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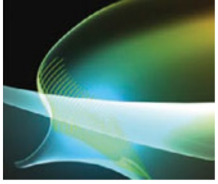

PERSPECTIVE

Prospects on large-scale manufacturing of solid-state batteries

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ABSTRACT

Widespread deployment of solid state batteries requires facile, high-throughput coating processes. Solid state batteries that utilize energy dense anodes may have similar manufacturing costs as traditional lithium ion batteries.

Widespread deployment of renewable energy and electrification of transportation are necessary to decrease greenhouse gas emissions. All solid-state batteries that employ a solid electrolyte, instead of a liquid electrolyte, are well suited for energy dense anodes (e.g., Li metal, Si, etc.) and may be capable of extending the current driving range of an electric vehicles by nearly 2 ×. However, to achieve giga-scale capacities relevant to the EV market large-scale manufacturing approaches are necessary. Solid-state batteries are likely to adopt coating techniques and processing approaches similar to solid oxide fuel cells and conventional battery systems. While control over microstructure, interfaces, and thickness are paramount for achieving long lifetimes, processing speed governs cost and scalability. This perspective highlights the state-of-the-art for solid-state battery manufacturing approaches and highlights the importance of utilizing conventional battery manufacturing approaches for achieving price parity in the near term. Decreasing material costs and improving cell architecture (biploar) may further decrease manufacturing costs.

Keywords scale-up/manufacturing; energy storage; interface; transportation

Introduction

Transportation accounts for ~ 19% of the global energy consumption and 23% of energy-related carbon dioxide (CO_2) emissions. [1] Replacing fossil fuel vehicles with electric vehicles (EV) could ameliorate some of these issues. However, gasoline-powered vehicles exceed 300 miles/tank, while most electric vehicles fall between 100 and 200 miles/charge. Furthermore, the current state-of-the-art lithium ion battery contains flammable liquids which make it susceptible to explosions at elevated temperatures (Fig. 1a). Thus, for widespread market penetration, there is growing interest in increasing the driving range and improving battery safety. All solid-state batteries could potentially address both the safety and range requirements necessary for EV adoption.[2] Solid-state batteries utilize a solid electrolyte instead of a flammable liquid electrolyte to move ions between the anode and cathode. There are two notable benefits associated with using a solid electrolyte. First, solid electrolytes operate more efficiently and safely at elevated temperatures than liquid electrolytes. Second, solid-state batteries can operate with a lithium metal anode which can enable a 2 × increase in energy density (e.g., drive range) (Fig. 1b). Until recently, solid-state batteries were thought to be impractical because of poor ion transport properties. However, new materials have demonstrated transport properties that are competitive with liquid electrolytes.[3,4] Yet there is still a significant lack in understanding about how solid-state batteries can be manufactured. [2,5,6]

Discussion

- Solid state batteries are receiving a tremendous amount of attention as safe, energy dense, alternatives to traditional lithium ion batteries. However, very little attention focuses on the challenges with processing air-sensitive glass/ceramic materials at giga-scale capacities with realistic geometries (thicknesses). Material cost, material performance, material selection, and processing speed will all likely impact the ultimate application for solid state batteries.

The battery manufacturing industry is expected to grow from 160 GWh to greater than 1000 GWh over the next decade. Resilient and cost-effective manufacturing methods are necessary to accommodate this growth. The majority of the manufacturing growth aims to scale up conventional lithium ion batteries (LIBs) based on liquid electrolytes (Fig. 1a). LIBs are comprised of two porous electrodes separated by a microporous polyolefins separator (e.g., polypropylene) (Fig. 1a). Both electrodes and the separator are flooded with a liquid electrolyte in a secondary filling step. The liquid electrolyte penetrates the porous electrodes and enables facile ion transport throughout the cell. In contrast, solid-state batteries do not have any liquids and thus require solid-state cathodes. Solid-state cathodes are composite materials comprised of a carbon additive, active electrode materials, and solid electrolyte materials (Fig. 1b). Pores in a solid-state cathode are ion-blocking, and thus, the cathode microstructure is important for achieving effective ion transport and material utilization.[7,8] Finally, SSBs utilize a solid electrolyte instead of a porous separator and energy dense anodes (Silicon/Li metal) instead of graphite.[9] Materials processing and handling and electrode manufacturing will differ greatly depending on the type of solid electrolyte and will be discussed in the next section.

