Influence of geometrical manufacturing tolerances on lithium-ion battery performance

Russell Broad | Ashley Fly

Department of Aeronautical and Automotive Engineering, Loughborough University, Loughborough, UK

Correspondence
Ashley Fly, Department of Aeronautical and Automotive Engineering, Loughborough University, Loughborough, UK.
Email: a.fly@lboro.ac.uk

Summary
The manufacture of lithium-ion battery cells consists of multiple production processes, all of which have tolerances that can affect cell performance. For battery packs that contain 100s or 1000s of individual cells, ensuring consistency and minimising variation between cells is important for reliability and lifetime. This study uses a numerical battery model to examine the influence of electrode coating thickness, calendering and electrode cutting tolerance on capacity, energy, resistance and voltage relaxation. Results show that electrode cutting tolerance has the largest affect upon cell performance characteristics, with calendering tolerance predominantly affecting the voltage relaxation period. For the simulated cell, the negative electrode coating tolerance mainly affects the cell capacity and energy, whereas the positive electrode coating tolerance has the most significant effect upon voltage relaxation. Multiple simulations were conducted with random tolerances of a Gaussian distribution applied to all processes simultaneously to represent a production run of 1000 cells. The resulting distribution of cell performance values is compared with manufacture datasheets to determine the expected reject rate for different tolerance SDs. For a reject rate of zero, the maximum tolerance SD must be less than 1% but need not be less than 0.5% for the geometrical parameters considered. Findings show that the electrode slitting, cutting and negative electrode coating tolerances require tighter manufacturing tolerance than calendering and positive electrode coating thickness.

KEYWORDS
battery, lithium-ion, manufacturing, modelling, tolerance

1 | INTRODUCTION
In recent years, there has been an increased interest in the use of electric vehicles, with many governments committing to phase out the sales of petrol and diesel vehicles. Meeting the need for electric vehicles will require significant increases in battery production, with multiple ‘giga-factories’ planned to manufacture lithium-ion battery cells for electric vehicles and an estimated 2900 GWh worldwide annual cell production capacity in 2030.

The performance of the individual lithium-ion cells within a vehicle battery pack must be consistent so that...
the entire pack can operate optimally and utilise its full capacity without excess interference from the battery management system. However, the performance of cells within the battery pack will vary due to tolerances of the different steps in the manufacturing process. There are a number of production processes involved in the manufacture of a cell and battery pack, summarised in various studies.\textsuperscript{5,6} Whilst several different variations of manufacturing techniques exist for lithium-ion batteries, the most commonly utilised method is wet coating of the electrodes.\textsuperscript{7} In this method, the electrodes are manufactured by applying a coat of active material ‘slurry’ to either an aluminium or copper current collector surface. The wet slurry is then dried and subsequently undergoes a calendaring process which compresses the active material between a pair of rollers to achieve the desired thickness, density and porosity.\textsuperscript{5} The resulting sheet of electrode is slit to the required width, then cut to form the individual electrodes. The individual electrodes are then either layered or rolled before being packed into the cell casing (pouch, prismatic or cylindrical), filled with electrolyte and sealed. Each of these different processes will have an associated tolerance which will lead to cell-to-cell performance variations in the final manufactured cells. Understanding the relationship between production process tolerance and cell performance can help improve consistency between cells and avoid the use of unnecessarily stringent tolerances that could reduce production speed or increase cost.

The influence of production tolerance on lithium-ion battery manufacturing has been studied by several different researchers. Yourey\textsuperscript{8} studied the impact of electrode loading and calendaring tolerances through a simple theoretical model, showing both parameters had significant effect upon the electrode porosity and subsequent amount of electrode required. The impact of tolerances was increased for thinner electrodes, designed for high power, low energy applications. Meyer et al\textsuperscript{9} experimentally investigated the influence of different calendaring force and speed on the electrode porosity and pore size distribution in graphite and lithium nickel manganese cobalt oxide (NMC) electrodes. An et al\textsuperscript{10} experimentally evaluated the cell-to-cell capacity variation in 5473 commercial cells, showing a strong correlation between measured capacity and cell mass, indicating the variation is likely due to changes in the active material mass due to manufacturing tolerances. Baumböfer et al\textsuperscript{11} used a set of 48 commercial cells to show how initial cell-to-cell variation due to manufacturing tolerances increased as the cells underwent cyclic ageing.

