

**COMPARATIVE THERMO-ELECTROCHEMICAL
INVESTIGATION OF COOLING STRATEGIES FOR
COMMERCIAL LITHIUM-ION CELLS**

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COMMERCIAL LITHIUM-ION CELLS**

by

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submitted

in fulfillment of the requirements of the degree of

Doctor of Philosophy

to the



INDIAN INSTITUTE OF TECHNOLOGY DELHI

March, 2025

I would like to dedicate this thesis to my loving family . . .

CERTIFICATE

This is to certify that the thesis entitled “**Comparative thermo-electrochemical investigation of cooling strategies for commercial lithium-ion cells**”, submitted by **Raghvendra Gupta** to the Indian Institute of Technology Delhi, for the award of the degree of **Doctor of Philosophy** is a record of the original, bonafide research work carried out by him under our supervision and guidance. The thesis has reached the standards fulfilling the requirements of the regulations related to the award of the degree.

The results contained in this thesis have not been submitted in part or in full to any other University or Institute for the award of any degree or diploma to the best of our knowledge.

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Raghvendra Gupta

ABSTRACT

Lithium-ion batteries are central to the rapid development of energy storage technologies due to their superior energy density, long cycle life, and efficiency. However, understanding their thermal behaviour and optimizing cooling mechanisms remain critical for enhancing performance and ensuring operational safety, especially under high C-rates and varying ambient conditions. This thesis employs experimental and modelling approaches to investigate the thermo-electrochemical behaviour of commercial 18650 Lithium-ion cells, focusing on discharge characteristics, thermal effects, and cooling techniques.

Experiments were performed on a cylindrical cell of nominal capacity of 2.9 Ah to characterize its performance in air with ambient temperatures ranging from 25 °C to 45 °C. These studies were carried out for a range of discharge rates (1C–4C). Measurements revealed that the cell surface temperature is highly sensitive to ambient conditions. Low ambient temperatures cause a disproportionate increase in surface temperature, likely due to the low ionic conductivity of the lithium-salt-based electrolyte. The discharge capacity was observed to be higher at elevated temperatures (at 45°C), with a maximum of 10% and a minimum of 46% (at 0°C) of the rated capacity at low temperatures to the cell's rated capacity at 0.5C rate. The discharge rate primarily influenced the discharge power, while the ambient temperature had a negligible impact on average discharge power within a single discharge cycle. In contrast, the discharge energy was strongly affected by both the discharge current and the ambient temperature.

An immersion-based cooling technique using mineral oil was considered to address the thermal challenges in the cells. The use of mineral oil was considered because of its high convective heat transfer coefficient and the possibility of ensuring greater uniformity in cell temperature. Comparative analysis with natural air cooling demonstrated that immersion cooling effectively reduced temperature increase and maintained cell temperatures within the safety limit of 50 °C. However, higher flow rates of the dielectric fluid slightly reduced discharge ca-

capacity and energy, indicating a trade-off between thermal management and the electrochemical performance of lithium-ion cells.

To attain a deeper understanding of heat diffusion within the cell and subsequent advection into the dielectric liquid, a physics-based model was developed. Simulations were performed to analyze cell behaviour under varying discharge rates and cooling conditions. The model incorporated detailed information obtained from cell tear-down analyses and was validated against experimental results. Subsequently, a low-fidelity, surrogate-based, reduced-order model was trained using datasets generated from the design of experiments (DOE) sampling points. The surrogate model efficiently explored the combined effects of discharge rate, ambient temperature, inlet fluid temperature, and flow rate on performance metrics. The findings indicated that the discharge rate significantly affects polarization, narrowing the operating voltage window and reducing discharge capacity and energy. The study revealed that at least 70% of the nominal discharge capacity can be delivered under galvanostatic conditions when operating cells up to 3C rate, irrespective of the liquid flow rate. The ambient temperature primarily influenced cell temperature, discharge capacity, and energy, while its effect on discharge power was minimal.

Sensitivity analyses revealed that immersion cooling is most effective in power-intensive applications, where maintaining the cell temperature within safe limits is critical. The flow rate of the cooling fluid inversely affected discharge capacity and energy, while its impact on power output was negligible. Moreover, response surface analyses provided insights into optimal operating conditions for achieving balanced performance and safety.

