

## Solid-state / Semi-solid Li-ion Battery Innovation & Patent Review

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- <u>Cell Design Concepts</u>
- Pack Engineering
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- Applications
- <u>Electrolyte Film Deposition Processes</u>

Benchmarking & Product Launch Risk Factors -

- Cells with Liquid vs. Semi-Solid vs. Solid Electrolytes
- Inherent Safety Key Risk Factors
- Energy Density Positive & Negative Electrode Active Material Selections
- Power Density Ion Conductivity of Solid / Semi-solid Electrolytes
- Longevity Risk of Crack Formation & Chemical Instability
- <u>Cell Size</u>
- Raw Materials & Manufacturing Processes

#### Predictions

Assessment of Companies

- <u>Ampcera</u>
- BASF
- Blue Current
- Blue Solutions
- BrightVolt
- <u>BYD</u>
- <u>Contemporary Amperex Technology Ltd. (CATL)</u>
- <u>Corning</u>
- Enpower Greentech
- Factorial Energy
- FDK / Fujitsu
- Foxconn (Hon Hai Precision Industry) / SolidEdge Solution
- Ganfeng Lithium / Zhejiang Fengli / Zhejiang Funlithium
- <u>GM</u>
- Hydro Québec
- Idemitsu Kosan
- Ilika Technologies
- ION Storage Systems
- JX Nippon Mining and Metals / Eneos
- LG Energy Solution / LG Chemical
- Maxell
- Mitsui Mining and Smelting / Mitsui Kinzoku
- Murata Manufacturing
- NGK Insulators
- Ohara
- Panasonic
- Piersica
- PolyPlus
- ProLogium Technology

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	New Energy Technology / Yichung Qingtao Energy Technology	
•	QuantumScape	
•	Sakuú	
•	Samsung	
•	SES Holdings	
•	Soelect	
•	Solid Power	
•	SVOLT Energy Technology / Fengchao Energy Technology /	
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## About the Author

**Pirmin Ulmann** obtained a diploma in chemistry from ETH Zurich (Switzerland) in 2004 and a PhD from Northwestern University (USA) in 2009. Thereafter, he was a JSPS Foreign Fellow in an ERATO academic-industrial project at the University of Tokyo (Japan). From 2010 to 2016, while working at a major battery materials manufacturer in Switzerland, he was a co-inventor of 7 patent families related to lithium-ion batteries. He was also in charge of a collaboration with the Paul Scherrer Institute, evaluated outside technologies for corporate strategy, and made customer visits to battery manufacturers in East Asia, North America & Europe. He holds the credential Stanford Certified Project Manager (SCPM) and has co-authored scientific articles with more than 2,000 citations.



## Introduction

#### Focus of this Review

In this review, technical options are discussed that are being evaluated by key solid-state / semisolid lithium-ion battery companies towards the launch of commercial products for various applications, in particular electronics and EVs. The analysis is based on a unique AI-supported screening approach for the identification of patent filings with high prospective commercial relevance, which are compared with public statements (incl. at conferences).

Comprehension of solid-state / semi-solid Li-ion battery technology decision trees allows for the identification of promising product development directions that have not yet been explored.

Patent portfolios by key commercial players have been classified into 6 categories:

- Level 1) Electrolyte & electrode patents
- Level 2) Cell patents (chemistry & architecture)
- Level 3A) Pack / form factor / packaging patents
- Level 3B) Application patents
- Level 3C) Reliability patents (e.g. mitigation of short circuits / heat & gas formation)
- Level 3D) Manufacturing patents (electrolytes, electrodes, cells)

A patent portfolio that covers all of these categories generally reflects a substantial product development effort that addresses all aspects necessary for a successful launch.

For tailored patent searches, the AI model used for preparation of this review is available to users on <u>b-science.net</u>.

Table 2: (projected) market launches for solid-state / semi-solid battery EVs; color labels: midnight blue: oxide / phosphate-based electrolytes (may contain polymers); mocha: sulfide-based electrolytes (may contain halides, polymers); teal: halide-based electrolytes (without sulfur); plum: polymer-based electrolytes (predominant component)

Company	Country	Year	Possible Electrolyte / Negative Electrode Type / Other Info
		20 entries	

# Benchmarking & Product Launch Risk Factors – Cells with Liquid vs. Semi-Solid vs. Solid Electrolytes

A battery fails commercially if any performance & safety characteristic or costs do not match the requirements of the corresponding application. Outperformance in one dimension usually does not compensate for the biggest weakness.