Solid-state batteries have properties and characteristics that are similar to both conventional LIBs and solid oxide fuel cells (Fig. 1c). Solid oxide fuel cells utilize a solid electrolyte and contain porous composite electrodes. Solid oxide fuel cells require the use of gas phase reactants and thus employ porous electrodes to achieve effective ion, electron, and gas transport. Solid-state battery manufacturing will likely be a hybrid approach which adopts processes from both conventional LIBs and solid oxide fuel cell communities. Currently, LIB cell and pouch manufacturing involves a large number of consecutive and continuous processes which can be described by three primary steps: (1) electrode processing, (2) cell production, and (3) cell conditioning.[5] (Fig. 1d). Conventional LIB electrodes are processed using scalable solution-processed approaches. A colloidal ink is formulated by mixing the active material (anode/cathode) with a nominal amount of polymer binder and conductive additives.[2,10,11] The ink is mixed using high-shear mixing approaches and then coated directly to a current collector via slot die or blade coating processes (Fig. 1d). Post-processing techniques (heating, solvent removal, cutting, and calendaring) are utilized to prepare electrodes with precise microstructural properties (density) and to remove any residual solvent.[12-14] Wet- or solution-based processing enables working speeds up to 25–50 m^2/min and have the potential to get as fast as 100 m^2/min (Fig. 1f). The roll-to-roll speed is dependent on the material properties, electrode thickness, ink formulation, substrate, and machine limitations (coating technique).[10,11] Ink formulation is a critical step in material processing and is often achieved through empirical approaches. Changing a material, solvent, processing approach, or speed can greatly change the formulation step. Rational approaches to ink engineering are necessary and may benefit greatly from machine learning techniques due to the large parameter space. Ink formulation vary greatly between solid oxide fuel cell and LIBs (Fig. 1e). LIBs tend to minimize inactive materials (e.g., binder/additives) and require low-viscosity inks compatible with high-throughput slot die (Fig. 1f). In contrast, solid oxide fuel cells incorporate dispersants and pore formers that lead to more viscous feedstock inks. Pore formers enable control over electrode microstructure which is critical for achieving effective transport. The rheological properties, governed by the ink formulation, are largely important for achieving uniform and scalable coating. Different rheological properties are advantageous for different coating techniques (slot die, doctor blade, screen printing, etc.) (Fig. 1f). Conventional LIBs use slot die or doctor blade approaches and solid oxide fuel cells use a combination of blade coating, tape casting, and screen printing. Solid-state batteries, depending on the type of solid electrolyte, may employ a range of coating techniques (Fig. 1f). Solid oxide fuel cells manufacturing approaches are typically slower than processing approaches used in lithium ion batteries. Thus, solid-state batteries that adopt identical or similar processing techniques to solid oxide fuel cells may face challenges in cost and scale. Batteries for electric vehicles will require giga-scale production, and slow processing and manufacturing approaches will require higher capital investments, larger plants, and greater human investments.

Solid electrolytes and materials processing challenges

The manufacturing approach for solid-state batteries is going to be highly dependent on the material properties of the solid electrolyte. There are a range of solid electrolytes materials currently being examined for solid-state batteries and generally include polymer, sulfide, oxides, and/or halides (Fig. 2a). Sulfides demonstrate excellent transport properties (>10 mS/cm), but scalable production may require the use of clean/dry room environments to avoid H_2S formation. Oxides are hard ceramics and resemble materials typically used in solid oxide fuel cells. Finally, polymer electrolytes are potentially the most mature solid electrolyte in terms of manufacturing and processing. Figure 2a provides a direct comparison of material tradeoffs (transport, stability, processing, and air stability). While there are more properties to consider, this highlights some of the intrinsic trade-offs that exist, depending on which material family is pursued for scalable solid-state batteries.

