Technical Report

Directions for Next-Generation Innovative Batteries Centering on All-Solid-State Batteries

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Abstract:

Research and development is proceeding worldwide for next-generation innovative batteries such as all-solid-state batteries. However even at Toyota, which has continued automotive battery R&D for more than 15 years and has the largest number of patent applications in the world, there are still many issues facing the commercialization of all-solid-state batteries, and the roadmap to commercialization continues to be delayed. This means that the key to commercialization is how much overall value can be extracted from the all-solid-state batteries compared to the current liquid-type lithium-ion batteries.

1. Current Status and Future Prospects of Next-Generation Innovative Battery Research

The ongoing worldwide shift from conventional vehicles to EVs suggests that EV evolution will in large part be predicted by the battery which emerges as the successor to the lithium-ion battery (LIB). As shown in **Table 1**, the many possibilities for this role include all-solid-state batteries, lithium-air batteries, and lithium-sulfur batteries. Universities, research institutes, and private companies throughout the world are working on research and development in this area.

	Type		Positive electrode	Negative electrode	Characteristics	Issues
	Lithium-air		Oxygen (air)	Metal lithium	High capacity Safety	• Super-short lifetime • Separating oxygen from air
	Lithium-sulfur		Sulfur Sulfur compounds	Metal lithium	High capacity Safety	• Low conductivity • Short lifetime
High expectations	Lithium-ion	1st generation	Ternary (nickel-cobalt-manganese)	Graphite	Safety	• For automotive use, only sulfide-type solid electrolyte can be used. • Ionic conductivity is an issue with oxide types.
		Next-generation	High voltage type	Metal lithium	High capacity Safety	

Table 1 — Types and characteristics of next-generation innovative batteries (author's work)

As shown in **Figure 1**, all-solid-state batteries are composed of positive and negative electrodes combined with a solid electrolyte used instead of the liquid electrolyte and separator function used in LIBs. In principle, all-solid-state batteries greatly improve safety since conventional liquid electrolyte made of flammable solvents are replaced by a noncombustible solid electrolyte. Lithium ions are the only substance that moves through the solid electrolyte of a charging or discharging all-solid-state battery, which should enable longer battery life through resistance to the side reactions (undesirable reactions that accompany the required battery reactions) that occur in LIBs.

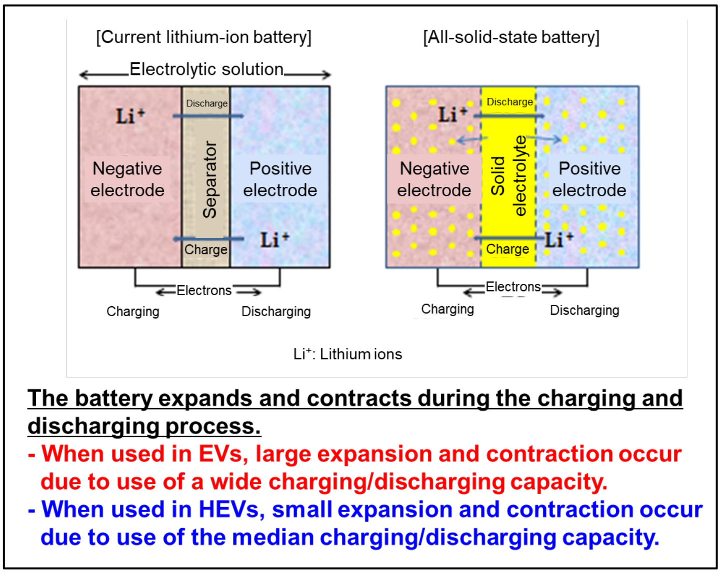

Figure 1 — Models of a liquid-type LIB and all-solid-state battery (author's work)

Solid electrolyte undergo component breakdown when exposed to high temperatures (60°C or above), and prevent sufficient battery output by increasing in electrical resistance when exposed to low temperatures (0°C or below). In contrast, the operating temperature range of solid electrolytes has a wide characteristic spanning low to high temperatures (-30°C to about 100°C). **Figure 2** shows the current status of development.

