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Loss Verification of Li-ion Battery Cell from Thermal Flow Measurements

Daniel Poposki[1*], Andrew Mandeya[1], Istaq Ahmed[1], Torbjörn Thiringer[2]

[1]*Volvo Group Trucks Technology*

[2]*Chalmers University of Technology*

* *Corresponding author, daniel.poposki@volvo.com*

Executive Summary

This paper investigates heat generation in lithium-ion battery cells, a critical factor influencing the performance, lifespan, and safety of batteries, particularly in electric vehicle applications. Using an isothermal heat conduction calorimeter, heat flow measurements were performed on a commercial 850 mAh lithium-ion pouch cell. The sensitivity of the calorimeter demonstrated a capability of measuring heat flow levels down to 6 mW. By quantifying the contributions of the irreversible and reversible heat components, the entropic profile of the cell was determined. This study highlights the necessity of accurate heat flow measurement and emphasizes challenges posed by heat leakage and measuring low power detection using this calorimetric method.

Keywords: Batteries, Measuring Methods & Equipment, Energy Storage Systems

1 Introduction

As the demand for electric vehicles (EVs) grows, development of battery technology is an essential part and at the core of modern electrification efforts. Their performance remains tightly connected to how well their thermal and electrical properties are understood. In particular, effective management of the heat generated during the operation of lithium-ion (Li-ion) batteries can significantly impact their efficiency, longevity, and overall safety, especially in applications such as electric vehicles [1].

Effectively managing heat generation within batteries is hence an essential part of any system design. To accurately predict and manage thermal behavior continues to be a challenging effort since it involves complex interactions between the materials, electrochemical processes, and environmental operating conditions [2].

This experimental study focuses on utilizing calorimetry for analysing heat generation in Li-ion battery cells and characterize their electrothermal behaviour. The results will be used to develop accurate battery models supporting the design of efficient thermal management systems. With this purpose in mind, this work aims more specifically at identifying the capabilities and limitations of the calorimetric method and setup used.

2 Methodology

To investigate and characterize the heat generation of a Li-ion battery cell, a custom-built isothermal heat conduction calorimeter is used to measure the heat flow in a commercial pouch cell. A well-insulated setup is essential to reduce heat leakage and hence improve the sensitivity of heat measurement signals. This section outlines the theoretical basis, calorimeter setup, and procedure used in this study.

2.1 Battery heat generation

In a Li-ion battery, heat generation can be categorized into two components, irreversible heat and reversible heat [3].

- **Irreversible heat** is generated due to energy losses from internal resistances and polarization effects. It is always exothermic, resulting in heat being released.
- **Reversible heat** arises from entropy changes during electrochemical redox reactions. It can be either exothermic or endothermic, which in a cell can vary depending on e.g. the state of charge and temperature.

The total heat generation Q can be expressed by

$$Q_{\text{generated}} = Q_{\text{irreversible}} + Q_{\text{reversible}} = I^2 R_{\text{cell}} + I T_{\text{cell}} \frac{\partial U_{\text{ocv}}}{\partial T_{\text{cell}}}, \tag{1}$$

where I is the rate of the current, R_{cell} is the ohmic resistance of the cell, T_{cell} is the temperature of the cell and $\partial U_{\text{ocv}} / \partial T_{\text{cell}}$ is the entropic coefficient. The entropic coefficient can be experimentally determined by calorimetry if the other parameters are known.

2.2 Calorimetric Setup

An overview of the calorimetric setup can be viewed in the illustrated schematic in Figure 1. The calorimeter is housed inside of a Styrofoam box to mitigate the temperature impact from the ambient environment. The calorimeter includes water-cooled heat sinks via a temperature regulated recirculating cooler. The calorimeter utilizes thermoelectric heat flow sensors to measure the heat dissipated from the test object. The calorimeter in turn consists of two sides labelled A and B. The test and reference sample are placed between side A and B with the two plates tightened using spring holders.

