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Evaluation of Lithium Battery Thermal Runaway Propagation

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16. Abstract

Lithium batteries have been shipped aboard aircraft with existing United Nations (UN) classification numbers for many years. Although the UN classifies lithium batteries as dangerous goods, current UN numbers for lithium batteries do not indicate what level of hazard each individual shipment may pose. Lithium batteries can exhibit varied temperature rise and propagation characteristics when heated to thermal runaway. Therefore, This study was conducted to characterize the propagation of cylindrical cells and pouch cells at various states-of-charge (SoCs) to determine or verify key test factors that should be considered for development of a lithium battery propagation test.

Six cells were placed in line with each other (denoted cell # 1 through cell #6) in an insulated box, and thermal runaway was initiated in cell #1. Once thermal runaway initiated, power to the heater was cut off and propagation characteristics were recorded.

Key findings included:

- The onset temperature of thermal runaway for the pouch cells depended on SoC. For example: A test at 20% SoC showed a thermal runaway onset temperature 53 °C to 83 °C higher than tests at 100% SoC.
- Pouch cells propagated faster than cylindrical cells.
- In the test at 20% SoC, the temperature of pouch cell #6 was as high as 108°C when pouch cell #1 went into thermal runaway and in the test at 100% SoC, pouch cell #6 was only 36.5 °C when pouch cell #1 went into thermal runaway. In other words, at a fixed heat rate, more time was required for cells at a lower SoC to enter thermal runaway. This indicated the need for more than six cells because too much heat was transferred to the system before propagation began.
- It was observed that the mass loss of each individual cell was fairly constant with slightly higher mass loss in the first cell.
- Cell voltage was a usable metric to pinpoint thermal runaway in the pouch cells but behaved erratically in the cylindrical.

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Contents

Figures

Tables

Acronyms

Acronym	Definition
Ah	Amp-hour
FAA	Federal Aviation Administration
PID	Proportional-integral-derivative
SoC	State of charge
UN	United Nations

Executive summary

Billions of lithium batteries are shipped annually with existing United Nations (UN) classification numbers throughout the globe [1]. They are characterized by the UN as dangerous goods, and when shipped, are classified by a number that distinguishes between lithium-ion and lithium-metal batteries, either individually packed or packed in or with equipment. Unfortunately, the current UN numbers for lithium batteries do not indicate what level of hazard each individual shipment may pose.

Lithium batteries can exhibit varied temperature rise and propagation characteristics when heated to thermal runaway. Therefore, this study was conducted to characterize thermal runaway propagation of cylindrical cells and pouch cells at various states-of-charge (SoCs) to determine or verify key test factors that should be considered for development of a standardized lithium battery propagation test.

Six cells were placed in line with each other (denoted cell #1 through cell #6) in an insulated box and thermal runaway was initiated in cell #1. Once thermal runaway initiated, power to the heater was cut off and propagation characteristics were recorded.

Key findings were that:

- The onset temperature of thermal runaway for the pouch cells depended on SoC. For example: A test at 20% SoC showed a thermal runaway onset temperature 53°C to 83°C higher than tests at 100% SoC.
- Pouch cells propagated faster than cylindrical cells.
- In the test at 20% SoC, the temperature of pouch cell #6 was as high as 108°C when pouch cell #1 went into thermal runaway and in the test at 100% SoC, pouch cell #6 was only 36.5°C when pouch cell #1 went into thermal runaway. In other words, at a fixed heat rate, more time was required for cells at a lower SoC to enter thermal runaway. This indicated the need for more than six cells because too much heat was transferred to the system before propagation began.
- It was observed that the mass loss of each individual cell was fairly constant with slightly higher mass loss in cell #1.
- Cell voltage was a usable metric to pinpoint thermal runaway in the pouch cells but behaved erratically in the cylindrical cells.
- The type and shape of the heater used to initiate thermal runaway may have an impact on the actual heating rate experienced by the cell.

1 Introduction

1.1 Background

As the use of electronic devices becomes more commonplace, the use of lithium batteries has become more prevalent to power them. As a result, the quantity shipped worldwide continues to increase. Additionally, new chemistries, form factors and designs continue to emerge.

Lithium batteries are known to undergo a process called thermal runaway, which is a self-sustaining uncontrolled increase of temperature within the battery, thus creating a fire hazard. However, the hazard of each of the different types of lithium battery is unique and can present a varied threat.

Current United Nations (UN) classifications used to classify the hazard of batteries during transportation do not differentiate various lithium batteries based on their potential hazard. As a result, extremely hazardous models of lithium batteries have the same shipment classification as extremely safe lithium batteries. Therefore, there is a need to establish a method to classify lithium batteries based on their hazard.

