Numerical and Experimental Investigation of Phase Change Material-Based Thermal Management for Mitigating Aging in Lithium-Ion Battery Packs

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Abstract: The paradigm shift from internal combustion engines to electric vehicles is driven primarily by advancements in lithium-ion cells (LICs) technology. As the advancement of the LICs in terms of long-life, high-energy density, high power density, high charge/discharge rate and cost-effective shows potential for future. This research examines a 3-series x 3-parallel (3s3p) based 21700-NMC lithium-ion battery pack module utilizing phase change material (PCM) as its thermal management solution. The module is evaluated for capacity degradation through a cycling test consisting of 600 continuous charge-discharge cycles. After 600 cycles at 1.5C charge and 2.5C discharge at room temperature, the SOH dropped by 24% without PCM and 19% with PCM. During operation, cell temperature rises, and PCM absorbs heat to keep it within the optimal range. PCM enhances battery performance and extends its lifespan. A deviation of ±5% is observed between the experimental and simulation results. This research emphasizes the effectiveness of PCMs in managing thermal challenges, enhancing the safety, reliability, and suitability of lithium-ion batteries for electric vehicles and other high-power applications.

Keywords: Lithium-Ion Cell; Cyclic Aging; Degradation; Equivalent Circuit Model; Phase Change Material; BMS.

1. Introduction

Lithium-ion cells (LIC) are now a pivotal source of energy for modern transportation, offering several advantages over traditional energy storage solutions. Their high energy density ensures compact and lightweight designs, making them ideal for electric vehicles (EVs). The prolonged lifespan of LICs reduces the need for frequent replacements, enhancing cost-effectiveness and sustainability. Additionally, their high discharge rates support powerful acceleration, while fast-charging capabilities enable reduced downtime, catering to the increasing need for efficient and eco-friendly transportation solutions [VIII]. LICs performance and lifespan depend heavily on precise assessment of the state of charge (SOC) and state of health (SOH). Precise algorithms for SOC and SOH estimation are vital for effective battery management, optimizing performance, and preventing failures [III]. However, operating C-rate and operation temperature parameters causes the degradation of the cell. For instance, a recent study [VII] highlights that higher C-rates (2C,3C) and low-temperature conditions (10°C) considerably increase the effects of aging, with lithium plating being the dominant cause of pressure increases. High temperatures can lead to thermal runaway in lithium-ion batteries, were excessive heat triggers uncontrollable reactions, causing further heat generation. This chain reaction can result in battery failure, fire, or even explosion [IX].
Maintaining the optimal temperature range is critical for ensuring the performance, safety, and lifespan of lithium-ion cells[X]. Battery Thermal Management Systems (BTMS) play a key role by preventing

overheating and uneven temperature distribution. Effective BTMS designs integrate active methods such as air and liquid cooling with passive methods like phase change materials (PCMs) and heat sinks [VI]. Current research emphasizes the adoption of advanced materials and intelligent control algorithms to enhance thermal regulation, reduce energy consumption, and prevent thermal runaway—especially relevant for electric vehicle (EV) applications. Accurate battery modeling is essential to predict behavior under a wide range of operating conditions, as it is not feasible to experimentally evaluate every scenario. Electrochemical models provide a comprehensive understanding of the internal processes and structural dynamics of lithium-ion cells. Alternatively, Equivalent Circuit Models (ECMs) offer a simplified representation of battery behavior through electrical analogies, enabling faster simulations. Data-driven models, using empirical data and machine learning techniques, have emerged as powerful tools for predicting performance and aging without explicitly modeling internal physics [V]. An investigation on temperature-dependent capacity loss in NMC and LMO cells, using a semi-empirical aging model, highlighted how cyclic aging behavior varies with temperature (4°C to 48°C), transitioning from decelerating to linear fade at higher temperatures [II]. Another study implemented a machine-learning framework to classify aging modes in NMC cells by analyzing electrochemical features, achieving 88% accuracy in identifying mechanisms like lithium inventory loss and cathode degradation after 225 cycles [I]. A twin-model framework was also developed to estimate battery lifetime by simulating capacity loss and resistance growth under real-world driving profiles, achieving RMS errors of 1.31% and 0.56% [IV].

To date, no known studies have experimentally or numerically analyzed the thermal optimization of 21700 NMC battery packs in a 3s3p configuration, incorporating cyclic aging effects.

Current research struggles to address cyclic aging effects in NMC-based battery packs. While PCM studies and high discharge impacts on cells exist, they overlook pack-level aging. Passive cooling causes hotspots and degradation, while active systems offer better control but face challenges due to complexity and cost, limiting real-world application.

The remainder of the paper is structured as follows: The second part of the paper covers the problem statement, followed by the third part, which outlines the elaborates on the experimental setup and methodologies. The fourth part explains the numerical approach, while the fifth part delves into the results and analysis, and the final part concludes the paper.

2. Problem Statement and Objective

Cell degradation occurs regularly during charge and discharge cycles, and this degradation is more pronounced if the cells' temperature is not maintained within the optimal range. The gradual degradation of lithium-ion cells leads to a rise in direct current internal resistance (DCIR), which subsequently reduces the cell's capacity. Higher operating temperature of the battery pack results in accelerated degradation and loss of battery pack capacity. Incorporating PCMs into these modules presents a viable strategy to enhance thermal performance. PCMs regulate heat by absorbing and releasing it during phase transitions between solid and liquid states. This characteristic permits them to control the battery's temperature modules by the absorption and release of surplus heat when necessary.

