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Multi-Electron Reaction-Boosted High Energy Density Batteries: Material and System Innovation

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DOI: 10.13208/j.electrochem.2219011 http://electrochem.xmu.edu.cn
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J. Electrochem. 2022, 28(12), 2219011 (1 of 12)

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Rui-Qi Guo1,2, Feng Wu1,2, Xin-Ran Wang1,2*, Ying Bai1*, Chuan Wu1,2* (1. Beijing Key Laboratory of Environmental Science and Engineering School of Material Science & Beijing Key Laboratory of Environmental Science and Engineering School of Materials Science & Beijing Key Laboratory of Envi $[{\bf L}]. \label{eq:2.1}$ $[{\bf Multi-Electron\textbf{ Reaction-Boosted\textbf{High Energy Density Batterickem.}xm. below}] \centering \begin{minipage}{0.9\textwidth} \begin{tabular}{l} \textbf{DOL: } 10.132085 \text{electrochem.} & \textbf{Mup2}^2 \\ \textbf{Multi-Electron\textbf{ Reaction-Boosted\textbf{High Energy Density Batterics:}} \\ \textbf{Material and System\textbf{Inovation} \\ \textbf{Rui-Qi Guu}^2, \textbf{Feng Wu}^2, \textbf{Xin-Ran Wang}^{127}, \textbf{Ying Bai}^1, \textbf{Chuan Wu$ **B** *R R*

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 Multi-Electron Reaction-Boosted High Energy Density Batteries:
 Material and System Innovation

Rui-Qi Guo¹², Feng Wu¹², Xin-Ran Wang^{12*}, Ying Ba **Multi-Electron Reaction-Boosted High Energy Density Batteries:**

Material and System Innovation

Rui-Qi Guo¹², Feng Wu¹², Xin-Ran Wang¹², Ying Bai², Chuan Wu¹²

(*I. Beijing Key Labordary of Environmental Scien* **Multi-Electron Reaction-Boosted High Energy Density Batteries:**

Material: **Material and System Innovation**

Rui-Qi Guo¹², Feng Wu¹², Xin-Ran Wang¹²⁹, Ying Bai^p, Chuan Wu¹²

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Rui-Qi Ciuo¹², Feng Wu¹², Xin-Ran Wang¹², Ying Bai^{1*}, Chuan Wu¹²²

(*I. Beijing Rey Labitationy of Benjinmentand Science and Engineering School of Materi* **FORMATE ARTERN AND SYSTEM INNOVATION**

Rui-Qi Guo¹³, Feng Wu¹³, Xin-Ran Wang¹³, Ying Bai^{1°}, Chuan Wu¹³^{*ⁿ*}.
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Engineering Beijing Institute of Technology Beijing 100081,* Rui-Qi Guo¹², Feng Wu¹², Xin-Ran Wang¹², Ying Bai¹, Chuan Wu¹²

(*I. Beijing key Laboratory of Environmental Science and Engineering School of Materials Science &

<i>Engineering Beijing Institute of Technology Be* **Instract:** The continuous development of the global energy structure transformation has put forward higher demands upon the

velopment of batteries. The improvements of the energy density have become one of the important **Abstract:** The continuous development of the global energy structure transformation has put forward higher dermands upon the
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under engineering proposes are comprehensively discussed, and udditionally, the appli their engineering prospects are comprehensively discussed, and additionally, the application potential and the main challenges currenty encoundered by solid-state batteries are abo introduced. Finally, this paper gives a rently encountered by solid-state batteries are also introduced. Finally, this paper gives a comprehensive outlook on the development
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this review has illustrated the principle of "multi-

electron reaction materials" and "multi-ion effect",

and summarized the development of multi-electron

where η means

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E_{g} = -\frac{nFE}{\sum M_{g}}\tag{1}
$$

(mol·g⁻¹), *n* represents the charge transfer number, *E*

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\nPrinciple of "multi-
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\nThen, $E_g = \frac{-nFE}{\sum M_g} \eta$

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this review has illustrated the principle of "multi-

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density (*E_i)* as an example, standard conditions. Taking the gravimetric energy

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fol density (*F*_c) as an example, it can be expressed by the six multiclectron reaction materials, such as ferrates,

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 $E_s = -\frac{nFE}{2M_s}$ (1) second "973" project in 2009-2013 has achieved

in which $\sum M_s$ is the total weight of reactants

(mol-g"), *n* represents the char 28(12), 2219011 (2 of 12)
 $E_g = \frac{-nFE}{\sum M_g} \eta$ (2)

where η means the activity of charge carriers, includ-

ing both cations and anions that participate during

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from delivering three-electron transfer per Fe atom^[7]. The
second "973" project in 2009-2013 has achieved a
huge leap in practical energy densities by transferring
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building up multi-electron reaction systems, such as
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is α -NaFeO₂ type layered structure, w

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ment the main focus. In addition, mu aversion-type electrochemistry and materials have

the hexagonal crystal system with the R-3m space

corone the main focus. In addition, multi-valence group. As shown in Figure 2a, Li and O atoms occu-

are groups and in become the main focus. In addition, multi-valence group. As shown in Figure 2a, Li and O atoms occur-
churge curries have played an important role in pythe 3 and 6 sites in the lattice, while transition
building up multicharge carriers have played an important role in

by the 3a and 6c sites in the lattice, while transition

building up multi-electron reaction systems, such as

emeli (TM) atoms and

values are all (TM) at the fol-

zion $mAh \cdot g^{-1}$, thus fulfilling high specific energy density 219011 (3 of 12)

), thus fulfilling high specific energy density

The crystal structure of the ternary material

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gonal crystal system with the $R-3m$ space

s shown in Figu $28(12)$, 2219011 (3 of 12)

mAh·g⁻¹), thus fulfilling high specific energy density

for LIBs. The crystal structure of the ternary material

is α -NaFeO₂ type layered structure, which belongs to

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mAh·g⁻¹), thus fulfilling high specific energy density

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mAh·g⁻¹), thus fulfilling high specific energy density

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con py the 3a and 6c sites in the lattice, while transition
metal (TM) atoms randomly occupy the 3b positions
to form a MO₆ octahedral structure^[8]. In fact, high Ni-
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interlayer will accelerate NiLi mixing, wh acancies produced by Li⁺ extraction from the

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g guest ions in lattice has b r will accelerate Ni/Li mixing, which agena-
structure transformation to spinel and rock-
tures with serious capacity attenuation^[9]. In-
g guest ions in lattice has been reported to
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mAh·g⁻¹), thus fulfilling high specific energy density