Several authors have also utilised numerical models to evaluate the sensitivity of manufacturing tolerances on the electrochemical performance. Schmidt et al\textsuperscript{12} used a multi-level model approach to link process parameters to battery model parameters then electrochemical properties through a pseudo-two-dimensional (P2D) model, considering discharge capacity, energy density and power density. This approach was then later expanded by Thomitzek et al\textsuperscript{13} to consider manufacturing costs and battery cell revenue, allowing targeted investment in improving manufacturing equipment. Laue et al\textsuperscript{14} utilised model-based uncertainty quantification, combined with a P2D electrochemical cell model to investigate cell-to-cell variation at different C-rates, demonstrating an increased capacity spread at higher C-rates. Recently, Lombardo et al\textsuperscript{7} produced an open-source online tool, the ‘ARTISTIC online calculator’ for linking manufacturing process to electrochemical performance, allowing users to specify different slurry, drying, calendaring and electrolyte impregnation parameters.

Variations in the calendering process were examined in the study by Kwade et al\textsuperscript{16} where the electrode pore size and distribution were exponentially correlated to the calendering line load. Kenney et al\textsuperscript{15} simulated the impact of electrode manufacturing tolerances of individual cells in a battery pack of cells connected in series, where it was found that they had a significant effect upon initial capacity, as well as capacity fade of the pack. The results suggested that the electrode thickness and porosity had the most significant effect.

The majority of battery manufacturing tolerance studies focus on the impact on cell capacity and energy. Whilst this is a key performance metric, at low C-rates, capacity is predominantly linked to the mass of active material and does not fully consider the influence of manufacturing tolerances on resistance and transient behaviour which will have an impact on performance at the pack level. For cells connected in series, a cell with a lower capacity will affect the entire series string to avoid over charging or discharging of the weaker cell.\textsuperscript{5} Variations in the ohmic resistances of cells in series strings will affect the overall voltage available from the string, as well as an imbalance in heat distribution between the cells for the common load current. For cells connected in a parallel arrangement, variations in ohmic resistance between the cells can cause current imbalances during charge (or discharge) and for variations in capacity, steady state current imbalances can occur.\textsuperscript{16} These current imbalances could cause premature cell ageing or present a potential safety issue, should an overcurrent condition occur for an individual cell. It is therefore important to consider the dynamic nature of these variations to understand the issues faced when cells are built into a battery pack for use in a real application.

In this paper, the influence of manufacturing tolerances from electrode coating thickness, calendaring and
electrode cutting on the performance of a lithium-ion cell are studied through the use of a numerical model parameterised around a commercial cell. These geometrical tolerances were chosen as they represent key components of the manufacturing process and can be reliably represented in battery numerical models. Section 2.1 introduces the numerical model and study methodology and Section 2.2 conducts a sensitivity analysis on individual production process tolerances for each electrode and different cell chemistries. Section 2.3 simulates a production scenario, where a gaussian distribution is applied to different process tolerances simultaneously for a range of SD values. The resulting distribution of the cell performance is determined and the reject rate is calculated for the desired upper and lower performance limits.

It is anticipated this work will provide a methodology to help inform the allocation of resources to manage specific process tolerances, allowing improvement in the sensitive tolerances, whilst avoiding wasting time, effort and cost in improving those that have no detrimental effect. As this work also determines cell transient response, the effects of process tolerances upon battery packs due to variations in internal current flow can be considered. In addition, by including a Gaussian distribution for combinations of process tolerances, the distribution of the performance parameters can be compared to allow management, rather than elimination of the process tolerances, to achieve the desired production reject rate.

