MEASUREMENT OF THE EFFECTIVE RADIAL THERMAL CONDUCTIVITIES OF 18650 AND 26650 LITHIUM-ION BATTERY CELLS

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ABSTRACT

Harmful incidents caused by lithium-ion batteries in the past decade have inspired research on the thermal management of these batteries. Several recent studies have developed thermal models of lithium-ion cells, and a key consideration in the models is the effective radial thermal conductivity of the cell. Prior studies use an effective conductivity close to 1 W/m-K, which accounts for all of the solid layers of the cell connected in series with no contact resistance between the layers. In a recent paper by Drake et al.[1], the radial thermal conductivities of 18650 and 26650 lithium-ion cells were theoretically inferred from transient thermal measurements to be 0.20 ± 0.01 and 0.15 ± 0.01 W m^{-1} K^{-1}, respectively. While researchers have used these values in their papers, no direct, steady state experimental values are found in the literature. After disassembling and modeling the cells, we were able to heat the center of the cells with nichrome wire and a power supply, and experimentally measure their radial thermal conductivities. For the 18650 cell, we calculated a thermal conductivity of 0.43 ± 0.07 W m^{-1} K^{-1}, while for the 22650 cell, we calculated a thermal conductivity of 0.20 ± 0.04 W m^{-1} K^{-1}. Our thermal conductivity values include the effects of the various solid layers as well as the interfaces between these layers. Both of our measured values are larger than Drake's reported values and they are significantly smaller than the values reported with perfect thermal contact between the layers. This latter finding suggests that including realistic, non-ideal, contact coefficients from layer-to-layer is important when modeling the radial transport of heat in cylindrical lithium ion battery cells.

INTRODUCTION

In recent years there has been increased scrutiny of lithium-ion batteries, partly because of incidents in which they have caused harmful fires. One such incident occurred in September of 2010, when the lithium batteries inside a Boeing 747-400F cargo aircraft near Dubai caught on fire, killing both of the crewmembers inside the airplane. Since 2006, there have been numerous mobile phone fires caused by their small lithium-ion batteries. The much-publicized incidents of the 2016 Samsung Galaxy S7 phones catching fire serve as a recent example. The cause of these fires is thermal runaway, a term that describes the rapid increase in temperature caused when the energy generated within a cell is larger than what can be dissipated by the cell[2-4]. Another problem with thermal runaway is that it can easily propagate from one cell to the next. This means that thermal runaway in one cell can proliferate to all of the cells in the battery, leading to a much more energetic and potentially catastrophic event. It is therefore important to understand the thermal pathway from cell-to-cell to develop the means to prevent propagation of a cell failure.

To gain further understanding of thermal runaway, researchers have developed analytical thermal models of individual lithium-ion cells and the batteries they comprise. A key consideration in these models is the effective radial thermal conductivity of the cell. The electrochemical portion of the cell is comprised of a many-layered "winding" which makes estimation of the effective radial thermal conductivity of the device difficult. Recently, Tanaka[5] used an effective radial conductivity of 1.02 W m^{-1} K^{-1} and Coman, et al.[6] used a radial thermal conductivity of 3.4 W m^{-1} K^{-1}. In each of these cases, the value accounts for the conductivities of the various layers of the winding of the cell but ignores the thermal contact resistance from layer to layer. Drake et al.[1] used an analytical model for the expected temperature curve as a function of time when a cylindrical Li-ion cell is subjected to radial heating on one of its outer surfaces. They determined the radial thermal conductivities of 18650 and 22650 lithium-ion cells by comparing their analytical results to the measured temperature response curves for both types of cells and obtained thermal conductivities of 0.20 ± 0.01 and 0.15 ± 0.01 W m^{-1} K^{-1}, respectively. Vishwakarma, et al.[7] present measured results from a flat 1-D layered geometry that indicate an effective conductivity of 0.24 W m^{-1} K^{-1} stating that the majority of the thermal resistance is in the contact from the cathode layer to the plastic separator layer.

