Quantitative Comparison of Thermal Influence among Laser Irradiation and Other Methods for Thermal Runaway of Traction Lithium Ion Battery

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Initiation methods for thermal propagation tests are being discussed in the EVS-GTR. In this paper, laser irradiation was compared with nail penetration and heater heating using a prismatic Li-ion battery module from the viewpoint of heat energy transfer to adjacent cells. Such transfer with laser irradiation was 0.025% of that of heater heating and the adjacent cell’s temperature rise was less than 1°C by thermal runaway of the target cell. This result demonstrates that laser irradiation is a rational method in terms of replicating single cell thermal runaway only due to the cell’s own energy.

KEY WORDS: EV and HV systems; lithium ion battery; electrical safety (A3)

1. Introduction

The trend toward stricter environmental regulations is accelerating in various countries. Some European countries have declared that sales of vehicles with internal combustion engines will be prohibited in the future, and the popularization of electric vehicles (EV) is expected to accelerate. Many current EVs use lithium ion batteries (LiBs) as the traction batteries. LiBs have a higher energy density than lead-acid batteries and Ni-MH batteries[1]. However, smoking and ignition accidents have occurred in the market for both traction and consumer-use LiBs[2-3], and further enhancement of safety is necessary for the popularization of EVs. Meanwhile, longer EV cruising ranges are also demanded, and there are expectations for even higher energy densities for traction LiBs in order to increase cruising ranges. High-energy density designs for LiBs generally tend to result in lower safety, which calls for advanced battery design to increase the energy density while still securing safety.

For this reason, the “thermal propagation test method” is being discussed in the UNECE EVS-GTR (Electric Vehicle Safety – Global Technical Regulation) as a method of verifying the safety of EV batteries. This test method forcibly induces thermal runaway of one cell on the hypothesis that an internal short circuit occurred in the cell due to some issue, and assesses whether vehicle occupants are exposed to a hazardous environment resulting from a thermal propagation[6]. Forced thermal runaway initiation methods of a target cell are currently being examined in the EVS-GTR. The forced internal short circuit method by nail penetration, the heater heating method, and the overcharge method are noted as initiation methods in the EVS-GTR Phase 1 document published in 2018.

In the forced internal short circuit method by nail penetration, external heat energy is not applied to the target cell, which makes this a rational method in that it replicates thermal runaway that occurs only due to the cell’s own energy starting from an internal short circuit. However, it is necessary to physically penetrate a nail into the target cell, so target cell selection is restricted by the battery pack layout.

The heater heating method offers easy test preparation due to the high degree of freedom of heater and cable installation, and has the merit of a greater degree of freedom of target cell selection. However, the cell is heated by external heat energy input, so there is the issue of heat transfer to adjacent cells before thermal runaway of the target cell. As a result, the heater heating method may result in a more severe assessment compared with the thermal runaway mode starting with the internal short circuit of a single cell.

The overcharge method has a high degree of freedom of target cell selection if electrical connection is available. Like heater heating, however, it may result in a more severe assessment due to external energy input. Furthermore, the overcharge method is not applicable to cells with a current interrupt device.

This study examined the initiation method using laser irradiation.
The laser irradiation method has already been adopted as a thermal propagation test initiation method in the industrial storage battery standard IEC 62619-2017[7]. Moreover, as it is a heating method with local and high power density, it is expected that excessive heat input to adjacent cells, which is an issue with the heater heating method, will be suppressed.

Various research is underway on the thermal runaway initiation method with laser irradiation[8-11], but there are no known reports of quantitative evaluation of the amount of heat transfer to adjacent cells.

This study used a traction prismatic LIB and quantitatively evaluated the heat energy transferred to an adjacent cell until thermal runaway initiation of the target cell with the laser irradiation, nail penetration, and heater heating methods as the initiation method.

In addition, the results of morphological analysis of a discharged cell after laser irradiation and analysis of the heat generation characteristics of the cell constituent materials were used to examine the process leading from the laser-irradiated part to thermal runaway of the entire cell.

