Battery aging and degradation remain as important challenges faced in efforts to extend battery lifetimes. Aging not only causes capacity fade and a reduced cycle life, but also imposes threats to safety and can lead to catastrophic failure of the entire system. Therefore, from both operational and safety perspectives, it is essential to monitor battery aging so that the exact level of the decay can be measured. To quantify aging, state of health (SOH) is often used, which is a parameter defined as the ratio between the measured capacity and the initially rated capacity of a cell. For instance, a pristine cell has a SOH of 1, while a cell having a SOH of 0.8 is considered highly degraded. There are a number of studies on battery diagnosis with different approaches.\textsuperscript{1–7} The most accurate method of determining SOH is to measure capacities through controlled discharge tests. A more practical method involves mathematical models which take measurable parameters, such as current, voltage, and temperature, as inputs.\textsuperscript{8–10} However, the accuracy of such methods depends on the complexity of the underlying model and can be further complicated by battery aging. Therefore, a practical and accurate stand-alone method is needed.

Cannarella\textsuperscript{6} has proposed a mechanical method of determining the SOH of a pouch cell using measured external stress. By measuring the initial peak stress and peak stress at a later time, with the known stress-SOH relationship, SOH can be determined. This method is based on the mechanically active nature of batteries.\textsuperscript{11,12} Batteries undergo volume changes during operation, such as electrode swelling, polymer deformation, and film growth, where the volume changes caused by the latter two are irreversible.\textsuperscript{13–19} Polymer deformation largely results from separators and binders.\textsuperscript{20–25} As the softest component inside the cell, the polymer accommodates the expansion from electrode swelling and gradually deforms. Film growth can take place in the form of a solid electrolyte interphase (SEI) or lithium plating, which are both important battery degradation mechanisms and both of which can lead to an irreversible increase in cell thickness.\textsuperscript{26–28} Because the loss of active lithium is directly related to the thickness change, a linear relationship between peak stress and capacity fade has been observed.\textsuperscript{6} The study\textsuperscript{6} suggests that with a known relationship between peak stress and SOH, it is possible to determine battery aging by only measuring the increase of the external stress through a stress sensor. This method can be potentially used to diagnose battery health and can provide indications of battery safety. However, one of the limitations to the prior work is that it has only been verified on one cycling range using batteries from the same batch.

In this paper, we look further into the fundamentals behind this linear relationship between stress and SOH and try to find correlation between stress and capacity loss. By conducting tests using commercial batteries with different compositions and cycled at various depths of discharge, we show that the stress-SOH relationship is composed of both linear film growth and non-linear stress relaxation. The linear slope depends not only on mechanical properties of batteries, but also on various cycling ranges. The overall stress-SOH relationship results from electrochemistry as well as mechanics.

**Experimental**

In this study, 500 mAh commercial pouch cells with graphite anodes and lithium cobalt oxide cathodes are used. The same experimental setup is used as described in the previous paper\textsuperscript{14} where a pouch cell is placed in series with a load cell under a designed fixture, as shown in Figure 1. The pouch cell is connected to a potentiostat while the load cell records measured stress. Before being placed and constrained into the fixture, each cell is fully discharged to 2.7 V. Clamping pressure can strongly affect battery cycle life as well as altering cell behavior. In the experiment, the clamping pressure has been carefully adjusted to 0.05 MPa on an Instron mechanical testing apparatus to ensure the same amount of initial stack pressure is applied on all tests. The pressure was chosen because in the previous work we found that an initial stack stress of 0.05 MPa yielded the longest cycle life compared to a higher initial stack stress or a system free of constraint.\textsuperscript{14} A pressure around 0.05 MPa ensures good contact free of constraint.\textsuperscript{14} A pressure around 0.05 MPa ensures good contact between electrodes without causing significant mechanical degradation. Bolts and nuts, along with thread locking adhesive, are used to constrain the cell assembly.