This research incorporated experimental investigations, physics-based simulations, and surrogate modelling to comprehensively understand the interplay between operational parameters, thermal behaviour, and performance in lithium-ion cells. The findings contribute to developing cell cooling strategies, enabling enhanced performance and safety across diverse applications reliant on lithium-ion technology.

लिथियम-आयन बैटरियां अपनी उच्च ऊर्जा घनत्व, लंबी चक्र आयु, और दक्षता के कारण ऊर्जा भंडारण प्रौद्योगिकियों के विकास में केंद्रीय भूमिका निभाती हैं। हालांकि, उनके तापीय व्यवहार को समझना और शीतलन तंत्र को अनुकूलित करना उच्च C-रेट और विभिन्न परिवेशी स्थितियों के तहत प्रदर्शन को बढ़ाने और संचालन सुरक्षा सुनिश्चित करने के लिए अत्यंत महत्वपूर्ण है। यह शोध प्रबंध वाणिज्यिक 18650 लिथियम-आयन सेल की थर्मो-इलेक्ट्रोकेमिकल प्रतिक्रिया की जांच करता है, जिसमें डिस्चार्ज विशेषताएँ, तापमान प्रभाव, और शीतलन तकनीकों का अध्ययन शामिल है, जो प्रायोगिक और मॉडलिंग दृष्टिकोण दोनों का उपयोग करता है। प्रायोगिक अध्ययन 2.9 Ah की नाममात्र क्षमता वाले एक बेलनाकार 18650 सेल पर किए गए, ताकि विभिन्न डिस्चार्ज करंट (1C-4C) और परिवेशीय तापमान (25°C-45°C) के तहत इसके प्रदर्शन का विश्लेषण किया जा सके। परिणामों से पता चला कि सतह का तापमान परिवेशीय स्थितियों के प्रति अत्यधिक संवेदनशील है, जिसमें कम परिवेशीय तापमान लिथियम-सॉल्ट आधारित इलेक्ट्रोलाइट में आयनिक चालकता में कमी के कारण सतह के तापमान में असमान वृद्धि का कारण बनता है। अध्ययन की गई स्थितियों में सतह तापमान में अधिकतम और न्यूनतम वृद्धि क्रमशः 16.05°C और 1°C पाई गई। डिस्चार्ज क्षमता उच्च तापमान पर अधिक पाई गई, जिसमें अधिकतम 100% और न्यूनतम 46% रेटेड क्षमता दर्ज की गई। डिस्चार्ज पावर मुख्य रूप से डिस्चार्ज रेट से प्रभावित हुई, जबकि परिवेशीय तापमान का प्रभाव एक चक्र के भीतर नगण्य पाया गया। दूसरी ओर, डिस्चार्ज ऊर्जा डिस्चार्ज करंट और परिवेशीय तापमान दोनों से अत्यधिक प्रभावित हुई, जिसमें अधिकतम 50% और न्यूनतम 20% नाममात्र ऊर्जा दर्ज की गई। थर्मल चुनौतियों का समाधान करने के लिए खनिज तेल का उपयोग करके एक इमर्शन-आधारित शीतलन तकनीक अपनाई गई, जिसे उच्च संवहन ताप हस्तांतरण गुणांक और समान तापमान वितरण बनाए रखने की क्षमता के कारण चुना गया। प्राकृतिक वायु शीतलन के साथ तुलनात्मक विश्लेषण से पता चला कि इमर्शन शीतलन प्रभावी रूप से तापमान वृद्धि को कम करता है और सेल के तापमान को 60°C की सुरक्षा सीमा के भीतर बनाए रखता है। हालांकि, शीतलन तरल के उच्च प्रवाह दरों ने डिस्चार्ज क्षमता और ऊर्जा को थोड़ा कम कर दिया, जो थर्मल प्रबंधन और इलेक्ट्रोकेमिकल प्रदर्शन के बीच समझौते को दर्शाता है। सेल के व्यवहार का अनुकरण करने के लिए एक भौतिक-आधारित मॉडल विकसित किया गया, जो विभिन्न डिस्चार्ज दरों और शीतलन परिस्थितियों के तहत कार्य करता है। इस मॉडल में सेल के टियरडाउन विश्लेषण से प्राप्त विस्तृत जानकारी को शामिल किया गया और प्रायोगिक परिणामों के विरुद्ध मान्य किया गया। इसके बाद, डिज़ाइन ऑफ़ एक्सपेरिमेंट्स (DOE) सैपलिंग पॉइंट्स से उत्पन्न डेटा सेट का उपयोग करके सरोगेट-आधारित मॉडल को प्रशिक्षित किया गया। यह मॉडल इनपुट परिचालन परिस्थितियों के संयुक्त प्रभाव को समझने के लिए दक्षतापूर्वक उपयोग किया गया। निष्कर्षों से पता चला कि डिस्चार्ज रेट ओवरपोटेंशियल को महत्वपूर्ण रूप से प्रभावित करता है, ऑपरेटिंग वोल्टेज विंडो को संकीर्ण करता है और डिस्चार्ज क्षमता और ऊर्जा को कम करता है। संवेदनशीलता विश्लेषण से पता चला कि इमर्शन शीतलन पावर-गहन अनुप्रयोगों में सबसे प्रभावी है, जहां सेल के तापमान को सुरक्षित सीमा के भीतर बनाए रखना महत्वपूर्ण है। शीतलन तरल के प्रवाह दर का डिस्चार्ज क्षमता और ऊर्जा पर विपरीत प्रभाव पड़ता है, जबकि इसका पावर आउटपुट पर प्रभाव नगण्य होता है। इसके अलावा, प्रतिक्रिया सतह विश्लेषण ने प्रदर्शन और सुरक्षा को संतुलित करने के लिए अनुकूल परिचालन