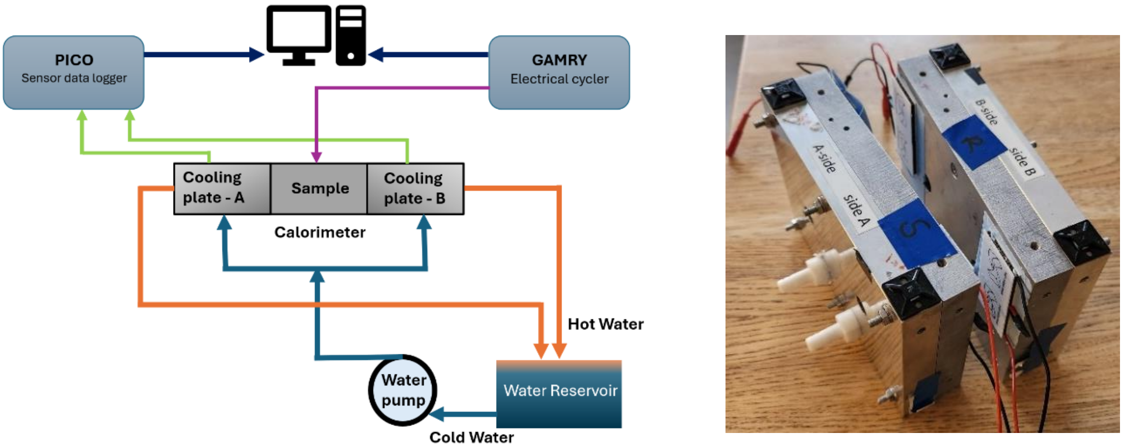

Figure 1: Overview of the experimental setup.

2.3 Measuring heat flow

Thermoelectric heat-flow sensors are integrated within the calorimeter to measure heat flux based on the Seebeck effect, where temperature differences across a sensor create measurable voltage differences. The sensor output voltage ΔU depends linearly on the temperature difference ΔT according to

$$\frac{\Delta U}{\Delta T} = a\,, \tag{2}$$

where a is the Seebeck coefficient in V/°K for a single sensor. The sensor's overall sensitivity depends on two main properties, the Seebeck coefficient, and the thermal conductance K of the device in W/°K [4]. The ratio of these parameters yields the calibration coefficient, ε defining how voltage measurements translate into actual heat flow through a plate consisting of N number of sensors,

$$\varepsilon = \frac{K}{Na}\,. \tag{3}$$

Accurate calibration of the calorimeter is essential to accurately interpret voltage signals as actual thermal power. To attain a calibration coefficient, electrical calibration was done by using a power resistor of known resistance, R_{cell}, electrically heated by a controlled current, I, where the thermal power, P, generated is given by

$$P = I^2R\,. \tag{4}$$

Throughout the experiment, a non-heated reference sample with similar thermal properties was included to measure background noise and support measurement reliability. Calibration results provide a calibration coefficient ε, enabling accurate heat flow determination from the measured voltages [5].

3 Results

This section presents the outcomes of the calibration and sensitivity analysis of the isothermal calorimeter. After calibration, the calorimeter is used to characterize the heat generation of a commercial Li-ion pouch cell rated at 850 mAh under varying C-rates and ambient temperatures. Contributions from both irreversible and reversible heat are also separated and analyzed.

3.1 Calorimeter calibration

In the first calibration experiment, a power resistor was utilized as heating element at various power levels including 1.25 W, 1 W, 0.625 W, 0.25 W and 0.125 W. The measurements from the heat flow sensors at side A and B for the test and reference samples are shown in Figure 2. In Figure 3, the ratio between measured voltage and power in a 10-minute window is presented. From this graph, the calibration coefficient ε can be found.

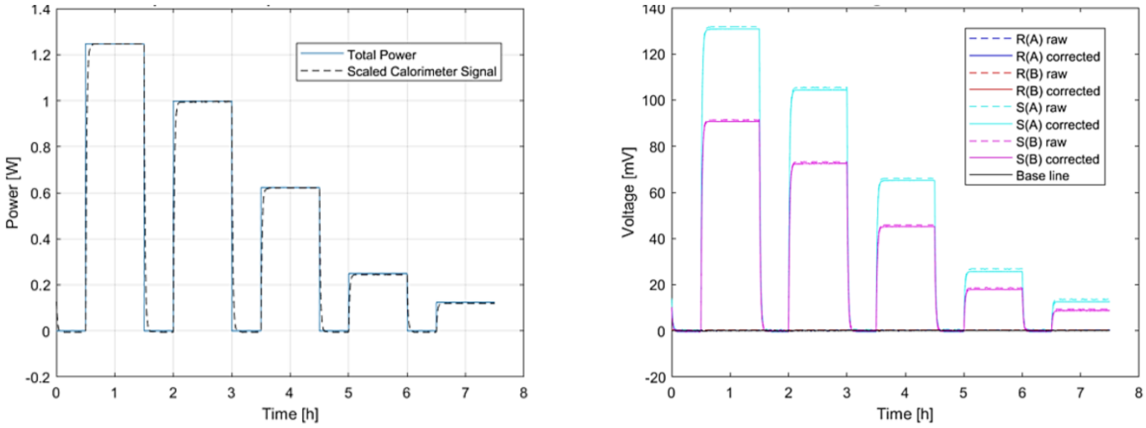

Figure 2: Heat flow measurement at various calibration power levels.

