

1 Manufacturing Scalability Implications of Materials Choice in Inorganic 2 Solid-State Batteries

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7 Abstract

8 The pursuit of scalable and manufacturable all-solid-state batteries continues to intensify, motivated by the
9 rapidly increasing demand for safe, dense electrical energy storage. In this Perspective, we describe the
10 numerous, often conflicting, implications of materials choices that have been made in the search for
11 effective mitigations to the interfacial instabilities plaguing solid-state batteries. Specifically, we show that
12 the manufacturing scalability of solid-state batteries can be governed by at least three principal
13 consequences of materials selection: (1) the availability, scaling capacity, and price volatility of the chosen
14 materials' constituents, (2) the manufacturing processes needed to integrate the chosen materials into full
15 cells, and (3) the cell performance that may be practically achieved with the chosen materials and processes.
16 While each of these factors is, in isolation, a pivotal determinant of manufacturing scalability, we show that
17 consideration and optimization of their collective effects and tradeoffs is necessary to more completely
18 chart a scalable pathway to manufacturing.

19 Context & Scale

20 With examples pulled from recent developments in solid-state batteries, we illustrate the consequences of
21 materials choice on materials availability, processing requirements and challenges, and resultant device
22 performance. We demonstrate that while each of these factors is, by itself, essential to understanding
23 manufacturing scalability, joint consideration of all three provides for a more comprehensive understanding
24 of the specific factors that could impede the scale up to production. Much of the recent activity in solid-
25 state battery research has been aimed at mitigating the various interfacial instabilities that currently prevent
26 the fabrication of a low cost, high performance device. With such a wide breadth of options, it can be
27 difficult for researchers to identify the most promising or scalable pathways forward. As such, we aim to
28 empower researchers to make more informed decisions by providing them with insights into how their
29 materials choices are likely to impact the manufacturing scalability of different interfacial mitigation
30 alternatives. In doing so, we hope that generalizable lessons about scalability can be extracted and applied
31 to subsequent challenges in solid-state battery development, thereby accelerating their scale up to
32 manufacturing.

33 Introduction

34 The demand for electrical energy storage is widely anticipated to escalate over the coming years.
35 Considerable growth in global renewable energy capacity and electrified mobility¹ necessitate an efficient,
36 cost effective, safe, and large-scale means of storing energy. While the cost and performance of traditional
37 lithium-ion batteries continues to improve², interest and effort in the development of all solid-state lithium
38 batteries has been accelerating. By enabling the use of thin, high capacity lithium metal anodes and thin,
39 solid electrolyte separators, solid-state batteries potentially provide for a higher energy density storage
40 solution. Moreover, by eliminating the need for volatile liquid electrolytes, solid-state batteries could also
41 offer a safer, more reliable alternative to current rechargeable batteries.

42 In the pursuit of sufficiently high performing solid-state batteries, a wide range of electrolyte chemistries
43 and structural archetypes are under investigation: oxide-based garnets³⁻⁸, LISICON-like (lithium superionic
44 conductor) and argyrodite-type sulfides⁹⁻¹⁹, NASICON-like (sodium superionic conductor)
45 phosphates^{9,11,12,14-16,20-26}, and perovskites^{11,12,14-16,20,21,23}, among others. Given this range of options, how
46 are promising pathways to large scale integration to be identified? The factors that can govern the
47 manufacturability and scalability of materials dependent technologies, like batteries, are considerably
48 varied²⁷. This is further compounded during the transition from lab to production, as the challenges that
49 confront battery scientists and engineers continue to evolve.

50 At present, the integration of solid electrolytes into full, solid-state battery cells remains largely an unsolved
51 challenge. Much effort is currently being directed toward improving the numerous instabilities that
52 continue to degrade the cathode and anode interfaces in these cells (high impedance, dendrite formation,
53 interphase formation and interdiffusion, etc.)²⁸⁻⁴⁶. But as with the diversity of candidate solid electrolytes,
54 a broad range of strategies has been reported in the literature to mitigate these interfacial problems. Much
55 of this work, for example, has focused on stabilizing the interface between the solid electrolyte and lithium
56 metal anodes through the use of artificial interlayers. Buffer layers have been incorporated through a variety
57 of methods including liquid phase deposition^{30,33,41-43,47}, evaporation^{32,33,39,40}, atomic layer deposition^{34,34,48},
58 sputtering^{35,38,49,50}, powder pressing⁴⁴, and melt deposition³⁷. Similar strategies have also been applied to
59 improve the interface between cathode active material particles and various solid electrolytes. Very thin
60 layers of certain oxides, for instance, have been applied to cathode particles to stabilize their interfaces with
61 sulfide solid electrolytes⁵¹⁻⁵⁷. Likewise, lithium borates have been found to provide a low melting point,
62 high conductivity, and high bonding material to improve the interface between cathode active materials and
63 garnet electrolytes^{28,29,58,59} under their required processing conditions.

64 This considerable materials diversity affords us the opportunity to examine how materials selection could
65 potentially impact manufacturing scalability. We thus review and analyze a selection of literature-reported,
66 solid-state batteries employing varied interfacial mitigation approaches and materials to show that the
67 manufacturing scalability of solid-state batteries is driven, at least in part, by: (1) the availability, scaling
68 capacity, and price volatility of the chosen materials' constituents, (2) the manufacturing processes
69 prescribed by the properties of the chosen materials, and (3) the specific capacities that can be extracted
70 from the chosen materials in practice. We find that exploration of any of these factors in isolation may miss
71 critical aspects of battery scalability. Rather, the numerous, sometimes diverging, consequences of
72 materials choice in solid-state battery design must be collectively evaluated such that their combined effects
73 and tradeoffs are considered.

74 **Materials and Availability**

75 Due to the dominant role of materials costs in overall solid-state lithium battery cost^{60,61}, supply chains for
76 the various required electrolyte precursors must be rapidly developed such that their production costs fall
77 by several orders of magnitude. Lab scale pricing for many of these precursors remains in the thousands or
78 tens of thousands of dollars per kg (at the proper purities). To reach the manufacturing scale materials cost
79 assumptions employed in this analysis, however, electrolyte precursor costs will have to be reduced by
80 approximately 100x. While this may be achievable for some of the relevant precursors in the near term,
81 sizeable deployment of solid-state batteries relying on a single electrolyte chemistry may ultimately be
82 constrained by the inability of precursor materials supply chains to achieve sizable economies of scale⁶².

