Characterization and modeling of the thermal mechanics of Lithium-ion battery cells

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Abstract

The thermal mechanics of Lithium-ion (Li-ion) batteries are explored with a focus on thermal swelling. Experiments show for the first time that the swelling shape of prismatic battery cells due to temperature variations is significantly different from that due to Li-ion intercalation in unconstrained conditions. In contrast to uniform and orthotropic Li-ion intercalation swelling in a direction perpendicular to electrodes, the nonuniform temperature distribution in the jellyroll and the gaps/voids between electrodes result in distinguishable different swelling shapes. A unique coupled thermal-structural analysis with a simple, but efficient 3-D finite numerical model is proposed to investigate the impact of temperature variations on the thermal behaviors of battery cells. Anisotropic heat conduction and temperature dependency of the coefficient of thermal expansion are taken into account and found to have an impact on temperature distribution and thermal expansion. Experimental validation of the proposed model clearly demonstrates that the coupled thermal-structural analysis with the proposed model can predict accurately the thermal swelling at unconstrained conditions. The solution at pack (constrained) conditions shows that the nonuniform temperature distribution of the jellyroll still plays a critical role for the thermal swelling shape, although the gaps/voids do not occur because of the constraints from spacers in the pack, suggesting that the estimation of core temperature is important. Such an accurate model, able to estimate cell thermal behavior, is beneficial to the design of Li-ion batteries, development of stress or strain sensors, optimum sensor placement, and thermal management of not only single battery cells but also battery packs.

Keywords

Lithium-ion battery; battery swelling shape; thermal expansion; the coefficient of thermal expansion; the coupled thermal-structural analysis

1. Introduction

1 A consumption of fossil fuel resources causes serious problems including the emission of
2 greenhouse gases and pollutants. The consumption of fossil fuels therefore creates other
3 environmental costs and this societal cost can no longer be negligible. To shift towards a low-
4 carbon world, mitigation efforts must occur across all countries from developing countries to
5 industrialized countries.

6 There are two promising solutions to mitigate environmental pollution and additional
7 environmental costs from fossil fuel consumption: 1) the cultivation of renewable farms such as
8 wind and solar farms to provide clean and eco-friendly energy, and 2) the electrification of
9 vehicles to enhance the efficiency of energy consumption in transportation. However, several
10 technological bottlenecks limit the viability and commercialization of these two solutions.
11 Energy produced from renewable farms has stochastic characteristics due to the inherent
12 intermittency of wind and solar energy sources. Vehicle electrification needs alternative energy
13 sources, which feature high energy/power capability, high efficiency, and a long lifetime.

14 Energy storage systems (ESSs) are an excellent way to break through the problems facing both
15 solutions. Especially, electrochemical energy storage systems are convenient and efficient for
16 storing electrical energy in the form of chemical energy. Among various types of electrochemical
17 energy storage systems on the market, Li-ion batteries (LIBs) are the most promising in the
18 sense that LIBs feature no memory effects, low self-discharge rate, broad temperature range of
19 operation, high power/energy density, and long cycle life [1-3].

20 These advantages have prompted on studies on hybrid solar-wind energy systems with ESSs [4-
21 8]. Several studies have focused on the optimum sizing of hybrid systems with advanced
22 optimization techniques considering resource variability and load uncertainty [9-17]. Dynamic
23 behaviors of hybrid systems have been characterized through simulations and experiments [18-
24 20]. Recent studies have also analyzed the influence of the vehicle-to-grid strategies on the aging
25 behavior of LIBs and suggested the efficient energy management and control methods for
26 vehicle-to-grid systems [21,22].

27 However, the high cost of energy storage systems still limits their application, especially for
28 vehicle electrification, even though vehicle electrification improves fuel efficiency. Therefore,
29 increasing battery pack power and charge/discharge levels is needed in battery packs and can be
30 obtained with advanced estimation methods for state of charge (SOC) [23-34]. Prolonging the
31 lifetime of battery packs with improved state of health (SOH) algorithms [35-41] can also reduce
32 costs because the total capacity of battery cells and packs should be minimized to reduce vehicle
33 weight. Moreover, intelligent energy and power management strategies based on the analysis of
34 overall energy flows should be sought [42-48] as well as the design optimization of battery packs
35 [49-52] to ensure economic feasibility of electric vehicles (EVs). High efficiency and fast battery
36 charge systems also accelerate the successful deployment of EVs on the market [53,54]. In
37 addition, experimental investigations on the performances of battery packs are indispensable [55,
38 56].

1 These needs require more intense use of LIBs leading to a large amount of heat generation which
2 increases the temperature of battery cells. Such excessive heat generation and uneven
3 temperature distribution not only reduce the efficiency of LIBs but also accelerates the
4 degradation of their life [57]. Moreover, overheating, combustion, or even explosion can occur
5 with the increase of thermal energy in the battery cells and packs [58,59]. The wide operational
6 range of LIBs and the often unpredictable pattern of thermal loads imposed further complicate
7 thermal issues. For example, a restricted range between 20°C and 40°C has been reported for
8 optimal performance and lifespan of LIBs [60], even though the desirable operational range for
9 transportation applications is between -10°C to 50°C [61].

10 Considerable efforts have been devoted to obtain a fundamental understanding of dynamic
11 thermal mechanics and to seek innovative and practical solutions [62-66]. Numerical simulations
12 based on computational fluid dynamics and finite element methods have been conducted to
13 predict the thermodynamics of LIB cells and packs [67-71]. A variety of heat transfer models
14 have been created and validated through experiments [72-75]. Moreover, lumped parameter
15 thermal models with novel thermal management schemes have been proposed for next-
16 generation battery management systems [76-81]. Recent studies also suggested the temperature-
17 compensate model with dual-particle-filter state of charge estimation algorithm and the coupled
18 dynamic electric thermal model to account for the thermal characteristics of LIBs [82,83].

19 In particular, recent attention has been paid to thermal expansion of host materials and cells
20 because the volume change of cells due to temperature variations has a similar order of
21 magnitude as that of cells due to Li-ion intercalation [84,85]. Thus, periodic thermal stress and
22 strain significantly affect not only the performance of individually battery cells but also the
23 efficiency of the whole battery pack, and the fatigue life of cells and packs [86]. However,
24 investigations of thermal expansion and important parameters related to thermal mechanics on
25 the cell-level, which are crucial to predict thermal mechanics of battery cells, are still few,
26 making it difficult to estimate the thermal stress and strain due to temperature variations,
27 whereas Li-ion intercalation-induced stress and strain have been studied [87-90]. Moreover,
28 modeling thermal expansion coupled with the thermodynamics of cells and mechanical
29 constraints has not been investigated in great detail, although characterizing thermal expansion
30 has become important to prolong the lifespan of LIBs. The limited information and the difficulty
31 in measuring thermal stress/strain on the cell-level, especially in pack conditions, stimulate
32 researchers to develop a 3-D thermal swelling model with a coupled thermal-mechanical
33 analysis technique to quantify the thermal stress and strain.

