Supplementary Information

Thermal Management Challenges in Lithium-Ion Batteries: Understanding Heat Generation Mechanisms

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Experimental

Cylindrical 18650 Lithium-Ion Battery Cells

This study examines cylindrical commercial 18650 Panasonic lithium-ion cells with a nominal capacity of 3.2 Ah. These cells utilize nickel cobalt aluminum (NCA) as the positive active material and graphite as the negative active material. The 18650 cell is cylindrical, measuring 18 mm in diameter and 65.0 mm in length. The specifications and characteristics of these cells, as outlined in their technical data sheets, are summarized in Tab.S1.

Tab.S1 Characteristic properties of the examined lithium-ion cells

Cell	**Type A**
Manufacture	Panasonic
Type	NCR18650B
Nominal Capacity (mAh)	3.35
Nominal Voltage (V)	3.7
Charging Voltage (V)	4.2
Discharging End Voltage (V)	3
Anode material	Graphite
Cathode material	NCA

Heat Generation Measurement

Heat generation during these cells' charging and discharging cycles is quantified using an isothermal calorimeter (TAM IV micro-calorimeter system, TA Instruments). The battery cell is placed in the sample holder on the calorimeter's sample side, which is connected to an external Arbin Instrument battery cycler (model BT-2000) via two pairs of wires for the charging and discharging processes. Heat loss through the cables is minimal, contributing to no more than 1% of the total heat generation. A dummy cell is situated on the reference side of the calorimeter. The isothermal calorimetry technique adiabatically isolates the battery from the environment, allowing for precise measurement of total heat generation while ensuring a constant temperature throughout the charge and discharge cycles [8].

Electrochemical measurements are conducted using the Arbin Instrument battery cycler. Cells are charged following a constant current/constant-voltage (CC-CV) protocol to a maximum voltage of 4.2V, and the voltage is held at this level until the current drops to 0.01C before being discharged at the same rate down to 3V.

Electrochemical Impedance Spectroscopy (EIS) Measurement

EIS measurements are conducted utilizing the Solartron Cell Test System (model 1470E) in conjunction with the VTL 4003 Votsch Temperature Test System. Initially, a current corresponding to 0.05C is applied for 1 hour to achieve a specific state of charge or discharge. Following this, the cell can rest for 3 hours to stabilize the voltage at a steady state. The EIS measurements are then performed under these steady-state conditions. The frequency range for the EIS is set from 5 MHz to 100 mHz, employing a sinusoidal voltage amplitude of 10 mV. The resulting EIS spectra are analyzed and fitted using an equivalent circuit model through ZView software.

EIS measurements are conducted at every 5% increment of SOC and DOD to capture the dynamic behavior of the commercial 18650 lithium-ion battery cells. This process follows a relaxation period to ensure that the cell voltage stabilizes at steady-state conditions, which allows for accurate measurement of the impedance characteristics. The measurements are performed at two different temperatures (20°C and 40°C) to evaluate the temperature-dependent effects on the cells' electrochemical performance and resistance behavior. The resulting impedance spectra are shown in Fig. S1.

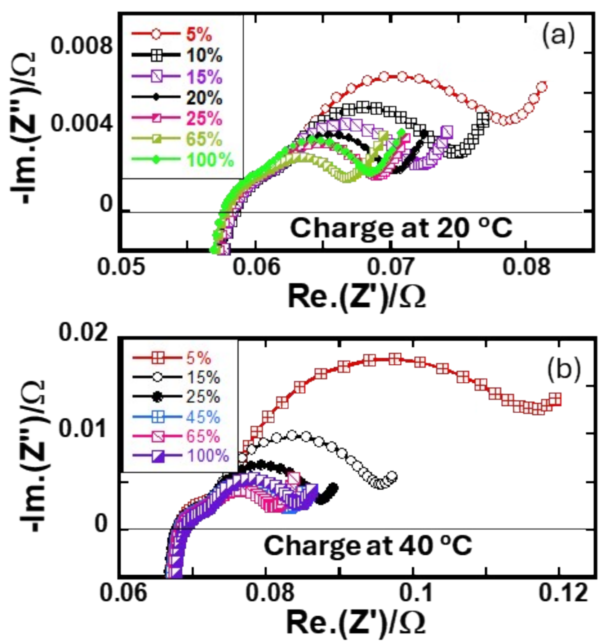


Fig. S1 Impedance spectra of commercial 18650 lithium-ion battery cells measured at (a) 20°C and (b) 40°C during charge.

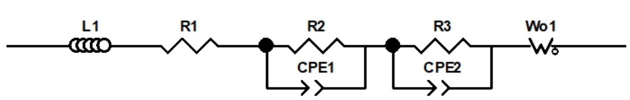


Fig. S2 Equivalent circuit model used to separate the individual resistance components of the commercial 18650 lithium-ion battery cells.

The equivalent circuit model employed to separate the individual resistance components is represented through the measured Nyquist plots, which reveal several distinct features:

L1: This component signifies the circuit's inductor, reflecting the battery's inductive behavior at higher frequencies. It indicates how the battery responds to changes in current, particularly in high-frequency scenarios.