anode materials, rather for LIBs. The crystal structure of the ternary material

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sentative electrode active materials are summarized in Table 1. 3 From Cation-to Anion-Redox Cat- hodes $n > 1$, $\eta > 1$): structure, sta- bility and safety According to Equation (1) (high E), the ternary cathode (LiNi _x Co _y Mn _{1-xy} O ₂ , NCM) has exhibited high operational voltage and theoretical capacity (~ 280)			interlayer will accelerate Ni/Li mixing, which aggra- vate the structure transformation to spinel and rock- salt structures with serious capacity attenuation ^[9] . In- troducing guest ions in lattice has been reported to minimize such problems, which can form strong bonds with host ions to suppress cation-mixing (Fig- ure 2b) ^[10] . More promisingly, designing a full concen- tration gradient (FCG) NCM allows for both high ca- pacity and long cycle life ^[11] . Being steadily protected		
			Table 1 Corresponding design strategy and electrochemical performance of representative electrode active materials	Capacity retention	
Electrode type	Material type	Designing strategy	Cycling performance	or voltage hysteresis (Li Li cells)	Ref.
Cathode	$LiNixCoyMn1-xyO2$	"Pillar ions" doping	$162.5 \text{ mA} \cdot \text{g}^{-1}$ at 1 C after 100 cycles	96.2%	$[10]$
		Concentration gradient regulation	$158.7 \text{ mA} \cdot \text{g}^{-1}$ at 1 C after 100 cycles	92.7%	$[12]$
	Li- and Mn-rich based	Surface modification	202 mAh \cdot g ⁻¹ at 1 C after 100 cycles	94.0%	$[18]$
		Doping modification	$222.2 \text{ mA} \cdot \text{g}^{-1}$ at 1 C after 200 cycles	89.2%	$[19]$
Anode	Silicon-based	Nano-crystallization by vapor phase growth	3500 mAh · g^{-1} at 0.2 C after 20 cycles		$[28]$
		Coating by dual-carbon shell	\sim 1350 mAh \cdot g ⁻¹ at 0.2 C after 1000 cycles	75.2%	$[29]$
	Li metal	Composite with carbon fiber and Ag coating	Cycled over 400 h at 1 $mA \cdot cm^{-2}$ with an area capacity of $1 \text{ mA} \cdot \text{cm}^{-2}$	80 mV	$[33]$
		PVDF-HFP coating	Cycled over 1600 h at 1 $mA \cdot cm^{-2}$ with an area capacity of $1 \text{ mA} \cdot \text{cm}^{-2}$	20 mV	$[34]$

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increasing attention due to their anion-redox (Equa-

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 $\#E\#(J. Electrochem.)$ 2022, 28(12), 2219011 (5 of 12)
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lead to the poor cycling stability in practical. There-
fore, Li-S batteries remain in the stage approaching
to commercialization. Recen $28(12)$, 2219011 (5 of 12)
trical conductivity and shuttle effect of polysulfides
lead to the poor cycling stability in practical. There-
fore, Li-S batteries remain in the stage approaching
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trical conductivity and shuttle effect of polysulfides

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fore, Li-S batteries remain in the stage approaching

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trical conductivity and shuttle effect of polysulfides
lead to the poor cycling stability in practical. There-
fore, Li-S batteries remain in the stage approaching
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trical conductivity and shuttle effect of polysulfides

lead to the poor cycling stability in practical. There-

fore, Li-S batteries remain in the stage approaching

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al conductivity and shuttle effect of polysulfides

to the poor cycling stability in practical. There-

Li-S batteries remain in the stage approaching

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11 conductivity and shuttle effect of polysulfides

1 to the poor cycling stability in practical. There-

1.1.4.5 batteries remain in the stage approaching

1.1.4.5 batteries remain in the stage ap cal conductivity and shuttle effect of polysulfides
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psulating S into n trical conductivity and shuttle effect of polysulfides
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capsul lead to the poor cycling stability in practical. There-
fore, Li-S batteries remain in the stage approaching
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capsulating S into nanomaterials for spatial confine-
ment and fore, Li-S batteries remain in the stage approaching
to commercialization. Recent works showed that en-
capsulating S into nanomaterials for spatial confine-
ment and introducing polar adsorption group into
battery compon to commercialization. Recent works showed that en-
capsulating S into nanomaterials for spatial confine-
ment and introducing polar adsorption group into
battery components are more feasible to suppress the
shuttle effect capsulating S into nanomaterials for spatial confinement and introducing polar adsorption group into battery components are more feasible to suppress the shuttle effect of polysulfides (Figure 3d). At present, researchers 电化学(*J. Electrochem.*) 2022, 28(12), 2219011 (5 of 12)

onstructed an O₂-type trical conductivity and shuttle effect of polysulfides

single-layer Li₂MnO₃ lead to the poor cycling stability in practical. There-

ne

ment and introducing polar adsorption group into
battery components are more feasible to suppress the
shuttle effect of polysulfides (Figure 3d). At present,
researchers are exploring the multi-electron reaction
mechanism ttery components are more feasible to suppress the
tuttle effect of polysulfides (Figure 3d). At present,
earchers are exploring the multi-electron reaction
chanism of S electrode to provide theoretical guid-
ce for the co shuttle effect of polysulfides (Figure 3d). At present,
researchers are exploring the multi-electron reaction
mechanism of S electrode to provide theoretical guid-
ance for the commercialization of Li-S batteries.
4 From

 $\# \# \# (J. \text{Electrochem.}) 2022, 28(12), 2219011 (6 of 12)$
ity than the conventional intercalation-based counter-
parts, e.g., silicon (Si)-based anode gives rise to an
ultra-high theoretical capacity (4200 mAh · g⁻¹ for
pure Si a then the conventional intercalation-based counter-

ity than the conventional intercalation-based counter-

(Figure 4b)^[28]; (2) Si/C composite. Apparently, the

parts, e.g., silicon (Si)-based anode gives rise to an
 $# \&L^{\#}(J. \n *Electrochem.*) 2022, 28(12), 2219011 (6 of 12)$ ity than the conventional intercalation-based counter-

(Figure 4b)¹²⁸¹; (2) Si/C composite. Apparently, the

parts, e.g., silicon (Si)-based anode gives rise to an
 ultra-high theoretical capacity (4200 mAh \cdot g⁻¹ for livers much reduced capacity than that of pure Si, yet **E** *(K²²*(*J. Electrochem.*) 2022, 28(12), 2219011 (6 of 12)

ity than the conventional intercalation-based counter-

(Figure 4b)^[28]; (2) Si/C composite. Apparently, the

parts, e.g., silicon (Si)-based anode gives then the conventional intercalation-based counter-
transfer, alloying-then the conventional intercalation-based counter-
(Figure 4b)⁽²⁸⁾; (2) Si/C composite. Apparently, the
parts, e.g., silicon (Si)-based anode gives **EVALUATION**
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ity than the conventional intercalation-based counter-
figure 4b)^[38]; (2) Si/C composite. Apparently, the
parts, e.g., silicon (Si)-based anode gives rise to an
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parts, e.g., silicon (Si)-based anode gives rise to an
c ± 0 the pulse is the increase of the mumber of class of active material consists, e.g., silicon (Si)-based anode gives rise to an equacity of Si/C (with limited Si less than 10%) de-
ultra-high theoretical capacity (42 **EVALUATION**
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pure Si anote). Actually, due to the multi-lectron it is the most commercialization available forms.
transfer pure Si anode). Actually, due to the multi-electron

it is the most commercialization available forms.