2. Experimental

2.1 Evaluation of Heat Transfer Amount to an Adjacent Cell Using an Assembled Battery

An assembled battery consisting of five unit cells connected serially as shown in Fig. 1 was fabricated to evaluate the thermal influence on adjacent cells in the assembled battery state. Table 1 shows the specifications of the cells used in this study.

The cells were numbered 1 to 5 in order from the negative side, and the center cell (No. 3 cell) was selected as the target cell. Polypropylene sheets with a thickness of 0.8 mm were sandwiched between the cells. In addition, a flat plate-type heat flux sensor (HF series made by Captec Enterprise) with the same area as the cells was installed on the target cell side of the No. 2 cell. This heat flux sensor outputs a DC voltage proportional to the value of the heat flux passing through the sensor surface. The sensor is 0.4 mm thick and has a thermal resistance value of 0.00015°C/W/m². The load application surface for each initiation method was set to the narrow face that is accessible in an on-board module. Temperatures were measured by attaching K-type thermocouples to the narrow faces on the sides opposite the load input of each cell.

Laser irradiation, nail penetration, and heater heating tests were conducted using this assembled battery. The conditions of each test were as follows.

2.1.1. Laser irradiation

A disk laser unit for machining applications was used to perform repeated laser irradiation in a pattern of 0.4 s irradiation and 1.0 s stop, with a working distance of 50 cm and a power setting of 500 W.

| Table 1. Battery cell specifications. |
|--------------------------------------|
| Rated capacity (1C)                  | 27 Ah                         |
| Nominal voltage                     | 3.7 V                         |
| Size                                 | 148 mm(W) × 26.5 mm(D) × 91 mm(H) |
| Case material                        | Aluminum                      |
| Cathode material                     | Lithium Nickel Cobalt Manganese Oxide |
| Anode material                       | Graphite                      |
| Separator material                   | Polypropylene/Polyethylene multilayer |

Fig. 1. Schematic Image of 5-cell stacked module.

The average power during the test was equivalent to 143 W. To prevent equipment damage due to light reflected by the cell case surface, an angle of approximately 8° was set between the laser beam axis and the cell case normal.

2.1.2. Nail penetration

The nail penetration test used a steel nail with a diameter of 3 mm and a tip angle of 60°. This nail penetrated to a depth of 25 mm from the cell case surface at a speed of 1 mm/s.

2.1.3. Heater heating

Heater heating used a 16 mm × 35 mm planar heater. This heater was fixed to the narrow face of the cell case and heating control was performed to maintain a heater temperature of 600°C.

Judgment of thermal runaway of the target cell conformed to the EVS-GTR. The judgment condition was set as satisfying both a cell voltage of less than 2 V and a temperature rise of more than 1°C/s at the temperature measurement point.

2.2. Analysis of Mechanism of Thermal Runaway Occurrence by Laser Irradiation

The mechanism of LIB thermal runaway occurrence by laser irradiation has been reported as originating from separator contraction near the irradiated part resulting in an internal short circuit [10]. In that report, laser irradiation was performed in the direction perpendicular to the electrode stacking direction in a cylindrical battery. By contrast, this study performed laser irradiation in the direction horizontal to the electrode stacking direction in a prismatic battery. As the cell shape and the laser irradiation direction to the electrode differ, this study
sought to confirm that thermal runaway occurs by the same mechanism, and examined the process by which thermal runaway occurs with low energy input.

A laser irradiation test using a discharged cell was performed to reproduce the cell internal state when thermal runaway occurs due to laser irradiation. The internal structure of the laser-irradiated cell was analyzed by X-ray computed tomography (CT) and the cell was disassembled to directly observe the electrode and separator.

To extrapolate the process from local heating by laser irradiation to cell thermal runaway, differential scanning calorimetry (DSC) was used to measure the cathode and anode in the discharged state with electrolyte present. Approximately 4 mg of electrode powder in the charged state and electrolyte collected from the cell were sealed at a weight ratio of approximately 1:1 under an argon atmosphere in a stainless steel pressure-resistant hermetically sealed vessel. Measurement was performed at a temperature rise rate of 10°C/s over the temperature range from 0°C to 400°C.

