A Rapid Low-Temperature Internal Heating Strategy with Optimal Frequency Based on Constant Polarization Voltage for Lithium-ion Batteries

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Abstract— The constant polarization voltage is managed for battery heating to achieve a good tradeoff between short heating time and less damage to battery lifetime based on an electro-thermal coupled model. The optimal frequency for maximum heat generation rate at a certain temperature is determined, which is different from the frequency for minimum total impedance. Heating under variable frequency is almost the same as under a constant frequency in terms of heating time and efficiency. However, engineering realization for variable frequency is more difficult, implying that constant frequency heating is a more promising candidate. The optimal frequency during the overall heating process, which is always lower than that at the initial temperature, can be evaluated from the intermediate temperature with low computational effort. Experimental results demonstrate that the heating time at the optimal frequency, corresponding to the maximum heat generation during the overall heating process, is the shortest with high efficiency. The battery is heated from -15.4°C to 5.6°C within 338s, an average temperature-rise rate of 3.73°C/min with an essentially uniform temperature distribution. The proposed heating strategy, which is experimentally verified with no apparent detrimental effect on battery health, is of great potential for rapidly improving operating performance of electric vehicles in cold weather.

Keywords—Lithium-ion battery, low temperature, constant polarization voltage, optimal frequency, internal heating

Nomenclature

$u_o(t)$	Terminal voltage (V)	Z	Total impedance (Ω)
$u_p(t)$	Total polarization voltage (V)	R_e	Real part of the impedance (Ω)
U_{ocv}	Open circuit voltage (V)	I_m	Imaginary part of the impedance (Ω)
U_p	Peak value of total polarization voltage (V)	i	Total current though the battery (A)

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I	Peak value of alternating current (A)	<i>Abbreviations</i>	
Q_I	Irreversible heat during a period (J)	BMS	Battery management system
θ	Phase angle (rad)	BT	Battery temperature
L	Inductance (H)	BTI	Battery temperature interval
R_b	Ohmic resistance (Ω)	CA	Current amplitude
C_p	Polarization capacitance (F)	DT	Desired temperature
R_p	Polarization resistance (Ω)	EV	Electric vehicle
ω	Angular frequency ($\text{rad}\cdot\text{s}^{-1}$)	EW	Electrochemical workstation
R_α	Resistance parameter (Ω)	FD	Frequency-dependent
R_β	Resistance parameter (Ω)	he	Highest efficiency
R_γ	Resistance parameter (Ω)	hp	Heating process
C_α	Capacitance parameter (F)	IC	Incremental capacity
C_β	Capacitance parameter (F)	IT	Intermediate temperature
C_γ	Capacitance parameter (F)	LIB	Lithium-ion battery
ω_o	Optimal angular frequency ($\text{rad}\cdot\text{s}^{-1}$)	NCM	$\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$
η	Heating efficiency (%)	RETC	Reduced electro-thermal coupled
T_a	Ambient temperature ($^\circ\text{C}$)	RIHTs	Repeated internal heating tests
A	Surface area of the battery (m^2)	RQ	Resistance and constant phase element
h	Heat transfer coefficient ($\text{W}\cdot\text{m}^{-2}\cdot\text{K}^{-1}$)	SAC	Sinusoidal alternating current
m	Battery mass (g)	SAPV	Sinusoidal alternating polarization voltage
C'_p	Average specific heat capacity ($\text{J}\cdot\text{g}^{-1}\cdot\text{K}^{-1}$)	SOC	State of charge
T	Battery temperature ($^\circ\text{C}$)	SOH	State of health
Q'_o	Heating energy under variable or constant frequency (J)	VF	Variable frequency

1 Introduction

The dramatically increased resistance of lithium-ion batteries (LIBs) at low temperature [1] not only results in the substantial loss of both pulse power and usable energy [2] but also makes Li-ion easily deposited [3], leading to significant reduction of LIB lifetime. The deteriorated performance at low temperature adversely restricts the feasibility and applicability of electric vehicles (EVs) [4]. Therefore, preheating the LIB to a suitable temperature, to improve the LIB poor performance [5, 6], is an essential function of an effective battery management system (BMS) for EVs operated in cold weather [7].

Internal heating, in which the heat is generated inside the battery itself [8], is more efficient and can achieve better uniformity of the temperature distribution within LIBs than external heating. Either direct current or alternating current can be used for internal heating. Alternating current, in contrast to direct current with restricted amplitude and duration [9], can heat batteries quickly and effectively, avoiding substantial change of the state of charge (SOC) [10].

Understanding the strong mutual coupling between electrical and thermal processes is critical for

developing rapid heating strategies of LIBs at low temperature [11, 12]. There are several possible models for developing effective internal heating strategies. For core heating, the internal heating power is specified as a constant or linear function of time using thermal finite element models [13]. But this model lacks the ability to predict nonlinear behaviors of LIBs. The electrochemical-thermal coupled model, which accurately characterizes the reaction mechanism within LIBs [14], is adopted to study internal heating strategies [15]. However, it contains many unknown variables and so it significantly increases computational complexity. The heat generation model [16] based on the equivalent electrical circuit can predict the temperature-rise in the frequency-domain during internal heating. Nevertheless, the model, including RQ and Warburg elements in the frequency-domain, has difficulty in calculating battery voltage in real time; as a result, the terminal voltage may surpass beyond the safe threshold. The reduced electro-thermal coupled (RETC) model presented in the literature [17], not only precisely predicts temperature-rise within LIBs, but also accurately calculates the terminal voltage of LIBs. Furthermore, it is simple with low computational complexity for development of rapid and safe internal heating strategies.