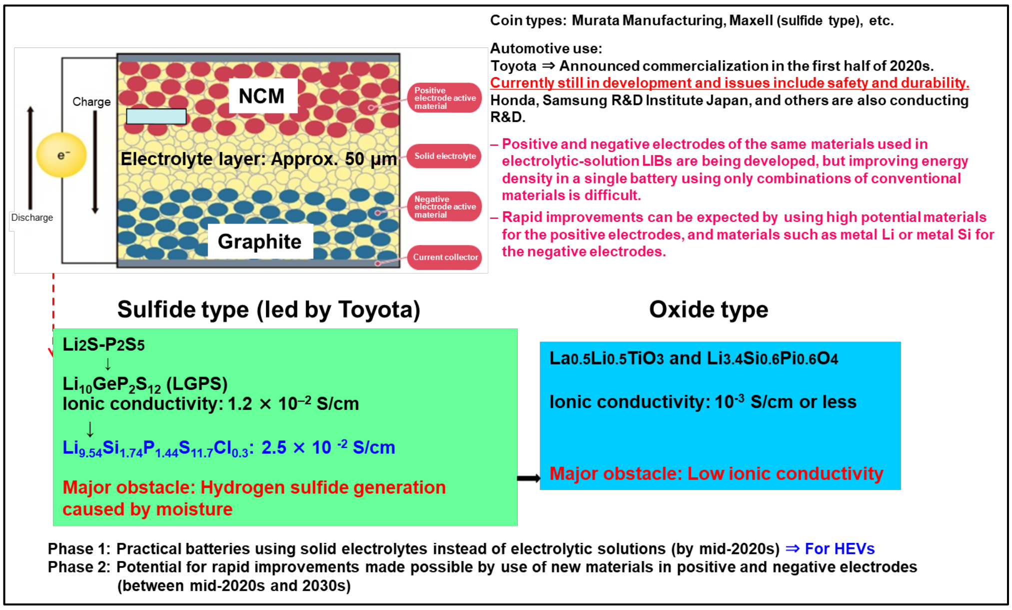

Figure 2 — Development status and future prospects of all-solid-state batteries (author's work)

2. Issues in Research and Development for All-Solid-State Batteries

While several sulfide-based solid electrolytes have been developed, they all have the problem of generating toxic hydrogen sulfide when they come into contact with water vapor-laden air. The establishment of rigorous production technologies in the electrode manufacturing process is essential.

Fig. 3 shows the all-solid-state battery production method. With compact batteries, a compression molding method can be used for solid powder electrolytes, and this field has advanced to the mass production and pilot production stage. However with the large-size batteries for automotive use where expectations are high, compression molding cannot be used due to the larger surface area of the electrode, and as a result a coating method is used. In this case, the positive electrode is formed as a single positive electrode unit as shown in the figure, and the negative electrode is created in the same way. The solid electrolyte layer that is used in place of a separator is also formed separately in sheet form, and the cell is created in three layers composed of the positive electrode, solid electrolyte layer, and negative electrode.