### Table 1: A summary of the governing equations of the P2D model\textsuperscript{18}

| Description       | Governing equations                                      | Boundary conditions                                      |
|-------------------|----------------------------------------------------------|----------------------------------------------------------|
| **Solid phase**   |                                                          |                                                          |
| Conservation of mass | \[ \frac{\partial c_s}{\partial t} - \frac{D_s}{r^2} \left( \frac{\partial}{\partial r} \frac{r^2 \frac{\partial c_s}{\partial r}}{\partial r} \right) \] | \[ \begin{align*} \frac{\partial c_s}{\partial r} &= 0, & r &= 0 \\ \frac{\partial c_s}{\partial r} &= \frac{j}{\sigma_s}, & r &= r_p \end{align*} \] |
| Conservation of charge | \[ \nabla \cdot (\sigma_s^{\text{eff}} \nabla \Phi_s) = a_j \] | \[ \begin{align*} \nabla \cdot (\sigma_s^{\text{eff}} \nabla \Phi_s) &= 0, & x &= L^- \\ \nabla \cdot (\sigma_s^{\text{eff}} \nabla \Phi_s) &= I, & x &= L^- + L^+ \end{align*} \] |
| **Electrolyte phase** |                                                          |                                                          |
| Conservation of mass | \[ \frac{\partial c_e}{\partial t} = \nabla \cdot (D_e^{\text{eff}} \nabla c_e) + \frac{1}{\sigma_s^{\text{eff}}} a_j \] | \[ \begin{align*} \frac{\partial c_e}{\partial x} &= \frac{2a_j}{\sigma_s^{\text{eff}}}, & x &= 0 \\ \frac{\partial c_e}{\partial x} &= 0, & x &= L^- + L^+ \end{align*} \] |
| Conservation of charge | \[ \nabla \cdot (\sigma_e^{\text{eff}} \nabla \Phi_e) + \nabla \cdot [\sigma_e^{\text{eff}} \nabla \ln (c_e)] = -a_j \] | \[ \begin{align*} \nabla \cdot (\sigma_e^{\text{eff}} \nabla \Phi_e) &= 0, & x &= L^- + L^+ \end{align*} \] |
| **Reaction kinetics** |                                                          |                                                          |
| Butler-Volmer | \[ j = k (c_e)^{a_j} \left( c_{e,\text{max}} - c_e \right)^{a_j} \left( c_{e,\text{min}} \right)^{a_j} \left\{ \exp \left[ \frac{a_j}{\sigma_s^{\text{eff}}} \left( \eta - \frac{R_{\text{max}}}{a_j} \right) \right] - \exp \left[ \frac{a_j}{\sigma_s^{\text{eff}}} \left( \eta - \frac{R_{\text{min}}}{a_j} \right) \right] \right\} \] |                                                          |
| Overpotential | \[ \eta = \Phi_s - \Phi_e - U \] |                                                          |

### 2.1 Numerical model

The P2D Doyle-Fuller-Newman (DFN) model\textsuperscript{17} was used in this work because of its ability to represent the different manufacturing tolerances through modifying simulation inputs and reflect the corresponding variation in capacity, energy, resistance and voltage relaxation. The governing equations and boundary conditions of the P2D model\textsuperscript{18} are illustrated in Table 1 (see Table 2 for nomenclature).

The open-source python package “PyBaMM”\textsuperscript{19,20} was used for numerical simulations\textsuperscript{19,20} with the baseline parameter set for a commercial cylindrical 21700 Ni\textsubscript{0.8}Mn\textsubscript{0.1}Co\textsubscript{0.1}O\textsubscript{2} (NMC811)/Graphite + Si (LGMS50\textsuperscript{21}) cell from Chen et al.\textsuperscript{22}

Six separate manufacturing processes were considered in this study; electrode height, electrode width (ie, jelly-roll length), positive electrode coating thickness, negative electrode coating thickness, positive electrode calendering and negative electrode calendering. These parameters were selected based on their ability to be represented within the P2D model input parameters and link to physically measurable variables during the production process such as lengths and thicknesses. There are many other important process tolerances within battery production process, such as material refining, slurry mixing, drying,
and electrolyte filling,\(^6\) however, these cannot be directly represented in the parameters of the P2D model without detailed models of the manufacturing processes themselves. Misalignment of the electrodes during assembly is replicated from the geometrical tolerances in electrode height and width but the effect of electrode overhang\(^2\) is not considered. The P2D model runs in an isothermal condition, so heat generation or heat transfer is not simulated. This study assumes that the production processes are operating correctly and does not analyse the effect of process quality issues or failure conditions. The influence of tolerances on the ability to manufacture the cell are also not considered. For example, it is assumed that the electrodes can still fit in the cell can, despite changes in the length and thickness.