Several studies have been performed for internal heating. It has been found that the heating process speeds up as the current amplitude increases [9]. Whereas, the detrimental situation, such as overcharging, in which the terminal voltage exceeds the safe threshold, may occur when the current reaches a high value, resulting in irreversible deterioration of LIBs. The polarization voltage, which is associated with the electrochemical reactions, must be taken into account when choosing the current [18], but it is often neglected [13, 15, 16]. It is essential to keep the polarization voltage in a proper range. The constant polarization voltage, preventing the terminal voltage from exceeding the safe threshold [19], is a preferred choice by increasing the current as the temperature rises. The constant polarization voltage not only maintains a high and homogeneous temperature-rise rate instead of the low and decreased temperature-rise rate reported in the literatures [7, 16], but also ensures quick heating of the battery without lifetime reduction.

The sinusoidal alternating current (SAC) heating technology, which is claimed to cause less damage to the battery, is a promising candidate for many applications [7], [13,15,16]. The low SAC frequencies, such as 1Hz [16], 60 Hz [15], as well as high SAC frequencies, like 20kHz [7, 13], are empirically employed in internal heating. However, the question of which frequency is most suitable to heat the battery and whether optimal heating frequency exists remains unanswered. Chen L.R., et al.

[20, 21] explored optimal frequencies during charging and discharging processes, which are the frequencies corresponding to the minimum total impedance by the electrochemical impedance analysis. Nevertheless, the frequency for the minimum total impedance differs from the optimal frequency for SAC heating. Therefore, it is essential to derive the frequency dependence of the heat generation rate for development of rapid internal heating strategies.

In this work, a rapid and effective low-temperature internal heating strategy is proposed with the following salient features. First, the constant SAPV amplitude is determined for heating the battery to achieve a suitable compromise between high and homogeneous temperature-rise rate, and less detrimental impact on battery health. Second, the optimal frequency for the maximum heat generation rate at a certain temperature is determined, which is different from the frequency for the minimum total impedance. Third, optimal frequency for the shortest heating time during the overall heating process, which is always lower than that at the initial heating temperature, can be accurately estimated from the intermediate temperature with a low computational effort. A rapid internal heating strategy with optimal frequency is proposed and experimentally validated. The battery is warmed up from -15.4°C to 5.6°C within 338s, where the average temperature-rise rate is $3.73^{\circ}\text{C}/\text{min}$, and the temperature distribution within the battery remains essentially uniform. Last but not least, there is no battery life reduction after 30 repeated heating tests because the polarization is kept suitable and constant, and the faradaic current approximates to zero with high frequency. The proposed self-heating strategy, thanks to its swiftness and feasibility, is highly promising for improving operating performance of EVs in cold conditions.

The remaining part of this paper is organized as follows. The rapid internal heating strategies are developed in Section 2. The experimental setup is briefly described in Section 3. The simulation and experimental results and discussions about internal heating are presented in detail in Section 4, followed by conclusions summarized in Section 5.

2 Internal Heating Strategies

2.1 Electro-Thermal Coupled Model

There are several approaches for establishing dynamic electro-thermal models to capture the thermal and electrical behavior of LIBs [12, 22-23]. Miranda et al. [24] presents an improved integrated model, which explains how current and temperature affect the thermal and electrical behavior. In order to investigate the optimal frequency for the maximum heat generation rate, the

electro-thermal model [17], which includes the temperature and frequency effects, may be a suitable choice for rapid internal heating. Fig.1 shows the reduced and accurate model, which is proposed in detail in [17]. The values of resistance ($R_p(f,T,S)$) and capacitance ($C_p(f,T,S)$) depend on SAC frequency, which are based on the FD equation as described by Eq.(10) in the appendix. The ohmic resistance (R_b) and the inductance (L) represent the response of LIBs at high frequency. Model parameters are dependent on the battery temperature (T) as expressed by Eq. (11) in the appendix. Model parameters are associated with SOC (S) [25], but this dependence is neglected due to the small change of SOC during internal heating. When the battery is subjected to the current, the heat generated from the electrical process dictates battery temperature evolution. The electrical process is affected by the battery temperature because of the temperature-dependent parameters, which has an impact on the heat generation rate and the temperature-rise rate. There exists strong mutual coupling between electrical and thermal processes during the operation of LIBs. It is essential to employ an electro-thermal coupled model for predicting the electrical and thermal performance of LIBs.

Fig.1. The reduced equivalent electrical circuit (RETC) model.