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reserv transfer, alloying-type anode inevitably suffers from Regulating Si/C structure is widely accepted, such as
severe volume expansion, e.g., 300% for Si. The huge core-shell structure, yolk shell structure, i.e. Typical-
vo severe volume expansion, e.g., 300% for Si. The huge core-shell structure, yolk-shell structure, etc. Typical-
volume change may lead to the accumulation of in- by, Yu' is team proposed a Si/C anade with two car-
termal s

EVACUAL EXACUATE (EXACUATE (Figure 4) 2022, 28(12), 2219011 (6 of 12)

sity than the conventional intercalation-based counter-

(Figure 4b)¹²⁸; (2) Si/C composite. Apparently, the

parts, e.g., silicon (Si)-based anod the $\mathcal{L}^{\infty}(L, Ebc, model, n)$. (Figure 4b)^[32], (2) 8icC composite. Apparently, the parts, e.g., silicon (65)-based and educes issent is equality of Si/C (with limited Si less than 10%) de-
ultra-high theoretical capacity **EVALUATION**
 EVALUATION the $E^2(z)$ Ebectwehem, 2022, 28(12), 2219011 (6 of 12)

iiy than the conventional intercalation-based counter-

(Figure 4b)^(a8); (2) Si/C composite. Apparently, the

ultra-high theoretical capacity (4200 mAh + z^{-1} f $t\mathbb{E}/\ell^2\ell$. *Leterseben*.) 222, 28(12), 2219011 (6 of 12)
up that, e.g., silicon (Si)-based andode gives rise to an
capacity of Si/C (with limited Si less than 10%) Δ
ultrain-
high theoretical capacity (4200 mMh⁺ g⁻¹ for
liver Si andode). Actually, due to the multi-electron
in its the most commercial reaction available forms.
transfer, allowing-type mode inevitably suffers from
Regulating Si/C structure is widely accepted, such as
transfer, allowing-type mode inevitably suffers from
Regulating Si/C structure is widely accepted, such as
transfer, allowing**Example 10**

interior of (Figure 4b)⁽²⁸⁾; (2) Si/C composite. Apparently, the

iion (Si)-based anode gives rise to an

caractity of Si/C (with limited Si less than 10%) de-

orientical capacity (4200 mAh·g⁻¹ for

liv onventional intercalation-based counter-

(Figure 4b)^[28]; (2) Si/C composite. Apparently, the

licon (Si)-based anode gives rise to an eapacity of Si/C (with limited Si less than 10%) de-

concretical capacity (4200 mA parts, e.g., silicon (Si)-based anode gives rise to an
equacity of Si/C (with limited Si less than 10%) de-
plure Si untochical capacity (4200 mAh · g⁻¹ for livers much reduced equacity than that of pure Si anode). Attu 28(12), 2219011 (6 of 12)

(Figure 4b)^[28]; (2) Si/C composite. Apparently, the

capacity of Si/C (with limited Si less than 10%) de-

livers much reduced capacity than that of pure Si, yet

it is the most commercializa $28(12)$, 2219011 (6 of 12)

(Figure 4b)^[28]; (2) Si/C composite. Apparently, the

capacity of Si/C (with limited Si less than 10%) de-

livers much reduced capacity than that of pure Si, yet

it is the most commercia 28(12), 2219011 (6 of 12)

(Figure 4b)^[28]; (2) Si/C composite. Apparently, the

capacity of Si/C (with limited Si less than 10%) de-

livers much reduced capacity than that of pure Si, yet

it is the most commercializa $28(12)$, 2219011 (6 of 12)

(Figure 4b)^[28]; (2) Si/C composite. Apparently, the

capacity of Si/C (with limited Si less than 10%) de-

livers much reduced capacity than that of pure Si, yet

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(Figure 4b)^[28]; (2) Si/C composite. Apparently, the

capacity of Si/C (with limited Si less than 10%) de-

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(Figure 4b)^[28]; (2) Si/C composite. Apparently, the

capacity of Si/C (with limited Si less than 10%) de-

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capacity of Si/C (with limited Si less than 10%) de-

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capacity of Si/C (with limited Si less than 10%) de-

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livers much reduced capacity than that of pure Si, yet

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capacity of Si/C (with limited Si less than 10%) de-

livers much reduced capacity than that of pure Si, yet

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(Figure 4b)^[28]; (2) Si/C composite. Apparently, the capacity of Si/C (with limited Si less than 10%) de-
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livers much reduced capacity than that of pure Si, yet
it is the most commercializa (Figure 4b)²⁸³; (2) Si/C composite. Apparently, the capacity of Si/C (with limited Si less than 10%) de-
livers much reduced capacity than that of pure Si, yet
it is the most commercialization available forms.
Regulatin (Figure 4b)¹²⁸⁾; (2) Si/C composite. Apparently, the capacity of Si/C (with limited Si less than 10%) delivers much reduced capacity than that of pure Si, yet it is the most commercialization available forms. Regulating Apparently, the
s than 10%) de-
aat of pure Si, yet
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anges capacity of Si/C (with limited Si less than 10%) de-
livers much reduced capacity than that of pure Si, yet
it is the most commercialization available forms.
Regulating Si/C structure is widely accepted, such as
core-shel livers much reduced capacity than that of pure Si, yet
it is the most commercialization available forms.
Regulating Si/C structure is widely accepted, such as
core-shell structure, yolk-shell structure, etc. Typical-
by, Regulating Si/C structure is widely accepted, such as
core-shell structure, yolk-shell structure, etc. Typical-
ly, Yu's team proposed a Si/C anode with two car-
bon cladding layers (Figure 4c). The internal carbon
shell core-shell structure, yolk-shell structure, etc. Typical-
ly, Yu's team proposed a Si/C anode with two car-
bon cladding layers (Figure 4c). The internal carbon
shell provides a limited internal void to allow a large
numbe 电化学(*J. Electrochem.*) 2022, 28(12), 2219011 (6 of 12)
calation-based counter-
(Figure 4b)^[28]; (2) Si/C composite. Apparently, the
anode gives rise to an capacity of Si/C (with limited Si less than 10%) de-
(4200 mAh

 $\#E\#(J. Electron) 2022, 28(12), 2219011 (7 of 12)$
remains challenging to achieve the balance among tion of the LiCl layer and interfacial side reactions
safety, cost and environment requirement. (Figure 4f).
Lithium metal is regar $#E \# (J. Electrochem.) 2022, 28(12), 2219011 (7 of 12)$ Fermains challenging to achieve the balance among

safety, cost and environment requirement.

Lithium metal is regarded as one of the most at-

tractive choices for anode due to i $\# \ell \neq (L \: \text{Electrochem.}) 2022, 28(12), 2219011 (7 of 12)$ mains challenging to achieve the balance among

diverse of the LiCl layer and interfacial side reactions

dety, cost and environment requirement.

Lithium metal is regarde the external content of SLE and the UCF (*LEtectrochem.*) 2022, 28(12), 2219011 (7 of 12)

Termains challenging to achieve the balance among tion of the LiCl layer and interfacial side reactions

safety, cost and environm $#I\&R\#(J. Electron) 2022, 28(12), 2219011 (7 of 12)
\nremains challenging to achieve the balance among
\nsafety, cost and environment requirement.
\nLithium metal is regarded as one of the most at-
\ncapcity of 3860 mAb·g⁻¹ and the lowest reduction
\nopacity of 3860 mAb·g⁻¹ and the lowest reduction
\nproductal (-3.04 V vs. SHE). It can theoretically pro-
\nyide a specific energy exceeding 400 Wh·kg⁻¹ when
\nparing with S or high-Ni NCM cathodes. However,
\nthe annlication of metallic lithium anode is still in the
\n
$$

$$$ $\pm 1/2$ (*L. Electrochem.*) 2022, 28(12), 2219011 (7 of 12)
 premains challenging to achieve the balance among tion of the LiCl layer and interfacial side reactions

safety, cost and environment requirement.

Lithium m **under the specific energy exceeding to a set of the Eight Chernochem,** 2022 **,** $28(12)$ **,** 2219011 **(7 of 12)

remains challenging to achieve the balance among tion of the LiCl layer and interfacial side reactions

safety EXAM CONTRON THE S ON THE S ORTER IN THE S OR THE INTERENT CONDUCT THE INTERENT CONDUCT THE INTERENT (SUPPRETENT CONDUCTS) THE SERVICE INTERENT CONDUCTS (Figure 41).