34 In this paper, we report experimental characterizations and a numerical three-dimensional
35 thermal swelling model of Li-ion cells. High-precision displacement sensors were used to
36 measure the cell-level swelling on the surface arising from temperature elevation of an
37 unconstrained graphite/nickel-manganese-cobalt-oxide (NMC) cell whose temperature was
38 regulated in a thermal chamber. The swelling shape due to temperature variations was found to
39 be different than that due to Li-ion intercalation. This difference might be due to nonuniform

1 temperature distribution of the jellyroll and the generation of gaps/voids between electrodes. To
2 account for these characteristics of Li-ion cells, several important parameters were estimated
3 through experiments. Moreover, the thermal swelling model incorporated actual mechanical
4 constraints of cells and packs. The coupled thermal-structural analysis was performed to predict
5 the nonuniform temperature and the thermal swelling. Experimental validations at unconstrained
6 conditions, i.e. free swelling conditions, showed that the proposed model accurately predicted
7 the thermal mechanics of Li-ion cells due to thermal loads.

8 Based on the proposed model, thermal mechanics of battery packs were predicted at constrained
9 conditions, i.e. pack conditions. The solutions at constrained conditions showed that the
10 nonuniform temperature distribution of the jellyroll still played a critical role for the thermal
11 swelling shape at pack conditions, even though the gaps/voids did not occur at pack conditions
12 because of mechanical constraints on the battery cells. Further, high core temperatures were
13 found to increase the magnitude of thermal swelling, suggesting that the prediction of the core
14 temperature is important to predict the dynamic thermal mechanics, which can be used in power
15 management schemes. This nonuniform temperature also resulted in nonuniform thermal
16 swelling over the surface, whereas Li-ion intercalation-induced swelling was uniform over the
17 surface due to uniform Li-ion diffusion over the surface [92]. This nonuniform temperature
18 creates a significant thermally induced reaction force, suggesting that the reaction force easily
19 captures this thermal dynamic behavior, whereas the amount of swelling is so small that it is
20 hard to measure in pack conditions. This observation suggests that the force due to the volume
21 change of the battery cell can become a sensitive gauge for characterizing the operational state.

22 23 **2. Experiments**

24
25 This study used a flat-wound type prismatic 5Ah Li-ion cell. Detailed information of the cells is
26 available in Ref. [85]. The free swelling of the cell was measured with high-precision contact-
27 type displacement sensors with 1 μm accuracy and 0.1 μm resolution (Keyence GT2-H12KL,
28 Japan). The sensor head creates a contact force on the battery surface of less than 0.3 N. The
29 sensor was used to measure a variety of locations on the battery surface with a fixture (Figure 1).
30 The fixture was made of aluminum (distinct from our previous work [85,91,92]).

31 The prismatic cell was constrained at its eight corners in the fixture (using dimples at the rear
32 side, and aluminum spring pushers at the front side), but was otherwise unconstrained as shown
33 in Figure 1. The fixture was placed inside a thermal chamber (ESPEC BTZ-133, Japan) with a
34 controlled desired temperature. Three thermocouples were used. One thermocouple was installed
35 near the center of the battery cell surface to measure the cell surface temperature (at a location
36 which avoids interference with the displacement sensors). Another thermocouple was installed
37 on the fixture to measure temperature variations in the fixture due to heat exchanged with the
38 battery through the corner contacts. The third thermocouple was installed in the air between the

1 fixture and the cell to measure near-surface ambient temperature. Note as mentioned above, the
2 spatial temperature distribution was measured at the surface of the same battery cell [94]. The
3 temperature variation over the surface was observed to be less than 0.5 °C. Hence, measuring
4 more points on the surface of the case would not yield much information for this battery cell
5 considering that the accuracy of thermocouples is 0.5°C.

6 A preliminary test was conducted to check the repeatability, thermal hysteresis, and the isotropic
7 expansion of the aluminum fixture. An aluminum block, which has the same size as the battery
8 cell, was installed instead of the cell because the thermal expansion of aluminum is available.
9 The temperature of the thermal chamber was regulated at 25°C at first. Then, the temperature
10 was changed to 5°C and incrementally changed to 45°C with 10°C increments, and finally
11 returned to 25°C. Each temperature variation was followed by a two hour rest time to ensure
12 thermal equilibrium. Relative thermal expansion in the z direction (Figure 1) was measured at
13 three locations labeled as 5, 12, and 23 in Figure 1 for each side of the battery cell.

14 Pulse excitation experiments were carried out to induce heating of the battery cell and
15 characterize the resulting thermal swelling shape. The battery was fully charged using a standard
16 CCCV protocol [94] prior to discharge. Then, the battery was discharged with a 2 A (0.4 C)
17 current for 1.25 hours to obtain a 50% SOC. This procedure was repeated three times to check
18 repeatability. Two currents, 50A and 75A, were used for pulse excitations to identify the thermal
19 swelling shape at different temperature elevations. The entire experiment was repeated with
20 sensors placed at locations labeled as 1 ... 26 in Figure 1 (rear side). One sensor was installed as
21 a reference at the center of the opposite side labeled as 16 in Figure 1.

22 Note that the thermal time constant for the prismatic cell used is hard to estimate with a
23 characteristic length and thermal diffusivity considering the complex structure of the jellyroll
24 and other components inside the cell. To address this, we first chose the pulse period. Previous
25 studies [94] have shown that for the type of cell we are using, a 5 s (or shorter) pulse time is
26 short enough to approximate pure thermal swelling. However, the pulse period should also be
27 short enough to ensure that the SOC variation during a pulse is small enough as to not create
28 significant Li-intercalation swelling. Thus, we used a pulse period of just 1 s (five times shorter).
29 At 15 C rate, these pulses change the SOC by only approximately 0.2% over each period. Hence,
30 the pulse period of 1 s ensures pure thermal swelling (and is smaller than the associated
31 characteristic time) and assures that the SOC variation is small.

32 Once we established the pulse period, we used preliminary pulse excitation experiments to
33 determine the relaxation period needed for reaching thermal equilibrium. We measured the
34 swelling as a function of time while pulse excitation was on for a very long time (over 3 hours,
35 estimated from [85]). We used the measured swelling to estimate the thermal relaxation constant.
36 These preliminary experiments showed that the time constant was 340 s. With this value, the
37 thermal swelling converged to within 1 μm (accuracy limit of our displacement sensors) of its

equilibrium in about 35 minutes. To ensure a long enough relaxation time, we chose a relaxation time four times larger, namely 2 hours in all our subsequent experiments.

