



Cite this: *Chem. Commun.*, 2021, 57, 12587

Configuring solid-state batteries to power electric vehicles: a deliberation on technology, chemistry and energy†

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Solid-state batteries (SSBs) have been widely regarded as a promising electrochemical energy storage technology to power electric vehicles (EVs) that raise battery safety and energy/power densities as kernel metrics to achieve high-safety, long-range and fast-charge operations. Governments around the world have set ambitious yet imperative goals on battery energy density; however, sluggish charge transport and challenging processing routes of SSBs raise doubts of whether they have the possibility to meet such targets. In this contribution, the battery development roadmap of China is set as the guideline to direct how material chemistries and processing parameters of SSBs need to be optimized to fulfill the requirements of battery energy density. Starting with the identification of bipolar cell configurations in SSBs, the blade cell dimension is then selected as an emerging cell format to clarify weight breakdown of a solid NCM523||Li cell. Quantifying energy densities of SSBs by varying key cell parameters reveals the importance of active material content, cathode layer thickness and solid-electrolyte–separator thickness, whereas the thicknesses of the lithium metal anode and bipolar current collector have mild impacts. Even in the pushing conditions (200 μm for the cathode layer and 20 μm for the solid electrolyte separator), high-nickel ternary (NCM) cathodes hardly meet the expectation of the battery development roadmap in terms of gravimetric energy density at a cell level, while lithium- and manganese-rich ternary (LM-NCM) and sulfur cathodes are feasible. In particular, solid lithium–sulfur batteries, which exhibit exciting gravimetric energy density yet inferior volumetric energy density, need to be well-positioned to adapt diverse application scenarios. This analysis unambiguously defines promising battery chemistries and establishes how key parameters of SSBs can be tailored to cooperatively follow the stringent targets of future battery development.

Received 10th August 2021,
Accepted 14th October 2021

DOI: 10.1039/d1cc04368d

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Introduction

The growing demand on long-range and safe electric vehicles (EVs), along with cheap and long-term stationary energy storage, has stimulated scientific and industrial efforts toward new battery concepts that fundamentally deviate from current lithium-ion (Li-

ion) batteries.^{1–5} These new battery concepts cover a range of material and technology diversities, such as Li metal batteries,^{6,7} solid-state batteries (SSBs),^{8–11} and emerging battery pack design,¹² by which the battery energy density (ED) and safety have the potential to surpass the state-of-the-art liquid Li-ion batteries (LIBs) that employ intercalation chemistry electrodes and aprotic electrolytes. In particular, the Li metal battery pairing of solid-state electrolytes (SSEs) has been roughly estimated to increase 70% and 40% volumetric energy density (v-ED) and gravimetric energy density (g-ED), respectively,¹³ while preventing battery thermal runaway and fire ignition, due to the rigid SSEs with intrinsic fire-proof features and dendrite-suppression properties.^{14,15} Besides, the replacement of liquid electrolytes by solid Li-ion conducting ceramics has additional benefits:^{13,16} (1) obviating the need of polymeric separators and thus reducing the material and processing costs; (2) achieving a unit cation transference number ($t_{\text{Li}^+} \approx 1$) and mitigating anion migration to curtail the build-up of a concentration gradient resistance in the internal cell;

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† Electronic supplementary information (ESI) available. See DOI: 10.1039/d1cc04368d

and (3) simplifying the packing process by alternatively stacking bipolar electrodes and SSEs.

The properties of solid electrolytes principally define the processing line and the selection of electrode materials.^{17,18} Among the various SSE families, ceramic oxides exhibit the exciting oxidative and reductive endurance (thermodynamically decomposing at the interfaces while kinetically limited), enabling the pairing of Li metal anodes and high-voltage cathodes, offering ample opportunities to increase ED and improve operational safety.^{19,20} However, pressing challenges, such as the brittleness induced resistive interfaces,^{21–24} high sintering temperature resulting in incompatibility with cathode materials,^{25–27} and heavier density dwarfing the overall ED,²⁸ impede their applications in well-performing SSBs. The sulfidic SSEs with favorable ductility allow a cold-pressing technique to create a dense electrolyte separator and an intimate contact among electrode ingredients, benefiting the ion conduction and processing routes.^{29,30} The remaining issues around sulfidic SSEs are to deal with humid sensitivity and interface stability against metallic Li anodes.^{31–33} Halide materials face a poor reduction stability with the Li anode, mild ionic conductivity, and a low oxidative potential, which largely narrow down the selection of cathode materials.^{34,35} Polymer electrolytes embrace sufficient ionic conductivity (normally higher than 60 °C) and mechanical flexibility, which favor implanting the current Li-ion battery production line to SSBs to some extent.^{8,36} However, the crystallization of polymer electrolytes under atmospheric conditions hinders the ion mobility.^{37,38} In addition, their capability to inhibit Li dendrite growth is still controversial due to low Young's modulus at room temperature and even fluidic properties at elevated temperature.^{39,40} Our group discovered lithium-rich anti-perovskites (LiRAP)⁴¹ that exhibited superionic conductivity ($>10^{-3}$ S cm⁻¹), materials sustainability (Li, Cl, O, F, etc.), compositional flexibility (large span of non-stoichiometric

composition to tailor ionic conductivity)^{35,42} and synthetic simplicity (<450 °C).⁴¹ In particular, the low melting point of LiRAP favors infiltration into dense electrodes at moderately elevated temperatures, which has the potential to reduce interfacial resistance at an industrial scale.⁴³ The current hindrance is how to improve the structure and surface stability under moist conditions.

