



Thermal behavior and failure mechanism of lithium ion cells during overcharge under adiabatic conditions



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HIGHLIGHTS

- The modified adiabatic method is used to measure the heat generation under overcharge.
- Side reactions contribute 80% heat to thermal runaway in the cases with cycling rate below 1.0 C.
- The inflection and maximum voltages increase linearly with the increasing current rates.
- The decomposed products of cathode materials are soluble with that of SiO_x.
- Lithium plating on anode is due to changes of distance between the cathode and anode.

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ABSTRACT

Cells in battery packs are easily overcharged when battery management system (BMS) is out of order, causing thermal runaway. However, the traditional calorimetry could not estimate dynamic overcharging heat release. In this study, commercial LiCoO₂ + Li(Ni_{0.5}Co_{0.2}Mn_{0.3})O₂/C + SiO_x cells are employed to investigate the dynamic thermal behaviors during overcharge under adiabatic condition by combining a multi-channel battery cyclor with an accelerating rate calorimeter. The results indicate that overcharging with galvanostatic - potentiostatic - galvanostatic regime is more dangerous than that with galvanostatic way. Side reactions contribute 80% heat to thermal runaway in cases below 1.0 C charging rate. To prevent the thermal runaway, the effective methods should be taken within 2 min to cool down the batteries as soon as the cells pass inflection point voltage. Hereinto, the inflection and maximum voltages increase linearly with the increasing current rates. By scanning electron microscope and energy dispersive spectrometer, the decomposed products of cathode materials are suspected to be soluble with SiO_x. The overcharge induced decomposition reaction of Li(Ni_{0.5}Co_{0.2}Mn_{0.3})O₂ is also proposed. These results can provide support for the safety designs of lithium ion batteries and BMS.

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1. Introduction

With the merits of high energy density, long cycle life, low memory effect, slow self-discharge rate and environmental friendliness, lithium ion batteries are widely used in portable electronic productions and has become one of the promising power sources for hybrid and electric vehicles (EV) [1,2]. However, the thermal runaway is still a bottleneck hindering its large-scale application. Moreover, thermal runaway of lithium ion batteries used in

aviation and shipping causes more severe consequences [3,4]. Many factors can cause the battery going to thermal runaway, such as, short circuit [5], heating, overcharge, and more. As one of the main factors, overcharge occurs not only in single cell due to inappropriate design of battery management system (BMS) [6] and malfunction of the charging control system, but also in battery packs affected by internal resistance difference of cells [7]. A single cell is often wrapped by heavy plastic outer layers, as used in cell phone, which offers a limited ventilation environment. This condition gets worse in large battery packs of EVs, since the distance between two cells can be as small as 3 mm [8]. Thousands of cells in battery packs with some inconsistency can easily accumulate heat during charging [9]. Therefore, it is significantly important

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Nomenclature

c	heat capacity of the cell, $1.0 \text{ J g}^{-1} \text{ K}^{-1}$	R_{η}	over-potential resistance of the cell, Ω
C	current rate, non-dimensional	SOC_{end}	state of charge of the cell at the end of overcharge process
C_c	charge capacity, Ah	t	time, s
C_n	the nominal capacity, Ah	t_1	duration of stage 2 of overcharge process under adiabatic condition, s
C_1	capacity of stage 2 of overcharge process under adiabatic condition, Ah	t_2	duration of stage 3 of overcharge process under adiabatic condition, s
C_2	capacity of stage 3 of overcharge process under adiabatic condition, Ah	T	temperature of the cell, $^{\circ}\text{C}$
E_{oc}	open circuit voltage at a certain SOC, V	T_0	temperature at the beginning of a certain period, $^{\circ}\text{C}$
I	current, A	T_{end}	temperature at the end of a certain period, $^{\circ}\text{C}$
m	mass, g	T_{i-p}	temperature of inflection point during overcharge, $^{\circ}\text{C}$
Q	accumulated heat of the cell, J	T_{max}	the maximum surface temperature of the cell, $^{\circ}\text{C}$
Q_h	heat of side reactions, J	T_{onset}	onset temperature of cell self accelerating, $^{\circ}\text{C}$
Q_p	over-potential heat, J	T_{tr}	temperature of thermal runaway, $^{\circ}\text{C}$
Q_{rev}	reversible heat, J	V	voltage, V
\dot{Q}_h	heat rate of side reactions, W	V_{max}	the maximum voltage value of the cell, V
Q_1, Q_2, Q_{end}	accumulated heat of a cell from beginning to V_{i-p} , V_{max} , and the end of overcharge, J	V_{i-p}	voltage of inflection point during overcharge, V
$Q'_1, Q'_2, Q'_{\text{end}}$	charge energy of a cell from beginning to V_{i-p} , V_{max} and the end of overcharge by cyler, J	η	contribution rate of side reaction heat to thermal runaway
r_1^2, r_2^2	variances of Eqs.	$ x $	absolute value of x , x represents $V, V_{\text{max}}, V_{i-p}, E_{\text{oc}}, C_n R_{\eta}$

to investigate the thermal behaviors and failure mechanism of lithium ion batteries during overcharge under quasi-adiabatic condition.