In this paper, we directly measure the radial thermal conductivities of both 18650 and 22650 lithium-ion cells and compare the measured results to a thermal model for the layered radial geometry including the contact resistance from the winding of the cell to the outer wall of the cell as reported by Gaitonde, et al.[8]

DISASSEMBLY AND MODELING

Before designing an experiment to measure the radial thermal conductivities of 18650 and 22650 cells, we discharged and disassembled a total of ten cells, eight INR18650-25R cells and two LFP26650P (K226P01) cells. All the 18650 cells were manufactured by Samsung®, while all the 22650 cells were manufactured by K2 Energy Solutions, Inc. To drain as much power from the cells as possible, we connected them to both 13.1 ohm and 5.6 ohm resistors over a few days. Then, using a dremel tool and vice, we cut off the top and bottom of each cell, making sure not to harm the cell's winding in the process. One important finding we made was that not all 18650 cells are constructed in the same way, even those made by the same manufacturer. Table 1 summarizes information about the ten cells we cut open, including notable observations. It is apparent that although all the 18650 cells were made by Samsung®, those with dark green plastic covers had metal spindles at the center of the winding and those with light green plastic covers had no spindles. For the sake of consistency, we only used 18650 cells with dark green plastic covers and spindles in our experiments. As for the 22650 cells, a notable finding was that some electrolyte bubbled out of these cells when they were being cut, which was not the case for the 18650 cells.

We then created simple thermal models of the cells using the equation for the thermal resistance of a cylindrical layer given by Fourier's Law:

$$R_{cyl} = \frac{\ln\left(\frac{R_2}{R_1}\right)}{2\pi k_1 L} \qquad (1)$$

Specifically, we measured the thickness of each cathode, anode, and plastic separator layer inside the winding of the cells. We categorized the layers by calling a bundle of four layers a "sheet" (see Figure 1). The layered structure of a "sheet" is identical for 18650 and 22650 cells: anode layer consisting of copper foil and copper foil coating (usually carbonaceous electrode), plastic separator, cathode layer consisting of aluminum foil and aluminum foil coating (usually $LiCoO_2$), and plastic separator. For the 18650 cell winding, we counted approximately 28 "sheets", while for the 22650 cell winding, we counted approximately 38 "sheets". Using these data and the thermal conductivities of the layers given by Chen, et al.[9], we calculated the thermal resistance of each cylindrical layer using Equation (1). This allowed us to calculate the cumulative resistance of the layers as a function of cell radius for both 18650 and 22650 cells, the graphs of which are plotted in Figure 2 and Figure 3. Note that three of the last four plotted points come from the respective thermal resistances of the extra plastic layer between the winding and case, the case itself, and the green plastic cover.

Using these data as well as contact resistances between the winding and case of the cell, anode and plastic separator, and cathode and plastic separator measured by previous researchers[7-8], we predicted an effective radial thermal conductivity of the cell. According to our models, this value was around 0.27 W/m-K for the 18650 cell and around 0.22 W/m-K for the 22650 cell. Using our model, we calculated the thermal conductivity of the 18650 cell disregarding the contact resistances from layer to layer within the winding and obtained 1.4 W/m-K, which is in the range of values that Dr. Tanaka and Coman et al. used in their papers[5-6].

Table 1. Summary of Ten Disassembled Cells

	Cell Type	Nominal Voltage (in V)	Observations
Cell 1	Li-ion, INR18650-25R	3.6	Dark green plastic cover; Had spindle
Cell 2	Li-ion, INR18650-25R	3.6	Light green plastic cover; Had no spindle
Cell 4	Li-ion, INR18650-25R	3.6	Dark green plastic cover; Had spindle
Cell 5	Li-ion, INR18650-25R	3.6	Light green plastic cover; Had no spindle
Cell 6	Li-ion, INR18650-25R	3.6	Light green plastic cover; Had no spindle
Cell 7	Li-ion, INR18650-25R	3.6	Dark green plastic cover; Had spindle
Cell 8	Li-ion, LFP26650P (K226P01)	3.2	Electrolyte bubbled out during cutting; Had no spindle

Cell 12	Li-ion, INR18650-25R	3.6	Dark green plastic cover; Had spindle
Cell 13	Li-ion, INR18650-25R	3.6	Dark green plastic cover; Had spindle
Cell 15	Li-ion, LFP26650P (K226P01)	3.2	Electrolyte bubbled out during cutting; Had no spindle