A UN committee comprised of delegates from the international community representing governments, industries, trade associations, and test laboratories from around the world was established to create a UN classification system for lithium batteries based on hazard. The committee determined that several studies should be performed to evaluate the potential hazards of lithium batteries. Many experiments were required from each of the participating test laboratories to refine a test method and verify consistency.

1.2 Scope

Lithium batteries can exhibit varied temperature rise characteristics and propagation characteristics when heated to thermal runaway. Based on FAA research [2], [3], [4] and past FAA experience, all of these characteristics can depend on various factors:

- the power rating and type of heater that is used and how it is controlled,
- how accurately the cells are brought to the desired SoC,
- how much surface contact each adjacent cell makes with the next cell,
- how well the overall enclosure is insulated, and
- whether the flammable gases ignite.

Due to the wide range of factors influencing thermal runaway behavior, it is important to understand how small differences in test setup can impact test results. All these factors were considered in developing the test configuration in this study.

Additionally, there were several key indicators that were considered to make a determination of whether a battery had gone into thermal runaway, including cell voltage drop, peak measured temperature, and rate of temperature rise.

Ensuring that appropriate thermal runaway indicators and test parameters were accurately measured was critical when evaluating the hazards of lithium batteries. This is a very significant factor for lithium batteries that do not exhibit a classic high energy event.

2 Objective

The objectives for this study were to characterize propagation of cylindrical cells and pouch cells at various states-of-charge (SoCs) and to determine or verify key factors that should be considered when developing a lithium battery propagation test.

3 Setup

Two types of lithium-ion cells were tested. The first test series evaluated cylindrical cells, followed by the second test series that evaluated pouch cells. The heating rate was controlled by a proportional-integral-derivative (PID) controller that was programmed specifically for each test. Initially, the controller was configured to stop heating at 200°C. This was later changed to a much higher temperature of 500°C based on test results, and it was ensured that the heater would not prematurely turn off before thermal runaway occurred. In all cases, power to the heater was turned off when cell #1 entered thermal runaway. All cells were charged to specific SoCs with an Arbin Instruments battery analyzer with predetermined constant-current, constant-voltage charge criteria. The Arbin Instruments system operated with a measurement accuracy within 0.01% and a control accuracy within 0.02%. For these tests, the instruments full scale voltage was 10Volts.

3.1 Cylindrical Cells

Tests were performed with 2.6Ah Li-ion 18650 cylindrical cells as shown in Figure 1. Six cells, denoted cells #1 through #6, were placed in-line with each other so that each cell was in contact with the next (Figure 2). One or two (depending on the desired heating rate) cylindrical 0.125″ diameter, 1.25″ length, 25-Watt cartridge heaters were attached with stainless steel wire to the 18650 cylindrical cell on one end (Figure 1). A 30-gage type-k thermocouple was attached to each cell, also shown in Figure 1. Additionally, the initiation cell (cell #1) required a second type-k thermocouple to provide feedback to a PID controller.

The battery configuration was placed in an insulated steel enclosure to reduce the heat transfer away from the cells (Figures 1, 2, and 3). Finally, some of the tests had voltage terminals installed on each cell to monitor the effect of thermal runaway on cell voltage (Figure 3).

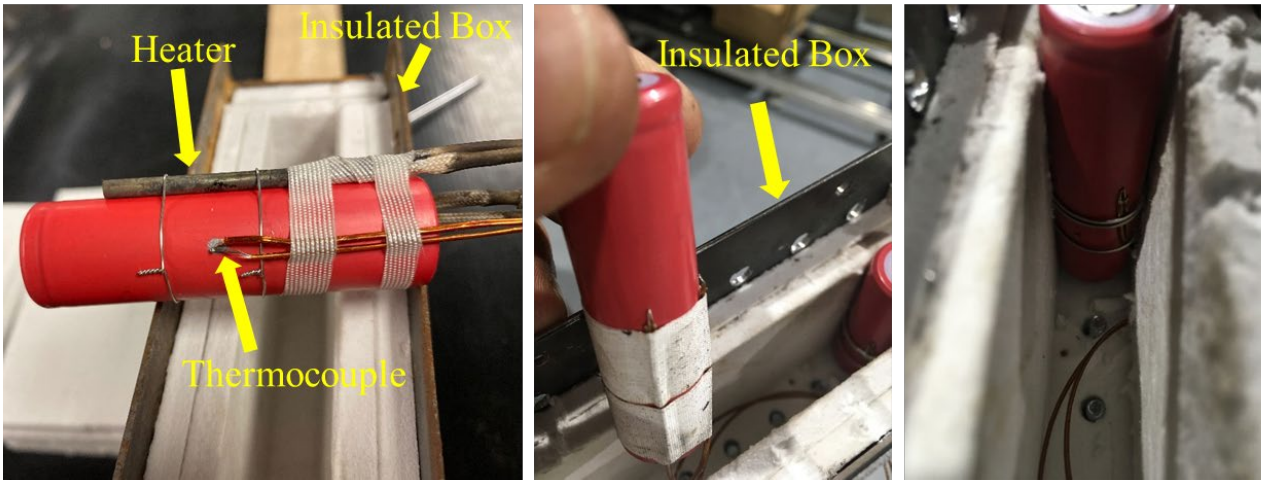

Figure 1. Photographs of setup including heater and thermocouple placement for the cylindrical cell tests

