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# A new charging mode of Li-ion batteries with $LiFePO_4$/C composites under low temperature

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**Abstract** Li-ion batteries with $LiFePO_4$/C composites are difficult to be charged at low temperatures. In order to improve the low temperature performance of $LiFePO_4$/C power batteries, the charge–discharge characteristics were studied at different temperatures, and a new charging mode under low temperature was proposed. In the new charging mode, the batteries were excited by current pulses with the charge rates between 0.75 C and 2 C, while the discharge rates between 3 and 4 C before the conventional charging (CC–CV). Results showed that the surface temperature of Li-ion battery ascended to 3 °C at the end of pulse cycling when the environment temperature was −10 °C. Comparing with the conventional charging, the whole charge time was cut by 36 min (23.4%) and the capacity was 7.1% more at the same discharge rate, respectively.

**Keywords** Charging mode · Li-ion battery · $LiFePO_4$ · Low temperature

## Introduction

Li-ion batteries have been applied in many portable electronic devices because of a high energy density. Meanwhile, Li-ion batteries with a high energy density are required for an improvement of high-rate performance in order to use high power devices as power sources of electric and hybrid vehicles [1]. Olivine-type $LiFePO_4$ is regarded as the most promising alternative cathode material for Li-ion batteries because of its advantages of higher theoretic capacity (170 mAh $g^{-1}$), lower cost, higher safety, and environmental friendliness [2, 3]. But still a number of papers have addressed the problems of poor low temperature performance of Li-ion batteries [4–9]. In General, both the power and energy of the Li-ion battery are substantially lost as the temperature falls below −10 °C, and charging of a discharged Li-ion battery is more difficult than discharging of a charged battery at low temperatures. Zhang et al. [7–9] explained the above phenomena of Li-ion batteries: (1) impedances of Li-ion cells are composed of bulk resistance ($R_b$), solid-state interface resistance ($R_{sei}$) and charge-transfer resistance ($R_{ct}$); (2) the $R_{ct}$, which can be linked to kinetics of the cell electrochemical reaction, is most significantly increased and becomes predominant as the temperature falls to below −10 °C; (3) the delithiated graphite and lithiated cathode, both of which correspond to a discharged state in a Li-ion battery, have much higher $R_{ct}$ at a low temperature than when charged. The successful improvement of the low temperature performance is mainly based on two approaches: (1) formulating new solvent mixtures to lower freezing temperature of the liquid electrolytes [6, 10, 11] and (2) replacing the existing $LiPF_6$ salt with $LiPF_6$ to reduce charge-transfer resistance of the batteries [9]. However, researches are rarely focused on the point of how to charge a Li-ion battery effectively and quickly at a low temperature.

The conventional Li-ion battery charging (CC–CV) occurs in two steps, the battery is charged at a constant current (e.g. 1 C) until the potential reaches the upper voltage limit followed by constant voltage charging until the current reaches a predetermined small value. At low temperatures the substantially high $R_{ct}$ produces high IR

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polarization, which brings the battery voltage rapidly to the upper voltage limit, and the constant voltage charging seriously extends the charging time.

In this work, the emphasis was on the surface temperature of Li-ion batteries. Although Yang, Sato, Saito and Viswanathan et al. have analyzed the thermal behaviors of secondary batteries for electric vehicles [12–16], the spotlight is outside the heat utilization. The heat produced by a Li-ion battery during discharge process is much more than the heat in the process of charging, so we took advantage of this electrochemical property and proposed a new charging mode under low temperature.

## Experimental

### Battery and equipment

The commercial Li-ion batteries, VLP27/80/112S-Fe from Voltix Energy Co., were used for all experiment. The battery voltage and capacity are rated at 3.2 V and 12 Ah, the maximum charging and discharging rates are 2 C and 4 C, respectively. The battery is fabricated with the electrodes, separator, and electrolyte. The negative electrode is graphite and the electrolyte is 1 mol/L $LiPF_6$ in EC + EMC + DMC (1:1:1 in volume). The tests were carried out on a Digatron universal battery tester (UBT) 300–060 produced by Digatron Firing Circuits, which is capable of charging and discharging the battery at a maximum rate of 300 A. It can operate in several modes such as constant-current and constant-voltage. It comprises voltage, current and temperature measurements and a computer interface. Table 1 summarizes the specifications of the UBT and the test arrangement is shown in Fig. 1. The software "Digatron BTS 600" allows programming the test-procedure and logging data like time, current, voltages, power, temperature, watt-hours, and ampere-hours. An environmental oven was used to provide a constant temperature environment for the tests at various temperatures. The Li-ion batteries were put in the oven under constant temperature for 2 h before use. A Fluke Ti32 Industrial-Commercial Thermal Imager was also used to detect surface temperature changes of the Li-ion battery.

