A Characteristic of the Thermal Runaway with Defects from Manufacturing process of the Lithium-ion batteries

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Abstract-- **The initial defect or performance deviation of the produced lithium-ion battery can be easily identified, but identifying a cell with a potential problem is a challenging study. A potential defect affects the performance and safety of the cell in the process of using. The batteries are applying to the varying application field requiring high capacity and power performance, many numbers of cell are connected in series and parallel for the system needs, wherein the problem of individual cells directly affects the performance and stability of the system. In other words, it is very important to understand the behavior characteristics of a potential cell to ensure the performance reliability and stability of the battery system. This research understood the potential thermal runaway (TR) behavior characteristics of cell through a TR experiment with pouch cell, which was manufactured by reproducing potential cell failure, and derived characteristics to identify and isolate failure cell.**

Index Terms-- **Cell defects, Lithium-ion batteries safety, Potential failure, Thermal runaway**

I. INTRODUCTION

Lithium-ion batteries (LIBs) are widely used in various fields. Particularly, recently, hundreds to thousands of battery cells are connected in series and parallel while being applied to high-capacity and high-power systems such as electric vehicles (EV) and energy storage system (ESS) [1]. However, due to characteristics of the manufacturing process of the battery cell, a deviation in the performance of individual cells may occur, and a potential defect may also occur inside the battery. Such cell characteristic imbalance not only reduces the performance of the battery system, but also affects the stability of fire and explosion due to thermal runaway (TR) of a specific cell in serious cases. As the number of connected cells increases [2], it is increasingly difficult to detect this problem, and research on the improvement of battery management technology for managing the imbalance between battery cells and diagnosing failure becomes important. In particular, countermeasures against TR, which are directly related to the battery fire accident, are needed, and it is very important to understand the process of TR to ensure the safety of the battery system.

TR can be caused by electrical, mechanical, and thermal abuse, and to understand these mechanism, systematic and quantitative analysis and experimental methods are presented for various measurement methods, including cell-level measurements and combination of electrolyte and electrode materials [3]. The abuse of a single cell may affect chemical reactions and physical changes in internal components such as an anode, a cathode, a separator, and an electrolyte, which may generate excessive heat and gas. In particular, phenomena such as smoke generation, jet fire, and explosion, which occur when large heat generation at high temperature conditions causes rapid self-heating of the cell, are related to TR [4]. The major events during TR occurring in a single cell include decomposition of a solid electrolyte interface (SEI), side reactions between an electrode and an electrolyte, damage to a separator, and decomposition of a cathode and electrolyte, and this process is closely related to a fire of a battery caused and controlled by a thermal phenomenon [5]. Zonghou Huang, et al. [6], studied the TR propagation in the difference two cathode LIB in the large format modules. They carried out detail analyzed of TR propagation characteristics with NCM and LFP modules. This study provided much information of the TR behavior with propagation time, risk, gas production and toxicity, mass loss, etc. Various research has been conducted on TR from a wide perspective, such as cause analysis [7], mechanism [8], propagation after TR [9], etc., and various methods, such as improved internal battery material and battery state technology [10-12], are proposed. The lithium-ion battery electrode manufacturing defects change electrochemical performance, and the effects of various electrode defects such as pinholes and non-uniformity were analyzed, but the results were based on coin cell and showed performance reduction rather than stability [13]. Yi Wu et al. [14], presented a method of identifying defects and structural deformation caused by battery manufacturing with a non-destructive method using computed tomography (CT). In the case of an accident in an application field to which an actual battery is applied, defects in a battery such as welding burr, foreign particles inside a cell, deflected electrode, and separator pores or puncturing could be identified by CT scanning.

Research on TR and methods of diagnosing battery internal defects have been studied a lot, respectively, but research on the relationship between internal defects and TR is not much known. It is suggested that TR is likely to occur due to internal defects, but understanding of the impact is still insufficient. Therefore, this research conducted a TR test on a normal battery cell and a cell with potential defects and compared the results to analyze its characteristics.