83 A key determinant of materials cost is the actual or perceived availability of the elements that constitute
84 that material's precursors. Given this, we evaluated the availability of several key constituents of three
85 common solid-state electrolytes: $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZO), $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$ (LGPS), and $\text{Li}_6\text{PS}_5\text{Cl}$. Our initial
86 screening of their availability produced a list of five, potentially constrained, elements: Ge, La, Nb, Ta, and
87 Zr. We acknowledge that additional elements other than those included here (Ta, Nb) are sometimes used

88 throughout the literature to dope LLZO—Al, Ca, Ga, and Tb, for example. However, using our natural
89 language processing pipeline⁶³⁻⁶⁶, we have determined that Ta-doped LLZO remains the most frequently
90 reported variant (the full LLZO corpus used in our analysis can be found online at synthesisproject.org),
91 likely due to its comparatively high ionic conductivity and processability⁴¹. Recent analysis of the literature
92 by Mahbub, et al., has likewise revealed that c-site modification (of which Ta-doping is one example) of
93 similar lithium garnets is correlated with reduced processing temperatures while still being able to maintain
94 suitably high ionic conductivities⁶⁷. We, therefore, focus our present analysis on Ta-doped LLZO as
95 reported in the work by Han, et al²⁹. While cathode active materials cost constitutes large fractions of cell
96 cost, we refer the reader to prior work done in this area⁶⁸ and instead focus here on the availability of
97 electrolyte and interfacial mitigation materials.

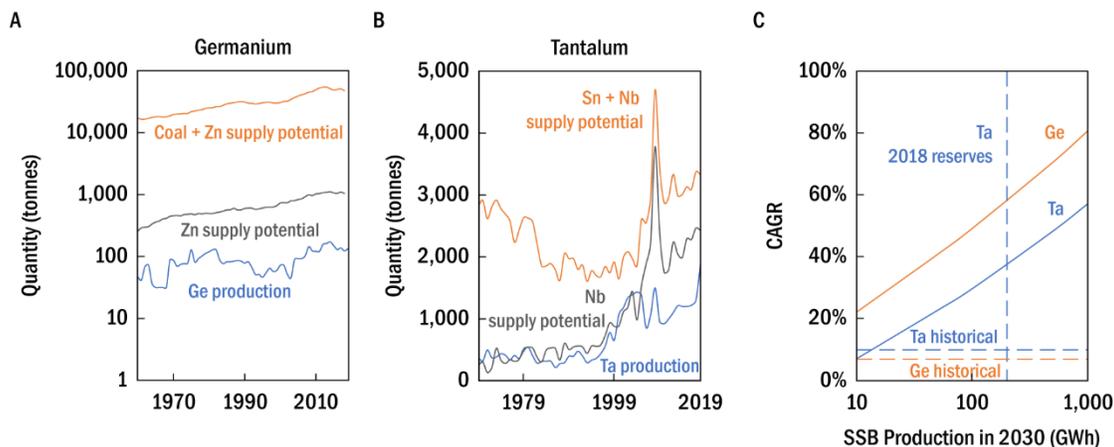
98 To evaluate materials availability, we examined the viability of significant scale up in the production of Ge,
99 La, Nb, Ta, and Zr, as informed by expected electric vehicle (EV) growth (method and full analysis
100 provided in Supplemental Information). Based on our initial screening, we find no major concern associated
101 with Nb or Zr. Ge and Ta, however, required further investigation (La raises minor availability concerns,
102 but due to data availability, we focus on Ge and Ta in our analysis). At a battery production volume of 100
103 GWh/yr (annual EV production in 2030⁶⁹⁻⁷¹), production of these critical elements must scale to 120 and
104 22 times their 2018 production, respectively⁷².

105 Both Ge and Ta are mined as by-products, metals mined and produced from the extraction of other
106 materials. Ge is produced as a by-product of coal ash and Zn, while Ta results from the production of Sn
107 and Nb. The supply of these by-products is contingent on the dynamics of their carrier materials⁷³. When
108 prices of by-products increase, but not those of the carriers, dedicated increases in carrier production is not
109 typically expected (assuming that production economics are driven by the price of the carrier), effectively
110 constraining the supply of the by-products. To gauge this, supply potential is used, the maximum by-
111 product content accompanying the production of the carrier⁷⁴. This provides an upper bound to the by-
112 product supply that could be obtained without a dedicated increase in carrier production. The supply
113 potentials of Ge and Ta are presented in Fig. 1.

114 Chiefly produced in China, Ge is a by-product of the processing of Zn ore, which is initially recovered by
115 leaching Zn-refining residues or coal ash, precipitating concentrate from this leachate (to GeCl_4), and then
116 hydrolyzing the concentrate to produce GeO_2 ⁷⁵. As the precursor commonly used to synthesize Ge-
117 containing solid-state electrolytes (such as LGPS) is GeS_2 , the obtained GeO_2 must then undergo further
118 processing. Unfortunately, however, less than 5% of the Ge contained in Zn concentrates ever reaches
119 refineries capable of extracting and producing Ge as described. The Ge supply potential from Zn and coal
120 ash is compared to Ge production from 1960-2018 in Fig. 1a. We see that the total potential supply of Ge
121 from Zn and coal is 100x what is currently produced. Accordingly, while the elemental availability of Ge
122 is not in question, facilities that can process this material are limited. Whether this capability can be rapidly
123 expanded will no doubt be dictated by profit maximization and environmental regulations. Moreover, any
124 aggressive increase in Ge processing capacity will also surely impact Ge price and, hence, LGPS cost.
125 Given these logistical constraints, Ge recycling from recovered scrap, which currently accounts for 30% of
126 world production, will need to play a more significant role.

127 Ta is derived as a by-product of Sn and Nb (two materials that, by themselves, are also relevant to solid-
128 state battery production), with the relevant Ta-containing precursor being Ta_2O_5 . A significant fraction of
129 Ta material trade is undocumented, as leading ore and concentrate producers rely on artisanal mining
130 operations, a recovery mode that has historically led to supply constraints. As presented in Fig. 1b, the
131 supply potential of Ta from Sn and Nb is less than double current production, indicating that Ta production
132 would, therefore, fail to meet projected increases in demand driven by the scale up of solid-state batteries.

133 However, sizeable Ta resources exist in Australia and Brazil, resources that are not currently being
 134 exploited. Interestingly, Ta could also be recovered as a by-product of Li from spodumene production in
 135 Australia and Canada, further entangling the supply chains of these various battery-relevant materials.