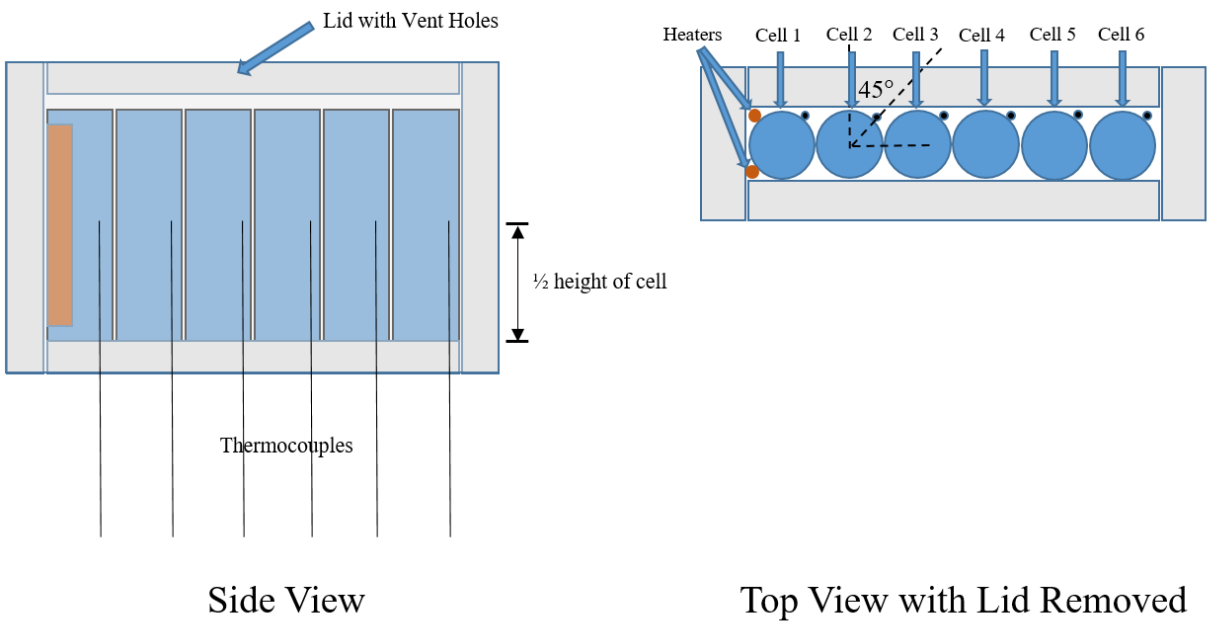

Figure 2. Illustration of test configuration for cylindrical cells

Figure 3. Photo of cylindrical cell test configuration showing voltage leads installed

3.2 Pouch Cells

Similar to the cylindrical cell tests, six pouch cells, denoted cells #1 through #6, were tested in a steel insulated box. The pouch cells had a capacity of 4.8Ah, and thermocouples were attached to the outer edge of each cell (Figures 4 and 5). This thermocouple location was shown in previous tests to be the location most representative of the cells' internal temperature [5].

Six pouch cells were placed in line with the larger flat surfaces making contact before being placed in the insulated box. Once the cells were in the box, loose insulation was stuffed behind the last cell to ensure that all cells were in direct contact with each other. The configuration was rigid so that pouch cells were unable to expand, and the first cell was heated with a 1.5″ by 4″, 75-Watt plate heater unless otherwise specified.

In some of the tests, two 1″ by 2″ polyimide heaters with adhesive backings were used instead of the plate heater. The total power output of both heaters was 40 Watts.