Lithium metal is regarded as one of the most at-
 \begin** the application of metallic lithium anode is still in the **Example 19** EVA: *Electrochem.* (2022, 28(12), 2219011 (7 of 12)

remains challenging to achieve the balance among tion of the LiCl layer and interfacial side reactions

stacky, cost and environment requirement.

(Figure $\pm \frac{1}{2}$ the $\frac{2}{2}$ (*k Mechrochem*) 2022, 28(12), 2219011 (7 of 12)

remains challenging to achieve the balance among tion of the LiCl layer and interfacial side reactions

statey, cost and environment requiremen **Example 1988**
 Example 1989 (Electrochem.) 2022, 28(12), 2219011 (7 of 12)
 Example 1989 (Figure 4f).
 Example 1989 (Figure 4f).
 Example 1989 (Figure 4f).
 Example 1989 (Figure 4f).
 Example 1999 (Figure 4f) **Example 10**
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streamins challenging to achieve the halance among tion of the LiCl layer and interfacial side reactions

safety, cost and environment requirement.

L Moreover, the active Li is also easily detached from the remains challenging to achieve the balance among
safety, cost and environment requirement.

Eightiom metal is regarded as one of the most at-

Traition metal is regarded as one of the most at-

Traition collector of the c ermains challenging to achieve the balance arison on the Lict layer and interfacial side reactions
safety, cost and environment requirement. (Figure 4f).
Lithium metal is regarded as one of the most at-
 $\frac{1}{5}$ **From Li** streyt, cost and environment required to solve these problems, and U and U Linum metal is regarded as one of the most at-

Linum and its responsed to the most at the content of the state of the most at

tractive choices for and due to its high theoretical

empericinal (-3.04 V vs. SHE). It can t tractive choices for anode due to its high theoretical
equation (31860 mAch $_2$ ⁻² and the lowest reduction
equation (31860 mAch $_2$ ⁻² and the lowest reduction
potential (-3.04 V vs. SHF). It can theoretically pro-
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potential (-3.04 V vs. SHF). It can theoretically pro-
potential (-3.04 V vs. SHF). It can theoretically pro-
prigging with S or high-Ni NCM cathodes. However, in thigh l potential (-5.04 V vs. Srits). It can theoretically pro-

by the a specific energy exceeding 400 Wh, kg⁻¹ when

a specific energy exceeding 400 Wh, kg⁻¹ when

the application of metallic lithium anode is still in the
 Use Y as SHT). It can theoretically pro-

i. Go the corresponse and Mh kg⁻¹ when

is corresponsed and OWh kg⁻¹ when

So thigh-Ni NCM cathodes. However,

so the profession of metallic lithium anode is still in the

bat Vote a spectre energy exceeding 400 Wir Kg² when ϵ when the shown is the formulation of metallic lithium and is still in the development of LBs. Solid-state lithium the application of metallic lithium ande is still i party with S or igni-N Pot reconsoss. However,
the application of metallic linkinum ande is still in the batteries (SSLBs) are expected to imhibit safety haz-
very initial stage due to its obvious defects (Figure and selu the approach or metalle inturn manoe is stul in the component of reacting the physical component of excellent e very minal stage oue to its convois decrets (rigure

and stage and to its convois decrets (rigure)

4d), such as the uncontrollable growth of Li dendries. The anticials As shown in Figure 5, current SSBs can be

In additio Ho, such as the uncontrolation growth of Latentines,

In addition, the unstable SEU is easily broken during

in divided into polymer and oxygen group inorganic

cycling, resulting in direct contact and the consump-

compo na and the unstable Est is easily broken attracts and the consumptional procedure of the system and the system of electrolyte, thereby shortening the cycle life. The materials.

Moreover, the active Li is also easily deta eyourg, resulting in direct contact and in economic values of the presentation of decoration of electrochemical performance (some resulting the speed in of electrochemical performance in the state of the metric of orm "de thermore interestive interestive interestive and the existence of the thermore incided the coefformation of the
content of the fluit includes the coefformation of Method from the SLI **Organic Polymer Electrolytes.**
CL and Moreover, the actres in is also easily encenee of momotometric and the set and the set and the set are the set as easy of the set are the set as the set are the set as the set of the set are the set of the set of the set eurent conserts of form cean in , when recuses the conservation of the properties, the conservation due to their good safety properties,
In order to solve these problems, a number of carbon deviativity, easy processing int CE and tuttier deteriorates the eycle performance", tensive attention inter to their good sates (proton interface containing the other and tuttier deterioration interface containing the protocomplement and excellent host In order to solve these problems, a number of carton incomer of carrolling, cas processing mo mins, and excellent
In order to solve the solven inplemented, being supposed to interfrictal contact, such as polyvinyl nitrile nost have been implemented, been ally supposed to unitarized counterpolation, by the buffer the volume expansion^[14, 21]. Moreover, they al-

buffer the volume expansion^[14, 21], Moreover, they al-

so have the ability ourier ine volume expansion²²²². Moreover, they all physochouse (τιλ), physicaly direct provides direct provides direct provides the basility to regulate the transports of election and Li* so that Li can be uniformly so have the abouty to regulate the furtaples of elec-
then and L is other L is an beniformly deposited in-
the (PVDF), and poly (vinylidene fluoride-co-bes-
side the host in the absence of dendrites. As shown in alluoropr from antal so that I can be untrompy appeared mi-

side the host in the absence of dendrites. As shown in the corvery, and poly quence none-co-tens-

Figure 4e, Zhang et al.^{px}l designed a Li-infused car-

every polymer-

电化学(*J. Electrochem.*) 2022, 28(12), 2219011 (7 of 12)

e the balance among tion of the LiCl layer and interfacial side reactions

quirement. (Figure 4f).

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to its bigh theoration

$\#I\# \# (J. Electrochem.) 2022, 28(12), 2219011 (7 of 12)$ ieve the balance among tion of the LiCl layer and interfacial side reactions

requirement. (Figure 4f).

d as one of the most at-
 5 From Liquid to Solid-State Lithi-

lue to ²⁸⁽¹²⁾, ²²¹⁹⁰¹¹ (7 of 12)
tion of the LiCl layer and interfacial side reactions
(Figure 4f).
**5 From Liquid to Solid-State Lithi-
um Batteries: High Safety and En-** $\frac{28(12), 2219011 (7 \text{ of } 12)}{25(12), 2219011 (7 \text{ of } 12)}$

(Figure 4f).
 **5 From Liquid to Solid-State Lithi-

um Batteries: High Safety and En-**

ergy Density ²⁸⁽¹²⁾, ²²¹⁹⁰¹¹ (7 of 12)

tion of the LiCl layer and interfacial side reactions

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 **5 From Liquid to Solid-State Lithi-

um Batteries: High Safety and En-

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tion of the LiCl layer and interfacial side reactions

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 **5 From Liquid to Solid-State Lithi-

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(Figure 4f).
**5 From Liquid to Solid-State Lithi-
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 From Liquid to Solid-State Lithi-
 **m Batteries: High Safety and En-

gy Density**

High levels of energy density and safety are re $\frac{28(12)}{2219011(7 \text{ of } 12)}$
tion of the LiCl layer and interfacial side reactions
(Figure 4f).
5 From Liquid to Solid-State Lithi-
um Batteries: High Safety and En-
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(Figure 4f).
 5 From Liquid to Solid-State Lithi-
 um Batteries: High Safety and En-
 ergy Density

High levels of energy density 28(12), 2219011 (7 of 12)

tion of the LiCl layer and interfacial side reactions

(Figure 4f).
 5 From Liquid to Solid-State Lithi-
 and Batteries: High Safety and En-
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High levels of energy density an 28(12), 2219011 (7 of 12)

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 5 From Liquid to Solid-State Lithi-
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High levels of energy density and 28(12), 2219011 (7 of 12)

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(Figure 4f).