This research proposes a novel thermal management solution utilizing composite PCM, leveraging its latent heat absorption properties to maintain optimal cell temperatures. This approach not only mitigates hotspot formation but also provides a practical, cost-effective solution for managing the thermal effects associated with cyclic aging of NMC battery packs. The proposed innovation demonstrates improved battery performance, enhanced safety, and extended lifespan, addressing the vital need for efficient and economical thermal management strategies in modern battery systems. This is a need of an hour for electric vehicle.

This study involves both numerical and experimental analyses of a 3x3 configuration in a 21700 lithium-ion battery pack module, focusing on cyclic aging.

The objectives of the works are

❖ Perform hybrid pulse power characterization (HPPC) on the cell to determine the open circuit voltage (OCV), resistance, and capacitance values, which are essential for developing a model for numerical analysis

❖ Evaluate the impact of cyclic aging under 1.5C charge and 2.5C discharge cycles at room temperature on a 3x3 LIC module, with and without PCM utilization.

❖ Conduct a comprehensive investigation of several factors influencing the thermal performance of the battery pack.

❖ Propose advanced cooling techniques for lithium-ion cell pack and recommend areas for further development in this field.

3. Experimental Setup and Procedures

Fig. 1 displays a picture of the battery pack module used in this research. A steel casing was designed to securely hold the cells in a compact arrangement, with a 3 mm spacing provided between each cell. Once the cells are properly arranged, the PCM is filled into the casing. This research uses 21700 cylindrical lithium-ion cells. A recent model of LICs SAMSUNG INR-21700-40T cell is used in this study. The Samsung 21700 lithium-ion cell with NMC chemistry is a high-performance battery cell commonly used in EVs, energy storage systems (ESS) and other applications. The 21700-form factor has a greater energy density and capacity than the previous 18650 cells, with dimensions of 21 mm in diameter and 70 mm in length. NMC chemistry provides an excellent balance of energy density, power output, and lifespan while maintaining safety. These cells are distinguished by their reliability and efficiency, turning them into a popular selection for modern energy solutions. The specification of the cells is highlighted in table 2.

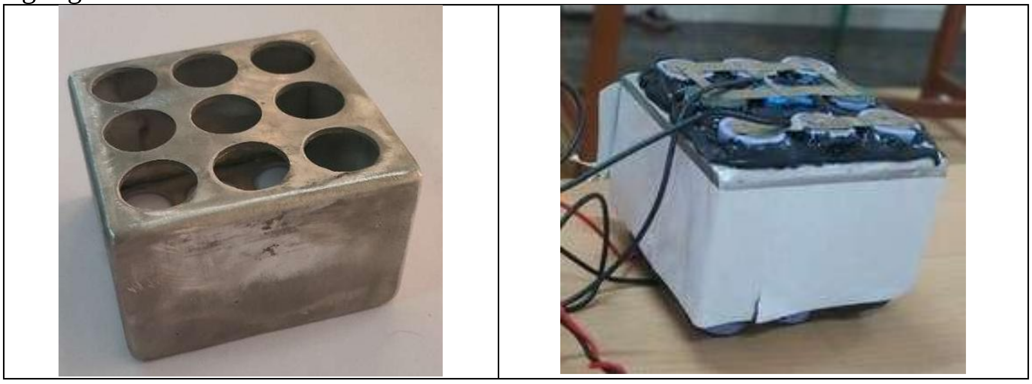


Fig 1: Battery Pack Module with 3x3 arrangement

This setup consists of three series-connected groups, each containing three cells connected in parallel. The series connection increases the overall voltage of the pack, while the parallel connection enhances the capacity and current-handling capability. To reduce changes in environmental conditions, strict control mechanisms are put in place, ensuring consistency across multiple trials. However, the experiments are not conducted under temperature-controlled conditions but are performed at ambient temperature, maintaining the highest possible consistency. To maintain consistency during testing, High-precision data recorders were used to closely monitor the temperature. To minimize inherent variances, all the battery cells used in the tests were from a single manufacture batch.

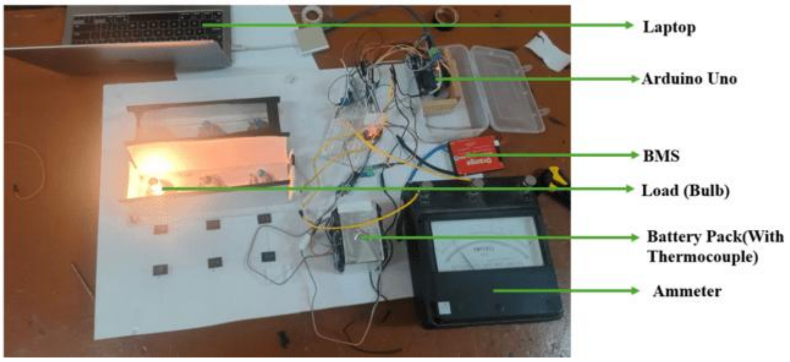


Fig. 2. Experimental setup of proposed methodology

The battery pack module was tested under controlled conditions to analyse its cyclic aging characteristics. The setup consisted of an Arduino board, which served as the central data acquisition system. Multiple sensors were connected to the battery pack, including voltage, current, and three temperature sensors, to monitor real-time performance parameters. Three thermocouples were strategically placed in many places within the battery pack to measure temperature variations. The thermocouples were positioned from the centre of the battery module to its outer surface, capturing temperature gradients across the pack. This setup allowed for precise monitoring of thermal distribution and identification of potential hotspots.