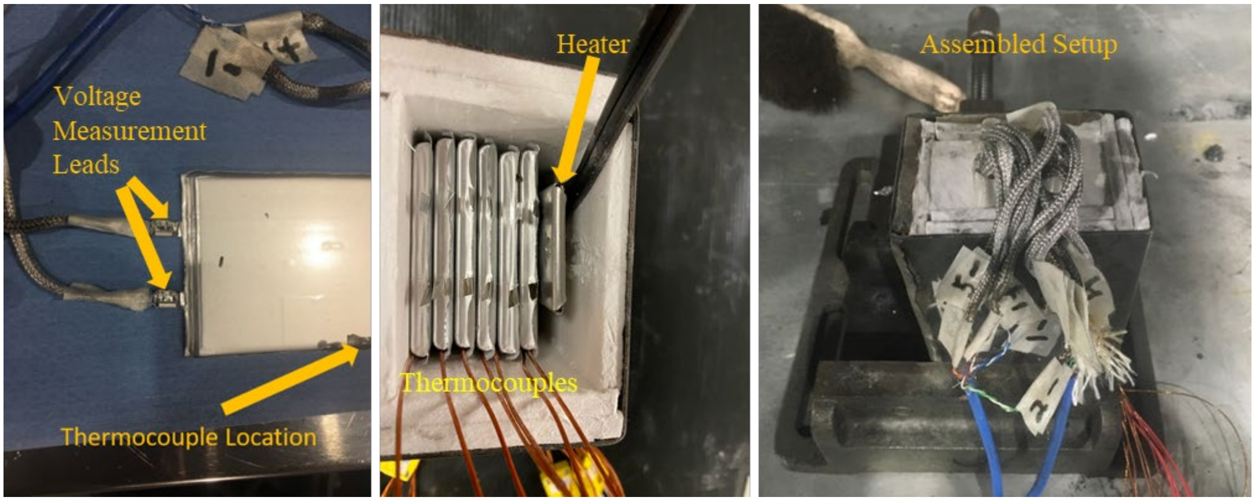

Figure 4. Photographs of the overall pouch cell test setup

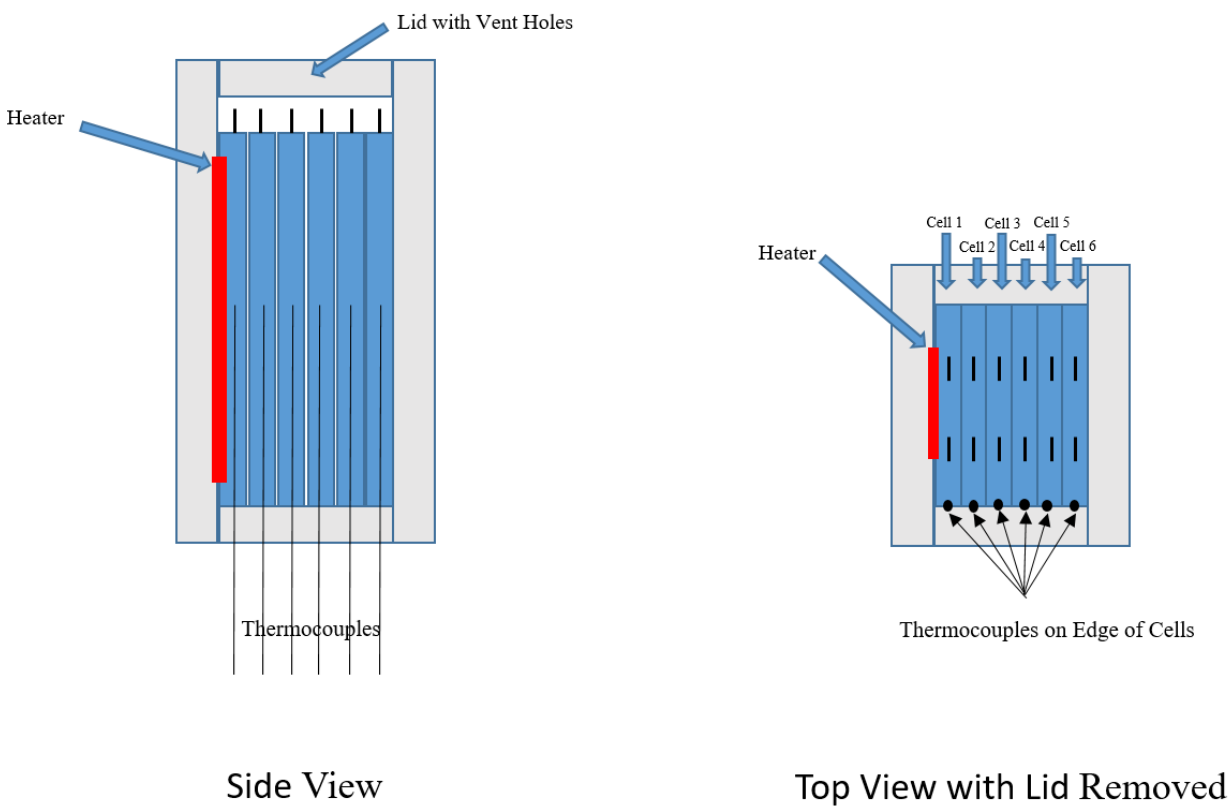

Figure 5. Illustration of test configuration for pouch cells

3.3 Procedure

Propagation tests were performed according to the following procedure using the previously described test setup. First, three tests were performed at 100% SoC and 20°C/min. Next, heat was added at 5°C/min and 20°C/min for a total of 5 additional tests, as shown in Table 1. Unless otherwise specified, once thermal runaway occurred, evidenced by a sharp increase in temperature beyond what was supplied by the heater, power to the heater was cut off. A summary of tests performed is shown in Table 1.