5 **From Liquid to Solid-State Lithi-**
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5 **From Liquid to Solid-State Lithi-**
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High levels of energy density and s **n** of the LiCl layer and interfacial side reactions
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 The Batteries: High Safety and En-
 The Batteries: High Safety and En-
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High levels of energy densi tion of the LiCl layer and interfacial side reactions
(Figure 4f).
 5 From Liquid to Solid-State Lithi-
 um Batteries: High Safety and En-
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High levels of energy density and safety are requisite for th (Figure 4f).
 5 From Liquid to Solid-State Lithi-
 um Batteries: High Safety and En-
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High levels of energy density and safety are requi-

site for the development of LIBs. Solid-state lithium

batteri 5 **From Liquid to Solid-State Lithi-**
 um Batteries: High Safety and En-
 ergy Density

High levels of energy density and safety are requisite for the development of LIBs. Solid-state lithium

batteries (SSLBs) are ex

since more in the assence or denotines. As shown in all subseque the transmitter (μ , ν) and μ a Figure 4e, Zhang et al.² designed a L_o-Hnused car-
bon fiher composite, where the surface of carbon be pairs of electrons on heterostoms in the polymer
framework (CF) was modified with a lithiophilic silver chains⁵⁹¹ from the reomposits, where the surface of carrow one pairs of eactnooming in the polymer framework (CF) was modified with a lithiophilic silver chains³⁹¹. The free volume is generated under the botal conting (CF/Ag). By Transwork (CP) was modulme what although thus although the change of the polymer change of CF/Ag. By comparison, the CF/Ag. Li exhibits segmental motion of the polymer chania, thereby re-
no dendrite and "deal Li" during **um Batteries: High Safety and En-**
 ergy Density

High levels of energy density and safety are requisite for the development of LIBs. Solid-state lithium

batteries (SSLBs) are expected to inhibit safety haz-

ands and **ergy Density**

High levels of energy density and safety are requisite for the development of LIBs. Solid-state lithium

batteries (SSLBs) are expected to inhibit safety haz-

ards and better inplement the multi-electron r **Figure 12** Extracted computes the development of LIBs. Solid-state lithium site for the development of LIBs. Solid-state lithium batteries (SSLBs) are expected to inhibit safety hazards and better implement the multi-ele righ rotest or diety during a site of the divelopment of LIBs. Solid-state lithium
site for the development of LIBs. Solid-state lithium
and better implement the multi-electron reaction
and better implement the multi-elect afluoropropylene) (PVDF-HFP) (Figure 5a). Li⁺ achi-Solvet in the minimism of the solvet and between
that is solvet in the minimism batteries (SSLBs) are expected to inhibit safety haz-
ards and better implement the multi-electron reaction
divided into polymer and oxygen gr batteric (BDEE) are valued to finite detectron reaction
and better implement the multi-electron reaction
materials. As shown in Figure 5, current SSBs can be
divided into polymer and oxygen group inorganic
compounds (oxide and was the mpelliont are main elected in the divided into polymer and oxygen group inorganic divided into polymer and oxygen group inorganic compounds (oxide, sulfide, etc.) according to the type of materials.
5.1 **Organ** materials. To shown in Figure 2, variant 2023 can be
divided into polymer and oxygen group inorganic
compounds (oxide, sulfide, etc.) according to the type
of materials.
Polymer-based solid electrolytes have received ex-
 alizing the conduction of the conductivity, every compounds (oxide, sulfide, etc.) according to the type of materials.
 5.1 Organic Polymer Electrolytes.

Polymer-based solid electrolytes have received extensive attenti compounts (state, statine, tee.) according to the yper
of materials.
5.1 Organic Polymer Electrolytes have received ex-
tensive attention due to their good safety properties,
flexibility, easy processing into films, and e 5.1 **Organic Polymer Electrolytes.**

Polymer-based solid electrolytes have received ex-

tensive attention due to their good safety properties,

flexibility, easy processing into films, and excellent

interfacial contact, **EXECT OF THE EXECT CONSTRANGES.** Polymer-based solid electrolytes have received extensive attention due to their good safety properties, flexibility, easy processing into films, and excellent interfacial contact, such as rolyinet-oased solid electrolytes have received ex-
tensive attention due to their good safety properties,
flexibility, easy processing into films, and excellent
interfacial contact, such as polyethylene oxide (PEO),
poly tensive attention tue to their good sately properties,
flexibility, easy processing into films, and excellent
interfacial contact, such as polyethylene oxide (PEO),
polycarbonate (PC), polyvinyl nitrile (PAN), poly-
methy nexiomity, easy processing into imits, and excenent
interfacial contact, such as polyethylene oxide (PEO),
polycarbonate (PC), polyvinyl nitrile (PAN), poly-
methyl methacrylate (PMMA), polyvinylidene difluo-
ride (PVDF), meriacial contact, such as polyemytene oxtee (FEO), polyearbonate (PC), polyinyl nitrile (PAN), polymethyl methacrylate (PMMA), polyvinylidene difluo-
ride (PVDF), and poly (vinylidene fluoride-co-hex-afthoropropylene) (PV polycaroniate (r.c.), polyvinyi initie (r.ars), poly-
methyl methacrylate (PMMA), polyvinylidene difluo-
ride (PVDF), and poly (vinylidene fluoride-co-hex-
afluoropropylene) (PVDF-HFP) (Figure 5a). Li⁺ achi-
eves polymer metally inetalactylate (FMMA), polyvinylatene dinato-
ride (PVDF), and poly (vinylidene fluoride-co-hex-
afluoropropylene) (PVDF-HFP) (Figure 5a). Li⁺ achi-
eves polymer-to-salt solvation by coordinating with
one pairs densed -- OH groups, which constitutes "Li+-solva-The (VDF), and poly (vinylidence indome-co-inex-
afluoropropylene) (PVDF-HFP) (Figure 5a). Li⁺ achi-
eves polymer-to-salt solvation by coordinating with
lone pairs of electrons on heteroatoms in the polymer
chains¹⁵⁸ anuoropropylene) ($\bf r$ v $\bf D1$ -11 $\bf r$) (Figure 5a). Et atmetes polymer-to-salt solvation by coordinating with lone pairs of electrons on heteroatoms in the polymer chains¹⁵⁸. The free volume is generated under the loc eves polymet-to-sant solvation by coordinating with
lone pairs of electrons on heteroatoms in the polymer
chains¹⁵⁹. The free volume is generated under the local
segmental motion of the polymer chain, thereby re-
alizin ble pairs of electrols on interbations in the polynical
chains^[58]. The free volume is generated under the local
segmental motion of the polymer chain, thereby realizing the conduction of Li⁺ within and between
chains chains \cdot . The free volume is generated under the local segmental motion of the polymer chain, thereby realizing the conduction of Li⁺ within and between chains^[56]. The polymer electrolytes are restrained by the lo by re-
tween
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HMP)
4.6 V),