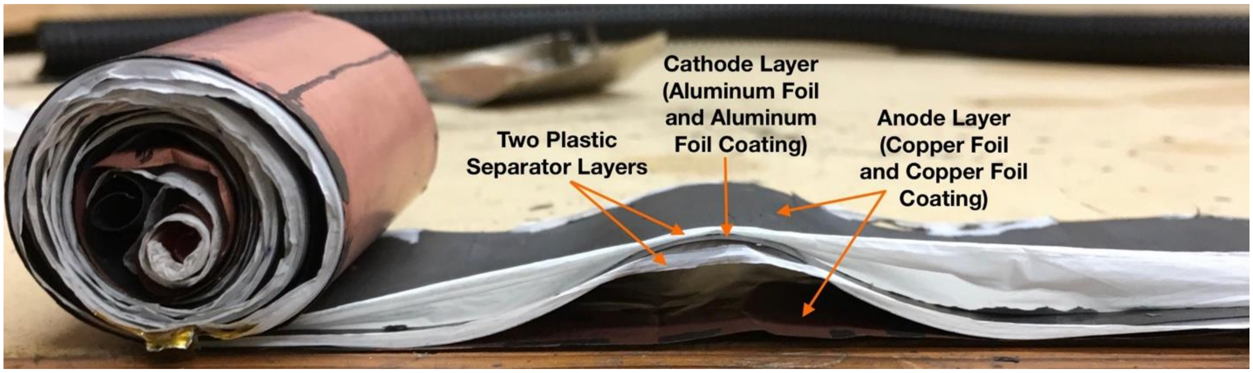

Figure 1. "Sheet" of four layers inside the winding of an 18650 cell.

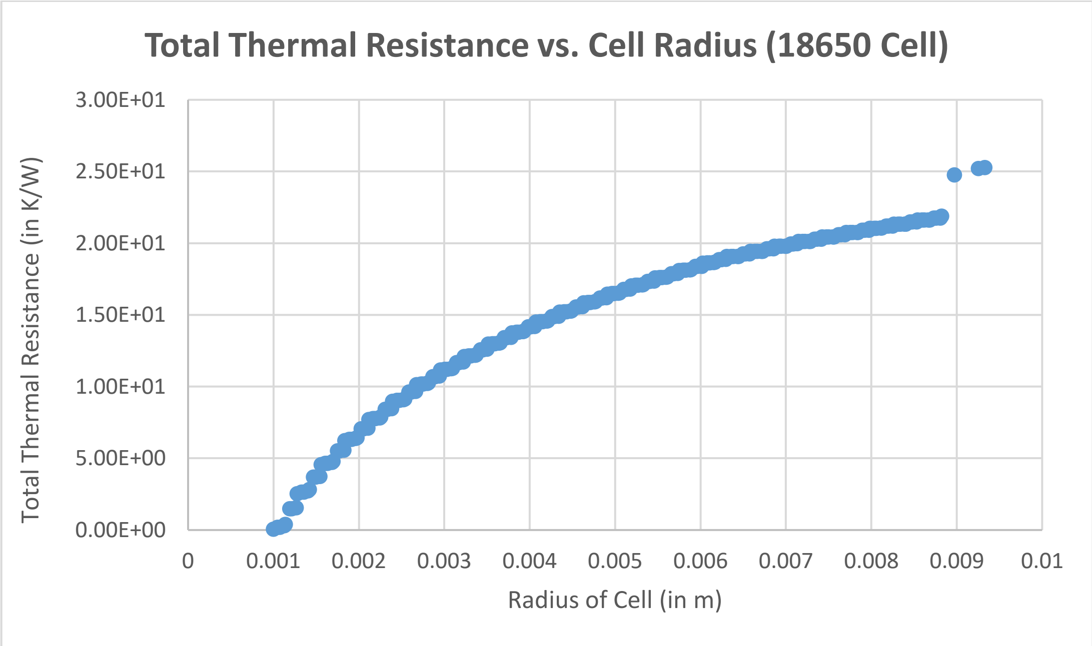

Figure 2. Plot of total thermal resistance versus cell radius for an 18650 cell.