136
 137 **Fig. 1. Materials availability and scaling requirements for Ge and Ta**
 138 (a) Supply potential for Ge from dominant carrier metals compared with current production. (b) Supply potential for
 139 Ta from dominant carrier metals compared with current production. (c) Compound annual growth rate (CAGR)
 140 needed to achieve a given solid-state battery production volume in 2030 in GWh. Vertical dashed line corresponds
 141 to Ta reserves in 2018. Horizontal dashed lines indicate maximum historical CAGRs.
 142

143 The considerable scaling required of Ge and Ta production prompts consideration of the staggering impact
 144 this will have on the supply chains of compounds containing these elements. Fig. 1c presents the required
 145 compound annual growth rate (CAGR) in metal production as a function of solid-state battery production
 146 in 2030 for each of these elements. The required quantity of each element is determined by a materials
 147 intensity that has been calculated based on electrolyte stoichiometry, electrolyte usage in the battery, and
 148 literature-reported battery capacity (details in Supplemental Information). The calculated CAGRs for Ge
 149 and Ta (Fig. 1c) to meet the projected surge in EV production far exceed their respective historical growth
 150 rates⁷⁶. The average 18-year CAGRs for Ge and Ta since 1972 are 1% and 4%, respectively (with maximum
 151 CAGRs of 7% and 10%, respectively). For 100 GWh/yr of annual solid-state battery production in 2030,
 152 the required CAGRs for Ge and Ta are ~50% and ~30%, respectively. Such unprecedented growth rates
 153 are likely to significantly impact the cost of all solid-state model cells discussed throughout this Perspective
 154 except for those based on argyrodite-type electrolytes. The vertical dotted line in Fig. 1c indicates the 2018
 155 reserves limit for Ta (reserves are the quantity which could be extracted or produced at the time of
 156 determination). As can be seen, for the projected growth in battery production, the possible short-term
 157 scarcity of Ta and subsequent availability-driven price increases could considerably impede the scale up of
 158 solid-state batteries based on Ta-doped variants of LLZO (in contrast, reserves and resources for Ge are
 159 found to be sufficient). Recycling could begin to play a role in meeting this demand. However, previous
 160 work indicates this role will likely be limited until 2040 due to vehicle and battery lifetimes⁷⁷. As
 161 researchers explore the use of particular materials, screening analyses can enable them to understand how
 162 the scaled resource use will impact primary supply based on the fraction of total end-use demand that the
 163 new application requires⁷⁸.

164 Given the lack of available data, we assumed a constant electrolyte price (\$50/kg^{60,61}) across the model
165 battery cells used throughout our analysis; this would not be the case in practice, however. Ge and Ta are
166 extremely high priced elements, exceeded only by Au, Sc, and platinum group metals (50-year average⁷⁹).
167 Moreover, both elements have exhibited high price volatility as well. Ta, for example, has a recent price
168 volatility of 40% (price variation of 40% of its mean over a 5-year period), driven by increased use in
169 electronics applications. Similarly, Ge has exhibited a 30% 5-year average volatility. The scale up of solid-
170 state batteries, therefore, may potentially be hindered not only by the logistical and operational barriers to
171 scaling the requisite precursor materials supply chains at unprecedented rates, but by the price uncertainty
172 and volatility of these high-priced elements as well.

173 Materials availability constraints are provide a significant source of uncertainty that will drive the total cost
174 of solid-state batteries. As has been previously noted for existing battery technologies, the dominance of
175 materials in the cost structure may set a practical lower bound on battery price⁸⁰. However, as materials
176 choices will, in turn, dictate the manufacturing processes that can or must be used, we consider the
177 implications of these choices on cell processing as well.

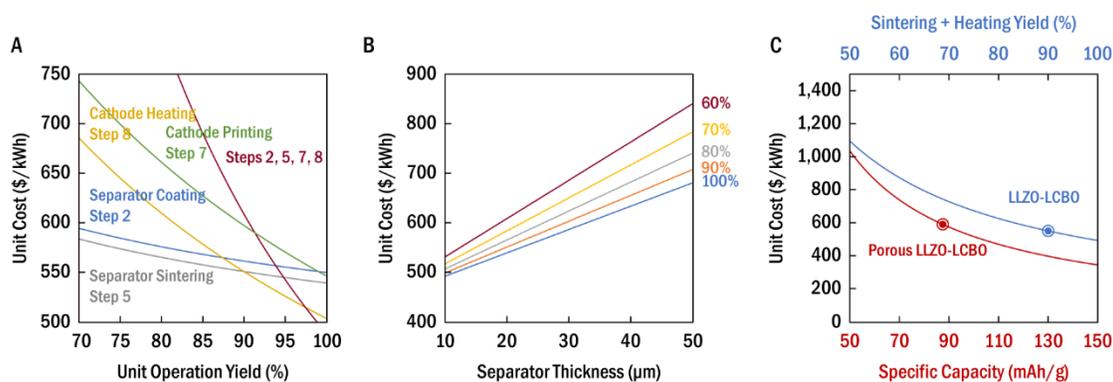
178 Materials and Processing

179 One of the key differences between sulfide and oxide solid electrolytes is the disparity in their elastic
180 moduli, with sulfide solid electrolytes typically exhibiting moduli an order of magnitude lower than those
181 of their oxide counterparts^{10,81-83}. This key difference in electrolyte materials properties has several
182 consequences. First, sulfide electrolytes may be better able to accommodate the volume changes that
183 accompany repeated charge and discharge cycles in an all-solid-state cell, thereby maintaining superior
184 solid-solid interfacial contact across the lifetime of the battery⁸⁴. However, this potentially enhanced ability
185 to withstand cycling-induced stresses could be offset by the lower fracture toughness of such materials, a
186 vulnerability that makes them especially sensitive to flaws and defects that may be introduced during
187 processing⁸³. Second, the contrasting mechanical behavior between oxides and sulfides allows for sulfide
188 sheets to be densified and bonded to device stacks through high pressure calendaring, while oxide sheets
189 must instead be sintered at high temperature for densification and adhesion, a critical process barrier that
190 can chemically degrade the interface with other materials, especially cathode active materials²⁸. Third,
191 while the lower modulus of sulfide electrolytes might enable very thin, large area sheets of these materials
192 to better withstand the rigors of large scale manufacturing^{85,86}, higher modulus oxide sheets might be more
193 prone to failure under the requisite large scale handling and processing conditions. Thin, large area, tape
194 cast sheets are well known to warp, crack, shrink, and delaminate upon heating, particularly when the slurry
195 composition and its processing conditions are not properly optimized⁸⁴⁻⁹⁸, making higher modulus oxide
196 sheets especially vulnerable to failure during their required high temperature sintering.