![Figure 1. A schematic of the experimental setup where capacity and mechanical stress are measured.](image-url)
ensure no loosening or displacement occurs over time. All tests are conducted under ambient room temperature.

To investigate the effects of battery type on stress-SOH relationships, two types of commercial batteries are used, which have the same nominal dimension and chemical reactions (C/LCO), but different structures and material compositions. They have different anode structures as shown in the SEM images in Figure 2, and use different separators. A detailed comparison of battery structures and compositions is provided in Table I. In the experiment, cells are aged between 2.7 V and 4.2 V, corresponding to a range of 0 to 100% state of charge (SOC), at a rate of C/2. Stress is monitored every 10 minutes and capacity is measured at a rate of C/10 for every 50 C/2 cycles. Subsequent characterization of cell components is conducted to reveal differences in material properties.

To investigate the effect of cycling ranges on stress-SOH relationships, tests are performed with four different cycling profiles using batteries from the same batch. Cells are cycled from a fully charged state to various depths of discharge (DOD). For instance, for a 500 mAh battery at C/2 and C/4, three cells are subjected to cycling at each condition to ensure repeatability of the results. The test modes and nominal cycling ranges are summarized in Table II.

**Results and Discussion**

Pouch cells are mechanically active, so by constraining the displacement, the internal stress can be easily measured. The overall irreversible stress change results from underlying stress mechanisms such that

\[
\Delta \sigma_{\text{total}} = \Delta \sigma_{\text{increase}} + \Delta \sigma_{\text{decrease}}
\]

Mechanisms causing irreversible stress increases include film growth at the anode and density change of the electrodes. Mechanisms that cause stress to decrease include stress relaxation, polymer deformation (binder or separator), and electrode fatigue. In this work, it is assumed that under normal cycling conditions, film growth is the dominant mechanism causing stress to increase, and the film may be composed of SEI layers, lithium plating, or a combination of both.

Starting with a simple model, we explore the effect of the film layer growth on the stress change, and then add a term for stress decrease to obtain a generalized relationship.

**Film growth model.**—The film growth model for the stress-SOH relationship is constructed based on following assumptions:

1. Batteries degrade solely from the gradual loss of active lithium.
2. The irreversible film growth is the sole source of stress increase inside the battery.
3. The battery modulus at peak stress is constant.

The first assumption is valid when the cycling rate is relatively low such that no significant degradation occurs. For batteries used in this work and in previous work, lithium-ion pouch cells mainly degrade through the loss of active lithium when they are cycled slower than C/2. A higher rate leads to larger irreversible structural changes, such as electrode cracking and loss of contact with current collectors, causing capacity to decay and the proposed method to break down. Within the context of this work, since all batteries are cycled below C/2, the first assumption is legitimate. The second assumption holds in the case that no other parasitic effects such as gas evolution at the electrodes are occurring. In a later section, we show that besides the film growth, there are mechanisms that lead to a decrease in stress which can be important as well, but we ignore them here. With respect to the third assumption, even though batteries exhibit a non-linear modulus, the modulus stays roughly the same when the battery is fully charged. Last but not least, underlying these

### Table I. A comparison between two types of commercial lithium-ion pouch cells with different dimensions and material compositions.

<table>
<thead>
<tr>
<th>Nominal dimension</th>
<th>Type A 25 mm × 35 mm × 6.5 mm</th>
<th>Type B 25 mm × 35 mm × 6.5 mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Measured thickness</td>
<td>Anode 2.9 ± 0.05 mm</td>
<td>2.7 ± 0.06 mm</td>
</tr>
<tr>
<td></td>
<td>Cathode 2.3 ± 0.05 mm</td>
<td>2.2 ± 0.06 mm</td>
</tr>
<tr>
<td></td>
<td>Separator 0.46 ± 0.07 mm</td>
<td>0.69 ± 0.02 mm</td>
</tr>
<tr>
<td></td>
<td>Pouch 0.29 ± 0.005 mm</td>
<td>0.21 ± 0.01 mm</td>
</tr>
<tr>
<td></td>
<td>Total 5.8 ± 0.1 mm</td>
<td>5.7 ± 0.2 mm</td>
</tr>
<tr>
<td>Separator</td>
<td>Layer thickness 10 μm</td>
<td>18 μm</td>
</tr>
<tr>
<td></td>
<td>Composition Polyethylene</td>
<td>Polypropylene</td>
</tr>
</tbody>
</table>