Table 1. Tests performed with cylindrical cells and pouch cells

	20% SoC	30% SoC	50% SoC	70% SoC	100% SoC
5°C/min					x1
20°C/min	x1	x1	x1	x1	x3

4 Discussion of Results

Results were analyzed for cylindrical cells and pouch cells individually to quantify several criteria. The criteria included the temperature of cell #6 when cell #1 underwent thermal runaway, whether flames were observed, how cell voltage behaved throughout propagation, and general trends of mass-loss from the cells.

Next, a comparison was made between pouch cells and cylindrical cells. Attention was given to the rate-of-propagation, maximum temperature, and mass loss between the two types of cells.

4.1 Cylindrical Cells

In the initial round of tests conducted with 18650 cylindrical cells at 100% SoC, temperature, voltage, and mass loss results were observed as shown in Figure 6. The maximum temperature remained fairly consistent from test-to-test as expected since all parameters/procedures were held constant from test-to-test.

The voltage showed somewhat sporadic behavior when each individual cell neared thermal runaway. At some points in the tests, 0 volt was reached prior to thermal runaway followed by a jump in voltage. This result showed that voltage drop may not be a suitable sole indicator of thermal runaway for cylindrical cells, because an individual may not know if voltage is going to remain at 0 or increase again. This may prevent the individual from knowing the appropriate time to disconnect power to the heater. This is important because failure to turn off the heater at a standardized time can lead to excessive heat being added to the system and inconsistent test results.

At 100% SoC, mass loss was fairly consistent from cell to cell. In two tests, the initiating cell (cell #1) had the highest mass loss. In a third test, mass loss of cell #1 was nearly equal to the other five cells. The higher mass loss of cell #1 in two of the three tests can be explained by the fact that more heat was added to that cell, and therefore more internal components burned or vaporized. More tests would be required to confirm this since one of the three tests did not follow this trend.

One goal of these tests was to determine how many cells were necessary for a propagation test. To reduce the amount of “preheating” in the system, the aim was to minimize the temperature of the last cell (cell #6) when the first cell (cell #1) underwent thermal runaway. In all tests, at all tested SoCs, the temperature of cell #6 was nearly ambient when the first cell underwent thermal runaway and cell #5 was within a few degrees of ambient. This result suggested that six cells should be sufficient for a cylindrical cell propagation test. Any reduction below six cells would require further testing and validation.

From all of the cylindrical cell tests, flames were only observed when the cells were charged to 100%. This was consistent with the higher temperatures and more violent reactions observed at increased SoC, as described in previous FAA studies [4], [3].