#### Table 6: targeted energy density

<b>Companies</b> (approximate cell capacity)	Approximate volumetric / gravimetric energy density (for >1 Ah cells unless if mentioned otherwise)	Positive electrode	Negative electrode
20 entries			

### Technology Decision Trees

Table 7: ion conductivity of solid electrolytes (as identified in patent applications, in public statements, or by reference to an academic publication); color labels: midnight blue: oxide / phosphate-based electrolytes (may contain polymers, may contain minor amount of halide); mocha: sulfide-based electrolytes (may contain halides, polymers); teal: halide-based electrolytes (without sulfur, may contain oxygen); plum: polymer-based electrolytes (predominant component)

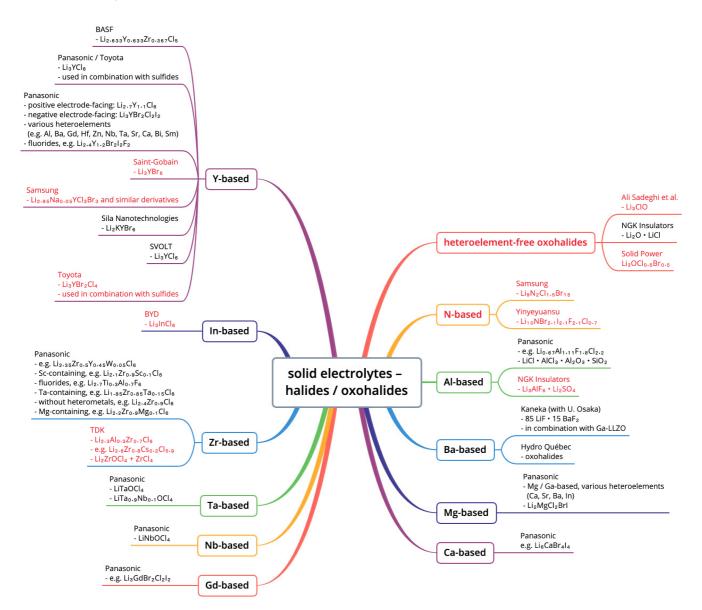
Companies	Possible electrolyte	Approximate ion conductivity at 25 °C unless if otherwise mentioned		
Škoda (VW)	'Li-glasses' based on complex mixture, e.g. $P_2O_5$ / LiCl / Li_2O /Al_2O_3 / B_2O_3 / Lil	5.5 × 10 <sup>-2</sup> S/cm		
Hydro Québec (licensed from Texas University / Porto University / Laboratório Nacional de Energia e Geologia)	'Li-glasses' based on dried LiOH, LiCl, Ba(OH)2	4 × 10 <sup>-2</sup> S/cm		
<u>Toyota</u>	Li <sub>9.54</sub> Si <sub>1.74</sub> P <sub>1.44</sub> S <sub>11.7</sub> Cl <sub>0.3</sub>	2.5 × 10 <sup>-2</sup> S/cm		
SVOLT	Li <sub>5.85</sub> P <sub>0.8</sub> Bi <sub>0.1</sub> Sn <sub>0.1</sub> S <sub>4.4</sub> O <sub>0.15</sub> Cl <sub>1.45</sub>	1.5 × 10 <sup>-2</sup> S/cm		
<u>Ampcera</u>	'sulfur-stuffed' argyrodite, $Li_8P_3S_{11+n}Cl$ or $Li_{8+2n}P_3S_{11+n}Cl$ , n > 0	>1.2 × 10 <sup>-2</sup> S/cm (public statement that presumably corresponds to the electrolyte on the left)		
Dynanonic	Supramolecular siloxane-PEO, coupled with click chemistry	1.2 × 10 <sup>-2</sup> S/cm		
118 additional entries				

#### Table 10: raw material / process aspects that could impact costs

Companies	Critical raw material or process aspects
19 e	entries



## Figure 12: technology decision tree – solid electrolytes – halides / oxohalides (in red: newly added branches as compared to prior review)



## Assessment of Companies

Author comments are displayed in maroon.

#### Contemporary Amperex Technology (CATL) – China

#### **Organization profile**

Contemporary Amperex Technology Limited (CATL, <u>https://www.catl.com/en/</u>) <u>is the world's</u> <u>largest Li-ion battery producer.</u> CATL was founded in 2011 in Ningde, China. In 2017, CATL has completed a <u>split from its parent company ATL/TDK</u>. <u>With BRUNP Recycling (subsidiary)</u>, CATL jointly develops positive electrode active materials.