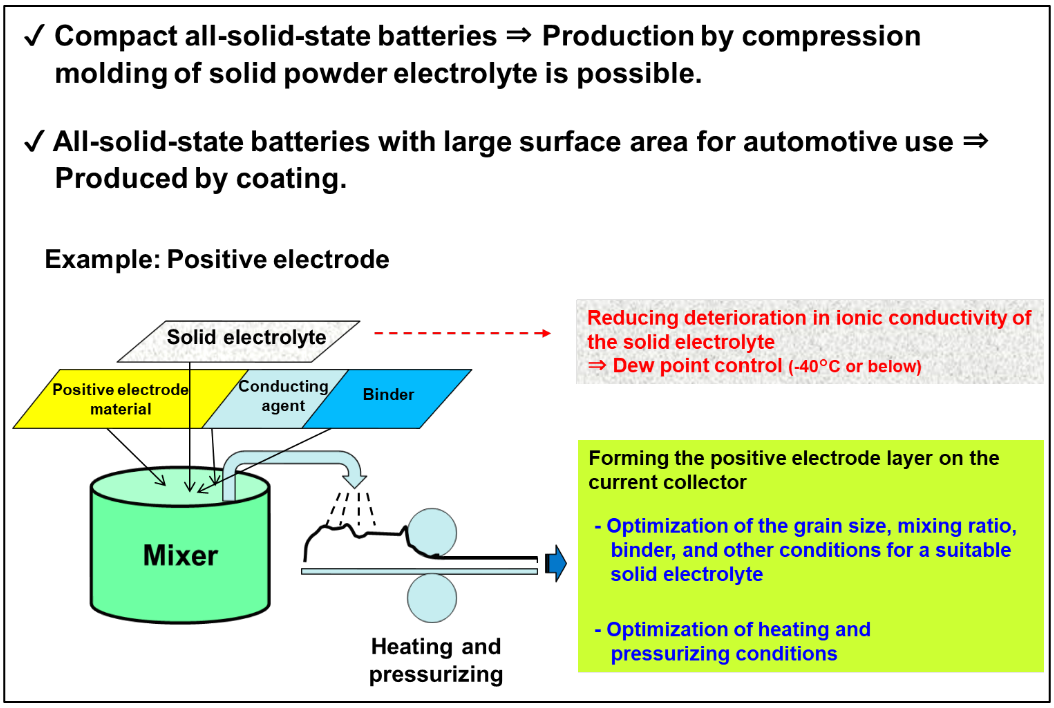

Figure 3 — Example of all-solid-state battery production method (author's work)

Because solid electrolytes are negatively affected by moisture, the production process requires dew point control that keeps the manufacturing equipment at a temperature of -40°C or below. Development of production technologies is also key because of the need to optimize the heating and pressurization conditions. In this way, while it is true that research into automotive all-solid-state batteries has made considerable progress, each new advance in research and development only seems to uncover new obstacles. **Figure 4** shows the overall issues that are faced.

At the same time as shown in **Table 2**, while the core material of the all-solid-state batteries where expectations are high is a solid electrolyte, the only companies that have announced a mass production target for automotive applications are major chemical material manufacturers Mitsui Mining & Smelting and Idemitsu Kosan. Although there are many chemical manufacturers in Japan, the lack of movement at the other manufacturers is likely due to focusing on risks over business opportunities because of patent problems and concerns about commercialization of all-solid-state batteries.

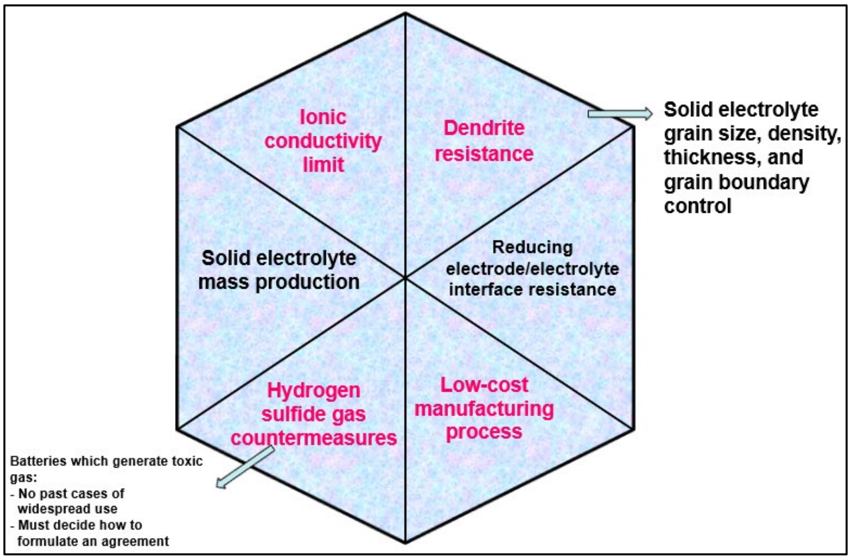

Figure 4 — Issues in development of all-solid-state batteries (author's work)

Table 2 — Trends in research and development of the key solid electrolyte materials (author's work)

Mitsui Mining & Smelting:

- Constructing a system for producing several tens of tons of argyrodite-type (Li-P-S-Cl) solid electrolytes annually at the Ageo Plant

- Target is commercialization for infrastructure and medical use through collaboration with Maxell.

- Currently conducting commercialization development for automotive use together with automobile manufacturers and battery manufacturers.

Idemitsu Kosan:

- Installed production equipment at the Ichihara Plant in Chiba Prefecture, and started production from autumn 2021.

- Has technologies for high-purity manufacture of lithium sulfide (Li_2S), which is the raw material for sulfide-type solid electrolytes.

