Supplementary Materials

A square wave alternating current preheating with high applicability and effectiveness of preventing lithium plating

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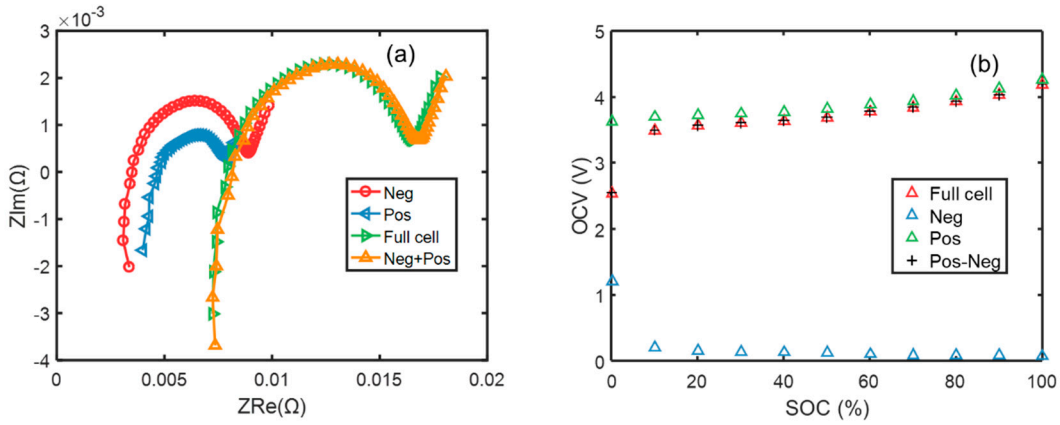
S1. The effectiveness of the reference electrode potential.

To validate the effectiveness of the reference electrode, the EIS of each electrode at 50% SOC were measured at 25 ℃. The OCV of each electrode at every 10% SOC was measured after the cell was kept for 3 hours. The results are presented in Fig. S1. As for the EIS results, the sum of positive electrode impedance and negative electrode impedance agrees well with the measured full cell impedance. Regarding the OCV change, the OCV difference between the positive electrode relative to the reference electrode and the negative electrode relative to the reference electrode coincides well with the measured OCV of the full cell. Such results well validate the effectiveness of the three-electrode lithium-ion cell.



Supplementary Figure S1. The validation of the effectiveness of the reference electrode. (a) The EIS of the positive electrode, the EIS of the negative electrode and the EIS of the full cell at 50% SOC; (b) The OCV change of each electrode at different SOC.

S2. Calibration of the equivalent heat transfer coefficient

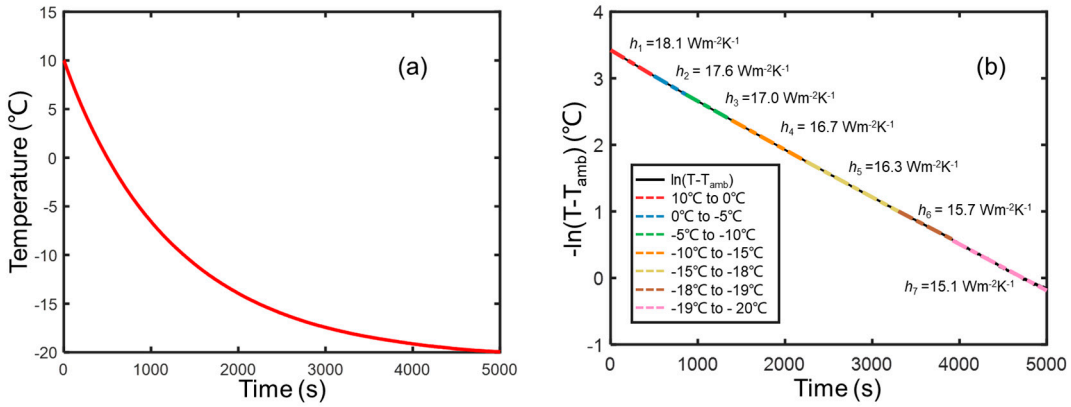
The equivalent heat transfer coefficient is calibrated using the cooling curve of the cell with the energy conversation equation,