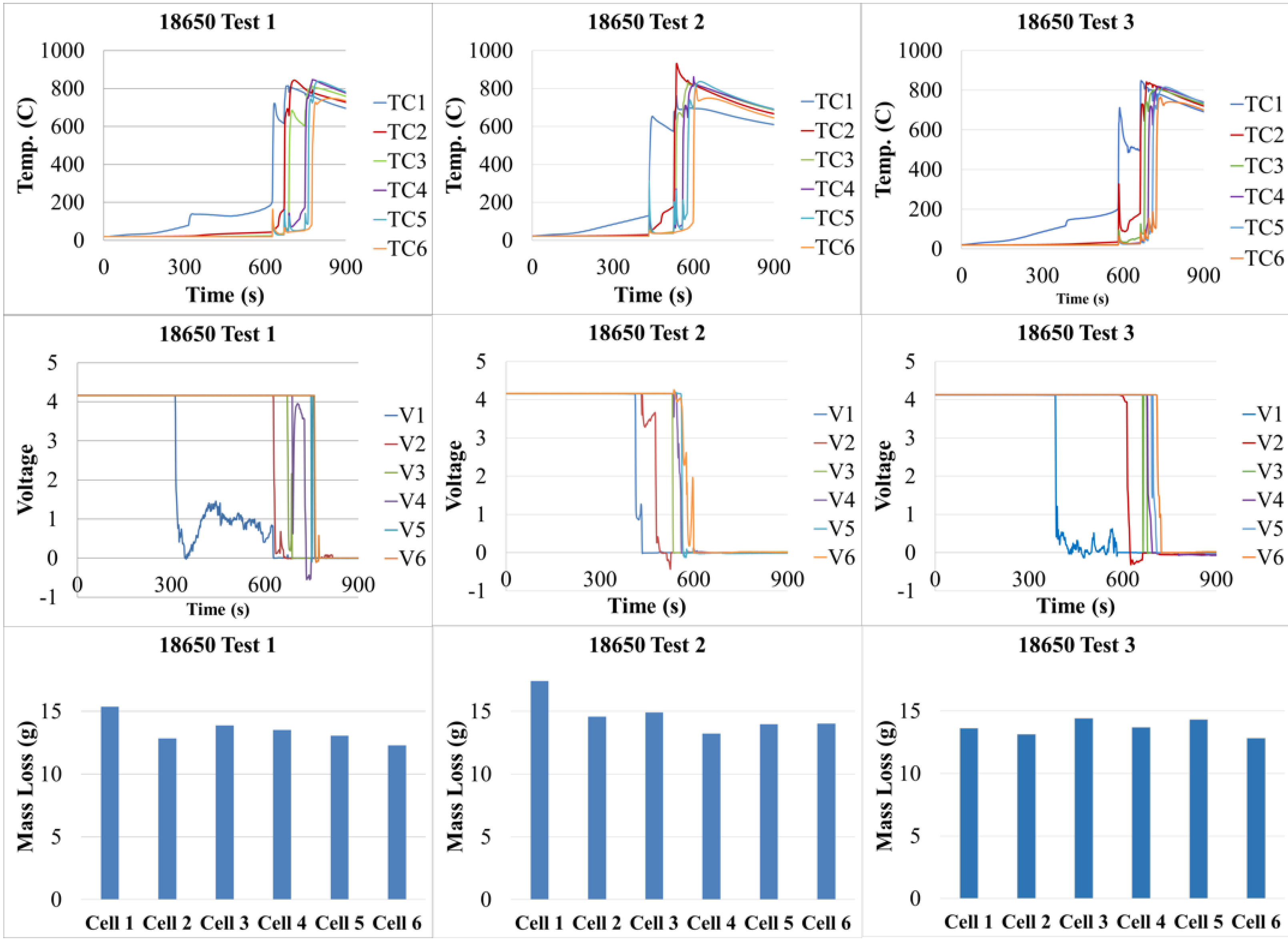

Figure 6. Cylindrical cells, tests performed at 100% SoC

4.2 Pouch Cells

As observed in cylindrical cell tests, flames were only observed in pouch cell tests at 100% SoC. In the other tests, large quantities of flammable gases [6] were emitted from the cells. In many of the thermal runaway events, it was speculated that cell temperatures *were* hot enough to ignite the gases close to the cell, but gas mixtures were too rich to support combustion. Further away from the cells, mixtures were ideal to support combustion, but temperatures were not high enough.

In the 100% SoC tests, voltage consistently dropped to 0 volt at the onset of thermal runaway (Figure 7). This was in contrast to cylindrical cells, which showed a much more sporadic voltage behavior and fell to 0 volt only to rise again moments later. Additionally, mass loss stayed fairly consistent from cell to cell with the first cell consistently losing slightly more mass (Figure 7).

Also shown in Figure 7, cells entered thermal runaway at 150°C to 180°C when at 100% SoC. However, at 30% SoC, the pouch cells did not go into thermal runaway when kept at 200°C for 30 minutes (Figure 8). It was later found that when cell #1 in the 20% SoC configuration was heated to 233°C, it went into thermal runaway and propagated thermal runaway to the remaining cells while releasing a significant quantity of flammable gas.

These results illustrated the dependence of SoC on thermal runaway onset temperature. To determine the thermal runaway hazard of a specific cell, it may be necessary to continue heating a cell well beyond 200°C to ensure there are no exothermic reactions that develop at higher-than-usual temperatures.

As with the cylindrical cells, one goal was to minimize the amount of heat that was transferred to the last cell (cell #6) when cell #1 underwent thermal runaway. Results showed, at a low SoC (20%), cell #6 reached a temperature of 108°C before cell #1 entered thermal runaway. However, at higher SoC (70% and 100%), cell #6 reached 49°C and 36.5°C, respectively. In other words, at a fixed heat rate, more time was required for cells at a lower SoC to enter thermal runaway. This behavior was likely related to the variation of thermal runaway onset temperature with SoC.

These results show that the number of cells required in a propagation test may also be dependent on SoC. More cells may be required for tests performed at lower SoCs than at higher SoCs.

Finally, at the manufacturer-specified voltage, polyimide heaters were only able to heat the first cell at 10 to 12°C/min. and therefore fell short of the specified rate of 20°C/min. This reinforced the value of the higher power output from the heater plates. It was possible that an increase of the supplied voltage to the polyimide heaters beyond the manufacturer specification may have been sufficient to increase the heating rate to 20°C/min. but was not verified.