### Table II. Cycling ranges and cycling rates used in this study.

<table>
<thead>
<tr>
<th>Cycling range (SOC)</th>
<th>75–100%</th>
<th>50–100%</th>
<th>25–100%</th>
<th>0–100%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Commercial type A batteries at C/2</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Commercial type B batteries at C/2 and C/4</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
three assumptions above, the model is based on the conceptual idea that stress and strain can be interconverted from a mechanics point of view. Conceptually, a stress increase is equivalent to compressing a material to a certain strain.

The previous data shows that both capacity fade and peak stress change as functions of $\sqrt{t}$. Therefore, SOH and change in stress can be experimentally related to $\sqrt{t}$ using:

$$1 - SOH = a \sqrt{t}$$

where $\frac{\Delta \sigma}{\Delta \sigma}$ is an experimentally fitted linear slope.

Since the only degradation mechanism is through the loss of active lithium that is used to construct the film layer, the thickness of film, $\Delta l_{film}$ can be related to the SOH through two proportionality:

$$\Delta l_i = k_1 (1 - SOH)$$

$$\Delta l_{film} = k_2 \Delta l_i$$

where $\Delta l_i$ is the thickness of irreversible film around one graphite particle and $\Delta l_{film}$ is the thickness of film around the anode. Parameter $k_1$ is a constant that relates the loss of the lithium to the thickness of one layer, which depends on the electrochemical reactions in forming these film layers, while parameter $k_2$ is a geometry factor relating the thickness of the film on one graphite particle to the total thickness of the film layer at the entire anode. After combining Eq. 4 and Eq. 5, and aggregating $k_1$ and $k_2$, we obtain the following equation:

$$\Delta l_{film} = k (1 - SOH)$$

Mechanically, the thickness of film can be converted to strain and to stress through a constant modulus:

$$\Delta \epsilon_{increase} = \frac{\Delta l_{film}}{l} = \frac{k}{l} (1 - SOH)$$

where $l$ is the thickness of the battery. With a constant modulus $E$, stress can be linearly related to capacity fade:

$$\Delta \sigma_{total} = \Delta \epsilon_{increase} E = \frac{k}{l} E (1 - SOH)$$

where $\frac{1}{l} E$ is the proportionality between stress increase, $\Delta \sigma_{total}$, and capacity fade, $1 - SOH$, and can be related to the experimental slope through:

$$\frac{k}{l} E = \frac{b}{a}$$

Note here that instead of absolute stress values, stress differences are used.

The above model indicates that if the film growth dominates, the stress-SOH relationship should be linear, which is consistent with what Cannarella has observed. The next question to address is whether the stress increase due to film growth is always the dominant mechanism and whether the linear slope is always the same. In fact, using tests conducted under various cycling ranges, we show that the mechanisms leading to stress decrease affect the overall stress-SOH relationship, and film growth is no longer the dominant mechanism.

**Effects of Cycling Ranges on Stress vs. SOH**

Batteries cycled with different profiles exhibit different stress-SOH behaviors and such behavior results collectively from film growth and stress relaxation. Figure 3 shows stress evolution profiles for batteries cycled with different depths of discharge. After every 50 cycles, the capacity is measured through a full C/10 cycle, as indicated by the lower stress dropping almost to 0. By plotting increases of peak stresses compared to the first cycle and the corresponding capacity losses, a relationship between stress increase and SOH can be obtained, which is shown in Figure 4. Each data point represents the increase in the peak stress at its corresponding capacity fade compared to a pristine cell. Details regarding the construction of the plot can be found in the previous work.