In addition to the choice of proper materials, the identification of a suitable processing line plays a decisive role to realize energy-dense and large format SSBs.^{18,27} The roll-to-roll method widely used in the mature fabrication of LIBs is not applicable to SSBs due to wrinkling and peeling off.²⁸ Pressed pellets dominate the fundamental research in the search of electrolyte materials and probing the charge transport mechanisms, but they are ruled out in the large format cell at the industrial scale, since the thick SSE separator and electrode layers significantly penalize battery EDs.^{27,44,45} The lamination of sheet-type electrodes and electrolytes seems to be the only plausible configuration to scale up cell size.^{46–48} Chemical coating,⁴⁹ gas phase deposition,^{50–52} and penetration of SSE solution^{53,54} have been proposed to fabricate sheet-type cells. However, a large number of uncertainties, in terms of material selection, nanoscale interface/interphase engineering, and cell configuration, remain to be addressed. In particular, the unique bipolar cell concept that has the potential to increase cell voltage and simplify module and pack design requires rational design to correspond to the complexity of SSBs.⁵⁵

Due to the complexities and uncertainties of the material selection, cell configuration and production line, the question arises of how to configure cell-level parameters of SSBs to follow the ambitious yet imperative battery projects proposed by governments around the world, such as Europe "Battery 2030+,"⁵⁶ China "Energy-saving and New Energy Vehicle Technology Roadmap 2.0" (abbreviated as "Roadmap 2.0 v"),⁵⁷



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and America “National Blueprint for Lithium Batteries 2021–2030”.⁵⁸ A mature battery technology that can be massively applied in the market has to simultaneously overcome performance (energy density, power density, and shelf/working life-time) and technical (cost, safety, and reliability) barriers.⁵⁹ Although the SSB concept has a long history, it is still in the research phase, and is not likely to concurrently overcome all their barriers in the short term. It is, therefore, necessary to prioritize the critical metrics among them, and direct the future research toward real-world energy-dense SSBs. ED is a measure of the amount of energy that can be reversibly stored and released per unit, and majorly determines the driving range of electric vehicles.^{60,61} Identification of cell parameters and configurations based on current empirical and theoretical knowledge is helpful to corroborate the superiority of SSBs over the state-of-the-art LIBs from an ED aspect.

Herein, a bottom-up analysis is performed, with the Roadmap 2.0 v as a target, to clarify large-format cell parameters and material chemistries in SSBs that can fulfill the requirements of electric vehicles in future markets. In this calculation, a blade cell with a bipolar configuration is adopted as a prototype, which is anticipated to maximize EDs on a cell level and to simplify the stacking process in the scalable fabrication. This prototype used in the analysis can be regarded as a rather optimistic scenario, which sets the lower bound that cell parameters need to be reached in order to align with the Roadmap 2.0 v. The sulfidic SSE is selected in the projection of battery EDs due to attributes of its easy processing and low density, although its issues of moisture sensitivity and Li anode compatibility need to be addressed. High ionic conductivity, Li compatibility and facile processibility are regarded as default characteristics for the sulfidic SSE. The cost of materials and processing schemes are out of the scope of the current work, and cost would not be the stumbling blocks for the wide adoption of SSBs in electric vehicles. Once the great breakthroughs are made in performance metrics, it is highly expected that their costs will be significantly reduced in the massive market, benefitting from long learning curves, which have been witnessed in LIBs.⁶²

Targets of automotive batteries in Roadmap 2.0 v

The Ministry of Industry and Information Technology of China and the China Society of Automotive Engineers jointly issued the Roadmap 2.0 v in 2020, which set the targets for automotive battery performances within 15 years. Compared with the old version (issued in 2016) of the roadmap (Table S1, ESI†) that has unified requirements of cell- and pack-level energy densities without considering diverse scenarios (*e.g.*, electric bus, electric truck, and electric passenger car), Roadmap 2.0 v refines the application scenarios, classifying automotive batteries into energy-dense, power-dense, and energy- and power-dense types, with specific requirements for different sets of batteries (Table S2, ESI†). Fig. 1 abstracts the ED targets of batteries in the next 15 years. Roadmap 2.0 v raises the safety level as the prerequisites, while sets an ambitious goal in the ED metric. For example, the ED in the high-level batteries of energy-dense group targets at 350 W h kg^{-1} in 2025, and is more aggressive in 2030 (400 W h kg^{-1}) and 2035 (500 W h kg^{-1}), which can be hardly achieved with the conventional LIBs with a graphite (Gr) anode.⁶³ This dilemma necessities a bold change in the key components of batteries instead of mild modification of matured active materials. Combining Li metal anodes and SSEs may pave a way to meet such stringent targets while maintaining the desired safety level, since SSBs take the advantages of intrinsically safe electrolytes and high-capacity metallic Li, which may be the game-changing power sources for long-range, high-safety and durable EVs. The following section performs a cell-level analysis to illustrate how key parameters influence the ED of SSBs with an emerging blade cell format.

Configuration and weight breakdown of a solid-state cell

Identification of the SSB configuration that is applicable in the large format cell is the first step to define the layout of

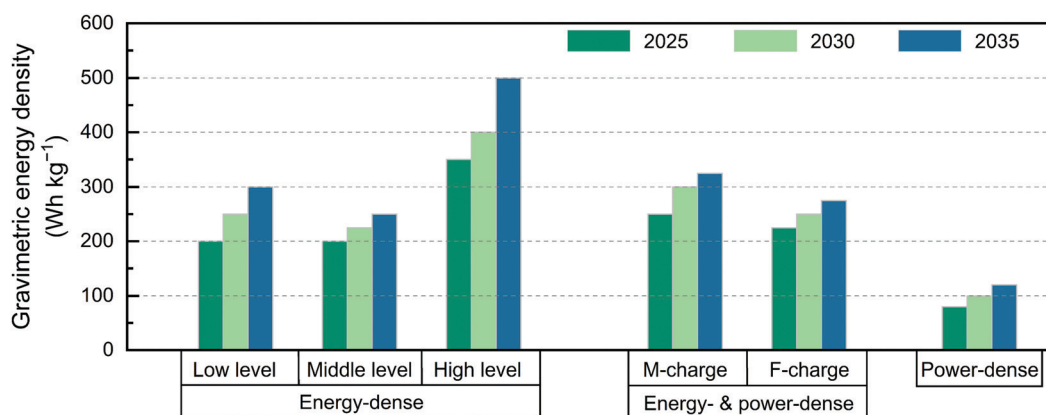


Fig. 1 Targets of the g-ED in Roadmap 2.0 v. Roadmap 2.0 v refines application scenarios, clustered into energy-dense, energy- and power-dense, and power-dense batteries. Each type is further divided into sub-types to fulfill diverse requirements. M-charge and F-charge represent medium charge and fast charge, respectively.

