The Effect of Volume Change on the Accessible Capacities of Porous Silicon-Graphite Composite Anodes

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Silicon-graphite (Si/C) composite anodes are used to increase total anode capacity while maintaining a tolerable degree of active material volume expansion. However, increasing the Si/C ratio does not directly lead to an increase in the accessible capacity because excessive volume expansion can lead to unacceptable cell pressure or electrode porosity. To predict the accessible capacity as a function of Si/C ratio, we integrated mechanical behavior for individual cell components into our previous battery model that couples mechanical and electrochemical phenomena and then simulated a full charge of a pouch cell with foam packing. The simulations were used to determine the anode accessible capacity as a function of Si/C ratio, based on practical pressure and porosity design limitations. The resulting predictions illustrate the tradeoff between the capacity gained by increasing the Si/C ratio and the accessible capacity lost based on the pressures that build up in the cell due to the interconnected mechanical and electrochemical phenomena. For a given set of cell and pack design requirements, battery designers can use these types of simulations as a tool to maximize accessible capacity of an operating cell by selecting appropriate cell/pack materials and identifying optimal Si/C ratios.

The desire to realize large scale and cost effective electric vehicle (EV) commercialization has caused several classes of anode active materials to be proposed in recent years, and silicon has received considerable attention as a result of its high theoretical capacity (4200 mAh/g for Li22Si5). However, developers have not yet been able to realize pure Si anodes with sufficient cycle life for commercial use, due in part to the large volume change that occurs during lithium intercalation. Silicon cycles through at least 4 stable alloy phases during a charging and discharging cycle, leading to a complete transformation in the crystal structure and a ~400% volume expansion between the fully charged and fully discharged states. The coupled high crystallographic strain and large volume change leads to Si fracturing and pulverization, which detaches Si from the electrode during discharge, rapidly decreasing capacity and electrical contact. In fact, metallic silicon loses nearly all of its capacity after only 10 cycles. To mitigate structural strain, and thus, reduce capacity loss with cycling, researchers are using silicon/graphite (Si/C) composites to incorporate the energy dense silicon active material without excessive expansion.

In order to accurately predict pressure and porosity to aid in practical design limitations, cell face pressure and electrode porosity. Most current production EVs contain the cells in a battery pack or module to maintain electrical conductivity and provide efficient cooling. Typical modules contain cooling fins, thermostats, foam separators, and repeating frame elements to hold the cells. Each module is connected to an adjacent module with bus bars and structural bars. As the cells expand and contract during cycling, the stresses generated can cause the materials in the battery module to deform and crack, resulting in a loss of electrical contact and therefore, decreased range, or a loss of heat transfer fluid, which could result in a loss of effective cooling and reduce thermal runaway. This cell face pressure also has an effect inside the cell, leading to decreased porosity, meaning higher ionic diffusion resistance across the electrode thickness; as well as the pulverization and detachment of the active material from the porous electrode matrix, reducing capacity.

In order to accurately predict pressure and porosity to aid in practical cell design, we incorporated mechanical behavior of the individual components of the cell as well as the cell packing itself into our previously developed model.

Recently, Zhang et al. conducted mechanical tests to evaluate the constitutive properties of porous electrodes, for both anode and cathode. Zhang’s experimental work gave insight into the mechanical behavior of the fully formed electrode as opposed to the mechanical behavior of the individual materials in the electrode. Using Zhang’s experimental results as an input to our model allowed us to mimic realistic mechanical behavior for the anode and cathode. Likewise, we incorporated component level mechanical behavior for the separator and a generic polyurethane packing material into our model to produce an accurate representation of mechanical behavior in a full battery pack.

The incorporation of these realistic component mechanical behaviors allowed us to develop practical dimensional predictions in our model.

