[941}] com-
pared with inorganic crystalline electrolytes. Contin-
uous efforts have been made to further improve their
mechanical/electrochemical stability and ionic con-
duc pared with inorganic crystalline electrolytes. Contin-
uous efforts have been made to further improve their
mechanical/electrochemical stability and ionic con-
ductivity, e.g., introducing mechanical reinforcement
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mechanical/electrochemical stability and ionic con-
ductivity, e.g., introducing mechanical reinforcement
blocks^[98], incorporating single-ion conductors that re-
pla mechanical/electrochemical stability and ionic con-
ductivity, e.g., introducing mechanical reinforcement
blocks^[98], incorporating single-ion conductors that re-
place lithium salts^[96], and double layer designs that
 ductivity, e.g., introducing mechanical reinforcement
blocks^[05], incorporating single-ion conductors that re-
place lithium salts^[06], and double layer designs that
balance the performance metrics^[97,98].
Recently, blocks^{[951}, incorporating single-ion conductors that re-
place lithium salts^{[961}, and double layer designs that
balance the performance metrics^[97,98].
Recently, several composite SSEs designs have
been proposed tha place lithium salts^[96], and double layer designs that
balance the performance metrics^[97,98].
Recently, several composite SSEs designs have
been proposed that combine ceramic nanoparticles/
nanowires with the polymer

rate.

Example 18 CM Example CONSTRATIONS EXAMPLE CONSTRATIONS EXAMPLE CONSTRANG AND CONSTRANG AND THE CONSTRANG AND THE CONSTRANG AND THE CO 1. Li metal **19** PLL composite electrolyte \bigotimes Immobilized anion $\mathcal{N}\mathcal{N}$ Free Li ions **C** Routine liquid electrolyte \bigotimes Mobile anion $\mathcal{N}\mathcal{N}$ Free Li ions **C** Routine liquid electrolyte \bigotimes Mobile a *N*/Free Li ions **Example and electrolyte** (Nobile anion

Figure 6 (a) Schematic of the electrochemical deposition behavior of the Li metal unode with the PLL solid electrolyte with immo-

bilized anions and (b) the rout (MOFs) with superior ionic conductivity and $Li⁺$ trans-**Figure 6** (a) Schematic of the electrochemical deposition behavior of the 1.4 metal anode with the PLI, solid dectrolyte with immo-
bilized unions and (b) the routine liquid electrolyte with mobile mions. Reproduced with **Figure 6** (a) Schematic of the electrochemical deposition behavior of the Li metal anode with the PLL solid electrolyte with immobilized anions and (b) the routine liquid electrolyte with mobile mions. Reproduced with pe bilized anions and (b) the routine liguid electrolyte with mobile anions. Reproduced with permission from ref¹⁹⁸⁸. Copyright 2017

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liquids, which is insufficient to fulfi National Asademy of Sciences. (color on line)

liquids, which is insufficient to fulfill the demand for methane (TFPM) building units through imine con-

batteries with high energy density and high charging densation, as liquids, which is insufficient to fulfill the demand for

methane (TFPM) building units through imine con-

batteries with high energy density and high charging

consation, as shown in Figure 7a. With PC filling the

cont liquids, which is insufficient to fulfill the demand for

methane (TFPM) building units through imine con-

batteries with high energy density and high charging

densation, as shown in Figure 7a. With PC filling the

rate liquids, which is insufficient to fulfill the demand for

methane (TFPM) building units through imine con-

batteries with high energy density and high charging

oreas, Li⁻exchanged MOF-688 exhibited a high intic

rate. batteries with high energy density and high charging densation, as shown in Figure 7a. With PC filling the conder of the practical density and the conder conder of the productivity of 4.0 x 10⁻⁴ Sc exhibited a high ionic rate. pores, Li⁻-exchanged MOF-688 exhibited a high in One of the practical approaches to address above
mentioned challenges is the use of quasi-solid-state fermee number of 0.87 a 298 K.
electrolytes (OSSEs.), i.e., ad One of the practical approaches to address above

conductivity of 4.0 × 10⁻⁴ S - cm⁻¹ and high Li' tr

mentioned challenges is the use of quasi-solid-state

electrolytes (OSSEs), i.e., adding a minimum amount In this

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and the characterizaconductivity of 4.0×10^{-4} S \cdot cm⁻¹ and high Li⁺ trans-**IF the Case of the Case of the Case of the Case of the PLL solid electrolyte** extinements and a new class of the PLL solid electrolyte with immo-
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Ethance (TFPM)

Example 12 Immobilized anion

electrolyte \bigotimes Mobile anion

electrolyte \bigotimes Mobile anion

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methane (TFPM) building units through imine con-

densatio **Example 12**
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electrolyte \bullet Mobile anion

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Subsect as a mode with the PLL solid electrolyte with immo-

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methane electrolyte $\frac{1}{200}$. Mobile anion.

electrolyte $\frac{1}{200}$. Mobile anion.