With different cycling profiles, stress-SOH relationships vary, partly because they have different stress relaxations, which play a significant role in the early stage. Stress relaxation occurs due to the viscoelasticity of polymer components, such as separators, pouches
Cycling ranges affect the average stress level inside the battery, leading to different stress relaxations. Since the stress is a dynamic quantity that varies over time, an integral average is used to represent the average stress level a battery experiences during cycling:

$$\bar{\sigma} = \frac{\int \sigma \, dt}{\int dt}$$  \[10\]

Note here that during 0–100% and 25–100% tests, stresses gradually increase, which can cause higher stress relaxations. However, stress relaxation is only relevant at the initial stage. Moreover, the additional stress relaxed due to the stress increase is much smaller than the stress increase due to the film growth. Therefore, in the analysis, it is legitimate to assume that the average stress stays roughly the same. Table III shows that the average stress is positively related to the stress increase as well as the non-linear stress decrease. Stress relaxation is assumed to be the dominant mechanism causing stresses to decrease and can be modeled in different mathematical forms.42 In this work, we empirically select a power law shown in Eq. 11 as it most accurately represents our experimental stress relaxations shown in Figure 5a.

$$\sigma_{\text{relax}} = \sigma_0 - c t^m$$  \[11\]

$$\Delta \sigma_{\text{decrease}} = -c t^m$$  \[12\]

Combining film growth in Eq. 1–2, 8–9 and stress relaxation in Eq. 12, we obtain the following equation that relates the total stress change to the SOH:

$$\sigma_{\text{change}} = \sigma_0 - c t^m$$

Figure 5. a) Stress relaxation as a function of different loaded stresses. Cells are initially loaded to 0.1 MPa, 0.5 MPa, 1 MPa, and 2 MPa at a strain rate of $10^{-3}$ s. Once the desired stress level is reached, cells are held at a constant displacement while stresses are free to relax. b) A log scale plot showing the experimental stress relaxation along with a fit using Eq. 11, where $\sigma_0$, $c$, and $m$ are determined by the best fit. Parameters $c$ and $m$ vary nonlinearly with the initial stress, $\sigma_0$.

Table III. A table summarizing stress relaxation during the first 50 cycles, average stress and average voltage during the first C/2 cycle, and fitted model coefficients for tests under various cycling ranges.

<table>
<thead>
<tr>
<th>Cycling range (SOC %)</th>
<th>Amount of stress relaxation (MPa)</th>
<th>Average stress $\bar{\sigma}$ (MPa)</th>
<th>Maximum stress at SOH = 0.95 $\sigma_{\text{max}}$ (MPa)</th>
<th>Average voltage (V)</th>
<th>$C_1$ (MPa%)</th>
<th>$C_2$ (MPa%)</th>
<th>$C_3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>75–100</td>
<td>0.069</td>
<td>0.33</td>
<td>0.53</td>
<td>4.09</td>
<td>$4.0 \times 10^{-3}$</td>
<td>$8.6 \times 10^{-4}$</td>
<td>0.1</td>
</tr>
<tr>
<td>50–100</td>
<td>0.033</td>
<td>0.29</td>
<td>0.5</td>
<td>4.04</td>
<td>$6.1 \times 10^{-3}$</td>
<td>$7.3 \times 10^{-4}$</td>
<td>0.1</td>
</tr>
<tr>
<td>25–100</td>
<td>0.0011</td>
<td>0.24</td>
<td>0.4</td>
<td>3.99</td>
<td>$1.2 \times 10^{-2}$</td>
<td>$2.2 \times 10^{-4}$</td>
<td>0.1</td>
</tr>
<tr>
<td>0–100</td>
<td>0</td>
<td>0.20</td>
<td>0.36</td>
<td>3.90</td>
<td>$3.2 \times 10^{-2}$</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

Stress-SOH model.—Building on the previous model, a term for stress decrease is added so that the model now includes the linear stress decrease:

$$\Delta \sigma_{\text{decrease}} = -c t^m$$  \[11\]

$\Delta \sigma_{\text{decrease}}$ represents our experimental stress relaxations shown in Figure 5a.