**Table 1** Specifications of the Digatron UBT 300-060 universal battery tester

| | |
|---|---|
| Maximum discharging current/A | 300 |
| Maximum charging current/A | 300 |
| Voltage range/V | 0–60 |
| Error (current measurement)/mA | ±0.5% but not better than ±50 |
| Error (voltage measurement)/mV | ±5 |
| Error (temperature measurement)/K | ±0.1 |

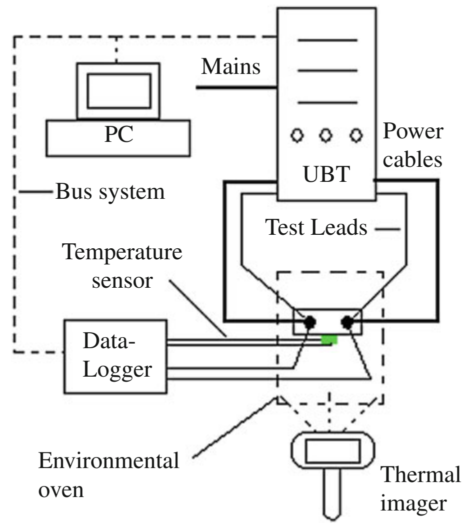


**Fig. 1** Arrangement for testing the Li-ion battery on the Digatron battery tester

### Experimental process

This research consisted of two experiments. For the first experiment, the Li-ion batteries were both charged and discharged at −10 °C, 0 °C, 10 °C and 20 °C. The charge process was under the following scheme: (1) the battery was charged at a constant current of 1 C to 3.6 V, (2) the battery voltage was held constant at 3.6 V until the current dropped to 1.0 A. The batteries were discharged according the following scheme: (1) the battery was discharged at the 3 C rate down to 2.1 V, (2) the battery was kept for 20 min at the open circuit, (3) the battery was discharged at the 0.2 C rate down to 2.1 V. Because of concentration polarization, the open circuit voltage of Li-ion battery rapidly rises after the end of discharging at high discharge rates, and the battery could not be discharged completely. Therefore, step (3) was added to the discharge process. From the first experiment, the charge-discharge characteristics of Li-ion batteries at different ambient temperatures were studied.

For the second experiment, the battery was excited by high-rate current pulses with the charge rates between 0.5 C and 2 C, while the discharge rates between 3 C and 4 C before the conventional charging (CC–CV) at −10 °C. The effects of pulse excitation on surface temperature of Li-ion batteries and conventional charging (CC–CV) were studied from the second experiment.

## Results and discussions

### Charging and discharging characteristics of Li-ion battery

Figure 2a compares the current curves of Li-ion batteries in charge process at various temperatures. It is shown that with the decreasing temperature, the charging time is increased rapidly. At 20 °C, the charging current maintains 12 A for 50 min and approximately 80 min are needed to fully charge the battery, while at −10 °C the charge process requires about 150 min. At low temperatures, the substantially high $R_{ct}$ produces high IR polarization as soon as a high current is applied to the discharged Li-ion battery. Owing to the high IR polarization, the charge voltage is immediately jumped up to the cut-off limit so that the constant-current charging ends and starts the constant-voltage charging. Thus, the charging of a discharged Li-ion battery is more difficult at low temperatures than at room temperature.

Figure 2b shows the discharge curves of Li-ion batteries at various temperatures. In the process of discharging at 3 C rate and −10 °C, the battery voltage is not falling all the time. In the initial stage of discharging, the voltage falls quickly, but 100 s later, it begins to rise and when the voltage reaches 2.53 V, it falls again until below 2.1 V. This phenomenon proves that the total resistance of the Li-ion battery is not constant in discharge process, and the change of resistance makes the fluctuation of voltage across the battery. However, this phenomenon is not obvious at 20 °C, which indicates that the impedance change is particularly evident at low temperatures and is mainly attributed to the change of internal temperature of the Li-ion battery (to be discussed latter).