2.2 Polarization Voltage

The polarization is caused by slow electrochemical reactions, ohmic hindrance, and mass transport limitations [26]. The electrochemical polarization, which is associated with the activation of the charge-transfer process [27], significantly increases and becomes dominant at low temperature, which easily leads to Li plating on the anode surface [28]. The ohmic drop dominates at high frequency while the diffusion polarization dominates at low frequency. In short, the contribution of each process to the total polarization of LIBs is dependent on SAC frequency and environmental condition. The sluggish charge-transfer process should be restricted to prevent Li-ion depositing at low temperature and the SAC with high frequency may be a promising candidate for internal heating.

On the other hand, from the outside of LIBs the total polarization, namely total over-potential, can be given in:

$$u_p(t) = u_o(t) - U_{OCV} \quad (1)$$

Fig.2. The characteristic of polarization voltage and heat generation rate (calculated): (a) under different amplitudes at -15°C , (b) under different frequencies at -15°C . (c) The evolution of battery temperature (calculated) and the real part of impedance during internal heating.

The total polarization voltage and the heat generation rate are small in the high current frequency region while they are both large at low frequency as shown in Fig.2 (a) based on the literature [17]. Fundamentally, the battery, operating at low frequency with heavy current, experiences the high polarization voltage especially under low temperature as shown in Fig.2 (a) and (b), easily leading to battery terminal voltage exceeding the safe voltage threshold [29]. This detrimental situation, which causes an unwanted side reaction, not only makes Li-ion to be deposited on the anode surface [27, 30], but also results in active material loss [18], leading to a substantial reduction of battery life. Consequently, it is essential to keep the polarization voltage in a proper range.

Fundamentally, to rapidly heat the battery at low temperature, the large amplitude of both polarization voltage and current is always desired. However, during heating with constant SAC amplitude, the polarization voltage gradually diminishes as the battery temperature rises. So the temperature-rise rate gradually decreases and the heating time duration extends as shown in Fig.2 (c). In order to keep a high heat generation rate with the rising temperature, the current amplitude should be updated. But the question that how much the current amplitude increases remains unknown. Therefore, it is critical to pre-set the alternating polarization voltage at a constant and high value as a goal. Then the maximum acceptable current can be computed from the given polarization voltage as Eq.(2). As the constant SAPV with suitable amplitude ensures a high and homogeneous temperature-rise rate, a good compromise is achieved between the short heating time and less damage to battery life.

$$I = \frac{U_p}{|Z|} = \frac{U_p}{\sqrt{R_e^2 + I_m^2}} \quad (2)$$

2.3 Optimal Frequency

When the battery is subjected to the SAC excitation, only irreversible heat is taken into account.

The irreversible heat during a SAC period can be governed as:

$$Q_1 = \int_0^{2\pi} u_o(t) i(t) \cos \theta dt = \int_0^{2\pi} (u_p(t) + U_{ocv}) \times i(t) \cos \theta dt = \int_0^{2\pi} u_p(t) i(t) \cos \theta dt \quad (3)$$

The irreversible heat generated from the real part of impedance during a SAC period, namely heat generation rate, can be also expressed as:

$$Q_1 = \frac{U_p}{\sqrt{2}} \frac{I}{\sqrt{2}} \cos \theta = \frac{U_p^2}{2} \times \frac{R_e}{|Z|^2} \quad (4)$$

It indicates that the irreversible heat increases with the increase of the polarization voltage when the impedance is kept the same. On the other hand, the greater the ratio between the real part of impedance and the square of the total impedance is, the greater the heat generation rate is when the polarization voltage remains at the same. So the ratio between the real part of impedance and the square of the total impedance, defined as $g(\omega) = \frac{R_e}{|Z|^2}$, is used in the deduction of the maximum heat generation rate.

In the frequency domain, the total impedance can be given in:

$$Z = \frac{R_p - j\omega R_p^2 C_p}{1 + \omega^2 R_p^2 C_p^2} + R_b + j\omega L \quad (5)$$

Therefore,

$$g(\omega) = \frac{R_b + \frac{R_\gamma + \phi(\omega)}{1 + \omega^2 (R_\gamma + \phi(\omega))^2 (C_\gamma + \varphi(\omega))^2}}{\left[R_b + \frac{R_\gamma + \phi(\omega)}{1 + \omega^2 (R_\gamma + \phi(\omega))^2 (C_\gamma + \varphi(\omega))^2} \right]^2 + \left[L\omega - \frac{\omega (R_\gamma + \phi(\omega))^2 (C_\gamma + \varphi(\omega))}{1 + \omega^2 (R_\gamma + \phi(\omega))^2 (C_\gamma + \varphi(\omega))^2} \right]^2} \quad (6)$$

where, $\phi(\omega) = \frac{2\pi R_\alpha}{2\pi R_\beta + \omega}$, $\varphi(\omega) = \frac{2\pi C_\alpha}{2\pi C_\beta + \omega}$

The maximum value of $g(\omega)$ can be numerically calculated by MATLAB and the optimal frequency for the maximum value is in the high frequency region. In order to reduce the computational effort, it is essential to develop the analytical expression for the optimal frequency.

The derivative of $g(\omega)$, namely $g'(\omega)$, is very complex and hard to calculate. Fundamentally, based on the electric circuit theory [31], if $R_\gamma \geq 5\phi(\omega)$, R_γ is considered to be much greater than $\phi(\omega)$.