स्थितियों में अंतर्दृष्टि प्रदान की। यह शोध प्रबंध प्रायोगिक जांच, भौतिक-आधारित सिमुलेशन, और सरोगेट मॉडलिंग को एकीकृत करता है, ताकि परिचालन मापदंडों, थर्मल व्यवहार, और प्रदर्शन के बीच अंतःक्रिया की व्यापक समझ प्रदान की जा सके। निष्कर्ष लिथियम-आयन प्रौद्योगिकी पर निर्भर विविध अनुप्रयोगों के लिए उन्नत बैटरी प्रबंधन रणनीतियों के विकास में योगदान करते हैं।

CONTENTS

CERTIFICATE	i
ACKNOWLEDGEMENTS	ii
ABSTRACT	iii
CONTENTS	vii
List of Figures	x
List of Tables	xiii
Nomenclature	xiv
1 Introduction	1
1.1 Background	1
1.1.1 Electrochemical energy storage devices	2
1.1.2 Historical perspective of battery	6
1.1.3 Lithium-ion based battery technology	7
1.1.4 Categorization of lithium-ion batteries	8
1.2 Motivation	11
1.3 Objectives	13
1.4 Outline of the thesis	14
2 Literature review	16
2.1 Introduction	16
2.2 Experimental studies	19
2.3 Different degradation mechanism in Li-ion cells	20
2.4 Thermo-electrochemical models	21

2.4.1	Pseudo-2D models	21
2.4.2	Thermal models	22
2.5	Energy conservation models	23
2.6	Local heat generation models	25
2.7	Cooling strategies for lithium-ion batteries	28
2.7.1	Water/glycol	30
2.7.2	Nanofluids	31
2.7.3	Dielectric fluids	32
2.8	Data-driven modelling	33
2.9	Conclusions from literature review	37
3	Methodology	38
3.1	Introduction	38
3.2	Experimental setup for cell testing	39
3.2.1	Immersion cooling system	41
3.3	Thermo-electrochemical modelling	42
3.3.1	Physics based modelling	43
3.4	Design of experiments	50
3.4.1	Design space and sampling techniques	50
3.4.2	Central Composite Design (CCD)	51
3.4.3	Latin Hypercube Sampling (LHS)	51
3.5	Surrogate modelling	52
3.5.1	Polynomial response surface model	52
3.5.2	Kriging	53
3.5.3	Sensitivity analysis	55
3.5.4	Methods for data analysis	56
4	Thermo-electrochemical behaviour: Temperature and discharge current effects	58
4.1	Introduction	58