197
198 The consequences of this disparity in mechanical properties is not trivial. And while the processing-driven
199 uncertainty in the ability of sulfide electrolytes to better withstand cycling stresses demands examination,
200 it is a question that currently lies outside the scope of our models. Instead, we analyze how the difficulty
201 of processing and handling extremely thin, large area, oxide electrolyte sheets could manifest by modeling
202 the possible manufacturing costs of two hypothetical cells informed by the work by Han, et al.²⁹ Our
203 analysis is motivated, in part, by the apparent difficulty in producing solid-state batteries based on thin,
204 garnet separators at high yield, even in industry⁹⁹⁻¹⁰⁵. In our model cells, $\text{Li}_{2.3-x}\text{C}_{0.7+x}\text{B}_{0.3-x}\text{O}_3$ (LCBO) is
205 used in the composite cathode as a low melting point, high binding, and suitably conductive “solder” at the
206 interface between cathode active material (LiCoO_2 , LCO) and solid electrolyte particles (Ta-doped LLZO).
207 Given that the solid electrolyte separator must be sintered at high temperature, it must be cast, dried, cut,
208 and sintered by itself first, with the much thicker composite cathode subsequently cast and processed on
209 top (this cell is referred to in our model as the “LLZO-LCBO” cell). However, at the low separator
210 thicknesses (here, 20 μm) required to capture the desired energy density benefits of solid-state batteries, the

211 handling and processing of brittle oxide sheets with areas exceeding 100 cm² can be difficult. Accordingly,
 212 prior work proposes an alternative processing approach. First, a thick, porous oxide electrolyte framework
 213 (a “catholyte”) is cast. Because it is 100 μm in thickness, researchers posit that it may be more easily handled
 214 and manipulated as a free-standing sheet than a 20 μm sheet of the same material^{60,106}. The thin electrolyte
 215 separator is then cast on top, and the two are cut and sintered together before cathode active material is
 216 backfilled or infiltrated into the porous electrolyte framework. This variant of the LLZO-LCBO cell will
 217 be referred to as the “porous LLZO-LCBO” cell. Details for these cells (and the other subsequent model
 218 cells presented throughout this Perspective) as well as an overview of our cost modeling approach are
 219 presented in the Supplemental Information. We note, however, that our manufacturing cost calculations
 220 for each model cell are based on specific, literature reported cell design and performance data. This enables
 221 us to map individual cell design and materials choices to their resulting cell capacities and, as a
 222 consequence, their attendant manufacturing costs. The challenge with this modeling approach, however, is
 223 that the calculated costs of cells whose performance data were recorded at different testing conditions are
 224 not directly comparable. A cell discharging 100 mAh/g at low rate and high temperature does not have the
 225 same performance as a cell discharging 100 mAh/g at high rate and low temperature. Yet, with equivalent
 226 materials and processing, these two cells would be calculated to have equivalent manufacturing costs in
 227 \$/kWh (we elaborate on the implications of this in the Supplemental Information and later in this
 228 discussion).

229
 230 In Fig. 2a, we illustrate the manufacturing cost of the LLZO-LCBO cell as a function of process yield (used
 231 here as a proxy for process difficulty, with higher yield representing greater processing ease) for each of
 232 the indicated process steps. The effects of yield are studied for four key processing steps in LLZO-LCBO
 233 cell assembly: separator coating and drying (step 2), separator sintering (step 5), cathode printing (step 7),
 234 and cathode heating (step 8). Steps are numbered with their positions in the overall manufacturing process
 235 flow (process flows for all our model cells are presented in Table S1). A fifth curve that illustrates how
 236 simultaneous changes in yield to all four of these process steps impacts cell cost is presented as well. These
 237 particular process steps were chosen, as tape cast sheets have been found to more likely fail during forming
 238 and heating operations.
 239



240
 241 **Fig. 2. Effects of process difficulty**
 242 (a) Manufacturing cost as a function of unit operation yield for key processing steps in the fabrication of the LLZO-
 243 LCBO cell. (b) Manufacturing cost as a function of separator thickness and separator sintering yield. (c)
 244 Manufacturing cost as a function of separator sintering + cathode heating yield for the LLZO-LCBO cell vs.
 245 manufacturing cost as a function of cell capacity for the porous LLZO-LCBO cell. Dots indicate baseline model
 246 values for each cell. Each x-axis corresponds to only the curve with the matching color. Specific capacity is based
 247 on active material mass in the cathode.

248
249 Our results indicate that modest drops in the yield of even a single process step can considerably increase
250 the overall manufacturing cost of these cells. The effect of decreases in separator sintering yield, as one
251 might expect due to the failure of these thin, large area, brittle oxide sheets during high temperature
252 processing, is somewhat dampened by the relatively early position of the separator sintering step in the
253 manufacturing process flow. In contrast, manufacturing cost is notably more sensitive to the yield of the
254 cathode heating step. For example, a 5% drop in cathode heating yield from its baseline value results in a
255 cost increase of ~\$30/kWh, a nontrivial increase considering that a commonly accepted target cost for such
256 batteries is \$100/kWh^{107,108}. Given the late position of cathode heating in the overall process flow—and its
257 position after other low yield process steps, like separator sintering—failures during cathode processing
258 have comparatively larger impacts. For example, when cells are lost during cathode heating, both composite
259 cathode material and solid electrolyte separator materials are lost, representing a significant fraction of
260 overall cell cost (see Fig. S1). In addition, many more sheets must be made in the numerous preceding
261 process steps to accommodate these losses late in the process flow. Unsurprisingly then, the sensitivity of
262 manufacturing cost to process yield is even more pronounced when the yield of all four critical heating
263 operations is varied (Steps 2+5+7+8 in Fig. 2a).

264
265 Thus, while the difficulty of obtaining, processing, and handling thin, large area, standalone oxide
266 electrolyte sheets could result in large increases in manufacturing cost, it is even more crucial that
267 downstream process steps maintain high yield. Achieving high yield during cathode heating, for example,
268 could be particularly difficult, as the device stack at this point in the process flow contains both the solid
269 electrolyte separator as well as a thick composite cathode, a complex mixed materials system with a variety
270 of solid-solid interfaces that could be especially vulnerable to thermally induced stresses and defects.
271 Mitigating such processing-induced flaws, especially at the interfaces, will no doubt be integral to ensuring
272 low unit production costs and high resulting device performance.