3. Phenomenological 3-D thermal swelling model

3.1. Constitutive equations

The total strain ϵ_{ij} consists of the mechanical strain ϵ_{ij}^{me} and two other types of strains as [95]

$$\epsilon_{ij} = \epsilon_{ij}^{me} + \epsilon_{ij}^T + \epsilon_{ij}^{Li}, \quad (1)$$

where ϵ_{ij}^T and ϵ_{ij}^{Li} denote the thermal strain and the diffusion induced strain. The mechanical strain can be expressed as [96]

$$\epsilon_{ij}^{me} = \frac{1}{E} ((1 + \nu)\sigma_{ij} - \nu\sigma_{kk}\delta_{ij}), \quad (2)$$

where E , ν , σ , and δ denote the modulus of elasticity, Poisson's ratio of the material, the stress components, and the Kronecker delta function respectively. The thermal strain can be expressed as [95]

$$\epsilon_{ij}^T = \alpha_{ij}\Delta T\delta_{ij}, \quad (3)$$

where α_{ij} and ΔT represent the coefficient of thermal expansion and the temperature variation. The diffusion induced strain (due to Li-ion intercalation) can be expressed as [95]

$$\epsilon_{ij}^{Li} = \frac{1}{3}\Delta c\Omega\delta_{ij}, \quad (4)$$

where Δc and Ω are the concentration variation of diffusion species and the partial molar volume representing volume expansion caused by the concentration of solute atoms. It should be noted that strains due to gas generation [97] or solid electrolyte interphase (SEI) formation [98] over long-term cycling are not considered in this work.

Equations (3) and (4) reveal an analogy between Li-ion intercalation-induced stress/strain and thermal stress/strain. Specifically, stresses caused by the diffusion of solute atoms lead to a stress-strain relation similar to that due to temperature gradients. Hence, Li-ion intercalation-induced stress/strain is theoretically analogous to thermal stress/strain in homogeneous isotropic materials. However, a different spatial distribution of strain can occur because of different spatial distributions between temperature ΔT and the Li-ion concentration Δc . The concentration of diffusion Δc is uniform at macro scale, especially at steady-state, suggesting that quasi-static experiments create a uniform concentration in the jellyroll for Li-ion diffusion. In contrast, the spatial distributions of temperature are clearly non-uniform, as observed in many previous

studies [99-101]. Hence, the spatial distribution of strain from temperature variations might be different than that from Li-ion intercalation. Note that swelling in the z direction (Figure 1) is of primary interest in this study because the Li-ion intercalation/deintercalation into the electrode occurs mainly in the direction perpendicular to the electrodes during charge and discharge. Moreover, X-ray tomography has shown that a gap exists between the case and the jellyroll in the x and y directions (Figure 1) [102], suggesting that the swelling of the jellyroll in the x and y directions is not observable in the surface of the case.

Contacts between the case and the jellyroll and between the case and the spacer further complicate the mechanical behavior. Contacts are nonlinear phenomena that affect the stiffness of the system, making it dependent on the contact status (areas of closed contact or gap). Therefore, these complex contacts create complex mechanical constraints which affect the overall swelling shape of battery cells [91].

In summary, constitutive equations theoretically suggest that the different shapes of Li-ion intercalation swelling and thermal swelling (details of experimental results are described in section 5) might occur for two reasons: nonuniform temperature distribution in cells [71], and generation of micro gaps/voids among electrodes [103]. This interpretation also suggests that these two phenomena should be accounted for the thermal swelling model. The coupled thermal-structural analysis, which is the distinct difference from previous studies, was performed to calculate nonuniform temperature distribution and thermal gradient of the jellyroll. Moreover, the jellyroll was modeled as two pieces and contact surfaces of two pieces allow separation in the center regions to account for the creation of micro gaps/voids. Details of the numerical three-dimensional thermal swelling model are described next.

3.2. Model description

The geometry of a prismatic battery cell was created to simulate the thermal mechanics by using detailed information of the aluminum case and jellyroll configurations (obtained in the past from the X-ray tomography and dissection). The configuration and geometry are the same as those used to develop the Li-ion intercalation swelling model in Ref. [92]. However, the jellyroll was modeled with two pieces of components (left sided figure in Figure 2), which is the key difference from Ref. [92].

Briefly, the jellyroll, shown as a yellow domain in Figure 2, is a roll over a flat surface instead of a cylindrical roll. One can envision a manufacturing process where the material is rolled onto a flat (rectangular) thin plate. The resulting shape is close to a cuboid, and far from a cylinder. This can also be viewed as a cylinder squeezed between two flat surfaces. Hence, the jellyroll is rolled but not cylindrical at all. It was modeled as a cuboid, meaning that the yellow domain

1 represents the jellyroll inside of the case, which consists of hundreds of cathode, anode, and separator layers, all resulting in equivalent mechanical and thermal properties of the jellyroll (Refs. [85] and [102] provide details of the jellyroll used in this prismatic battery cell). Note that the rounded parts of the jellyroll at its edges (top and bottom sides) do not contact the case; thereby, their swelling or presence has negligible effect the expansion of the case. Additional details regarding this modeling approach, its effectiveness and validation can be found in Refs. [71] and [92]. Note also that individually modeling each thin electrode layer of the jellyroll requires extremely heavy computational efforts considering the enormous increase in the mesh and the hundreds of contacts among electrodes. This feature is the one of the main reasons why there is still little study on the thermal mechanics at the cell-level. For example, the computational time for free swelling and constrained swelling with the proposed simple model takes over a day of CPU time to perform because of the several nonlinear contact conditions among components, even though the model is relatively simple compared with actual cells.

The size and location of the jellyroll inside a battery cell was estimated from the X-ray tomography [102]. Information from the dissection showed that the active material region is slightly larger than the overall contact region between the jellyroll and the battery case. Hence, the contact region was approximated as the active material region. To distinguish the contact region (the center region of the jellyroll, a purple domain in Figure 2) from the noncontact region (top and bottom of the jellyroll), a 100 μm gap was modeled at the top and bottom sides of the jellyroll to define the region where the jelly roll does not contact the case (see inset figure in Figure 2).

The jellyroll was modeled with two pieces of components (left sided figure in Figure 2) to account for the creation of micro gaps/voids. Note that micro gaps/voids might result from the combined effects of mechanical constraints and the expansion of each material layer. The jellyroll cannot experience any voids/gaps at its edges because the top and bottom sides are wound, and the left and right sides are clamped with current collectors and bus bars. In contrast, there is no bonding material between the electrode surfaces in the center region of the jellyroll and thereby gaps/voids easily occur there between electrodes. To account for these features, appropriate contact conditions were imposed to the two pieces of the jellyroll. Bonded contact was used at the four edges (top, bottom, left, and right edges) to account for the lack of separation at the top and bottom sides (due to the wound shape) and the clamping with current collectors and bus bars at the left and right sides (which do not allow separation between electrodes at the edges). In contrast, separation was allowed on the contact surface between the two pieces of the jellyroll, i.e. the contacted surface between piece A of the jellyroll and piece B of the jellyroll in Figure 2.