3. Results and Discussion

3.1. Evaluation of Heat Transfer Amount to an Adjacent Cell Using an Assembled Battery

Figure 2 (a) to (c) respectively show the transitions in voltage and temperature when thermal runaway of the target cell was induced by the laser irradiation, nail penetration, and heater heating methods.

In the laser irradiation test, a hole opened in the cell case 1 s after the start of laser irradiation, and after 5 s the cell safety valve opened and a sudden voltage drop and temperature rise of the cell surface were observed. Thermal runaway of the target cell was judged at 9 s after the start of irradiation. The temperature rise of the adjacent cells at that time was less than 1°C. Laser irradiation was stopped 10 s after the start of irradiation. The highest temperature of the target cell was 245°C, and the highest temperatures of the adjacent cells were 107°C for the No. 2 cell and 114°C for the No. 4 cell. Thermal propagation to adjacent cells did not occur.

In the nail penetration test, voltage drop and temperature rise were observed 20 s after the start of the test, that is to say the time at which the nail penetrated to 20 mm from the cell case surface. Thermal runaway of the target cell was judged at 26 s after the start of the test. The temperature rise of the adjacent cells at that time was less than 1°C. The highest temperature of the target cell was 209°C, and the highest temperatures of the adjacent cells were 106°C for the No. 2 cell and 111°C for the No. 4 cell. Thermal propagation to adjacent cells did not occur even with nail penetration.

In the heater heating test, thermal runaway of the target cell occurred 28 min 17 s after the start of heating. The highest power applied to the heater was 481 W, and the average power throughout the test was 241 W. Thermal propagation to adjacent cells occurred.

Fig. 2. Voltage and Temperature chart for (a) Laser irradiation (b) Nail penetration and (c) Heater Heating. Inset: close caption around sharp voltage drop.
after thermal runaway of the trigger cell, and all five cells eventually experienced thermal runaway. The temperatures of the adjacent cells at the time of thermal runaway of the target cell had risen to 89°C for the No. 2 cell and 88°C for the No. 4 cell. Normally, heater heating should efficiently transmit heat to the electrode element inside the cell. However, in the center cell of the assembled battery, the narrow face and bottom face to which the heater can be installed do not directly contact the electrode element, which is thought to be why it took such a long time until thermal runaway of the target cell.

The amount of external energy input until thermal runaway in each test was 1.43 kJ for laser irradiation, 39 J for nail penetration, and 410 kJ for heater heating. The laser beam is reflected by the cell case surface and the heater radiates heat to the surroundings, so less energy was actually input than these values. In addition, the amount of energy input for nail penetration was calculated from the mechanical work, that is to say the product of load (N) x displacement (m). The input energy amounts compared with the amount of electrical energy of the test cell (350 kJ) were 0.40% for laser irradiation, 0.011% for nail penetration, and 117% for heater heating.

Figure 3 shows the temperature of an adjacent cell at the start of the test, at the time of thermal runaway of the target cell, and the highest temperature reached during the test for each test. The temperature rise of the adjacent cell from the start of the test until thermal runaway of the target cell was less than 1°C for laser irradiation and nail penetration, but was 67°C for heater heating. The amount of heat transfer to the adjacent cell until thermal runaway of the target cell, measured using the heat flux sensor, was 0.030 kJ for laser irradiation, 0.040 kJ for nail penetration, and 1.2 x 10^5 kJ for heater heating. The laser irradiation method enabled reduction of the amount of heat transfer to the adjacent cell until thermal runaway of the target cell to 0.025% that of the heater heating method.

These measurements demonstrated a method for quantitatively evaluating the amount of heat transfer to adjacent cells until the start of thermal runaway of the target cell in a prismatic cell.

3.2. Analysis of Mechanism of Thermal Runaway Occurrence by Laser Irradiation

Figure 4 shows the temperature and voltage data when a discharged cell was irradiated with a laser. A hole opened in the cell case and the laser beam reached the electrode element 50 s after the start of irradiation. Unlike the charged case, however, there was no sudden voltage drop or temperature rise when the laser beam directly irradiated the electrode element. Laser irradiation was stopped when the voltage dropped by approximately 80 mV at the time 177 s after the start of irradiation. The voltage continued to drop even after laser irradiation stopped, which confirmed that a soft short circuit had occurred inside the cell.