II. EXPERIMENTAL SETUP

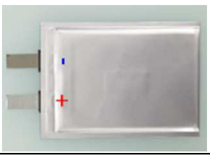
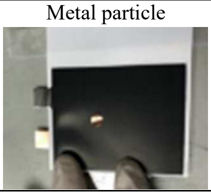
A pouch cell sample representing the internal failure of three types of battery cells was manufactured for research, and a TR experiment was conducted with the corresponding cell.

A. Conditions of the defect cell

In order to confirm the relationship between the internal defect of the lithium-ion battery and the TR, a potential defect holding cell that may cause an internal short circuit, which provides a direct cause of the TR occurrence, was considered. An internal short circuit is one of the main factors that seriously affect the safety of the battery. An internal short circuit refers to a direct or indirect connection between an anode and a cathode inside a battery cell, which may occur even when the battery is damaged or otherwise. If an internal short circuit occurs, a short circuit current instantaneously flows at a short circuit position in a cell, resulting in a fire or explosion of the cell caused by structural damage and side reaction caused by the temperature rise.

In this study, a potential defect in a battery cell was applied assuming three cases in which an internal short circuit may be caused by a potential defect. The first is a faulty form in which the separator of the battery cell is damaged. A battery cell was manufactured by forming a small hole in the battery separator. The second is a fault form in which foreign substances, especially metallic foreign substances, are inserted. A battery cell was manufactured by inserting a thin copper thin film onto the cathode active material of the battery. Finally, a battery cell was manufactured by forming a burr at the edge of the anode current collector by forming a burr. All cells were manufactured with a pouch type battery cell of 2Ah, and after manufacturing, the battery cell was fully charged and discharged to verify the initial performance, it was verified that there is no significant difference in performance from a normal cell and then charged with 100% state-of-charge (SOC). The test case of the produced battery cells is listed in Table 1.

TABLE I
THE CONDITIONS OF TEST CASE

Case number		Fault state & method	Heat condition
1	1-1	Normal 	Continuity
	1-2		
2	2-1		Discontinuity
	2-2		
3		Pinhole 	Continuity
4		Metal particle 	
5		Burr	

B. Experimental set

As an experimental method for TR, a penetration method, a heating method, and the like are applied. Since the penetration method may affect TR progress depending on the penetration position and speed, this research applied a method to use a heating pad. A heating pad was attached to one side of the manufactured pouch cell and slowly heated to induce TR of the cell. At this time, when the heating rate is rapid, TR may be induced due to damage to the attachment surface of the cell, so the heating amount in which the temperature change occurs was supplied to the cell while gradually adjusting the heating amount of the heating pad.

The experimental cases are largely divided into four categories, cases 1 and 2 are experiments using normal cells, and cases 3, 4, and 5 are experiments on cells to which separator damage, metal particle insertion, and burr damage are applied, respectively. The experiment was conducted to review the progression of TR according to potential internal damage to the cell. In addition, in the experiments on the normal cells of cases 1 and 2, the heating of the heating pad was continued until TR generation and the heating of the heating pad was stopped at the time when gas ejection was confirmed. The experiment was conducted to determine whether TR persists in the process of TR, when heating is stopped after gas ejection. The configuration of the set up for the experiment is shown in Fig. 1.