Another material family which has attracted a lot of attention is composite or hybrid solid electrolytes. A hybrid or composite solid electrolyte is composed of an ion conducting organic polymer and an inorganic material (i.e., SiO_2, $Li_{1+x}Al_xTi_{2-x}(PO_4)_3$). The addition of an inorganic components has been widely explored, in battery and membranes in water applications, as a way to increase the electrolyte's mechanical properties and/or critical current density.[15-20] These materials are often denoted as composite electrolytes or composite membranes when the inorganic additive is 'inert' or non-ion conducting. An alternative approach is to combine a polymer ion conductor with an inorganic ion conductor (e.g., hybrid electrolyte). This approach may enable the fast processing speeds

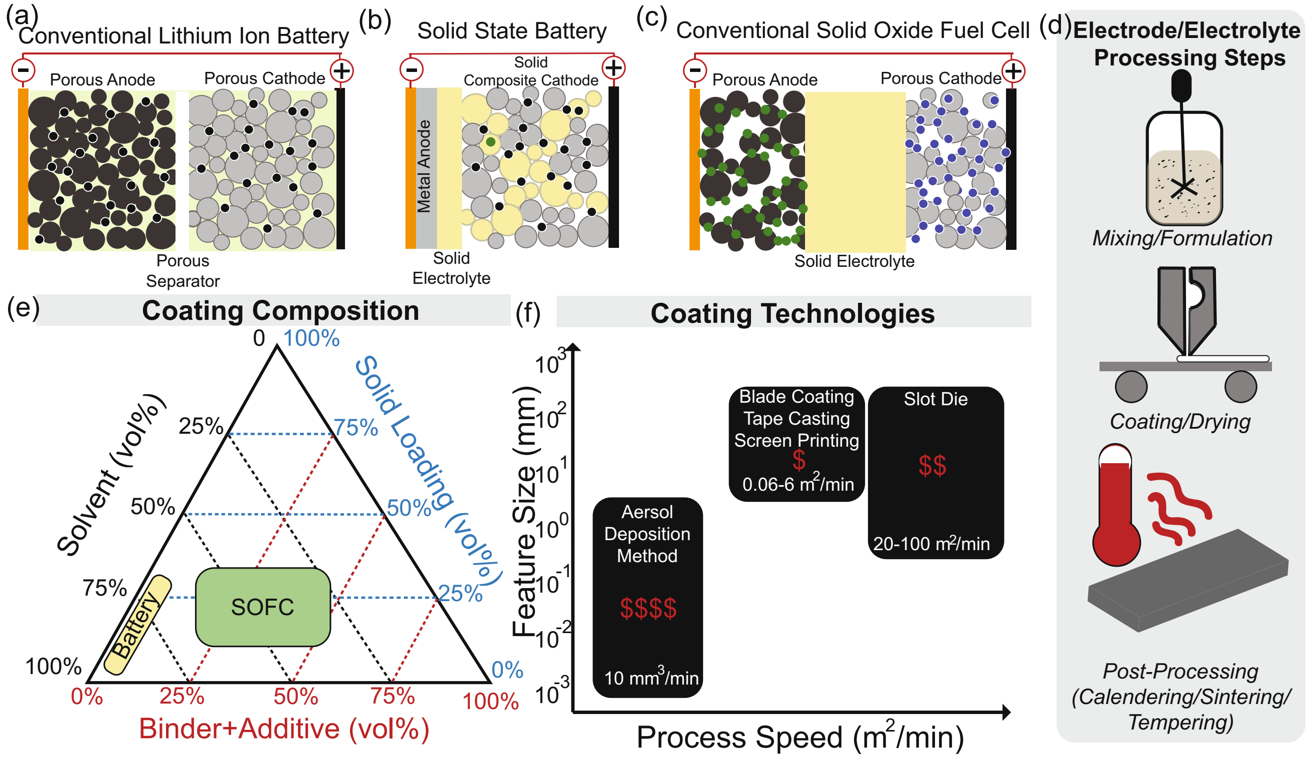

Figure 1. Generic architecture for a conventional lithium ion battery (a), a solid-state battery (b), and a solid oxide fuel cell electrode assembly (c). Most electrochemical systems utilize solution processing approaches for manufacturing (d). The feedstock for solution-based manufacturing approach is an ink. Ink composition (e) is engineered for part performance and for the specific processing technique utilized (f).