273
274 With the production cost of LLZO-LCBO cells so dependent on the yield of even just a few, key process
275 steps, serious consideration must be given to alternative manufacturing strategies. For instance, it is
276 potentially appealing to simply use a thicker solid electrolyte separator that can be cast and sintered with
277 greater success (i.e., higher yield). However, given the sensitivity of cell manufacturing cost to separator
278 thickness (shown as a function of separator sintering yield in Fig. 2b), such a simplistic strategy might not
279 offer a clearly advantageous alternative. Take, for example, a scenario in which a 20 μm separator can be
280 sintered with a yield of 80% and a separator with twice that thickness that can be sintered with a yield of
281 100%. Despite the considerable increase in process yield that is captured when using a thicker separator,
282 the increase in solid electrolyte materials usage and the concomitant decrease in cell energy density result
283 in a nearly \$70/kWh net increase in total cell production cost.

284 The hypothetical fabrication of our porous LLZO-LCBO cell also potentially offers a more easily processed
285 alternative. Despite the promise of easier manufacturing, however, reports of similar devices in the
286 literature reveal an essential uncertainty. How successfully can cathode active material be backfilled or
287 infiltrated into a thick, porous, solid electrolyte framework? Given that battery performance hinges on the
288 quality of these solid-solid interfaces⁸⁴, ensuring intimate contact between cathode active material and solid
289 electrolyte across their entire interfacial area is imperative. In one report, for example, a similar porous
290 LLZO device structure was made. Active material precursor solution was then backfilled into the porous
291 electrolyte¹⁰⁹. It was necessary to repeat this process several times to achieve the desired active material
292 loading. Ultimately, solid-state cells produced in this way could not be extensively cycled. Moreover,
293 while this report illustrates the challenges attendant to a manufacturing approach that relies on the
294 backfilling of cathode material into a porous electrolyte, doing so at production scale and speed and with
295 the cathode slurry modeled here instead of the reported precursor solution could be more difficult.

296 To examine the tradeoffs between the production of the LLZO-LCBO cell and that of the porous LLZO-
297 LCBO cell, we compare the manufacturing costs of each in Fig. 2c. The production cost of each cell is
298 plotted as a function of each cell's key processing uncertainty. That is, the cost of the LLZO-LCBO cell is
299 plotted as a function of the process yields for its separator sintering and cathode heating steps (similar to
300 the analysis presented in Fig. 2a), while the cost of the porous LLZO-LCBO cell is plotted as a function of
301 cell specific capacity. In this latter case, capacity is used as a proxy for the quality of the backfilled cathode-
302 electrolyte interface, given the critical role that maintaining solid cathode-solid electrolyte contact has in
303 cell performance⁸⁴.

304 As shown in Fig. 2c, the production cost for a porous LLZO-LCBO cell varies markedly with the resulting
305 capacity of the cell, similar in magnitude to the sensitivity of LLZO-LCBO cell cost to sintering and heating
306 process yield. The dots on each curve represent their baseline values in our models (for both cells, capacity
307 = 87 mAh/g, sintering + heating yield = 90%), as informed by the literature^{29,60}. Accordingly, at these
308 baseline values, neither approach appears universally preferable to the other. Rather, a tradeoff between
309 processability and cell performance exists. The LLZO-LCBO cell will be favored if it is more difficult to
310 successfully and intimately backfill active material into the porous LLZO-LCBO cell (thus resulting in
311 decreased cell capacity) than it is to handle and process thin, large area, brittle oxide electrolyte sheets (thus
312 resulting in decreased process yield).

313 We note that an alternative use of thick, porous electrolyte layers has been described in the literature. Rather
314 than infiltrating such porous electrolyte layers with cathode active material, they can, instead, be backfilled
315 with lithium metal to form a composite anode¹¹⁰⁻¹¹². As with the use case described in Fig. 2, the initial
316 fabrication of a thick, porous oxide electrolyte layer provides for a potentially more easily manufacturable
317 cell. Moreover, the electrolyte's porosity can also serve to accommodate the large volume changes that are
318 expected to occur at the anode during cycling. While employing a thicker composite anode than would
319 otherwise be used in an ideal cell structure (e.g., using a thin, lithium foil anode, as our model assumes)
320 reduces the achievable energy density, this could, in fact, be outweighed by an even more substantial
321 increase in manufacturability afforded by the cell design.

322 The preceding discussion has revealed that not only do materials choice and materials properties dictate the
323 processes that can or must be used (e.g., calendaring vs. sintering) as well as the challenges that could
324 detrimentally impact the yield and, hence, cost of those processes (e.g., failure during heating), but that
325 these considerations must ultimately be weighed against the performance that can be extracted from those
326 materials in order to make meaningful determinations about scalability.