A single production process can affect multiple P2D model parameters. For example, tolerances in the coating process will cause variations in the volume of active and non-active material formed in the electrode. When the electrode is subsequently passed through the calendering process, any excess material will still be calendered to the correct thickness. This will result in the pores of the material being over compressed to accommodate the additional material. A positive coating tolerance will result in the following cell structural property changes:

1. An increase in the electrode active material volume.
2. No change in the electrode thickness after the subsequent calendering process.
3. A reduction in the electrode porosity post-calendering.

The porosity reduction due to over-calendering can be determined by considering all the active material fractional parts, where \(\varepsilon_e = \text{porosity (electrolyte fraction by volume)}\), \(\varepsilon_s = \text{active material fraction by volume} \) and \(\varepsilon_n = \text{non-active material fraction by volume} \). By definition, all of the fractional parts add up to 1, so rearranging the terms:

\[
1 - \varepsilon_e = \varepsilon_s + \varepsilon_n,
\]

if the slurry coating thickness is over-applied with tolerance of \(t\%\), then any extra thickness will be subsequently calendered to the correct size, resulting in the electrode porosity being reduced to accommodate the additional active and non-active material. The resulting active material fraction \(\varepsilon_{s1}\), non-active material fraction \(\varepsilon_{n1}\), and material porosity \(\varepsilon_{e1}\) can be calculated as:

\[
\varepsilon_{s1} = \varepsilon_s \left(1 + \frac{t}{100}\right), \quad \varepsilon_{n1} = \varepsilon_n \left(1 + \frac{t}{100}\right),
\]

\[
\varepsilon_{e1} = 1 - \varepsilon_{s1} - \varepsilon_{n1}.
\]

Therefore, the resulting porosity \(\varepsilon_{e1}\) becomes:

\[
\varepsilon_{e1} = 1 - \left(1 + \frac{t}{100}\right) (\varepsilon_s + \varepsilon_n).
\]

This can be put in terms of the nominal porosity value \((\varepsilon_e)\) as shown in Equation (1)

\[
\varepsilon_{e1} = 1 - \left(1 + \frac{t}{100}\right) (1 - \varepsilon_e), \quad (1)
\]

The same equation can be used for the calendering process tolerances but in this case, the amount of active material has remained constant but the electrode volume has been affected by the calendering tolerance. It should be noted that for negative calendering tolerances, this equation will cease to apply if the tolerance is of such magnitude to prevent any actual calendering to occur.
The relationship between the six manufacturing processes and P2D model parameters is summarised in Table 3 and a diagram showing the electrode geometrical parameters is shown in Figure 1.

The model was simulated using 20 nodes for the positive electrode, negative electrode and separator, 30 nodes for the positive and negative particle radius and 10 nodes for the current collector. The CasADi solver option in PyBaMM was used with a relative tolerance of $1 \times 10^{-6}$. The setup of the virtual experiments is shown in Figure 2, which illustrates how the model was utilised.

For each process parameter, different tolerances ranging from $-5\%$ to $+5\%$ from the nominal value (Table 4) were simulated in $1\%$ steps to evaluate the trend in cell performance deviation over the tolerance range. The entire parameter set for the P2D model can be found in Table VII of Chen et al. For each tolerance value a constant current discharge test and pulse discharge test were simulated to analyse the steady state and dynamic performances, respectively. The constant current discharge consisted of a C/50 discharge based on the nominal cell capacity and was used to determine the capacity and energy of the cell. The pulse test consisted of a 200 s 1C discharge pulse followed by an 800 s relaxation period until the cell reached its lower voltage threshold of 2.5 V. Additional details of the simulation procedure are seen in Tables S1 and S2 and the code is available through the link in the acknowledgements. The simulated discharge voltage for variation of the electrode height parameter is shown in Figure 3 for tolerances ranging between $-5\%$ and $+5\%$ of the nominal value.

### 2.2 Data analysis

The performance change with tolerance was evaluated through the discharge capacity, energy and transient performance. Discharge capacity and energy were calculated directly from the C/50 constant current time and voltage data. For the pulse discharge, data were separated into individual pulses then the voltage response analysed to determine the ohmic resistance and rise time (voltage relaxation), shown in Figure 4. Ohmic resistance was measured from the magnitude of the voltage change from both the falling and rising edges of the voltage response as the current was applied and removed. Voltage measurements were taken 0.1 s after the current was applied and calculated using Ohm’s law as shown in Equation (2) where $R =$ cell ohmic resistance, $\Delta V =$ change in cell voltage, $\Delta I =$ change in cell current.