When $\omega > 320\pi^1$, it can be said that $R_\gamma \gg \phi(\omega)$ and $C_\gamma \gg \varphi(\omega)$. Thereby, the $g(\omega)$ can be deduced as $g_1(\omega)$.

¹ The angular frequency is dependent on the model parameters and the battery type.

$$g_1(\omega) = \frac{R_b + \frac{R_\gamma}{1 + \omega^2 R_\gamma^2 C_\gamma^2}}{\left(R_b + \frac{R_\gamma}{1 + \omega^2 R_\gamma^2 C_\gamma^2}\right)^2 + \left(L\omega - \frac{\omega R_\gamma^2 C_\gamma}{1 + \omega^2 R_\gamma^2 C_\gamma^2}\right)^2} \quad (7)$$

Solving the equation $\frac{dg_1(\omega)}{d\omega} = 0$, the meaningful root can be expressed as:

$$\omega_0 = \sqrt[2]{\frac{L \times 2\sqrt{R_\gamma^5 \times (R_\gamma + R_b)} - L \times R_\gamma^3 - L \times R_\gamma^2 \times R_b + C_\gamma R_\gamma R_b \times 2\sqrt{R_\gamma^5 \times (R_\gamma + R_b)}}{C_\gamma^2 R_\gamma^4 R_b L}} \quad (8)$$

Substituting model parameters' values in $g'(\omega)$, it can be seen $g'(\omega) > 0$, if $0 < \omega < \omega_0$; while $g'(\omega) < 0$, if $\omega > \omega_0$. Hence, $g(\omega) = \frac{R_e}{|Z|^2}$ is at the maximum value, when the angular frequency of SAC is ω_0 . The optimal angular frequency, namely ω_0 , is obtained for rapid internal heating. In that case, the temperature-rise rate and the heat generation rate are maximal.

2.4 Internal Heating Strategies

Since the kinetic and transport processes are highly temperature-dependent, the model parameters vary so significantly that the optimal frequency alters with the rising temperature during internal heating. In order to heat the battery quickly, the heating frequency should be changed to track the optimal frequency, achieving the maximum heat generation rate at any temperature.

On the other hand, the SAC frequency variation under different temperatures during internal heating places an extremely high requirement on the performance of power electronic device compared to constant SAC frequency. Thus, it is essential to analyze the total heat generation and heating time during internal heating under constant SAC frequency.

The heating efficiency as Eq. (9) is introduced to evaluate the heating strategies. It can be expressed as the ratio between the energy for battery temperature-rise and the total active energy of electrical work. The heating time and efficiency are used to assess the heating strategies. Understanding the theoretical characteristic contributes to manufacturers select, implement and optimize viable internally self-heating strategies.

$$\eta = \frac{\int mC'_p dT}{Q_o} = \frac{\int mC'_p dT}{\int mC'_p dT + \int hA(T - T_a)dt} \quad (9)$$

3 Experimental

The tested batteries with NCM cathode material and the experimental platform are the same as that presented in the literature [17]. After soaked at -15.4°C for more than 5h, the tested battery is

internally heated from -15.4°C using the increasing SAC with various sets of frequencies as shown in Table 1. During the process the SAPV amplitude is kept constant. The heating process ends when battery temperature reaches above 5.6°C . The battery temperature is measured by five T-type thermocouples, the average of which is used as the LIB experimental temperature. In order to investigate the influence of the rapid internal heating on battery health, the battery is repeatedly heated with SAC of 1377Hz and 0.5V for 30 times. Before the repeated heating, the tested battery is soaked for 5h under 25°C and then it is charged and discharged with different current-rates as shown in Fig.3. The cut-off voltages of charging and discharging are 4.2V and 2.5V, respectively, while the cut-off current during the constant voltage charging is $1/20C$. In between the charging period and discharging period, the battery rests for an hour. After the battery has been heated 30 times, the tested battery is soaked for 5h under 25°C , and then charged and discharged with the same method as shown in Fig.3.

Table 1. Experimental details.

Test	Item	Value
	Initial temperature	-15.4°C
Preheating tests	Voltage parameters	0.5V 500Hz; 0.5V 1200Hz; 0.5V 1377Hz; 0.5V 1567Hz; 0.5V 1743Hz; 0.5V 3kHz
Other tests	Performance tests	1/20C, 1/3C, 1/2C charge and discharge
	Life tests	0.5V 1377Hz repeated heating