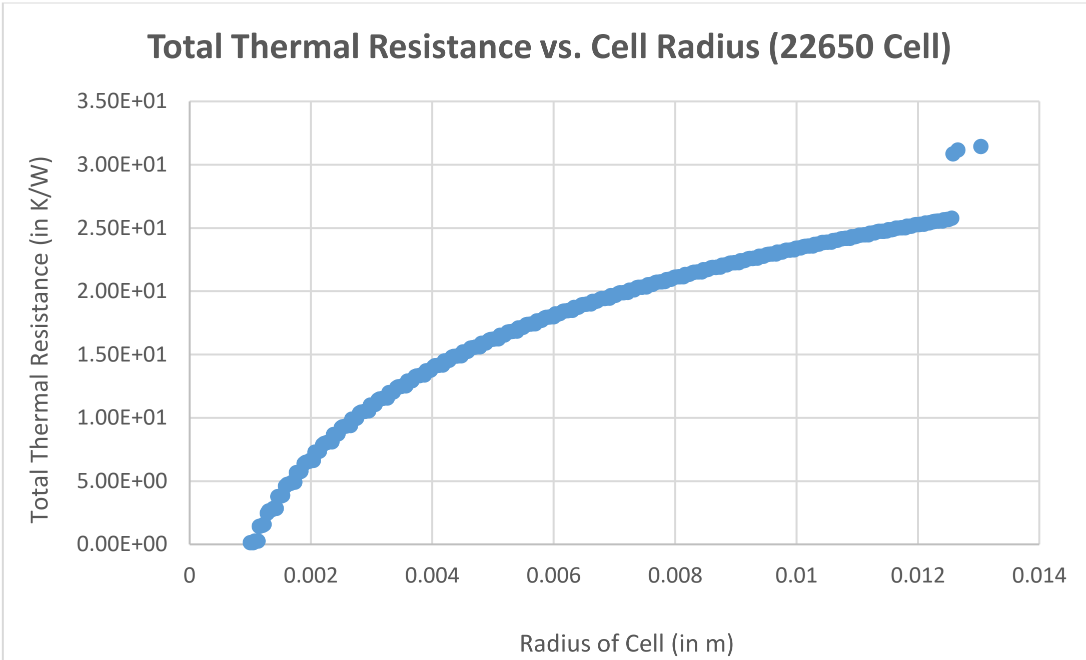

Figure 3. Plot of total thermal resistance versus cell radius for a 22650 cell.

METHODS

After disassembling and modeling the 18650 and 22650 cells, we designed an experiment to measure their effective radial thermal conductivities. We inserted 20 AWG nichrome wire into the gap at the center of each cell's winding and heated the wire using a DC power supply at varying currents. We coated four type K thermocouples (two 36 AWG and two 20 AWG) with SteelStik™ conductive putty for accurate temperature measurements and placed them inside the center of the winding as well as outside the case of the cell. These thermocouples measured the temperature difference between the center and case of the cell when it was heated from inside. This temperature difference was used in the following steady-state, one-dimensional heat conduction equation for cylindrical objects to calculate k_{eff}, the effective radial thermal conductivity of the cell:

$$\dot{Q} = \frac{2\pi k_{eff} L\,(T_1 - T_2)}{\ln\left(\frac{R_2}{R_1}\right)} \qquad (2)$$

The first experiments we conducted on the 18650 cell consisted of putting the nichrome wire inside the spindle at the center of the cell's winding. Since different manufacturers make 18650 cells differently (some with and some without spindles), we used only cells that contained spindles inside their windings. For each trial, the cell was suspended from a height of 6 inches using monofilament fishing line to provide thermal isolation. The DC power supply was used to supply a constant current to the nichrome wire inside the center of the cell, and a multimeter was used to independently measure the voltage difference across the wire. Two thermocouples were placed inside the spindle of the cell and on top of the green plastic cover, and a data logger was used to record the temperature measurements every second. Insulation was added to both ends of the cell to decrease heat conduction in the axial direction. Pictures of the experimental setup are given in Figure 4. In each experiment, the power supply connected to the nichrome wire was left on until the temperature measurements from the four thermocouples became stable. The power supply was then disconnected and the cell was left to cool down in preparation for the next trial. The measured steady-state temperature difference between the inside and outside thermocouples was used along with the lengths and radii of the cell and spindle to calculate the effective radial thermal conductivity of the cell using Equation (2).

The next set of experiments we conducted on the 18650 cell consisted of taking out the spindle in the center of the cell's winding and placing the nichrome wire inside the resulting gap. Everything else was kept the same. Our models suggest that the thermal resistance between the spindle and winding is quite high; hence, experiments done on cells without their spindles give more accurate measurements of the effective radial thermal conductivity. The subsequent experiments on the 18650 cell consisted of combinations of removing the spindle, the green plastic cover, and the case of the cell.

We also conducted similar experiments on the 22650 cell. This cell is bigger and does not contain a spindle, so we inserted the nichrome wire into the gap in the center of the cell's winding. In addition, unlike the green plastic cover of the 18650 cell, the cardboard cover of the 22650 cell slides off very easily. Thus, all of our experiments on the 22650 cell were conducted without the cardboard cover or the spindle. A picture of the experimental setup is given in Figure 5.