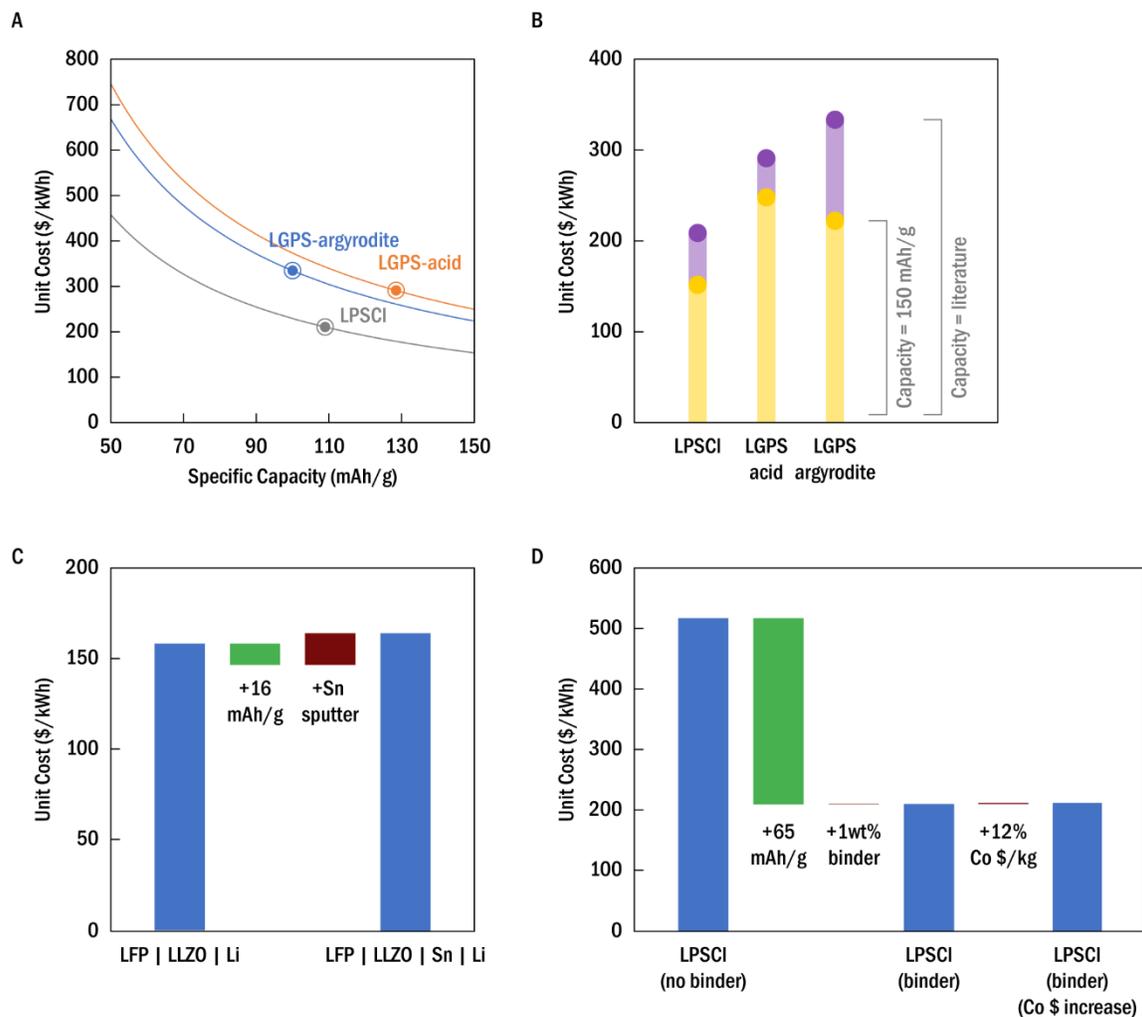
327 **Materials and Performance**

328 While materials and processing are key drivers of manufacturing scalability, how they combine to yield
329 device performance is equally as critical. Battery cost models of the type used here frequently rely on a
330 constant, theoretical or ideal cell capacity and a generalized cell configuration when evaluating
331 manufacturing cost. As such models are typically being used to inform process and production facility
332 optimization at scale, the use of such standards and simplifications to capture the maximum attainable
333 performance (or lowest manufacturing cost) for a finalized device is entirely appropriate. However, as our
334 goal here is to illustrate the scalability implications of various material and interfacial mitigation options
335 currently being investigated in the research literature, our model requires separate performance data for
336 each specific device design. That is, rather than using a theoretical or standard specific capacity for the
337 cathode active materials used in our model batteries, we must instead map the literature reported cycling
338 data for the cells on which our models are based to the specific materials and processing that produced
339 those results (including, for example, specific choices for the electrolyte separator, composite cathode

340 compositions, and whatever interfacial mitigations or buffer layers were required to achieve the reported
341 cell performance).

342 In Fig. 3a we present the manufacturing cost as a function of cell specific capacity for two sulfide-^{43,44} and
343 one argyrodite-based¹¹³ model batteries that represent an illustrative diversity of interfacial mitigation
344 strategies. In the first sulfide-based model cell, the lithium anode is treated with phosphoric acid to enhance
345 its chemical stability with the LGPS electrolyte. This cell will be referred to as the “LGPS-acid” cell. In
346 the other, a Sn-substituted version of the argyrodite electrolyte $\text{Li}_6\text{PS}_5\text{I}$ is introduced as a stabilizing anode
347 interlayer (subsequently referred to as the “LGPS-argyrodite” cell). A third, argyrodite-type cell using a
348 $\text{Li}_6\text{PS}_5\text{Cl}$ electrolyte is modeled. In this example, no additional anode interfacial material is introduced, but
349 the study on which our model is based investigated the effect of binder content in the composite cathode.
350 This cell will be referred to as the “LPSCI” cell.

351 Cost is a strong function of cell specific capacity. Even modest decreases in extracted capacity (e.g., ~10
352 mAh/g) can result in cost increases of nearly \$50/kWh. When the commonly accepted target cost for these
353 batteries is \$100/kWh^{107,108}, such large increases in manufacturing cost can be ill afforded. Given these
354 literature reported cell capacities, for solid-state batteries to be scalable, significant additional gains in
355 performance must be captured with extremely low additional materials and processing costs.