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methane (TFPM) building units through imine con-

densation, as shown in Figure 7a performed the Linesian methanology entroducing the Linesian methanology entroducing the Linesian from ref ^[160]. Copyright 2017
methane (TFPM) building units through imine con-
densation, as shown in Figure 7a. With PC of the Li metal anode with the PLL solid electrolyte with immo-
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methane (TFPM) building units through imine con-
densation, as shown in Figure 7a. With PC fil ms. Reproduced with permission from ref^{[160}]. Copyright 2017
methane (TFPM) building units through imine con-
densation, as shown in Figure 7a. With PC filling the
pores, Li'-exchanged MOF-688 exhibited a high ionic
con methane (TFPM) building units through imine con-
densation, as shown in Figure 7a. With PC filling the
pores, Li¹-exchanged MOF-688 exhibited a high ionic
conductivity of 4.0×10^{-4} S · cm⁻¹ and high Li⁺ trans-
f methane (TFPM) building units through imine con-
densation, as shown in Figure 7a. With PC filling the
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f methane (TFPM) building units through imine con-
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fer densation, as shown in Figure 7a. With PC filling the
pores, Li⁺-exchanged MOF-688 exhibited a high ionic
conductivity of 4.0×10^{-4} S·cm⁻¹ and high Li⁺ trans-
ference number of 0.87 at 298 K.
In this new class o pores, Li⁺-exchanged MOF-688 exhibited a high ionic
conductivity of 4.0×10^{-4} S · cm⁻¹ and high Li⁺ trans-
ference number of 0.87 at 298 K.
In this new class of promising prototype QSSEs, it
is important to unde conductivity of 4.0×10^{-4} S \cdot cm⁻¹ and high Li⁺ trans-
ference number of 0.87 at 298 K.
In this new class of promising prototype QSSEs, it
is important to understand the transport and conduc-
tion mechanisms, e

Property (1.4) Synchetic strategy, structure illustration and key performance of QSSE has a **state-of-the-art (0.19** $\frac{1}{2}$ and $\frac{1}{2}$ and $\frac{1}{2}$ and $\frac{1}{2}$ are *s* and MOF-688. Reprinded with premised in th

 $\begin{array}{lll\hline \mathbb{Q}_{\text{c}}&\mathbb{S}_{\text{c}}\mathbb{Z}_{\text{c}}&\mathbb{S}_{\text{c}}\mathbb{Z}_{\text{c}}&\mathbb{S}_{\text{c}}\mathbb{Z}_{\text{c}}&\mathbb{S}_{\text{c}}\mathbb{Z}_{\text{c}}&\mathbb{S}_{\text{c}}\mathbb{Z}_{\text{c}}&\mathbb{S}_{\text{c}}\mathbb{Z}_{\text{c}}&\mathbb{S}_{\text{c}}\mathbb{Z}_{\text{c}}&\mathbb{S}_{\text{c}}\mathbb{Z}_{\$ **Example 19**
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 Figure 7(a) Synthetic strategy, structure illustration and key performance of QSSE's hased on MOF-688 (SSEE)

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Figure 7 (a) Synthetic strategy, structure illustration and key performance of QSSE's hased on MOF-Figure 7 (a) Symbolic strategies interesting the conduction of the extended