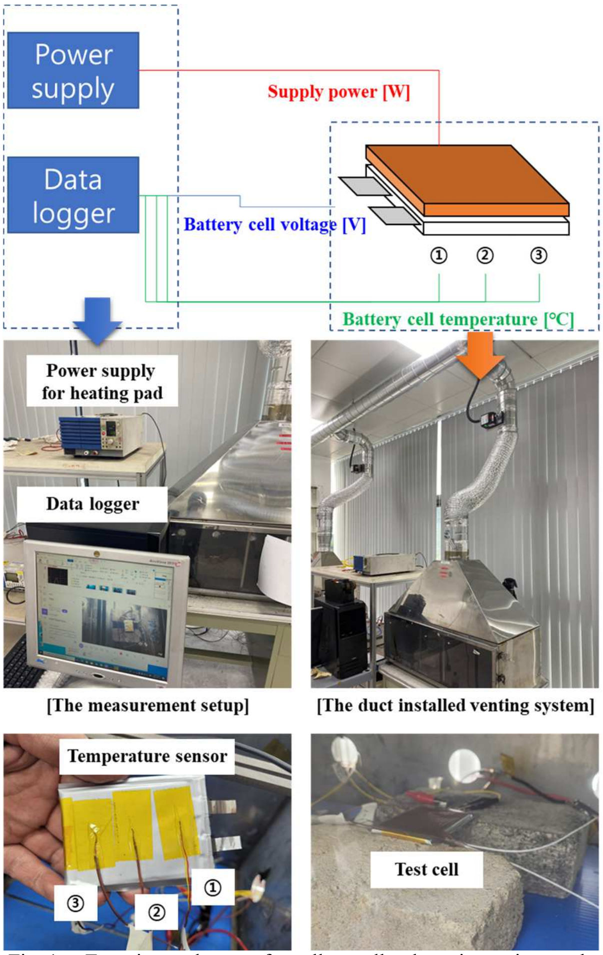


Fig. 1. Experimental set-up for cell-to-cell voltage inconsistency by cycle life test.

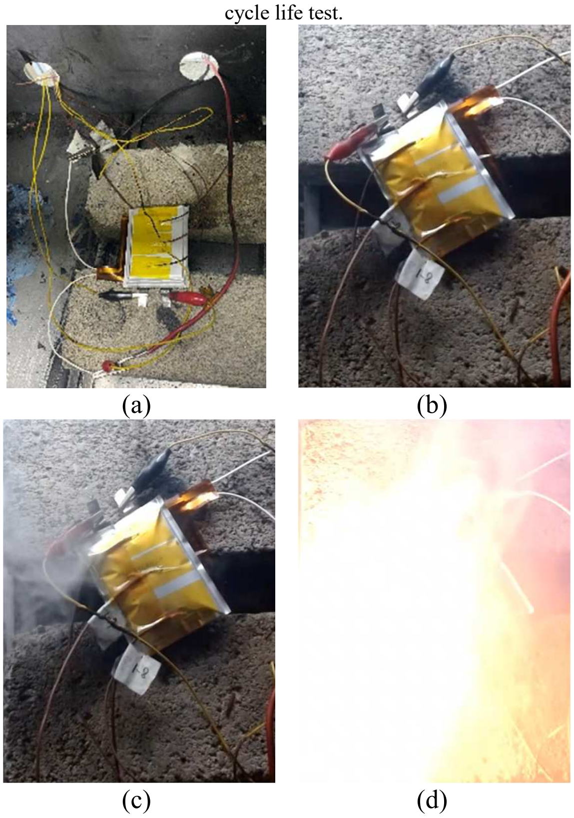


Fig. 2: Temperature-voltage variations along the process of TR: (a) normal, (b) swelling, (c) gas venting and, (d) TR

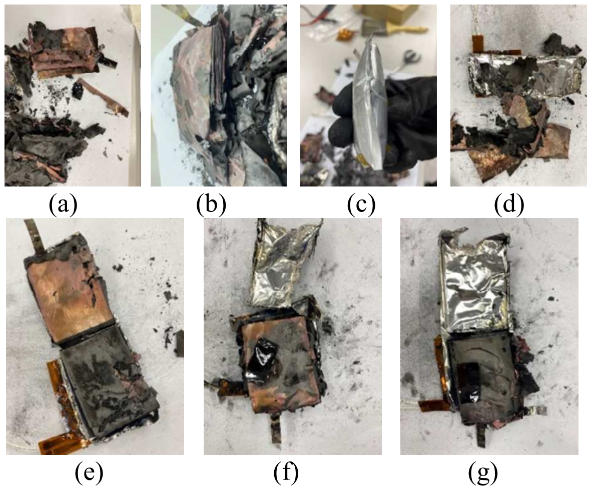


Fig. 3: Status of cells after experimental results: (a) case 1-1, (b) case 1-2, (c) case 2-1, (d) case 2-2, (e) case 3, (f) case 4, (g) case 5