Participation of other material manufacturers:

- Focusing on business risk more than business opportunities?

3. Expectations of All-Solid-State Batteries

Toyota was the first in the world to begin research into all-solid-state batteries for automotive use, and has been pursuing it for more than 15 years. In 2021, Toyota announced that there were large obstacles to the commercialization of all-solid-state batteries for EV use in terms of safety, the solid electrolyte material, and production technologies, and that it would postpone their use for EVs and would instead first commercialize such batteries for HEVs.

As shown in Figure 1, both liquid-type LIBs and all-solid-state batteries undergo volumetric expansion and contraction with repeated charging and discharging. In particular, for EV use, the amount of expansion and contraction is larger due to the wide range of battery capacity used. In the case of liquid-type batteries, a stable interface is maintained by forming a solid-liquid interface where the electrolyte contacts the surfaces of the positive electrode and negative electrode particles. On the other hand, because an all-solid-state battery forms a solid-solid interface between the positive and negative electrode particles and the solid electrolyte, if a slight gap is produced between the solids when expansion and contraction occur, a stable interface cannot be maintained. This increases the interface resistance and significantly reduces battery performance.

This issue is one major reason why Toyota has postponed commercialization of such batteries for EV use. In response, the company reversed direction and decided to prioritize batteries for HEVs. In the case of HEV batteries, because the median zone of the battery capacity is used, the battery undergoes little expansion and contraction, and there is no large effect on formation of the interface. Therefore this reversal of direction is reasonable.

As the global market is heading toward an EV shift, it is necessary to understand what will happen in the HEV market. This is shown in **Table 3**. The conclusion is that calculations show that the number of HEVs and PHEVs will be more than 20 million in 2035 alone. Conversely the global shift to EVs involves risk, and the lower part of Table 3 shows some specific examples.

Table 3 — Predicted developments for HEVs in the market (author's work)

2021:
EV: 4.6 million
(2.2× previous year,
China: 2.91 million,
NEV: 3.52 million,
Total: 26.27 million)
Germany: 340,000
USA: 490,000
Japan: 20,000
HEV: 3.10 million
(up 33% from
previous year)

2022:
EV: 7.89 million
PHEV: 2.74 million

⇒

2035:
Approx. 30 million vehicles across China ⇒ *HEV 15 million + PHEV 15 million × X%*

2021 HEV: Globally 3.1 million vehicles ⇒
Toyota 2030: EV 3.5 million vehicles + *HEV & PHEV 6 million vehicles*

Nissan 2030: *50% of global sales are EV + HEV (80% in EU).*

Honda: *Primarily HEVs even after 2030*

2035: China market and Toyota alone [*China 15 million vehicles + PHEV + 6 million vehicles*]

Obstacles to wider use of EVs
- Although EVs are a good match for France and Northern Europe, they are ineffective in countries which rely primarily on thermal power generation.
- However EV subsidies are increasing: France, USA, Japan. The scenarios for expanding use require decreasing subsidies and achieving an independent business by the time subsidies reach zero. Increasing subsidies is moving in the opposite direction.
- When driving at more than 200 km/hr on a German autobahn, the actual cruising distance is half of the mode. If the number of vehicles waiting to be charged at a rapid charging station is ten or more per station, what is the required time?
- Germany has reversed its EV shift policy and the EU has also agreed to permit engine vehicles that use synthetic fuels.
- In the same way that China approved HEV (the result of lobbying activities by automobile manufacturers), HEV would be overwhelmingly more effective in the India and Southeast Asia markets. It is necessary for Toyota, Honda, Nissan, and Suzuki to collaborate and promote the effects of HEV through lobbying activities targeting leaders and involving heads of states.

While increased safety may be the benefit of all-solid-state electrolytes, this benefit alone is not enough. The reason is that safety is already ensured by the Japanese LIBs currently being used in vehicles. The biggest benefit of all-solid-state batteries is the major improvement in energy density they could provide by using electrode materials that can't be used with LIB liquid electrolyte. These materials include metal lithium for negative electrodes, and high-voltage materials for positive electrodes. By greatly increasing the vehicle range per charge, this greater energy density could potentially enable a full-scale shift toward EVs.

