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Anode-Side Strategies for Raising the Energy Density of Solid-State Lithium Metal Batteries: A Cell-Level Quantitative Assessment

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Abstract. A solid electrolyte allows lithium metal to replace the graphite host of a conventional cell. Lithium metal promises a higher energy density. Published work on the anode side rarely converts that promise into the energy density of a complete cell. This paper builds a single-layer cell model that converts anode-side design choices into gravimetric and volumetric energy density. A reduction of the lithium foil from 50 to 20 micrometres raises volumetric energy density by 19.5 % and gravimetric energy density by 3.5 %. Removing the foil entirely raises them by 37.3 and 6.0 %. Excess lithium costs stack thickness rather than stack mass. An interlayer that enables this reduction breaks even at a thickness equal to the lithium it removes, which is 30 micrometres in the reference case. A 5 micrometre interlayer retains 15.7 of the 19.5 percentage points available. The volumetric gain has a closed form containing only thicknesses, so no assumed density enters it. The same cell draws 4 milliamperes per square centimetre at a one-hour charge rate and 8 at a half hour rate, which sets the current an anode interface must sustain. The practical target is an interlayer far thinner than the lithium it removes, combined with a threshold current above the demand of the cell.

Keywords: Solid-state battery, lithium metal anode, energy density, interlayer, critical current density

1. Introduction

A solid electrolyte allows a cell to carry lithium metal in place of a graphite host. Lithium metal stores 3860 milliampere hours per gram of charge, while graphite stores 372. The host material disappears from the stack, and the anode then occupies less space for the same charge. Volume rather than mass constrains a vehicle battery pack, so the expected benefit of this change is mainly a volume benefit.

The anode side of a solid-state cell limits how much of that benefit survives. A cell carries more lithium than one cycle consumes, because three failure modes remove lithium or reduce the working area. A lithium filament nucleates inside the electrolyte and grows until it bridges the two electrodes. A void form at the interface during stripping, because lithium leaves the surface faster than creep

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replenishes it [1]. Contact is then lost over part of the area, and the remaining area carries a higher local current. Extra lithium delays the point at which these losses end the life of the cell [2].

That extra lithium is inactive material. It occupies volume and mass in the stack but stores no charge beyond what the cathode can supply. The ratio of anode capacity to cathode capacity, the N/P ratio, measures the excess. One micrometre of lithium foil holds 0.206 milliampere hours per square centimetre, so a 50 micrometre foil paired with a 4 milliampere hour per square centimetre cathode gives an N/P ratio of 2.58.

Reported strategies for cutting the excess fall into three methods. The first reduces the lithium inventory directly, through a thinner foil or a cell assembled without an anode. The second places a thin layer between the lithium and the electrolyte. The third replaces the flat foil with a three-dimensional host or an alloy. Interlayers are described by the problem each one solves. A lithiophilic layer improves wetting, so that lithium spreads over the electrolyte instead of touching it at isolated points [3]. An electron-blocking layer raises the electronic resistance of the interface and suppresses the reduction of lithium ions inside the solid. A chemically stable layer converts on contact with lithium into a product that blocks electrons rather than conducting them [4]. Reported interlayer thicknesses span nanometres to tens of micrometres[5].

Two quantities dominate the evaluation of these strategies. The area specific resistance, measured by impedance spectroscopy, averages the interfacial resistance over the contact area. The critical current density marks the current above which the cell shorts, and the author of each study chooses the criterion that defines that failure [6]. Both quantities are sensitive to conditions that studies often leave unstated. Fabrication pressure and contact hold time change the reported critical current density [7]. Stack pressure and temperature change it again [8]. A symmetric cell and a full cell built from the same materials do not agree, because the cathode alters the stack pressure as it cycles [9]. A filament can also melt under its own heat, so a short appears and disappears while the cell seems to cycle normally [10]. The threshold therefore follows the test protocol as much as the interface [11], and cumulative areal capacity has been proposed to replace it [12].

Neither quantity states what a cell of a given areal capacity actually demands, and neither converts an anode-side strategy into cell-level energy density. This paper builds a transparent single-layer cell model and uses it to answer three questions. The first question asks how much energy density a reduction in lithium excess delivers, and how that gain divides between mass and volume. The second asks how thick an interlayer may be before its own thickness cancels the gain it enables. The third asks what current density an anode interface must sustain to serve a practical cell. The scope covers inorganic solid electrolytes and lithium metal anodes.