Figure 6. Stress-SOH relationships for capacity measurements collected at a C/10 rate for every 5, 10, and 25 cycles. Each data point corresponds to a C/10 capacity measurement. In between, cells are cycled at a 25% depth of discharge at a C/2 rate. As the capacity is measured more often, a smaller stress relaxation is observed.
the capacity fade:

$$\Delta \sigma_{\text{total}} = (1 - \text{SOH}) - C_1 (1 - \text{SOH})^{C_2}$$  \[13\]

Here in Eq. 13, the change in stress decrease may be affected by the change in stress increase, but such coupling effects between $\Delta \sigma_{\text{increase}}$ and $\Delta \sigma_{\text{decrease}}$ are small. Therefore, it is assumed that the effects from stress increase and effects from stress decrease are independent, and they can be added linearly. After aggregating the parameters from Eq. 13 into three tunable coefficients, we obtain the following equation that relates stress change to capacity fade:

$$\Delta \sigma_{\text{total}} = C_1 (1 - \text{SOH}) - C_2 (1 - \text{SOH})^{C_3}$$  \[14\]

where $C_1 = \frac{1}{c} = \frac{k}{E}$.

**Model fitting and parameters.**—Since our model is based on the assumption that under normal operating conditions, batteries degrade mainly through the loss of active lithium through a gradual film growth, the following analysis is restricted to the portion of data before multiple degradation mechanisms occur, indicated by the onset of continuous capacity loss without any further volumetric increase. For instance, for 0–100% SOC tests, multiple degradation mechanisms occur after the SOH drops below 92%. Beyond that point, the cell degradation causes changes to intrinsic properties, causing the method to break down. Nevertheless, this method has obvious benefits in the short term, as it can correlate stress increase to capacity fade.

The model confirms the hypothesis that the observed stress-SOH relationships result from both the linear film growth and the non-linear stress relaxation, which is related to the average stress level. The model is fitted using the least squares method, and the fitted model is shown in Figure 7 while coefficients are presented in Table III. The fitted C values are physical parameters, where $C_1$ represents the correlation between the stress increase and the capacity fade, while $C_2$ and $C_3$ describe the amount of stress relaxation. In tests with large depth of discharge, $C_1$ is much larger than $C_2$, indicating that the stress increase dominates over the stress decrease. This is the reason why in the previous work, stress relaxation contributions are negligible and a simple linear relation holds. $C_2$ describes the magnitude of the stress relaxation and scales with the average stress level. $C_1$ represents the non-linear stress effect from the relaxation and its near-zero value confirms that stress relaxation is more relevant at the initial stage. Figure 7c shows the relative effects of film growth and stress relaxation. The linear film growth is overshadowed by the non-linear stress relaxation at the beginning, but gradually becomes more dominant. The above analysis is confirmed with strong correlations in all tests using different types of batteries.

As the depth of discharge increases, the slope of the film growth increases, as indicated by $C_1$. Such slope differences arise from differences in the moduli as a result of different peak stresses. Figure 3a shows that batteries cycled under various ranges evolve to different peak stress levels. For instance, 75–100% tests have a lower peak stress profile than 0–100%. The reason $C_1$ scales negatively with the average stress but positively with the maximum stress is that a higher average stress leads to a larger stress relaxation, which then leads to a smaller peak stress at the fully charged state where stress and SOH measurements are collected. Because batteries behave non-linearly, a smaller peak strain leads to a smaller modulus and therefore a smaller slope through Eq. 14. In other words, average stress determines $C_2$ while maximum stress affects $C_1$.