$$R = \frac{\Delta V}{\Delta I}. \quad (2)$$

The rest period rise time (voltage relaxation) was measured as the time taken for the cell voltage to reach

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**Table 3**  Relationship between positive tolerances in cell process parameters and P2D model parameters

| Process parameter tolerance percentage $t$ | Effect of process tolerance upon P2D model parameters |
|-------------------------------------------|-----------------------------------------------------|
| Electrode coating                         | $(\frac{t}{100} + 1) \times \varepsilon_s$          |
| Electrode calendering                     | $(\frac{t}{100} + 1) \times z$                       |
| Electrode slitting                        | $\varepsilon_s$                                      |
| Electrode cutting                         | $(\frac{t}{100} + 1) \times x$                       |
| Electrode porousity $\varepsilon_e$       | $1 - (1 + \frac{t}{100})(1 - \varepsilon_e)$        |

---

**Figure 1**  Simplified diagram of cell components showing geometrical parameters (not to scale)
67% of its final rest period voltage from a starting value of 0.1 s after the current was removed. This threshold was chosen by inspection of the results, so that a useful amount of the response time could be measured from the curve without passing the curve ‘knee’, where small variations in terminal voltage would have a large effect upon the measured time values. This is illustrated in Figure 4, where the start of the rise time measurement (diamond marker), final value (triangle marker) and 67% threshold (round marker) are shown for different electrode height tolerances. The selection of these values is designed to encompass the charge transfer and diffusion effects of the manufacturing tolerances studied.

2.3 Gaussian distribution of process tolerances

To investigate the potential interaction of multiple tolerances on cell capacity and energy, and to better replicate a realistic manufacturing scenario, the numerical model was modified to generate random tolerance values with a common Gaussian distribution. To obtain an acceptable level of distribution resolution, 1000 simulation runs were performed with random combinations of process tolerance values. An example

![Diagram showing setup of virtual experiments](image)

**TABLE 4** Nominal values of the P2D model parameters affected by process tolerances

| P2D model parameter                  | Nominal value | Units |
|--------------------------------------|---------------|-------|
| Negative electrode thickness         | 8.52e−05      | m     |
| Negative electrode porosity          | 0.25          | ...   |
| Negative electrode active material volume fraction | 0.75          | ...   |
| Positive electrode thickness         | 7.56e−05      | m     |
| Positive electrode porosity          | 0.335         | ...   |
| Positive electrode active material volume fraction | 0.665         | ...   |
| Electrode height                     | 0.065         | m     |
| Electrode width                      | 1.58          | m     |
distribution of input value tolerances is shown in Figure 5 for a SD of 1%.

The process was repeated for process tolerance SD values of 0.1%, 0.5%, 1%, 2%, 3%, 4% and 5% with the reject rate being calculated for each value using the threshold values shown in Table 5. The manufacturer’s datasheet values for the energy capacity showed a nominal value of 18.2 Wh and a minimum value of 17.6 Wh, which equated to a permitted variation of −3.3%. This variation was applied to both the nominal energy discharge and capacity discharge values that were obtained from the simulation using the parameter set of Chen et al to obtain lower and upper threshold values for the discharge capacity and energy capacity.

3 | RESULTS AND ANALYSIS

3.1 | Single tolerance sensitivity

The model described in Section 2.1 was used to simulate tolerance ranges for each individual parameter in isolation, ranging from −5% to +5% deviation from the nominal value in 1% increments. The relationship between the tolerance percentage value and the percentage variation of the cell performance value was plotted and a linear fit was first attempted using the least squares method. If the linear fit was considered poor ($R^2 < 0.99$) then a parabolic fit was used instead. For the parabolic functions, the largest magnitude of gradient over the tolerance range was found by using the coefficients from the fit line function to create a differential function to allow the gradient to be established for each tolerance value used, with the worst-case value taken as the measure of sensitivity (Equation 3). This approach allowed the sensitivity values for the process parameters to be directly compared.

$$\text{Sensitivity value} = \frac{\Delta \text{cell performance variation}\%}{\Delta \text{tolerance}\%}. \quad (3)$$

Figure 6 shows the influence of normalised electrode height variation on the cell discharge capacity. In this case a direct 1:1 ratio between electrode height variation and capacity was seen, since the electrode height linearly affects the active material volume of both electrodes.