The experiment in all cases was completed within about 30 minutes after the start of the experiment, and the voltage and temperature of the cell measured. And then, the process of performing the experiment was recorded as a video, and the results of normal cells and potentially defective cells were compared according to the TR process.

III. Experimental Results

As for the results of the experiment, the analysis of the progress of TR identified in the experimental results was performed first, and the voltage and temperature measured in each experimental case were reviewed. In addition, analysis of the experimental results was performed to analyze the characteristics of cells with potential defects and normal cells.

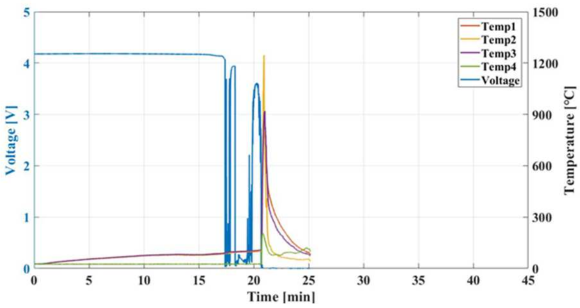
A. Process of the TR

Fig. 2 shows the progress of TR confirmed through the experiment. The TR progress could be divided into a section in which the temperature gradually rises when a cell is heated by a heating pad, a section in which the cell expands and swelling occurs, a section in which an external case of the cell is damaged and gas is ejected, and a section in which the cell is fired. In the case of case 2-1 in which gas ejection is confirmed and heating is stopped, firing did not occur after gas ejection, but in the case of case 2-2, firing was led to firing. That is, except for the case of Example 2-1, the process of heating-swelling-gas ejection-firing was confirmed in all cases. In Example 2-1, after swelling of a cell is expanded and swelled, and then gas is slowly ejected due to damage to an external case, swelling of the expanded cell is reduced, and then the experiment is terminated without a specific phenomenon. In all other cases, as a rapid gas eruption progressed after swelling, an explosion occurred along with an instantaneous fire, and the cell burned down.

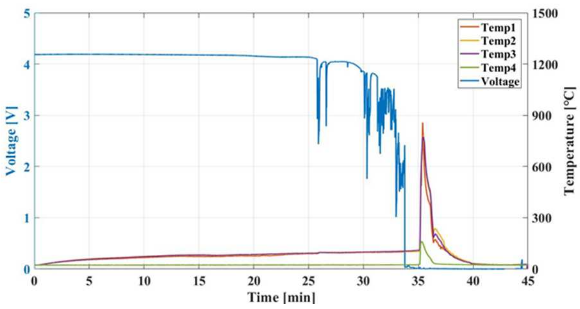
B. Results of the experiment

The experimental results with four normal cells (case 1-1, case 1-2, case 2-1, case 2-2) and three potential failures cells (case 3, case 4, case 5) are presented in Fig. 2 and Fig, 3. Only in the case of the normal cells, it was verified that a voltage would drop sharply at a point where gas is emitted, and in both normal cells and fault cells, the voltage would drop and recover repeatedly, and then TR would start. In particular, when gas release was confirmed and heating was stopped, it was confirmed that TR could stop or continue. In case 1-1 and case 2-2, TR proceeded within 30 minutes and caused an explosion, but in case 1-2, TR occurred after 30 minutes. In case 1-2, the gas emission time continued for 10 minutes from the point of about 25 minutes when gas emission began to the point of about 35 minutes when thermal runaway occurred.