**Unique capability:** 1) supramolecular ionic liquid / polymer / lithium salt electrolyte membranes with very favorable ionic conductivity (up to  $2.4 \times 10^{-3}$  S/cm) and high boiling point (>438 °C), along with corresponding cells with lithium metal negative electrodes; 2) sulfide electrolyte-based lithium metal cells based on  $\geq$ 5 complementary concepts to mitigate various failure modes.

**Leap of faith:** 1) the toxicity of triphenylene-containing electrolytes will be acceptable; 2) the risk of toxic hydrogen sulfide gas emissions when sulfide electrolytes are in contact with water or moisture will not be a showstopper during production, operation and / or recycling.

**Comment:** approaches 1) and 2) could finally enable the operation of lithium metal negative electrodes at room temperature and below with favorable fast charge / discharge characteristics (along with favorable energy density).

#### News reports & press releases

This information is included in the full version.

#### General patent portfolio characteristics

42 new patent families by CATL related to semi-solid or solid-state Li-ion batteries have been published since 2022 (level 1: 21, level 2: 24, level 3A: 8, level 3B: 1, level 3C: 11, level 3D: 9). Polymer / oligomer and sulfide electrolytes constitute key patenting focus areas (Figure CA-1).

Figure CA-1: Al-based classification of patent families by CATL published since 2022 related to solid electrolytes categories 1-5. Patents without direct relation to one category (e.g. because of solid-state cell packaging focus) were excluded.

CATEGORY	PATENT COUNT	PERCENTAGE	VISUAL
Category 5: Polymers/Oligomers	16	51.6%	
Category 4: Sulfides	10	32.3%	
Category 6: Halides/Oxyhalides	3	9.7%	•
Category 1: Oxides	1	3.2%	
Category 2: Oxide/Polymer Composites	1	3.2%	



#### Key Polymer Electrolyte Product Development Concepts

Figure CA-2: AI-based polymer electrolyte product development concept identification (CATL)



Figure CA-2 illustrates how CATL pursues a range of polymer / oligomer-related product development approaches, among which 2 concepts appear complementary, even though no patent has been identified yet that confirms their simultaneous use.

#### Concept 1: Supramolecular Ionic Liquids, WO 2022021231 A1 (EPO / Google)

#### **KEY FINDINGS**

 $2.4 \times 10^{-3}$  S/cm ionic conductivity in membranes based on supramolecular assembly of ionchannels, comparably low density, based on abundant precursors.

#### TECHNICAL DESCRIPTION

Benzophenanthrene-based supramolecular ionic liquids with  $\pi$ - $\pi$  stacking creating ordered ion transport pathways. Ether side chains (1-16 carbons, see Figure CA-3) balance molecular assembly with ion mobility. Synthesis via Williamson reaction and FeCl<sub>3</sub> cyclization, resulting in 6.5 × 10<sup>-3</sup> S/cm bulk conductivity at 25°C.

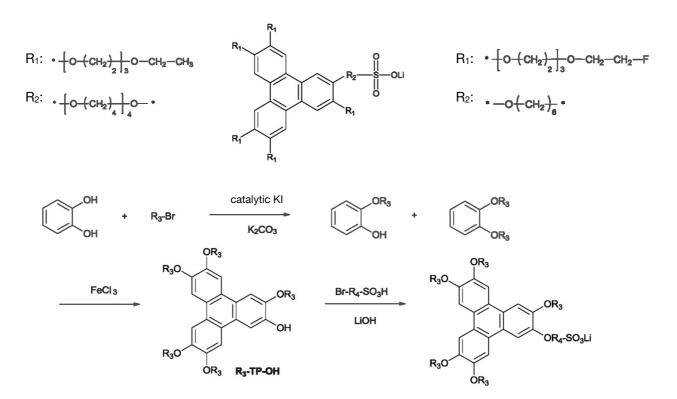
#### BACKGROUND INFORMATION

Patent WO 2022021231 A1 addresses the polymer ionic conductivity bottleneck. A selfassembly approach results in near-liquid conductivity ( $2.4 \times 10^{-3}$  S/cm in membrane), while offering safety advantages for which solid-state Li-ion batteries are known (triphenylene shown in Figure CA-3 exhibits a boiling point of 438°C). Manufacturing is based on organic chemistry and comparably abundant precursors.

#### ELECTRODE CONFIGURATION

Negative electrode: lithium metal | Positive electrode: NMC811.