The case, shown as a transparent white domain in Figure 2, was modeled using measured dimensions. Two halves of a spacer, transparent blue domains in Figure 2, made from polybutylene terephthalate (PBT) were also modeled using measured dimensions and assembled with the cell (using contact with the cell) on each side of the battery cell to replicate the actual

1 pack conditions. Several battery cells and spacers are assembled in series in battery pack
2 conditions [102]. This repetitive structure ensures that the model with one cell constrained
3 between two half spacers simulates one of several cells and the corresponding spacers in pack
4 conditions with reduced computational effort. All components were modeled as homogeneous
5 solids. Other components such as the bus bars and terminals were not modeled (for simplicity),
6 assuming that these un-modeled components have relatively small and local effects. This
7 assumption was used and validated in Ref. [92].

8 No separation with slip was imposed between the jellyroll and the case, between the case and the
9 spacer, and between the spacer and the exterior walls considering the physical contacts of the
10 battery pack. Since relatively large deflections were applied, geometric nonlinearity was
11 considered in the solution process, i.e. the coordinates of the element nodes were updated during
12 the loading steps with respect to the current deformations. A Hex-dominant meshing method was
13 used in ANSYS to generate the mesh of the model. This method generated hexahedral meshes
14 for the jellyroll because of its simple geometry. In contrast, tetrahedral elements were created for
15 the case and the spacer because these components had more complex shapes than the jellyroll.

16 Three simulations were carried out with a structural analysis package of ANSYS. First, a
17 coupled thermal-structural analysis was conducted with the cell model consisting of the jellyroll
18 and the case to simulate the swelling of the battery cell at unconstrained conditions, i.e. free
19 swelling conditions. The resulting predictions were validated with measured swelling shapes.
20 Second, a structural analysis was conducted with the pack model consisting of four components,
21 namely the jellyroll, the case, and two half spacers, over an elevated temperature to predict the
22 thermal response of the battery cell at constrained conditions assuming that the jellyroll
23 expanded uniformly. The results were compared also to Li-ion intercalation swelling over SOC
24 to show the different shapes these two kinds of swelling create. Third, the coupled thermal-
25 structural analysis was conducted with the pack model at constrained conditions. The results
26 demonstrated that the thermal swelling shape obtained from the coupled thermal-structural
27 analysis is different than the thermal swelling shape from the uniform expansion of the jellyroll
28 (second simulation). Therefore, it is important to conduct the coupled thermal-structural analysis
29 to predict accurately the thermal mechanics of battery cells. This model also estimates the
30 thermally induced forces in pack conditions, which affect the lifetime of battery cells.

31 Specifically, thermal analysis was performed first with a constant heat generation in the jellyroll,
32 yellow domain in Figure 2, to estimate the nonuniform temperature distribution and the thermal
33 gradients in a battery cell. These results were fed into a structural analysis to estimate thermal
34 swelling. The amount of heat generated in the jellyroll due to entropy change and Joule heating
35 [104] was estimated from a coupled electro-thermal model [94]. Joule heating is the major
36 source of heat in pulse excitation experiments with a short pulse period because Joule heating is
37 irreversible, whereas entropy heat is reversible. Joule heating is exothermic regardless of the
38 charge process. Therefore, Joule heating is always a heat source. In contrast, entropy heat is
39 endothermic, i.e. heat sink during charge and exothermic, i.e. heat source during discharge.

Therefore, the total amount of heat from entropy heat in repeated charge/discharge cycles with a short period is approximately zero.

Heat conduction was imposed for all contacted surfaces between the two pieces of the jellyroll, between the jellyroll and the case, and between the case and the spacer. Heat convection was also imposed at the surface of the outer case to account for air cooling in the thermal chamber. Specifically, all outer surfaces of the case were modeled as exposed to heat convection with air (for unconstrained cell conditions) because heat exchange occurs at all outer surfaces of the case. In contrast, heat convection was imposed for the noncontact region of the case and the plastic spacers for constrained conditions because heat convection only occurs in the noncontact region of the case and the plastic spacers. The heat transfer coefficient (or film coefficient) h was calibrated to match the measured temperature at the center of the case. The estimated values were $58 \text{ Wm}^{-2}\text{K}^{-1}$ for 10 C pulse excitation experiment and $60.5 \text{ Wm}^{-2}\text{K}^{-1}$ for 15 C pulse excitation experiment. Note that the thermal chamber regulates the temperature and not the heat flow; a higher heat flow is needed for a faster cooling of battery cells to maintain constant temperature.

4. Estimation of properties

The structural and thermal properties used in the model are those listed in Table 1.

Table 1. Structural and thermal properties of a flat-wound type prismatic 5 Ah Li-ion cell used in this study. (See also Ref. [92] for more information on the evolution of the modulus of elasticity of the jellyroll)

Volume	Jellyroll	Case	Spacer
Material	Multiple	Aluminum	PBT
E [MPa]	6.5 (at 0.5 SOC)	68,000	3,600
ν	0.3	0.33	0.44
$\rho [\text{kg}\cdot\text{m}^{-3}]$	2,550	2,770	1,620
$\alpha [1\text{e}^{-4}\text{K}^{-1}]$	$13+0.29\Delta T$	0.23	0.41
k [$\text{Wm}^{-1}\text{K}^{-1}$]	x and y: 22 z: 0.43 (for center) and 9.5 (for left and right edges)	149	0.24
Reference	[92,107,108]	[109,110]	[111]

4.1. Structural properties

Microstructure transformations of electrodes lead to the evolution of material properties due to phase transitions [105,106]. In particular, layered structures such as graphite intercalation compounds show significant changes of material properties, whereas the spinel and olivine structures such as LiMn_2O_4 and LiFePO_4 vary minimally upon lithiation [107]. Our previous work also shows lithiation induced stiffening of the equivalent stiffness (and of the modulus of elasticity) of the jellyroll over SOC [92].

The estimated modulus of elasticity at 0.5 SOC is 6.5 MPa from Ref. [92]. This value was used for the jellyroll because the pulse excitations were carried out at 0.5 SOC. The value of the Poisson's ratio for the jellyroll was 0.3 in that the average Poisson's ratio value reported in the literature for all electrode materials is around 0.3 and does not significantly change with Li concentration [107]. Moreover, Poisson's ratio for a separator and electrolyte is 0.35 and 0.25 [108], suggesting that the equivalent Poisson's ratio of the jellyroll has a value around 0.3.

The modulus of elasticity and Poisson's ratio used for the aluminum case were 68 GPa and 0.33; the ANSYS material database provided property values for a variety of materials such as steel and aluminum taken from a variety of sources [109,110].

The modulus of elasticity and Poisson's ratio used for the spacer were 3.6 GPa and 0.44; the modulus of elasticity was measured from nanoindentation [92], and Poisson's ratio was available in Ref. [111]. Limited by the availability of published data, and given current understanding of thermal characteristics of structural properties for electrode materials, these structural properties were assumed to be constant in terms of temperature in the operational range of battery cells and packs at this stage of the work.