4. Future Prospects for the Battery Industry

Automotive LIBs are an important economic security issue. In the book *The King of Batteries*[1)], I explained the history of batteries in Japan, which led the world with a fusion of science and technology. I also explained how the rapid rise of Korea and China has cast a shadow over Japan's relative competitiveness.

The Japanese government has been advocating a "science and technology-leading nation," a "battery-leading nation," and an "intellectual property-leading nation," but it appears that these were just words. If the battery industry rivalry between Japan, South Korea, and China continues to be fierce, the South Koreans and Chinese with their superior speed and investment power will establish themselves as a battery powerhouse, even in the area of automotive batteries, and the Japanese battery industry will be in danger of going into decline. We must prevent the battery industry from becoming like the semiconductor industry. In contrast to China's battery industry supported by an immense subsidy system, the South Korean major conglomerates' battery industry showing a presence with a sense of speed and scale, the EU's battery industry with an accelerated rise led by the government, and the U.S. battery industry under the Biden administration clearly stating its commitment to economic security, the time is approaching for the Japanese government to provide direct support to the Japanese battery industry, which is inferior in scale and investment power. I believe that the starting point of economic security is not only research on next-generation batteries, but also direct support that fosters the investment capacity of the battery industry to sustain it.

In November 2021, the Ministry of Economy, Trade and Industry (METI) launched the Public-Private Council for Storage Battery Industry Strategy Study. I was involved as an expert member, and provided a range of opinions during the period leading up to the final report on August 31, 2022. As shown in **Table 4**, the Japanese battery industry is working towards securing a final goal of 150 GWh of LIB production capacity in Japan and 450 GWh overseas. For this purpose, it is clear that the government and private sector must invest 3.2 trillion yen in the battery industry, consisting of 2 trillion yen in the battery business and 1.2 trillion yen in the components and materials business. As a preliminary stage, the government formulated a policy of direct support for investment in the Japanese battery industry in November 2021. This is a large step forward that has not happened in the past.

At the same time, I directly suggested to METI that national investment in all-solid-state batteries, which it had been leaning toward before, was not necessarily the correct course. The course change by METI to invest more in the current liquid-type LIB business than in all-solid-state batteries as of March 2022 was a positive result of this study.

Table 4 — Recommendations of the Public-Private Council for Storage Battery Industry Strategy Study and future actions (author's work)

- Japan, where national government support is completely absent compared to overseas competitors, is in a perilous position: In contrast to the opinion that if the oxygen saturation of the industry were measured, it would be 93%, METI recognized that it would be 89% (Sept. 3, 2021).
  ⇒ Nov. 4, 2021: National government announces direct support for investment in the battery industry.
  Aug. 2022: 3.2 trillion yen in the battery industry is needed (2 trillion yen in the battery business and 1.2 trillion yen in the components and materials business).
- Statement correcting the direction that had been focused on all-solid-state batteries (until Sept. 2021): Will the Japanese battery industry still be alive when all-solid-state batteries are commercialized?
  ⇒ METI reconsidered and changed focus to the current liquid-type LIB business in the 3rd Study report (March 28, 2022).
- Statement of continuation in the 6th Study report (August 31, 2022):
  (1) Expansion of investment and production capacity, and price competition, are unavoidable.
  (2) • Japan has lagged behind in the development of automotive LIBs. Education on United Nations regulation ECE R100-02 Part II is mandatory. Japan's own development standards are needed. Overseas EVs are entering Japan.
  • Japan standards for stationary storage batteries are needed. Batteries have entered Japan from overseas, and there have been cases of fires and recalls.
  (3) It is necessary to secure personnel by evolving the battery industry in a more appealing direction.
  April 25, 2023 "Public-Private Council for Storage Battery Industry Strategy Study" 03.pdf (meti.go.jp)
- From FY 2023, this is continuing as the "Storage Battery Industry Strategy Promotion Council." Storage batteries are "designated critical materials" and 331.6 billion yen is allocated to them in the FY 2022 supplementary budget.

A requirement for improving the resilience of the Japanese battery industry is making the current liquid-type LIB business a foundational cornerstone. Based on that, all-solid-state batteries must be approached from a long-term perspective, and it is necessary to objectively observe to determine whether there are prospects for their success.

Reference

1） Noboru Sato: *The King of Batteries*, Nikkei Business Publications, Inc., September, 2020