$$mc\frac{dT}{dt} = -hS(T - T_{amb}) \qquad \text{Eq. (S.1)}$$

If the h is a constant value, Eq. (S.1) has a solution as

$$\ln(T - T_{amb}) = -\frac{hS}{mc}t + const \qquad \text{Eq. (S.2)}$$

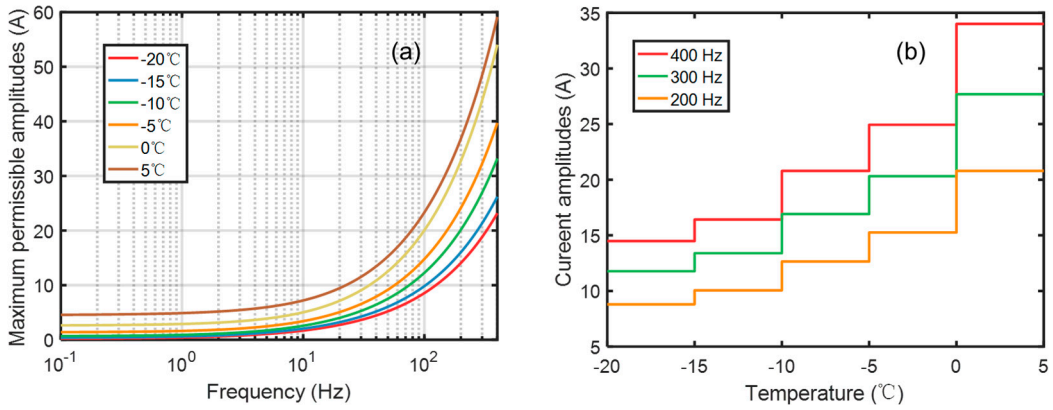
Fig. S2(a) depicts the cooling curves in the experimental thermal insulation condition. Several segmentations of the equivalent heat transfer coefficient h are divided into different temperature regions. The equivalent heat transfer coefficient h is calibrated with the first-order linear fitting function in MATLAB. The values of h at different temperature regions are displayed in Fig. S2(b).



Supplementary Figure S2. The cell temperature T, ln(T−T_{amb}) and the calibrated equivalent heat transfer coefficient h with time during cooling: a) Cell temperature T; b) The value of ln(T−T_{amb}) and the calibrated equivalent heat transfer coefficient h.

S3. Preheating method based on the full cell impedance control strategy

Based on the full cell impedance control strategy, the negative electrode impedance is substituted by the full cell impedance in the scenario of using SWAC, so as to acquire the operation boundaries for preventing lithium deposition. Utilizing EEC parameters of the full cell, as shown in Fig. 4 in the main text of the manuscript, the operation boundaries for preventing lithium deposition, in term of frequency and maximum permissible current amplitude I_{ac}^{Full}, are calculated by Eq. (19) in the main text of the manuscript. The results are presented in Fig. S3(a). According to the maximum permissible current amplitudes on the operation boundaries, the temperature-adaptive procedure for the method based on full cell impedance control strategy is formulated at the frequency of 200 Hz, 300 Hz, 400 Hz. The temperature interval is 5 ℃. The results are presented in Fig. S3(b).



Supplementary Figure S3. (a) Operation boundaries for preventing lithium deposition calculated using the full cell impedance; (b) The temperature-

adaptive procedure for the method based on the full cell impedance control strategy, the frequency is 200 Hz, 300 Hz and 400 Hz, the temperature interval is 5 ℃.