2. Methodology

2.1. Structure cell model

The model defines a single-layer stack per unit of electrode area, in the manner of a published benchmarking study of all-solid-state cells [13]. The stack holds a composite cathode, a solid electrolyte separator, an optional interlayer, a lithium foil, an aluminium current collector and a copper current collector. Areal energy follows from the cathode areal capacity and the average discharge voltage,

$$E = Q * V \tag{1}$$

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Where E is a real energy in milliwatt hours per square centimetre, Q is cathode areal capacity in milliampere hours per square centimetre and V is average discharge voltage in volts. The cathode is sized by the capacity it must deliver rather than by an assumed thickness. Its composite mass per unit area and its thickness follow as:

$$m_c=\frac{Q}{q_c*f_c},\ t_c=\frac{m_c}{\rho_c} \tag{2}$$

Where q_c is the specific capacity of the active material, f_c is the active mass fraction and ρ_c is the density of the composite. Every other layer is specified by thickness, so the mass per unit area of layer i is:

$$m_i=t_i*\rho_i \tag{3}$$

Gravimetric energy density and volumetric energy density follow by dividing the areal energy by the total mass and the total thickness of the stack,

$$w = \frac{E}{\sum m_i} \tag{4}$$

$$v = \frac{E}{\sum t_i} \tag{5}$$

Lithium thickness converts to capacity through the density and the theoretical specific capacity of lithium.

$$q_{Li} = t_{Li}*\rho_{Li}*C_{Li} \tag{6}$$

which gives 0.206 milliampere hours per square centimetre for each micrometre of foil. The ratio of anode capacity to cathode capacity follows as q_{Li} divided by Q. Areal energy does not change when only the anode is altered, so the volumetric gain from removing a lithium thickness dt, the relative gain in volumetric energy density, g has the closed form:

$$g = \frac{\Delta t}{\sum t_i - \Delta t} \tag{7}$$

where the sum runs over the stack before the removal.

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2.2. Assumptions and model boundary

Table 1 lists every parameter with its value and its origin. The results in Section 3 follow from that table alone. The model omits the packaging, the tabs and the repetition of layers in a real cell, so absolute values are not predictions for a pack. Every result below is a relative change between two anode configurations, which is insensitive to the omitted terms. The cathode areal capacity and the electrolyte thickness are swept, because both alter the share of the stack that the anode occupies. A published cell-level assessment of lithium requirement provides the external check on the model [14].

Table 1. Model parameters, values and origins

Parameter	Value	Unit
Lithium density	0.534	g * cm^{-3}
Lithium specific capacity	3860	mAh * g^{-1}
Cathode areal capacity	4	mAh * cm^{-2}
Average discharge voltage	3.8	V
Cathode active specific capacity	200	mAh * g^{-1}
Cathode active mass fraction	0.7	-
Cathode composite density	3.4	g * cm^{-3}
Electrolyte thickness	30	μm
Electrolyte density	1.9	g * cm^{-3}
Aluminium collector thickness	12	μm
Copper collector thickness	8	μm
Lithium foil thickness	0 to 50	μm
Interlayer thickness	0 to 50	μm
Interlayer density	4	g * cm^{-3}

3. Results and discussion

3.1. The reference cell and the effect of lithium excess

Table 2. Stack composition of the reference cell at two lithium foil thicknesses

Component	Thickness / μm	Mass / mg cm^{-2}
Composite cathode	84.0	28.57
Solid electrolyte	30.0	5.70
Aluminium collector	12.0	3.24
Copper collector	8.0	7.17
Lithium foil, 50 μm	50.0	2.67

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Table 2. (continued)

Total with 50 µm foil	184.0	47.35
Lithium foil, 20 µm	20.0	1.07
Total with 20 µm foil	154.0	45.75