Figure 2c shows the discharge capacity and the dc resistance ($R_{dc}$) corresponding to the ambient temperatures. The discharge capacities at −10, 0, 10, and 20 °C are 4.84, 8.52, 10.06, 10.93 Ah, respectively. As can be seen, the discharge capacity at −10 °C loses almost 56% of its discharge capacity at 20 °C, and there exists a rapid decrease in the relative capacity as the temperature falls to below 0 °C. Thus, the main problems of the low temperature performance of Li-ion batteries are not only the long charging time but also the great loss of discharge capacity (which originates from the inner resistance [7]). It can also be found that the $R_{dc}$ is most significantly increased with decreasing temperature. The $R_{dc}$ at −10 °C is more than 150% of the $R_{dc}$ at 20 °C.

The change of surface temperature of the Li-ion battery at ambient temperature of −10 °C is shown in Figs. 2d and 8. The change of surface temperature is not obvious in charge process, which means a small amount of heat generated by the battery, however, the surface temperature rises rapidly in discharge process. It can be found that the

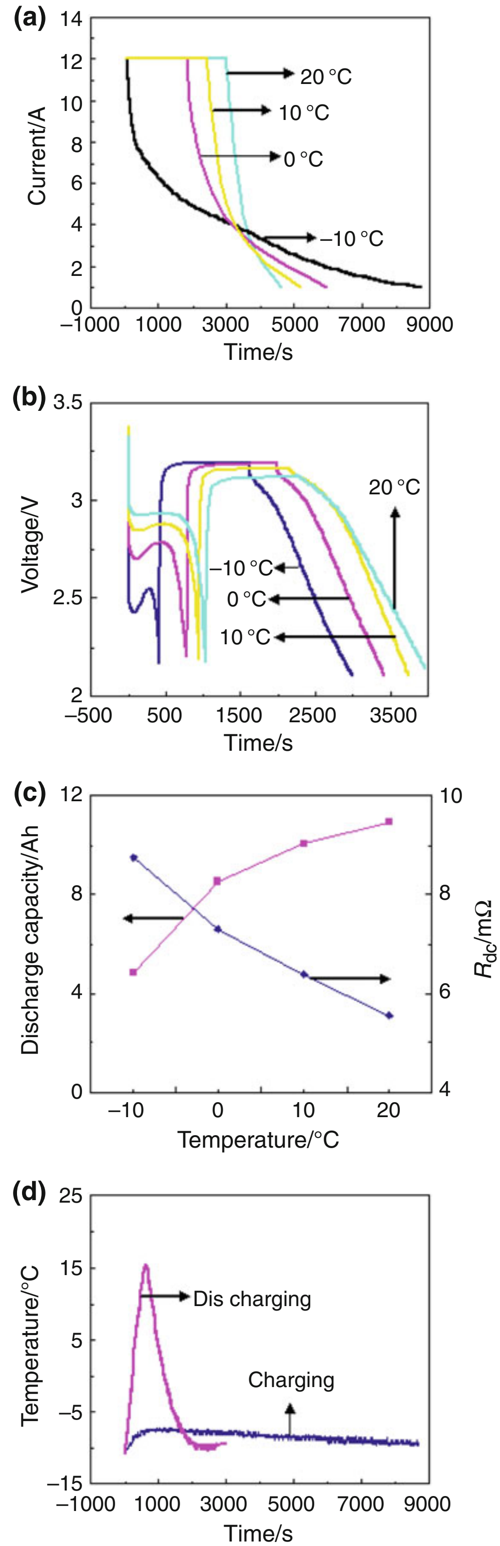


**Fig. 2** The charge–discharge characteristic of Li-ion batteries: **a** charging current with respect to time at different temperatures; **b** the voltage curves during discharge process at different temperatures; **c** the discharge capacity and the dc resistance corresponding to the ambient temperature; **d** the surface temperature curves in the charge and discharge process at −10 °C

surface temperature of the Li-ion battery is higher than 15 °C at the end of discharge process. After the end of discharging at 3 C, the surface temperature falls quickly to −10 °C, and then it rises again with the discharging current of 2.4 A (0.2 C), but this phenomenon is not obvious, which indicates that the value of discharging current determines the ascent rate of internal temperature of the Li-ion battery. Additionally, the change of the surface temperature in Fig. 2d can be used to explain the V-shape discharge voltage curves in Fig. 2b. Because at low temperatures the $R_{ct}$ is considered to be a predominant factor to influence the cycling performance of the Li-ion battery [8], the heat generated by the battery in the early stage of discharging increases inner temperature of the battery and makes $R_{ct}$ decrease consequently ($R_{sei}$ and $R_b$ also decrease), which leads to the decrease of voltage across the total resistance. It should be point out that although the total resistance also changes depending on battery voltage, the influence of voltage is small in the first half of discharge process [8].