associated with polymers and good transport properties associated with inorganic ion conductors. However, the role the inorganic constituent plays on ionic transport does not follow laws of superposition suggesting that transport in these hybrid systems is complex and governed by the underlying interactions between the inorganic and organic phases.[19–23] Thus, there is a significant knowledge gap in our understanding about ionic transport in hybrid and composite solid electrolytes. Hybrid solid electrolytes are composed of three different material phases that can the ion transport pathway: (1) polymer, (2) ceramic, and/or (3) the interphase (Fig. 2b).[24–26] The interphase is a region surrounding the inorganic polymer which has distinct material properties due to polymer confinement and the formation of a space charge layer. Ideally, ion transport would occur through the highly conducting inorganic phase. However, in practice, the ion (Li+) primarily transports through the interphase region because of an interfacial resistance (R_{int}) between the polymer and ceramic phases.[26–29] This resistance prevents transport between the two materials. Numerous reports confirm this surface-driven transport mechanism using nuclear magnetic resonance, modeling, and electrochemical techniques.[24,30]

Despite the rapid improvement in solid electrolyte transport properties, competitive energy densities are necessary to displace the current state of the art. The volumetric energy density of a Li-metal solid-state battery decreases as the electrolyte thickness increases and the electrode loading decreases (Fig. 2c). Figure 2c estimates the impact of energy density on solid electrolyte thickness and cathode loading assuming a low density solid polymer electrolyte. The black horizontal line on Fig. 2c represents the energy density of the current state-of-the-art lithium ion battery (graphite anode and liquid electrolyte). Calculations in Fig.2c are replicated from McCloskey et al.[31] assuming an electrolyte with a electrolyte density slightly greater than polyethylene oxide, an NCA cathode, and varying cathode loadings. Solid electrolyte thicknesses below 40μm (10 mg/cm^2), 125μm (20 mg/cm^2), and 225μm (30 mg/cm^2) are necessary in order for a solid-state (polymer-based electrolyte) Li-metal battery to exceed the state of the art (liquid electrolytes) (Fig. 2c). These 'break-even' thicknesses will decrease for denser solid electrolytes. Typically, inorganic electrolytes (LLZO/LATP) are around 700–1500μm. While achieving thin electrolytes is possible using pulsed laser and aerosol deposition approaches, slow processing speeds increase battery costs.[32] Thus, solid electrolytes that have similar form factors to traditional separators and can utilize existing manufacturing processes (roll-to-roll coatings) are ideal. Low-density hybrid electrolytes can achieve competitive energy densities at thicknesses between 80 and 150μm and thus represent promising future material systems for scalable manufacturing.

Electrolyte thickness, electrode microstructure, and interfaces need to be controlled in solid-state batteries during materials processing and/or manufacturing. Electrolyte thickness, as demonstrated in the previous section plays a significant role on achievable energy density. Electrode microstructures is also very important. Conventional electrodes are porous and infiltrated

