

Research papers

Thermo-electrochemical performance of lithium-ion cells with immersion cooling under various operating conditions

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ABSTRACT

The safe operation of a device relying on lithium-ion batteries depends on a robust cooling system. While the simplest systems still rely on convective cooling using air to regulate temperature, immersion-based cooling has garnered attention due to its potential to ensure uniform temperature distribution in a cell and mitigate thermal runaway. Though past research focused on temperature regulation, the apparent negative impact of immersion cooling on electrochemical performance at high currents remains unaddressed. In this work, the thermo-electrochemical behaviour of a commercial lithium-ion cell, which is convectively cooled by air and a dielectric liquid in a confined space, is compared. The effect of discharge current (0.5C–4C), ambient temperature (25 °C–45 °C), and flow rate of the dielectric liquid (200 ml/min–1000 ml/min) on the thermo-electrochemical performance is elucidated. Using experiments, physics-based simulations and data-driven modelling, the role of these operational parameters is explained. Heat removal by air through free convection leads to a lower heat transfer rate and a higher cell temperature, correspondingly enhanced electrochemical kinetics, but is more likely to lead to catastrophic failure. In contrast, immersion cooling by dielectric liquid limits the rise in cell temperature accompanied by relatively lower discharge capacity and energy, demonstrating the trade-off between safety and performance.

1. Introduction

Lithium-ion batteries (LIBs) have evolved with applications ranging from small consumer electronics to heavy electric vehicles, grid energy storage systems, and defence sectors. Governments are also promoting the use of electric vehicles by providing incentives to manufacturers and consumers [1–4]. The scale of production and continuous technological advancement have led to a reduction in the production cost of LIBs. However, safety, high power and energy density, life span, operating temperature, and recycling are challenges to overcome for the widespread adoption of Lithium-ion batteries in EVs [5].

The discharge current and ambient temperature play a vital role in the safety and performance of the battery packs. The optimal temperature range for Lithium-ion batteries is believed to be 15 °C to 35 °C [6]. Outside this range, the role of degradation mechanisms dominates, due to which safety is compromised. The role of temperature and discharge current is intricately tied due to their combined effect on heat generation in a Lithium-ion cell [7]. A high discharge current leads to substantial irreversible heat generation inside the cells, increasing their temperature [8,9]. The increased temperature affects the reaction kinetics, leading to a higher rate of side reactions and loss of lithium inventory [10–13]. If the cell temperature is allowed

to increase uncontrollably, it may lead to burning of the separator and decomposition of the solid electrolyte interphase (SEI) [14,15]. Furthermore, the decomposition of the electrolyte results in the thermal runaway of the cell [16,17], which can propagate to other cells, leading to catastrophic failure of the battery pack. Clearly, an effective cooling system is essential to achieve the desirable features of a long battery life, high power, and safe battery operation [18–20].

Fig. 1 presents an overview of various cooling systems explored by various research groups to manage cell temperature. This includes the classification based on the heat transfer medium and based on power consumption [20–29]. Convective air cooling is frequently used in two-wheeler applications and even in some four-wheel applications [22, 23,30]. Nowadays, due to high power and safety requirements, indirect or direct liquid cooling systems are incorporated with battery packs [31,32]. These cooling systems are primarily based on liquid jackets, cold plates, heat pipes, phase change material, or immersion-based cooling [24–27].

Natural air, direct and indirect liquid, and phase-change cooling present distinct advantages in battery thermal management based on material composition and active or passive operation. Natural air cooling, a passive method, relies on convective heat transfer to ambient air,

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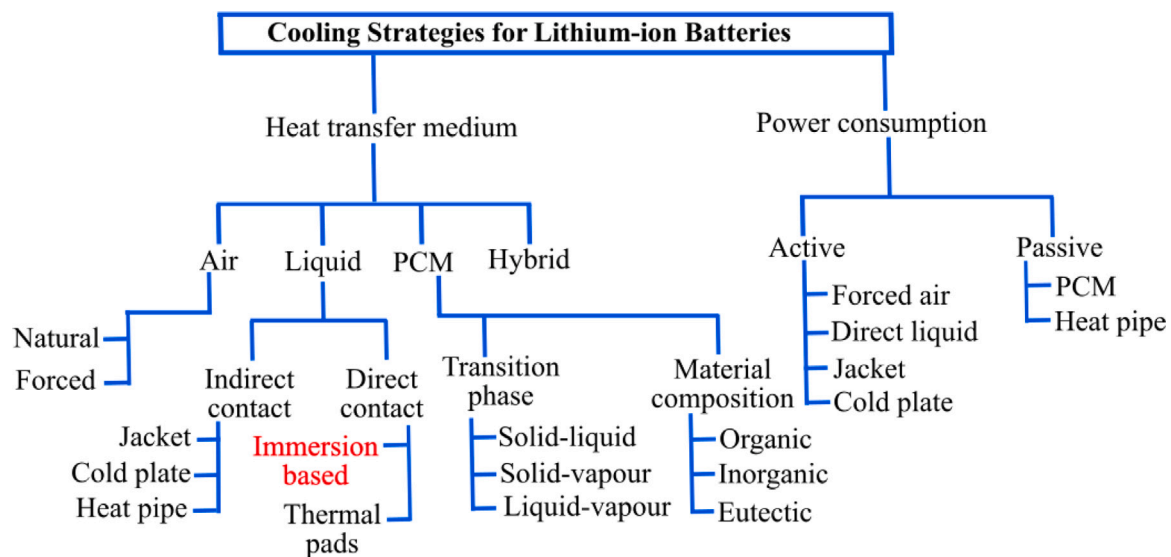


Fig. 1. Various cooling system strategies for thermal management of lithium-ion batteries.

typically requiring only flow pathways and essential heat sink [23,30]. However, this method is less efficient, especially under high power loads where high heat dissipation is needed. In contrast, direct liquid cooling actively circulates a liquid coolant, such as a glycol-water mixture, around the battery cells. This enables a higher heat transfer coefficient due to the coolant's thermal properties and close contact with cell surfaces [20]. Indirect liquid cooling separates the coolant from the cells via metal or plastic interfaces, offering electrical insulation and reduced coolant contamination risk, albeit with slightly lower heat transfer efficiency than direct methods. Phase-change cooling employs materials with high latent heat properties (e.g., paraffin wax or composite phase-change materials) to absorb excess heat during melting, providing effective passive cooling while requiring less energy input [25]. However, this method faces challenges in sustained high-temperature conditions due to the finite melting/freezing cycles. Selecting any of these approaches depends on trade-offs in the availability of space, material compatibility, effectiveness of cooling, and power requirements imposed on the battery.

It is now well known that there exists a non-uniform temperature variation among the cells of a battery module [33]. This makes cells prone to localized degradation and premature failure when cooled with air [22]. A recent study [28] has highlighted the role of immersion-based cooling systems in effectively mitigating the temperature rise in a cell and compared it with forced air cooling. In a similar work, researchers explored the role of dielectric liquid cooling and charging rate on the rise in cell temperature [29,34]. Earlier work in our group, utilizing experiments and data-driven techniques, has shown that ambient temperature has a significant influence on discharge capacity and very little effect on discharge power [9].

The aforementioned studies were primarily focused on observing the temperature rise when discharging cells at various C-rates, with a limited exploration of the effect of the cooling system on the battery's electrochemical performance, particularly from the interplay of discharge rate with coolant temperature and its flow rate. Furthermore, the role of the choice of cooling fluid (gas or liquid) in competing objectives, namely cell safety (quantified in terms of cell temperature rise) and electrochemical performance (quantified through discharge capacity, power and energy) has not been reported so far. Thus, there is a pressing need to investigate the influence of C-rate, ambient temperature, and dielectric fluid flow rate on the thermo-electrochemical performance of lithium-ion batteries.

The primary objectives of this work are: (a) to comprehensively examine how ambient temperature, discharge current, and dielectric

fluid flow rate influence the thermo-electrochemical performance of lithium-ion cells under immersion cooling conditions, and (b) perform a comparative exercise with convective cooling using air. In this work, a combination of experiments, thermo-electrochemical modelling and data-driven techniques have been employed to develop a simplified representation of cell performance and thermal safety. We have relied on surrogate modelling to estimate essential thermo-electrochemical parameters, namely cell temperature, discharge capacity, average discharge power, and discharge energy. This article is structured into the following sections. Section 2 delineates the methodology, encompassing details on the experimental setup, design of experiments, thermo-electrochemical modelling, and surrogate-based data-driven modelling. Section 3 presents the results and discussions pertaining to the thermo-electrochemical performance of commercial cylindrical cells, while Section 4 provides a summary of conclusions drawn from the study and outlines potential avenues for future research.

2. Methodology

This work utilizes thermo-electrochemical physics-based modelling [35,36] and data-driven techniques [37–39] as tools to analyse the performance and safety of lithium-ion batteries. Physics-based models and their governing equations are derived from the conservation of species and charge within the battery cell. These equations incorporate numerous internal parameters such as the thickness of positive and negative electrodes and separator thickness, particle radius, and thermophysical properties. These model inputs are typically determined through advanced material characterization methods like field-emission scanning electron microscopy (FESEM), energy dispersive X-ray spectroscopy (EDX) and differential scanning calorimetry (DSC) analyses.

To estimate various thermo-physical performance indicators, a combination of design of experiments (DOE) and surrogate modelling techniques have been applied. The data to be used for training and testing of the surrogate models was obtained from an experimentally validated physics-based electrochemical model coupled with a three-dimensional thermal model that was implemented in the COMSOL Multiphysics toolbox.

The surrogate model aims to predict outputs in scenarios where simulations were not performed. Furthermore, the reduced-order surrogate model is significantly faster to evaluate than conventional physics-based models. In fact, surrogate modelling requires a relatively small number of training and test data points compared with other machine learning methods, making it particularly appealing for modelling the thermo-electrochemical response of commercial lithium-ion cells [40, 41].

