ONLINE SUPPLEMENTARY MATERIAL

Preventing Cell-to-Cell Propagation of Thermal Runaway in Lithium-Ion Batteries

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DESCRIPTION OF SUPPLEMENTARY FIGURES AND TABLES.

Supplementary Figure S1 is a CT-scan image illustrating the effects of pre-TR vent on the structural integrity of the cell structure. Supplementary Figure S2 shows the test setup used in collecting spectral and video images of materials venting from a heated, fully charged cell during pre-TR and TR events. Supplementary Figure S3 presents a GC-MS chromatogram of the material vented from the cell, collected in a bottle and extracted with acetonitrile. Supplementary Figure S4 shows the 3D CAD geometry of the cell assembly for the battery used in the modeling. The vent channels in a 3s15p (3 cells in series with 15 strings in parallel) battery are shown in pink. The circled-units represent the cathode end, and the un-circled units the anode ends of each cell. The vent channels are designed so that in the event of TR, the ejecta flowing through the channel will impact the cathode sides and the anode sides alternatively. The mesh near the trigger cell and channel exiting into the plenum are shown in red. The left-most end cell along the parallel strings is the trigger cell (shown in blue), and the receiving cells are in black. Cells are assumed to vent at the cathode end through a 6-mm diameter hole. Supplementary Table 1 lists the input parameters used in CFD simulation of ejecta flow, following TR in the trigger cell. The solid particles in the ejecta are assumed to be nanometer size, therefore they track with the gas flowing through the ejecta channel. The CFD flow predictions (Supplementary Figure S5) highlight the contours corresponding to the pressure, temperature, velocity and Mach number along the vent channel. The eighth cell is the trigger cell, symmetric flow is assumed, and the results are shown only for seven cells away from the trigger. Only the cells along the vent channel (15p) are modeled in the CFD simulations since the ejecta are assumed to not flow from one channel to another. The simulation results show that the ejecta come out of the cell at supersonic velocity. The velocity drops as the ejecta traverse the channel, and increases as expected, when it exits into the plenum. The pressure and temperature continue to drop along the channel and when the ejecta exit into the plenum. Supplementary Figure S6 shows the time-domain thermal simulation results derived using the CFD results (data from Figure S5) as one set of inputs, and thermal properties of the cell and its surroundings (Supplementary Table 2) as a second set of inputs. Inputs for the reaction kinetics parameters of thermochemical reactions inside the cell were obtained from Hatchard et al.51 Thermal simulation results are shown only for a total of 4 cells along the channel; the remaining cells along the channels were negligibly impacted by the TR in the trigger cell. The time-

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domain simulation was made in increments of 0.1 second, but only three panels, corresponding to three different times with reference to the thermal runaway start time \( T_{TR} \) are shown. In each panel of Supplementary Figure S6, the third cell from the left is the trigger cell. At \( T_{TR} \) the cell’s surface temperature registered a value in excess of 400 \(^\circ\)C. At that instant, the adjacent cells also experienced a substantial increase in temperature. One second before TR (time = \( T_{TR} - 1 \) s), the adjacent cells were still at ambient (21 \(^\circ\)C). More interestingly, 2 seconds after TR (time = \( T_{TR} + 2 \) s), the receiving cells started cooling and dropped below 170 \(^\circ\)C, suggesting that no irreversible thermal runaway reaction should occur. Based on the high-speed video camera recordings (Supplementary Figure S7) typical TR event lasts for less than 1 second. Data in Supplementary Figure S6 showed that TR propagation should not occur, if the cell is assumed not to vent before TR, and all thermal energy associated with TR is carried immediately after TR by the ejecta coming out of the trigger cell.

