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Research Paper

Non-uniform effect on the thermal/aging performance of Lithium-ion pouch battery

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HIGHLIGHTS

- A new method is proposed to get more detail of resistance variation.
- Effect of non-uniform aging is simulated and analyzed.
- Difference of capacity between the uniform and non-uniform aging battery is explained.

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ABSTRACT

Non-uniform distribution of current density and temperature is inevitable especially in high C-rate and it can lead to bad performance of battery. Therefore, the non-uniform effect (non-uniform temperature, current density and aging) on the pouch battery performance is studied with experiment and simulation. A new method, which measures the direct current resistance (DCR) based on the discharge curve, is proposed to get more detail of resistance variation. The measurement shows that the resistance of Lithium-ion pouch battery with non-uniform temperature is similar to that of average temperature. Then, effect of non-uniform aging is simulated based on the electro-thermal coupled model. It is found that battery suffering non-uniform aging has smaller discharging capacity relatively. The main cause for the discharge capacity reduction between the uniform and non-uniform aging battery is the big difference of local stoichiometry of cathode electrode $\theta_{LiFePO4}$. The capacity reduction in 1 C rate occupies about 6% of the permissible capacity loss. Finally, tabs of battery are changed in order to acquire uniform temperature distribution. Battery whose tabs are put on the middle of the top side and the bottom side has a better performance in the opinion of thermal analysis.

1. Introduction

Lithium ion batteries with its increasing specific energy, steady high temperature performance and good cycle-life are gaining popularity for Electric Vehicles (EVs) and Hybrid Electric Vehicles (HEVs) applications. With the increasing demand of high energy density, large format and high-rate charging, heat generation rate will increase but the cooling specific surface area may decrease. That will bring a great challenge to the battery thermal management system (BTMS). In large format lithium batteries, including the cylindrical and prismatic ones, the pathways of the current and the heat generation are not equal at different location of electrodes [1]. Non-uniform distribution of current density and temperature [1,2] is inevitable especially in high C-rate (where C is the rate at which the full charge capacity is delivered in 1 h). The non-uniformity of current density can be caused by temperature gradient [3] and the location of tabs [4]. An in-plane SOC distribution was visualized in Ref. [5] using synchrotron X-ray micro diffraction, which implies that higher currents exist near the tab regions. Many researchers are devoted to measure the current distribution by adding some tabs to the electrode [6,7]. Osswald's [2] team analyzed the non-uniform distribution of SOC (state of charge) by measuring of local current with in-situ measure method. Guangsheng Zhang's [8] team showed that the number of tabs can have a strong impact on the current distribution. Non-uniform distributions will lead to under-utilization of active materials in a large cell, thereby reducing its energy density.

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Nomenclature

dV/dQ	the differential voltage
$I_{Average}$	the average discharge current
I_{ocv}	the discharge current at the open circuit condition
I_{test}	the discharge current at specific condition
Q	the capacity of the battery
Q_{graph_max}	the maximum capacity for graphite anode electrode
Q_{graph_nom}	initial nominal capacity for graphite electrode
$Q_{LiFePO4\_max}$	the maximum capacity for LiFePO4 cathode electrode
$Q_{LiFePO4\_nom}$	the initial nominal capacity for cathode electrode
Q_{Li_graph}	the present capacity for graphite electrode
$Q_{Li\_LiFePO4}$	the present capacity for cathode electrode
Q_{Li_nom}	initial nominal capacity for lithium inventory
R	the total resistance of the battery
$R_{\_25^\circ C}$	the resistances measured with the new method at 25 °C
$R_{\_31^\circ C}$	the resistances measured with the new method at 31 °C
$R_{\_nonuniform}$	the resistances measured with the new method at non-uniform temperature distribution
$R_{\_25^\circ C\_HPPC}$	the resistance measured with HPPC method at 25 °C.
U_n	the equilibrium potential of graphite electrodes after discharge
U_{ocv}	the open circuit voltage
U_p	the equilibrium potential of $LiFePO_4$ electrodes after discharge
U_{test}	the terminal voltage of the measurement
(x,y)	the coordinate of the cell element
θ_{graph}	the local stoichiometry of graphite anode electrode
$\theta_{LiFePO4}$	the local stoichiometry of $LiFePO_4$ cathode electrode

Moreover, high current density near the tabs will lead to overcharge or over-discharge, which will create the localized failure [9].