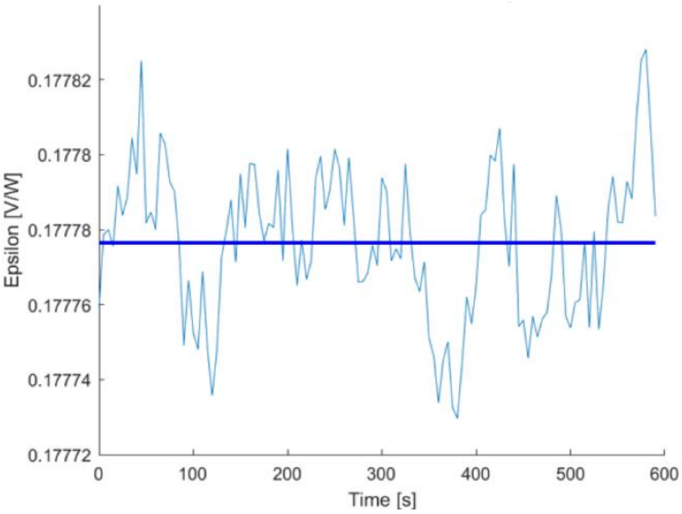

Figure 3: Calibration coefficient ε is found as the ratio between voltage and power averaged in a 10-minute window.

The voltage response from the thermal heat flow sensors in Figure 2 can be observed to be proportional to the power supplied to the power resistor used as the calibration test sample.

Furthermore, it was observed that the power resistor in this experimental setup produced more heat on side A than side B due to the physical properties of the resistor being asymmetrical.

To investigate how low thermal flows can be measured, a second calibration experiment was performed to investigate the limitations in terms of the calorimeter's sensitivity. During this calibration experiment, it was observed that power levels could be measured down to 6 mW while remaining distinguishable from the reference noise. The corresponding calibration coefficient ε for this power level is found from the signal in Figure 4.

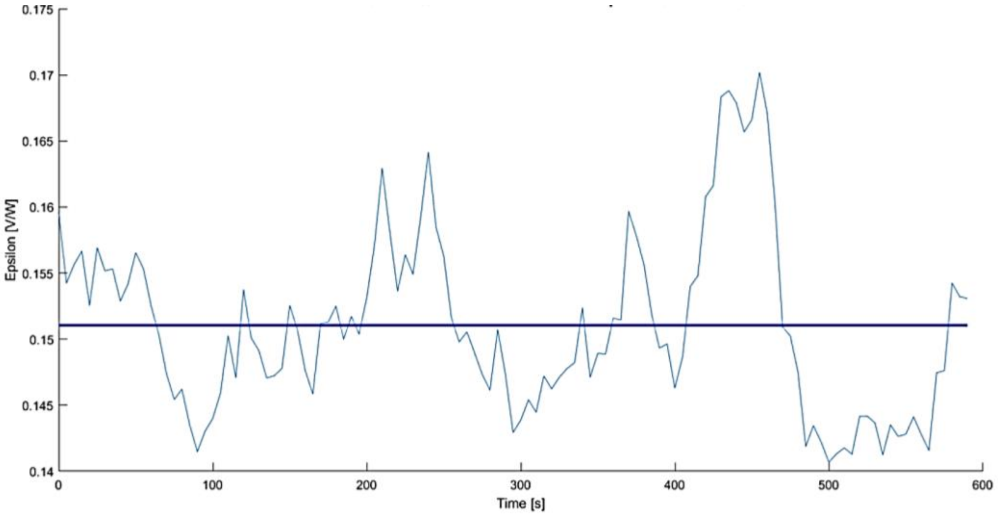

Figure 4: Calibration coefficient ε is found as the ratio between voltage and power at 6 mW in a 10-minute window.

The calibration coefficient is averaged at $\varepsilon = 0.151$ V/W for the 6 mW power level. At power levels lower than 6 mW, there was no clear distinction between the sample and reference signal. Hence, 6 mW is determined to be the limitation of the sensitivity of this calorimeter.

3.2 Battery heat generation

This section describes the heat flow behavior of the 850 mAh lithium-ion pouch cell when cycled under different C-rates and temperatures. Calorimetric data was used to separate reversible and irreversible heat contributions.