A laptop was interfaced with the Arduino board to continuously fetch and keep the gathered information for later examination and visualization of SOC, SOH, voltage, current and temperature. During the discharge phase, an electrical bulb was employed as a resistive load to draw current from the battery pack. A series of switches were incorporated into the setup to control the operation of the bulb, allowing it to be turned on and off as needed. Additionally, the switches enabled fine-tuning of the C-rate by adjusting the current draw, ensuring that the discharge rate could be set to the required value for each cycle. For the charging process, a charger was employed to replenish the battery capacity following each cycle. The Arduino Uno the perfect control centre for this adaptable and practical system because of It has fourteen digital I/O pins and six analog input ports. Fig. 3 shows the thermocouple, DC electrical loads, data recorders, and batteries. The temperature of the battery is upheld at 29°C, with changes limited to +/- 0.5 °C, to maintain reduce environmental effects and maintain a consistent experimental setup. This setup allowed precise monitoring of battery behaviour throughout the cyclic aging tests, ensuring accurate evaluation of capacity degradation under the specified charging and discharging conditions. The thermocouple and voltage sensor calibration procedures and parameters are displayed in Table 1.

Table 1. Thermocouple calibration procedures and parameters

Measurement	Calibration Method
Temperature Sensor	Preparation: Clean the thermocouple and prepare a calibrated reference thermometer. Setup: Place the thermocouple and reference thermometer in a controlled temperature environment. Measurement: Record the thermocouple's output at a known reference temperature. Adjustment: Compare readings with the reference thermometer and apply necessary corrections. Validation: Test at multiple temperatures to confirm accuracy across the range.
Voltage (Data Logger)	Preparation: Ensure the voltage sensor is properly connected and free from faults. Prepare a calibrated reference voltmeter. Setup: Connect the voltage sensor to a stable DC voltage source. Place the reference voltmeter in parallel with the sensor. Measurement: Measure the voltage sensor's response to a predefined input voltage. Adjustment: Contrast the sensor's measurements with the reference voltmeter and apply necessary corrections. Validation: Test at multiple voltage levels to verify accuracy across the operational range.

The battery pack was discharged in Constant Current (CC) mode, maintaining the required C-rate with a cutoff voltage of 2.5V to prevent over-discharge [21]. For the charging process, the Constant Current - Constant Voltage (CC-CV) mode was employed, with a cutoff voltage of 4.2V to ensure proper charging and avoid overcharging, thereby protecting the cells from potential damage. These voltage limits were crucial in maintaining the integrity of the cells throughout the test. Throughout the test, three thermocouples' temperatures data are recorded, and the gathered data is analysed. Fig 2 illustrates the experimental setup on the battery pack for measuring temperature, voltage and current under cyclic aging [22-23]. Before stating the experiment, the battery pack is kept in the stable test section box. The practical significance of the 3×3 LIC module design in real-world applications, particularly in ESS and EVs, led to its selection for this study. In order to stabilize the chemical substance inside the batteries, lessen the unpredictability of chemical constituents, and preserve cell performance, a steady temperature is regulated within designated test section enclosures before initiating experiments.

This 3×3 configuration pack module serves as a foundational study for researchers aiming to develop EV battery packs with PCM, aligning seamlessly aligned with the research goals across various aspects. Firstly, it aids in understanding the practical implications of PCM integration under cyclic charge and discharge conditions, highlighting its effect on battery performance and longevity. In addition, the chosen configuration enables the exploration of various factors that influence thermal performance, including cell configuration, discharge C-rates, environmental factors, and the characteristics of the PCM. This approach provides a comprehensive insight into thermal dynamics. The knowledge obtained from analysing the thermal performance of the 3×3 LIC pack can be applied to the development of advanced thermal management solutions for EV battery pack, supporting the goal of offering suggestions for improved thermal management. Ultimately, selecting the 3×3 LIC module is a strategic decision based on its real-world significance and its coherence with the study goals, assisting an in-depth study of thermal management techniques for LIC pack in EV uses.

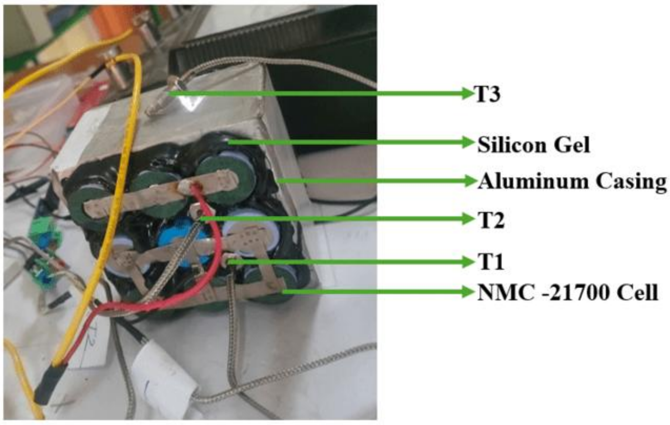


Fig. 3. Temperature measurement setup for measuring

The LICs pack is placed within the battery testing facility for multiple charge and discharge trials.