 $\#E\#$ (*L Electrochem.*) 2022, 28(12), 2219011 (8 of 12)
flexible-rigid mechanical properties. Furthermore, the ide electrolytes are unstable to Li metals, and interfa-
first-principles calculation reveals the mechani \pm *l*^{2 \neq}*(I. Electrochem.*) 2022, 28(12), 2219011 (8 of 12)
flexible-rigid mechanical properties. Furthermore, the ide electrolytes are unstable to Li metals, and interfa-
first-principles calculation reveals the the suppression of the suppression of probability $\mathbb{E}(t)$. *Electrochem.*) 2022, 28(12), 2219011 (8 of 12)
flexible-rigid mechanical properties. Furthermore, the ide electrolytes are unstable to Li metals, and interfacomposition due to the 3D structure. As a result, high ⁱE*K²*²(*J. Electrochem.*) 2022, 28(12), 2219011 (8 of 12)

flexible-rigid mechanical properties. Furthermore, the ide electrolytes are unstable to Li metals, and

first-principles calculation reveals the mechanism th $\mathcal{H}\mathcal{H}^{\infty}(L$ *Electrochem.*) 2022, 28(12), 2219011 (8 of 12)
 flexible-rigid mechanical properties. Furthermore, the ide electrolytes are unstable to Li metals, and interfa-

first-principles calculation reve **EVALUATION**
 EXALUATE: ELAT (Example 1902) 22, 28(12), 2219011 (8 of 12)
 Elexible-rigid mechanical properties. Furthermore, the ide electrolytes are unstable to Li metals, and interfa-

first-principles calculatio **ELECT Electrochem.**) 2022, 28(12), 2219011 (8 of 12)
 Elexible-rigid mechanical properties. Furthermore, the ide electrolytes are unstable to Li metals, and interfa-

first-principles calculation reveals the mechanism **Existing and model and model and model and model and model and the situal polymetrics and interfa-
first-principles calculation reveals the mechanism for ide electrolytes are unstable to Li metals, and interfa-
first-pri** $\text{ft}E^{\#}(L\text{~}Electrochem})$ 2022, 28(12), 2219011 (8 of 12)

flexible-rigid mechanical properties. Furthermore, the

ide electrolytes are unstable to Li metals, and interfa-

first-principles calculation reveals the mechanism **Example 12**
 Exidentical properties. Furthermore, the ide electrolytes are unstable to Li metals, and interfact

frist-principles calculation reveals the mechanism for ide electrolytes are unstable to Li metals, and in **EVALUAT UNIT (EXAMPLE 1998)**
 EXAMPLE 10 (E^2 (*L Electrochem*,) 2022, 28(12), 2219011 (8 of 12)
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First-principles calculation reveals the mechanism for

tied cleretrolytes are unste 10^{-4} S \cdot cm⁻¹ at room temperature. Furthermore, a sol**id:** $\mathcal{L}E\llap{\pm}k$ (*L Electrochem.*) 2022, 28(12), 2219011 (8 of 12)
 flexible-rigid mechanical properties. Furthermore, the ide electrolytes are unstable to Li metals, and interfa-

first-principles calculation thereafter in the thermoderation 2022, 28(12), 2219011 (8 of 12)

flexible-rigid mechanical properties. Furthermore, the

ide electrolytes are unstable to Li metals, and interfa-

first-principles calculation reveals the The
xible-rigid mechanical properties. Furthermore, the ide electrolytes are unstable to Li metals, and interfa-
first-principles calculation reveals the mechanism for ide electrolytes are unstable to Li metals, and inter flexible-rigid mechanical properties. Furthermore, the ide electrolytes are unstable to Li metals, and interfa-
first-principles calculation reveals the mechanism for call modifications are required¹⁶⁹. Current research any Li anode oxidation or capacity decay^[38]. In order ionic conductivity (even over 10^{-2} S -et o achieve sufficiently high electrochemical window, ever, it suffers from narrowed voltage r. Change at al.¹⁹⁹⁹ ulitze

²/*Electrochem.*) 2022, 28(12), 2219011 (8 of 12)

Furthermore, the ide electrolytes are unstable to Li metals, and interfa-

e mechanism for cial modifications are required^[40]. Current researches

high-voltage de-
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 I mechanical properties. Furthermore, the ide electrolytes are unstable to Li metals, and interfa-

cs calculation reveals th first-principles calculation reveals the mechanism for

eig modifications are required¹⁶⁸. Current researches

density of proton-induced high-voltage de-

egencally adopt proton-induced highers on composition of protonthe suppression of proton-induced high-voltage de-

cornepsifon due to the 3D structure. As a result, high surface to stabilize the interface

composition due to the 3D structure. As a result, high surface to stabilize th composition due to the 3D structure. As a result, high

ionic conductivity over 0.1 in Semi- at room temperator or

incited in Semi- at room temperator or the stability of the stability

ionic conductivity over 0.1 in Sem ionic conductivity over 0.1 mS·cm⁻¹ at room tempera-

oretically, Li et al.⁽⁶¹⁾ used first principles calculations

breaking through the inherent obstacles faced by noly-

the ly prevent the suitable coaling layer can ture is achieved with 4.6 V stable voltage window,

for prove that the suitable coating theyer can success-

breaking through the inhetent obstacles faced by poly-

imaging through the interest of the double properties co breaking through the inherent obstacles faced by poly-
met fuctory transfer from lithium metal
mere electrolyte⁵⁷¹, More recently, Wu et al. has pro-
nois to inchibit the dendrite growth (Figure 56). Ex-
posed an *in-si* mer clectrolytes^[97]. More recently, Wu et al. has pro-

to SSE to inhibit the dendritic growth (Figure Sc). Ex-

(LAGP) inorganic and lot yields (het ($P(0)$), perimentally, Z hu et al.¹⁰⁰ sprayed connervial beron
 posed an *in-situ* polymerization of Li_nAl_nGe₁₅(Po₁), perimentally, Zhu et al.⁽ⁱⁱ⁾ sprayed commercial boron con-

(LAGP) inorganic and polytelythere glycol metha-

uride-based release agent (BNRA) on LATP to con (LAGP) inorganic and poly(eltylene glyool metha-

nitride-based release agent (BNRA) on LATP to con-

ery plate) (PEGMA) based hybrid electrolyte system. struct a composite protective layer. The chemical in-

The ionic co erylate) (PEGMA) based hybrid electrolyte system. struct a composite protective layer. The chemical in-
The ionic conductivity was greatly improved to 2.37x reneation between ISN and Li forms Li—N bonds, reneation
10⁻¹ S The ionic conductivity was greatly improved to 2.37×

teraction between BN and Li forms Li—N bonds, re-

id-s'-c-m⁻¹ at room temperature. Furthermore, a sol-

id-selectrolyte-lithium integrated anode was synthe-

Li' mi 10⁻¹ S·cm⁻¹ at room temperature. Furthermore, a sol-

diectrolyte-linium iniegrated anode was synthe-

inigration and interfacial constrained is sized thereating band interfacial compared

sized thereather, which can id-electrolyte-lithium integrated anode was synths-

Li'migration and interfacial compatibility. Compared

sized threather, which can support the assembly of with botic-based are boty higher-based electrolytes, sul-

sSLBs sized thereafter, which can support the assembly of

sixtels in ambient conduction (30% humidity) without fide-based electrolytes show higher room-temperature

sixtels in ambient conductivity (see the conductivity (even o SSLBs in ambient condition (30% humidity) without

male coidation or capacity density and the chosen extent in the coincity (con over 10 -8 cm⁻¹⁹^{tam}. However,

to achieve sufficiently high electrochemical window,

to to achieve sufficiently high electrochemical window, ever, it suffers from narrowed voltage range, interface
Cheng et al.²⁰¹ utilized dual-salt components (LiBOB issue and poor air stability¹⁶⁻⁸⁰. In particular, sulfi e et al.¹⁹⁰ utilized dual-satt components (LiBOB issue and poor air stability¹⁴⁷⁴⁹¹). In particular, sulf