electrodes and electrolytes. In the conventional cell design, cathode and anode layers are double-coated on aluminum (Al) and copper (Cu) foils, respectively, and then stack together through flat winding or z-folding with separators inserted between them (Fig. 2a).⁶⁴ The welding joints are needed for the connection between tabs and current collectors (CCs), in which additional space has to be reserved.⁶⁵ The overall cell voltage equals the individual cell unit in such stacking. In the bipolar design, the anode layer and cathode layer share the same CC, as indicated in the inset of Fig. 2b.⁶⁶ The bipolar layer and the electrolyte layer are alternately stacked together. The current flows from the outermost layers, obviating the need of tabs and thus saving space for additional gains in ED.¹⁷ Another advantage of bipolar design allows a serial stacking of cells, leading to the increase of the overall cell voltage by the sum of individual cell voltages to achieve considerably high cell-level voltage, which probably facilitates the design of a battery module or a pack.

A direct comparison of LIBs and SSBs is conducted on the basis of a blade cell size, which represents emerging cell design for high ED (Fig. 2c). The blade cell is featured by the large ratio of length to height, with depth and height comparable to the popular cell dimension.¹² It is of notice that the real-world blade cell combines both serial and parallel connections to balance cell voltage, ED, and potential safety issues.⁶⁷ The current analysis ignores such complexities, and exploits direct stacking of sheet-type electrodes and electrolytes, as indicated in Fig. 2a and b. An Al-plastic film is assumed as the housing material to pack cell components due to the light-weight feature. The number of cell units is adapted to the depth of the blade cell.

In the initial tentative analysis, liquid LIBs with a NCM523||Gr chemistry adopt typical electrode compositions of commercial cells,⁶³ and detailed values are listed in Tables S3 and S4 (ESI†). The thicknesses of the cathode layer and separator are 100 and 16 μm , respectively, and the anode thickness can be calculated according to the cathode areal capacity and capacity ratio of negative to positive electrodes (N/P). In the SSBs, the liquid electrolyte in the cathode is substituted with the sulfidic SSE. The thickness of an SSE separator is assumed to be 100 μm , significantly higher than the polymer separator, yet is currently acceptable considering the benchmark of SSBs recently documented by Richter and Janek.⁶⁸ Reducing the thickness of the SSE separator is kernel to enhance ED of SSBs, which will be tailored later.

Based on the above assumption, the weight breakdown of liquid NCM523||Gr and solid NCM523||Li cells are presented in Fig. 3c. A noticeable change is the electrolyte portion in the cells, in which the solid and liquid electrolytes account for 36.7% and 14.7% in SSBs and LIBs, respectively. This significant variation is attributed to the heavy density of the solid electrolyte and the thick SSE separator. Despite the greater N/P (3.0) in SSBs than LIBs (1.05), the high-capacity Li metal anode (3861 mA h g^{-1} for Li vs. 360 mA h g^{-1} for Gr) only occupies 3.7%, much lower than Gr in LIBs (23.3%). It is noticed that the solid-state NCM523||Li cell cannot surpass the liquid NCM523||Gr in both g-ED (263 W h kg^{-1} vs. 288 W h kg^{-1}) and v-ED (609 W h L^{-1} vs. 733 W h L^{-1}), and also is far behind the requirements of high-level batteries in the Roadmap 2.0 v. The inferior ED of SSBs with presented parameters drives

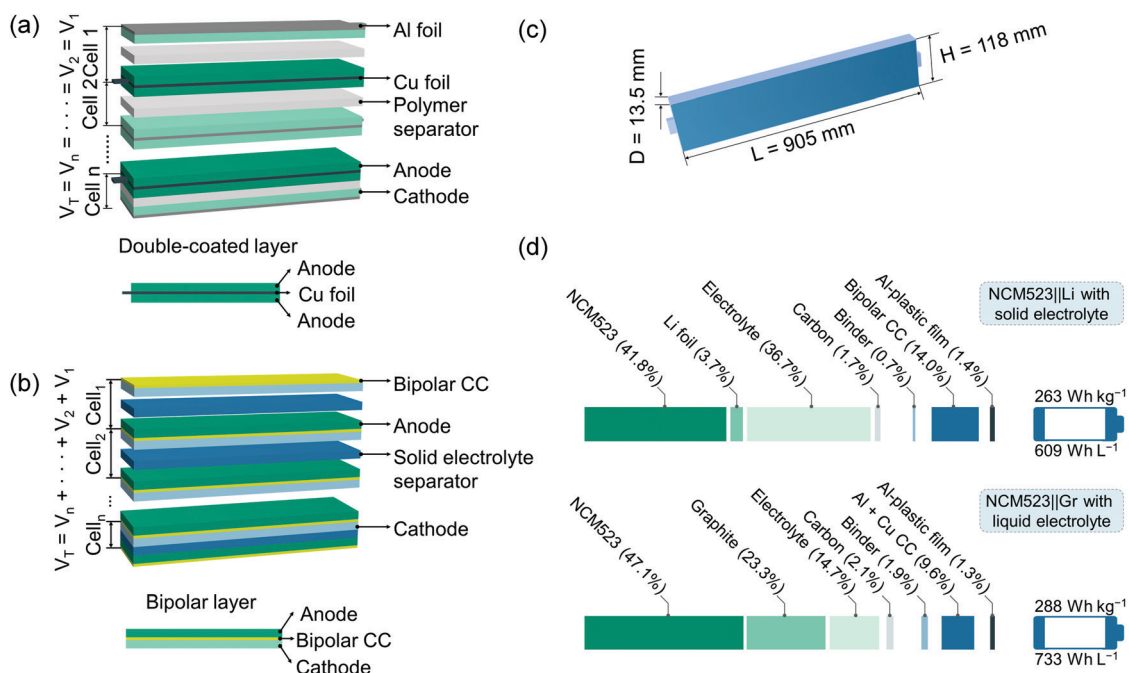


Fig. 2 Schematic illustrations of (a) parallel stacking for conventional LIBs and (b) bipolar stacking for SSBs. (c) Dimensions of a blade cell to estimate EDs of LIBs and SSBs. (d) Weight breakdown of a solid NCM523||Li cell and a liquid NCM523||Gr cell, and the resulting g-ED and v-ED.