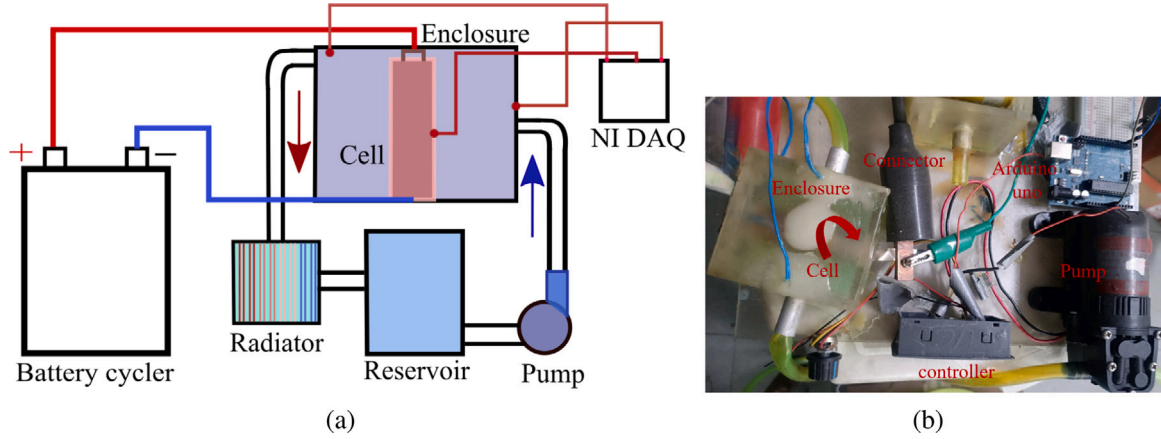


Fig. 2. Immersion-based cooling system setup (a) schematic and (b) cooling enclosure connections.

Table 1
Thermophysical properties of HiTech Therm 60 mineral oil.

Temperature (°C)	Specific heat (J/kg K)	Density (kg/m ³)	Viscosity (mPa s)	Thermal conductivity (W/m K)
20	1867	876	48	0.134
30	1894	865	30	0.133
40	1935	860	20	0.131
50	2018	857	14.3	0.130
60	2044	845	10.3	0.129

2.1. Experimental setup

This study utilized commercial 18650-type lithium-ion cells (make: Samsung) with a rated capacity of 2.9 Ah to generate raw data for validating the physics-based model. The experimental setup consisted of an Arbin battery cycler capable of discharging cells up to 60 A current attached to a computer to record the instantaneous voltage response. Temperature measurements were carried out using T-type thermocouples interfaced with a National Instruments (NI) data acquisition system (DAQ), specifically the NI9214 module and NI 9171 chassis. The NI DAQ was instrumental in signal acquisition and processing, with surface temperature data being recorded at the mid-length of the lithium-ion cell.

The experimental setup, illustrated in Fig. 2, incorporated an acrylic enclosure with dimensions of 6 cm × 6 cm × 6 cm. The enclosure dimensions were meticulously designed to ensure a fully flooded condition and maintain laminar flow, optimizing the system to achieve the maximum convective heat transfer coefficient. The cubic acrylic chamber had a wall thickness of 5 mm. This enclosure accommodated vertically placed 18650 cells, each measuring 6.5 cm long. This configuration was suitable to facilitate uniform heat dissipation across the cell surface. The setup also included an air-cooled radiator, a reservoir and a pump to circulate the dielectric liquid. An Arduino Uno microcontroller regulated the pump's flow rate. Immersion cooling experiments were performed by submerging the cell in mineral oil (HiTech Therm 60) whose variation in thermo-physical properties with temperature is described in Table 1. Experiments were also performed on cells in the acrylic enclosure to mimic the heat dissipation by the cell into quiescent air through free convection in narrow confinement. This was necessary for an effective comparison of the two thermal management approaches considered in this work.

A standardized protocol was followed for the initial charging and resting of the cell prior to initiating the discharge tests. The cells were charged with a constant current corresponding to a rate of 0.5C until the upper cutoff voltage (4.2V) was achieved, after which the charging process changed to a constant voltage mode until the current decayed

below the 0.05C rate. The cells were observed to have reached thermal equilibrium within the enclosure when rested for a duration of 2 h under open circuit conditions. Following this, cells were discharged at different constant current discharge rates (0.5 to 4C), ambient temperatures (25 °C to 45 °C), and dielectric fluid flow rates (200 ml/min to 1000 ml/min) up to the lower cutoff voltage of 2.8 V. Upon complete discharge, the cells were allowed to cool to ambient temperature via free convection, typically completed within 30 min. These experiments under controlled conditions were subsequently used to validate the mathematical model under air and liquid-cooled scenarios.

2.2. Mathematical model

A one-dimensional isothermal electrochemical model based on the Newman framework [42], as depicted in Fig. 3, was implemented in COMSOL Multiphysics. The validation of the electrochemical model has already been presented in our earlier papers [36,43]. The standard electrochemical equations used in the model are detailed in Table 2. The electrochemical model was integrated with a volume-averaged three-dimensional heat conduction model. Spatially varying thermal properties were employed to achieve a realistic temperature distribution within the cell [44]. The laminar flow around the cylindrical cell was assumed to be governed by the Navier–Stokes equations for a Newtonian fluid, with the maximum Reynolds number based on the cell diameter for cross-flow arrangement (achieved for the highest flow rate) calculated to be 2.5. The heat generated by the electrochemical processes was calculated from the pseudo-2D model and incorporated as a source term in the thermal model. The spatial-averaged cell temperature obtained from the solution of the coupled heat diffusion, fluid flow, and heat advection equations was fed into the electrochemical model, facilitating the coupling of thermo-electrochemical phenomena.

In the pseudo-2D model, the local molar reaction flux density $j_{f,i}$ depends on the local overpotential η_i and is given by the Butler-Volmer relationship,

$$j_{f,i} = k_i (C_{max,i} - C_{s,i})^{\alpha_a} C_{s,i}^{\alpha_c} C_e^{\alpha_a} \left[\exp\left(\frac{\alpha_a F \eta_i}{RT}\right) - \exp\left(-\frac{\alpha_c F \eta_i}{RT}\right) \right]. \quad (1)$$

The index $i \in \{n, s, p\}$ represents the negative electrode, separator and positive electrode, respectively. The overpotential is given by $\eta_i = \phi_{s,i} - \phi_{e,i} - U_i$. The effective conductivities ($\kappa_e^{\text{eff}}, \sigma_e^{\text{eff}}$) and diffusivities ($D_e^{\text{eff}}, D_s^{\text{eff}}$) of the electrolyte and electrodes account for the local volume fractions and tortuosity.

Furthermore, the heat generated within the cell [5,36] is computed using

$$\dot{Q}_{\text{local}}(x, t) = a_{s,j} j_{n,i} \eta_j + a_{s,j} j_{n,i} T \frac{\partial U_j}{\partial T} + \sigma_s^{\text{eff}} \nabla \phi_s \cdot \nabla \phi_s + (\kappa_e^{\text{eff}} \nabla \phi_e + \kappa_D^{\text{eff}} \nabla \ln C_e) \cdot \nabla \phi_e \quad (2)$$

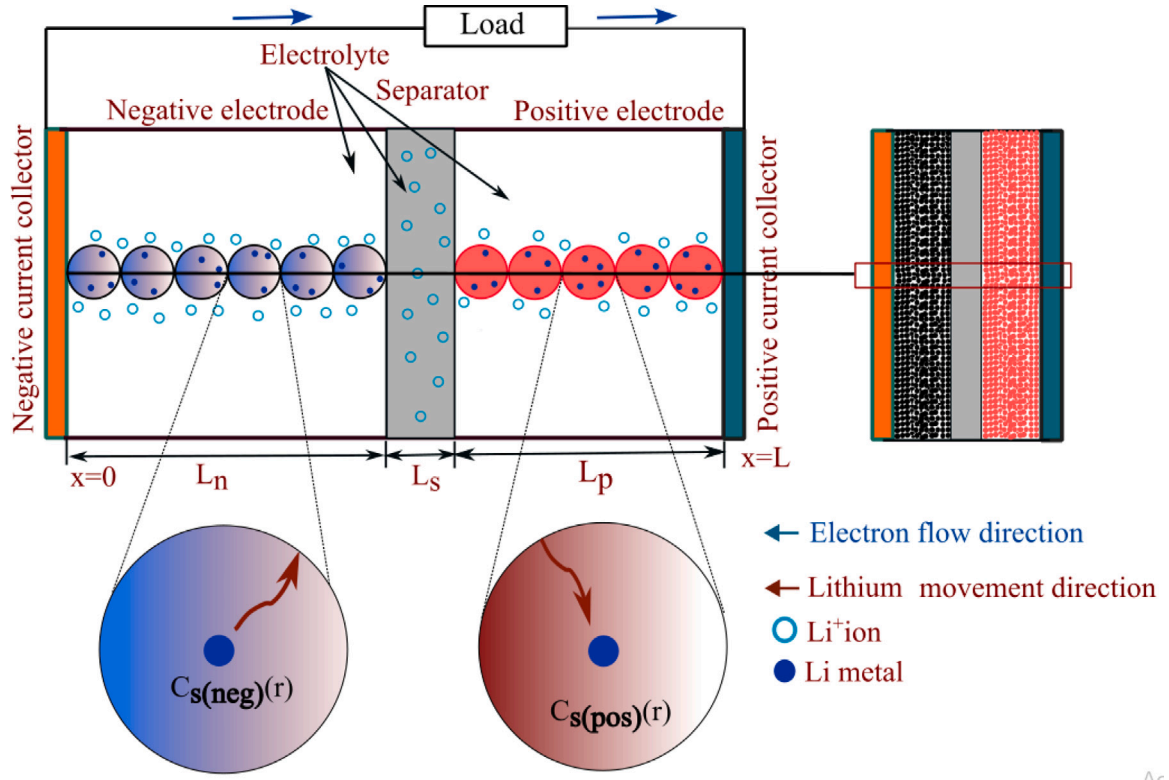


Fig. 3. Pseudo-2D representation of the Lithium-ion cell.

Table 2
Species and charge conservation equations.