Many related works were conducted to explore the failure mechanism of lithium ion battery during overcharge. However, the experiments were done in non-adiabatic condition and the adiabatic calorimeters were only used to evaluate the thermal risk of cells which were pre-overcharged to a certain voltage outside the calorimeters. Leising et al. [10] divided the overcharge process into four stages by a constant overcharge experiment of the LiCoO₂ (LCO)/graphite (C) cell in ambient temperature and considered that thermal runaway is determined by enlarged resistance. Based on above research, Ohsaki et al. [11] in detailed analyzed the gas production of the four regions of LCO battery and found that the gas generation emerges ahead of the increase of surface temperature, and the main reactions emerge in the cathode side. However, these reactions were thought to heat up the cell. The occurrence of thermal runaway depends on the reaction between deposited lithium and electrolyte, which agrees with Arora's conclusion [12], but is different from that of Leising's group. Feng et al. [13] pre-overcharged a 25 Ah battery in the ambient environment and then tested it in the extended volume-accelerating rate calorimeter (EV-ARC) to study its thermal runaway features. Doughty and Roth [14] and Lamb et al. [15] studied the thermal behavior of components inside 18,650 LCO/C cell in different state of charge (SOC) by accelerating rate calorimeter (ARC) and found the battery is more unstable with higher SOC. Jhu et al. [16] used another adiabatic calorimetry vent sizing package 2 (VSP2) to study thermal runaway potential of 4.2 V 18,650 batteries and Wang et al. [17] used ARC to study the thermal response of battery cycling, but the calorimeters have not been applied for dynamic overcharge study.

In addition to these traditional methods, researchers have also used other methods to observe to get more information about the actual overcharge process. Sasaki et al. [18] overcharged lithium nickel cobalt aluminum oxide (NCA)/C cell and proved that magnesium doping has a good effect on the electrochemical stability of NCA materials through in-situ X-ray diffraction (XRD) and scanning electron microscope (SEM). Erol et al. [19] applied electrochemical impedance spectroscopy (EIS) method to study the impedance of cells with different SOC. Snook et al. [20] coated

the LCO/adhesive/conductive carbon onto the aluminum foil current collector. And then SEM was used to observe the dissolution of cobalt. Park et al. [21] found the reaction of organic electrolyte on Li(Ni_{0.31}Co_{0.32}Mn_{0.28}Al_{0.09})O₂ surface by Fourier transform infrared spectroscopy (FTIR) and X-ray photoelectron spectroscopy (XPS), and more decomposition reaction of the electrolyte solvent than lithium salt was found. In addition, spectroscopic methods such as Raman spectra [22] and other electrochemical research methods such as the rotating disk electrode [23] have also been used.

Although the failure process during overcharge was not fully understood, these exploration works provide a better understanding on the changes after a cell is overcharged, which makes a contribution to battery design and manufacture. Another undergoing work is on the dynamic investigation of batteries during overcharge. However, few have reported on thermal behavior during the overcharge process under adiabatic conditions, and it is necessary to understand how the heat is accumulated during overcharge process. This is critical to understand the thermal runaway and failure process in case of overcharge and then the thermal runaway prevention as well.

In this work, a modified adiabatic calorimetry method by combining a battery cyler with the ARC was used to investigate the heat accumulation and failure mechanism of commercial lithium ion cells during overcharge with different current rates under quasi-adiabatic conditions. The characteristics of voltage and temperature were studied in detail. The outputs can be used in thermal management design on large-scale module and could provide estimation method for the overcharge state according to the energy utilization coefficient.

2. Methodology

2.1. Sample identification

The prismatic commercial lithium ion cell used in the experiments was with the size of 52 mm × 34 mm × 5 mm and the nominal cell capacity is 1.0 Ah. For detailed information of the electrode active material, 100% charged commercial lithium ion cells were disassembled in argon-filled glove box, where the water content

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