Model Setup

During charging, Li+ ions move from the cathode to the Si/C composite anode resulting in an expansion of the active material. For this study, we assumed low-rate charging of the anode to focus on...
maximizing volumetric and gravimetric energy density in order to aid in selection of silicon composites. Performance studies that incorporate high-rate charging and discharging of cells with silicon composite anodes will be the focus of future work.

As shown previously by Garrick and Weidner,\textsuperscript{8–10} the volume change of the anode is based on the material balance governing equation shown below:

\[
\frac{\partial}{\partial t} \left( -V \right) + \frac{\partial}{\partial t} \iota = \frac{s \Delta \hat{V}}{nF} j \tag{1}
\]

The right side of the equal sign represents the volume change due to the intercalation of lithium into the anode active material, which then either affects the electrode layer dimensional changes (second term on the left) or porosity changes (first term on the left). The average volumetric current density, \( j \), and the anode porosity and strain, \( \epsilon \) and \( \psi \), respectively, are the variables linking electrochemical behavior to mechanical behavior. The equations governing \( j \) and all relevant electrochemical initial conditions and boundary conditions have been taken from previous works.\textsuperscript{16–19}

Before we can properly couple mechanical and electrochemical behavior, we must accurately represent the mechanical behavior of the cell and each of its components. Each cell component will exhibit a different strain response to a given pressure, and the total cell face pressure is generated based on the relative compliance and thickness of each component. Therefore, the individual component thicknesses and their respective stress-strain responses must be incorporated in our calculations.

Our previous model incorporated porous rock mechanics to describe the strain response from an applied stress for our electrode component.\textsuperscript{8} Here, we utilize a cubic spline interpolation function, \( f_{\text{cubic}} \), to describe the strain response for each porous component: the anode, cathode, separator, and foam packing. The stress-strain response of most porous materials can be described as a second or third order polynomial when considering the pressure ranges applicable to an EV battery pack during normal operation. Therefore, the cubic spline interpolation would produce a more accurate representation of the material behavior than a linear interpolation. These interpolation functions, built from experimentally measured mechanical responses, provide a realistic mechanical behavior input for each porous component.

In this simulation, the porous anode and cathode have two phenomena that contribute to their overall strain. The first is the electrode compression from an externally applied pressure. This description is the previously mentioned anode or cathode intercalation function, \( f_{\text{c}} \) or \( f_{\text{s}} \), respectively. The second phenomenon is the electrode expansion caused by the active material volume expansion upon lithium intercalation, \( \psi \). Because we assume a constant low charge rate, we can correlate current density to average state of charge of the electrodes, \( \tau_{\pm} \). We also assume that the volume change is linearly dependent on state of charge and that the individual active materials in the anode composite lithiate at approximately the same rate. Therefore, we use the following governing equation to predict the active material volume change’s contribution to the overall strain response for the electrodes.

\[
\psi_{\pm} = \left[ \frac{\Delta \hat{V}}{V_0} \right] \tau_{\pm} \tag{2}
\]

\( \left[ \frac{\Delta \hat{V}}{V_0} \right] \) is the ratio of the total active material volume change to the un lithiated active material volume. For the composite anode, \( \left[ \frac{\Delta \hat{V}}{V_0} \right] \), is calculated as the sum of the individual active material molar volume expansion ratios multiplied by their respective volume fractions.

\[
\left[ \frac{\Delta \hat{V}}{V_0} \right] = \left[ \frac{\Delta \hat{V}}{V_0} \right]_{c} + \frac{\Delta \hat{V}}{V_0}_{s} \tag{3}
\]

Combining the two strain phenomena produces the overall electrode strain response in an operating cell. This equation can then be incorporated for both anode and cathode mechanics.

\[
\psi_{\pm} = (\psi_{\text{li}, \pm} + \psi_{\text{i}, \pm}) = -f_{\pm}(\sigma) + \left[ \frac{\Delta \hat{V}}{V_0} \right] \tau_{\pm} \tag{4}
\]

The strain for the anode and cathode current collectors is assumed to follow linear elastic mechanics:

\[
\psi = \frac{\sigma}{E} \tag{5}
\]

The Young’s Moduli, \( E \), used for the copper anode current collector and aluminum cathode current collectors are significantly high. These high Young’s Moduli values rendered the current collectors virtually incompressible compared to the other components at the low pressure windows needed for practical cell operation.