Figure 7 (a) Symbolic strategy, sturtune illustration and key perfirmance of CSSE is based on MODF-G88 (strategy and MOF-G88 (one-fold), and the **From the properties are deemed by the content of the summation of the summation of the summation and the synchromation of OSSH band on MOF-688. Reprinted with permission
time first, Copyright 2019 American Chemical Socie Figure 7** (a) Symbetic strategy, structure illustration and key performance of QSSE based on MOT-688. Reprinted with permission
from ref¹⁸⁶⁹. Copyright 2019 American Chemical Society, (b) Scheme of solvent-assisted hop **Figure 7.** (a) syntanic statespy, subturbultion and \approx per perminent of CSSE (and MOT-658 (energet \approx (a) syntanic membersion and \approx (b) scheme of solvent-assisted hopping. (c) Schemes of MOT-658 (the membersion an three-fold and proposed one-fold interpenetating structures. (d) lonic conductivity comparison plot of MOF-688 (three-fold) MMI-688 (ene-fold), and the conductivity range of liquid curbonate electrospas, (highlighted), Rep and MOF-688 (one-fold), and the conductivity range of liquid curbonate electrolytes (highlighted). Reprimed with permission from

ref¹⁰⁸¹. Copyright 2022 The Authors. (color on line)

hopping (> 77%), as illustrated in ref¹⁹⁶⁶. Copyright 2022 The Authors, (color on line)

hopping (> 77%), as illustrated in Figure 7b. Based on

the revealent mechanism of Li motion, a hypothe-spectroscopy (TERS)^(n/8) has even pushed the limit of

sized hopping ($>77\%$), as illustrated in Figure 7b. Based on
the exclusively, the utilization of tip-enhanced Raman
sized MOF design with a non-interpententing struct-
naisolatom of Lifeston and the previous profession (TERS) hopping (\sim 77%), as illustrated in Figure 7b. Based on
the prevalent mechanism of Li¹ motion, a hypothe-spectroscopy (TFRS)²⁰⁰⁹ has even pushed the limit of
sized MOF design with a non-interprenetrating struc-
inte hopping (> 77%), as illustrated in Figure 7b. Based on

sized the prevalent mechanism of Li⁺ motion, a hypothe-

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e of solvent-assisted hopping. (c) Schemes of MOF-688 QSSEs

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ante electrolytes mance of QSSE based on MOF-688. Reprinted with permission
e of solvent-assisted hopping. (c) Schemes of MOF-688 QSSEs
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face. Recently, the utilization of tip-enhanced Raman
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interfacial/interphasial characterization to the face. Recently, the utilization of tip-enhanced Raman spectroscopy (TERS)^[110] has even pushed the limit of interfacial/interphasial characterization to the nano-scale chemical and topographical heterogeneity of the SEI face. Recently, the utilization of tip-enhanced Raman
spectroscopy (TERS)^[110] has even pushed the limit of
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SEI face. Recently, the utilization of tip-enhanced Raman
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SEI. face. Recently, the utilization of tip-enhanced Raman
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SEI spectroscopy (TERS)^[110] has even pushed the limit of interfacial/interphasial characterization to the nano-
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with experiments in these reports. With the innova- $\frac{dE}{E^* (L \: Electron})$
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systems that are challenging to obtain from experi-

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predict of the research and development of ments¹¹⁰¹). Therefore, theoretical simulations are ex-

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当前和下一代锂离子电池电解液的 原子尺度微观认识和研究进展

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摘要: 电解液及构筑电极电解液界面对于开发和应用高比容量储能系统至关重要。具体来说,电解液的机械(抗 压性、粘度)、热(热导率和热容)、化学(溶解性、活度、反应性)、输运和电化学(界面及界面层)等性质,与其所组成 的储能器件的性能直接相关。目前,大量的实验研究通过调控电解液的物理和/或化学组成来改善电解液性能,以 满足新型电极材料的工作运行。与此同时,理论模拟方法近年来得到了迅速发展,使人们可以从原子尺度来理解 电解液在控制离子输运和构筑功能化界面的作用。站在理论模拟研究的前沿上, 人们可以利用其所揭示的机理 性认识对新型电解液开展理性设计。本文首先总结了传统电解液的组成、溶剂化结构和输运性质以及电极电解液 界面层的形成机理,进一步讨论了利用新型电解液设计稳定电极电解液界面层的方法,包括使用电解液添加剂、 高浓电解液和固态电解质,并着重讨论了对这些新型电解液体系进行原子尺度模拟的最新进展,为了解和认识电 解液提供更为基本的理解,并为未来电解液的设计提供系统的指导。最后,作者对新型电解液的理论筛选进行了 展望。 しんしゃくん かんしゅう しんしゃく しんしゃく しんしゅう

关键词: 锂离子电池:电解液:原子模拟:固体电解质界面层:固态电解质