## Heat generation during charge and discharge process

Viswanathan et al. have analyzed the reversible and irreversible heat of Li-ion batteries during charge and discharge process [12]. A battery releases and absorbs heat during charge and discharge reactions. If the battery reaction progresses in reverse, then the discharge and charge reactions are the reverse of one another, and so the intake and release of heat is reversed as well. The reaction heat value $Q_r$ can be expressed with Eq. 1

$$Q_r = T\Delta S\frac{I}{nF} \tag{1}$$

where $I$ is the current density (A cm$^{-2}$), $T$ is temperature (K), $\Delta S$ is entropy change (J mol$^{-1}$ K$^{-1}$), $n$ equals the number of electrons per reaction, and $F$ is the Faraday constant. Based on one $Li^+$ exchange during charge or discharge, the entropy change is given by

$$\Delta S = \frac{nF\partial E}{\partial T} \tag{2}$$

where $E$ is the open circuit potential. The battery reaction also results in side reactions, self-discharge and such due to electrolyte decomposition, which are manifested as thermal factor $Q_s$ [13]. Also added is Joule heat $Q_J$, which is caused by the battery's electrical resistance component. Total heat generation in the battery reaction, $Q_t$, is therefore expressed with Eq. 3.

$$Q_t = Q_r + Q_s + Q_J \tag{3}$$

During charging, the reaction heat $Q_r$ in a Li-ion battery corresponds to the heat absorbed when lithium ions from the positive electrode are intercalated by the negative electrode. In addition, a quantity of heat equivalent to this is generated during discharge process, and the endothermic reaction will not appear.

Heat generation that is dependent on the battery's internal resistance is manifested as Joule heat $Q_J$. The Joule heat, which is irreversible, is always exothermic during charge and discharge process. In addition, $Q_s$ is small enough to disregard in comparison to other quantities of heat [13, 14]. The thermal behavior during charge and discharge can be used to explain the surface temperature curves in Fig. 2d and the images in Fig. 8. During discharging, both the heat of reaction and Joule heat are exothermic, and the temperature rises continuously. During charging, however, the reaction heat is endothermic and the total heat generation is small.

Therefore, based on the results from the first experiment, we suggest that the charge performance of Li-ion batteries at low temperatures may be improved with current pulses. If a battery is cycled at high discharge rates before the conventional charging (CC–CV), the heat produced by the battery in discharging should make the temperature inside it rise rapidly and make the total resistance decrease (especially for $R_{ct}$).

## The effect of pulse excitation

The first test of pulse experiment was divided into four groups: P6-36, P9-24, P9-36, and P9-48 (see Table 2), and the surface temperature curves of Li-ion batteries are shown in Fig. 3.

**Table 2** The first pulse test on Li-ion batteries

| Group number | Charging current/A | Discharging current/A | Cut-off voltage/V | Cycle number |
|---|---|---|---|---|
| P6-36 | 6 (0.5 C) | 36 (3 C) | 2.1–3.6 | 10 |
| P9-24 | 9 (0.75 C) | 24 (2 C) | 2.1–3.6 | 10 |
| P9-36 | 9 (0.75 C) | 36 (3 C) | 2.1–3.6 | 10 |
| P9-48 | 9 (0.75 C) | 48 (4 C) | 2.1–3.6 | 10 |