As the current density near tabs is larger, temperature near the tabs location is higher because the joule heat, irreversible electrochemical reaction heat and reversible heat is positively related to current. The temperature and temperature difference can become more serious when they are packed together as the specific area of the packs will decrease. Although a number of models have been implemented in order to study such non-uniformity, its effect on the batteries' performance is seldom described quantitatively. Cycle life of the battery is strongly depending on the temperature according to many cycle aging experiments [10,11]. High temperature will lead to rapid side reaction rate and rapid growth rate of SEI (solid electrolyte interface) at the anode. The non-uniform temperature distribution will lead to localized aging. More seriously, both high current density and high temperature occur at the position near tabs. That will accelerate the localized aging rapidly.

Temperature can also make a great difference to the batteries' capacity, resistance [12]. The non-uniformity may change the capacity and resistance of the battery. However, the temperature distribution of batteries is uniform during such parameters measurement. That may bring errors to the battery pack design, BMS (battery management system) measurement and aging evaluation. Therefore, it is necessary to study the deviation of the parameters between the uniform situation and non-uniform situation.

In this work, the resistance and capacity of the battery with non-uniform temperature is measured. Then, the performance of non-uniform aging is simulated and analyzed. Finally, the position of tabs is changed and thermal behaviour of single battery and battery pack is studied.

2. Experiment

2.1. Method

Resistance measurement is quite important for the study of battery performance. Electrochemical impedance spectroscopy (EIS) is widely used to measure the detail of the resistance, but this method is limited for the large format battery as the increasing cost of the equipment. Another method is the Hybrid Pulse Power Characterization (HPPC) Test which is used to measure the direct current resistance (DCR) [13–15]. The DRC is the sum of pure ohmic resistance $\Delta V_{ohmic}/\Delta I$ and polarization resistance $\Delta V_1/\Delta I$, which is the result of the charge transfer reaction. The ohmic resistance can be measured by recording the immediate drop of voltage (ΔV_{ohmic}) after the applying current pulse. In the next several seconds, the voltage continues to drop because the lithium surface concentration of the active mass particles changes. The HPPC test is simple but always only some points (such as SOC = 0%, SOC = 50%, SOC = 100%) are chosen for measured. Some detail during the discharge/charge is overlooked in this way.

As a result, we propose a new method to measure the direct current resistance (DCR) based on the discharge curve. Firstly, a 20 Ah LiFePO4/graphite pouch battery is charged with C/3 rate to the cutoff voltage 3.65 V using LANDCT2001B. The battery is followed with constant voltage charge (3.65 V) until the current reaches C/20. Secondly, the fully charged battery rests for 3 h to reach the electrochemical equilibrium. Thirdly, the rest battery is discharged at C/25 (0.8 A) rate to the cutoff voltage (2.70 V) and the discharge voltage curve is recorded with Agilent 34972A. With the same procedure, the battery is then discharged at C/4 rate (5 A). Small discharge rate (C/4) will not lead to obvious temperature rise (less than 1 K). It can eliminate the effect of temperature rise in the way. During the whole measurement, the battery is placed into a temperature chamber and the ambient temperature is controlled.

The resistance of the battery can be calculated by equation:

$$R = \frac{\Delta U}{\Delta I} = \frac{U_{ocv} - U_{test}}{I_{test} - I_{ocv}} \quad (1)$$

where the U_{ocv} and U_{test} are the voltage of the battery measured at the same SOC but different discharge current. I_{ocv} and I_{test} are the discharge current. The voltage at C/25 rate is approximate to the open circuit voltage (OCV).

As U_{ocv} and U_{test} come from two different voltage curves, the remained problem is how to ensure that these two voltages are measured at the same SOC. To solve the problem, the dV/dQ curve is used because the peak in the dV/dQ curve represents the phase transform [16]. The differential voltage (dV/dQ) against capacity for individual electrodes is shown in Ref. [17]. The negative electrode dV/dQ curve shows peaks for the graphite staging while the dV/dQ curve for the $FePO_4$ electrode is fairly constant, with little variation. Therefore, the peaks in Fig. 1 almost come from the graphite electrode. As can be seen, the x-coordinates of peak #1 and peak #1′ in the differential voltage curve are different. The distance between two peaks in the same dV/dQ curve (such as peak #1 and peak #2, or peak #1′ and peak #2′) is explained to represent the amount of lithium that can be stored in the graphite electrode for a particular stage [17]. However, as no extra aging experiment