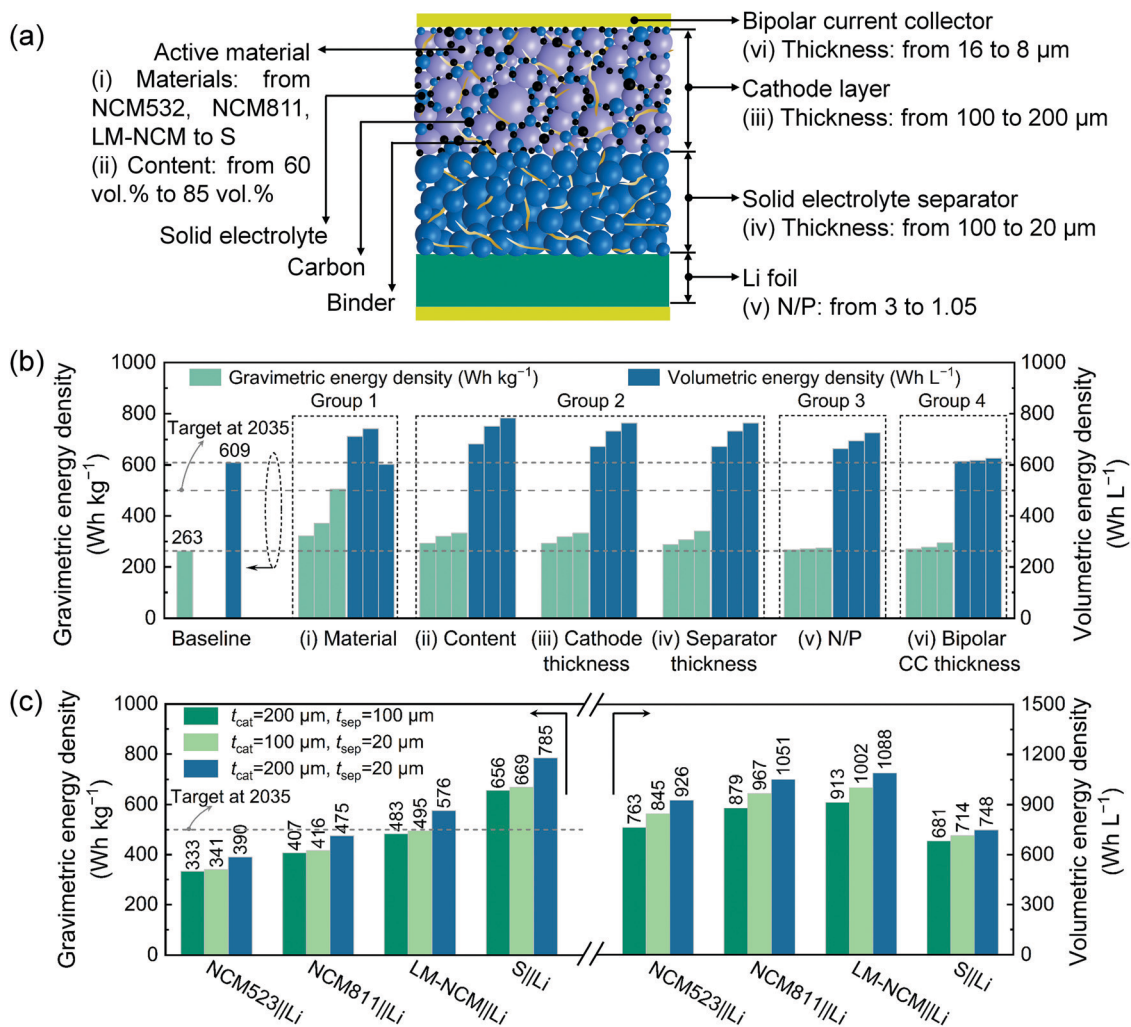


Fig. 3 (a) Schematic cross-section of a unit solid-state cell configuration and key parameters that are intended to be changed for high-energy-density SSBs. (b) g-ED and v-ED of SSBs as a function of key parameters. (c) Updating g-ED and v-ED by varying the thicknesses of the cathode layer (t_{cat}) and the solid electrolyte separator (t_{sep}).

intense effort to tailor electrode/electrolyte composition and thickness for higher EDs.

Material chemistries and processing parameters of the solid-state cell

To quantify the effects of individual parameters on the overall ED of a blade cell, the NCM523||Li cell with the parameters in Fig. 2d is selected as a baseline. Parameters that are intended to be optimized are schematically presented in Fig. 3a. Discovering and developing novel battery materials that can store more energy per unit without penalizing other properties (such as safety, structure stability and electrolyte compatibility) have dominated battery fields, and a large spectrum of material families have caught wide attention. Some of them have been successfully commercialized (NCM with moderate nickel content), while others (NCM with high nickel content, and sulfur) are underway. In this work, the active materials including matured

NCM 532, maturing NCM811, and emerging lithium- and manganese-rich nickel-cobalt-manganese oxides (LM-NCM) and sulfur (S) are selected to assess their potential in ED.

A high content of active materials in electrodes without affecting the reaction kinetics has been pursued, as it essentially dictates the amount of energy that can be stored in the limited electrode space, as well as reduce other cell components, for example, SSEs. The currently commercialized Li-ion batteries include ~60 vol% active materials, with 30–35 vol% left for liquid electrolytes.⁶³ The active materials content in this analysis ranges from 60 vol% to an aggressive value of 85 vol%, which has been occasionally reported,^{69,70} reflecting a progressive improvement in the next 15 years.