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graph LR
    Start([Start]) --> D1[1/3C Discharge]
    D1 --> R1[Rest]
    R1 --> C1[1/3C CCCV Charge]
    C1 --> R2[Rest]
    R2 --> D2{≥ 3 Times}
    D2 -- Yes --> D3[1/3C Discharge]
    D2 -- No --> D1
    D3 --> R3[Rest]
    R3 --> C2[1/2C CCCV Charge]
    C2 --> R4[Rest]
    R4 --> D4[1/2C Discharge]
    D4 --> R5[Rest]
    R5 --> D5[1/20C Discharge]
    D5 --> R6[Rest]
    R6 --> C3[1/20C Charge]
    C3 --> R7[Rest]
    R7 --> D6[1/20C Discharge]
    D6 --> R8[Rest]
    R8 --> End([End])
  
```

Fig.3. The charging and discharging methods before and after the 30 RIHTs

4 Results and Discussions

4.1 Analysis of Internal Heating Strategies

4.1.1 Optimal Frequency at A Certain Temperature

The optimal frequency (f_o) for the maximum heat generation rate can be calculated by Eq.(8) and it turns out to be 1567Hz at -15.4°C. The heat generation rate of LIBs increases with the growing frequency until 1567Hz and decreases thereafter in high frequency region as shown in Fig. 4. The frequency for the minimum total impedance is 1743Hz, which is higher than the optimal frequency (f_o), implying that the optimal frequency (f_o) is not equal to the frequency for the minimum total impedance.

Fig.4. The evolution of the calculated heat generation rate and the total impedance with SAC frequency

The heat generation rate is proportional to the square of the SAPV amplitude as shown in Fig. 4. Therefore, in order to shorten heating time, we should choose the optimal frequency and high polarization voltage. However, the polarization voltage must be restricted to a proper range in case the battery overcharges or over-discharges, causing dramatic battery life reduction. For heating the battery with a good compromise between short heating time and less damage to battery life, the SAPV amplitude is selected as 0.5V in this paper.

4.1.2 Internal Heating with Variable Frequency

The model parameters are different and the heat generation rate changes at various temperatures as shown in Fig. 5. The heat generation rate in the low frequency region increases with the rising temperature. The heat generation rate at very high frequency almost keeps the same because the inductance is independent of temperature [17, 32]. The optimal frequency (f_o) also changes with the battery temperature as shown in Table 2. The maximum heat generation rate increases while the optimal frequency decreases when the battery temperature reduces, owing to the decrease of battery impedance. In order to shorten heating time, the heating frequency should be changed to track the optimal frequency (f_o) with the rising temperature, achieving the maximum heat generation rate at any temperature.

Fig.5. The evolution of the calculated heat generation rate with SAC frequency.

Table 2. Optimal frequency and heat generation rate at different temperatures.

Temperature(°C)	-15	-10	-5	0	5
f_o (Hz)	1559	1459	1377	1299	1233
Heat generation rate (W)	4.12	4.30	4.47	4.64	4.81

In Fig. 6, battery temperature interval (BTI) indicates the value of the temperature increases in °C before the optimal frequency is reduced to a lower value. Battery temperature rises from -15.4°C until to 5.6°C and the optimal frequency correspondingly changes under different BTIs. Heating time increases while heating efficiency reduces, when the BTI goes up as shown in Table 3. For example, the optimal frequency is reduced from 1567Hz to 1228Hz with the BTI of 1°C. Battery temperature rises linearly and the average temperature-rise rate is 3.53°C/min, where the heating efficiency is 82.64%.

Fig.6. The evolution of the calculated temperature-rise and frequency with different BTIs.

Table 3. The results with different BTIs.

BTI(°C)	5	3	1	0.5
Efficiency(%)	82.6344	82.6353	82.6357	82.6358
Time(s)	357.95	357.93	357.92	357.91

4.1.3 Internal Heating with Constant Frequency

It is concluded that the optimal frequency ($f_{o,hp}$) during the internal heating process changes with

the desired temperature (DT) if constant frequency heating is adopted. Table 4 shows that the optimal frequency ($f_{o,hp}$) decreases with the increasing DT, when the initial temperature is kept the same as -15.4°C . As the DT is changed, the optimal frequency ($f_{o,hp}$) needs to be calculated repeatedly. The computation is complicated and time-consuming. However, it may be possible that the optimal frequency (f_o) at a certain temperature is close to the optimal frequency ($f_{o,hp}$). The intermediate temperature (IT) is between the initial temperature and DT. The optimal frequency ($f_{o,IT}$) at the intermediate temperature is almost the same as the optimal frequency ($f_{o,hp}$) during the internal heating process as shown in Table 4. Therefore, it is very simple and convenient that the optimal frequency ($f_{o,hp}$) is evaluated from the optimal frequency ($f_{o,IT}$) instead of complicated calculation during the overall process when heating with constant frequency is adopted.

Table 4. The optimal frequency with different heating DTs.

DT ($^{\circ}\text{C}$)	-5	0	5.6	10	15	20
$f_{o,hp}$ (Hz)	1465	1422	1369	1331	1296	1262
IT ($^{\circ}\text{C}$)	-10.2	-7.7	-4.9	-2.7	-0.2	2.3
$f_{o,IT}$ (Hz)	1467	1423	1377	1341	1305	1271

When the battery needs to be heated from -15.4°C to 5.6°C , the heating efficiency and heating time at various SAC frequencies is different as illustrated in Fig. 7. It indicates that the low heating efficiency and long heating time is observed under low frequency and very high frequency. It is included that the frequency for the shortest time is 1369Hz and the frequency ($f_{o,he}$) for the highest efficiency is 1325Hz.

Fig.7. The evolution (calculated) of heating efficiency and heating time with SAC frequency

4.1.4 Comparison about Internal Heating Strategies