3.2.1 Effect of varying C-rate

The total heat flow was measured at various C-rates. Figure 5 shows the measured heat flow in charging direction for 0.1C, 0.5C, 1C and 2C at 22°C. Similarly, Figure 6 shows the measured heat flow throughout the discharge direction, 0.1C, 0.5C, 1C, 2C and 3C.

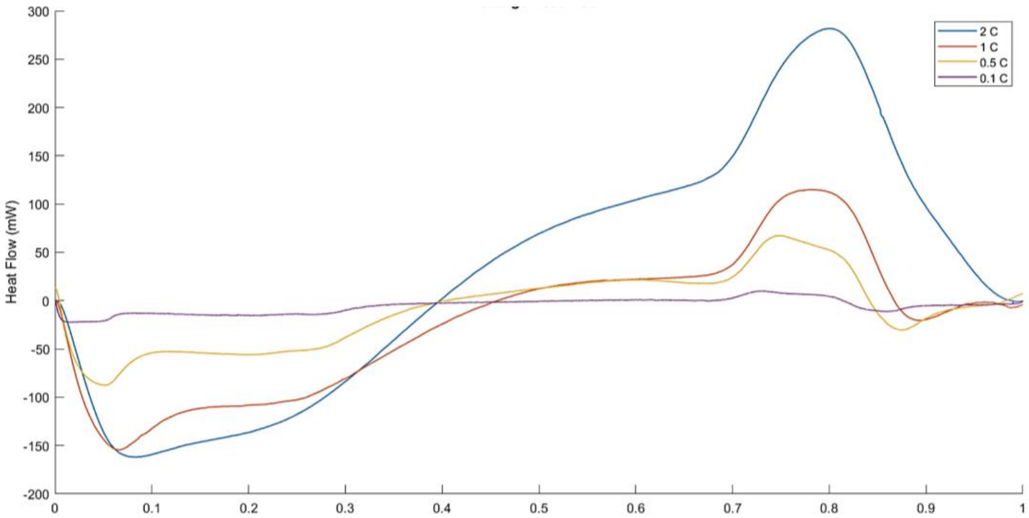

Figure 5: C-rate vs SOC in charge direction at 22°C.

For the heat generation throughout the charging of the battery, both endothermic and exothermic behaviour was observed. In Figure 5, clear endothermic behaviour was observed between 0-40 % State of Charge (SoC) range, while exothermic heat flow was measured above 40% SoC.

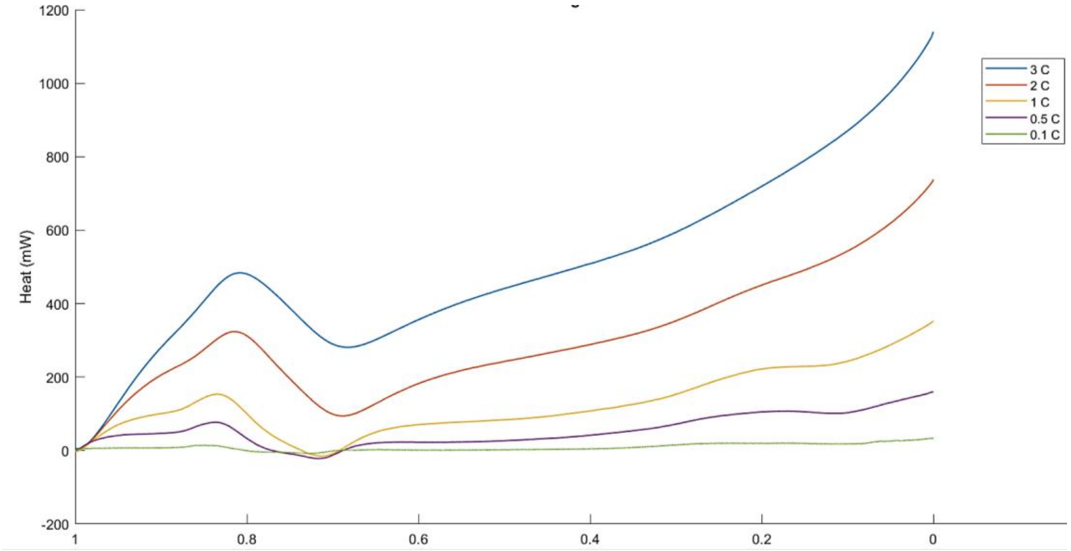

Figure 6: C-rate vs SOC in discharge direction at 22°C.