(a) Aluminium Casing (b) PCM

(c) Lithium-ion Cell (d) Battery Module Assembly

Fig. 4. (a) Aluminium Casing (b) PCM (c) Lithium-ion Cell (d) Battery Module Assembly

The experiment on battery module discharge is conducted in a controlled environment to guarantee precise output. Prior to testing, the initial parameters are verified to confirm they are stable and within normal limits, while data is continuously captured across the process. Fig. 4 illustrates the virtual battery module of the 3×3 cell configuration, and Table 2 presents the key characteristics from the battery configuration utilized in this study.

Table 2. Features of the Samsung INR-21700-40T battery

Battery Parameters	Parameter	Value
Capacity (Ah)	Typical	4
	Minimum	3.9
Voltage (V)	Nominal Voltage	3.60
	Voltage Cut-off for Charge	4.20
	Voltage Cut-off for Discharge	2.50
Operating temperatures (ºC)	Charge	0-45
	Discharge	-50-70
Dimension (mm)	Diameter	21.2
	Height	70

HPPC test on the single cell

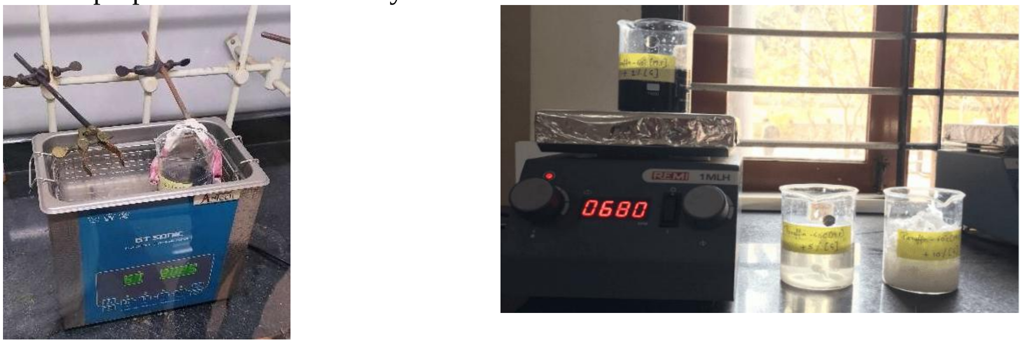
A lithium-ion cell was subjected to the HPPC test in order to assess its dynamic performance at distinct SOC levels. The test was conducted from 100% SOC to 10% SOC, with measurements recorded at 10% intervals. At each SOC level, the cell was subjected to a current pulse at a 0.5C rate (2A) under controlled ambient conditions maintained at 25°C.

Cyclic Aging of Lithium-ion Battery Module

The battery module is conditioned to a fully charged state before initiating the cycling process. In this study, the charging process is conducted using a current of 1.5C (18 Ampere) with a cutoff set at 600mA. Discharging begins only when the cell temperature drops below 30°C to ensure thermal stability. During discharging, a current of 2.5C (30 Ampere) is applied, with the process terminating either at a voltage cutoff of 2.5V or when the cell temperature reaches 75°C, whichever occurs first. Charging is resumed only after the cell temperature has cooled to below 30°C, maintaining safe operating conditions throughout the cycle. This rest period allows the chemical reactions within the cells to stabilize. Continuous performance metrics of the battery pack are documented under these conditions.

Material Development

In this study, a composite PCM is developed by combining paraffin wax with 10% by weight graphene nanoparticles to enhance its thermal conductivity. In the preparation of the composite PCM, paraffin wax and graphene nanoparticles are blended using an Ultrasonicator and a magnetic stirrer. The Ultrasonicator is used for 60 minutes maintaining 75 ^{o}C to disperse the graphene nanoparticles evenly in the wax by applying ultrasonic waves, which break down nanoparticle agglomerates and ensure a homogeneous mixture. The magnetic stirrer is then employed to continuously agitate the mixture, preventing the nanoparticles from settling and ensuring uniform distribution throughout the paraffin wax. This combination of ultrasonic and magnetic mixing techniques ensures that the composite PCM has enhanced thermal properties and consistency.



(a) Ultrasonicator (b) Magnetic Stirrer

(c) Paraffin Wax Mixing (d) Paraffin wax

Fig. 5. (a) Ultrasonic cleaner setup (b) Magnetic stirrer (c) paraffin wax mixing (d) paraffin wax

The use of an Ultrasonicator and magnetic stirrer ensures the effective dispersion of graphene nanoparticles in paraffin wax, enhancing the thermal properties of the composite PCM. This method guarantees a homogeneous mixture, optimizing the performance and consistency of the PCM for various thermal management applications. Fig 5 illustrates the material development experimental setup of this study.

Table 3. Characteristics of materials

Specification	CR+60 (Paraffin Wax)
Colour	Colourless
Interaction with metal	Inert
Latent heat	180 KJ/kg
Melting Point	60 ^{o}C
Max operating temperature	90 ^{o}C
Freezing Point	58 ^{o}C
Sp heat at solid	2.2 KJ/kgK
Sp heat at liquid	1.6 KJ/kgK
Solid thermal conductivity	0.3 W/mK
Liquid thermal conductivity	0.25 W/mK
Expansion of volume	5
Density at solid	1160 kg/m^3
Density at liquid	1220 kg/m^3
Combustible.	No

Congruent melting	Yes
Thermal life cycle	10000

While the cleaning process, high-frequency sound waves are released by the ultrasonic cleaner that cause the creation and cavitation bubble collapse. This phenomenon generates waves of shock that efficiently remove contaminants from the surfaces of the battery packs. After the set cleaning duration is complete, the ultrasonic cleaner is turned off, and the battery module are carefully taken out of the beaker. This test configuration leverages the benefits of ultrasonic cleaning, including its effectiveness, non-abrasive nature, and versatility, while also enabling the evaluation of how discharging affects the cleanliness of 3×3 battery packs. The material properties are detailed in Table 3.