The thick cathode layer promises greater areal capacity and higher active material ratio of a unit cell, which jointly contribute to the ED. However, a thick cathode layer requires fully considering lithium/electron transport properties and processibility, since a thick cathode layer would lead to sluggish electrode reaction kinetics and crack. A thickness of 100 μm

is normal for a cathode layer for automotive LIBs.⁷¹ Ito⁷² and Chen⁷³ have recorded the cathode layers with 250 μm and 300 μm , respectively. The cathode layer with a 200 μm thickness seems to be conceivable at the pushing condition.

Flexible microporous polyolefin-based separators with $<20\ \mu\text{m}$ have been widely used in automotive Li-ion batteries, which is far thinner than the SSE separator used in SSBs, which is in the level of 100–600 μm .⁶⁸ The thick SSE separator significantly impedes effective Li transport between the cathode and the anode, and undermines the ED. Reduction of the thickness of the SSE separator is a demanding task for both academic and industrial communities due to the high cost of solid electrolyte materials and critical determination of EDs. Processing of a thin electrolyte separator on the level of the polymer separator of LIBs is a consensus to realize high-ED SSBs, and 20 μm is assumed as the most optimistic condition in the calculation. It is of note that a too thin solid electrolyte separator (like 20 μm) would affect the safety of batteries in the practical operation of SSBs, such as ease of crack and lithium dendrite penetration. Battery safety is the prioritized consideration in the final practical applications, which underlines the urgency of high quality of the solid electrolyte separator. Therefore, the thin solid electrolyte separator with additional benefits of elastic, homogeneous, robust, and self-healing properties are required to ensure sufficient safety.

A limited Li supply is not only beneficial for the battery ED, but also is a key indicator for the side reactions that cause short cycle life and battery failure.⁷⁴ According to Li's calculation,⁷⁵ the Li metal anode will not be competitive to the graphite anode if the nonlithium volume fraction exceeds 70%, which hints $\sim 233\%$ excess of Li, that is $N/P \approx 3.33$, will be meaningless in terms of battery ED. Herein, the N/P of SSBs is assumed to reach the level of LIBs ($N/P = 1.05$) as the best-case condition to fully tap the potential of SSBs.

The bipolar CC plays a key role in the support of cathode and anode layers, and should be resistive against both oxidation and reduction within the working voltage window during battery cycling. The thickness and property of bipolar CC do not attract enough attention, probably due to undeveloped test protocols in SSBs. Most of the SSB test employs a pellet in a special designed cell. If SSBs enter into pre-phase commercialization, bipolar CC should be well developed to fulfill multiple requirements. An Al/Cu clad foil with a thickness of 10/10 μm has been demonstrated in the bipolar electrode fabrication.⁷⁶ Assuming a well-developed gas phase deposition technique is applied to deposit a nanometer Cu layer on an 8 μm Al foil, the thickness of such advanced bipolar CC can be possibly reduced to $\sim 8\ \mu\text{m}$. The detailed variation of key parameters is listed in Table S5 (ESI[†]).

The above assumptions lead to the charting of g-ED/v-ED in Fig. 3b, which can be clustered into four groups. Varying active materials constitutes Group 1, in which significant changes in g-ED and v-ED can be identified. The maturing NCM 811||Li cell gives rise to improved g-ED (322 W h kg^{-1}) and v-ED (711 W h L^{-1}), and can be further enhanced by employment of an emerging LM-NCM cathode, exhibiting 372 W h kg^{-1} and

741 W h L^{-1} , respectively. The solid-state Li-S batteries deliver an inferior v-ED of 603 W h L^{-1} , which is not as impressive as their g-ED (505 W h kg^{-1}). This is essentially attributed to a much lower density of elemental S. Group 2 consists of three parameters: active material content, cathode thickness, and solid-electrolyte-separator thickness, which exhibit gradual boosts of g-ED and v-ED. Group 3 ($N/P = 3\text{--}1.05$) shows a mild improvement in g-ED (from 263 W h kg^{-1} to 274 W h kg^{-1}) and observable enhancement in v-ED (from 609 W h kg^{-1} to 724 W h L^{-1}), respectively, as the Li anode only accounts for $\sim 3.7\ \text{wt}\%$ in the cell, while takes up to $\sim 24\ \text{vol}\%$ in the entire blade cell. The thickness of bipolar CC, belonging to Group 4, exerts a limited effect on both g-ED (from 263 W h kg^{-1} to 294 W h kg^{-1}) and v-ED (from 609 W h kg^{-1} to 627 W h L^{-1}). The limited ED changes upon the variation of the bipolar CC thickness implies that engineering surface properties and mechanical strength to render better electrochemical performances seems to be more meaningful than devoting costly efforts to reduce the bipolar CC thickness for higher ED. Variables dictating g-ED and v-ED are provided in Table S6 (ESI[†]).

The current calculation indicates an insufficiency of solely varying a single parameter to follow the aggressive targets of the Roadmap 2.0 v, which advocates optimization of multiple aspects of SSBs to boost ED. Screening key parameters of SSBs reveals that Group 1 and Group 2 exert more pronounced impacts on ED than Group 3 and Group 4. While the active material and liquid electrolyte in commercial automotive LIBs accounts for $\sim 65\ \text{vol}\%$ and $\sim 30\ \text{vol}\%$ in the cathode layer, respectively, enhancing active materials in SSBs from 60 vol% to 85 vol% may bring a mountain of challenges in materials chemistries involving SSE, binder and conductive agent. Accordingly, two parameters, that include the thicknesses of the cathode layer (t_{cat}) and the SSE separator (t_{sep}), are herein taken into consideration to estimate EDs of SSBs with various active materials (NCM 811, LM-NCM and S) through concurrently pushing them into stringent conditions ($t_{\text{cat}} = 200\ \mu\text{m}$ and $t_{\text{sep}} = 20\ \mu\text{m}$).