When the battery needs to be heated from -15.4°C to 5.6°C under our thermal insulation condition, the heating strategies with variable frequency (VF) and different constant frequencies, as discussed above and shown in Table 5, are compared in terms of heating time and heating efficiency as shown in Fig.8. The heating time is the longest and the heating efficiency is the lowest under the

SAC with the frequency for minimum total impedance, which is not suitable for battery heating. The frequency during the VF heating is changed for every degree increase of the battery temperature. It is concluded that the heating time is the shortest under the VF condition. The heating time is the shortest at 1377Hz among different constant frequency conditions. The heating time at constant frequency of 1377Hz is 0.16s less while the heating efficiency is 0.4% lower than that under the VF condition, implying that the constant frequency heating can almost achieve the same target as measured by heating time and heating efficiency. However, taking engineering implementation into account, the internal heating strategy with VF is more difficult and requires a higher computational effort. Thereby, the heating strategy with constant frequency may be a preferred choice.

Fig.8. Comparison of internal heating strategies (from calculation) in terms of heating time and efficiency.

Table 5. The optimal frequency for different goals.

Optimal frequency	Frequency(Hz)
Maximum heating efficiency	1325
Minimum heating time (intermediate temperature)	1377
Initial temperature (15.4°C)	1567
Minimum total impedance	1743

On the other hand, the small difference between the VF and the constant frequency condition indicates that the heating time and heating efficiency is almost the same in the frequency range from 1567Hz to 1228Hz. Although not all cells in a battery pack are the same, for instance inconsistent cell impedance between different cells, it is not difficult to obtain the optimal frequency for the overall battery pack.

The heating efficiency and heating time with 1325Hz is almost the same as that with 1377Hz. However, the frequency of 1325Hz is obtained from the overall heating process and it is time-consuming. So the frequency of 1377Hz may be better with its uncomplicated characteristic because it can be evaluated from the intermediate temperature.

4.2 Verification for Internal Heating Strategies

4.2.1 Constant Frequency and Variable Amplitude of Alternating Current

In order to keep the SAPV amplitude constant, the battery can be subjected to the SAC with variable amplitude when the battery temperature rises. Taking the same SAPV as a goal, where the U_p is fixed at 0.5V, an acceptable alternating current is developed by Eq.(2). The initial LIB voltage is 3.672 V, corresponding to 50% SOC, and the terminal voltage is between 3.172V and 4.172V during internal heating.

First of all, when the battery is soaked at -15.4°C for more than 5h, the SAC amplitude can be calculated, which turns out to be 16.40A, and the constant optimal frequency is 1377Hz. The battery subjected to the SAC with the calculated amplitude and frequency is warmed up and the LIB temperature gradually rises. When the LIB temperature reaches -14.4°C , corresponding to 1°C temperature-rise, the SAC amplitude is calculated again, which turns out to be 16.54A. Next, the battery is heated with the SAC amplitude of 16.54A. Then, as long as the LIB temperature rises 1°C , the SAC amplitude is updated again by Eq.(2). The whole process continues until the LIB temperature reaches 5.6°C .

Fig.9 shows that the battery is heated from -15.4°C to 5.6°C within 360s. The LIB temperature is almost a straight line up and the average slope is about $3.51^{\circ}\text{C}/\text{min}$. The terminal voltage is kept between 3.172V and 4.172V during the overall heating process. The SAC amplitude increases in a stepwise fashion, because the resistance gradually decreases with the increase of the LIB temperature. In fact, in the process of constant SAC amplitude, when the LIB temperature gradually goes up, the resistance decreases step by step, leading to the terminal voltage diminishing below 4.172V as shown in Fig.9 (b). When the LIB temperature rises by 1°C , the amplitude is updated and the terminal voltage achieves 4.172V. Hence, if the amplitude increases when the LIB temperature changes by 0.5°C , the time duration that the polarization voltage is below 0.5V will reduce and the temperature-rise rate accordingly increases.

Fig.9. Heating simulation results with constant frequency and variable amplitude of SAC (a) temperature and amplitude evolution (b) voltage and current evolution.

The SAC amplitude increases in a stepwise fashion at different speed with various BTIs as shown in Fig.10, where CA and BT represent current amplitude and battery temperature respectively. The greater the BTI is, the longer the heating time is. The time duration of constant SAC amplitude is gradually reduced with the decrease of BTI. The SAC amplitude change is not step-by-step but almost a straight line up when BTI is 0.05°C . In that case, all polarization voltage is nearly 0.5V and the average temperature-rise rate is $3.53^{\circ}\text{C}/\text{min}$, which is larger than that under other BTIs.

Fig.10. Temperature and current amplitude evolution calculated with different BTIs.

Taking the constant SAPV amplitude as a goal, the acceptable alternating current is developed and updated for a high heat generation rate and a homogeneous temperature-rise rate. The SAC

amplitude is calculated each time when BTI is changed by $n^{\circ}\text{C}$. The smaller the n is, the less the time is for the polarization voltage to be below 0.5V, which means that the heat generation rate correspondingly increases. When n approaches to zero, the SAC amplitude is calculated in real-time with the decrease of the LIB impedance, implying that the SAC amplitude continuously increases and the SAPV amplitude is completely kept constant.