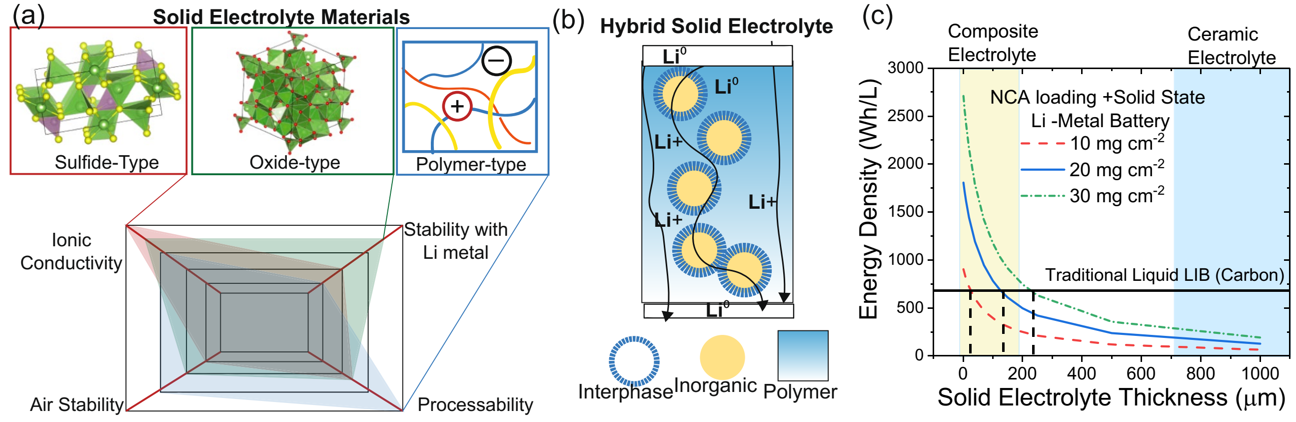

Figure 2. Examples of material families being explored for solid electrolyte include oxide, sulfide, and polymers. There are various trade-offs to consider in materials selection (a). Hybrid solid electrolytes combine an inorganic ion conductor with a polymer ion conductor. Interactions between the two phases governs ion transport mechanisms (b). For solid-state batteries to exceed the state-of-the-art Li-ion battery, the electrolyte needs to be thin (c).

with a liquid electrolyte. Percolating pores allow for effective ion transport within the cathode. Unlike conventional cathodes, solid-state cathodes are composite materials comprised of solid electrolyte and cathode material. Thus, the microstructure is critical for establishing transport pathways within the cathode. Finally, interfaces are important for long-lasting solid-state batteries. Non-uniform or irregular interfaces can lead to local ionic flux which accelerate chemo-mechanical degradation models.[9,33,34]

There are two primary process chains that are being explored for solid-state batteries and are highly dependent on the material choice for the electrolyte.[34,35] The first approach highlighted in Figure 3 is widely employed in ceramics or oxides used as dielectric capacitors and solid oxide fuel cells.[36,37] This approach involves tape casting a composite cathode and sintering to achieve a dense component. Next, a thin solid electrolyte can be processed directly on the composite cathode using a deposition process.[6,38,39] Aerosol deposition enables exquisite control over electrolyte thickness and is a promising approach for achieving control over the interface. However, it was recently shown that aerosol deposition techniques would need to exceed 1000 mm^3 min^{-1} to be a viable manufacturing approach. This is two orders of magnitude greater than the state-of-the-art aerosol deposition technology (10 mm^3 min^{-1}). Maturation of this technology may enable these rates and is a much needed open area of research. The second approach to solid-state battery manufacturing is known as the trilayer approach. This approach is widely employed in solid oxide fuel cells and involves creating one layer of electrode with a pore former, sintering away the pore former, and infilling the cathode with solid electrolyte (Fig. 3). Alternatively, two layers of solid electrolyte can be processed directly on top of each other (one with a pore former, and one without). Upon sintering, a porous framework will be left behind that can be infilled with a cathode. This enables interconnected ion transport pathways in the cathode. Both of these approaches are being explored for all inorganic solid electrolytes. Yet these techniques rely on processing techniques that are inherently slower (tape casting, screen printing, etc.) than slot die used in conventional processing approaches and may require multiple passes for effective filling of porous structures. Ultimately, slower processing speeds will drive up the cost of the manufacturing process. The two processing strategies above assume that the anode is lithium metal, silicon, or an anode-less geometry. Integration strategies will be vastly different than conventional lithium ion batteries and be highly dependent on the material properties (mechanical, chemical, etc.) of the solid electrolyte.

While it would be ideal if solid-state batteries could adopt similar manufacturing and/or coating processes to conventional lithium ion batteries, it is unlikely that a perfect replication is possible. There are several notable differences between the two types of batteries that will require, at the very least, different post-processing approaches. Most notably, control over interfaces, integration of energy dense anodes, and/or anode-less geometries will definitely impact stacking, calendaring, and processing. Lithium metal processing adds another complexity that currently does not exist in battery manufacturing. Li metal manufacturing can occur either via calendering and rolling bulk material or via a extrusion process.[5,6] The cost of lithium metal varies widely (250–1000\$/kg)[40] and processing lithium metal to viable thicknesses within a clean environment is a challenge. Thus, there is great promise in anode-less approaches.