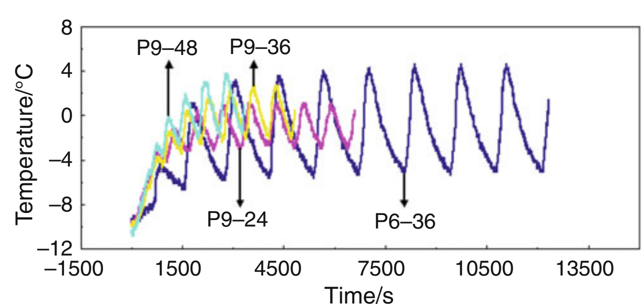


**Fig. 3** The surface temperature curves with different pulse currents at −10 °C

In the early stages of charge–discharge cycles, the surface temperature of Li-ion batteries rises rapidly, however, as the process continues, there appear periodic fluctuations in surface temperature, and the temperature does not go up anymore. The maximum surface temperature which exists among the four groups is about 5 °C. It can also be seen from Fig. 3 that the value of discharging current determines the ascent rate of surface temperature, when the charging current remains unchanged (0.75 C). The larger the discharging current is, the higher the surface temperature will be. Nevertheless, the battery's temperature drops quickly during charge process. In order to analyze the relationship between the charging current and the ascent rate of surface temperature, we extract the 4th discharge–charge cycle in P9-36, as shown in Fig. 4. Since the slow heat transfer from the battery to the temperature detector, the variation of surface temperature slightly lags behind the change of current in Fig. 4. However, the periods of temperature rising and dropping are approximately equal to the periods of discharging and charging respectively. In order to analyze the temperature variation, this lag is disregarded.

In Fig. 4, $T_0$ is the initial surface temperature following the last cycle, $T_1$ is the surface temperature of the Li-ion battery at the end of charge process and $T_2$ is the surface temperature at the end of discharge process which is also the termination of this cycle. Meanwhile, $t_1$ is the charging time in this cycle and $t_2$ is the discharging time. So, we obtain Eqs. 4 and 5:

$$T_0 - T_1 = k_d t_1 \quad (4)$$

$$T_2 - T_1 = k_r t_2 \quad (5)$$

where $k_d$ is the temperature dropping coefficient, $k_r$ the temperature rising coefficient. It is clear that $k_d$ is determined by battery's physicochemical properties, the difference in temperature between the environment and the battery, and the charging current. The temperature rising coefficient $k_r$ is also associated with battery's physicochemical properties, the difference in temperature between the battery and the environment, and the discharging current. Moreover, the larger the discharging current is, the higher $k_r$ will be. Besides, we can also get the equations of the charging time $t_1$ and the discharging time $t_2$:

$$t_1 = \frac{\int I_{ch} dt}{I_{ch}} = \frac{Q_c}{I_{ch}} \quad (6)$$

$$t_2 = \frac{\eta \int I_{ch} dt}{I_{dch}} = \frac{\eta Q_c}{I_{dch}} \quad (7)$$

where $Q_c$ and $\eta$ are the charge capacity of the Li-ion battery and the charging coefficient respectively, $I_{ch}$ is the charging current and $I_{dch}$ is the discharging current. Contrary to the relations between battery temperature and $Q_c$, $Q_c$ decreases as $I_{ch}$ increases. Therefore, the ascent rate of surface temperature in one cycle can be expressed with Eq. 8.

$$\frac{T_2 - T_0}{t_1 + t_2} = k_r - \frac{k_d + k_r}{\eta I_{ch} + I_{dch}} I_{dch} \quad (8)$$

In order to analyze the influence of charging current on $k_d$ and $\eta$, the values of $k_d$ and $\eta$ in P6-36 and P9-36 are plotted as a function of cycle number in Fig. 5. It can be found that in the early stages of cycles, the $k_d$ and $\eta$ vary according to the charging current, however, five cycles later, the $k_d$ and