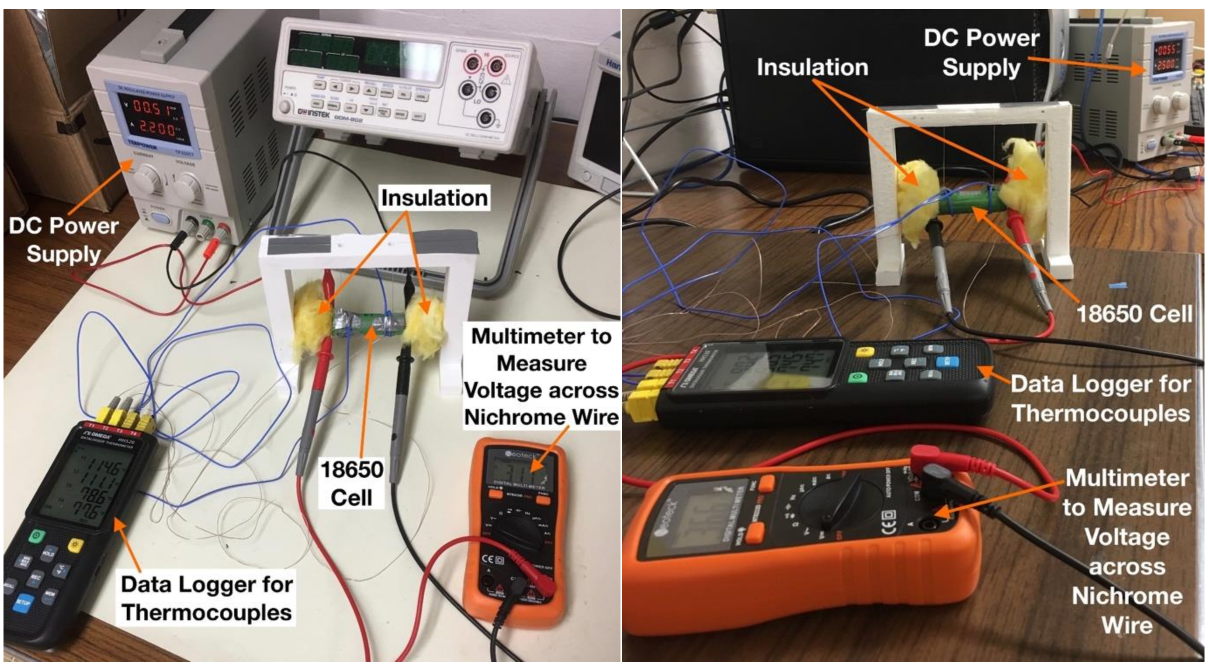

Figure 4. The experimental setup for an 18650 cell.

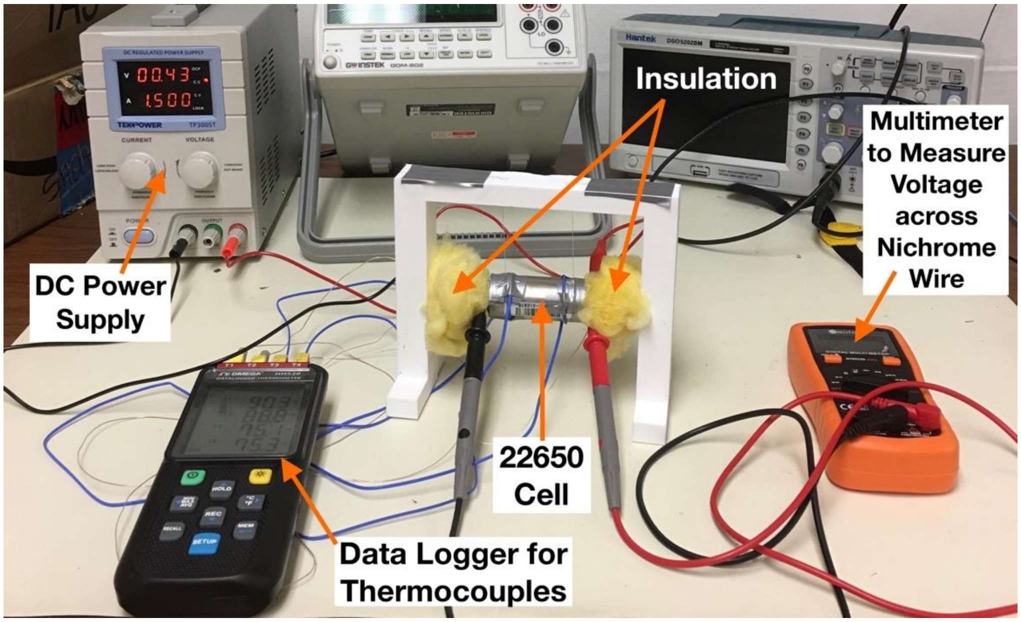

Figure 5. The experimental setup for a 22650 cell.