Techno-economic considerations

Conventional battery manufacturing is expected to grow from 160 to 1000 GWh over the next decade to accommodate the projected growth in the electric vehicle market.[5] Decreasing manufacturing costs and increasing manufacturing speed are paramount for further cost reduction at the pack level (< 100\$/kWh).[5,6,41–43] With the rise in battery manufacturing, it is conceivable and ideal that solid-state batteries utilize existing or similar infrastructure to conventional lithium ion batteries. Generally, battery manufacturing encompasses three primary tasks: (1) electrode production, (2) cell production, and (3) cell conditioning. While the

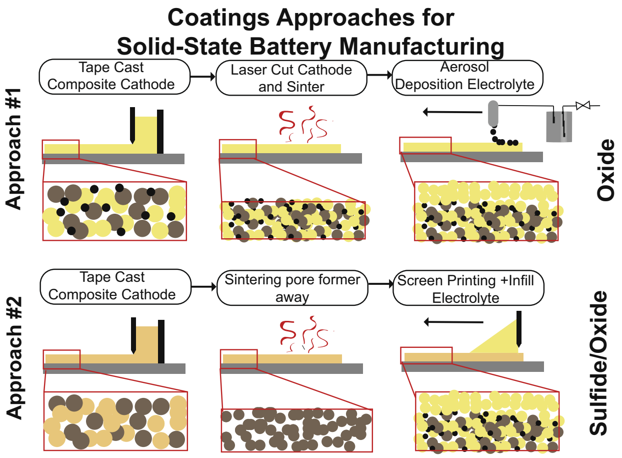

Figure 3. Potential coating approaches for inorganic solid electrolytes (sulfides and/or oxides).

later two processes (cell production and cell conditioning) will be affected by the solid electrolyte material choice, in our present discussion, we only reflect on changes that occur in the electrode processing stage (or electrolyte production) and how transitioning to solid-state batteries may impact battery manufacturing economics. Electrodes are processed using traditional coating processes where the coating technology either coats a single side or both sides of a current collector (Fig. 1f). Typically, slot die or blade coating technologies are implemented accordingly to scale. The manufacturing of the anode and cathode are spatially separated, but follow very similar processing steps (mixing, coating, drying, and calendaring). If the solid electrolyte is processed using a coating process, there will be three coating processes instead of two. To evaluate the impact of an additional coating process on battery manufacturing, we considered the economic framework previously developed by Duffner et al.[5] The analysis is integrated and includes a process model, operating model, and financial model. The process model correlates the product's (e.g., battery cell) dimension (assumption from[42]) and composition with manufacturing technical parameters (the assumptions of coating speed, coater width, machine downtime, scrap rate, and life cycle, etc. from[41]). In our analysis, we assume conservative electrode materials: lithium nickel cobalt aluminum oxides cathode, graphite anode, and lithium thiophosphate (LPS) solid electrolyte. All physical properties for electrodes were extracted from Nelson et al.,[42] and solid electrolyte properties were taken from Schnell et al.[38] The process model combined with characteristic machine properties is used to identify cycle time (the interval required for each product in the coating process). The cycle time can then be used to estimate machine capacity which can be integrated into an operating model. The operating model estimates operating costs required for a plant to meet state-of-the-art production rates. In particular, it considers the interplay between plant footprint, machine run time, and labor usage. In addition, the operating model considers facility-wide operating parameters such as working days per year, shifts per day, hours of shift, hours of paid/unpaid breaks, etc. All model assumptions were taken from Ciez et al.[43] and Sakti et al.[41] Finally, the financial model provides an estimate of specific costs (\$/kWh) accrued as a result of material, operating, and financial considerations (specific materials costs,[38, 42] machine costs,[41] surcharge rates of energy, maintenance and fixed overhead,[43] labor and building space costs[5], and annual discounted rate of building and machine[43]) (Fig. 4).[44]