Description	Governing equations	Boundary conditions	Initial conditions
Species conservation in electrolyte and separator	$\epsilon_{e,i} \frac{\partial C_e}{\partial t} = \frac{\partial}{\partial x} \left(D_{e,i}^{\text{eff}} \frac{\partial C_e}{\partial x} \right) + a_{s,i} j_{f,i} (1 - t_+^0)$ $i \in \{n, p\}$ $\epsilon_{e,s} \frac{\partial C_e}{\partial t} = \frac{\partial}{\partial x} \left(D_{e,s}^{\text{eff}} \frac{\partial C_e}{\partial x} \right)$	$-D_{e,n}^{\text{eff}} \frac{\partial C_e}{\partial x} \Big _{x=0^+} = 0$ $-D_{e,n}^{\text{eff}} \frac{\partial C_e}{\partial x} \Big _{x=L_n^-} = -D_{e,s}^{\text{eff}} \frac{\partial C_e}{\partial x} \Big _{x=L_n^+}$ $-D_{e,s}^{\text{eff}} \frac{\partial C_e}{\partial x} \Big _{x=L_n+L_s^-} = -D_{e,p}^{\text{eff}} \frac{\partial C_e}{\partial x} \Big _{x=L_n+L_s^+}$ $-D_{e,p}^{\text{eff}} \frac{\partial C_e}{\partial x} \Big _{x=L^-} = 0$	$C_e = C_{0,e}$
Charge balance for electrolyte	$\frac{\partial}{\partial x} \left[-\kappa_i^{\text{eff}} \frac{\partial \phi_e}{\partial x} - \frac{2\kappa_i^{\text{eff}} RT}{F} (t_+^0 - 1) \frac{\partial \ln C_e}{\partial x} \right] - a_{s,i} F j_{f,i} = 0$ $i \in \{n, p\}$ $\frac{\partial}{\partial x} \left[-\kappa_s^{\text{eff}} \frac{\partial \phi_e}{\partial x} - \frac{2\kappa_s^{\text{eff}} RT}{F} (t_+^0 - 1) \frac{\partial \ln C_e}{\partial x} \right] = 0$	$i_e = -\kappa_i^{\text{eff}} \frac{\partial \phi_e}{\partial x} - \frac{2\kappa_i^{\text{eff}} RT}{F} (t_+^0 - 1) \frac{\partial \ln C_e}{\partial x} \Big _{x=0, L^-} = 0$ $i_e _{x=L_n^-} = i_e _{x=L_n^+}$ $i_e _{x=L_n+L_s^-} = i_e _{x=L_n+L_s^+}$	
Species conservation in electrodes	$\frac{\partial C_{p,i}}{\partial t} = D_{s,i} \left[\frac{\partial^2 C_{p,i}}{\partial r^2} + \frac{2}{r} \frac{\partial C_{p,i}}{\partial r} \right] \quad i \in \{n, p\}$	$\frac{\partial C_{p,i}}{\partial r} \Big _{r=0} = 0$ $-D_{s,i} \frac{\partial C_{p,i}}{\partial r} \Big _{r=R_i} = j_{f,i}$	$C_{p,i} = C_{0,i}$
Charge balance for electrodes	$\frac{\partial}{\partial x} \left[-\sigma_i^{\text{eff}} \frac{\partial \phi_{s,i}}{\partial x} \right] + a_{s,i} F j_{f,i} = 0, \quad i \in \{n, p\}$	$-\sigma_i^{\text{eff}} \frac{\partial \phi_{s,i}}{\partial x} \Big _{x=0, L} = I_{\text{app}}$ $\frac{\partial \phi_{s,i}}{\partial x} \Big _{x=L_n^-, L_n+L_s^+} = 0$	

The first term on the right-hand side term in Eq. (2) represents the irreversible heat of reaction, the second term is the reversible heat of reaction, and the last two terms are the ohmic heat in the electrode and the electrolyte, respectively.

The temperature dependence of various electrochemical properties $\xi \in \{D_s, D_e, \kappa\}$ are considered to follow the Arrhenius relationship [36] as

$$\xi(T) = \xi(T_{\text{ref}}) \exp \left[\frac{E_{a,\xi}}{R} \left(\frac{1}{T_{\text{ref}}} - \frac{1}{T} \right) \right], \quad (3)$$

where $E_{a,\xi}$ is the corresponding activation energy.

The instantaneous values of the electrochemical variables are used to determine the local heat generation (Eq. (2)), which is then homogenized using Eq. (4) to obtain the volume-averaged heat generation variable $\dot{Q}_{\text{vol}}$.

$$\dot{Q}_{\text{vol}} = \frac{1}{A_{\text{cell}} L_{\text{cell}}} \int_0^{L_{\text{cell}}} \dot{Q}_{\text{local}} A_{\text{cell}} dx \quad (4)$$

Table 3
Battery thermophysical properties.
Source: Taken from [46].

Parameter	Value	Description
$k_{\text{batt}}(\theta)$	60.9 W/(m K)	Battery thermal conductivity, angular
$k_{\text{batt}}(r)$	0.945 W/(m K)	Battery thermal conductivity, radial
ρ_{batt}	2664.3 kg/m ³	Battery density
$C_{p,\text{batt}}$	1081.8 J/(kg K)	Battery heat capacity

The homogenized form of the local energy balance equation for the cell gives [45]

$$\frac{\partial(\rho C_p^{\text{eff}} T)}{\partial t} = \nabla \cdot (k^{\text{eff}} \nabla T) + \dot{Q}_{\text{vol}} \quad (5)$$

The parameters listed in Table 3 are used to determine the effective properties of the cell mentioned in Eq. (5). The calculation of the

Table 4
Battery modelling parameters for electrode and electrolyte.

Parameter	Negative electrode	Positive electrode	Description
r_p	11.6×10^{-6} [m]	30×10^{-6} [m]	Particle radius
D_s^a	6.4×10^{-14} [m ² /s]	3.3×10^{-13} [m ² /s]	Solid phase Li-diffusivity
K_s^a	100 [S/m]	3.8 [S/m]	Solid phase conductivity
c_s	25500 [mol/m ³]	36248 [mol/m ³]	Solid phase concentration
$c_{s,0}$	20528 [mol/m ³]	8047.1 [mol/m ³]	Initial solid lithium concentration
L	80×10^{-6} [m]	56×10^{-6} [m]	electrode thickness
L_{cc}	1.1×10^{-5} [m]	2×10^{-5} [m]	Current collector thickness
t_+^0	0.363	0.363	Cationic transport number
D_l^a	6×10^{-11} [m ² /s]	6×10^{-11} [m ² /s]	Salt diffusivity in electrolyte
ε_s^a	0.63	0.77	Solid phase volume fraction
ε_l^a	0.27	0.23	Electrolyte phase volume fraction
ε_{filler}	$1 - \varepsilon_s - \varepsilon_l$	0	Filler volume fraction
c_0^a	2000 [mol/m ³]	2000 [mol/m ³]	Initial electrolyte salt concentration
k_i^a	2×10^{-11} [m/s]	2×10^{-11} [m/s]	Reaction rate coefficient
L_s	17×10^{-6} [m]		Separator thickness
d_{can}	0.25 [mm]		Thickness of battery canister
r_{batt}	9 [mm]		Battery radius
h_{batt}	65 [mm]		Battery height
$r_{mandrel}$	2 [mm]		Mandrel radius
L_{batt}	153×10^{-6} [m]		Cell thickness

^a Values obtained from Tranter et al. [46].

effective properties follows from a well-known procedure [46]:

$$\rho c_p^{eff} = \sum_i \varepsilon_i \rho_i c_{p,i} \quad (6)$$

$$k^{eff} = \sum_i \varepsilon_i k_i \quad (7)$$

Here, i is the cell's component, which includes the cathode, anode and separator.

The thermophysical and electrochemical properties utilized as inputs to solve the mathematical equations are detailed in Tables 3 and 4, along with geometric dimensions, including the electrode and separator thicknesses, characteristic particle sizes and the mandrel radius. These dimensions, including particle radii, electrode thicknesses, cell can dimensions, and cell height, were determined through a cell teardown study. Additional parameters, including solid-phase diffusivities and thermal properties listed in Table 3, were adapted from Tranter et al. [46] and subsequently optimized for our model, including adjustments to the electrode volume fraction using cell balancing and OCV profile.

2.3. Grid independence test

The assembly comprising the cell and the acrylic enclosure was modelled for numerical simulations, as illustrated in Fig. 4. A user-controlled meshing strategy was implemented, employing a mix of triangular and tetrahedral elements. Specifically, the entire domain was discretized with tetrahedral elements, while the inlet and outlet regions were meshed with triangular elements.

A grid independence test optimized the total number of elements utilized in the simulation. Initially, an adaptive coarse mesh was selected, featuring minimum and maximum element sizes of 0.576 cm and 2.74 cm, respectively, with an element growth rate of 1.6. Simulation results with two other progressively fine meshes with minimum (maximum) element sizes of 0.4 cm (2.16 cm) and 0.26 cm (1.44 cm) were compared with results from the coarser mesh. The results revealed that the percentage change in output variables, including discharge capacity, average discharge power, and energy was less than 0.15% relative to the fine mesh. Consequently, the fine mesh comprising 89,743 elements was selected for performing detailed simulations.

2.4. Design of experiments

The design of experiments (DOE) technique was employed to determine the conditions under which simulations were to be conducted,

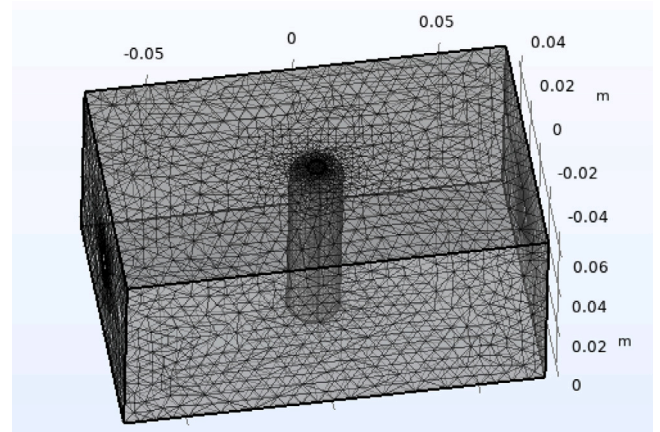


Fig. 4. Meshing of 3D thermo-electrochemical domain.

aiming to minimize the total number of such data points required for training a surrogate model [47,48]. Table 5 presents the minimum and maximum values of the design (i.e., input) variables that have been chosen as parameters for formulating surrogate models in this work. Two key techniques, namely central composite design (CCD) and Latin hypercube sampling (LHS), were utilized for selecting sampling points on this design space [49,50]. CCD, also known as the Box and Wilson design, strategically distributes samples across the edges, centre points of edges, and the centre of the entire design space. On the other hand, LHS is a constrained stratified sampling method that allocates a number of sample data points (denoted by N) with equal marginal probabilities (of $1/N$). In LHS, the distribution of samples can vary in terms of uniformity to ensure equal sampling precision across each dimension. This approach enhances accuracy and reduces variance [51,52].