IV. Numerical Method

In this research, the equivalent circuit model (ECM) is employed to evaluate the performance of lithium-ion cells, focusing on cyclic aging. The model simulates the cell's electrical behaviour, capturing key parameters like resistance, capacitance, and open-circuit voltage (OCV). Cyclic aging is studied by simulating the impact of repeated charge-discharge cycles on the cell's capacity loss and internal resistance. By incorporating aging effects into the ECM, the study offers insightful information about the prolonged performance and degradation mechanisms of lithium-ion cells.

HPPC Test

The outcomes of the HPPC test form the basis for the SAMSUNG 21700-40T cell model. Fig 6 illustrates the voltage response of the cell during the test, which involves applying a current pulse of 1C (4A) for 360 seconds, followed by a rest period of 30 minutes. This procedure provides essential data for modelling the cell's dynamic behaviour, including its capacity, internal resistance, and transient response characteristics.

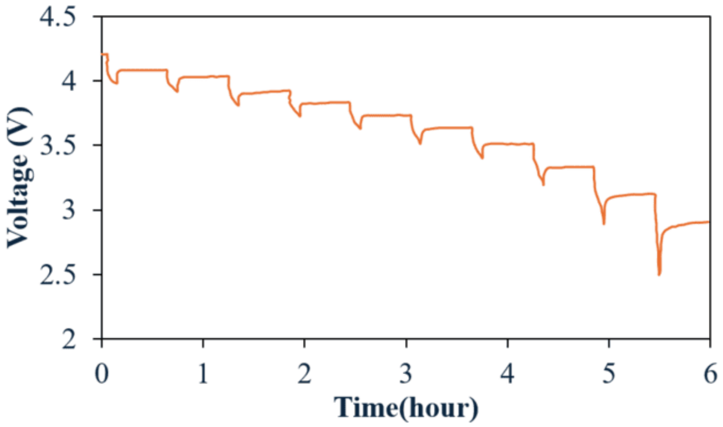


Fig. 6. HPPC test: Voltage response with respect to current pulse

Equivalent Circuit Model

The 2RC model, also referred as the Thevenin model is represented as illustrated in Fig 7, is a widely used equivalent circuit representation of a lithium-ion battery. This model is particularly effective for simulating dynamic battery behaviour, making it valuable for applications in battery management systems (BMS) and system-level simulations. Beyond this the model is also useful for calendar and cyclic aging modelling and prediction.

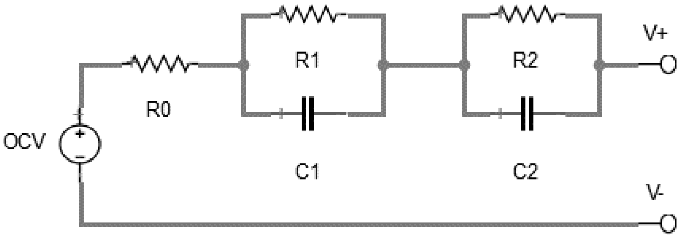


Fig 7. Thevenin model for lithium-ion cell.

Components of 2RC Model:

➢ Open Circuit Voltage (OCV): Represents the equilibrium voltage of the cell at a given SOC. It is independent of the load and is determined experimentally HPPC for different SOC levels.

- Internal Resistance (R_0): Models the ohmic losses (resistance) due to the internal resistance of the cell. Includes contributions from electrolyte resistance, electrode materials, and current collectors.
- Polarization Resistance (R_1): Resistance arising from charge transfer and electrochemical reactions taking place at the junction between the electrode and the electrolyte.
- Diffusion Resistance (R_2): Limitation of ion movement within the solid electrode materials and the liquid electrolyte.
- Capacitance (C_1 & C_2): Represent the capacitances in the two RC networks. These capacitors, combined with their associated resistances (R_1 & R_2), help capture the dynamic response of the cell voltage during charge and discharge.

Voltage Prediction:

The cell voltage is modelled as:

$$V_{out} = V_{OCV} - I.R_o - V_{R1C1} - V_{R2C2} \quad (1)$$

Where:

V_{out}- Output voltage of the cell (Voltage)

V_{OCV}– OCV of the cell (Voltage)

I – Current (Ampere)

R_o – Ohmic resistance (Ohm)

V_{R1C1} – Voltage across R_1C_1 branch (Voltage)

V_{R2C2} – Voltage across R_2C_2 branch (Voltage)

Equation (1) is time dependent and can be rewritten as:

$$V(t) = V_{OCV}(s(t)) - I(t).R_o - I_{R1}(t).R_1 - I_{R2}(t).R_2 \quad (2)$$

Where, $s(t)$ - State of Charge

$$s(t) = s(t0) - \frac{1}{Q}\int_{t0}^{t} I(t)dt \quad (3)$$

Where, Q – total capacity of cell (Ah)

To derive an expression for $I_{R1}(t)$, we note that the current through the resistor R_1 and the current through the capacitor C_1 must together equal the total current $I(t)$.