In summary of our model, we assume that each component will experience the same stress, known as the cell face pressure. This pressure variable, along with each component’s strain response, which is described by an interpolation, Eq. 4, or Eq. 5, is what links each individual component strain. Then, as the active materials expand and contract upon cell operation, described by Eq. 1, changes in electrode dimensions, pressure, and porosity will occur. These changes are fully detailed in our previous work.\textsuperscript{8–10,20} The degree to which the anode active material will expand is determined by the Si/C ratio and Eq. 3; as more silicon is incorporated into the active material, the total anode volume expansion will increase. Additionally, the individual component strain responses and thicknesses can be used to determine the overall cell displacement, or cell strain, as a function of cell pressure.

Results and Discussion

We produced a simulation to represent pouch cells with foam packing, as would be seen in an EV battery pack. The simulation incorporates the mechanics of all internal cell components as well as the packing itself. This enables us to observe the effect that packing will have on pressure generation in the cell as well as the overall cell strain during operation. Figure 1 shows the cell geometry considered in our model. The cross-sectional area of each cell component is considered to be uniform with negligible end effects, therefore we solved our model in 1D across the x-axis. Initial component thicknesses reflect the thicknesses seen in current-production large format pouch cells with the foam packing thickness being 30% of the total cell thickness. The initial cell face pressure is 0 kPa, meaning each component’s initial thickness is unaffected by a compressive strain response. Either end of the geometry is held in a fixed position meaning the total geometry length, including packing, is held constant.

COMSOL Multiphysics was used to run our simulation using a Time Dependent solver that was set to run through a full charging of a balanced cell from 2.5 to 4.2 V, representing a 0 and 1 state of charge of the cell, respectively. The physics of the cell were simulated numerically, using the Partial Differential Equation interface. This model is solved in one dimension along the thickness direction of the cell, with
respect to voltage, electrolyte concentration, strain, reaction rate, and porosity; where uniformity was assumed in the other two dimensions. Lithium concentration in the solid phase was solved using a pseudo two-dimensional (P2D) method,\textsuperscript{18} where one dimension is the length of the cell along the x-axis and the pseudo second dimension was solved along the radius of a theoretical active material particle. The electrochemical governing equations are connected to the mechanical governing equations through the electrodes linked by dependent variables: porosity, stress, and state of charge.

When considering an expanding active material, the pressure generation and decrease in porosity are two major design limitations that need to be considered when comparing to a traditional graphite anode. Therefore, we observed the effects of varying the Si/C ratio in the initial anode composite porosity. The anode used here contained 5% binder and an initial porosity of 0.37. This initial porosity was chosen based on a previous study,\textsuperscript{13} however, the model is versatile and can incorporate lower or higher initial porosities if desired. The overall volume change ratio was calculated with Eq. 3, using experimentally measured molar volume expansions for silicon and graphite from literature, 280% and 10%, respectively.\textsuperscript{21–23} The composite anode was assumed to be a fully mixed composite with no variations in local electrode composition. It was also assumed that the Si and C lithiate at approximately the same rate. We used a 3% expansion volume ratio for an NMC cathode.\textsuperscript{24} We chose design limitations for both pressure and porosity based on EV pack design limitations. Pressure limitations, 250 kPa and 500 kPa, are chosen to represent pressures where the module or pack may see cracking, or pressures where cells may be at risk of shorting if any dendrite growth or lithium plating is seen. Porosity limitation, 0.25, was chosen to represent power cells that may require a minimum porosity requirement to avoid ion diffusion resistances, whereas energy cells that sees low charge/discharge rates could still operate properly at a lower porosities.