4.2.2 Constant Frequency and Constant Amplitude of Alternating Voltage

In order to ensure the constant SAPV amplitude, the output voltage is directly controlled in the heating control circuit as shown in Fig.11, in which EW represents an electrochemical workstation carrying out SAC excitation, while PP241 is an external potentiostat for amplification of the current. The DC voltage is set as 3.672V, which is the same as U_{ocv} , and the disturbance voltage with the peak amplitude of 0.5V and the frequency of 1377Hz is generated in the EW. As the temperature is increased, the SAC amplitude is also continuously increased to ensure the constant output alternating voltage. Moreover, by using the internal heating strategy under an alternating voltage with constant frequency and constant amplitude, the battery temperature is not needed as feedback. It is convenient that only the electrical measurements, namely voltage and current, are employed as feedback.

Fig.11. Schematic view of heating for keeping the SAPV amplitude constant.

The comparative analysis of voltage error shows that simulation results match well with experimental data under -15°C as shown in Fig.12, where the mean error and the maximum relative error are 29.23mV and 2.19%, respectively. The increase of error may result from the high SAC amplitude and the distortion of SAC profiles. But the error is small enough to meet the requirement for application in EVs. Although there are some spikes, which may be from the error of the controller or the recording error of voltage data, the peak voltage is less than 4.2V. Table 6 exhibits the error at different temperatures during heating. Obviously, the higher the LIB temperature is, the higher the precision of voltage prediction is, indicating that the RETC model has a high accuracy. The experimental and simulation results demonstrate that the LIB terminal voltage is controlled well to guarantee the constant SAPV amplitude, preventing battery aging and safety issues. At the same time,

the SAC amplitude continuously increases to achieve high heat generation rate and maintain homogeneous temperature-rise rate.

Fig.12. Voltage evolution and error distribution during internal heating at 1377Hz

Table 6. Simulation errors of the RETC model with the SAC excitation

Temperature (°C)	-15	-8	-1	5
$ \epsilon _{\text{avg}}(\text{mV})$	29.2	22.9	17.9	15.2
$ \epsilon _{\text{max}}(\%)$	2.19	2.08	1.98	1.80

The proposed internal heating strategy, keeping the SAPV amplitude constant with the optimal frequency, is used to rapidly warm the LIB up to a proper temperature. Fig. 13 shows good agreement between model predictions and experimental data with the maximum absolute error of 1.64°C. Experimental results indicate that the battery is heated from -15.4°C to 5.6°C within 338s. The LIB temperature as a function of time is almost a straight line up because of the growing SAC amplitude, where the average temperature-rise rate is 3.73°C/min. The heating efficiency in the experiment reaches 82.81% and the total heating energy is 0.4435Wh. Simulation results show that the average temperature-rise rate is 3.54°C/min and is smaller. This may result from the voltage error of the model prediction.

Fig.13. Temperature evolution and error distribution during internal heating.

During internal heating the BT was measured using five T-type thermocouples along the axis direction of the battery as shown in Fig.14. The maximum temperature variation among the five thermocouples is below 1.6°C, whereas the battery temperature-rise is above 20°C, which reinforces

the assumption of the lumped thermal model within the RETC model. The small difference of the BT confirms that the proposed heating strategy can evenly heat the battery to a suitable temperature without high temperature gradient.

Fig.14. Temperature distribution at five separate locations during internal heating experiment.

The validation of internal heating at other frequencies is carried out and the outcome is illustrated in Fig.15. The simulation temperatures show good agreement with the experimental measurements, where the maximum absolute error is below 1.72°C at various frequencies. Table 7 shows the temperature-rise rate, heating efficiency and heating energy at different frequencies. The heating efficiency is the highest and heating energy is the minimum at 1377Hz. The same trends between the estimated and measured temperature at different frequencies validate that the most effective frequency is 1377Hz, the heating time of which is the shortest.

Fig.15. (a) Temperature evolution (b) Error distribution at different frequencies

Table 7. The results of the heating experiments at different frequencies.

Frequency (Hz)	500	1200	1377	1567	1743	3000
Temperature-rise rate (°C/min)	3.48	3.72	3.73	3.61	3.52	3.39
Efficiency (%)	82.02	82.73	82.81	82.32	81.97	81.12
Energy (Wh)	0.4532	0.4440	0.4435	0.4467	0.4494	0.4524

If a low frequency or very high frequency is selected, the time duration needed to heat battery for the same DT will extend and the average temperature-rise rate will increase. The results of experimental validation demonstrate that the average heat generation rate with SAC at the optimal frequency (1377Hz) is maximum under the constant SAPV amplitude.

4.3 The Concern on Battery Health for Internal Heating