4.2	Results and discussion	60
4.2.1	Core and surface temperature measurement and estimation	62
4.2.2	Discharge capacity estimation	65
4.2.3	Average discharge power estimation	66
4.2.4	Discharge energy estimation	68
4.2.5	Sensitivity analysis	69
4.3	Conclusions	71
5	Comparative study of immersion vs. air cooling for Lithium-ion cells	73
5.1	Introduction	73
5.2	Results and discussion	74
5.2.1	Experimental results	75
5.2.2	Simulation results	77
5.2.3	Surrogate model	79
5.3	Conclusions	87
6	Conclusions and future work	89
6.1	Conclusions	89
6.1.1	Thermo-electrochemical behaviour and heat generation	89
6.1.2	Impact of cooling and fluid flow rate	90
6.2	Future scope of work	91
	References	93
	List of Publications	110
	Biodata of the Author	114

List of Figures

1.1	Electrochemical energy storage systems	4
1.2	Working of electrochemical system	5
1.3	Specific energy comparison of different chemistries	8
1.4	Different applications of lithium-ion batteries	9
1.5	Types of cells by their form factors	10
1.6	Working of lithium-ion cells	10
2.1	Different lithium-ion batteries models.	18
2.2	Classification of Cooling System Strategies for Lithium-ion Batteries	29
2.3	Various cooling system strategies for thermal management of lithium-ion batteries.	30
3.1	CT scan image of unprobed cell from (a) top and (b) front, and probed cell from (c) top (d) front.	39
3.2	Working of lithium-ion cells	40
3.3	Immersion-based cooling system setup (a) schematic and (b) cooling chamber connections.	42
3.4	Pseudo-2D representation of the lithium-ion cell.	43
3.5	EDX image of NMC electrode.	47
3.6	FESEM images of (a) negative (graphite) and (b) positive electrode (NMC) at 1500x magnification, and (c) graphite and (d) NMC electrode at 5000x magnification.	48
3.7	Negative and positive electrode (a) Open circuit potential, and (b) entropic coefficient.	48
3.8	Surrogate modelling (a) flowchart and (b) an illustration of a fitted response surface as a function of design variables.	52

4.1	EIS curve comparison of probed and unprobed cells at (a) SoC=0 and (b) SoC=1.	60
4.2	Fresh cell discharge capacity comparison of probed and unprobed cells.	61
4.3	(a) Measured surface temperature comparison of the cell at three specified locations for nine CCD points, (b) maximum and (c) average core and surface temperature comparison at all DOE points.	63
4.4	Voltage vs discharge capacity for nine CCD points.	64
4.5	Contour plots of maximum (a) core temperature, (b) surface temperature, time-averaged (c) core temperature, and (d) surface temperature, and the symbols represent the DOE points where the experiments have been performed.	65
4.6	Contour plot of (a) maximum discharge capacity, time-averaged (b) discharge power and (c) discharge energy.	67
4.7	Sensitivity bar chart for thermo-electrochemical variables showing (a) main sensitivity and (b) total sensitivity.	71
5.1	The flowchart of chapter structure.	74
5.2	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.	76
5.3	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.	76
5.4	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.	77
5.5	Temperature distribution visualization (a) in 3D entire domain and (b) 3D cut plane	78

5.6	Contour plots of cell temperatures at (a) 200 ml/min, (b) 600 ml/min, and (c) 1000 ml/min.	80
5.7	Contour plot of cells discharge capacity at (a) 200 ml/min, (b) 600 ml/min, and (c) 1000 ml/min.	81
5.8	Contour plot of cells average discharge power at (a) 200 ml/min, (b) 600 ml/min, and (c) 1000 ml/min.	83
5.9	Contour plot of cells discharge energy at (a) 200 ml/min, (b) 600 ml/min, and (c) 1000 ml/min.	84
5.10	(a) Cut plane temperature and (b) heat flux dependency with inlet fluid temperature.	85
5.11	Effect of C-rate at 25 °C and 1000 ml/min flow rate on (a) Irreversible, (b) Reversible, and (c) Heat of mixing.	86