After obtaining the model parameters using various material characterization techniques and literature, simulations were conducted on the selected DOE points accordingly, and the resulting output data was utilized to train reduced-order models.

2.5. Surrogate modelling

In formulating a surrogate model, sampling points are divided into training and test data sets. The training data sets are used to train the model, and after establishing the model, testing data sets are used

Table 5

The range of design variables used in the experiments and simulations.

Design variables	Minimum	Maximum
Discharge current	0.5C	4C
Inlet temperature	25 °C	45 °C
Dielectric fluid flow rate	200 ml/min	1000 ml/min

to analyse the model's accuracy [37,52]. Various types of surrogate models are available, such as polynomial response surface model, kriging, support vector regression, and radial basis neural network [52]. The present study incorporated the polynomial response surface (PRS) model.

The polynomial response surface models the predicted output $\hat{y}(\mathbf{x})$ in terms of the independent variables using a linear combination of polynomial basis function [37]:

$$\hat{y}(\mathbf{x}) = \beta_0 + \sum_{i=1}^n \beta_i x_i + \sum_{i=1}^n \beta_{ii} x_i^2 + \sum_{i=1}^{n-1} \sum_{j=i+1}^n \beta_{ij} x_i x_j + \dots \quad (8)$$

The model includes linear, quadratic, and higher-order interaction terms up to the desired degree. PRS models can handle the non-linear relationship between the input and output variables and measure the prediction uncertainty, which is useful for decision-making due to its ability to provide unbiased and minimum variance estimates.

This work has invoked several techniques to quantify the error in constructing surrogate models for various performance measures. A standard method is to quantify the prediction error sum of squares (PRESS). This method utilizes the training dataset exclusively, implementing the leave-one-out strategy to create multiple new models. Subsequently, each removed point is used as a test point to assess the model constructed by training on all the other data points.

$$\text{PRESS} = \left(\sqrt{\frac{\sum_{i=1}^N (y_i - \hat{y}_{(-i)})^2}{N}} \right) \times 100 \quad (9)$$

Here, N is the number of sampling points, y_i is the output vector for i th data, and $\hat{y}_{(-i)}$ is the estimated value of the output variable using the surrogate model constructed by leaving out the i th point.

Additionally, other evaluation techniques, such as the relative test point error (TPE), residual sum of squares (RSS), and root mean square error (RMSE) were integrated into the modelling process. RSS quantifies the difference between the observed output y_i and the predicted output $\hat{y}_i$ for N data points. RMSE represents the square root of the average of squared differences between the observed output y_i and the predicted output $\hat{y}_i$ for N data points. TPE is another measure of model accuracy in which test points that are not employed for training the surrogate model are used to assess the error in output prediction. The equations required for computing these error metrics are provided below [37].

$$\text{RSS} = \sum_{i=1}^N (y_i - \hat{y}_i)^2 \quad (10)$$

$$\text{RMSE} = \sqrt{\frac{1}{N} \sum_{i=1}^N (y_i - \hat{y}_i)^2} \quad (11)$$

$$\text{TPE}_i = \left| \frac{(y_i - \hat{y}_i)}{y_i} \right| \times 100 \quad (12)$$

2.6. Performance indicators

The temperature at mid-length of each lithium-ion cell was directly measured by a thermocouple interfaced with DAQ, whereas the other performance indicators such as discharge capacity, average discharge power and discharge energy were calculated using the instantaneous voltage and current data recorded by the battery cycler during galvanostatic discharging. The measures used to characterize the system

Table 6

Equations and descriptions for data analysis.

Performance indicators	Description
$Q_{\text{dch}} = \int_{t_0}^{t_1} I(t) dt$	Discharge capacity (Q_{dch}): Calculated using the coulomb counting method, representing the integral of discharge current $I(t)$ over the discharge cycle from time t_0 to t_1 .
$\bar{P}_{\text{dch}} = \frac{1}{\Delta t_d} \int_{t_0}^{t_1} V(t) I(t) dt$	Average discharge power ($\bar{P}_{\text{dch}}$): Obtained by time averaging the product of instantaneous voltage $V(t)$ and discharge current $I(t)$ over the discharge duration $\Delta t_d = t_1 - t_0$.
$E_{\text{dch}} = \bar{P}_{\text{dch}} \cdot \Delta t_d$	Discharge energy (E_{dch}): Calculated as the product of average discharge power $\bar{P}_{\text{dch}}$ and discharge duration Δt_d .

performance have been adopted from our previous publication [9]. The equations and descriptions in Table 6 describe the data analysis procedure. Q_{dch} refers to the discharge capacity of the commercial cell, while $\bar{P}_{\text{dch}}$ and E_{dch} denote its average discharge power and energy, respectively.

3. Results and discussion

Commercial battery packs are enclosed within a casing, insulated electrically and provided with mechanical protection. Without an active cooling mechanism, the convective heat transfer coefficient within the enclosure remains notably low. The discharge capacity of the cells depends significantly on the ambient temperature, showing an increase with increasing ambient temperature up to a specific threshold [53, 54]. However, elevated cell temperature poses a direct threat to both safety and performance. Consequently, the implementation of an active cooling system becomes imperative.

The outcomes of the present study are presented in this section and organized sequentially. The study begins with conducting experiments on commercial 18650-type lithium-ion cells under natural (i.e., free) convection cooling by air in an enclosure, followed by active immersion conditions. Subsequently, the implementation and validation of the thermo-electrochemical mathematical model are discussed. Further, the section covers the construction of the DOE and the execution of simulations on sampling points. Finally, surrogate-based data-driven methods are used to construct polynomial response surfaces for various cell performance indicators described in Table 6.

3.1. Experimental results

Fig. 5(a) depicts the voltage vs. discharge capacities for cells operating within an acrylic enclosure with air and in open ambient conditions for C-rates ranging from 1C to 4C. While the discharge capacity is relatively higher for cells being discharged inside the acrylic enclosure up to 2C, the trend reverses for 3C and 4C rates. This indicates that the lower rate of heat transfer from the cell to the surrounding medium has a significant effect on the voltage characteristics of the cell. This argument is supported by the variation in temperature rise shown in Fig. 5(b). This figure depicts that the cell temperature with acrylic chamber is slightly higher for 1C and 2C discharge rates when compared with open-air (i.e., unconfined) conditions. However, the gap widens at 3C and 4C rates, indicating a likelihood of acceleration in side reactions and deterioration in long-term cell performance. This underscores the necessity of an active cooling system, particularly at higher C rates, to mitigate safety concerns arising from uncontrolled temperature rise within the cells.

Fig. 6 shows a similar comparison between cooling by mineral oil or ambient air in the acrylic enclosure. Fig. 6(a) shows the cell voltage vs. discharge capacity variation for the cell with the mineral oil at 400 ml/min flow rate and inlet temperature of 25 °C and compares it with the data obtained from testing under open-air conditions. As can be

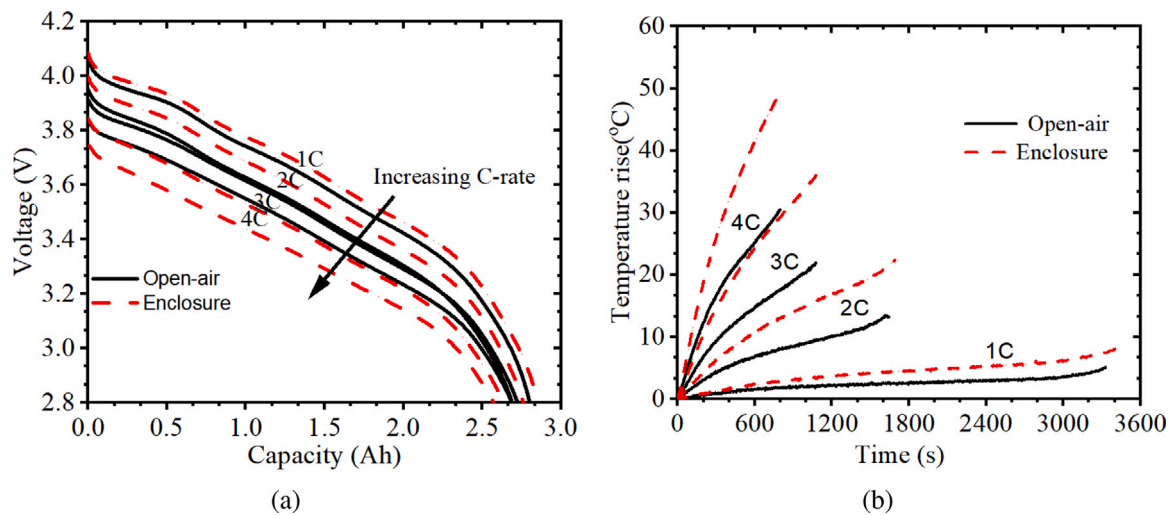


Fig. 5. A comparison of (a) voltage vs. discharge capacity, and (b) temperature rise vs. discharge time for cells with air as the coolant under free convection either within the acrylic enclosure or in open-air at 25 °C ambient temperature.