RESULTS AND DISCUSSION

We conducted a total of twenty trials on two 18650 cells and eight trials on two 22650 cells. Data from these trials is summarized in Table 2 and Table 3.

For the 18650 cells, the data is organized by combining experiments done with or without the spindle, the green plastic, and the case. The power input is calculated by multiplying the current input from the power supply by the voltage difference across the nichrome wire measured by the multimeter at the cell. The average ΔT is the average difference between the thermocouple measurements from the inside and outside of the cell, and the average cell temperature is the average of all four thermocouple measurements at their peaks. The values in red are from experiments done on cells without their spindles but with green plastic covers.

For the 22650 cells, all the experiments were done in the same way: case, no spindle, and no cardboard cover. The values in red are from experiments with the highest power input.

Table 2. Experimental Data for Two 18650 Cells

	Spindle?	Green Plastic?	Case?	Power Input (in W)	Average ΔT (in K)	Average Cell Temperature (in ℃)	Average k_{eff} of Cell (in W/m*K)
Cell 13 Experiment (18650)	Y	Y	Y	0.34	9.8	32.5	0.21 ± 0.04
	Y	Y	Y	0.66	16.6	39.3	0.24 ± 0.03
	Y	Y	Y	0.70	22.2	43.2	0.19 ± 0.02
	Y	Y	Y	0.93	28.4	49.4	0.20 ± 0.01
	N	Y	Y	0.35	4.0	30.0	0.52 ± 0.14
	N	Y	Y	0.59	7.6	35.8	0.47 ± 0.08
	N	Y	Y	0.91	13.7	42.0	0.40 ± 0.06
	N	Y	Y	1.4	19.3	50.6	0.42 ± 0.05
	Y	N	Y	0.60	10.2	38.7	0.35 ± 0.09
	Y	N	Y	0.93	11.3	44.7	0.50 ± 0.10
	Y	N	N	0.77	21.3	44.8	0.22 ± 0.02
	Y	N	N	0.85	17.4	41.1	0.29 ± 0.05
Cell 12 Experiment (18650)							
	Y	Y	Y	0.36	6.9	32.6	0.31 ± 0.05
	Y	Y	Y	0.67	12.6	38.0	0.32 ± 0.04
	Y	Y	Y	0.80	14.7	41.6	0.33 ± 0.03
	N	Y	Y	0.73	11.2	39.7	0.39 ± 0.09
	N	Y	Y	1.0	14.8	42.8	0.41 ± 0.05
	N	Y	Y	1.5	21.7	51.7	0.41 ± 0.03
	N	N	Y	0.59	12.6	39.1	0.28 ± 0.04
	N	N	Y	0.92	20.4	48.0	0.27 ± 0.03

Table 3. Experimental Data for Two 22650 Cells

	Spindle?	Cardboard Cover?	Case?	Power Input (in W)	Average ΔT (in K)	Average Cell Temperature (in ℃)	Average k_{eff} of Cell (in W/m*K)
Cell 8 Experiment (22650)	N	N	Y	0.35	13.2	34.0	0.19 ± 0.02
	N	N	Y	0.58	22.7	41.8	0.18 ± 0.01
	N	N	Y	0.86	27.3	46.9	0.22 ± 0.02
	N	N	Y	1.0	32.6	51.6	0.23 ± 0.01
Cell 15 Experiment (22650)	N	N	Y	0.35	10.5	33.1	0.23 ± 0.04
	N	N	Y	0.61	21.8	42.0	0.20 ± 0.02
	N	N	Y	0.79	19.2	42.8	0.29 ± 0.02
	N	N	Y	0.98	38.3	55.6	0.18 ± 0.02