In Figure 6, it can be observed that the discharge profile for various C-rates is predominantly exothermic.

3.2.2 Effect of varying temperature

The effect of changing the initial temperature of the battery has on the total heat generation from the cell was studied at 10°C, 22°C and 30°C. Figure 7 shows the measured heat flow during 0.1C charging.

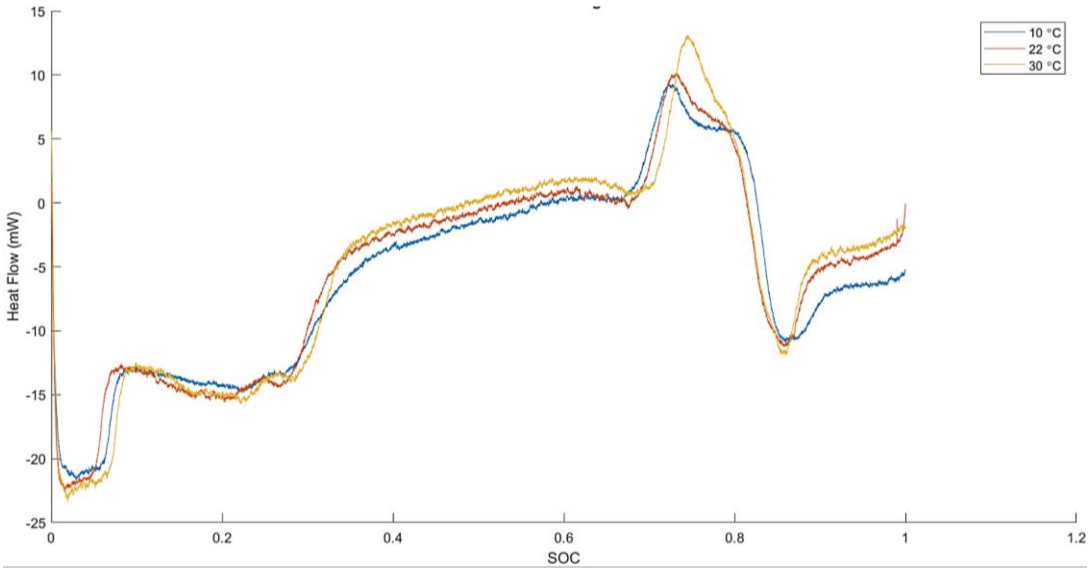

Figure 7: Heat flow generation during 0.1C charge at 10°C, 22°C and 30°C.

At 0.1C, the total heat generation was lowest at 10°C when compared to the other temperatures due to the reversible heat generation component playing a more significant role at lower C-rates. Similarly, Figure 8 shows the heat flow at 1C charging.

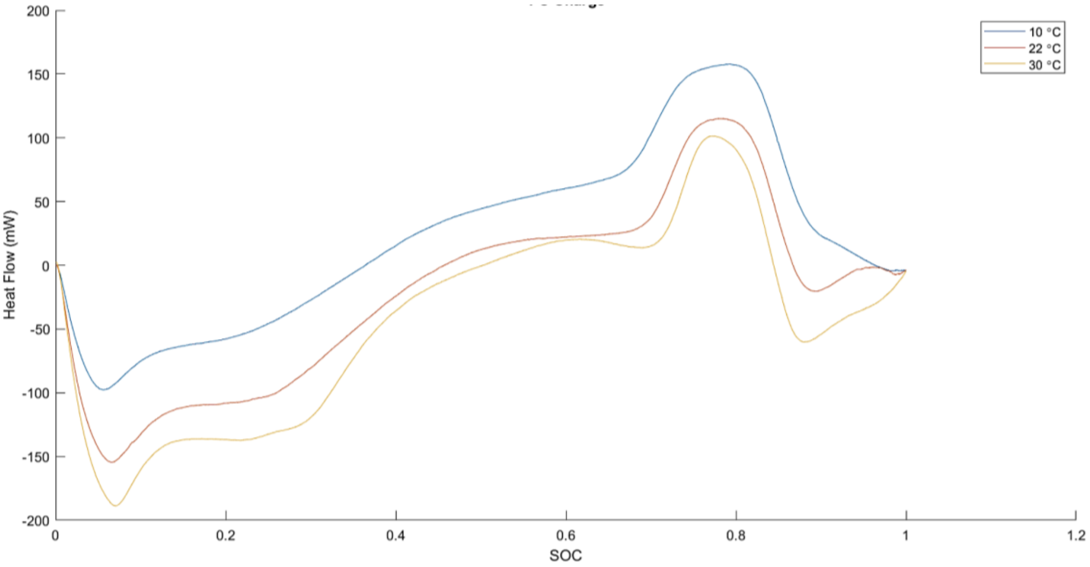

Figure 8: Heat flow generation during 1C charge at 10°C, 22°C and 30°C.