g-EN and v-ED with different sets of conditions are charted in Fig. 3c. Even in the demanding cell conditions, SSBs with NCM523 and NCM 811 cathodes hardly exceed 500 W h kg^{-1} at the cell level. Therefore, the SSB with the high-nickel NCM cathode may be ruled out in the development of high-level type batteries, which defines an energy density of 500 W h kg^{-1} in Roadmap 2.0 v (Fig. 1). However, this calculation is based on the 60 vol% content of NCM811 in the cathode (in the comparable level of current liquid NCM811||Li cell). If the content of NCM811 in the cathode is more than 80 vol% (may be very challenging in case the thickness is 200 μm), the g-ED would be more than 550 W h kg^{-1} , which will fulfill the requirements of Roadmap 2.0. In contrast, solid LM-NCM||Li and S||Li cells are more attractive, with some possibilities to align with the ED targets of Roadmap 2.0 v. In particular, the solid S||Li cell exhibits considerably high g-ED, reaching to more than 780 W h kg^{-1} . This exciting value is based on the breakthroughs in charge transport/transfer in the bulk/interface, as well as excellent processing techniques. However, it is worth

noting that the v-ED of solid S||Li cells, which is more weighted in the industrial community, will never outperform the NCM family in this calculation. Considering impressive g-ED, inferior v-ED and the low cost of sulfur, solid Li-S batteries need to be well-positioned to adapt the diversified applications.

The practical application of SSBs in electric vehicles is an open question, as it is related to materials, chemistries, mechanics, electronics, *etc.* which deviates from current Li-ion batteries with liquid electrolytes. From energy chemistry perspectives, the SSBs can exert an energy-density superiority over liquid Li-ion batteries in the case of the lithium metal anode to be used. However, bringing the proof-of-concept high-energy SSBs to a reality requires overcoming a mountain of challenges, such as affordable solid electrolyte materials, thin yet robust electrolyte separator, acceptable electrode reaction kinetics, and cost-effective processing routines for cell components, any failure of which may retard the application of SSBs in electric vehicles. In our opinion, the booming development of materials, deepening understanding of working principles from the atom to the system, and long-term research funding would progressively address such currently considered big challenges, and lead to a bright future of SSBs, as we have witnessed the critical leap of battery ED from $\sim 90 \text{ W h kg}^{-1}$ (the first commercialized cell in 1991) to the current value of 270 W h kg^{-1} , which was considered to be impossible 30 years ago.

Conclusion and outlook

Ambitious yet imperative targets of battery technologies that are expected to promote penetration of electric vehicles into the vehicle market motivate a bold change of key battery components to rationally improve the energy density and safety of batteries. Solid-state batteries (SSBs) with pairing of intrinsically safe solid-state electrolytes (SSEs) and a high-capacity Li metal anode represent research trends under this context. Prioritizing material chemistry and evaluating key parameters of SSBs from a practical perspective are the initial steps to position SSBs well in the roadmap of battery development. This work performs chemistry, technology and energy analysis of SSBs based on the emerging blade cell dimension, and decouples decisive parameters from inappreciable ones, the former of which exert more impacts on the battery energy density. The calculation reveals that a significant increase of energy density to the level of Roadmap 2.0 v requires pushing key parameters to stringent conditions, such as $200 \mu\text{m}$ for the cathode layer thickness and $20 \mu\text{m}$ for the solid-electrolyte-separator thickness. Even in such conditions, the family of high-nickel NCM cathodes is ruled out in the development of high-level batteries due to insufficient gravimetric energy density. In contrast, lithium- and manganese-rich ternary and sulfur cathodes have the possibility to surpass the requirements. However, the low volumetric energy density of solid Li-S batteries may weaken the widespread applications in long-range electric vehicles to some extent. The current work hopes to figure out critical material chemistries and real-world cell parameters to

align with the harsh requirements of future SSB in the energy density metric.

Author contributions

Long Kong: conceptualization, methodology, formal analysis, writing – original draft, writing – review and editing and funding acquisition. Liping Wang and Jinlong Zhu: investigation and funding acquisition. Juncao Bian, Wei Xia, Ruo Zhao and Haibin Lin: investigation and writing – original draft. Yusheng Zhao: writing – review and editing and funding acquisition.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

This work was supported by the Shenzhen Key Laboratory of Solid State Batteries (ZDSYS20180208184346531), Shenzhen Science and Technology Program (No. KQTD20200820113047086), Guangdong-Hong Kong-Macao Joint Laboratory (2019B121205001), and Guangdong Provincial Key Laboratory of Energy Materials for Electric Power (2018B030322001).

Notes and references

- 1 L. Trahey, F. R. Brushett, N. P. Balsara, G. Ceder, L. Cheng, Y.-M. Chiang, N. T. Hahn, B. J. Ingram, S. D. Minteer, J. S. Moore, K. T. Mueller, L. F. Nazar, K. A. Persson, D. J. Siegel, K. Xu, K. R. Zavadil, V. Srinivasan and G. W. Crabtree, *Proc. Natl. Acad. Sci. U. S. A.*, 2020, **117**, 12550–12557.
- 2 T. Placke, R. Kloepsch, S. Dühnen and M. Winter, *J. Solid State Electrochem.*, 2017, **21**, 1939–1964.
- 3 L. Kong, C. Yan, J. Q. Huang, M. Q. Zhao, M. M. Titirici, R. Xiang and Q. Zhang, *Energy Environ. Mater.*, 2018, **1**, 100–112.
- 4 E. Faegh, B. Ng, D. Hayman and W. E. Mustain, *Nat. Energy*, 2021, **6**, 21–29.
- 5 L. Xu, W. Liu, Y. B. Hu and L. L. Luo, *Chem. Commun.*, 2020, **56**, 13301–13312.
- 6 X.-B. Cheng, R. Zhang, C.-Z. Zhao and Q. Zhang, *Chem. Rev.*, 2017, **117**, 10403–10473.
- 7 D. H. Kim, M. H. Lee, B. G. Kim, S. M. Lee and J. H. Choi, *Chem. Commun.*, 2020, **56**, 13040–13043.
- 8 L.-Z. Fan, H. He and C.-W. Nan, *Nat. Rev. Mater.*, 2021, 1–17.
- 9 M. Pasta, D. Armstrong, Z. L. Brown, J. Bu, M. R. Castell, P. Chen, A. Cocks, S. A. Corr, E. J. Cussen and E. Darnbrough, *J. Phys. Energy*, 2020, **2**, 032008.
- 10 X.-B. Cheng, C.-Z. Zhao, Y.-X. Yao, H. Liu and Q. Zhang, *Chem*, 2019, **5**, 74–96.
- 11 J. Koettgen, C. J. Bartel and G. Ceder, *Chem. Commun.*, 2020, **56**, 1952–1955.
- 12 X.-G. Yang, T. Liu and C.-Y. Wang, *Nat. Energy*, 2021, **6**, 176–185.
- 13 J. Janek and W. G. Zeier, *Nat. Energy*, 2016, **1**, 16141.
- 14 M. Jia, N. Zhao, H. Huo and X. Guo, *Electrochem. Energy Rev.*, 2020, 1–34.
- 15 A. Manthiram, X. Yu and S. Wang, *Nat. Rev. Mater.*, 2017, **2**, 1–16.
- 16 Q. Zhao, S. Stalin, C.-Z. Zhao and L. A. Archer, *Nat. Rev. Mater.*, 2020, **5**, 229–252.
- 17 J. Schnell, T. Günther, T. Knoche, C. Vieider, L. Köhler, A. Just, M. Keller, S. Passerini and G. Reinhart, *J. Power Sources*, 2018, **382**, 160–175.
- 18 C. Singer, J. Schnell and G. Reinhart, *Energy Technol.*, 2021, **9**, 2000665.