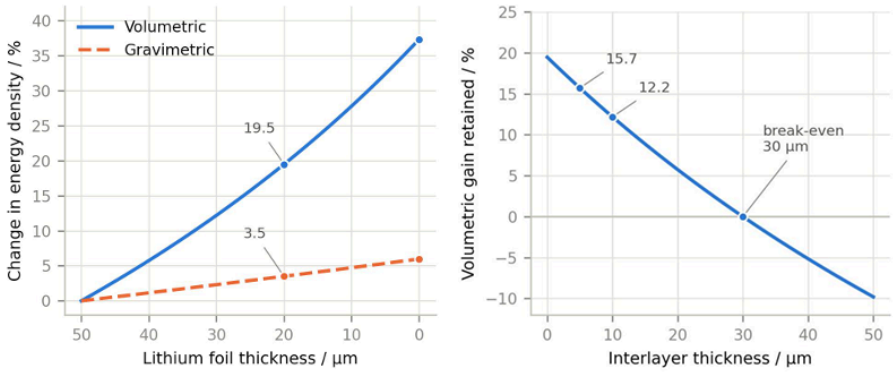
Table 2 gives the stack of the reference cell. The cathode delivers 4 mAh cm^{-2}, which calls for 20.00 mg cm^{-2} of active material and 28.57 mg cm^{-2} of composite by equation 2. That composite is 84.0 micrometres thick. The cell delivers 15.2 mWh cm^{-2} of areal energy by equation 1. A 50 micrometre foil brings the stack to 47.35 mg cm^{-2} and 184.0 micrometres, which gives 321.0 Wh kg^{-1} and 825.9 Wh L^{-1} by equation 4 and 5. A 20 micrometre foil brings the stack to 45.75 mg cm^{-2} and 154.0 micrometres, which gives 332.3 Wh kg^{-1} and 986.8 Wh L^{-1}.

3.2. Lithium excess and interlayer thickness

A reduction in the lithium foil raises volumetric energy density far more than gravimetric energy density. The reduction from 50 to 20 micrometres raises volumetric energy density by 19.5% and gravimetric energy density by 3.5%. Removing the foil entirely raises them by 37.3 and 6.0 %. Figure 1a shows both quantities across the range.

The cause appears directly in Table 2. The 30 micrometres of lithium removed weigh 1.60 milligrams per square centimetre, which is 3.4% of the stack mass. The same layer is 16.3% of the stack thickness. Excess lithium costs thickness rather than mass, and the asymmetry in Figure 1a follows from that contrast.

An interlayer occupies thickness of its own, so it returns less than the full gain. Figure 1b shows the volumetric gain retained against interlayer thickness, for an interlayer that allows the foil to fall from 50 to 20 micrometres. A 1 micrometre interlayer retains 18.7 of the 19.5 percentage points available. A 5 micrometre interlayer retains 15.7 points, and a 10 micrometre interlayer retains 12.2 points. The retained gain reaches zero at 30 micrometres, a thickness equal to the lithium removed. An interlayer thicker than the lithium it replaces lowers the energy density of the cell.



(a) Energy density against lithium foil thickness

(b) Volumetric gain retained against interlayer thickness for a foil reduced from 50 to 20 µm

Figure 1. Energy density of the reference cell relative to a 50 µm lithium foil

This break-even criterion ranks the three types of strategy. Vapour-deposited and formed interlayers occupy nanometres to a few hundred nanometres, two orders of magnitude below the lithium they allow a designer to remove, so almost the whole gain survives. Three-dimensional hosts

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and thick alloy layers occupy tens of micrometres and approach the break-even thickness, so part of the gain pays for the strategy itself.

3.3. The current density an interface must sustain

A cell demands a current density set by its areal capacity and its charge rate. The reference cell of 4 milliampere hours per square centimetre draws 4 milliamperes per square centimetre at a one-hour rate and 8 milliamperes per square centimetre at a half hour rate. Figure 2 plots this demand against areal capacity for three charge rates.