Next, $I_{C1}(t) = C_1.\frac{dV_{C1}}{dt}$, which gives

$$I(t) = I_{R1}(t) + C_1.\frac{dV_{C1}(t)}{dt} \quad (4)$$

Then, since $V_{C1}(t) = R_1.I_{R1}(t)$,

$$I_{R1}(t) + R_1C_1\frac{dI_{R1}(t)}{dt} = I(t) \quad (5)$$

$$\frac{dI_{R1}(t)}{dt} = -\frac{1}{R_1C_1}I_{R1}(t) + \frac{1}{R_1C_1}I(t) \quad (6)$$

The given differential equation can be directly used to simulate $I_{R1}(t)$, this approach allows for accurate representation of the circuit's dynamic response.

Temperature Prediction:

The equivalent circuit model (ECM) predicts the temperature in a lithium-ion cell by integrating its electrical and thermal behaviours. The ECM simulates the cell's electrical response, including current, voltage, and internal resistances. Heat generation within the cell primarily arises from two sources: Joule heat (irreversible heat), resulting from current flow through internal resistance, and reaction heat (reversible heat), associated with electrochemical reactions and entropy changes. This generated heat is then coupled with a thermal model, enabling the calculation of temperature rise within the cell during operation.

The governing thermal equation is:

$$mc\frac{dT}{dt} = Q_{gen} - Q_{loss} \quad (7)$$

Where, m: Mass of the cell, c: Specific heat capacity of the cell, T:Cell temperature, Q_{gen}: Total heat generated, Q_{loss}: Heat dissipated to the PCM.

Total heat generated can be expressed as:

$$Q_{gen} = I^2 R_{total} + IT\frac{dV_{OCV}}{dT} \qquad (8)$$

Where, $R_{total} = R_o + R_1 + R_2$: Total resistance in the model, I: Current through the cell, T: Cell temperature, $\frac{dV_{OCV}}{dT}$: Entropy coefficient.

Heat loss or heat dissipated to the PCM can be expressed as:

$$Q_{loss} = \frac{T - T_{ambient}}{R_{th}} \qquad (9)$$

Where, T: Cell temperature, $T_{ambient}$: Ambient temperature, R_{th}: Thermal resistance of the cell

Cyclic Aging Prediction:

By incorporating calendar and cyclic aging phenomena, the ECM enables precise prediction of capacity fade and resistance growth under various operating conditions. Numerical simulations in this study leverage the ECM to analyse the effect of temperature, SOC, and cycle count on the aging characteristics of the cell. The integration of aging effects into the ECM allows for enhanced modelling accuracy, supporting establishing practical methods for enhancing battery longevity and performance. The following equations outline the mathematical representation of aging phenomena, capturing the effect of calendar and cyclic aging on the key electrical parameters of the ECM.

The calendar aging is a function of SOC, temperature and time.

The capacity loss resulting from calendar aging can be expressed as:

$$L_t = \int g(soc).h(temp).f(t - t_{shift})dt \qquad (10)$$

The cyclic aging is dependent on SOC, temperature and charge.

Capacity degradation caused by cyclic aging can be represented as:

$$Lq = \int g(soc).h(temp).f(q)dq \qquad (11)$$

Resistance increase due to calendar aging is expressed as:

$$R_t = \int g(soc).h(temp).f(t - t_{shift})dt \qquad (12)$$

Resistance increase due to cyclic aging is expressed as:

$$Rq = \int g(soc).h(temp).f(q)dq \qquad (13)$$

Where, t: Time, q: Charge, $\frac{L_t}{L_q}$: Capacity loss due to time/charge, $\frac{R_t}{R_q}$: Resistance increase due to time/charge.

Finally, the cumulative effects of cyclic and calendar aging on capacity loss and resistance growth can be represented as:

$$Cap = Cap_{fresh} \times m_c = Cap_{fresh} \times \left[L_{init} - \left(L_t + L_q\right)\right] \qquad (14)$$

$$Res = Res_{fresh} \times m_c = Res_{fresh} \times \left[R_{init} - \left(R_t + R_q\right)\right] \qquad (15)$$

State of Health (SOH):

The SOH of a battery is generally expressed as a percentage that indicates the remaining capacity or ability of the battery to hold charge compared to its original capacity when new.

The general formula for calculating SOH is:

$$SOH = \left(\frac{\text{Currrent Capacity}}{\text{Intial Capacity}}\right) \times 100 \qquad (16)$$

5. Results and Discussion

The findings of this study highlight the impact of cyclic aging on the performance of lithium-ion cells, as analysed through the equivalent circuit model (ECM). This section will offer a detailed analysis of numerical and experimental data of 21700 NMC lithium-ion cell. The combination of numerical simulations and experimental data provides a comprehensive perspective, allowing for an in-depth evaluation of the effectiveness of different cooling strategies. This analysis sheds light on the complexities of the battery's thermal behaviour while highlighting its overall impact on safety and performance enhancement. By delving into these aspects, it seeks to provide meaningful insights and drive further progress in lithium-ion cell technology.

Assessment of Numerical Outcomes

Based on the numerical results from the 3x3 battery module, the ECM proves to be a reliable approach for modelling LICs battery packs. The ECM is a simplified representation of a complex system, by incorporating electrical elements like resistors, capacitors, and voltage sources. ECMs balance complexity along with efficient computation, rendering them ideal for real-time implementations and iterative system design in electric vehicle battery pack or energy storage systems.