- 19 J. Schnell, F. Tietz, C. Singer, A. Hofer, N. Billot and G. Reinhart, *Energy Environ. Sci.*, 2019, **12**, 1818–1833.
- 20 Y. Lu, C.-Z. Zhao, H. Yuan, X.-B. Cheng, J.-Q. Huang and Q. Zhang, *Adv. Funct. Mater.*, 2021, **31**, 2009925.
- 21 Z. Gao, H. Sun, L. Fu, F. Ye, Y. Zhang, W. Luo and Y. Huang, *Adv. Mater.*, 2018, **30**, 1705702.
- 22 A. Banerjee, X. Wang, C. Fang, E. A. Wu and Y. S. Meng, *Chem. Rev.*, 2020, **120**, 6878–6933.
- 23 X. Ke, Y. Wang, G. Ren and C. Yuan, *Energy Storage Mater.*, 2020, **26**, 313–324.
- 24 J. Han and J. C. Kim, *Chem. Commun.*, 2020, **56**, 15197–15200.
- 25 C. Wang, W. Ping, Q. Bai, H. Cui, R. Hensleigh, R. Wang, A. H. Brozena, Z. Xu, J. Dai and Y. Pei, *Science*, 2020, **368**, 521–526.
- 26 T. Famprikis, P. Canepa, J. A. Dawson, M. S. Islam and C. Masquelier, *Nat. Mater.*, 2019, **18**, 1278–1291.
- 27 M. Balaish, J. C. Gonzalez-Rosillo, K. J. Kim, Y. Zhu, Z. D. Hood and J. L. Rupp, *Nat. Energy*, 2021, **6**, 227–239.
- 28 L. Liu, J. Xu, S. Wang, F. Wu, H. Li and L. Chen, *eTransp.*, 2019, **1**, 100010.
- 29 K. H. Park, Q. Bai, D. H. Kim, D. Y. Oh, Y. Zhu, Y. Mo and Y. S. Jung, *Adv. Energy Mater.*, 2018, **8**, 1800035.
- 30 S. Chen, D. Xie, G. Liu, J. P. Mwizerwa, Q. Zhang, Y. Zhao, X. Xu and X. Yao, *Energy Storage Mater.*, 2018, **14**, 58–74.
- 31 K. J. Kim, M. Balaish, M. Wadaguchi, L. Kong and J. L. Rupp, *Adv. Energy Mater.*, 2021, **11**, 2002689.
- 32 Y. Li, D. Zhang, X. Xu, Z. Wang, Z. Liu, J. Shen, J. Liu and M. Zhu, *J. Energy Chem.*, 2021, **60**, 32–60.
- 33 S. S. Shin, J. S. Kim, S. Choi, H. I. Ji, K. J. Yoon, J. H. Lee, K. Y. Chung and H. Kim, *Chem. Commun.*, 2021, **57**, 3453–3456.
- 34 X. Li, J. Liang, X. Yang, K. R. Adair, C. Wang, F. Zhao and X. Sun, *Energy Environ. Sci.*, 2020, **13**, 1429–1461.
- 35 L. Yin, H. Yuan, L. Kong, Z. Lu and Y. Zhao, *Chem. Commun.*, 2020, **56**, 1251–1254.
- 36 L. Li, Y. Deng and G. Chen, *J. Energy Chem.*, 2020, **50**, 154–177.
- 37 M. Zhu, J. Wu, Y. Wang, M. Song, L. Long, S. H. Siyal, X. Yang and G. Sui, *J. Energy Chem.*, 2019, **37**, 126–142.
- 38 S. B. Aziz, T. J. Woo, M. Kadir and H. M. Ahmed, *J. Sci.: Adv. Mater. Devices*, 2018, **3**, 1–17.
- 39 P. Yao, H. Yu, Z. Ding, Y. Liu, J. Lu, M. Lavorgna, J. Wu and X. Liu, *Front. Chem.*, 2019, **7**, 522.
- 40 P. Barai, K. Higa and V. Srinivasan, *Phys. Chem. Chem. Phys.*, 2017, **19**, 20493–20505.
- 41 Y. Zhao and L. L. Daemen, *J. Am. Chem. Soc.*, 2012, **134**, 15042–15047.
- 42 J. A. Dawson, H. Chen and M. S. Islam, *J. Phys. Chem. C*, 2018, **122**, 23978–23984.
- 43 Y. Xiao, K. Turcheniuk, A. Narla, A.-Y. Song, X. Ren, A. Magasinski, A. Jain, S. Huang, H. Lee and G. Yushin, *Nat. Mater.*, 2021, **20**, 984–990.
- 44 L. Kong, C. Tang, H. J. Peng, J. Q. Huang and Q. Zhang, *Smart Mater.*, 2020, **1**, 1–35.
- 45 R. Sahore, Z. Du, X. C. Chen, W. B. Hawley, A. S. Westover and N. J. Dudney, *ACS Energy Lett.*, 2021, **6**, 2240–2247.
- 46 M. Yamamoto, M. Takahashi, Y. Terauchi, Y. Kobayashi, S. Ikeda and A. Sakuda, *J. Ceram. Soc. Jpn.*, 2017, **125**, 391–395.
- 47 J. Kasemchainan and P. G. Bruce, *Johnson Matthey Technol. Rev.*, 2018, **62**, 177–180.
- 48 H. Yuan, H.-X. Nan, C.-Z. Zhao, G.-L. Zhu, Y. Lu, X.-B. Cheng, Q.-B. Liu, C.-X. He, J.-Q. Huang and Q. Zhang, *Batteries Supercaps*, 2020, **3**, 596–603.