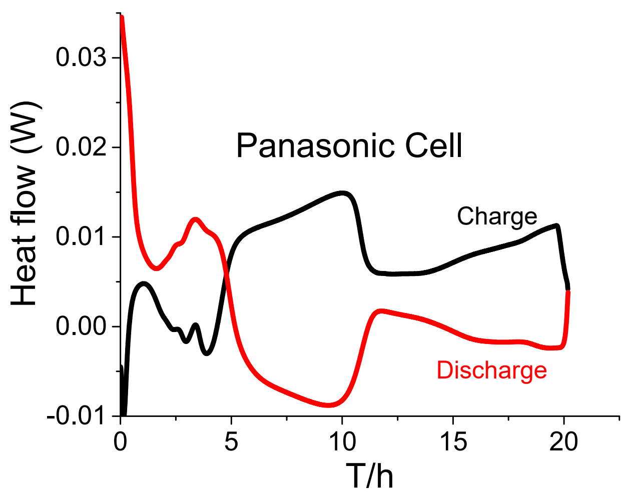
R1: The high-frequency intercept on the Nyquist plot corresponds to the ionic resistance of the electrolyte solution. This value also encompasses a minor contribution from the solid-electrolyte interphase (SEI) resistance, both of which play a crucial role in determining the overall performance and efficiency of the battery.

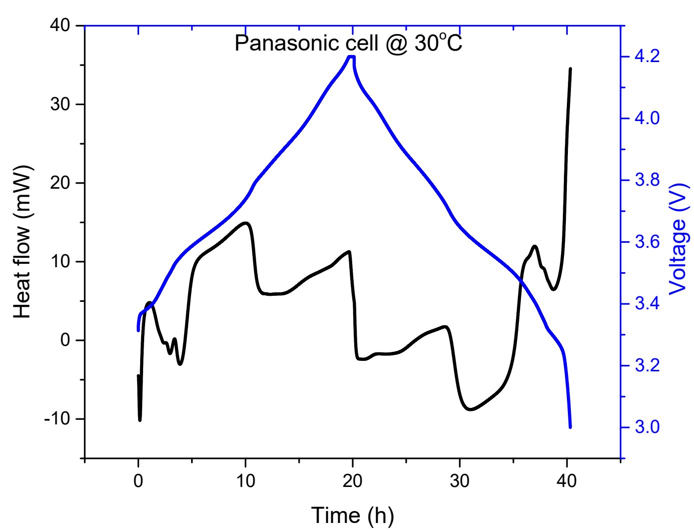
R2: The first semicircle observed at medium to high frequencies is attributed to the charge transfer resistance at the anode/electrolyte interface. This component offers insight into the effectiveness of charge carrier movement across this critical interface during battery operation, directly impacting power delivery and efficiency.

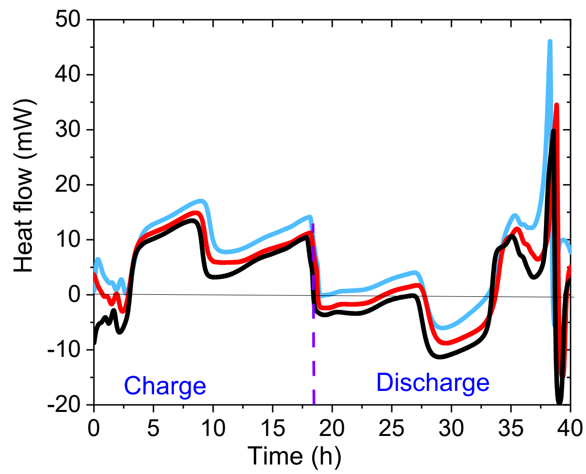
R3: The second semicircle at medium to low frequencies indicates the charge transfer reaction at the cathode/electrolyte interface. Notably, this semicircle is characterized by a higher associated capacitance value (10^{-5} F) compared to the first semicircle (10^{-9} F), suggesting a more pronounced capacitive effect in this region, which can influence the overall energy storage capabilities of the battery.

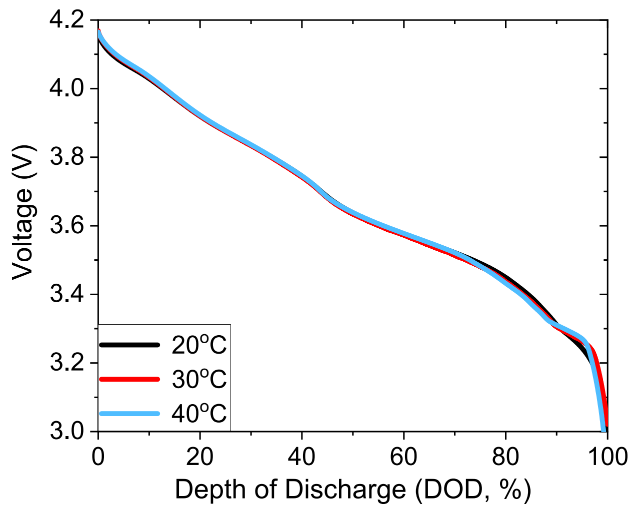
W01: This component represents the Warburg response observed in the lower frequency region of the Nyquist plots. The Warburg impedance illustrates the diffusion processes of lithium ions within the battery materials, highlighting how ionic transport contributes significantly to the battery's overall impedance and response time.

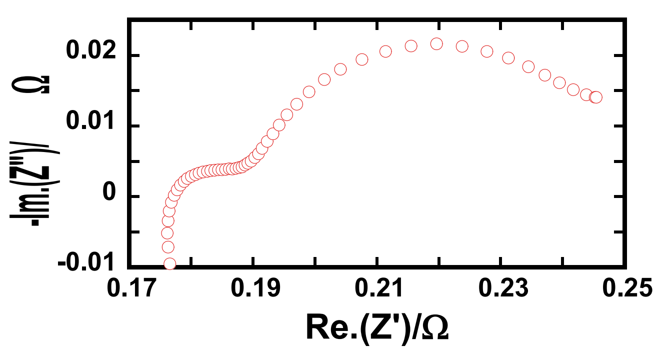
Data Availability






Heat flow (mW)
Time (h)
Charge
Discharge


Voltage (V)
Depth of Discharge (DOD, %)
20°C
30°C
40°C


-Im.(Z'')/Ω
Re.(Z')/Ω