Figure 5 shows an X-ray CT image of a discharged cell after laser irradiation. A hole approximately 1 mm in diameter and approximately 25 mm deep from the end of the electrode element opened in the laser-irradiated part. In addition, spherical particles with a high X-ray absorption rate were present around the hole. This is thought to be due to melting and re-coagulation of the copper current collector foil and other material melted by the laser irradiation.

Figure 6 shows a photograph of an electrode extracted from the disassembled discharged cell. The white part is the separator between the cathode and anode, the black part is the anode active material layer, and the copper foil on the bottom is the anode current collector foil. In addition, the arrow in the figure indicates the laser irradiation direction. Thermal damage to the separator in a concentric pattern from the irradiated part was observed, which confirmed that heat is transferred to the surroundings centering on the irradiated part.

![Fig. 3. Temperature of adjacent cell (No.2) at each point in time.](image)

![Fig. 4. (a)Voltage and cell bottom temperature during and after laser irradiation and (b) magnified voltage data during laser irradiation for discharged cell.](image)
The separator has a multilayer structure consisting of polyethylene (melting point: approximately 130°C) and polypropylene (melting point: approximately 160°C) as shown in Table 1. As confirmation of the appearance showed no heat damage to the separator in area A in the figure, it is presumed that this area was less than the polyethylene melting point. The presence of the separator could not be confirmed in area C, so it is presumed that this area was the polypropylene melting point or more. In the middle area B, only the polyethylene shutdown layer melted, so it is presumed that the temperature of this area was between the polyethylene and polypropylene melting points.

Figure 7 shows the DSC curves of the charged cathode with electrolyte and the charged anode with electrolyte. It was confirmed that heat generation on the anode starts at approximately 170°C or more, and then heat generation on the cathode starts at approximately 230°C or more.

In the discharged cell, only a short circuit resulting in a voltage drop of approximately 80 mV occurred even when the laser was irradiated directly to the electrode element for 127 s. By contrast, a hard short circuit occurred with the voltage falling below 2 V in 9 s and reaching 0 V in 14 s after the laser was irradiated to the electrode element in the fully charged state.

A report by Matthew et al. [13] using an internal short-circuit instigator confirmed that even if a state with a separator partially missing in the cell is reproduced, a sudden voltage drop does not occur as a result of a short circuit between the cathode active material layer and the anode active material layer, and only a gradual voltage drop occurs. This is mainly due to the high electron resistance of the cathode active material layer, and indicates that a hard short circuit does not occur simply due to partial omission of the separator.

In this study as well, a hard short circuit leading to a voltage drop of the entire cell was not observed in the discharged cell with only partial melting of the separator and metal melting around the hole.

From the above, it is thought that joule heat due to a soft short circuit is generated when a laser irradiates the electrode in a charged cell, and that heat generation occurs first on the anode heated by the laser beam near the irradiated part, and then on the cathode. Furthermore, it is presumed that the heat generated from the electrode around the irradiated part heats the surrounding electrodes, and heat generation propagates inside the cell, leading to thermal runaway of the entire cell.

The laser irradiation method can input heat energy intensively to the irradiated part and induce heat generation from the electrode element locality. This enables inducing of cell thermal runaway with energy input of just 0.40% of the electrical energy of the cell. This method replicates thermal runaway by only the cell’s own energy generated by an internal short circuit, so it can be considered rational. This method is expected to contribute to appropriate battery evaluation without excessive testing demands on products.
4. Conclusion

A method that uses laser irradiation to initiate thermal runaway of a traction LIB was compared with the nail penetration and heater heating methods from the viewpoint of evaluating the amount of heat transfer to adjacent cells until the start of thermal runaway of the target cell. In the heater heating method, the temperature of the adjacent cell rose by 67°C from the initial level by the start of thermal runaway of the target cell, and eventually thermal propagation occurred. In the laser irradiation and nail penetration methods, the temperature rise of adjacent cells was less than 1°C at the time of thermal runaway initiation of the target cell, and thermal propagation did not occur.