For the mechanical properties of the anode and cathode, realistic stress-strain curves were obtained from Zhang’s previous study.\textsuperscript{13} Likewise, we incorporated separator data\textsuperscript{14} and selected a generic polyurethane foam,\textsuperscript{15} to represent the packing region for this study because a softer material is needed to account for the large volume change of a Si/C composite anode. Each of these components’ stress-strain interpolation functions were assigned to their individual component domains within the model geometry in COMSOL. The copper anode and aluminum cathode current collectors followed linear elastic mechanics, Equation 5, using Young’s Moduli of 110 GPa and 70 GPa, respectively.\textsuperscript{25}

Simulating pressure generation.—Initially, we simulated a full charge of a cell containing a composite anode with silicon percentages from 0% to 25% to highlight the increase in pressure generation within the cell. Figure 2a shows the resulting pressures generated as a function of anode state of charge for composite anodes with varying percentages of silicon in a cell with foam packing. The pressure increases as a function of state of charge, similar to the results seen in earlier studies that consider single electrodes.\textsuperscript{10} Here, we illustrate how increasing the silicon composition in the anode results in differing rates of pressure generation due to the change in active material volumetric expansion. Even the predictions with 0% silicon show moderate pressure generation over a charge due to the assumed 10% expansion of graphite. Figure 2b shows the same simulation if the total cell volume was fixed (i.e., the foam packing volume would be rigid and hold the cell volume constant). This second simulation’s purpose is to show the importance of including packing mechanical behavior in a coupled mechanical and electrochemical simulation. When we assume a fixed anode volume as in Figure 2b, we only see an exponential increase in the pressure, which is representative of the sum of the components inside the cell. When considering 0% silicon, or a pure graphite anode, for fixed cell and foam-packed cell, we have pressures of 392.3 kPa and 89.1 kPa at the end of a full charge, respectively. These details indicate that even for anodes with less significant expansion, in this case, graphite with 10% expansion, we still observe the significant impact that the foam packing mechanical response has on the prediction of the full cell mechanics, and why most battery packs containing pouch cells with graphite anodes have foam to contain the cell expansion. As seen here, if a fixed cell volume is assumed, the pure graphite anode would not meet a 250 kPa pressure design limitation after a full charge. Hence, prismatic battery designers will leave space within the rigid casing to allow for cell material expansion. Additionally, from Figure 2a, the pressure generation follows a unique profile which includes an initial linear increase in pressure, a plateau, and then exponential increase. This profile aligns with the stress-strain of our generic foam packing, which is the most compliant component of our simulation geometry.

Accessible capacities based on pressure.—Dash and Pannala’s study indicates that increasing silicon percentage in the composite anode will only lead to an increase in gravimetric capacity.\textsuperscript{7} This is because their gravimetric capacity calculations are unaffected by the increasing initial porosity required to allow the active material to fully expand into the void space. Whereas, volumetric capacity will increase until a “threshold value”, or the silicon percentage to maximize volumetric capacity. This threshold value is found where the increasing capacity from adding silicon meets the decreasing capacity from increasing initial porosity. We built upon their concept for calculating threshold values by two means: first, we account for the impact that mechanical limitations will have on the anode, second, we consider that we need to maintain a porosity large enough to avoid significant ionic resistances instead of letting the active material completely expand into the void space.

To determine the accessible anode capacities, we must determine the anode’s operational state of charge, τ, range where the pressure limitations are avoided. In effect, the state of charge at which the pressure meets the design limitation would be the selected cutoff point.