iciClO₁ to conhance the electrochemical stability based electrolytes have one cletrochemical stab

wow of PEO d LiClO.) to enhance the electrochemical stability
hased electrolytes have poor electrochemical stability
nodow of PEO-hased composite electrolytes up to and compatibility with high-voltage can
host SU(give
V. The addition window of PEO-based composite electrolytes up to

and compatibility with high-voltage cathoeds (Figure

4.7 V. The addition of the dauble salt component 5b. Furthermore, sulfide-based electrolytes are not

formed stable S 4.7 V. The addition of the double salt component 5f). Furthermore, sulfide-based electrolytes are not formed stable SF1 and cathode electrolyte interfaces resistant to the air, which react with H₁O in the informed stabl formed stable SFI and cathode electrolyte interface resistant to the air, which react with H₂O in the air
(CEI) at the anode and cathode interfaces, respective-
and relation of Li-B-O/C-O species originated from the sta (CEI) at the anode and cathode interfaces, respective-
on trelation (SS gas. Compared with inorganic
oralism can divident compute the solid electrolyte, quasis-ofid electrolyte candinal for-
oralism of LiBOB effectively p Iy. The multiplied polymer-ion interactions and for-

solid electrolyte, quasi-solid electrolyte or gel elec-

mation of Li-B-OC-O species originated from the trolyte exhibit better processing performance, flexi-

(oxalat mation of Li-B-O/C-O species originated from the trolyte exhibit better processing performance, flexi-
(oxalato borate group of Li-BOB effectively promoto bility and more importantly the acceptably high ionic
the electroc (oxalato)borate group of LiBOB effectively promote

inlity and more importantly the acceptably high ionic

electrochemical stability of the PEO based com-

conductivity at room temperature, which can guaran-

posite clect 28(12), 2219011 (8 of 12)
ide electrolytes are unstable to Li metals, and interfa-
cial modifications are required^[40]. Current researches
generally adopt porous structures or coating layers on
surface to stabilize the 28(12), 2219011 (8 of 12)
ide electrolytes are unstable to Li metals, and interfa-
cial modifications are required^[40]. Current researches
generally adopt porous structures or coating layers on
surface to stabilize the 28(12), 2219011 (8 of 12)
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ide electrolytes are unstable to Li metals, and interfa-

cial modifications are required^[40]. Current researches

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cial modifications are required^[40]. Current researches
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ide electrolytes are unstable to Li metals, and interfa-
cial modifications are required^[49]. Current researches
generally adopt porous structures or coating layers on
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cial modifications are required^[40]. Current researches
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cial modifications are required^[40]. Current researches
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ide electrolytes are unstable to Li metals, and interfa-

cial modifications are required¹⁴⁰. Current researches

generally adopt porous structures or coating layers on

surface to stabilize t ide electrolytes are unstable to Li metals, and interfa-
cial modifications are required^[40]. Current researches
generally adopt porous structures or coating layers on
surface to stabilize the interface of Li metal^{[41,} ide electrolytes are unstable to Li metals, and interfa-
cial modifications are required^[40]. Current researches
generally adopt porous structures or coating layers on
surface to stabilize the interface of Li metal^{[41,} ionic conductivity (even over 10^{-2} S \cdot cm⁻¹)^[44-46]. Hownd interfa-
researches
g layers on
 $1^{[41,42]}$. The-
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al in cial modifications are required¹⁴⁰. Current researches
generally adopt porous structures or coating layers on
surface to stabilize the interface of Li metal^[41,42]. The-
oretically, Li et al.¹⁴¹ used first principle generally adopt porous structures or coating layers on
surface to stabilize the interface of Li metal^[41,42]. The-
oretically, Li et al.^[41] used first principles calculations
to prove that the suitable coating layer surface to stabilize the interface of Li metal^[41,42]. The-
oretically, Li et al.^[41] used first principles calculations
to prove that the suitable coating layer can success-
fully prevent the electron transfer from l oretically, Li et al.⁽⁴¹) used first principles calculations
to prove that the suitable coating layer can success-
fully prevent the electron transfer from lithium metal
to SSE to inhibit the dendritic growth (Figure 5e to prove that the suitable coating layer can success-
fully prevent the electron transfer from lithium metal
to SSE to inhibit the dendritic growth (Figure 5e). Ex-
perimentally, Zhu et al.^[43] sprayed commercial boron
 fully prevent the electron transfer from lithium metal
to SSE to inhibit the dendritic growth (Figure 5e). Ex-
perimentally, Zhu et al.^[43] sprayed commercial boron
nitride-based release agent (BNRA) on LATP to con-
str to SSE to inhibit the dendritic growth (Figure 5e). Ex-
perimentally, Zhu et al.^(a3) sprayed commercial boron
nitride-based release agent (BNRA) on LATP to con-
struct a composite protective layer. The chemical in-
tera perimentally, Zhu et al.⁽⁴³⁾ sprayed commercial boron
nitride-based release agent (BNRA) on LATP to con-
struct a composite protective layer. The chemical in-
teraction between BN and Li forms Li—N bonds, re-
duces the nitride-based release agent (BNRA) on LATP to construct a composite protective layer. The chemical in-
teraction between BN and Li forms Li—N bonds, re-
duces the interfacial resistance, and thereby promotes
Li⁺ migrati struct a composite protective layer. The chemical in-
teraction between BN and Li forms Li—N bonds, re-
duces the interfacial resistance, and thereby promotes
Li⁺ migration and interfacial compatibility. Compared
with o teraction between BN and Li forms Li—N bonds, re-
duces the interfacial resistance, and thereby promotes
Li^{*} migration and interfacial compatibility. Compared
with oxide-based electrolytes show higher room-temperature
i duces the interfacial resistance, and thereby promotes
Li^{*} migration and interfacial compatibility. Compared
with oxide-based and polymer-based electrolytes, sul-
fide-based electrolytes show higher room-temperature
ion Li⁺ migration and interfacial compatibility. Compared
with oxide-based and polymer-based electrolytes, sul-
fide-based electrolytes show higher room-temperature
ionic conductivity (even over 10^{-2} S \cdot cm⁻¹)^{(44,4} with oxide-based and polymer-based electrolytes, sul-
fide-based electrolytes show higher room-temperature
ionic conductivity (even over 10^{-2} S·cm⁻¹)⁽⁴⁴⁻⁶⁰). How-
ever, it suffers from narrowed voltage range, inter fide-based electrolytes show higher room-temperature
ionic conductivity (even over 10^{-2} S·cm⁻¹)^(44,46). How-
ever, it suffers from narrowed voltage range, interface
issue and poor air stability^{(47,40}). In particu ionic conductivity (even over 10^{-2} S \cdot cm⁻¹)^{(44,46}). How-
ever, it suffers from narrowed voltage range, interface
issue and poor air stability^{(47,49}). In particular, sulfide-
based electrolytes have poor elect ever, it suffers from narrowed voltage range, interface
issue and poor air stability^($\pi x \omega$). In particular, sulfide-
based electrolytes have poor electrochemical stability
and compatibility with high-voltage cathodes issue and poor air stability^{(*a*7,*a*9). In particular, sulfide-
based electrolytes have poor electrochemical stability
and compatibility with high-voltage cathodes (Figure
5f). Furthermore, sulfide-based electrolytes ar} eed electrolytes have poor electrochemical stability
d compatibility with high-voltage cathodes (Figure
r. Furthermore, sulfide-based electrolytes are not
is tant to the air, which react with H₂O in the air
d release tox and compatibility with high-voltage cathodes (Figure 5f). Furthermore, sulfide-based electrolytes are not resistant to the air, which react with H₂O in the air and release toxic H₂S gas. Compared with inorganic solid e 5f). Furthermore, sulfide-based electrolytes are not
resistant to the air, which react with H₂O in the air
and release toxic H₂S gas. Compared with inorganic
solid electrolyte, quasi-solid electrolyte or gel elec-
tro resistant to the air, which react with H₂O in the air
and release toxic H₂S gas. Compared with inorganic
solid electrolyte, quasi-solid electrolyte or gel elec-
trolyte exhibit better processing performance, flexi-
bil and release toxic H₂S gas. Compared with inorganic
solid electrolyte, quasi-solid electrolyte or gel elec-
trolyte exhibit better processing performance, flexi-
bility and more importantly the acceptably high ionic
condu solid electrolyte, quasi-solid electrolyte or gel electrolyte exhibit better processing performance, flexi-
bility and more importantly the acceptably high ionic
conductivity at room temperature, which can guaran-
tee suff trolyte exhibit better processing performance, flexi-
bility and more importantly the acceptably high ionic
conductivity at room temperature, which can guaran-
tee sufficient ion conduction, form a stable contact
between t bility and more importantly the acceptably high ionic
conductivity at room temperature, which can guaran-
tee sufficient ion conduction, form a stable contact
between the electrode and effectively avoid battery
failure cau 电化学(*J. Electrochem.*) 2022, 28(12), 2219011 (8 of 12)