The parameters for the CFD simulations used to determine the amount of DMC solvent depositing on the receiving cells located along the vent channel are listed in Supplementary Table 3. The battery is 3s15p configuration, and the trigger cell is located at the top of the parallel strings. The vent channel is open on both ends, with the walls of the battery compartment located 2 mm away from the open ends. In the simulation, the trigger cell was located close to the wall, and when the cell vented, part of the solvent could flow out toward the wall, and the rest into the vent channel. The orientation of the vent channel was configured vertical with respect to the ground plane. As stated in Supplementary Table 1, inside the vent channel, the temperature was <90 \(^\circ\)C, below the boiling point of DMC. Therefore, in the simulation, DMC inside the channel was assumed to be in liquid phase. Supplementary Table 4 lists the amount of DMC deposited on the first 10 receiving cells away from the trigger cell, as obtained from the CFD simulation.
Supplementary Figure S1. CT-scan images of LG HG2 18650 Li-ion cells before and after pre-TR venting (by heating to 141°C). (A) and (A’): middle cross-section of a control cell near its cathode. CID disk and scored-disk, located below the cathode, are visible. (B) and (B’): Vented cell shows ruptured CID disk and the popped-up scored-disk. The cross-sectional area of the scored vent disk is 9.1 mm$^2$ through which venting gas must come out first; if the six circular vents around the scored disk are also opened, each one of them would account for 1.6 mm$^2$ open area for the gas to escape. (C): The space between the CID disk and the positive terminal has a cross-sectional area of 10.5 mm$^2$. (D): The positive terminal has four vent valves. The unrolled view of the vent valves shows that after venting, three of the four vent valves partially opened (shown in orange), for a total cross-sectional area of 3.75 mm$^2$ in the escape path for the venting gas.
Supplementary Figure S2. FTIR and hyperspectral data collection setup showing the spectrometers focused at a 12 cm x 12 cm window in front of the venting cell.
Supplementary Figure S3. GC-MS data of a solution of material vented from the cell and collected in a bottle and extracted with acetonitrile (left). DMC is the main compound in the vented material along with lower amounts of EC and PC.
**Supplementary Figure S4.** CAD drawing of vent channel geometry for the simulated 3s15p battery. The left-most cell (shown in blue) is the trigger cell. The vent channel, shown in red, is folded to show the end that exits into the plenum.
Supplementary Figure S5. Ejecta flow along the vent channels as predicted by CFD simulation.
Supplementary Figure S6. Thermal simulation results for the 3s15p battery, shown for four different cells only in three separate panels at three different times. The trigger cell is 3rd from the left. TR starts at time = $T_{TR}$ s. One second before TR, the adjacent cells showed little increase in temperature. Two seconds after TR, temperature of the adjacent cells fell well below 170 °C, with no indication of TR.
Supplementary Figure S7. Single frame snapshots from two high-speed video cameras recording the entire pre-TR vent and subsequent TR events. Snapshots are selected when the flames were closer to their maxima. (A) TR in LG HG2 18650 cell generates a large flame. (B) Pre-TR venting lasts for 300 ms based on time-duration of DMC burning. (C) If DMC is vented before TR, the flame during a TR is much smaller.
**Supplementary Table 1.** Parameters used in CFD simulation of ejecta flow following TR with no pre-TR venting

| Parameter Description       | Parametric Value | Reference        |
|----------------------------|------------------|------------------|
| Initial Inflow Pressure     | 12.5 atm         | Jhu et al.\(^{52}\) |
| Final Inflow Pressure       | 1.5 atm          | Assumed          |
| Total ejecta volume         | 5.4 L at STP     | Rickman et al.\(^{53}\) |
| Ejecta flow time            | 250 ms           | Data from test in Fig. 7 |
| Ejecta Temperature          | 1250 °C          | Data from test in Fig. 2 |
| Gas Mixture Composition     | H\(_2\) (30%), CO\(_2\) (24.9%), CO (27.6%), C\(_2\)H\(_4\) (7.7%), CH\(_4\) (8.6%), C\(_2\)H\(_6\) (1.2%) | Rickman et al.\(^{53}\) |
| Gas-Solid ratio in the ejecta | 44%:66%         | NREL             |
| Cross Section of Vent hole  | 6-mm diameter    | Data from test in Fig. 2 |

**Supplementary Table 2.** Thermal properties of cell and battery materials

| MATERIAL                        | P (LBM/FT\(^3\)) | PCP (BTU/R-FT\(^3\)) | K (BTU/HR-FT-R) |
|---------------------------------|------------------|-----------------------|-----------------|
| Jellyroll (inner to the cell)   | 161              | 32                    | 1.96            |
| Steel (cell wall)               | 494              | 51                    | 8               |
| FR4 (PC board)                  | 115              | 26                    | 0.17            |
| G4 (housing)                    | 112              | 26                    | 0.17            |
| Air (between cells, PC boards and housing) | 0.075 | 0.055 | 0.0055 |
### Supplementary Table 3. Parameters used in CFD simulation of the pre-TR venting of DMC

| Parameter                          | Quantity          | Reference                          |
|------------------------------------|-------------------|------------------------------------|
| Mass of DMC                        | 2.1 g             | Weight loss (Sec. 4)               |
| Initial pressure / Final Pressure  | 1224 kPa / 101 kPa| Ref. [12, 13]                      |
| Duration of venting                | 300 milliseconds  | Data from Fig. 5B (Sec. 4)         |
| Temperature at the vent            | 95 °C             | Temperature measurement (Sec. 4)    |
| Temperature in the vent channel    | <90 °C            | Temperature measurement (Sec. 4)    |
| Vent hole area                     | 3.33 to 3.75 mm² | CT Scan Fig. 4D (Sec. 4)           |

### Supplementary Table 4. Mass of DMC deposited on the first 10 receiving cells away from the trigger cell.

| Cell # | 1    | 2    | 3    | 4    | 5    | 6    | 7    | 8    | 9    | 10   | Top | Sum  |
|--------|------|------|------|------|------|------|------|------|------|------|-----|------|
| Amount deposited (gram)            | 0.271 | 0.283| 0.391| 0.332| 0.221| 0.126| 0.057| 0.033| 0.011| 0.003| 0.326| 2.058|
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