Figure CA-3: top – two triphenylene derivatives with (optionally fluorinated) ethylene oxide / lithium sulfonate groups. The derivative on the left exhibits particularly favorable ion conductivity ( $6.5 \times 10^{-3}$  S/cm), while the derivative on the right exhibits particularly favorable cycling stability, bottom – synthesis procedure, OR<sub>3</sub> = R<sub>1</sub>, OR<sub>4</sub> = R<sub>2</sub> (CATL)



## The full version includes a discussion of polymer electrolyte product development concepts 2-5.

#### Potential Synergies Between Concepts

R&D concepts 1 (supramolecular ionic liquids) and 3 (gradient crosslinking systems) offer synergies in terms of overcoming the fundamental trade-offs in solid-state battery development. Concept 1 addresses the critical ionic conductivity bottleneck that has limited solid-state batteries, achieving near-liquid ionic transport through self-assembled molecular architectures. Concept 3 solves mechanical integrity and safety challenges by providing spatially optimized structural reinforcement. The combination could enable solid-state batteries that achieve both liquid-like ionic conductivity and superior structural integrity – previously mutually exclusive characteristics.

#### Possible Material / Cell / Process Characteristics (Projection Based on Public Information)

- **Electrolyte:** benzophenanthrene-based supramolecular ionic liquids with  $\pi$ - $\pi$  stacking architecture and ether side chains (1-16 carbons) that exhibit an ionic conductivity of 6.5 × 10<sup>-3</sup> S/cm at 25°C (WO 2022021231 A1, Example 22). When integrated with a polymer matrix (PEO/PVDF/LiTFSI at 10-80 : 100 : 5-40 by mass), the composite electrolyte exhibits 2.4 × 10<sup>-3</sup> S/cm conductivity. Gradient acrylate crosslinking systems (WO 2024243875 A1) might be incorporated into side chains to tune mechanical characteristics.
- **Negative electrode:** lithium metal on copper foil (WO 2022021231 A1).
- **Positive electrode:** NMC811 with conductive carbon (2 mass%) and PVDF binder (2 mass%) on aluminum current collector (WO 2022021231 A1).
- **Design:** prismatic stacked multilayer cells with favorable deformation resistance (WO 2024243875 A1).
- Process:
  - 1) Supramolecular ionic liquid synthesis via Williamson reaction and FeCl<sub>3</sub> cyclization (WO 2022021231 A1).
  - 2) Sequential or parallel (such as with multi-slurry feeder) injection gradient crosslinking film formation (WO 2024243875 A1).
  - 3) Hot pressing (1-20 MPa, 50-100°C) with spatially controlled crosslinking density.
  - 4) Vacuum annealing (60-80°C, 1-8 h).
  - 5) Electrode and electrolyte layer lamination through cold-pressing (250 MPa, 25 °C, 2 min) to obtain multi-layer cells.
  - 6) Prismatic cell encapsulation.

Supporting inventions listed in Figure CA-2 (bottom) might additionally be employed in this context.

#### Key Sulfide Electrolyte Product Development Concepts

The full version includes a discussion of sulfide electrolyte product development concepts 1-5, synergies and possible material / cell / process characteristics.



#### Highly Relevant Inventions Covered in Recent Triweekly Patent Updates

The full version includes a summary and discussion of 3 additional highly relevant all-solid / semi-solid Li-ion battery electrolyte patents that have been identified during our triweekly patent screening process.

#### **AI-based Patent Summaries**

The adjacent Excel file contains AI-based patent summaries for all patents mentioned in this chapter, classified in terms of electrolyte type (Figure CA-1) and <u>level</u> 1 (electrode / electrolyte patents) to <u>level</u> 3D (manufacturing patents).

### Al Patent Analysis Methodology

The patent information source for this review is the European Patent Office (EPO), <u>which covers</u> patent filings from more than 100 patent offices around the world. >3M patent documents are included in the b-science.net database that were published since 1980, which either contain the words 'battery' or 'batteries' in the title or abstract, or were assigned to one of the energy storage-related CPC (cooperative patent classification) or IPC (international patent classification) codes: H01M (batteries & fuel cells) or H01G (capacitors). An AI model was defined for commercially relevant Li-ion battery solid / semi-solid / gel electrolytes. Patent documents were grouped into patent families and scored with the AI model. An AI relevancy score cutoff value of 40 was applied (100: very relevant, 0: not relevant). For companies covered with a chapter, AI scores between 35 and 45 were checked manually and false-positives / false-negatives were corrected if necessary.

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