From Figure 8, the total heat generated is observed to be higher at 10°C for higher C-rates. This can mainly be attributed to the fact that the irreversible heat attributed to ohmic losses dominates at higher C-rates, since the ohmic resistance of the battery is larger at lower temperatures.

3.2.3 Reversible and irreversible heat generation

The reversible and irreversible heat components can be separated by estimating and subtracting the irreversible heat from the total measured heat flow using equation 1. Figure 9 shows the total heat flow measured from the battery throughout charging and discharging with 0.1 C.

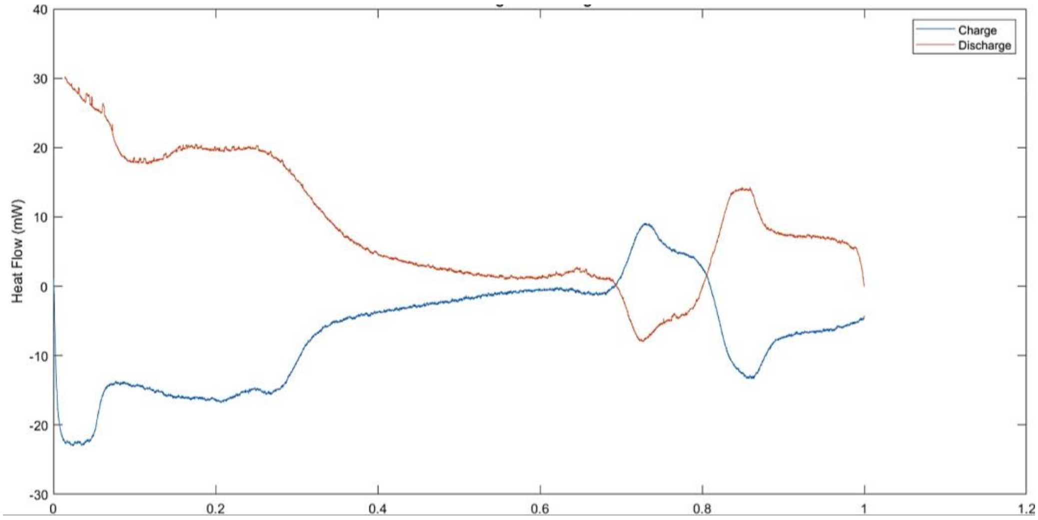

Figure 9: Total heat flow vs SoC at 0.1C charge and discharge at 22 °C.

To estimate the irreversible heat contribution, the open circuit voltage U_{OCV} can be subtracted from the battery terminal voltage U_{cell} during operation according to

$$Q_{\mathrm{irreversible}} = I\,(U_{\mathrm{cell}} - U_{\mathrm{OCV}})\,. \tag{5}$$

From the data presented in Figure 10 and Figure 11, the estimated irreversible heat generation can be subtracted from the total heat generation measured in the calorimeter to ultimately estimate the reversible heat contribution during charge and discharge process respectively.

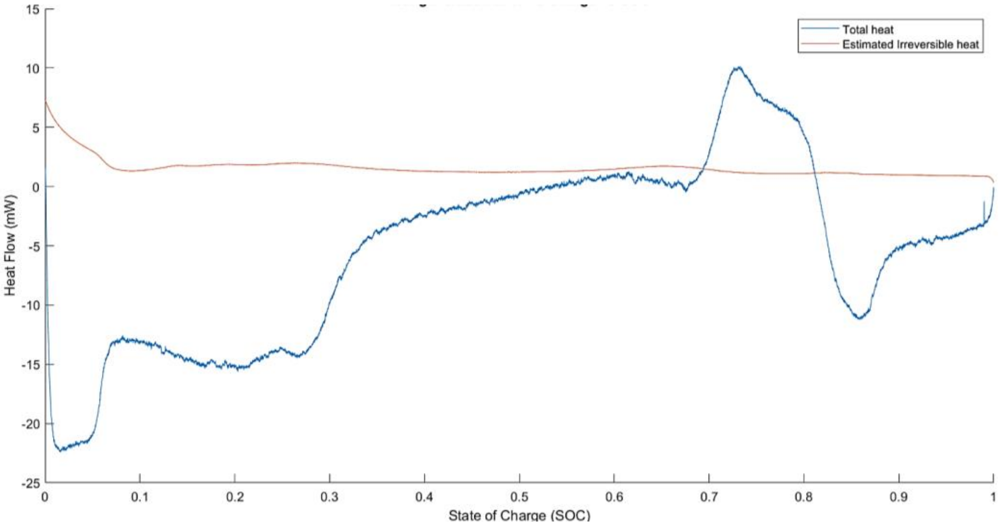

Figure 10: Charge total heat flow vs estimated irreversible heat flow.