The cyclic aging models is calibrated using the specifications and data provided in a battery's technical data sheet as a foundation to develop and validate cyclic aging models. The technical data sheet specifies the cycle life performance of the cell as follows: The cell retains a capacity of ≥ 2,400mAh after 250 cycles, which corresponds to 60% of the standard capacity at room temperature. During cycling, the cell is charged at a current of 6A using a CC-CV method with a cut-off voltage at 4.20V and 100mA. It is discharged at a current of 35A with a cut-off voltage of 2.5V, both at room temperature. These conditions define the testing protocol for evaluating the battery's cycle life performance.

Fig 8. illustrates the cyclic aging calibration of the cell, demonstrating a strong correlation with the data provided in the technical data sheet. The calibrated model accurately predicts the battery's capacity retention and degradation trends under the specified cycling conditions. This alignment confirms the model's validity for simulating the cell's performance, ensuring reliable predictions for charge-discharge cycles at various operating conditions.

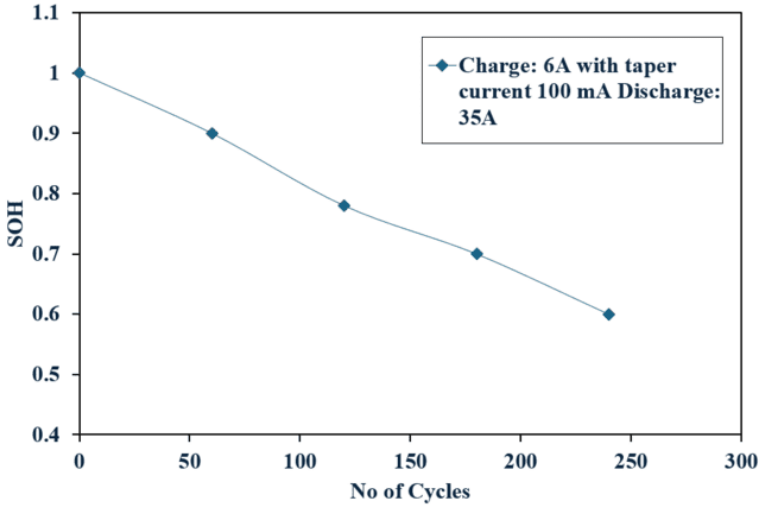


Fig 8. Calibration of cyclic aging for the cell

Fig 9 illustrates the cyclic aging behaviour under specific conditions: charging at 1.5C with a 600mA cutoff and discharging at 2.5C, terminating at 2.5V or 75°C. Both processes commence only when the cell temperature drops below 30°C to ensure safe operation.

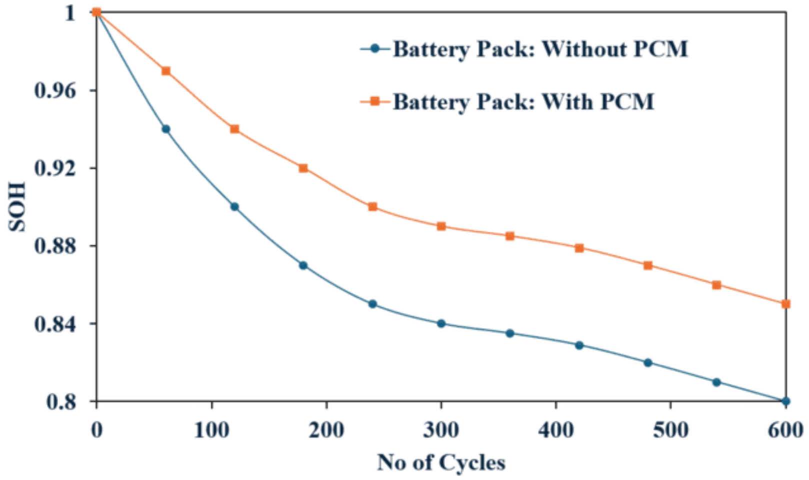


Fig 9. Cyclic aging of battery pack at room temperature.

In this graph, it is clearly observed that the battery pack degradation with PCM is lesser than without PCM. This demonstrates the effectiveness of PCM in reducing thermal stress and slowing down the aging process of the battery pack. An improvement of 5% is observed with the use of PCM.

Experimental Results

The experimental results offer crucial information about the performance and degradation behaviour of the tested battery pack module. Capacity fade and internal resistance growth were observed as expected, consistent with cyclic aging models calibrated to match the experimental data. The experiment is conducted with and without PCM material and its impact on thermal behaviour is evaluated.

Fig 10. Shows the cyclic aging of battery pack at room temperature, here the battery pack degradation is more for the pack without PCM, and 6% lesser degradation is observed for pack with PCM. This shows that the maintaining of the cell temperature is optimum value, helps in minimizing the degradation of the battery module. This effect is observed because the PCM helps regulate the cell temperature by absorbing and dissipating excess heat during charge and discharge cycles. By preventing temperature spikes, the PCM reduces thermal stress on the cells, thereby slowing down the electrochemical degradation processes and enhancing the battery pack's overall longevity.