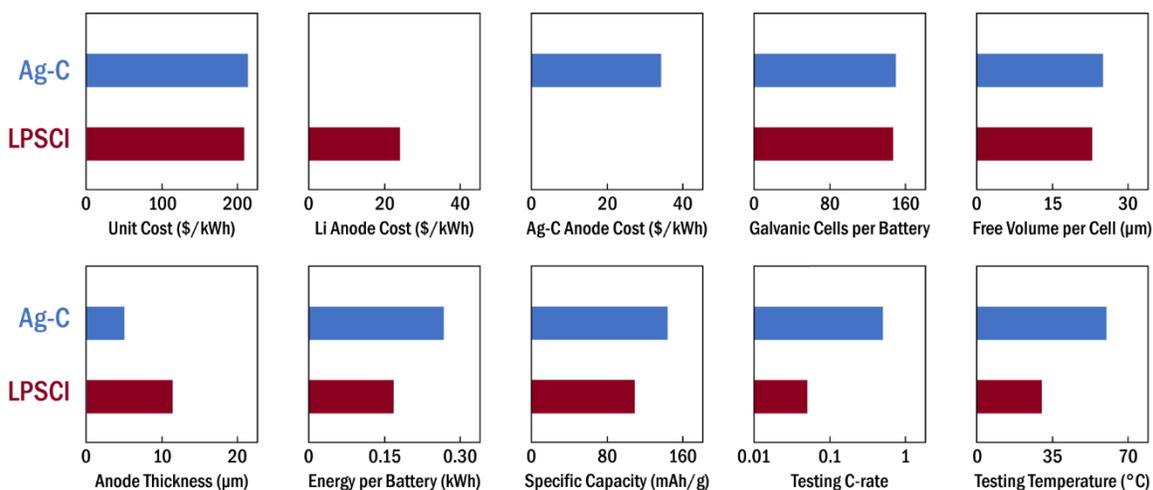


356
 357 **Fig. 3. Tradeoff between cell performance and cell processing cost**
 358 (a) Manufacturing cost of each model cell as a function of cell specific capacity. Dots indicate baseline model
 359 values for each cell. (b) Manufacturing cost as a function of their literature-reported specific capacities (purple dots)
 360 as well as a higher, hypothetical benchmark capacity of 150 mAh/g (yellow dots). The latter is used to isolate cost
 361 differences due only to underlying differences in materials and manufacturing, rather than differences in cell
 362 performance as well. (c) Manufacturing cost of an LFP | LLZO | Li cell utilizing a sputtered tin interlayer at the
 363 anode. Presented as a waterfall plot to illustrate the cost savings due to an increase in capacity upon insertion of the
 364 Sn interlayer as well as the cost increase due to the cost of sputtering the Sn interlayer itself. The manufacturing
 365 cost of this cell cannot be directly compared (in absolute terms) to that of our other model cells given that its high
 366 reported capacity relies on the use of small amounts of liquid electrolyte at the cathode-separator interface.
 367 Moreover, as this volume was unspecified in the report, it has also been excluded from the cost. (d) The LPSCI
 368 model cell discussed throughout this Perspective. Presented as a waterfall plot to illustrate the cost savings due to an
 369 increase in capacity upon the addition of 1 wt% binder to the composite cathode as well as the cost increase due to
 370 the cost of including 1 wt% binder in the composite cathode. Also illustrated is the effect of a 12% increase in the
 371 price of cobalt. Specific capacity is based on active material mass in the cathode in (a)-(d).
 372

373 The substantial impact of cell capacity on manufacturing cost and, critically, the importance of using cell
374 performance data that is specific to each device design and interfacial mitigation strategy is further
375 examined in Fig. 3b. Here, cell cost is presented as a function of both the literature-reported specific
376 capacity for each cell (purple dots) as well as a higher, hypothetical, benchmark cell capacity = 150 mAh/g
377 (yellow dots). Differences in cost among the three cells using their literature-reported capacities reflect
378 both differences in the materials and processing used to construct each cell as well as differences in the
379 observed cell capacities. However, when each cell is set to a standard capacity of 150 mAh/g in our cost
380 models, the remaining differences in cost between the various cells reflect only the underlying disparities
381 in cell materials and manufacturing. For instance, while the LGPS-argyrodite cell is, nominally, more
382 costly than the LGPS-acid cell, when both cells are set to the same, hypothetical benchmark capacity, the
383 cost of the LGPS-argyrodite cell becomes lower than that of the LGPS-acid cell. This is because treatment
384 with phosphoric acid is a costlier solution to anode interfacial chemical stability than the use of a thin
385 argyrodite interlayer, due to both higher materials costs as well as higher processing costs. Despite this,
386 because the LGPS-acid cell outperforms the LGPS-argyrodite cell, the total cost of the LGPS-acid cell is,
387 in reality, lower. In other words, while it uses a costlier interfacial modification process (see Fig. S1(b)),
388 the LGPS-acid cell captures enough additional cell performance to ultimately offset this cost premium and
389 produce the more cost attractive cell design. Crucially, this underscores the need to consider the materials
390 and processing costs of the cell as well as the literature reported performance that results from those specific
391 materials and processing choices when evaluating battery scalability.

392 These examples illustrate a critical tradeoff in cell design: while it is possible to employ materials and
393 process solutions that increase cell capacity, it is also possible that the additional materials and processing
394 costs to incorporate those solutions can, in fact, offset any captured gains in cell performance. Both factors
395 must be considered and balanced. In applying solutions to the challenges that currently plague solid-state
396 batteries, gains in performance must far outstrip attendant gains in manufacturing cost.

397 Fig. 3c and 3d present this central tension with two contrasting examples. In the first, data is taken from
398 the literature report of an LFP | LLZO | Li cell that uses a sputtered Sn interlayer to decrease the interfacial
399 resistance between the lithium metal anode and the garnet electrolyte³⁶ (referred to hereafter as the “LLZO-
400 Sn” cell). Upon insertion of Sn into the cell stack, the resultant increase in cell capacity (+16 mAh/g)
401 decreased total cell cost by \$16/kWh (cost reductions are indicated in green). This cost savings, however,
402 is ultimately offset by the cost to deposit the Sn interlayer itself (~\$23/kWh, cost increases are indicated in
403 red). In our model, the Sn cost is especially conservative, reflecting merely the price of tin metal and not
404 the added cost of fabricating it into a sputtering target. Thus, while inclusion of the Sn interlayer increased
405 the performance of the cell, the associated cost savings were ultimately negated by the high cost of the
406 materials and processing to achieve those performance gains. Our model LPSCI cell offers a contrasting
407 example. Upon the addition of just 1 wt% binder to the composite cathode, an increase in performance of
408 approximately 65 mAh/g was observed. This represents a cost reduction of \$309/kWh. The cost to
409 implement this change, however, amounted to less than \$1/kWh, yielding a total net reduction in cost of
410 approximately \$308/kWh, more than 99% of the cost savings captured by the increase in cell capacity.
411 These examples illustrate that it is often not clear or intuitive how materials choices will impact overall cell
412 cost. Seemingly promising design choices and interfacial mitigations that increase cell performance can be,
413 practically speaking, negated by their high manufacturing costs. On the other hand, even simple design
414 choices—such as the use of an appropriate binder to enhance interfacial contact between cathode and
415 electrolyte particles—can provide an extremely low cost pathway to large capacity gains that, in turn, yield
416 considerable reductions in total cell cost.



417
 418 **Fig. 4. Consequences of materials choice**
 419 Manufacturing cost of Ag-C and LPSCI model cells as well as comparisons between the two cells for several key
 420 underlying consequences of the disparity in anode materials selection. Specific capacity is based on active material
 421 mass in the cathode.
 422