Figure 7 shows the relative variation in cell discharge capacity and energy for the six different geometric tolerances considered for the LGM50 cell parameter set. Electrode height, width and negative electrode coating
thickness tolerance are seen to have the largest influence on discharge capacity, with a close to 1:1 relationship between parameter variation and capacity. The significant variation with negative electrode coating relative to the positive electrode implies the negative electrode is the limiting electrode in the cell.

Although the variation with negative electrode calendaring tolerance was insignificant, a parabolic characteristic can be observed in Figure 7. This was investigated further by analysing the effects of the underlying characteristics of the active material volume fraction and porosity parameters independently from the thickness parameter. It was found that these parameters had characteristics of opposite polarity but with close to a 1:1 magnitude, almost cancelling the effect of each other.

For the transient analysis, Figure 8A demonstrates how the variation of electrode height influences the cell’s ohmic resistance at different DOD, where DOD is...
calculated relative the cells actual capacity rather than the nominal cell capacity. Increasing electrode height is seen to decrease the cell's resistance since the cross-sectional area of the cell increases, both reducing cell resistance and current density. The variation is seen to increase at high DOD, as the rising resistance at high DOD means small variations in cell parameters have a more significant impact on resistance.

Figure 8B shows the variation in rise time (voltage relaxation) with changing positive electrode calendering at different DOD. Higher levels of calendering are seen to increase voltage rise time due to a reduction in the porosity of the electrode. The irregular variation in rest time change with DOD can be attributed to the variation in diffusivity of the active materials at different levels of lithiation. This can be seen by looking at the absolute values of rest period rise time in Figure 9 where the rise time varies significantly with DOD. This reflects the significant variation in diffusion coefficient measured through half-cell galvanic intermittent titration technique (GITT) analysis for the same cell's materials by Chen et al.22

The ohmic resistance and rise time results were analysed at low, medium and high DOD values of 20%, 50% and 80%, as shown by the red, black and blue dashed lines, respectively, in Figure 10A. A sensitivity value for each of these plots was obtained by finding the gradient of the respective fit lines. For non-linear variation, such as the calendering tolerance in Figure 10B, the steepest gradient was chosen.

The sensitivity results for the steady state discharge tests and step load pulse discharge tests are combined
and summarised using a heatmap, shown in Figure 11. For clarity, ohmic resistance values for the falling voltage edge at 50% DOD were used.

In Figure 11 it can be seen that tolerances in electrode height and width have a sensitivity value of 1.00 (% change of value per % variation in parameter) for discharge capacity and energy. This is simply due to these tolerances directly affecting the electrode size and the amount of active material contained within the electrodes. This results in a larger or smaller cell being constructed due to these tolerances, which also partly explains the ohmic resistance sensitivity of $-0.78$. It was found that this is also due to the DOD level of the cell. For the rise time sensitivity value of $-1.38$, it suggests that there is a significant strong negative correlation with increasing electrode size. The underlying data was reviewed and it was discovered that the rise time results for these process tolerances exhibited significant noise (Figure 12), reducing the confidence in these sensitivity values, although there was still a trend. This effect is thought to be due to the LGM50 cell positive electrode having a nickel rich composition of $Li_{0.833}Ni_{0.80}Mn_{0.08}Co_{0.08}Al_{0.04}O_2$, which causes variations in the voltage profile due to changes in the oxidation states for the transition metals within the positive electrode. As each tolerance value being simulated has a corresponding different charge capacity, the times at which the rise time measurements are made are at different DOD values, corresponding to different oxidation states and diffusion properties for the active material.