- 49 A. Sakuda, K. Kuratani, M. Yamamoto, M. Takahashi, T. Takeuchi and H. Kobayashi, *J. Electrochem. Soc.*, 2017, **164**, A2474.
- 50 C. Loh, R. Djenadic, M. Bruns, O. Clemens and H. Hahn, *J. Electrochem. Soc.*, 2016, **164**, A6131.
- 51 M. Saccoccio, J. Yu, Z. Lu, S. C. Kwok, J. Wang, K. K. Yeung, M. M. Yuen and F. Ciucci, *J. Power Sources*, 2017, **365**, 43–52.
- 52 D. Hanft, J. Exner, M. Schubert, T. Stöcker, P. Fuierer and R. Moos, *J. Ceram. Sci. Technol.*, 2015, **6**, 147–182.
- 53 G. L. Zhu, C. Z. Zhao, H. J. Peng, H. Yuan, J. K. Hu, H. X. Nan, Y. Lu, X. Y. Liu, J. Q. Huang and C. He, *Adv. Funct. Mater.*, 2021, 2101985.
- 54 D. H. Kim, Y.-H. Lee, Y. B. Song, H. Kwak, S.-Y. Lee and Y. S. Jung, *ACS Energy Lett.*, 2020, **5**, 718–727.
- 55 K. N. Jung, H. S. Shin, M. S. Park and J. W. Lee, *ChemElectroChem*, 2019, **6**, 3842–3859.
- 56 EUROBAT. BATTERY 2030+. (Accessed: 7/12/2021); <https://battery2030.eu/research/roadmap/>.
- 57 China Society of Automotive Engineers. Energy-saving and New Energy Vehicle Technology Roadmap 2.0 officially released. (Accessed: 7/12/2021); <http://en.sae-china.org/a3967.html>.
- 58 Federal Consortium for Advanced Batteries. National Blueprint for Lithium Batteries 2021–2030. (Accessed: 7/12/2021); https://www.energy.gov/sites/default/files/2021-06/FCAB%20National%20Blueprint%20Lithium%20Batteries%200621_0.pdf.
- 59 Y. Ding, Z. P. Cano, A. Yu, J. Lu and Z. Chen, *Electrochem. Energy Rev.*, 2019, **2**, 1–28.
- 60 Z. P. Cano, D. Banham, S. Ye, A. Hintennach, J. Lu, M. Fowler and Z. Chen, *Nat. Energy*, 2018, **3**, 279–289.
- 61 P. Liu, R. Ross and A. Newman, *MRS Energy Sustain.*, 2015, **2**, E12.
- 62 B. Nykvist and M. Nilsson, *Nat. Clim. Change*, 2015, **5**, 329–332.
- 63 D. Andre, S.-J. Kim, P. Lamp, S. F. Lux, F. Maglia, O. Paschos and B. Stiaszny, *J. Mater. Chem. A*, 2015, **3**, 6709–6732.
- 64 L. Xu, Y. Lu, C.-Z. Zhao, H. Yuan, G.-L. Zhu, L.-P. Hou, Q. Zhang and J.-Q. Huang, *Adv. Energy Mater.*, 2021, **11**, 2002360.
- 65 J. Schnell, H. Knörzer, A. J. Imbsweiler and G. Reinhart, *Energy Technol.*, 2020, **8**, 1901237.
- 66 Y.-S. Hu, *Nat. Energy*, 2016, **1**, 16042.
- 67 C.-F. Wang, L. He, H.-J. Sun, P. Lu and Y. Zhu, *China Pat.*, CN110165118A, 2020.
- 68 S. Randau, D. A. Weber, O. Kötz, R. Koerver, P. Braun, A. Weber, E. Ivers-Tiffée, T. Adermann, J. Kulisch and W. G. Zeier, *Nat. Energy*, 2020, **5**, 259–270.
- 69 H. Wakayama, H. Yonekura and Y. Kawai, *Chem. Mater.*, 2016, **28**, 4453–4459.
- 70 D. H. Kim, D. Y. Oh, K. H. Park, Y. E. Choi, Y. J. Nam, H. A. Lee, S.-M. Lee and Y. S. Jung, *Nano Lett.*, 2017, **17**, 3013–3020.
- 71 P. A. Nelson, K. G. Gallagher, I. D. Bloom and D. W. Dees, Argonne National Lab. (ANL), Argonne, IL (United States), 2012.
- 72 S. Ito, S. Fujiki, T. Yamada, Y. Aihara, Y. Park, T. Y. Kim, S.-W. Baek, J.-M. Lee, S. Doo and N. Machida, *J. Power Sources*, 2014, **248**, 943–950.
- 73 R.-J. Chen, Y.-B. Zhang, T. Liu, B.-Q. Xu, Y.-H. Lin, C.-W. Nan and Y. Shen, *ACS Appl. Mater. Interfaces*, 2017, **9**, 9654–9661.
- 74 C. Yang, P. Li, J. Yu, L.-D. Zhao and L. Kong, *Energy*, 2020, **201**, 117718.
- 75 S. Li, M. Jiang, Y. Xie, H. Xu, J. Jia and J. Li, *Adv. Mater.*, 2018, **30**, 1706375.
- 76 H. S. Shin, W. G. Ryu, M. S. Park, K. N. Jung, H. Kim and J. W. Lee, *ChemSusChem*, 2018, **11**, 3184–3190.