In cases 2-1 and 2-2, the heating of the heating pad was stopped after confirming the gas ejection in the same manner. However, in the result, in Example 2-1, the voltage recovered to a certain level and there was no firing, whereas in Example 2-2, firing and explosion occurred after the voltage drop. As a result of confirming the difference in this case in the video, it was confirmed that in both cases, the gas was slowly emitted in case 2-1 when the external case was damaged and the gas was ejected, while in case 2-2 the fire and explosion occurred as a rapid gas eruption occurred. In other words, case 2-1 stopped TR when the heating was stopped due to the gradual internal reaction, but in case 2-2, it can be inferred that TR continued even when the heating was stopped due to the rapid internal reaction, which also showed a difference in the tendency of voltage drop in the two cases. In the case results of the normal cell, rapid voltage drops and recovery occur repeatedly and then voltage reaches 0 V (cases 1-1 and 1-2), and voltage recovers after voltage drops and does not reach 0 V (cases 2-1). It was confirmed that the voltage reaches 0 V after the voltage is partially recovered after



(a)



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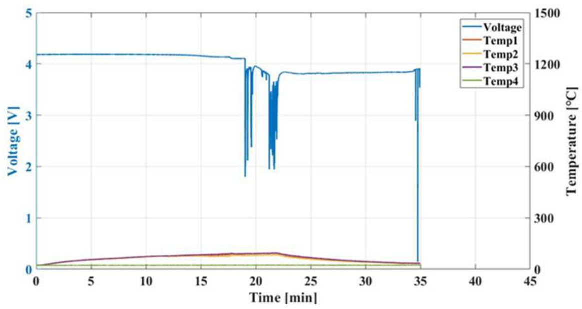

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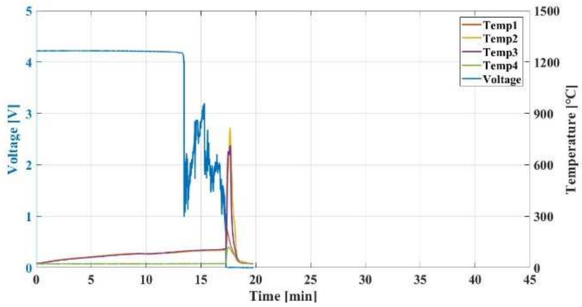
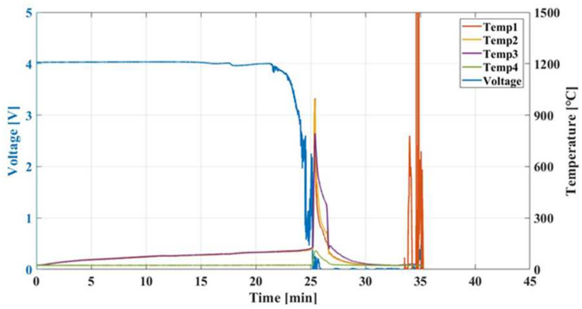


(d)

Fig. 4: Voltage and temperature measured in normal cells: (a) case 1-1, (b) case 1-2, (c) case 2-1, (d) case 2-2.

the rapid voltage drops (case 2-2). When the voltage reaches 0V, it is estimated that the cell is completely short-circuited, and the internal short-circuit occurs suddenly as the internal structure of the cell is damaged by heating, or the short-circuit develop in some parts gradually.

Experimental results like as those of the normal cell were also confirmed in the test results of cells with potential failures. After the swelling of the cell, a rapid gas eruption occurred, and then firing and explosion proceeded continuously. It could be expected that a faulty cell also received a rapid reaction inside the cell by heating, and it could be verified that TR was continuously performed as