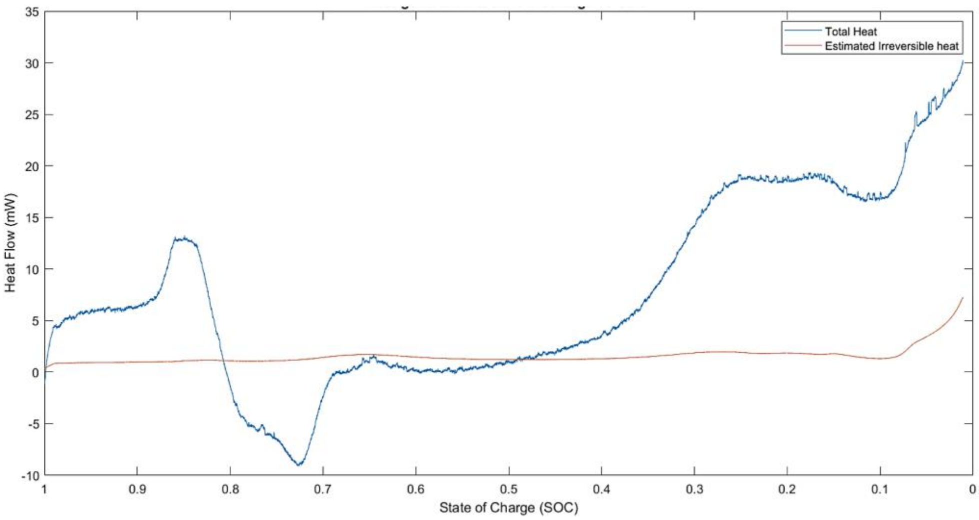

Figure 11: Discharge total heat flow generated vs estimated irreversible heat flow.

The results lead to the entropic profile of the battery in Figure 12, which is described with the entropic coefficient $\partial U_{\mathrm{ocv}}/\ \partial T_{\mathrm{cell}}$ derived from equation 1.

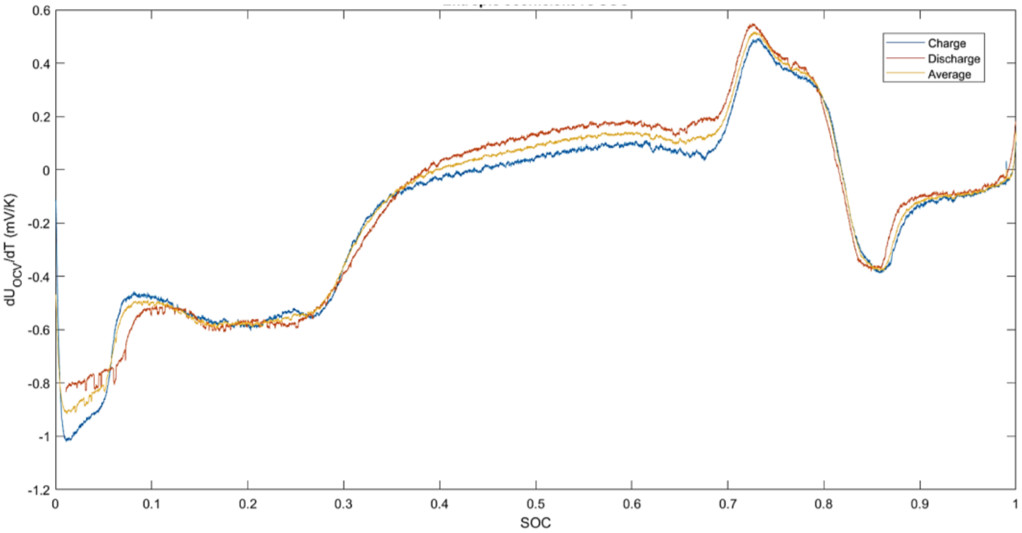

Figure 12: Entropic profile comparison between charge and discharge.