Figure 2. Cell pressure as a function of τ for a cell within (a) foam packing or (b) a fixed volume with an initial anode porosity of 0.37. Solid and dashed curves represent incremental 5% increases in silicon composition in the Si/C composite, from 0% to 25%. The solid horizontal lines represent pressure limitations of 250 kPa and 500 kPa.
The horizontal lines in Figure 2 illustrate where the state of charge cutoff points would be for a pressure limitation of 250 kPa and 500 kPa. The state of charge cutoff can then be used to determine the accessible capacity. The accessible capacity is calculated by multiplying the total anode volume at a given state of charge by the volumetric capacity of the anode, and then dividing this product by the total mass of the anode materials (including binder) and volumetric capacity.

The accessible gravimetric and volumetric capacities for a cell with flexible foam packing are shown in Figure 3. From Figure 3, we see a linear increase in anode capacity with increasing percent silicon. This trend follows Eq. 6, which is the calculation of theoretical capacity unaffected by the pressure cutoff. Then, at a certain silicon composition, we observe a non-linear decrease in anode capacity, caused by the pressure cutoff limitation occurring at decreasing \( \tau \) with increasing silicon composition. Using our simulation of a cell with foam packing, we determined the threshold capacity to be 461 mAh/g at 5% silicon and 689 mAh/g at 14.5% silicon for the 250 kPa and 500 kPa limitations, respectively. For this given cell geometry and chemical design, these would represent the ideal silicon percentages to use without generating damaging pressures. However, this model is also able to tune chemistries, component thicknesses, and packing thicknesses. Therefore, the model can act as a tool for engineers designing cells, choosing foam packing, and determining pack structure requirements.

**Simulating porosity change.**—As previously mentioned, another mechanical design limitation is electrode porosity. There are minimum porosities required to avoid significant ionic resistance in the electrolyte, and these ideal design porosities would vary depending on the function of the cell; high power cells would require a higher porosity due to the need for significant diffusion of ions through the electrolyte. On the other hand, energy cells see lower C-rates, meaning that in the high strain-rate region, the dimensional growth contribution in Eq. 1, or the second term on the left side of the equal sign, would represent either power or energy cell design requirements. As we did in the pressure study, we can calculate accessible capacities using Eq. 6 and the porosity cutoff condition. Figure 5 shows the resulting volumetric capacities as a function of state of charge for each silicon percentage, showing initial porosity of the anode from 0.25 to 0.3. Figure 4 shows the resulting plot from the 25% silicon composition.

As expected, in Figure 4 the porosity initially decreases due to the expanding active material in the anode, and because silicon expands more than graphite, we see a large porosity change as a function of state of charge with a high silicon composition in the active material. However, near a state of charge of 0.3, we observe a slight increase in porosity until a state of charge of about 0.45. This is another indication of the foam’s mechanical behavior. With a 25% silicon composition, the anode active material expansion generates pressures linearly until the pressures reach the foam’s “plateau” region in its stress-strain response. The “plateau” represents a region of high strain-rate, in other words, large increases in strain, or material expansion, generate much less pressure as the foam is extremely compliant in this region. This means that in the high strain-rate region, the dimensional growth contribution in Eq. 1, or the second term on the left side of the equal sign, will be favored heavily because of the relatively constant cell face pressure, and therefore the porosity will increase. After a porosity of 0.45, we see the porosity exponentially decrease as pressures quickly develop after the high strain-rate region.

**Accessible capacities based on porosity.**—Similar to the pressure design limitation analysis, we assigned a 0.25 porosity cutoff to represent either power or energy cell design requirements. As we did in the pressure study, we can calculate accessible capacities using Eq. 6 and the porosity cutoff condition. Figure 5 shows the resulting volumetric capacities as a function of state of charge for each silicon percentage.
Component strain prediction.—In addition to pressure and porosity limitations, practical cell and pack design will require a total strain limitation of the cell or its individual components. Because the anode active material expansion is linked to the mechanical behavior of all cell components, we can observe the component strain (compression/expansion) as a function of state of charge. Figure 6 illustrates this concept for a 5% silicon Si/C composite with a generic foam packing. As can be seen in Figure 6, as the cell charges, the anode expands and in turn compresses the other internal components. The most significant compressive strain is seen with the cathode, caused by the pressure generation and the de-lithiation of the cathode active material. The packing compresses to accommodate for the overall cell expansion, so the displacement of the anode does not directly correlate to the compression of the cathode and separator. For a more rigid foam packing, we would see similar component displacements without much cell or packing displacement. From this simulation, we can translate anode and cathode expansion and contraction to separator displacement. In this case the separator is being displaced by nearly 17 microns, which is over half of a typical separator thickness. The repeated displacement of the separator can lead to the wearing of the edges of the separator, a critical safety factor for lifetime cell usage.