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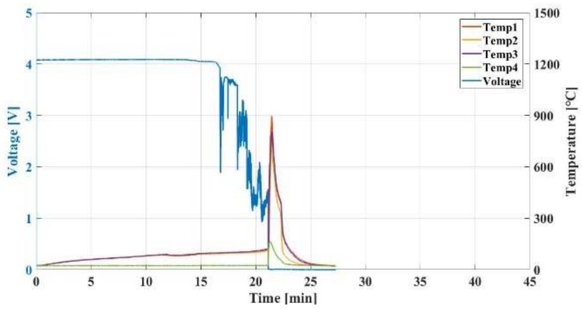

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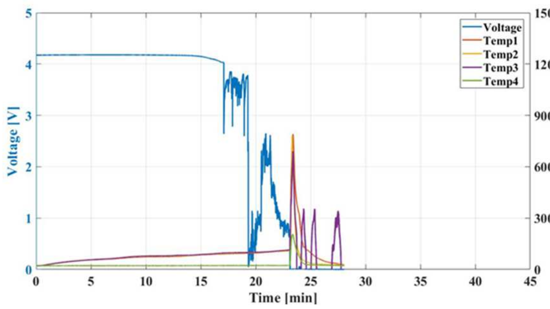


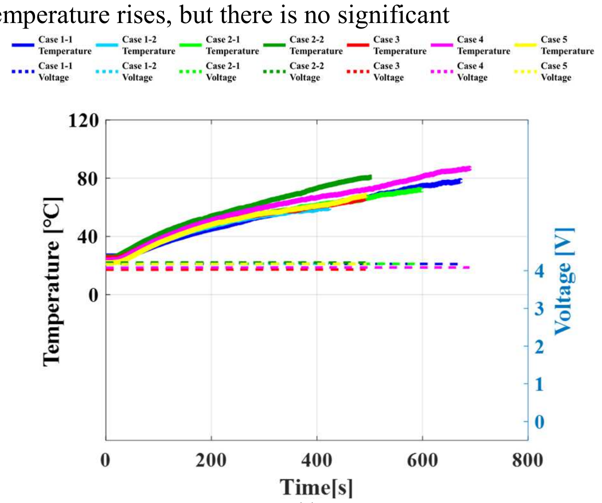
(c)

Fig. 5: Voltage and temperature measured in potential failures cells: (a) case 3, (b) case 4, (c) case 5.

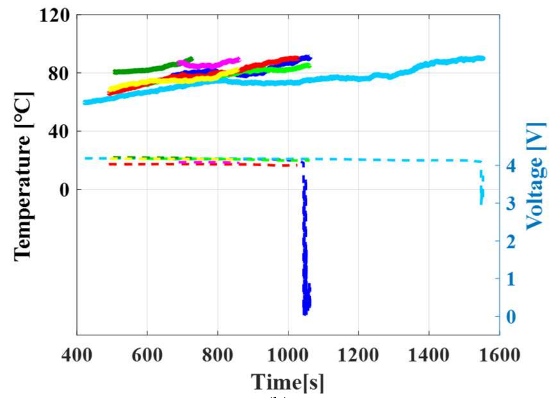
the voltage drop occurred and not recovered even in the tendency of voltage drop. Particularly, the experimental results of a cell with separator damage show a relatively continuous and gentle tendency of voltage drop. This is estimated to indicate a tendency of a gradual voltage drop, not a tendency of a rapid voltage drops, as the potentially existing separator damage area is expanded by heating. If there is a metal insertion therein, it can be found that a relatively gradual voltage drops proceeds and then reaches 0V as compared with a case caused by damage to the burr.

C. Analysis of the experimental results

In all experimental measurement results, the voltage and temperature measured in the experimental results were classified according to the progress of TR. The measurement data for each section are classified into a heating section, a swelling generation section, a gas ejection section, and a firing section, and the results are presented in Fig. 6. The solid line represents the measurement results of temperature (left axis) and the dotted line represents the voltage (right axis). The measurement results for each interval clearly expresses the experimental results. In the heating section (Fig. 6 (a)), the temperature rises, but there is no significant



(a)