However, the change of the modulus alone cannot fully account for the slope difference. In fact, both modulus and electrochemistry are contributing to the slope. To demonstrate this idea, a simple calculation is performed to find the moduli at stress levels corresponding to a 5% capacity fade. Results are presented in Table IV. The modulus change is much smaller than the slope change, suggesting that $k$ in Eq. 14 must be different. Such a difference can be inferred from the difference in the average voltage. SEI layer and lithium plating are two common contributions to the surface film layer. The particular composition of the surface film will depend on the voltage level or cycling range which causes $k$ to differ. Furthermore, even though substantial efforts have been made, the composition and the formation of SEI layers remain highly debated subjects, as are their electrochemical and mechanical properties, which can be further complicated by composition differences at different voltages. Nevertheless, it is well agreed that the film thickness, as well as the film’s porosity and morphology, are voltage dependent. Thus, because of these complexities arising in forming this passivating layer, the parameter $k$ as shown in Eq. 9 and Eq. 14, can vary among the tests, affecting the linear slopes.

**Effects of Battery Types on Stress vs. SOH**

Besides cycling ranges, the linear slope also varies on battery type. Commercial lithium-ion pouch cells yield qualitatively similar results on stress-SOH relationships, but they differ quantitatively. Such quantitative variation is mainly due to the difference in their mechanical properties. This idea is demonstrated by comparing stress-SOH behavior from two different types of batteries which are cycled from fully charged to fully discharged states. As shown in Table I, the two types of batteries have different dimensions and material compositions. Since during the full cycle range, the linear film growth is the dominant mechanism, we can neglect the non-linear stress decrease and focus on comparing linear slopes. Batteries with different material compositions behave mechanically differently, but they all yield qualitatively similar results in terms of the straight line as previously observed. By plotting stresses at the fully charged state (4.2 V) and the fully discharged state (2.7 V), we

---

**Table IV.** A table summarizing values for parameter $C_1$, moduli, maximum stress, and average voltage under various cycling ranges. $C_1$ scales nonlinearly with the modulus and the average voltage.

<table>
<thead>
<tr>
<th>Cycling range (SOC %)</th>
<th>$C_1$ (MPa%)</th>
<th>Modulus (MPa)</th>
<th>Maximum stress at SOH = 0.95 (MPa)</th>
<th>Average voltage (V)</th>
</tr>
</thead>
<tbody>
<tr>
<td>75–100</td>
<td>$4.0 \times 10^{-3}$</td>
<td>42</td>
<td>0.53</td>
<td>4.09</td>
</tr>
<tr>
<td>50–100</td>
<td>$6.1 \times 10^{-3}$</td>
<td>45</td>
<td>0.5</td>
<td>4.04</td>
</tr>
<tr>
<td>25–100</td>
<td>$1.2 \times 10^{-2}$</td>
<td>53</td>
<td>0.4</td>
<td>3.99</td>
</tr>
<tr>
<td>0–100</td>
<td>$3.2 \times 10^{-2}$</td>
<td>57</td>
<td>0.36</td>
<td>3.90</td>
</tr>
</tbody>
</table>
obtain two curves that represent a battery's stress evolution during cycling. Figure 8a shows representative stress evolution profiles for two types of batteries cycled between fully charged and fully discharged states at a C/2 rate. The increase of stress is an indicator of battery aging as shown in the literature. Batteries from type A evolve to higher stresses than batteries from type B, as indicated by the maximum and minimum stress curves. Note here that for the same type of battery, the rate of stress buildup can indicate the degradation rate. However, since both battery type A and type B degrade at a similar rate, the difference in the stress magnitudes is mainly due to the different material properties inside batteries. For instance, batteries from type B are less mechanically stable than those from type A, as indicated by the flat lower stress curve, suggesting softening inside the battery. Such mechanical softening can arise from either separator, polymer binder, or electrode layer sliding. With all variations in battery materials, stress evolves differently from type to type.

Batteries all exhibit a linear trend between stress and SOH. Figure 8b shows stress-SOH relationships for two types of batteries. Note here that we only plot the range where the major degradation mechanism is the gradual film growth. For all batteries tested, peak stresses vary linearly with capacity fade but the slopes are different. Since slopes is the gradual film growth. For all batteries tested, peak stresses vary linearly with capacity fade but the slopes are different. Since slopes evolves differently from type to type.