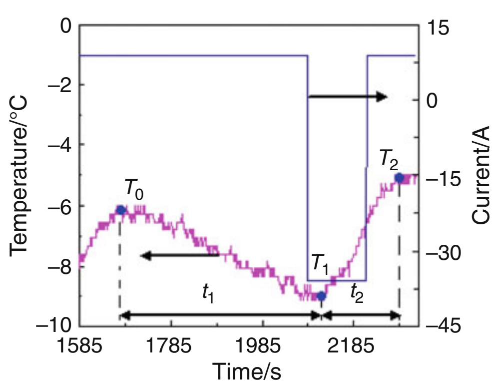


**Fig. 4** The battery surface temperature and pulse current curves (P9-36)

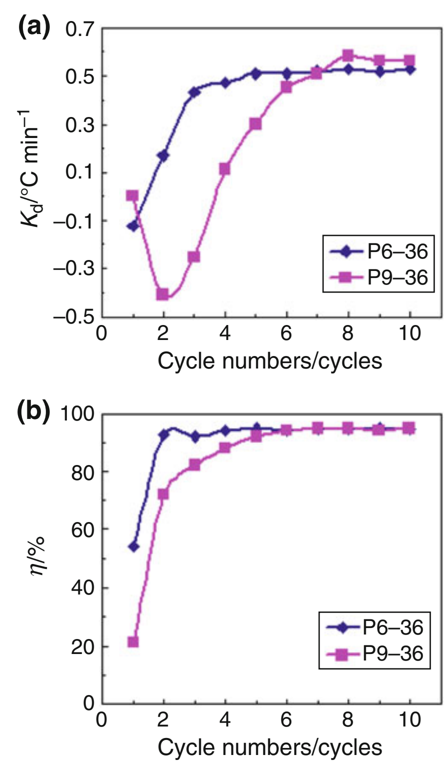


**Fig. 5** $k_d$ and $\eta$ as a function of cycle number

$\eta$ become stable at about 0.54 K/min and 94%, respectively, and are no longer influenced by charging current. Therefore, we may conclude from Eq. 8 that if the charging current is increased after several cycles, the ascent rate of battery temperature would be improved.

The Li-ion battery is difficult to be charged in the first cycle at low temperatures. If big charging current is used at the beginning of pulse experiment, the electrical quantity charged into the battery may approach zero, which creates a phenomenon that the battery is cycled without any change of surface temperature. So the charging current should not be too large in the initial stage of pulse experiment. The second test of pulse experiment was divided into three groups: N1, N2, and N3 (see Table 3). The discharging current was still 36 A (3 C), and the cut-off voltages were 2.1 V and 3.6 V, respectively. The charging current increased with cycle number, and the surface temperature curves of Li-ion batteries are shown in Fig. 6.

Among the three groups, the rising rate of charging current is the quickest in N3 and the slowest in N1, so the change of surface temperature is most distinctive in N3. The results of the second test just prove the correctness of Eq. 8. Comparing Figs. 6 and 3, it can be found that the way to increase charging current with cycles remarkably improves the ascent rate of surface temperature. Especially in N3, the surface temperature of Li-ion battery is over 5 °C in half an hour.

**Table 3** The second pulse test on Li-ion batteries

| Cycle index | Charging current | | |
|---|---|---|---|
| | N1 | N2 | N3 |
| 1 | 0.75 C | 0.75 C | 0.75 C |
| 2 | 0.75 C | 0.75 C | 0.75 C |
| 3 | 0.79 C | 0.79 C | 0.83 C |
| 4 | 0.79 C | 0.79 C | 0.92 C |
| 5 | 0.83 C | 1.00 C | 1.00 C |
| 6 | 0.83 C | 0.92 C | 1.08 C |
| 7 | 0.88 C | 1.08 C | 1.17 C |
| 8 | 0.88 C | 1.17 C | 1.25 C |
| 9 | 0.92 C | 1.25 C | 1.33 C |
| 10 | 0.92 C | 1.33 C | 1.42 C |
| 11 | 0.96 C | 1.42 C | 1.5 C |
| 12 | 0.96 C | 1.50 C | 1.58 C |
| 13 | 1.00 C | 1.50 C | 1.67 C |
| 14 | 1.00 C | 1.50 C | 1.75 C |
| 15 | 1.08 C | 1.50 C | 1.83 C |
| 16 | 1.17 C | 1.50 C | 1.92 C |
| 17 | 1.17 C | 1.50 C | 2.00 C |
| 18 | 1.17 C | 1.50 C | 2.00 C |