4.2. The equivalent coefficient of thermal expansion

There are two issues in the estimation of the equivalent coefficient of thermal expansion for the jellyroll: (1) determining whether the expansion is isotropic or orthotropic, and (2) revealing whether the equivalent coefficient of thermal expansion depends on temperature.

Thermal expansion in the through-plane direction (z direction in Figure 2) is different than that for the in-plane directions because of the layered structure of the jellyroll and despite the fact that each component of the jellyroll has isotropic thermal expansion characteristics. Cathodes, anodes, and separators are stacked in series for the through-plane direction. Therefore, the total expansion is the sum of the expansions of each electrode. In contrast, electrodes and separators are stacked in parallel for the in-plane direction. Hence, the separator plays a critical role in the in-plane direction because it also has the largest coefficient of thermal expansion. The anisotropy of the thermal expansion of polyethylene and polypropylene, which are the main materials for

separators, further complicates estimating the equivalent coefficient of thermal expansion of the jellyroll [112,113].

Quasi-static thermal swelling measured at the center of the battery cell (near location 12 in Figure 1, see also [91]) ranging from 25°C to 45°C was used to calibrate the equivalent coefficient of thermal expansion of the jellyroll for the through-plane direction (α_{thro}) in the model. Uniform temperature elevation, which was the same conditions for experiments, was imposed in the static structural analysis to the jellyroll and case from 25°C to 45°C with 5°C increments. Then, the equivalent coefficient of thermal expansion of the jellyroll for the through-plane (α_{thro}) was calibrated to match the experimental measurements as shown in Figure 3. The estimated equivalent coefficient of thermal expansion for the through-plane direction not only is similar in order of magnitude with that for polyethylene and polypropylene as aforementioned in Ref. [91], but also shows temperature dependency, which is the general thermal characteristic of polymer materials [114], suggesting that separators play a critical role in the thermal mechanics of the battery cell. This calibration also suggests modifying the constitutive equation for thermal swelling of the jellyroll as

$$\epsilon_{ij}^T = \alpha_{ij}(T)\Delta T\delta_{ij}. \quad (5)$$

This modification implies that the constitutive equation for thermal swelling of the jellyroll is nonlinear, whereas that of the case and the spacer is linear.

To estimate the equivalent coefficient of thermal expansion for the in-plane (α_{in}) and evaluate the effects of through-plane thermal expansion caused by in-plane thermal expansion, four sets of thermal expansion values ($\alpha_x, \alpha_y, \alpha_z$) were analyzed, namely values of (0, 0, 13), (0, 13, 13), (13, 0, 13), and (13, 13, 13) [10^{-4}K^{-1}] (axes x, y , and z are shown in Figure 1). Finite element analyses show that the swelling shapes are almost the same for all these cases. The maximum difference for swelling in the z direction is below 2 μm , which is below the repeatability of measurement (details for repeatability are shown in the next section). This small difference suggests that the effects of in-plane thermal expansion on the through-plane thermal expansion is negligible because no constraints are imposed on the jellyroll and there is a gap between the case and the jellyroll in the x and y directions.

In summary, the equivalent coefficient of thermal expansion of the jellyroll for the through-plane is the only important parameter in the thermal swelling model, and that coefficient is calibrated through experiments.

The coefficients of thermal expansion for aluminum and PBT were set to $23 \cdot 10^{-6} \text{ K}^{-1}$ (value from ANSYS datasheets) and $41 \cdot 10^{-6} \text{ K}^{-1}$ (value provided by the manufacturer). The case and spacer were assumed to expand isotopically.

4.3. Thermal conductivity

The thermal conductivity of the jellyroll depends on the direction of the heat flow [116]. The heat transfer is anisotropic because the current-collector layers within the jellyroll have much higher thermal diffusivity than the separator. Thus, the equivalent thermal conductivity of the jellyroll for the in-plane direction ($k_{eq_{in}}$) is higher than that for the through-plane direction ($k_{eq_{thro}}$). Theoretically, the equivalent thermal conductivity for the in-plane direction can be calculated using the following formula [116] (because cathodes, anodes, and separators are thermally stacked in parallel)

$$k_{eq_{in}} \sum t_i = \sum k_i t_i, \quad (6)$$

where k_i , and t_i denote the thermal conductivity and the thickness of each material layer. The equivalent thermal conductivity for the through-plane direction can be calculated using the following formula [116] (because electrodes and separators are thermally stacked in series)

$$\frac{\sum t_i}{k_{eq_{thro}}} = \sum \frac{t_i}{k_i}. \quad (7)$$

Equations (6) and (7) show how anisotropic thermal conductivity can be taken into account in a macro-homogeneous thermal model of the jellyroll. It can be inferred from these equations that the material, which features the lowest thermal conductivity, influences the overall thermal characteristics the most, especially for the through-plane direction. Therefore, separators and electrolytes play a critical role for heat flow in the through-plane direction because the thermal conductivity for separators and electrolytes is much smaller than that for other materials [108].

The equivalent thermal conductivity of the jellyroll for the in-plane direction is $22 \text{ Wm}^{-1}\text{K}^{-1}$ (value from Ref. [94]). The estimation of the equivalent thermal conductivity for the through-plane direction is more complicated. The jellyroll is composed of layers thermally connected in series with the case. Moreover, current collectors only exist at the left and right sides of the jellyroll, whereas all components are thermally connected in series in the jellyroll, suggesting that the equivalent thermal conductivity is different in the left and right sides of the jellyroll compared to the central area.

To calculate the thermal conductivity for the each region, three-step calculations were conducted. First, the equivalent thermal conductivity of the jellyroll for the through-plane direction was estimated using Eq. (7) with the equivalent thermal conductivity of the cell estimated from our previous work [91] ($0.73 \text{ Wm}^{-1}\text{K}^{-1}$). The case and jellyroll are thermally connected in series in the z direction. The resulting estimated equivalent thermal conductivity of the jellyroll for the through-plane direction is $0.65 \text{ Wm}^{-1}\text{K}^{-1}$. Second, the equivalent thermal conductivity for the central region was calibrated to match the magnitude of thermal swelling for 10 C pulse experiments. The resulting value was $0.43 \text{ Wm}^{-1}\text{K}^{-1}$. Third, the equivalent thermal conductivity

for the edge region was calculated using Eq. (6) with the value of the equivalent thermal conductivity for the entire jellyroll of $0.65 \text{ Wm}^{-1}\text{K}^{-1}$ and the value of the equivalent thermal conductivity for the center region of $0.43 \text{ Wm}^{-1}\text{K}^{-1}$. This calculation can be performed because the edge region and the central region can be considered as thermally connected in parallel in the z direction. The resulting estimate for the equivalent thermal conductivity of the jellyroll for the edge region was $9.5 \text{ Wm}^{-1}\text{K}^{-1}$. Finally, these estimated values were validated with different experiments, i.e. 15 C pulse excitation experiments (details are shown in the next section).