erties. Furthermore, the ide electrolytes are unstable to Li metals, and interfa-

rals the mechanism for ide electrolytes are required^[40]. Current researches

u

is capacity, low average delithiation potential, and detective interaction potential capacity, low average delithiation potential, and detective interaction potential, for the main terrado of declene interaction capacity, Example 2022
 Example 2022 EXERCT CONSULTER CON COMPRESE ACTES AC Contention
 Continuously and the industrial in the industrial interference in solid-state be Example 19 (EE) formed by the O/C-O species
 Example 19 and the content to developeration of the main three technology routes in solid-state batteries, (a) Common polymer based solid electrolyte systems; (b) Synthetic **Figure 5** Overview diagram of the main three technology routes in solid-state butteries. (a) Common polymer based solid electrolyte systems; (b) Synthetic scheme of PHMP composite solid electrolyte. Reproduced with permi **Figure 5** Overino diagram of the main three technology routes in solid-state batteries. (a) Common polymer based solid clear-
Figure 3, the main three technology routes in solid allocated with permission of Ref. 37, copyr **EXERCT SERVE AND THE SAFET CONDUCTS AND THE SAFET CONDUCTS (SERVED AND THE SAFET CONDUCTS OF THE SAFET CONDUCTS (SERVED AND THE SAFET CONDUCTS) And the safety performance. Reproduced with permission of Ref. 45, copyright** SCRICM CONDITION DETROIT CONDITION DETROIT CONDITION TO DETROIT ON A SALE SURVEY OF LABORATION SUGGERED MORE CONSUMING THE SURVEY COLLECT STREAM VALUE CONTINUES IN SURVEY COLLECT STREAM SURVEY OF A SURVEY COLLECT STREAM S SCRICH PROCESS CONDITIONS CONDITIONS OF ORDER CONDITIONS (PROCESS ORDER CONDUCTS) $\frac{p_{\text{S}}}{p_{\text{S}}}}$ is $\frac{p_{\text{S}}}{p_{\text{S}}}}$ and solid-state batteries. (a) Common polymer based solid electrolyte. Reproduced with permi

intely systems; (b) Synthetic scheme of PHMP compose solid electrolyte. Reproduced with permassion of Ref. 37, copyright 2021, Electric interface is able structure of the film of the mission of Ref. 39, copyright 2021, Ele Whey (e) scontinuus
The Microsoften and the anisomator of the anisomation of the system and the system and popularization of the R 39, copyright 2012, Elsevier. (d) Schematic diagram of typical oxide and sulfade electrolyt by extractively a constrained unique and supprementation of Ref. 45, copyright 2022, Royal Society of the specified points and supprement of the specified composite components. Composite and metal-based components and Nef Chemikity, (c) Schematic illustration of the role of protective layers on suppressing Li denditie formation in SSEs. Reproduced with permission of Ref. 41, copyrign 2022, Elsevier, (i) Schematic diagram of interface instab permission of Ref. 41, copyright 2022, Elsevier. (f) Schematie diagram of interface instability between sulfide electrolyte and high voltage cathode. Reproduced with permission of Ref. 48, copyright 2022, wiley. (color on voltage cathode. Reproduced with permission of Ref. 48, copyright 2022, Wiley. (color on line)

ic capacity, low average delithiation potential, and ductivity and the safety performance of the elec-

multi-clectron reactio ic enpacity, low average delithiation potential, and ductivity and the safety performance of the clearentified-celeroon reaction characteristies are becoming trolyte itself still need more efforts to optimize. In the main

Example 18 The magnitude of the majorithm is the majorithm of the majorithm of the majorithm of the majorithm is the majorithm of the m EXAMPRED FOR ADDETERT CONDUCT THEORY CONDUCTS ADDETERT IS A SAMPLE REPOID CONDIC, Repointing volid-state batteries. (a) Common polymer based solid electrom
In solid-state batteries. (a) Common polymer based solid electr **EXAMPLE TO A COMOVE ASSEMALL CONDUCT ASSEMALL CONDUCT**
 EXAMPLE ASSEMALL ASSEMALL ASSEMALL ASSEMALL ASSEMALL ASSEMALL AND A CONDUCT A CONDUCT A CONDUCT A CONDUCT A CONDUCT A CONDUCT A MOREOVER SURFAPT AND A SURFAPT AND a solid-state batteries. (a) Common polymer based solid elec-
rolyte. Reproduced with permission of Ref. 37, copyright 2022,
high voltage cycling performance. Reproduced with permission
ral oxide and sulfide electrolytes c is solid-state batteries. (a) Common polymer based solid elec-
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and indeper cycling performance. Reproduced with permission
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thigh voltage cycling performance. Reproduced with permission
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多电子反应材料推动高能量密度电池发展: 材料与体系创新

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摘要: 全球能源结构转型推动了电化学储能系统的飞速发展, 提高能量密度是发展新型二次电池的重要方向和 研究热点。然而,受限于传统的嵌入式反应,锂离子电池在能量密度上已经逐渐达到极限。要发展更高能量密度 的新型二次电池,需要在新理论、新材料和新体系上进行突破。基于此,本文总结了 20 年来多电子反应材料概念 的形成、理论的发展、材料创制的历程。在"轻元素多电子反应"和"多离子效应"核心设计准则的指导下,具有上述 特征的电极材料与电池结构不断发展迭代,引领了高能量密度电池的发展方向。从阳离子氧化还原到阴阳离子 协同氧化还原,从嵌入式反应到合金化反应,从传统有机液态体系到电池固态化,本文梳理了典型的多电子反应 正负极材料的结构特性、体系创新和工程化前景,剖析了多电子反应电极材料的瓶颈问题,并分析了电池固态化 发展所面临的挑战。最后,对高能量密度电池的未来发展趋势和难点进行了归纳与展望。

关键词: 多电子反应; 高能量密度; 锂离子电池; 电池固态化; 电极材料体系