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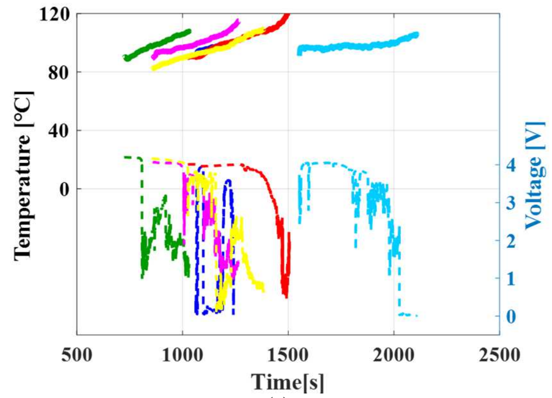

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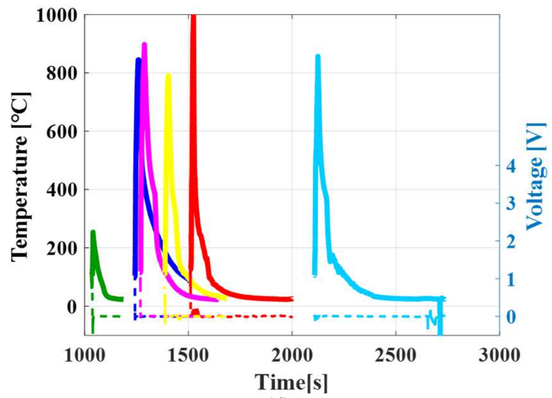

(d)

Fig. 6: Temperature-voltage variations along the process of TR: (a) normal, (b) swelling, (c) gas venting and, (d) TR

voltage change. In the swelling section (Fig. 6 (b)), a voltage change was confirmed in some cells, but in most cases, a result similar to that of the heating section was confirmed. In the gas ejection section (Fig. 6 (c)), continuous temperature rise, and sudden voltage change are confirmed, and in the firing section (Fig. 6(d)), a voltage of 0V and a high temperature rise are confirmed in all cases. In all cases, swelling and gas eruptions occur around 60 to 80°C, and the temperature rises gradually in the swelling section, and it can be seen that rapid temperature rises, large voltage fluctuations, and subsequent firing occur in the gas eruption section.

The TR process of a normal cell and a cell representing a potential failure could not be specified in the analysis of voltage and temperature measured in the experimental results, but the tendency of temperature increase and voltage change for each section could be confirmed during the TR process. If the trend of sudden voltage change is estimated to be connected to internal phenomenon, it can be estimated that the point where gas eruption starts is deformation of the cell due to internal side reaction or electrolyte vaporization by heating, resulting in a rapid voltage change and temperature rise.

IV. Conclusions

This research investigates the characteristics with experiments, in the process of battery TR to analyze the effect of potential defects in the manufacturing process of battery cell on battery performance and safety. The experimental results showed that there was no significant difference in the TR process between the normal cell and the potentially defective cell, but it was confirmed that the voltage of the normal cell was changed rapidly just before the gas was emitted. In addition, when the heating was continued and the heating was stopped after confirming the gas emission, it was confirmed that TR did not occur or occurred at a low temperature. In all cases, it is difficult to verify the possibility of TR of a battery, but even if the battery temperature rises and TR occurs, it is verified that if proper heat dissipation is performed, the damage of TR could be minimized, and transition prevented.

This study analyzed the experimental results of TR that occurs under different conditions through the analysis of the change trend of voltage and temperature measured in the experiment and suggested that TR that proceeds inside the cell could be estimated through the trend of voltage change and temperature change. Further studies will be conducted to further analyze voltage behavior and thermal behavior to study TR progress of cells with potential faults and normal cells. Based on analysis and thermal modeling of heating and self-heating of battery cells, TR modeling methods and TR diagnostic methods will be secured.

Acknowledgment

This research was supported by the Korea Electric Power Research Institute (R21XO02-09) and Korea Institute of Energy Technology Evaluation and Planning (No. 20210501010020).

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