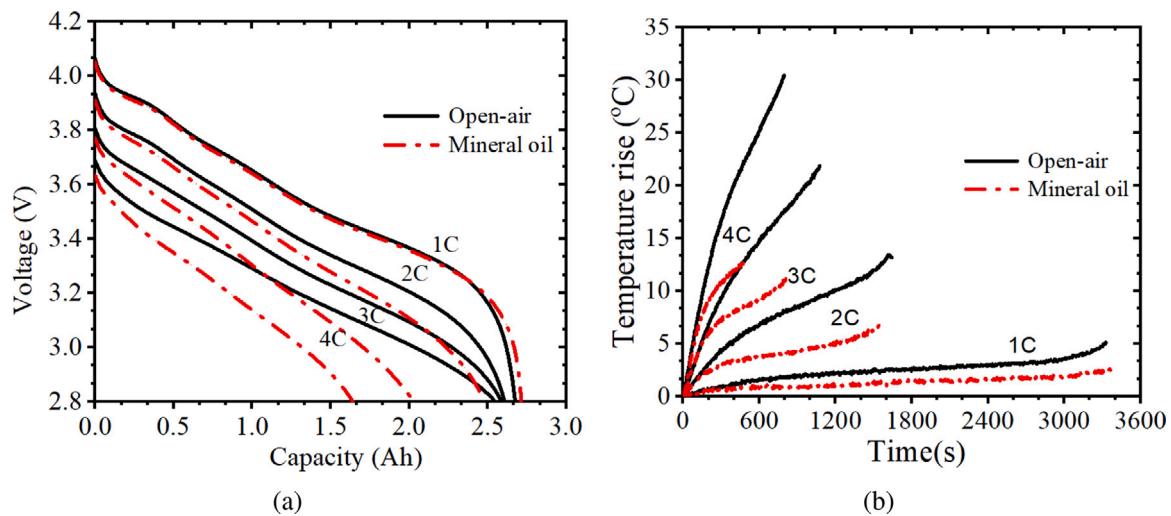


Fig. 6. A comparison of (a) voltage vs. discharge capacity, and (b) temperature rise vs. discharge time for cells operating inside the acrylic enclosure with mineral oil as the coolant against those in open-air at 25 °C ambient temperature.

observed, the discharge capacity is lower for mineral oil cooling when compared to that of the cell operating under ambient air conditions. This happens as mineral oil effectively suppresses temperature rise, which is shown in Fig. 6(b), resulting in slower reaction kinetics. This demonstrates the side-effect of the immersion-based cooling system, which in the present case limits the temperature rise to 14 °C at 4C rate. Further, there is a marginal difference in discharge capacity when comparing open-air cooling with mineral oil system at 1C discharge rate, which is promising for certain stationary storage or EV applications where operation below this rate may be desired.

3.2. Simulation results

The 1D electrochemical and 3D thermal and hydrodynamics models were coupled and implemented in COMSOL Multiphysics. Fig. 7 demonstrates a comparison of the simulation results with experimental measurements. Fig. 7(a) and (b) compare the cell voltage as a function of time obtained from simulations with experiments for air cooling by natural convection and mineral oil, respectively, within the enclosure. This comparison validates the electrochemical model employed in this

work. The thermal model was validated by comparing the temperature rise predicted by the simulations against the actual thermocouple measurements. As shown in Fig. 7(c) and (d), the temperature rise obtained from simulations is compared with measurements for air and mineral oil as surrounding fluids at 25 °C, respectively. These results reveal a good agreement between simulations and experiments. The match of experiments and simulation results for mineral oil is relatively better than that for air. This is because the difference between the maximum and average temperature with mineral oil is smaller than that with air, which is clear from Fig. 7(c) and (d).

Fig. 8(a) shows the temperature distribution within the cell and in the mineral oil at $t = 821$ s. The flow streamlines are also shown by solid lines with arrows representing the direction of flow. The spatial temperature variation within the cell is also apparent from Fig. 8(b). These simulations were conducted at 4C rate with a mineral oil inlet temperature of 25 °C and a flow rate of 200 ml/min. The temperature on the left surface of the cell is relatively higher than on the right surface, attributed to the existence of a fluid stagnation point on the left equatorial position on the cylinder surface, which inhibits heat removal by convection. Additionally, the abrupt expansion at the fluid

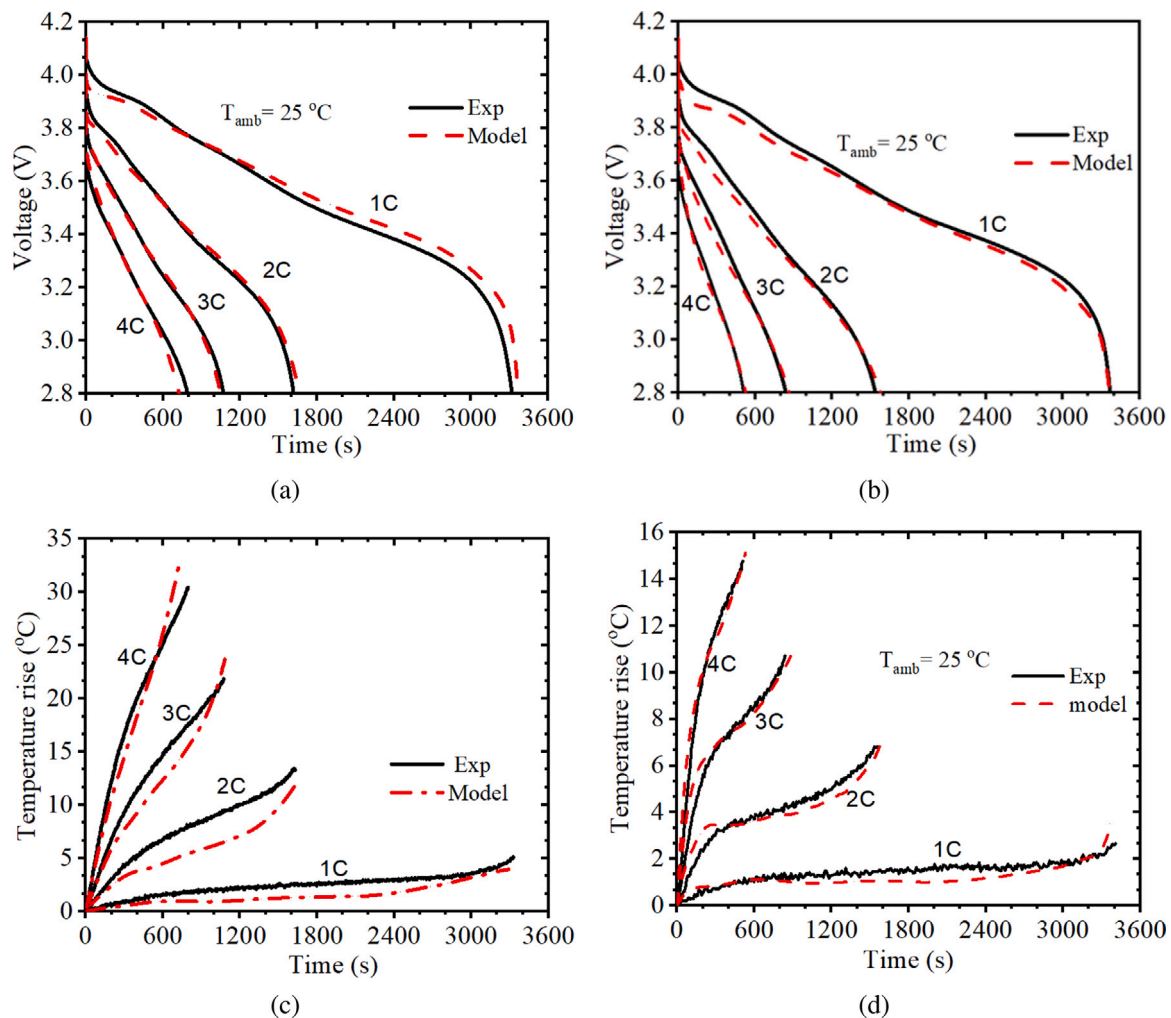


Fig. 7. Comparison of simulated cell voltage versus discharge capacity results with experimental data of (a) cells operating in open-air, (b) cells operating inside the acrylic enclosure with mineral oil. (c-d) shows the model validation against experimental data, showcasing the temperature versus discharge time for cells operating in the open air and within an acrylic enclosure filled with mineral oil.

inlet induces a negative pressure within the enclosure, forming a vortex in the cooling liquid within the chamber. The maximum temperature rise under these conditions was observed to surpass 32°C . A point to note is that the temperature rise for the flow rate of 1000 ml/min is 10.5°C only, which is consistent with the 10°C temperature rise reported by [55] under similar conditions. However, this is achieved at the cost of a decline in discharge capacity, as was shown in Fig. 6(a).

3.3. Surrogate model

The validated mathematical model was employed to generate training data for developing surrogate models for different performance indicators. Initially, fitting reduced-order models was attempted with 60 DOE points, which were divided into two subgroups of 54 training and 6 test points. These data, when used to train the surrogate model, resulted in PRESS and TPE $> 30\%$, implying that the training data was insufficient to achieve an accurate PRS model. Hence, the DOE was refined by adding more LHS points, and a total of 155 training points and 15 test points were deemed sufficient to achieve PRESS and TPE $< 5\%$. The thermophysical performance indicators are assessed using fourth-degree polynomial response surface models and the details are as follows.

3.3.1. Surface temperature estimation

A PRS surrogate model for cell temperature estimation was formulated with a PRESS value of 4.6%, a relative test point error (TPE) of 1.9%, and a root mean square error (RMSE) of 0.24. Fig. 9 shows the contours of cell temperature as a function of the discharge current and mineral oil temperature for three different flow rate conditions corresponding to (a) 200, (b) 600 and (c) 1000 ml/min , respectively, obtained from the estimates of this surrogate model. As is apparent, the highest temperature in the cell develops for 4C discharge rate and 45°C liquid inlet temperature. This also agrees with the fact that the surface temperature of a cell increases with C rate due to increased irreversible and reversible contributions to heat generation [5]. As will be shown soon, the rise in temperature directly affects the electrochemical kinetics and safety of the battery. An elevated cell temperature can enhance reaction kinetics, reduce electrolyte viscosity, and consequently lower ion transport resistance. This results in reduced internal resistance under given C rate and flow conditions.