The negative electrode calendering tolerance has a negligible sensitivity value of $-0.10$ or less for discharge capacity and energy results. For the ohmic resistance value a sensitivity value of 0.44 was recorded. It was expected that the ohmic resistance would reduce with an increased calendering, due to the resulting reduced porosity causing an increase in electrical contact within the active material. This positive increase in ohmic resistance could be due to the increased tortuosity of the active material due to the reduced porosity caused by the over calendering. This would make it more difficult for the lithium to be extracted from the graphite structure of the negative electrode during the high current load used during the test, resulting in an increase in the electrode-electrolyte interface resistance. The rise time sensitivity of $-0.44\%$ per % suggests that the rise time reduces by a moderate amount as calendering increases. The underlying data was reviewed and was
The negative electrode coating tolerance showed sensitivity values of 0.98 and 0.91 for discharge capacity and energy, respectively, demonstrating a significant correlation with the negative electrode coating tolerance. It also suggests that the negative electrode is the limiting electrode within the cell. This is because an increase in this tolerance causes extra active material to be applied to the electrode with a resulting increase in discharge and energy capacity. After the calendering process, the additional material will result in further compression of the active material. This will reduce its porosity and explain why these sensitivity values are not unity values. With sensitivity values of $-0.18$ and $-0.17$ for ohmic resistance and rest period rise time, respectively, there is no significant effect of the negative electrode calendering tolerances. This indicates that the negative electrode is not the rate limiting electrode in the cell.

The positive electrode calendering tolerance was seen to have a negligible effect upon the discharge capacity and energy of less than $-0.01$. This suggests that the positive electrode is not the limiting electrode. For the ohmic resistance, the sensitivity value of $-0.16$ shows that negligible effect of the positive electrode calendering tolerances. The rest period rise time sensitivity value of $-0.51$ suggests there is a moderate effect of the positive electrode calendering tolerances.

not found to be excessively noisy, allowing confidence on the values. An explanation for this rise time reduction with increased calendering is that the thinner electrode caused by the over-calendering has reduced the distance required for lithium transport between the electrodes, requiring a reduced lithium concentration gradient for the same current and improved voltage relaxation.
The sensitivity values for the tolerances in positive electrode coating are 0.02 and 0.09 for the discharge capacity and energy, respectively, again suggesting the positive electrode already has sufficient active material and is not the limiting electrode. This result is unexpected, as a ratio of 1.1:1 is often used for amount of negative electrode active material relative to the positive electrode to avoid lithium plating. With a sensitivity value of −0.13, there is also no significant effect of these tolerances upon the ohmic resistance. The small reduction observed could be due to the additional active material particles being subject to additional compression after the calendering process, reducing the physical ohmic resistance between the particles, at the expense of the porosity. For the rest period rise time, the sensitivity value of −1.68 is very significant and the largest sensitivity value from all the results in Figure 11. One possible reason for this large sensitivity value is the combination of additional electrode active material and resulting over-calendering, requiring larger lithium concentration gradients ($\nabla c_l$) to drive diffusion compared to over-calendering alone without the additional active material.

The sensitivities shown in Figure 11 are specific to the simulation parameter set used, based on the LGM50 21700 cell. It must be noted that these cell parameters were obtained from the tear down, inspection and analysis of a single cell. The actual nominal parameters or the expected production tolerances of this cell are not publicly known and will be slightly different from those obtained.

To investigate how manufacturing sensitivities will change with different cells, the process was repeated for three additional parameter sets, shown in Figure 13. Figure 13A uses the parameter set from Marquis et al.26 for a cell having a negative electrode of graphite, a positive electrode of lithium cobalt oxide and a LiPF6 in EC: DMC electrolyte, Figure 13B the parameter set from Ecker et al27 for a Kokam SLPB 75106100 7.5 Ah high energy pouch cell, and Figure 13C the parameter set from Ramadass et al28 for an unspecified LiCoO2/graphite cell.

![Graph of Performance Characteristic Variation](image1)

![Graph of Performance Characteristic Variation](image2)

![Graph of Performance Characteristic Variation](image3)

**Figure 13** Sensitivity analysis for different cell parameter sets. (A) Marquis results summary.26 (B) Ecker results summary.27 (C) Ramadass results summary28.
The results in Figure 13 for the additional parameter sets all show that the discharge and energy capacity values have a sensitivity of 1.00 and for the electrode height and width variations for the same reasons explained for Figure 11. The ohmic resistance values also all have a sensitivity of approximately 1. The results show different limiting electrodes with Figure 13A,C showing a sensitivity correlation for the positive electrode coating tolerance variations, suggesting that the positive electrode is the limiting electrode, whereas Figure 13B suggests that the negative electrode is limiting. Figure 13C also shows very high sensitivity values of 8.48 and 10.65 for the rest period rise time, with increases in the positive electrode calendering and coating tolerances respectively. This demonstrates how cell design can influence the impact of manufacturing tolerances and also how tolerances associated to specific manufacturing tolerances can be targeted to minimise cell-to-cell variation, particularly those involving the limiting electrode.