Although the possible lifetime reduction of LIBs is taken into consideration when determining SAC parameters, the possibility of life degradation also needs to be experimentally tested. Both the amplitude and the frequency of SAC may result in life reduction [33, 34]. During the SAC heating process, a maximum current about 7C is recorded because of the low resistance at the optimal frequency. Fig.16 shows incremental capacity (IC) curves at 25°C before and after 30 repeated internal heating tests (RIHTs). Each IC peak has a unique shape, intensity and position [35], which stands for electrochemical processes within the battery. Any variation in shape, intensity and position of an IC peak represents a symptom of battery aging [36]. However, all the peaks are almost the same, implying that no obvious life reduction is observed. Moreover, the discharge capacity at different current rates after 30 RIHTs increases to some degree as shown in Table 8. This is because that the SAC with high frequency improves the active material utilization giving the battery higher discharge capacity [37]. On the other hand, the decrease rate of battery capacity from the first 30 pulse charging-discharging cycles is the same as that during all the pulse charging-discharging cycles as presented in [37]. It is concluded that the aging rate of LIBs with the proposed heating strategy is less

than that presented in [37, 38]. Therefore, the experimental results and the comparison with the existing literatures indicate that there is no obvious detrimental effect on the state of health (SOH) of LIBs after the proposed internal heating.

The life reduction at low temperature mainly results from Li plating on the anode surface instead of intercalation [28]. The SAC with high frequency promotes the intense suppression of the charge-transfer process [39]. It makes the electrochemical polarization and diffusion polarization approximate to zero so that it has little possibility for Li depositing on the anode surface. On the other hand, the fundamental factor that determines Li deposition is the amplitude of the faradaic charging current [34]. The high current recorded is only instantaneous and the larger part of the current is leaked through the non-faradaic path via the electrical double layer because of the high frequency. Much of the polarization voltage is caused by the ohmic drop, which does not cause Li plating. Therefore, based on the suitable and constant polarization voltage, there is no Li plating and no detrimental effect on the SOH of LIBs when the SAC heating with the optimal frequency is adopted.

Fig.16. IC curves during discharging with different current rates in the experiment.

Table 8. The discharge capacity with different current rates.

Current rates	1/20C	1/3C	1/2C
Before RIHTs(Ah)	2.927	2.749	2.710
After RIHTs (Ah)	2.932	2.765	2.727

5 Conclusions

A rapid and effective internal heating strategy with the optimal frequency is proposed based on constant SAPV amplitude when the RETC model is applied, which has the following salient features:

(a) *A good tradeoff.* To achieve high and homogeneous temperature-rise rate, the SAC amplitude is gradually increased to keep the SAPV amplitude constant, when the temperature rises. The suitable and constant SAPV is a preferred choice for effectively heating the battery with a good balance between the short heating time and less damage to battery lifetime.

(b) *Optimal frequency.* The optimal frequency for maximum heat generation rate at -15.4°C is obtained as 1567Hz, which is lower than the frequency for the minimum total impedance. Taking heating time, heating efficiency, and engineering realization into account, heating under a constant frequency is more promising than under variable frequency. The optimal frequency for the shortest heating time during the overall heating process is always lower than that at the initial temperature. It depends on the desired heating temperature and it approximates that at the intermediate temperature. Therefore, it is convenient and simple that the optimal frequency is estimated from the intermediate temperature with low computational effort.

(c) *Heating rapidly and effectively.* The experimental battery, which is subjected to the SAC with the optimal frequency of 1377Hz, is heated from -15.4°C to 5.6°C within 338s. The temperature distribution within the battery remains essentially uniform. The well matched experimental and simulation results confirm that the average temperature-rise rate at the optimal frequency is maximum and reaches $3.73^{\circ}\text{C}/\text{min}$.

(d) *No detrimental effect on SOH.* Neither apparent IC peak variation nor noticeable capacity loss is found after 30 RIHTs owing to the constant SAVP amplitude and the nearly zero faradaic current with high frequency. The proposed heating strategy with suitably determined parameters is demonstrated to have no detrimental effect on SOH of LIBs, providing considerable potential for improving operating performance of EVs in cold weather.

Further study will be conducted on how to determine the amplitude of the additional direct current component of the heating current so that the heat generation rate at high and low SOC is higher while minimizing the risk of life degradation. Moreover, the method and the power electronic equipment to generate the SAC in practical conditions for battery packs are being studied.

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Appendix

The frequency-dependent (FD) equation in the RETC model is expressed as:

$$Z_p = \frac{Z_\alpha}{Z_\beta + f} + Z_\gamma \quad (10)$$

where, Z_p represents resistance ($R_p(f)$) or capacitance ($C_p(f)$).

The model parameters in the RETC model are temperature-dependent, which can be written as:

$$\ln Z_x(T) = \lambda \times \frac{1000}{T} + \kappa \quad (11)$$

where, Z_x represents the parameters ($R_\alpha, R_\beta, R_\gamma$) of resistance ($R_p(f)$), the parameters ($C_\alpha, C_\beta, C_\gamma$) of capacitance ($C_p(f)$), or the ohmic resistance (R_b).

Fitting results of electrical parameters in the RETC model are shown in Table 9.

Table 9. Fitting results of electrical parameters in the RETC model

	$R_\alpha(\Omega)$	$C_\alpha(F)$	$R_\beta(\Omega)$	$C_\beta(F)$	$R_\gamma(\Omega)$	$C_\gamma(F)$	$R_b(m\Omega)$	$L(\mu H)$
λ	-6.26	6.38	23.05	2.24	-15.87	5.80	0.90	0
κ	1.58	-1.64	-5.58	-0.45	3.16	-2.38	0.63	0.447

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