Coolant flow rate is crucial in the thermal management of cells. Increasing the immersion fluid flow rate enhances convective heat transfer from the cell to the liquid, improving heat dissipation and reducing cell temperature. In Fig. 9(a)–9(c), it is evident that increasing the flow rate is not an effective solution to suppress the temperature below a certain value. In a comparison of Fig. 9(a) and (b), the increase

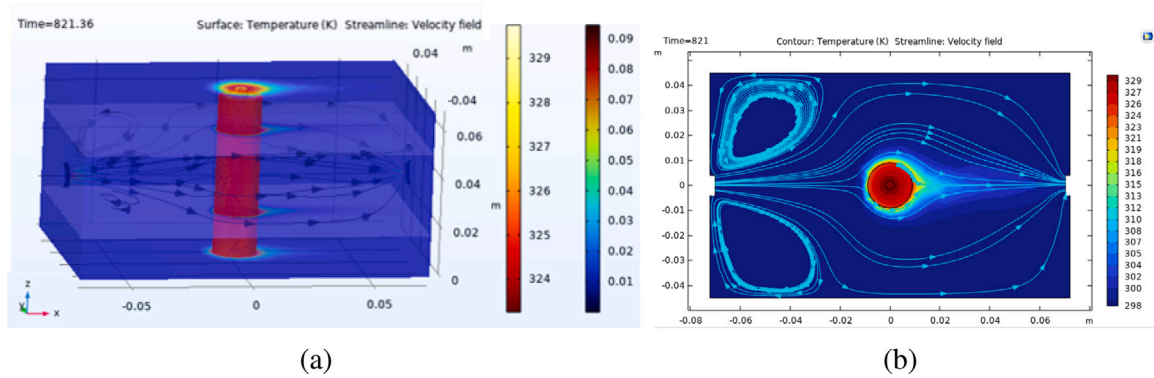


Fig. 8. Temperature distribution visualization in (a) the entire domain, and (b) a plane cut through the mid-section of the cell. The solid lines represent the flow streamlines, with arrows indicating the direction of fluid flow.

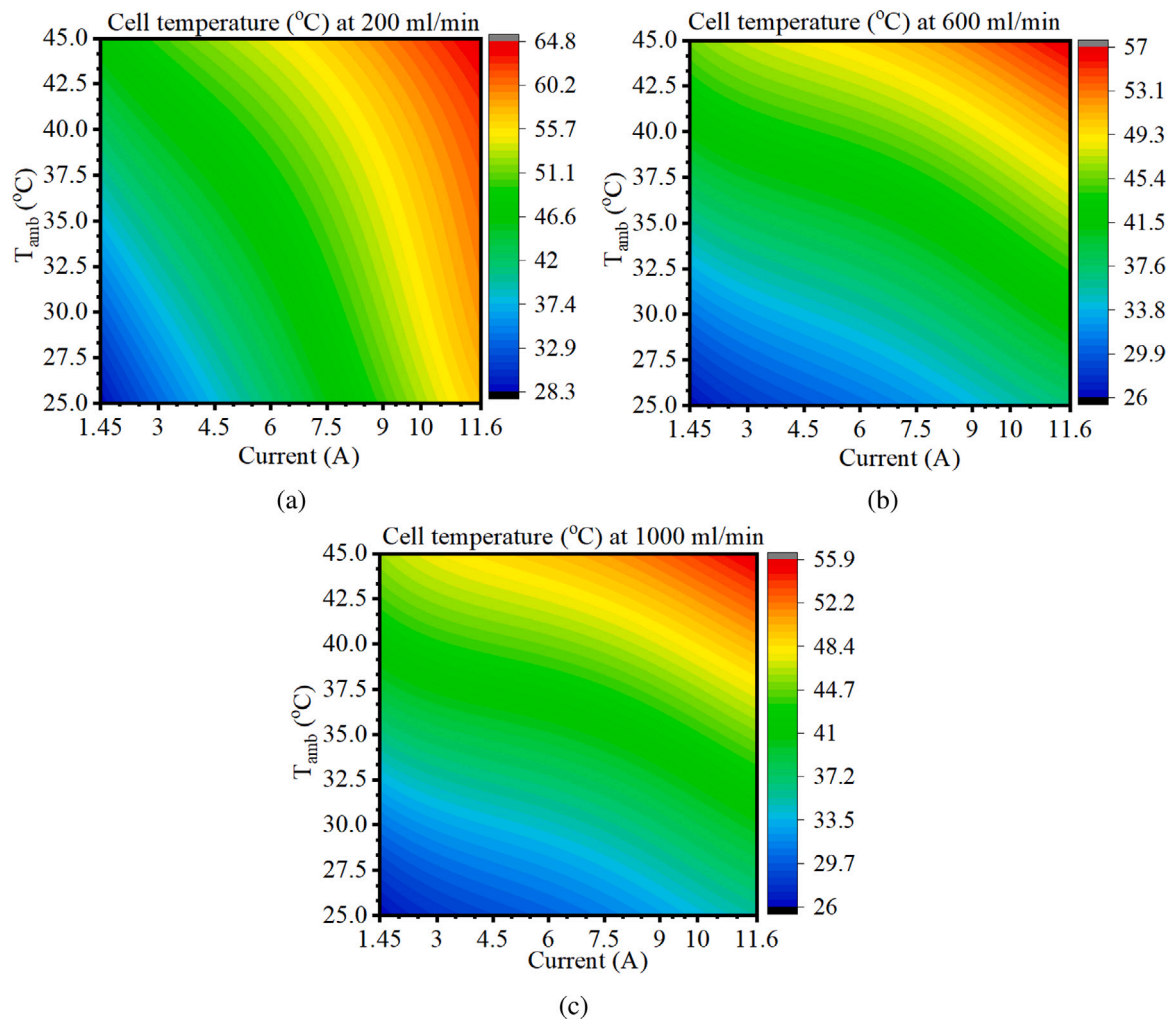


Fig. 9. Contour plots of cell temperatures at (a) 200 ml/min, (b) 600 ml/min, and (c) 1000 ml/min.

in flow rate significantly reduces the cell temperature from 65 °C to 57 °C due to increased heat dissipation. However, comparing Fig. 9(b) and (c), it is clear that a further increase in fluid flow rate has a marginal effect on the cell temperature. This may have happened because the heat transfer rate has reached saturation. We observed that the 600 ml/min flow rate is optimal and that there is an insignificant benefit towards reducing the cell temperature by increasing it any further.

3.3.2. Discharge capacity estimation

The discharge rate, inlet fluid temperature, and flow rate directly influence the discharge capacity of a battery. Using the training dataset, a surrogate model was developed to estimate discharge capacity as a function of these design variables, which had a PRESS value of 4.7% and a relative TPE of 0.35%. Using this model, the variation of discharge capacity with discharge rate and mineral oil inlet temperature is shown in Fig. 10 for three different flow rates. This figure shows

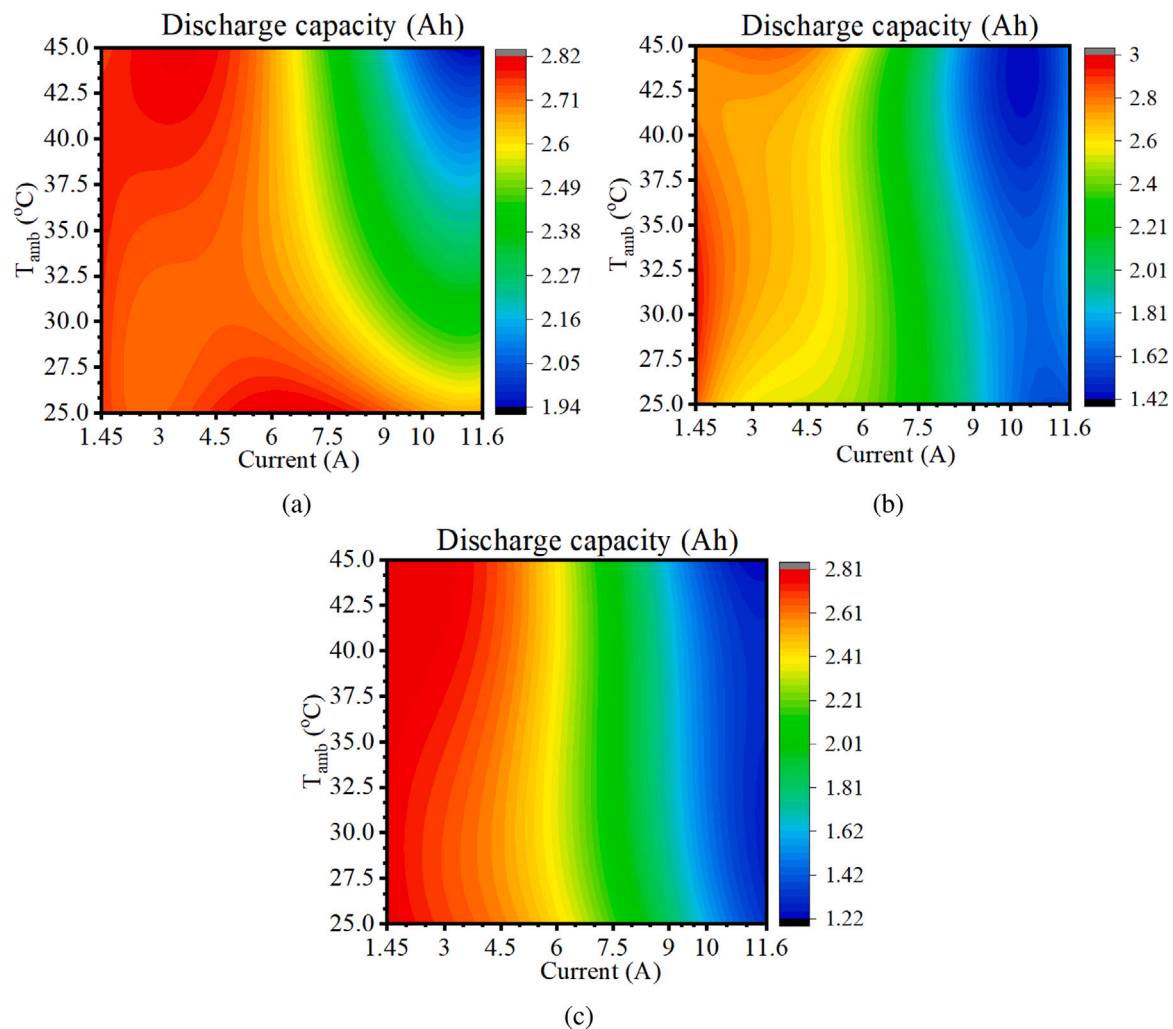


Fig. 10. Contour plots of cell discharge capacity at (a) 200 ml/min, (b) 600 ml/min, and (c) 1000 ml/min.

that the C-rate and fluid flow rate are the primary factors affecting cell capacity. An increase in the C-rate results in higher overpotential and hence a reduced voltage operating window (apparent from the voltage drop shown in Fig. 5(a)). The inlet fluid temperature also directly affects the overpotential as the activation overpotential is relatively lower for a higher temperature. This results from faster reaction kinetics and ion transport rates with increasing temperatures, leading to higher discharge capacity.