423 Finally, in Fig. 4, we illustrate multiple consequences of a difference in anode materials selection and how
 424 they ultimately contribute to overall manufacturing cost. Here we compare our previous LPSCI model cell
 425 with another example based on the recent report of a cell (also utilizing an $\text{Li}_6\text{PS}_5\text{Cl}$ electrolyte) in which
 426 excess Li foil is instead replaced by a thin Ag-C nanocomposite layer¹¹⁴ (referred to hereafter as the “Ag-
 427 C” cell). This report serves as a valuable, additional example of the key performance-processing tradeoffs
 428 under discussion here while using a contemporary, high performing, large format pouch cell fabricated with
 429 traditional battery and fuel cell slurry coating processes similar to those in our model. The total
 430 manufacturing cost of both cells is remarkably similar, despite numerous key underlying disparities that
 431 result from this difference in anode materials choice. For example, while both cells utilize $\text{Li}_6\text{PS}_5\text{Cl}$
 432 electrolyte and Ni-rich NMC cathode active materials, the Ag-C cell contains no excess lithium foil. The
 433 \$24/kWh cost savings captured by the lack of lithium, however, is exceeded by the \$34/kWh materials and
 434 processing cost of incorporating a Ag-C anode layer in its place. Moreover, both batteries can accommodate
 435 a similar number of galvanic cells in their cell stacks, a consequence of their comparable free volume
 436 requirements and the relatively small difference in thickness between the Ag-C anode layer and the excess
 437 lithium foil used in the LPSCI cell. Critically, however, the higher specific capacity exhibited by the Ag-
 438 C cell (due, in part, to a slightly higher cathode active material loading and the use of applied pressure
 439 during cycle testing) serves to offset the higher cost of the Ag-C layer as a replacement for traditional
 440 lithium foil. It is important to note here that the literature reported capacities for these two cells were
 441 recorded at different testing conditions and are, therefore, not strictly comparable. While the Ag-C cell was
 442 cycled at 0.5C, likely resulting in lower capacities than would otherwise be recorded at lower C-rates, the
 443 cell was also tested at elevated temperature. Similarly, while the performance of the LPSCI cell was
 444 measured at a lower rate (0.05C), it was tested at a comparatively lower temperature as well. Thus, while
 445 the capacities of these two cells aren’t directly comparable, they are included here to illustrate how the
 446 numerous, sometimes divergent, effects of even a single materials choice (in this case, the anode) must be
 447 collectively weighed. Moreover, these capacities were measured at C-rates and temperatures with offsetting
 448 effects on cell performance.

449 This problem—finding relevant and diverse sets of “state-of-the-art” solid-state batteries with directly
450 comparable cell design and testing data—remained a persistent challenge throughout the course of our
451 analysis. In the absence of literature reports with equivalent or controlled cell configurations, electrode
452 compositions, component thicknesses, and cycling conditions, manufacturing costs calculated from these
453 specific, literature reported cell design and performance data cannot be meaningfully compared across
454 reports. This reflects a much broader concern in the battery literature: the lack of standardization in test
455 cell design and characterization protocols remains a significant barrier to accurate, widespread, and
456 insightful analysis of battery cost, manufacturability, and scalability. Without the ability to directly
457 compare cell performance (i.e., performance captured under equivalent testing conditions) as well as the
458 resulting costs of those cells across different reports, researchers are ill equipped to discern which materials
459 and process solutions are truly promising and worthy of subsequent attention. Greater benchmarking and
460 standardization^{115–117} would serve the community well.

461 Final Thoughts

462 We end by reviewing the combined effects of the three factors we’ve discussed in this Perspective: (1) the
463 availability and scaling capacity of the chosen materials’ constituents, (2) the cost of processing the chosen
464 materials into full cells at high volume and yield, and (3) the performance that may be practically extracted
465 from those materials. Take, for example, Fig. 3d, which presents the relative effects of these three factors
466 for our model LPSCl cell. As already discussed, the cost impacts of a materials choice (in this case, a
467 binder) are presented. Use of the binder significantly increased cell capacity while the materials and
468 manufacturing costs for its incorporation were marginal. Moreover, by constructing and modeling a process
469 flow in which the cell layers are calendared, we’ve already embodied a key aspect of the mechanical
470 properties that drive sulfide solid electrolyte processing. Finally, to integrate the effects of materials
471 availability, we illustrate how the price volatility of cobalt impacts cell cost. Since the $\text{Li}_6\text{PS}_5\text{Cl}$ electrolyte
472 doesn’t appear to suffer from any considerable materials availability constraints, we examined the
473 sensitivity of the NMC cathode to its constituent metals. For example, an increase in cobalt price
474 approximately equal to recent cobalt price volatility⁶⁸ yielded an increase in our NMC cost of less than
475 \$1/kg, an expected result given that prior work has already illustrated the relative insensitivity of Ni-rich
476 NMC to the price of cobalt⁸⁰. This small increase in NMC price increased total cell cost by ~\$1.50/kWh.
477 Ultimately, when the collective effects of these drivers were considered, gains in performance were found
478 to far outweigh the attendant materials and manufacturing costs or the possible cost increases due to
479 materials availability constraints.

480 Scrutiny of any of these elements in isolation would have failed to capture the comprehensive understanding
481 necessary to inform scale up. Low cost and high performance cells might fail to scale if materials supply
482 chains are severely and unpredictably constrained. Likewise, high performance cells made from readily
483 available materials might fail to scale if the properties of those materials require that costly or challenging
484 manufacturing processes be used during cell integration. Collective examination of the numerous,
485 sometimes conflicting, consequences of materials choice, for both the electrolyte and its interfacial
486 mitigations, is necessary to accurately weigh the various key tradeoffs that are likely to drive the
487 manufacturing and scale up of solid-state batteries.

488 While the present analysis makes clear the importance of using low cost materials and processes to capture
489 large gains in cell performance, the practical realization of this strategy may be challenging. It is not clear
490 whether the requisite ore processing and refining capacity will be able to scale at historically unprecedented
491 rates, or whether historical and availability driven volatility in the underlying materials prices will continue.
492 Further complicating matters, without significant additional increases in cell capacity over what has already
493 been reported in the literature, materials supply and growth requirements will remain historically

494 unprecedented and possibly unattainable. Higher cell energy density serves to reduce not only the overall
495 manufacturing costs for each kWh produced, but critically, it also reduces the mass of material required to
496 produce each kWh (i.e., the materials intensity). It is therefore not sufficient to focus solely on increasing
497 materials supply or on enhancing cell capacity, these drivers are coupled and must be jointly improved.

498 Despite these challenges, recently published work offers several promising pathways that align with aspects
499 of our current analysis. Higher voltage charging of current NMC and LCO cathode materials enabled by
500 high-voltage-stable electrolytes or coating materials¹¹⁸, novel cathode chemistries with higher energy
501 content¹¹⁹, and higher loading densities in the composite cathode¹²⁰ could significantly boost the attainable
502 specific capacities. Moreover, if such materials can be incorporated into full battery cells inexpensively—
503 for instance, through rapid and low cost processing routes¹²¹—practical, large scale manufacturing of all-
504 solid-state batteries could indeed be realized.

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