4 Discussion and conclusions

The results demonstrated the use of an isothermal heat conduction calorimeter to measure heat generation in an 850 mAh commercial lithium-ion pouch cell and to characterize its entropic profile. The calorimeter demonstrated a capability to measure heat flows as low as 6 mW. Below this power level, the noise in the measurement signal is too significant to obtain reliable sensor readings. The insulation of the calorimetric setup and the stability of the external ambient temperature were shown to be crucial in reducing the signal-to-noise ratio in the heat flow measurements.

However, the calorimetric setup did prove capable of characterizing the 850 mAh Li-ion pouch cell. A differentiation between irreversible and reversible heat contributions was made to analyze the entropic profile of the cell at 0.1C, where the contribution from irreversible heat generation is relatively small. The measured entropic profile followed typical patterns observed in similar Li-ion batteries, with both endothermic and exothermic behavior as a function of the state of charge, attributed to phase transitions occurring within the electrode materials.

While this study focused on a flat-format pouch cell, future work could aim to extend this methodology to cells with other geometries, such as cylindrical cells, which pose challenges in thermal contact and heat dissipation. This will likely require specially designed fixtures to ensure consistent thermal coupling and to reduce measurement uncertainty. To further push the boundaries of heat flow detection, efforts to improve the sensitivity of the calorimeter beyond 6 mW would also enable effective testing of small prototype cells, which are often used in research but are difficult to characterize thermally due to their low heat generation. Exploring other techniques such as adiabatic calorimetry could improve the ability to accurately measure small heat flows, as heat exchange with the surroundings would be minimized.

Additionally, the calibration coefficient showed sensitivity to power level variations. While the system displayed consistent results at higher input powers, slight deviations at lower levels suggest that calibration accuracy may vary depending on the magnitude of the heat source. This introduces a potential source of error when analyzing very low thermal outputs and emphasizes a need to minimize such deviations.

In summary, this work demonstrates the viability of isothermal calorimetry for thermal characterization of commercial lithium-ion cells. The methodology enabled separation of reversible and irreversible heat generation mechanisms and captured thermodynamic behaviors of the cell. However, challenges remain in extending this approach to measuring lower heat flows and more complex cell formats. Addressing these limitations through improved design, insulation, and calibration will allow for broader application of calorimetric studies in both research and industrial battery development.

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Authors

Daniel Poposki received M.Sc. degree in Electric Power Engineering from Chalmers University of Technology in the city of Gothenburg in 2019. After graduation, he spent three years working with battery development at Volvo Group Trucks Technology, after which he decided to learn more about batteries through research as an industrial Ph.D. candidate at Chalmers University of Technology while remaining to be a team member in the Battery Cell Technology team in Volvo Group Trucks Technology. The focus of his research is on experimental characterization of battery cells and studying heterogeneous degradation effects on large format cells.

Andrew Mandeya received a B.Sc degree in Chemical Engineering from the University of Cape Town in 2017, and a M.Sc in Sustainable Energy Systems at Chalmers University of Technology in 2023. The focus of his M.Sc degree was on calorimetric measurements on a smaller calorimeter device measuring small format pouch cells at Volvo Group Trucks Technology. Currently, he is in the team for battery cell evaluation and analysis within Volvo Group's department of Energy Storage Engineering.

Torbjörn Thiringer works at Chalmers university of Technology, in Göteborg Sweden, as a professor in applied power electronics. He took his M.Sc. and Ph.D. at Chalmers University of technology in 1989 and 1996 respectively. His areas of interest include the modeling, control and grid integration of wind energy converters into power grids, battery technology from detailed cell modelling to system aspects, as well as power electronics and drives for other types of applications, such as electrified vehicles, buildings and industrial applications.

Istaq Ahmed completed M.Sc and PhD from Chalmer University of Technology, Gothenburg, Sweden. He worked as post-doctoral research fellow (2008-2010) in collaboration with Rutherford Appleton lab, UK & Chalmer University of Technology, Sweden. In 2010, he started corporate carrier in the field of heavy-duty automotive industry at Volvo Technology in Sweden focusing on electric vehicles. He worked 14 years at Volvo Technology until 2024 when he moved to Swedish Electric Transport Laboratory (SEEL). In his both academic and corporate carrier, Istaq Ahmed published 30+ journal papers and 20+ industrial patents (mainly for Volvo Technology) in the field of both fuel cells and lithium ion batteries. Today, he is mainly working with the characterisation and safety of the lithium-ion batteries for automotive applications.