3.2 Gaussian distribution of process tolerances

The simulated test results were used to determine the discharge and energy capacity values for each simulation run in the same way as Section 3.1 but with randomised tolerances of Gaussian distribution as discussed in Section 2.3. Figure 14 shows an example distribution from 1000 simulation runs for the 1% SD of individual process tolerances. The blue dashed vertical lines represent the positive and negative SD of the results and the red dashed vertical lines represent the lower and upper reject thresholds from Table 5. The relationship between the SD value and reject rate was then plotted for both discharge and energy capacity, shown in Figure 15.

From the example in Figure 14, the results for both the discharge capacity and energy capacity distributions are very similar. The distribution appears to be of Gaussian type (as with the process variations used in the simulation) and symmetrical about the nominal value, as indicated by the central blue dashed line at 0% variation. For the common 1% SD used for the process variations in this example, a SD of 1.7% variation is observed from the blue dashed lines shown in the results. This is influenced by the sensitivity of the process variations summarised in Figure 11 along with the probability distribution of the process variation value simulated. This demonstrates the
potential compounding, or stacking, of multiple tolerance parameters in the model. It can also be seen that the reject rate for this example is small from the distribution of the results outside the upper and lower reject thresholds shown by red dashed line in the results.

From Figure 15, the simulated cell reject rate be seen with the process variation SD for both the discharge and energy capacity values. It can be seen that the results are virtually identical for both charge and energy capacity with approximately 2% difference at the 2% to 4% SD range. Above a SD of 1% it appears that the reject rate increases at a reduced rate and is expected to tend to 100% as the SD tends to infinity.

At a SD value of 0.5% or below, the reject rate is zero. From this it can be seen that in order to achieve zero rejects, the common SD must be better than 1%, although need not be better than 0.5%. The analysis in Figure 15 considers the same tolerance deviation for each of the parameters for simplicity. However, these parameters could be independently varied to reflect the tolerance of each individual manufacturing process and provide a more representative estimation of reject rate for a specific manufacturing scenario.

4 | CONCLUSIONS

The influence of six geometrical manufacturing tolerances on the performance of a lithium-ion battery have been analysed through the means of a numerical model. Results from a commercial NMC811/Graphite + Si 21700 cell show the most significant process tolerances upon the cell performance were due to dimensional changes in the electrode height or width, which directly affected the amount of active material present in the cell for both electrodes. A positive tolerance directly increased the discharge and energy capacity and significantly reduced the ohmic resistance due to an increase in active material and electrode area.

Calendering tolerance (which directly affects the electrode thickness) was seen to have little effect upon the discharge and energy capacity values when applied to either the positive or negative electrode, but a moderate variation in ohmic resistance for the negative electrode and changes in rest period rise time (voltage relaxation) for both positive and negative electrodes. These results imply that whilst calendering has minimal influence on capacity, it could lead to current imbalance in cells connected in parallel.

Electrode coating tolerances had significant effect upon the discharge and energy capacity values when applied to the negative electrode, but not to the positive electrode, implying the cell is limited by the negative. However, positive electrode coating tolerance was seen to have a significant effect on the voltage relaxation of the cell.

For a simulated production run of 1000 cells with a zero-reject rate for the desired discharge and energy capacity values, the common tolerance SD value must be better than 1%, although it is not necessary for it to be less than 0.5%. Using this in conjunction with the sensitivity analysis it can be concluded that the electrode slitting, electrode cutting and negative electrode coating tolerances must be controlled at a SD of 0.5% to 1.0%. The negative electrode calendering tolerance, positive electrode calendering tolerance and positive electrode coating tolerance do not have a significant effect upon production yield and do not need to be as tightly controlled to obtain a low reject rate.

The findings of this work demonstrate the importance of understanding the impact of tolerances upon cell performance and the use of numerical models to highlight the most sensitive tolerances. It is anticipated this work will help direct the allocation of resources to the most sensitive processes within battery manufacturing, so that the desired performance variation and reject rate can be achieved.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

DATA AVAILABILITY STATEMENT

The underlying data and code used in this work can be found here: www.doi.org/10.17028/rd.lboro.19745614.

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**SUPPORTING INFORMATION**

Additional supporting information can be found online in the Supporting Information section at the end of this article.

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