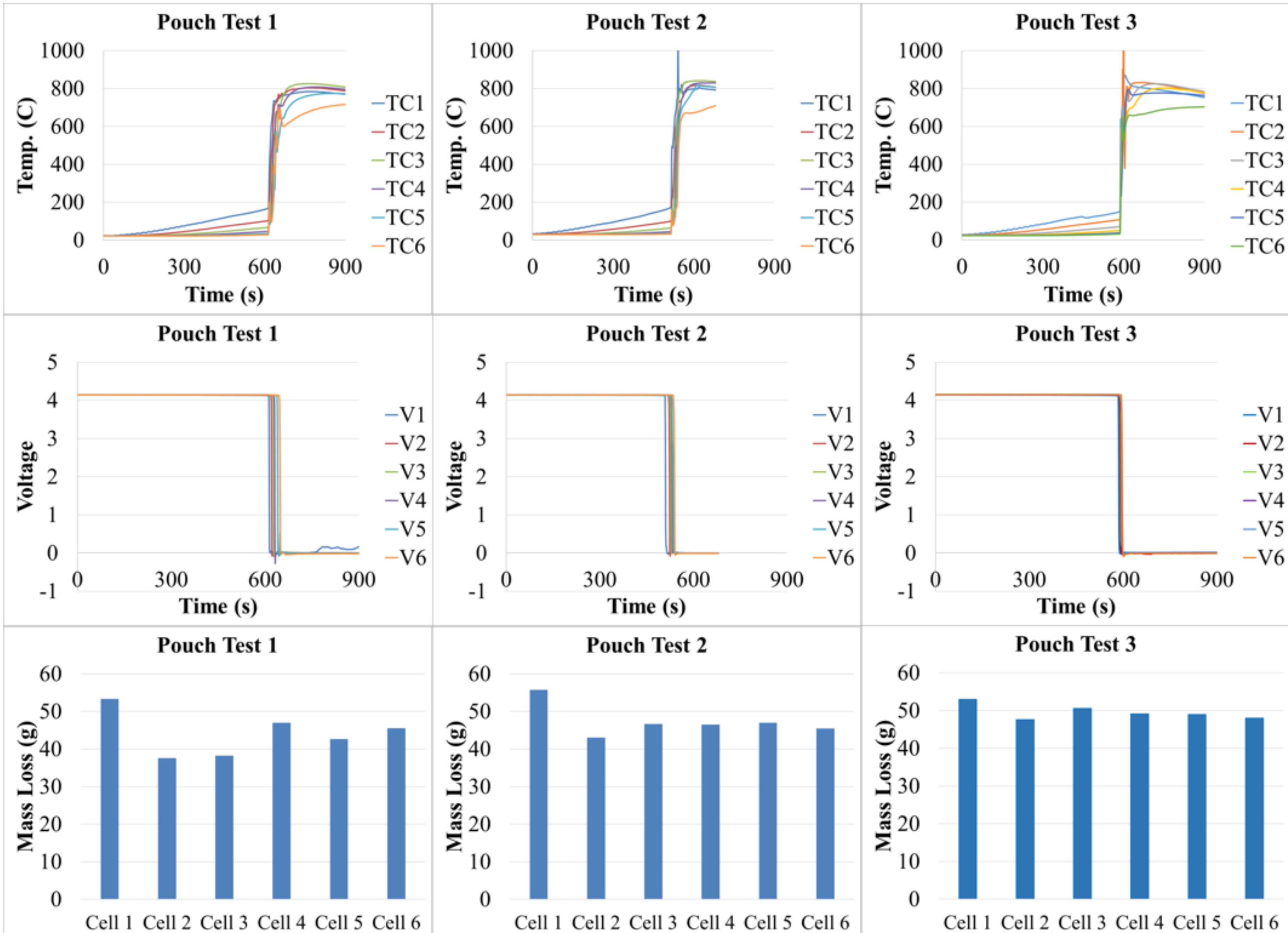

Figure 7. Pouch cells, tests performed at 100% SoC

Untested | 20% SoC, TR onset temp.: 233C | 30% SoC, heater only heated to 200C | 50% SoC, TR onset temp.: 150C | 100% SoC, TR onset temp: 150C to 180C

Figure 8. Images of various pouch cell scenarios

4.3 Pouch Cells vs Cylindrical Cells

Results showed that the rate of propagation of pouch cells was much faster than cylindrical cells (Figure 9). This can be attributed to the larger contact area and increased cell-to-cell heat transfer for the pouch cell configuration.