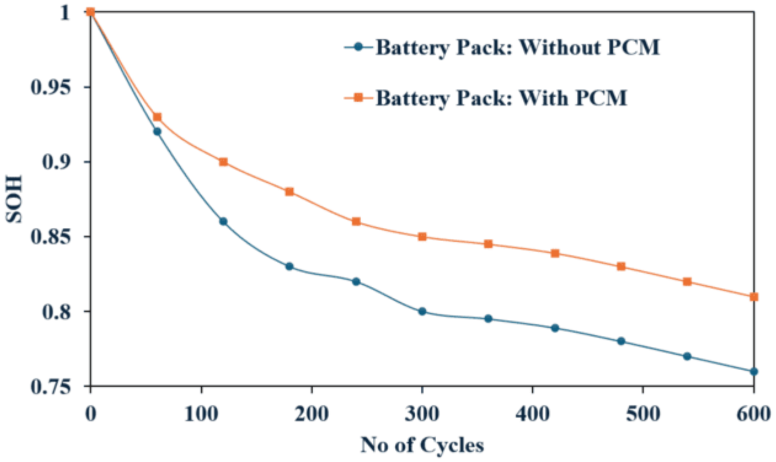


Fig 10. Cyclic aging of battery pack at room temperature for 600 cycles.

Validation of the Model

Fig 11 shows the cyclic aging results were validated by comparing experimental data with simulated predictions under the same operating conditions. The validation involved analysing capacity degradation across charge-discharge cycles for battery packs with and without PCM. Key parameters, such as current rates (1.5C charging and 2.5C discharging) and temperature thresholds, were matched in both experimental and simulated setups. The results showed a strong agreement between experimental and numerical data, verifying the model's validity in capturing the influence of cycling conditions on cell degradation. This validation demonstrates the robustness of the methodology in evaluating cyclic aging behaviour.

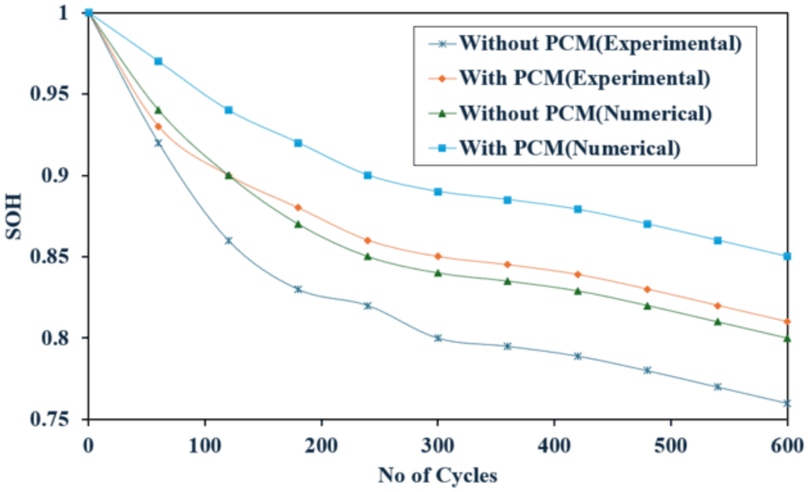


Fig 11: Validation of Cyclic Aging Results: Experimental vs. Simulated Capacity Degradation under Charge-Discharge Cycles

6. Conclusion

This study explores the effects of cyclic aging on the thermal performance of a battery pack system and utilizing a simulation data for validating it with experimental data. The established cell model effectively represents the thermal behaviour of 3×3 battery modules, both with and without the inclusion of PCMs.

An in-depth analysis of the key parameters impacting the thermal and aging dynamics of the battery pack module results in the following conclusions:

- This study investigated the cyclic aging behaviour of lithium-ion battery packs subjected to 1.5C charging and 2.5C discharging at room temperature. After 600 cycles, the battery pack without PCM exhibited a 24% capacity loss, whereas the pack equipped with PCM showed a reduced capacity loss of 19%. Reflecting a 6% improvement in capacity retention
- These results highlight the benefits of PCM in enhancing thermal management, particularly under elevated temperatures, thereby improving the sustained performance and reliability of lithium-ion cells.
- These outcomes highlight the significance of temperature control in improving battery performance and longevity, offering key insights for the design and optimization of high-power battery systems.
- Investigating temperature fluctuations across various surfaces within a lithium-ion battery module provides essential data for future research, helping to pinpoint key heat-sensitive areas that require attention in the development of advanced cooling solutions. Moreover, incorporating PCMs into these modules serves as an effective thermal management strategy, maintaining stable temperatures even during aging.
- PCMs maintain battery temperature within the ideal operating limits by absorbing heat via both sensible and latent heat sources.
- This paves the way for an expanded study to investigate the behaviour of PCM-based battery packs under both low and high-temperature conditions, focusing on cyclic aging. Additionally, further research can be conducted by varying the charge and discharge C-rates to assess their impact on battery pack performance. It is vital to mention that the present study is limited to a 3×3 battery pack configuration utilizing paraffin wax and graphene nanoparticle-based PCM. The following indicates the work's future course.

Future work for this research could focus on several key areas to advance understanding and application. First, extended studies on calendar and cyclic aging at extreme temperatures (-20°C to 60°C) should be performed, incorporating microstructural analysis and advanced electrochemical-thermal modelling to better predict aging mechanisms. Second, the optimization of PCM properties, such as thermal conductivity and phase change range, and their integration into battery packs should be explored, including hybrid PCM systems and mechanical design improvements. These efforts will contribute to more efficient, durable, and eco-friendly battery systems for electric vehicles.

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Conflict of interest

The authors declare no conflict of interest related to this research.

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