The results indicate that data from a single set of experiments done in the same way are fairly consistent. It is worth noting that the data does vary quite a bit from one set of experiments to another, especially for the two 18650 cells. For instance, the k_{eff} values from the last two trials done on Battery 12 (case, no green plastic, and no spindle) are significantly lower than the three trials above (case, green plastic, and no spindle). The difference between experiments may result from the thermocouple placement inside the cell. Although we did put SteelStik™ putty on the thermocouples before inserting them into the center of the winding, in some trials, they may have been in better contact with the nichrome wire instead of the winding. This would have led to a higher temperature difference between the inside and outside of the cell, which would result in smaller k_{eff} values. Likewise, thermocouples that are in better contact with the winding instead of the nichrome wire would lead to a lower temperature difference between the inside and outside of the cell and thus higher k_{eff} values.

From our models, the thermal resistance between the spindle and the winding is quite high, so we believe experiments done on cells without their spindles give more accurate measurements of the effective radial thermal conductivity. In these experiments, this thermal resistance is neglected, and heat is conducted only through the winding and case. On the other hand, we disregarded the k_{eff} values from the last two trials of Battery 12 (case, no green plastic, and no spindle) for the following reason: When the cells are placed next to each other inside a battery, each one of them has a green plastic cover on the outside. In the event of a thermal runaway, heat would conduct through all parts of the cell, including the case and the green plastic cover. As a result, for thermal models, it is necessary to find the effective conductivity of the entire cell, including the green plastic cover. Thus, if we average the k_{eff} values from only the experiments done without the spindle but with the green plastic cover (the red measurements in Table 2), the effective radial thermal conductivity of 18650 cells is 0.43 ± 0.07 W/m-K.

As for the 22650 cell, because all our experiments were conducted without the spindle, we believe experiments done at the highest power input give the most accurate measurements of the effective radial thermal conductivity. Higher power input causes a greater temperature difference between the inside and outside of the cell, which in turn leads to a smaller uncertainty. Thus, if we average the k_{eff} values from only these experiments on the 22650 cells (the red measurements in Table 3), we get that the effective radial thermal conductivity of 22650 cells is 0.20 ± 0.04 W/m-K.

UNCERTAINTY CALCULATIONS

Uncertainty calculations for our experimental data were computed using a root-sum-square (RSS) method on all the variables used to calculate k_{eff} in Equation (2):

$$\frac{\Delta k_{eff}}{k_{eff}} = \sqrt{\left(\frac{\Delta(\Delta T)}{\Delta T}\right)^2 + \left(\frac{\Delta V}{V}\right)^2 + \left(\frac{\Delta I}{I}\right)^2 + \left(\frac{\Delta R_2}{R_2}\right)^2 + \left(\frac{\Delta R_1}{R_1}\right)^2 + \left(\frac{\Delta L}{L}\right)^2} \qquad (3)$$

To calculate the $\Delta(\Delta T)$ term, the average temperature difference between the inside and outside thermocouples was subtracted from the highest temperature difference between the two sets of thermocouples. One degree Celsius was then added to this value because each of the two sets of thermocouples readings have an accuracy of around ±0.5 ℃. As for the rest of the terms, datasheets were used to find the accuracies of the multimeter, power supply, micrometer, and dial calipers.

COMPARISONS WITH PRIOR WORK

Previous studies of thermal propagation in lithium ion batteries, such as that reported by Rickman, et al.[3], have used a radial thermal conductivity of 1 W/m-K for the cell winding and have used a contact heat transfer coefficient between the winding and the cell wall. In such efforts, a value of approximately 50 W/m^2-K has provided reasonable correlation to thermal propagation test results. This value is, however, in conflict with that reported by Gaitonde, et al.[8], which was closer to 670 W/m^2-K and was for the actual contact coefficient and not for the materials on either side of the interface. Figure 6 shows a reasonable explanation for this difference: the relatively large increase in thermal resistance occurs due to the thick (0.15 mm) insulating separator layer that comes between the cell winding and the cell wall. If we calculate an effective contact coefficient between the winding and the cell wall by including the lower measured radial thermal conductivity of the 18650 cell winding (0.43 W/m-K), the measured contact coefficient reported by Gaitonde, et al.[8], and this thick outer separator layer, we obtain a value of 36 W/ m^2-K. This value is in reasonable agreement with previous values from higher level model correlations.