The ability to model anode porosity, pressure in the cell, and strain of cell and individual cell components is imperative for practical pack design. The simulations here show realistic mechanical behavior of the battery and its components which can be used to optimize the design of battery systems using Si/C composite chemistries. In the future, the model can be experimentally validated and applied to cell and battery pack design.

Conclusions

The objective here was to determine the accessible capacities of varying Si/C composite ratios while appropriately accounting for the volume change of the electrode active materials. This was achieved by using a battery model that couples electrochemical and mechanical phenomena and incorporating realistic mechanical behavior for each cell component. We emulated realistic mechanics by linking experimentally measured mechanical behavior for each component. Then, we simulated a full charge of a pouch cell within a generic foam packing for anodes with varying Si/C ratios. Our simulations allowed us to predict mechanical changes (i.e., porosity, strain, and pressure) that occurred while charging. Then, the accessible capacities were determined based on realistic design limitations, cell face pressure and anode porosity. This study revealed that, when incorporating the full cell and packing in mechanical predictions, there are possibilities to incorporate more silicon in the composite anodes used in automotive battery pack applications. Throughout this study, we observed the extremely significant effect that packing material has on the mechanical predictions in the cell. For this particular model study of Si/C composites, using conservative 250 kPa pressure and 0.25 porosity cutoffs, we found that an appropriate balance of initial anode porosity and silicon percentage in the anode active material could lead to anodes with ~59% more capacity than traditional graphite anodes. This modeling process could be used to predict and account for anode composite volume change for cell and pack design applications. Additionally, this model can be tuned to simulate and optimize other features of the cell, such as cell component thicknesses, different electrode/electrolyte chemistries, and different packing thicknesses/materials.

Future Work

We plan to study the effect that significant active material volume change will have at non-uniform reaction rates. We will also continue an experimental approach to refine our model in several areas. We will conduct mechanical studies on large format pouch cells to validate our model as well as develop a model to predict the realistic component and cell level mechanics based on fundamental material properties. We will also study the relative lithiation rates and volume expansion rates for both Si and C in the same electrode.

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List of Symbols

- E: Young’s Modulus, Pa
- f: Function describing strain response to applied pressure
- F: Faraday’s constant, 96485 C/mol
- j: Average current density, A/cm²

Figure 6. Individual component (anode, cathode, separator, and foam packing) and the overall cell expansion/compression as a function of state of charge.
Electrons transferred in the reaction

\( Q \)

Theoretical Capacity

\( s \)

Stoichiometric coefficient

\( t \)

Time, s

\( x \)

Mass fraction

\( y \)

Volume fraction

**Greek and Special**

\( \Delta V \)

Total molar volume expansion of the active material particle, cm\(^3\)/mol

\( \Delta V_0 \)

Ratio of the total active material volume change to the un lithiated active material volume

\( \varepsilon \)

Porosity

\( \varphi \)

Strain

\( \varphi_i \)

Strain response from lithium intercalation

\( \varphi_{st} \)

Strain response from applied stress

\( \sigma \)

Stress, Pa

\( \tau \)

State of charge of the local electrode

**Subscripts and Superscripts**

\( 0 \)

Initial

\( \text{Anode} \)

\( \text{Cathode} \)

\( \text{Graphitic carbon} \)

\( \text{Cell Component} \)

\( \text{Silicon} \)

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