Figure 4 considers costs associated with the electrode processing step and assumes three identical coating steps for the electrodes and solid electrolytes (instead of two used in conventional batteries). The cell size and design is outlined in Argonne National Lab's BatPac model[41–43] Since we do not consider cell production and conditioning steps, we present a normalized manufacturing cost. The normalized manufacturing costs estimate the increase in electrode processing and are normalized to a best case or low-cost scenario (two coating processes at 100 m/min). The bar graph shows how the cost is distributed for a solid-state battery depending on the coating speed. At high coating speeds (100 m/min), material costs dominate and at low coating speeds (1 m/min) annualized machine and building investment dominates. The manufacturing speed is governed by the coating technology (e.g., machine capabilities) as well as the feedstock material composition (e.g., ink). Under a specific manufacturing capacity with a fixed overall material cost, increasing coating speed decreases the cycle time of each product. Therefore, the annual investment in machines (proportional to the required number of machines) as well as the parasite costs relevant to plant footprint, manpower, machine maintenance, and overhead are reduced. Coating costs associated with solid-state batteries are higher than conventional lithium ion batteries at all process speeds. However, the margin decreases with increasing process speeds. There are additional costs to consider when the solid electrolyte is air- and moisture-sensitive. Materials that are sensitive will require additional investments in dry rooms and the footprint, labor, and energy costs will result in higher costs. In our analysis, we do not assume these costs, and thus, the values represent a 'optimal scenario'.[5] However, these estimates assume a graphite anode. Solid-state batteries that integrate energy dense anodes (such as silicon or lithium metal) maybe able to overcome this cost differential. Bipolar electrodes, an advanced battery structure, enable electrodes connected in series without external accessories like taps and wires. It may reduce the battery volume and weight and the relevant material costs[45] Additional manufacturing strategies, such as multi-material processing may also enable decreases in manufacturing and processing costs. However, regardless of the approach, processing speed is going to be critical to achieve giga-capacities and enable solid-state batteries in electric vehicle applications.

Conclusions

The battery manufacturing industry is expected to grow by an order of magnitude in the next decade. Battery manufacturing involves three primary processes: (1) electrode production, (2) cell production, and (3) cell conditioning. All of these processes will be altered for solid-state batteries and are highly dependent

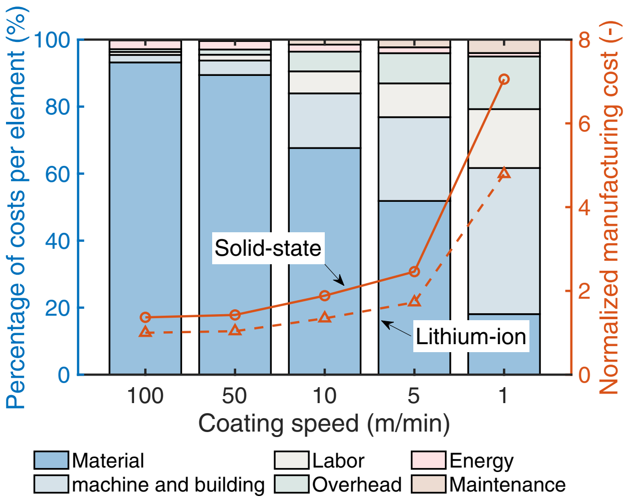

Figure 4. Percentage of cost per element as a result of process speed for solid-state batteries manufacturing (bar graph). Normalized manufacturing cost for conventional lithium ion and solid-state lithium ion batteries as a function of coating speed.

on the material properties of the solid electrolyte. It is likely that solid-state batteries will adopt manufacturing approaches from both the solid oxide fuel cell and conventional battery manufacturing community. Ultimately, advanced coating technologies are necessary to achieve control over microstructure, interfaces, and form factor. Immature technologies such as aerosol deposition are promising techniques for control over interfaces and microstructure, but face significant challenges with scale. For solid-state batteries to be a cost-effective alternative to conventional lithium ion batteries, it is critical to achieve processing speeds exceeding 20 m/min and incorporate energy dense anodes.

Acknowledgments

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