Note that the equivalent thermal conductivity of the jellyroll for the edge region is much higher because separators do not exist at the edge region. The dependency of the thermal conductivity on temperature is also assumed to be negligible in the operational range because measured thermal conductivities of the separator at 25°C and 45°C show a small difference [117]. Note again that the thermal conductivity of separators is dominant and strongly affects the overall heat flow, especially for the through-plane direction, because it has the lowest value.

The thermal conductivity for the aluminum case and PBT spacer was set to $149 \text{ Wm}^{-1}\text{K}^{-1}$ (value from ANSYS datasheets) and $0.24 \text{ Wm}^{-1}\text{K}^{-1}$ (value from Ref. [111]). It was assumed that the temperature dependency of thermal conductivity is negligible for both materials.

5. Results and Discussion

5.1. Characterization of the thermal behavior of the fixture

Figure 4 shows the measured displacement from a preliminary test performed with an aluminum fixture and an aluminum block. Lines show measurements from the front side in Figure 1, while dashed lines show measurements from the rear side. The initial displacement at 25°C was set to zero (sensor measure relative displacements). Measured data from all sensors converge to zero when the temperature was changed from other values back to 25°C . The maximum difference observed at 25°C is below $1\mu\text{m}$, which is the accuracy of the displacement sensor. Measured displacements decrease when temperature increases. The fixture holds the displacement sensor, and hence the sensor indicates a lower value when the fixture expands. Measurements at the same face show the same displacement, suggesting that the fixture expands isotopically.

Measurements from the front side are slightly different than those from the rear side. This difference is because of (1) the different fixture length for each side and (2) the architecture of the fixture which allows the aluminum block to move into one direction (by the use of spring pushers at its corners). Note that thermal expansion depends on the material length, i.e. $\Delta L = \alpha L \Delta T$, as well as temperature variation. Hence, measurements from the front side can be expressed as

$$\Delta L_{measured}^{rear} = -\Delta L_{fixture}^{rear} + \Delta L_{sensor}, \quad (8)$$

where $\Delta L_{measured}^{rear}$, $\Delta L_{fixture}^{rear}$, and ΔL_{sensor} are the expansions measured from a sensor at the rear side, the expansions of the fixture at the rear side, and the expansions of the sensor respectively. Similarly, measurements from the front side can be expressed as

$$\Delta L_{measured}^{front} = -\Delta L_{fixture}^{front} + \Delta L_{sensor} + \Delta L_{Al}, \quad (9)$$

where $\Delta L_{measured}^{front}$, $\Delta L_{fixture}^{front}$, and ΔL_{Al} are the expansion measured from a sensor at the front side, the expansions of the fixture at the front side, and the expansion of the aluminum block. Thus, measurements from the rear include the expansion of the fixture and that of the sensor (Eq. (8)). In contrast, measurements from the front side include the expansion of the fixture, that of the sensor, and that of the aluminum block (Eq. (9)). The thermal characteristic of the sensor is available in Ref. [91].

The thermal expansion of the fixture for both sides was calculated with measured values of $\Delta L_{measured}^{front}$ and $\Delta L_{measured}^{rear}$ by using Eqs. (8) and (9). Results are shown in the inset of Figure 4. The thermal expansion varies linearly with the temperature in the temperature range. In this inset figure, the slope is αL because the thermal expansion is $\Delta L = \alpha L \Delta T$; L_F and L_R denote length of the fixture in the z direction (Figure 1) for the front and rear sides. To verify the applicability of these equations and the accuracy of the measured L values for each side, the coefficient of thermal expansion was calculated for six locations, and the estimated α was compared to the known value for aluminum. The average and standard deviation are $23.8 [10^{-6}K^{-1}]$ and $0.5 [10^{-6}K^{-1}]$, whereas the coefficient of thermal expansion for the aluminum used is $23.6 [10^{-6}K^{-1}]$. The maximum and minimum values calculated/identified for the coefficient of thermal expansion were $24.4 [10^{-6}K^{-1}]$ and $22.9 [10^{-6}K^{-1}]$, suggesting that the difference is negligible.

5.2. Thermal swelling at free conditions

The temperature of the fixture increased during the pulse excitation because heat flows from the battery cell to the fixture through contacts. Hence, the fixture thermally expanded, and the measured data should be calibrated to accurately estimate the thermal swelling of the battery cell. Equation (9) is thus modified when experiments are conducted with the battery cell. The pure thermal swelling of the battery cell measured at the rear side was calibrated with the following equation

$$\Delta L_{measured} = -\Delta L_{fixture} + \Delta L_{sensor} + \Delta L_{cell}, \quad (10)$$

where ΔL_{cell} is the relative thermal expansion of the battery cell at a sensor location with respect to the reference point of the fixture (in Figure 1) for the rear side. Note that the swelling

measured at the front side edge of the case is not zero because the temperature of the edge also increases due to heat conduction, and because the stiffness of the spring pushers is low. However, the swelling at the front side edge of the case is negligible because the aluminum case is empty near the edges, and thus the thermal expansion of the aluminum case within 10°C temperature variation is below 1 μm . (The difference of the measured thermal swelling between the front and rear sides is shown in Figure 4). However, the 2-D thermal swelling shape was created with data measured at the rear side (to minimize uncertainty due to measurement errors). This feature explains why the measured data from the front side were used only as a reference. This reference was always measured during the experiment to check the variability in measuring the thermal swelling.

Figure 5 (a) illustrates the 2-D thermal swelling shape obtained from usage-induced elevated temperature, i.e. experiment for 50A (10C) pulse excitation. The swelling shape does not have flat regions, which differs from the shape of Li-ion intercalation swelling [92]. This difference might be due to two physics behind the cell's expansion as aforementioned.

First, Li-ion intercalation swelling is caused by orthotropic expansion, and is uniform across the jellyroll. Hence, that swelling occurs mainly in a direction perpendicular to the electrodes, which leads to a flat shape in the central region. In contrast, usage-induced temperature fluctuations are not uniform. For example, the center region is the hottest, whereas the edge region is the coolest due to the different thermal boundary conditions (Figure 6 (a)), even though the variation is not very significant. This small variation on the surface suggests that monitoring temperature of the surface center is enough for predicting thermal swelling. The spatial temperature distribution measured at the surface of the same battery cell in Ref. [94] also supports this hypothesis; temperature variation over the surface is less than 0.5°C at low thermal convection conditions. Hence, measuring many points on the surface is not very informative for this battery cell considering that the accuracy of thermocouples is 0.5°C. In contrast, thermal gradients are more significant through the thickness of the battery cell, i.e. the z direction in Figure 1. The core temperature of the jellyroll increases up to 5°C from 25°C although the surface temperature of the case increases only up to 3°C (Figure 6 (c)). This significant temperature variation through the battery cells leads to nonuniform thermal swelling of the jellyroll and thereby different thermal swelling results on the surface of the battery cell.