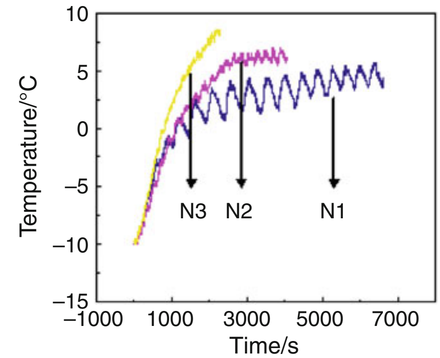


**Fig. 6** The surface temperature curves with different pulse currents at −10 °C

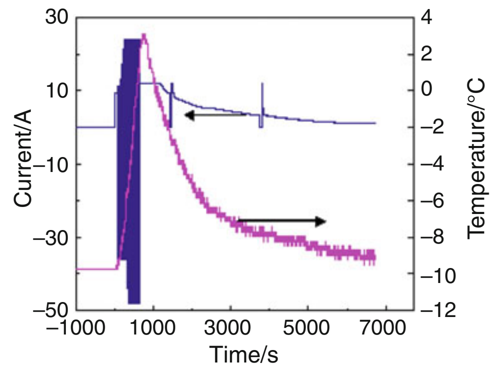


**Fig. 7** The surface temperature and current at −10 °C

In order to further accelerate the ascent rate of battery temperature and shorten the time of pulse process, the discharging current was changed to 4 C (48 A) in the last 12 cycles of N3. After the pulse process, the battery was charged at 1 C (12 A) until the potential reached the upper voltage limit (3.6 V) followed by constant voltage charging until the current reached 1 A. At the same time, to overcome concentration polarization, two short relaxation periods were applied during the charge process, and the time of relaxation was 2 min. The whole process is shown in Fig. 7. It can be found from Figs. 7 and 8 that the surface temperature of Li-ion battery exceeds 3 °C at the end of pulse process, and the time of charging at 1 C exceeds 10 min. The whole charge time is 114 min, which is cut by 36 min (23.4%) comparing with the conventional charging (CC–CV) at −10 °C. The battery was also discharged under the scheme in the first experiment and the discharge capacity was 5.18 Ah, which was increased by 7.1% comparing with the conventional charging. Therefore, the new charging mode remarkably improved the charge efficiency at −10 °C.

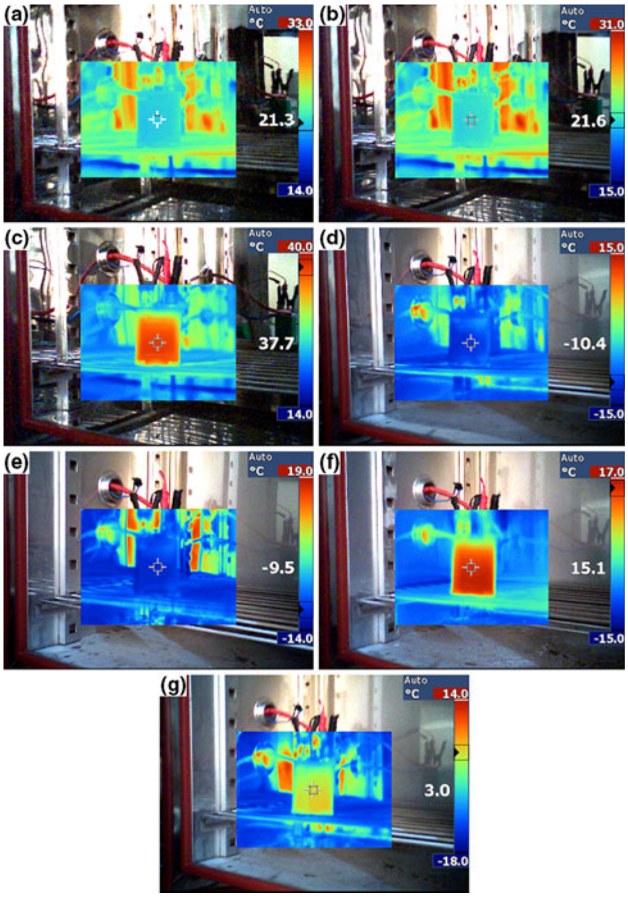


**Fig. 8** The surface images of Li-ion battery: **a** static state at 21 °C; **b** at the end of charging at 1 C (21 °C); **c** at the end of discharging at 3 C (21 °C); **d** static state at −10 °C; **e** at the end of charging at 1 C (−10 °C); **f** at the end of discharging at 3 C (−10 °C); **g** at the end of pulse excitation in the new charging method

## Conclusions

At low temperatures, the substantially high inner resistance results in the poor performance of Li-ion battery, and that the much larger resistance at the fully discharged state makes the charge process very difficult. The heat produced by the battery in discharging can make the temperature inside it rise rapidly. Therefore, we made use of this electrochemical property and suggested a new charging mode for Li-ion batteries. The battery was excited by high-rate pulses with the charge rates between 0.75 C and 2 C and the discharge rates between 3 C and 4 C before the conventional charging (CC-CV) at −10 °C. The surface temperature of Li-ion battery ascended to 3 °C at the end of pulse cycling. Comparing with the conventional charging, the whole charge time was cut by 36 min (23.4%) and the capacity was 7.1% more at the same discharge rate.

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