In addition, a method was demonstrated quantitatively measuring the amount of heat flowing to adjacent cells before thermal runaway of the trigger cell occurs, and the amount of heat transfer to an adjacent cell was quantitatively evaluated for each initiation method. The results confirmed that laser irradiation enables reduction of the amount of heat transfer to an adjacent cell until thermal runaway of the target cell to 0.025% that of heater heating.

As the laser irradiation method can induce local heat generation with low energy input, it can thus be considered a rational method from the viewpoint of replicating thermal runaway by only the cell’s own energy generated by an internal short circuit.

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References

1. Heide Budde-Meiwes, Julia Drillkens, Benedikt Lunz, Jens Muenrix, Susanne Rothgang, Julia Kowal and Dirk Uwe Sauer, A review of current automotive battery technology and future prospects, Proceedings of the Institution of Mechanical Engineers Part D Journal of Automobile Engineering, 227(5), 761–776 (2013), DOI:10.1177/0954407013485567

2. Laura Bravo Diaz, Xuanze He, Zhenwen Hu, Francesco Restuccia, Monica Marinescu, Jorge Varela Barreras, Yatish Patel, Gregory Offer and Guillermo Rein, Review—Meta-Review of Fire Safety of Lithium-Ion Batteries: Industry Challenges and Research Contributions, J. Electrochem. Soc., 167 090559 (2020), DOI:10.1149/1945-7111/abaf8b9

3. Peiyi Sun, Roeland Bisschop, Huichang Niu and Xinyan Huang, A Review of Battery Fires in Electric Vehicles, Fire Technology 56, 1361–1410 (2020), DOI: 10.1007/s10694-019-00944-3

4. Thomas Barth and Robert Swaim, NTSB Investigations on EV Fires, EVS 16th session (2018), https://wiki.unece.org/display/trans/EVS+16th+session (Accessed on 27 October 2021)

5. EVENTS WITH SMOKE, FIRE, EXTREME HEAT OR EXPLOSION INVOLVING LITHIUM BATTERIES, FAA website, https://www.faa.gov/hazmat/resources/lithium_batteries/media/Battery_incident_chart.pdf (Accessed on 27 October 2021)

6. UNECE GTR No. 20 (Electric Vehicle Safety (EVS))

7. IEC62619 (2017)

8. Sayoko Shironita, Hideki Tsuruga, Keizoh Honda, Kenichiroh Koshika and Minoru Umeda, Thermal runaway characteristics of a LiFePO4-based lithium-ion secondary battery using the laser-irradiation method, Journal of Energy Storage, 40 102715 (2021), DOI: 10.1016/j.est.2021.102715

9. John J. Darst, Eric C. Darcy Romil Patil and Safan Abbassi, Triggering TR in Li-Ion Cells with Laser Radiation, NASA Aerospace Battery Workshop (2018)

10. Lamb Joshua, Ko Jonathan, Torres-Castro Loraine, Stanley June, Grosso Christopher and Gray Lucas, Li-Ion Battery Propagation Trigger Technique Development/Igniter Development, NHTSA - Vehicle Safety Research (2020), https://rosap.nrl.navy.mil/view.dot/43945 (Accessed on 27 October 2021)

11. H. Döring and M. Wörz, Initializing of thermal runaway for lithium-ion cells, JRC Workshop: Safer Li-Ion Batteries by Preventing Thermal Propagation? (2018), https://ec.europa.eu/jrc/sites/default/files/initializing-of-thermal-runaway-for-lithium-ion-cells.pdf (Accessed on 27 October 2021)

12. Fredrik Larsson, Petra Andersson and Bengt-Erik Mellander, Lithium-Ion Battery Aspects on Fires in Electrified Vehicles on the Basis of Experimental Abuse Tests, batteries, 2(2) 9 (2016), DOI: 10.3390/batteries2020009

13. Matthew Keyser, Dirk Long, Ahmad Pesaran, Eric Darcy, Mark Shoesmith and Ben McCarthy, NREL/NASA Internal Short-Circuit Instigator in Lithium Ion Cells, 228th ECS Conference (2018), https://www.nrel.gov/docs/fy17osti/66958.pdf (Accessed on 27 October 2021)