Second, the different mechanical constraints applied to the jellyroll at the edge and in the central region create gaps/voids. The model prediction clearly shows the creation of the gaps/voids, and therefore agrees with measurements as shown in Figure 5 (b). For easy comparison, the same legend is used for Figure 5 (a) and (b). The maximum gap is 15 μm at the center (a value which is approximately 30% of the maximum measured swelling) and has a shape similar to the swelling of the case (not shown here for the sake of brevity), suggesting that gaps/voids play a critical role for thermal swelling at unconstrained conditions.

Figure 5 (c) shows the uncertainty and the repeatability of the 10C pulse experiments for all locations. The variability in the measured swelling at the rear face is generally below 2 μm (the bottom of Figure 5 (c)). The difference between the maximum and minimum measured swelling and the standard deviation of swelling at a reference location (on the front face of the cell, labeled 16 in Figure 1) are 3.2 μm and 1.1 μm . These results confirm that the experiments are reliable and repeatable considering the accuracy of the displacement sensor and the noise of the thermal couples used to control the environmental temperature.

The model prediction (Figure 5 (e)) was validated with 15 C pulse excitation experiments (Figure 5 (d)) because the equivalent thermal conductivity for the through-plane direction was estimated from 10 C pulse excitation experiment. The model predictions are in excellent agreement with the measurements. The thermal swelling at 15 C pulse excitation is more rounded in the edge regions in comparison to the thermal swelling at 10 C pulse excitation. That can be explained by the fact that a large amount of swelling fills/eliminates more gaps/voids at the edges of the jellyroll, and thus the shape is more smooth and rounded. However, this shape is still convex and elliptical. That shape differs from the shape of Li-ion intercalation swelling, suggesting that nonuniform temperature distribution and gaps/voids effect contributes to the shape of thermal swelling. The maximum gap is 38 μm in the center, a value which is approximately 30% of the maximum swelling measured at the center.

Figure 5 (f) shows the uncertainty and repeatability of the 15 C pulse experiments for all locations. The variability in the measured swelling at the rear face is generally below 3 μm (the bottom of Figure 5 (f)). The difference between the maximum and minimum measurements and the standard deviation of swelling at a reference location (on the front face of the cell, labeled 16 in Figure 1) are 4.3 μm and 1.4 μm . These re-confirm that experiments are reliable and repeatable.

One more interesting phenomenon is shown in the temperature measurements and observed in the thermal analysis. The surface temperature increases 3°C for 10 C pulse excitation and 6°C for 15 C pulse excitation, whereas the temperature at the center of the battery cell increases over 20°C [94] with similar pulse profiles from initial temperature, i.e. 25°C. This difference clearly demonstrates the importance of cooling the battery cell. The estimated heat transfer coefficient in this study is around 60 $\text{Wm}^{-2}\text{K}^{-1}$, whereas that in Ref. [94] is to 6 $\text{Wm}^{-2}\text{K}^{-1}$ (a value similar to a natural convection). The cooling capacity for the chamber used in this study was ten times larger than that used in Ref. [94]. Therefore, it can be inferred that the heat dissipation from the appropriate thermal management mitigates the thermal stress and strain. On the contrary, this strong forced convection and the lower thermal conductivity of the jellyroll result in greater temperature gradients through the battery cell (the bottom of Figure 6 (c) and (d)). The core temperature is 4°C higher than the maximum surface temperature in this study. That is because a forced convection results in a lower, but more severe nonuniform, temperature distribution through the battery cell due to the lower Biot number [71]. Nonuniform degradation might be induced by this nonuniform temperature distribution in the jellyroll. However, the average

temperature is a stronger determinant of cell aging than temperature uniformity [118]. Hence, a thermal management system with forced convection is expected to improve battery lifetime.

5.3. Thermal swelling in the pack condition

The new 3-D model proposed is useful for simulating a variety of operational conditions. The model is particularly useful for sensor placement in thermal sensing or in stress sensing used in emerging methods for battery health management, for state of health estimation, and for state of charge estimation. Based on the battery cell model, optimal location or optimal number of sensors can be determined.

Another related example showing the effectiveness of the proposed model is the simulation of a battery pack to predict the coupled thermal-mechanical effects on the battery cell such as the nonuniform temperature distribution inside the cell and the resulting thermal stress/strain. To observe the thermodynamic-related phenomena, the approach proposed in this paper was used for calculating the thermal swelling at pack conditions.

The results presented are also useful, in addition to the proposed methodology. For example, the results shown in Figure 7 (structural analysis with uniform thermal expansion of the jellyroll, below) and in Figure 8 (coupled thermal-structural analysis, below) allow for the first time an accurate, quantitative comparison between swelling due to temperature elevation and swelling due to Li-ion intercalation. This comparison shows the difference of these two swelling shapes and also quantifies the swelling from these two different sources. The comparison between the uniform thermal swelling and nonuniform swelling shows the importance of the coupled thermal-structural analysis because nonuniform temperature distribution and mechanical constraints result in significantly different thermal stress and strain in pack conditions.

For example, Figure 7 (a) compares the swelling due to uniform temperature variations with the swelling due to Li-ion intercalation in the center region, i.e. in a region without contact with the spacer, at pack conditions. The Li-ion intercalation swelling over SOC was obtained from Ref. [92] assuming that the cell was discharged with 0.2 C. Ref. [92] already showed that the discharge with 0.2 C (1A) did not cause significant heating. The battery-cell surface remained within 0.1°C of the 25°C ambient temperature during discharge. Data at 0.2 C therefore allow direct correlations to be made between swelling and Li-ion intercalation in a cell sandwich without significant convolution by thermal expansion. Diamonds denote Li-ion intercalation swelling over SOC (the bottom axis of Figure 7 (a)), while circles denote thermal swelling over temperature elevation (the top axis of Figure 7 (a)) for each side.

Thermal swelling shows a weak nonlinear behavior over the temperature elevation because the constitutive equation of the jellyroll is weakly nonlinear (Eq.(5)). In contrast, Li-ion intercalation

swelling shows a strong nonlinear behavior over SOC due to phase transitions. Moreover, the swelling due to temperature elevation is similar in order of magnitude with the swelling due to Li-ion intercalation, suggesting that thermal stress and strain are very important for the safety and the lifetime of the battery.

In addition, Figure 7 (b) shows the swelling shape on the surface of the case at an elevated temperature and pack conditions. The swelling shape is the same as that due to Li-ion intercalation. This similarity can be explained by the fact that large gaps/voids cannot be created in pack conditions between electrodes in the center due to the spacer which compresses the case. The constitutive equation of thermal strain is theoretically identical to that of diffusion-induced strain when no gaps/voids occur. Relatively large and uniform swelling is observed in the center region in between areas of contact with the spacer. This observation reveals a key fact, namely that geometric constraints on the battery cell swelling due to spacers are important in pack conditions. Moreover, the pattern of swelling is repetitive, and the amount of swelling (especially in the areas where there is no contact with the spacers) is the same as in the center region (where there is contact between the jellyroll and the casing). Thus, an appropriate location for placing displacement or strain sensors is in the center region, at a location without contact with the spacer, even in pack conditions. However, the quantitative swelling is very small, which requires high-precision displacement or strain sensors. This small amount of swelling at constrained conditions suggests that the force due to the volume change of the battery cell can be a great alternative to characterizing the dynamic operational states.