List of Tables

1.1	Energy storage systems	2
2.1	Comparison of thermal physical properties of different coolants	30
2.2	Thermal properties of dielectric fluids used in immersion cooling	33
2.3	Data-driven modeling techniques in lithium-ion batteries	36
3.1	Thermophysical properties of HiTech-Therm 60 mineral oil	42
3.2	Species and charge conservation equations	45
3.3	Battery thermophysical properties	46
3.4	Relative composition of nickel, manganese, and cobalt obtained in different samples.	46
3.5	Range of input variables	49
3.6	Cell modelling parameters for electrode and electrolyte.	49
4.1	The range of input variables used in the experiments.	59
4.2	The temperature rise of cells at different discharging C-rates when cells are covered with and without sleeves.	62
4.3	The maximum and minimum values of various thermo-electrochemical performance outcomes at different input design data points.	69
4.4	Parameters of the fitted kriging model	69

Nomenclature

Abbreviations

BMS	battery management system
CPE	constant phase element
DMC	dimethyl carbonate
DMP	differential mechanical parameter
DTV	differential thermal voltametry
DV	differential voltage
ECM	equivalent-circuit model
EC	ethylene carbonate
EIS	electrochemical impedance spectroscopy
EV	electric vehicle
FDM	finite difference method
GPR	Gaussian process regression
IC	incremental capacity
OCP	open-circuit potential
P2D	pseudo-2D
PDE	partial differential equation
RUL	remaining useful life

SEI solid electrolyte interface

SoC state-of-charge

SoH state-of-health

SPM single-particle model

SVM support vector machine

Symbols

α_a, α_c anodic/cathodic charge transfer coefficients

$\dot{q}_{\text{mix},j}$ rate of heat generation due to mixing per unit volume in an electrode i (W/m^3)

\dot{q}_g rate of heat generation per unit volume (W/m^3)

$\epsilon_{e,i}$ electrolyte volume-fraction in electrode i

$\epsilon_{s,i}$ active material volume-fraction in electrode i

η_i surface overpotential for lithium intercalation/deintercalation reaction in electrode i (V)

λ_i thermal conductivity of domain i ($\text{W}/\text{m}\cdot\text{K}$)

$\phi_{e,i}$ ionic potential in the electrolyte phase (V)

$\phi_{s,i}$ electrical potential in electrode i (V)

ρ_i density of domain i (kg/m^3)

σ_i electrical conductivity for electrode i (S/m)

$a_{s,i}$ specific interfacial area for an electrode i (m^2/m^3)

C_e lithium concentration in the electrolyte (mol/m^3)

$C_{\text{max},i}$ maximum lithium concentration for an electrode i (mol/m^3)

$C_{p,i}$	lithium concentration in an electrode particle i (mol/m ³)
$c_{p,i}$	specific heat capacity of domain i (J/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)
$E_{a,i}$	activation energy for property i (J/mol)
F	Faraday's constant (C/mol)
H_{par}	total enthalpy stored in electrode particle i (J)
$H_{v,j}$	enthalpy per unit volume in an electrode i (J/m ³)
I_{app}	applied current(A/m ²)
$i_{0,j}$	exchange current density for lithium intercalation/deintercalation reaction at electrode j (A/m ² .s)
$j_{0,i}$	exchange molar flux density for lithium intercalation/deintercalation reaction at electrode i (mol/m ² .s)
$j_{f,i}$	interfacial molar flux density for lithium intercalation/deintercalation reaction at electrode i (mol/m ² .s)
k	rate constant for lithium intercalation/deintercalation reaction (m ² /s)
L_i	thickness of cell component i (m)
R	universal gas constant (J/mol.K)
$R_{s,i}$	particle size for electrode i (m)
T	temperature (K)

t	time (s)
T_{amb}	ambient temperature (K)
t_+^0	cation transfer coefficient
$U_{H,i}$	enthalpy potential for electrode i (V)
U_i	open-circuit potential in electrode i (V)
V	cell voltage (V)
y_i	state-of-charge in electrode i

Subscripts

e	electrolyte
n	negative electrode
p	positive electrode
s	separator
ref	reference value

Superscripts

0	initial condition
avg	average value
eff	effective property