As shown in Fig. 10, for a given discharge rate and inlet temperature, the discharge capacity of the battery varies with flow rate quite substantially. At a low discharge rate, such as 0.5C ($= 1.45$ A), the effect of the fluid temperature and the fluid flow rate is not apparent, with a capacity of ≈ 2.8 Ah being achieved. On the other hand, the lowest achievable capacity happens to be for the high discharge rate and the highest fluid temperature considered in this work. However, the difference is that the minimum achievable discharge capacity decreases with an increase in the fluid flow rate, as can be observed by comparing the lower bounds in Fig. 10(a)–(c). Based on the results of the surrogate model, it is inferred that more than 70% of the nominal discharge capacity can be achieved under galvanostatic conditions when operating these cells up to 3C rate with any choice of the liquid flow rate.

3.3.3. Average discharge power estimation

The average discharge power is quantified by the product of discharge current and operating voltage, averaged over the total discharge period. In an earlier study, it was shown that this metric is predominantly influenced by the discharge current, as variations in current

directly affect the power output [9].

The training data points were used to formulate a suitable surrogate model for average discharge power, and the model exhibited a PRESS and TPE of 4.2% and 1.8%, respectively. Using this model, contours of constant discharge power have been plotted and shown in Fig. 11 for different flow rates. The figures show that in contrast to cell temperature and discharge capacity, the dielectric liquid temperature and flow rate have minimal impact on the average discharge power. The discharge power demonstrates an approximately linear relationship with discharge current, regardless of variations in flow rate and temperature, as depicted in Fig. 11(a)–(c). The maximum average discharge power stabilizes around 41 W at 4C rate, independent of the inlet temperature and flow rate conditions.

3.3.4. Discharge energy estimation

The discharge energy quantified in this work represents the total energy delivered by the cell under the galvanostatic discharge operation. The simulation data was used to train a reduced-order surrogate model to estimate the discharge energy, with the PRESS and TPE values of 6% and 5.8%, respectively. Fig. 12 shows that the three design variables (inlet fluid flow rate, inlet temperature and discharge rate) have a significant influence over the discharge energy, a behaviour similar to discharge capacity. This is in contrast to the average discharge power which depends primarily on the discharge rate. The discharge energy is higher for low discharge rates, irrespective of the choice of the liquid temperature or flow rate. The discharge energy is also lowest when

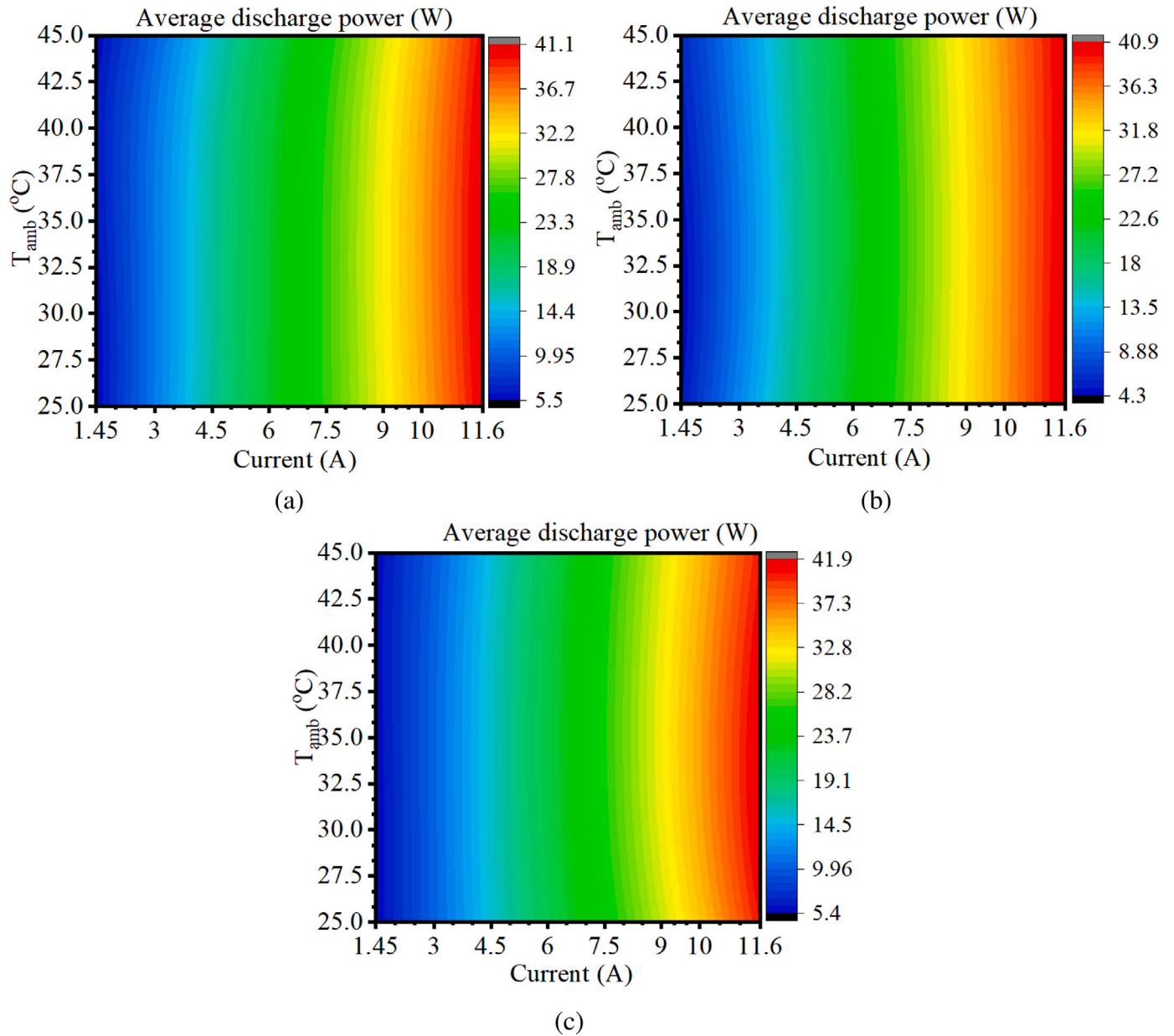


Fig. 11. Contour plot of cells average discharge power at (a) 200 ml/min, (b) 600 ml/min, and (c) 1000 ml/min.

operating the cell at high C-rates. However, the discharge energy shows a significant drop when the flow rate is increased from 200 ml/min to 600 ml/min, particularly at discharge rate more than 1C (≈ 2.8 A). Moreover, Fig. 12(c) shows that the discharge energy increases as the temperature of the liquid coolant increases for discharge rate > 10 A and flow rate of 1000 ml/min.

The temperature distribution at the mid-plane of the cell at $t = 850$ s at a current of 10.5 A, a coolant temperature of 42.5°C , and flow rate of 1000 ml/min is shown in Fig. 13(a). This figure shows that the temperature variation within the cell is of $O(10^{\circ}\text{C})$. Fig. 13(b) shows the variation of outward heat flux from the cell to the liquid and the maximum temperature within the cell as a function of time for two different liquid temperatures and the same flow rate. It is evident that the outward heat flux is notably higher at the relatively lower coolant temperature, despite the convective heat transfer coefficient being approximately similar for both cases. The higher heat flux at 28°C results in a reduction of the cell temperature, leading to slower electrochemical kinetics, an increase in internal resistance, and a correspondingly higher heat generation rate. Consequently, this

effect reduces the total discharge duration and, therefore, lowers the discharge energy of the cell at lower coolant temperatures.

4. Conclusions

This study on immersion cooling explored the role of discharge rate, inlet fluid temperature, and dielectric liquid flow rate on the thermo-electrochemical performance of commercial 18650-type lithium-ion cells to identify an optimal balance between performance and safety. Cell testing for discharge rates between 1C-4C and standard air cooling in a confined enclosure revealed a substantial increase in cell temperature, necessitating an active cooling system. An immersion-based cooling technique was chosen due to its relatively higher convective heat transfer coefficient and capability to enforce a uniform temperature distribution within the cell. Experiments were performed with the cell placed inside an acrylic enclosure filled with mineral oil and whose flow rate could be controlled using a pump. Cells were discharged at different rates, and a comparative analysis was conducted between cells operating under ambient air and immersion cooling conditions. The