To characterize the thermal mechanics and identify the discrepancy from a uniform thermal swelling at the battery pack during operation, the steady state response of 15 C pulse excitation at pack condition was predicted (Figure 8) through the coupled thermal-structural analysis.

Figures 8 (a) and (b) show the temperature distribution of the surface on the cell and the spacer in the x - y plane. For clarity, Figure 8 (a) uses the same legend as Figure 6 (b). The overall trend is the same. However, slightly lower temperature distribution (around 0.1°C to 0.2°C lower) is observed in Figure 8 (a), even though the contact surface of the case was reduced due to the contact surface with the spacer. In addition, the contact surface of the spacer has the same temperature as the surface of the case, while the surfaces in contact with air have the same temperature as the ambient temperature (Figure 8 (b)). These results can be explained by the fact that the spacer also emits heat, which is transferred from the case to the environment.

Figure 8 (c) shows the thermal swelling of the case. As expected (from the thermal swelling shape at free swelling conditions), a larger and more nonuniform swelling is observed in the central regions compared to the same tests at constrained conditions (Figure 7 (c)). Remarkably, the order of magnitude of the swelling due to pulse excitations is much larger than that due to uniform temperature changes, even though the surface temperature of the case is the same. This observation supports the hypothesis that the thermal swelling shape of the surface can be mainly attributed to the nonuniform temperature distribution of the jellyroll as well as the creation of

gaps/voids between electrodes. However, gaps/voids are not created at constrained conditions because of the force exerted by the spacer on the case, suggesting that nonuniform temperature is only important at constrained conditions. Further, the ratio between the nonuniform swelling and uniform swelling at the center is similar to the ratio between the core temperature and the surface temperature, suggesting that measuring thermal swelling can be a useful mean to predict the core temperature.

The estimated reaction force shows more significant difference than swelling; 193 N for uniform temperature and 747 N for nonuniform temperature of the jellyroll even if the surface temperature of the case is the same. This large difference suggests again that the measured force can be a sensitive metric for identifying the state of battery cells and packs.

The thermal behavior of the spacer is also different. The spacer thermally expands in the pulse excitation case (Figure 8 (d)). In contrast, the spacer is compressed in the case of uniform temperature changes (Figure 7 (c)). These features might be because the temperature of the spacer increased in the pulse excitation case, while the spacer remains at a constant temperature (same as the ambient temperature) in the case of uniform temperature changes.

In summary, a coupled thermal-structural analyses should be carried out with approaches such as the proposed model to predict accurately the thermal mechanics including the thermal swelling during operation at pack conditions. This analysis also suggests not only that the thermal gradient and the core temperature through the battery can be estimated from thermal swelling, but also that the measured force can be a more sensitive metric for identifying the dynamic state. This has significant implications for emerging force-based methods for state of health and state of charge estimation, that are expected to have a significant impact on the operation of modern LIBs in transportation applications (e.g. electrified vehicles).

6. Conclusions

The thermal mechanics of a commercial prismatic battery cell were investigated via simulations and experiments. To characterize the critical properties of the thermal swelling, pulse excitation experiments with constant SOC were carried out. Different shapes were found to occur when swelling is caused by thermal variations compared to the shapes from electrochemical reactions, i.e. Li-ion intercalation. A nonuniform temperature distribution (especially through the thickness of the battery cell) and gaps/voids create this difference. Based on the experimental characterization, a macroscopic finite element model was proposed. Comparisons between experiments and model predictions successfully demonstrated that the model accurately predicted thermal swelling at unconstrained conditions. Simulations of thermal swelling at pack conditions showed that elevated core temperature and nonuniform temperature distributions play a critical role, whereas gaps/voids disappear at pack conditions due to the constraints caused by the spacer, suggesting that the coupled thermal-structural analysis is indispensable to accurately

1 predict the thermal swelling during operation at pack conditions. In future work, a variety of
2 simulations can be carried out with the proposed thermal swelling model. These include analyses
3 of dynamic thermal mechanics at different cooling conditions, studies of thermal runaway, and
4 investigations of nonuniform degradation induced by temperature distributions in the jellyroll.

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Figure captions

Figure 1. Schematic diagram of the experimental setup showing the fixture, the cell, and the sensor locations 1...26.

Figure 2. The geometry of the prismatic battery cell used is shown. The yellow, transparent white, and transparent blue domains represent the jellyroll, the case, and the half spacer respectively. The purple domain represents the contact region between the jellyroll and the case. The inset figure shows the micro-gap at the top and bottom sides of the jellyroll.

Figure 3. Quasi-static thermal swelling at the center of the battery cell; symbols show experimental measurements, while the solid line shows simulation results.

Figure 4. Thermal expansion of the fixture with an aluminum block due to temperature variations; solid lines show the measured thermal expansion measured at locations labeled 5, 12, and 23 on the front side in Figure 1, whereas dashed lines show measured thermal expansion at locations labeled 5, 12, and 23 on the rear side in Figure 1.

Figure 5. Thermal swelling shape on the surface at 50 A (10 C) pulse excitation from (a) measurements, (b) model predictions, (c) the top shows the maximum, average, and minimum measured swelling at each location, the bottom shows the difference between maximum and minimum measured swelling at each location for 50A pulse excitation. Thermal swelling shape on the surface at 75 A (15 C) pulse excitation for (d) measurements, (e) model predictions, (f) the top shows the maximum, average, and minimum measured swelling at each location, the bottom shows the difference between maximum and minimum measured swelling at each location for 75 A pulse excitation.

Figure 6. Temperature distribution on the case surface at (a) 50 A, and (b) 75 A pulse excitation; front view and cross-section view along the centerline of local temperature distribution for the jellyroll at (c) 50 A and (d) 75 A pulse excitation.

Figure 7. (a) Swelling in the center between the areas of contact with the spacer from two different sources at pack conditions; Li-ion intercalation (diamonds) and temperature elevation (circles). Swelling on the surface of (b) the case and (c) the spacer from the elevated temperature (ΔT : 6 °C) in the xy plane at pack conditions.

Figure 8. Temperature distribution of the surface on (a) the cell and (b) the spacer in the xy plane during a 15 C pulse excitation at pack conditions. Swelling of the surface on (a) the cell and (b) the spacer in the xy plane during a 15 C pulse excitation at pack conditions.

Figure 1

Figure 2

Two pieces of the jellyroll

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8