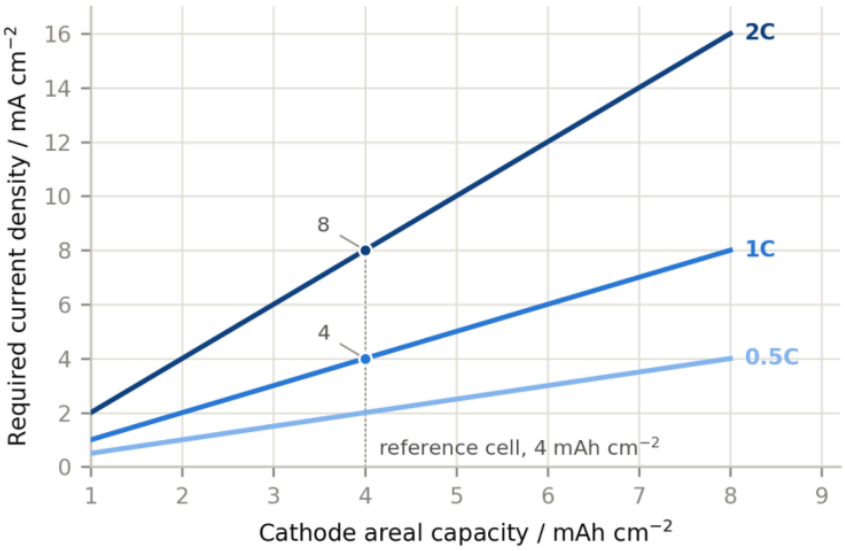


Figure 2. Current density required by a cell against its cathode areal capacity at three charge rates

This demand is the quantity a reported threshold should be measured against, and published reports of anode interfaces rarely state it. A threshold quoted on its own cannot be read as sufficient or insufficient, because the areal capacity and the charge rate of the intended cell are not given.

The comparison is obstructed from the other side as well. A reported threshold rises with stack pressure and with temperature [7, 8], so a study that omits either condition leaves a reader unable to transfer the value to a cell operated at ambient temperature and moderate pressure. A threshold measured in a symmetric cell transfers no better, because the cathode of a full cell alters the stack pressure as it cycles [9]. A dynamic soft short can also pass for continued cycling, which places a reported threshold above the true one [10]. Reported values and cell demands therefore sit on the same axis in Figure 2 without yet being comparable [11].

A low reported threshold therefore has two possible causes. The interface may genuinely fail below the demand of a practical cell. The interface may instead clear the demand under conditions the report does not state. The threshold alone does not separate the two cases. The comparison in Figure 2 becomes direct once a report gives the stack pressure, the temperature and the areal capacity of the intended cell alongside the threshold.

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3.4. Sensitivity and outlook

Equation 7 explains how the volumetric benefit varies with cell design. The benefit equals the removed lithium thickness divided by the thickness of the remaining stack, so any design that thins the rest of the stack raises it. A cathode areal capacity of 2 milliampere hours per square centimetre thins the cathode and lifts the same reduction from 19.5 to 26.8 %. An electrolyte 250 micrometres thick lowers it to 8.0 %. The volumetric result carries no dependence on the density of any layer, because equation 7 contains only thicknesses.

This dependence sets where anode-side work pays. A thick pressed-pellet electrolyte dominates the stack, so trimming the lithium changes little. A thin supported electrolyte leaves the anode as a large share of what remains, and the same trimming returns three times as much.

The trend also sets the direction of travel. Solid electrolyte layers are reported at ever smaller thicknesses as processing improves, and every micrometre removed from the separator raises the value of a micrometre removed from the anode. A thinner electrolyte and a thinner anode each raise the value of the other. An interlayer thin enough today stays thin enough as the stack shrinks, whereas a host that is affordable in a pellet cell will not survive the move to a thin-film stack. A design rule follows. An anode strategy should be judged by its thickness relative to the lithium it removes, and that ratio should be reported alongside the electrochemical result.

4. Conclusion

In conclusion, anode-side optimisation of a solid-state lithium metal cell buys volume rather than mass. A reduction in lithium excess from an N/P ratio of 2.58 to 1.03 raises volumetric energy density by 19.5 % and gravimetric energy density by 3.5 %. An interlayer that enables such a reduction returns 15.7 of those 19.5 percentage points when it is 5 micrometres thick, and returns nothing at 30 micrometres. The volumetric benefit depends only on layer thicknesses, so it grows as solid electrolyte layers become thinner. The reference cell demands 4 to 8 milliamperes per square centimetre, and an anode interface must clear that threshold to serve it. Reported thresholds cannot yet be compared with that demand. Reports often omit the stack pressure and the temperature.

Three steps would extend this work. The model should next represent a multilayer pouch cell with packaging and tabs, so that the relative gains become pack-level figures. Sodium metal stores about half as much charge per micrometre of foil as lithium. Excess sodium therefore costs more thickness, and the same analysis should matter more for sodium metal cells. Threshold measurements at ambient temperature and moderate stack pressure would allow reported interfaces to be compared directly with the demand of a cell. Two recommendations follow. An anode interlayer should be judged by its thickness relative to the lithium it removes, not by its absolute thickness. A report of an interlayer should state the stack pressure, the temperature and the intended areal capacity alongside every threshold measurement.

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