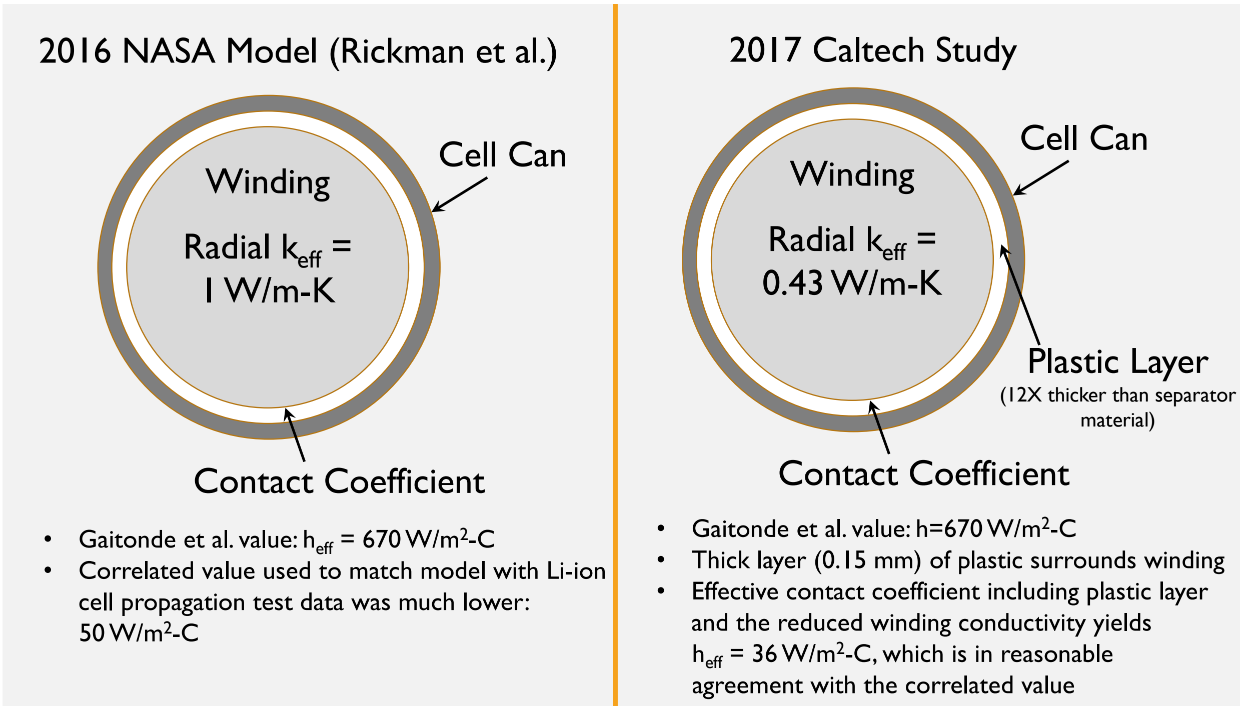

Figure 6. Differences between 2016 Rickman et al. Study and Our 2017 Study

CONCLUSIONS

Our measured radial thermal conductivity values for the 18650 and 22650 cells are greater than the ones theoretically determined by Drake et al., which suggests that the cells can conduct heat better than previously thought. Our experimental values are also significantly smaller than the values reported with perfect thermal contact between the layers[5-6], which suggests that including realistic, non-ideal, contact coefficients from layer-to-layer is important when modeling the radial transport of heat in cylindrical lithium ion battery cells.

If we compare our experimental values to those predicted by our thermal models, we see that the predicted value of 0.22 W/m-K for the 22650 cell is close to our experimental value, while the predicted value of 0.27 W/m-K for the 18650 cell is significantly smaller than our experimental value. Out of the many factors in our model that could account for this disagreement, the contact resistance between the cathode and plastic separator may account for the discrepancy. Vishwakarma et al.[7] reported that the contact resistance of the cathode-separator interface accounts for around 88% of the total thermal resistance in the cell. However, it is possible that this interfacial resistance is lower than that measured by Vishwakarma et al. since their measurements were done at a very low contact pressure, approximately 0.14 bar (2 psi). If the actual contact pressure between the winding layers is higher, the interfacial resistance would be reduced. According to our calculations, if this resistance is 1.87 times lower, then the predicted radial thermal conductivity of the 18650 cell becomes 0.43 W/m-K, which is what we obtained experimentally.

In summary, this research provides direct measurements of the effective radial thermal conductivities of 18650 and 22650 lithium-ion cells, which should lead to better models of thermal runaway propagation.

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