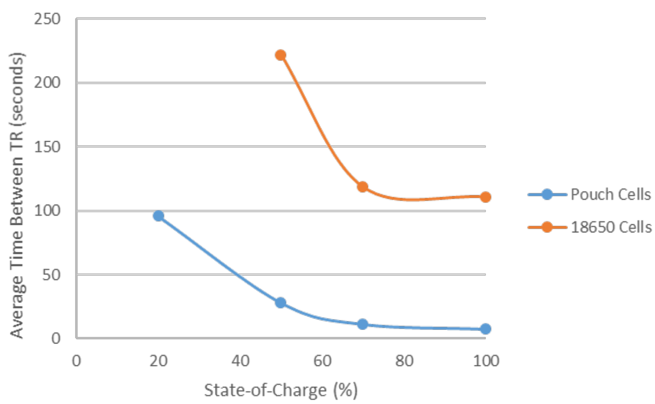

Figure 9. Propagation rate for cylindrical cells and pouch cells

All the cells tested were shown to have an increased temperature with increased SoC, and the maximum temperatures were similar for cylindrical cells and pouch cells (Figure 10). This was consistent with results previously published in other FAA research [3].

Thermal runaway propagation occurred for the pouch cells from 20% SoC through 100% SoC and for cylindrical cells from 50% SoC through 100% SoC. This further showed the greater tendency of pouch cells to propagate when compared to cylindrical cells. Because pouch cells had greater surface contact and heat transfer, it was also likely that they required more initial heat input into the first cell because much more heat was being dissipated to its neighboring cell.

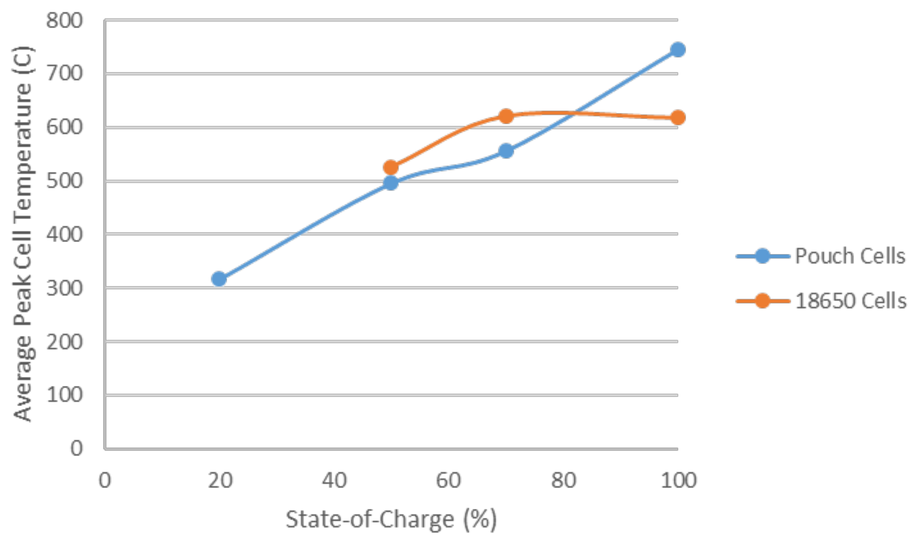

Figure 10. Peak temperature vs SoC for pouch and cylindrical cells

5 Summary

5.1 Thermal Runaway Onset Temperature

The onset temperature of pouch cell thermal runaway was dependent on SoC. The test at 20% SoC showed a thermal runaway onset temperature that was 53°C to 83°C higher than the tests at 100% SoC. Therefore, it may be necessary in a thermal runaway test to heat cells far beyond 200°C to verify that hazards are sufficiently identified.

5.2 Quantity of Cells Required for a Standardized Propagation Test

Pouch cells propagated faster than cylindrical cells due to greater surface contact from cell to cell. These results indicated a need for more cells to be included in a pouch cell thermal runaway propagation test than in a cylindrical cell propagation test. However, a quantity of five or six cells appeared to be sufficient for a cylindrical cell propagation test.

In the test at 20% SoC, the temperature of pouch cell #6 was elevated to 108°C when cell #1 went into thermal runaway and in the test at 100% SoC, pouch cell #6 was elevated to 36.5°C when cell #1 underwent thermal runaway. These results also indicated that more cells may be necessary in a propagation test at lower SoCs.

5.3 Summary of Additional Findings

- The mass loss of each cell remained fairly consistent from cell to cell during a propagation test and was generally higher in the first cell.
- In these tests, voltage appeared to be a usable metric to determine thermal runaway onset in pouch cells but would be more questionable in cylindrical cells because voltage

occasionally dropped to 0 volt, giving an impression that the heater should be turned off, only to jump back up moments later.

- Maximum achieved temperature increased with an increase in SoC and was similar for both cylindrical and pouch cells. This was consistent with previous FAA studies.
- Metal flat-plate heaters may be necessary due to their higher power density if the desired heating rate cannot be achieved with flexible polyimide heaters.

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