Mechanical testing reveals that the moduli are different between the two types of cells. During the tests, cells are compressively loaded to 1 MPa at a strain rate of $10^{-5}$ /s, which corresponds to the average strain rate inside cells cycled at 1C rate, and then unloaded until the stress is below 0.005 MPa. Such a loading-unloading cycle is repeated for 20 cycles to eliminate any initial variations in the mechanical behavior. Figure 9a reveals that batteries from type A are mechanically stiffer than those from type B, leading to larger elastic moduli at the same strain level. Characterization of individual cell components reveals that the modulus of type A batteries is higher mainly because they use thinner separators. The measured modulus of a full cell can be approximated as the modulus of a composite using the Reuss Model, where the composite modulus largely depends on the volume fraction of the softest component in the system. With the same product dimensions, a thicker separator leads to a larger volume fraction of separator, leading to a smaller overall modulus.

There are also other sources of variations inside batteries, such as the electrode geometry and the mechanical stability, which can affect the slope difference. Characterization of cell components reveals geometrical variations inside the anodes as shown in Figure 2. The morphology of graphite particles are different. Type B anodes use elongated graphite particles while type A anodes use particles with a more symmetric spherical shape. Such variations in particle geometry may not only affect the mechanical strength of the anode and therefore the overall modulus, but also can affect the $k_2$ geometry factor. However, the direction and the percentage of contribution are difficult to quantify. Furthermore, mechanisms causing stress to decrease, such as softening due to low mechanical stability, can result in a smaller peak strain and consequently a smaller modulus.

After comparing the slope to the modulus, we determine that differences in slopes are primarily due to differences in moduli. By measuring the expansion of a battery at the fully charged state and converting expansion to strain, we can calculate the modulus at the peak strain (Figure 9). The peak strain is calculated at a SOH of 1 and it is assumed that the corresponding modulus does not vary significantly over cycling. Figure 9b shows the corresponding moduli and their ratio. Note here that the exact number can depend on where the mechanical stress-strain curve is initialized, and can depend on the cycle. However, within reasonable initial positions, the ratio between the modulus of type A and the modulus of type B ranges between 2.38 to 3.57, which is roughly a ±20% deviation from the real slope.
Thus, we conclude that the slopes of different battery types are mainly dictated by the moduli.

Conclusions

This work highlights mechanical and electrochemical coupling inside batteries through stress and capacity fade under various scenarios. In this work, we investigate the effects that battery type and cycling range have on the long term stress-SOH relationship and we show that the overall stress change inside the batteries is composed of both stress-increasing and stress-decreasing mechanisms. Through investigating the overall stress-SOH relationships using batteries cycled to various depths of discharge, we assert that the overall stress-SOH relationship results from the combination of a linear effect due to film growth and a non-linear effect due to stress relaxation, where the latter is the dominant process at the initial stage and gradually gives way to the rising importance of film growth.

By constructing a stress-SOH model, we demonstrate that both the modulus and the electrochemistry contribute to the experimentally observed slopes, and the slopes depend on cycling range as well as battery type. Cycling ranges affect the average stress level inside batteries, leading to different stress relaxation and peak stress, and therefore influence slopes. Cycling ranges also lead to different average voltages, which likely affect the compositions and properties of the thin surface films. In addition, different battery types yield qualitatively similar results for the stress-SOH relationship, but vary quantitatively due to mechanical and geometrical differences in cell components, such as differences in moduli. The linear slope of the stress-SOH curve will also depend on the average voltage during cycling, causing the properties of the film, such as compositions and porosity, to differ. Through the results obtained in this work, one can gain a better understanding of the links between mechanical evolution and electrochemical performance, enabling more accurate characterization of the state of health in Li-ion batteries.

Acknowledgments

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