S4. Preheating method based on the terminal voltage strategy

Based on the EEC model, the terminal voltage V excited by the SWAC with 0 phase angle can be calculated by,

$$V(t)=\frac{4}{\pi}\left|\sum_{n=1}^{128}U_n^{\text{full}}\right| \quad \text{Eq. (S.3)}$$

where $U_n^{\text{full}}=\frac{I_{\text{SWAC}}}{2n-1}\sin((2n-1)\omega t+\varphi_n^{\text{full}})\cdot\left(Z_{\text{Re}}^{\text{neg}}((2n-1)\omega)+Z_{\text{Re}}^{\text{pos}}((2n-1)\omega)\right)$,

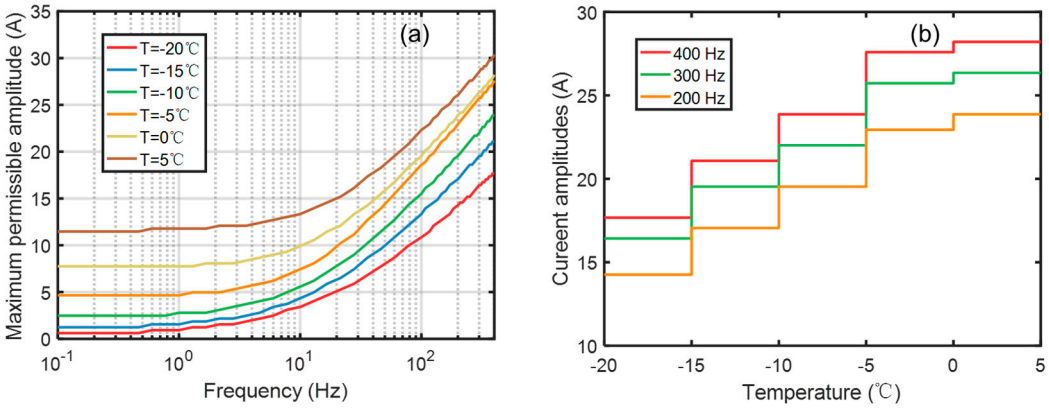
$\varphi_n^{\text{full}}=\arctan(\frac{\text{Re}(Z_{\text{Re}}^{\text{neg}}((2n-1)\omega)+Z_{\text{Re}}^{\text{pos}}((2n-1)\omega))}{\text{Im}(Z_{\text{Re}}^{\text{neg}}((2n-1)\omega)+Z_{\text{Re}}^{\text{pos}}((2n-1)\omega))})$; $Z_{\text{Re}}^{\text{neg}}, Z_{\text{Re}}^{\text{pos}}$ are calculated using Eq. (6) in the main text of the manuscript; n = (1, 2, ⋯128).

Based on the terminal voltage control strategy, the terminal voltage in preheating process should be within working voltage range. Therefore, the maximum permissible amplitude I_{ac}^{Terminal} can be calculated by,

$$\begin{cases}\min(V(t))\geq U_{\text{lower\_limit}}\\ \max(V(t))\leq U_{\text{upper\_limit}}\end{cases} \quad \text{Eq. (S.4)}$$

Utilizing the EEC parameters of the negative electrode and the positive electrode, as shown in Fig. 4 in the main text of the manuscript, the operation boundaries for preventing lithium deposition, in term of frequency and maximum permissible current I_{ac}^{Terminal}, are calculated using Eq. (S3) and Eq. (S4). The results are presented in Fig. S4(a). According to the maximum permissible

current amplitudes on operation boundaries for preventing lithium deposition, the temperature-adaptive procedure for the method based on terminal voltage control strategy is formulated at the frequency of 200 Hz, 300 Hz, 400 Hz. The temperature interval is 5 ℃. The results are presented in Fig. S4(b).



Supplementary Figure S4. (a) Operation boundaries for preventing lithium deposition based on the terminal voltage control; (b) The temperature-adaptive procedure for the method based on the terminal voltage control strategy, the frequency is 200 Hz, 300 Hz and 400 Hz, the temperature interval is 5 ℃.