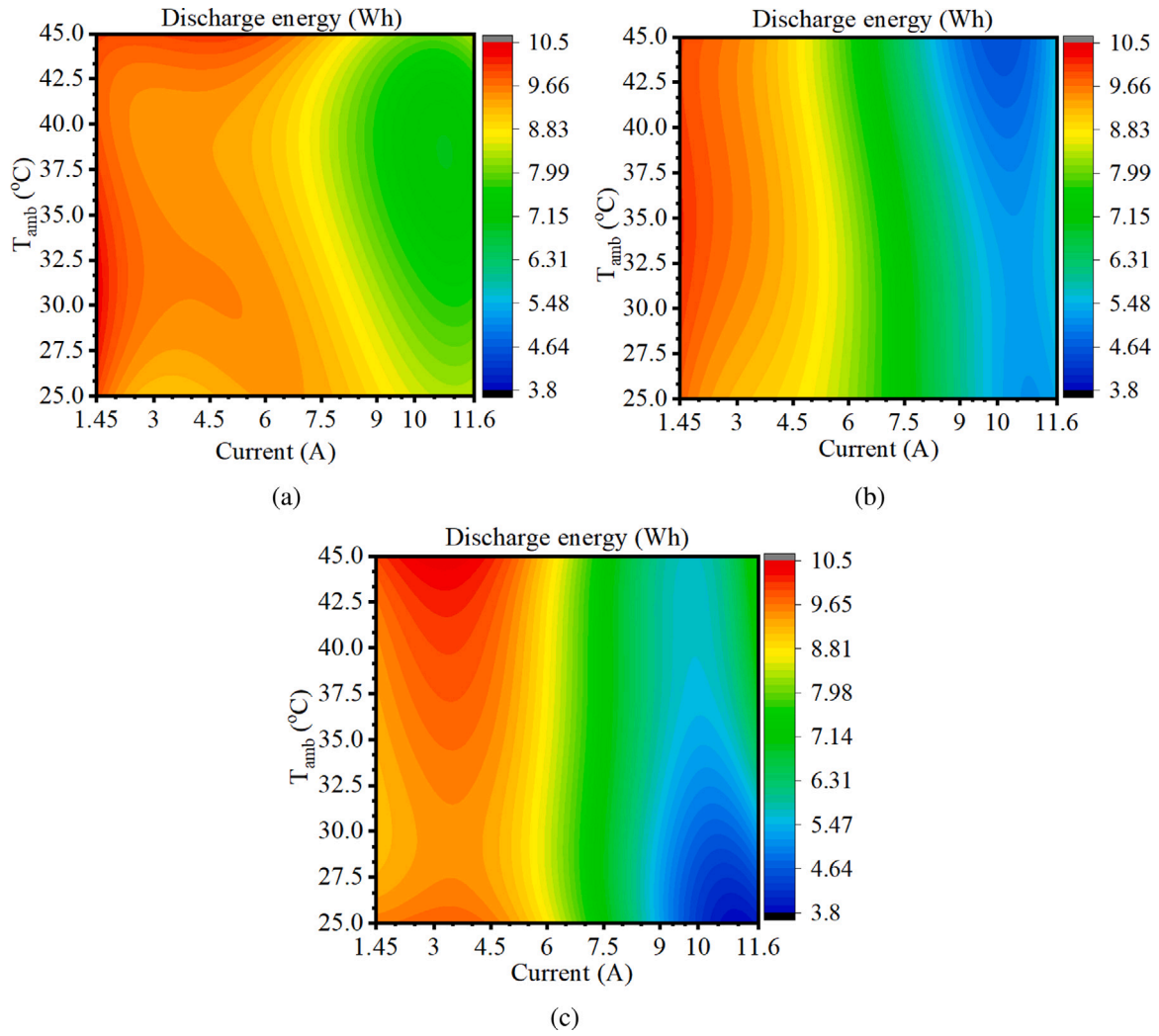


Fig. 12. Contour plot of cells discharge energy at (a) 200 ml/min, (b) 600 ml/min, and (c) 1000 ml/min.

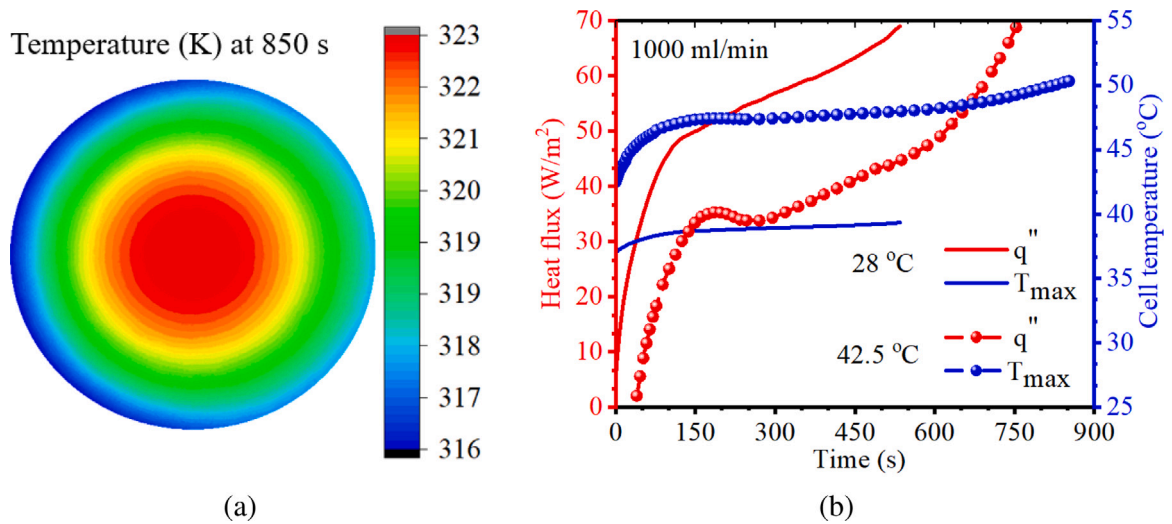


Fig. 13. (a) Mid-section temperature variation within the cell at inlet flow rate of 1000 ml/min, discharge rate of 10.5 A and coolant temperature of 42.5 °C at the end of discharge. (b) Temporal variation of heat flux from the cell and the cell temperature for 10.5 A and 1000 ml/min for inlet fluid temperatures of 28 °C and 42.5 °C.

performance metrics that were compared included cell temperature, discharge capacity, average discharge power, and average discharge energy. Our experiments demonstrate that an active cooling system effectively reduces temperature rise, ensures a lower difference between maximum and average temperature within the cell, and enhances safety. However, an increased cooling liquid flow rate also leads to lowered capacity and discharge energy compared to those achieved under free convection in air.

Since experiments are inherently time-consuming, we relied on mathematical modelling of the physics governing cell behaviour and heat transfer between the two media to describe the effect of varying operating parameters on cell performance. A design of experiments approach was followed to determine the conditions under which simulations were to be conducted, aiming to minimize the total number of such data points required for training the surrogate model. The physics-based model was developed and validated against experimental results. Subsequently, simulations were performed for conditions required by DOE sampling points to generate a dataset for the training of surrogate models that could enable a better understanding of the combined influence of input operating conditions on performance metrics.

The inlet fluid temperature had a significant influence on cell performance by affecting electrochemical reaction kinetics and ion transport rate. The flow rate also inversely affects the discharge capacity and discharge energy, while its effect on discharge power is insignificant. This implies that immersion cooling will have the most impact in power-intensive applications where the cell temperature has to be maintained within 60 °C.

The discharge rate, on the other hand, significantly affects total overpotential, and the operating voltage window narrows with an increase in this operational variable, resulting in lower discharge capacity and energy. A close examination of the performance indicators used in this study revealed that a discharge rate of less than 3C can provide up to 70% of discharge capacity with any temperature or flow rate of the immersion liquid.

This work was an attempt to provide a deeper understanding of the trade-off between thermal safety and cell performance for immersion cooling-based thermal management systems. Future work could involve studying the impact of phase-change in the immersion liquid, extending the system to conditions when heating is required under subzero temperatures, and long-term cycling assessment of cell performance under immersion cooling.

Nomenclature

DDM	data driven modelling
DOE	design of experiments
LHS	latin hypercube sampling
LIBs	lithium-ion batteries
PRESS	prediction error sum of squares
ROM	reduced order modelling
TPE	training point error

Symbols

$a_{s,i}$	Specific interfacial area for an electrode i (m^2/m^3)
C_e	Lithium concentration in the electrolyte (mol/m^3)
$C_{p,i}$	Lithium concentration in an electrode particle i (mol/m^3)
$C_{p,\text{batt}}$	Battery heat capacity ($\text{J}/(\text{kg K})$)
$C_{s,i}$	Lithium concentration on the surface of electrode particle i (mol/m^3)

D_e	Lithium diffusion coefficient in electrolyte (m^2/s)
$D_{s,i}$	Lithium diffusion coefficient in electrode i (m^2/s)
E_{dch}	discharge energy (Wh)
$E_{a,i}$	Activation energy for property i (J/mol)
F	Faraday's constant (C/mol)
I_{app}	Applied current density (A/m^2)
$j_{f,i}$	Interfacial molar flux density for lithium intercalation/deintercalation reaction at electrode i ($\text{mol}/\text{m}^2 \text{ s}$)
k_i	Rate constant for lithium intercalation/deintercalation reaction (m^2/s)
$k_{\text{batt}}(\theta)$	Battery thermal conductivity, angular ($\text{W}/(\text{m K})$)
$k_{\text{batt}}(r)$	Battery thermal conductivity, radial ($\text{W}/(\text{m K})$)
k^{eff}	Effective thermal conductivity of domain i ($\text{W}/\text{m K}$)
L_i	Thickness of cell component i (m)
P_{dch}	discharge power (W)
$\bar{P}_{dch}$	time-averaged discharge power of cell (W)
$\dot{Q}_{vol}$	Volume-averaged heat generation (W/m^3)
$\dot{Q}_{local}$	Heat generation rate per unit volume within the cell (W/m^3)
Q_{dch}	discharge capacity (Ah)
R	Universal gas constant ($\text{J}/\text{mol K}$)
T	Temperature (K)
T_{amb}	Inlet fluid/ambient temperature (°C)
t	Time (s)
t_+^0	Cation transfer coefficient
U_i	Open-circuit potential in electrode i (V)
V	Cell voltage (V)

Greek symbols

α_a, α_c	Anodic/cathodic charge transfer coefficients
$e_{e,i}$	Electrolyte volume fraction in electrode i
$e_{s,i}$	Active material volume fraction in electrode i
η_i	Surface overpotential for lithium intercalation/deintercalation reaction in electrode i (V)
κ	Ionic conductivity of the electrolyte (S/m)
ϕ_e	Ionic potential in the electrolyte phase (V)
ϕ_s	Electrical potential in electrode i (V)
ρ_i	Density of domain i (kg/m^3)
ρ_{batt}	Battery density (kg/m^3)
σ_i	Electrical conductivity for electrode i (S/m)

Subscripts

amb	Ambient
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<i>e</i>	Electrolyte
<i>dch</i>	discharge
<i>n</i>	Negative electrode
<i>p</i>	Positive electrode
<i>s</i>	Separator
<i>ref</i>	Reference value

Superscripts

0	Initial condition
avg	Average value
eff	Effective property

CRedit authorship contribution statement

Raghvendra Gupta: Writing – original draft, Visualization, Validation, Software, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Supreet Singh Bahga:** Writing – review & editing, Supervision, Resources, Conceptualization. **Amit Gupta:** Writing – review & editing, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Amit Gupta reports financial support was provided by India Ministry of Science & Technology Department of Science and Technology. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

Data will be made available on request.

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