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Incorporating Copper and Carbon Nanotube Nanoparticles into Phase Change Materials for Enhanced Thermal Management in Batteries

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Abstract

The primary objective of this study was to explore the impact of integrating nano-additives on heat transfer enhancement within an LHTES (Latent Heat Thermal Energy Storage) system. Our findings revealed that introducing a minimal amount of nano-materials (less than 5.0 vol%) into the Phase Change Material (PCM) led to a remarkable alignment between experimental correlations and mixture models. Furthermore, employing the mixing model allowed for the accurate prediction of outcomes. In the current context of scientific research, there is a strong endorsement for the widespread adoption of Electric Vehicles (EVs) as a sustainable alternative to Internal Combustion Engines (ICEs), crucial for advancing decarbonization and mitigating climate-related crises. Lithium-ion batteries are the predominant choice for EVs and various electrical devices. However, the operational challenges posed by high temperatures, impacting their lifespan, charge/discharge cycles, and the risk of thermal runaway, necessitate effective thermal management systems. Given this background, our study focuses on simulating the heat dissipation of a single cylindrical Li-ion battery cell employing a Nano-enhanced Phase Change Material (NePCM)-based cooling system. We also introduce a novel fin design in this work. Through comprehensive analysis, we examine the performance of battery modules utilizing PCM and NePCM at different discharge rates, both with and without the newly proposed fin design. Our research demonstrates that the incorporation of fins and nanoparticles into PCM significantly enhances heat transfer and reduces the charging time compared to the base PCM. Notably, carbon-based nanoparticles outshine their metal-based counterparts in terms of melting rate and maintaining a uniform temperature profile within the battery.

Keywords: *Battery Thermal Management, Carbon Nanotubes, Copper, Fins, Li-ion Battery, Nano enhanced PCM (NePCM)*

1.0 Introduction

In response to challenges related to energy scarcity and environmental degradation, the automotive industry has redirected its efforts towards the development and implementation of Electric Vehicles (EVs) as an alternative to conventional internal combustion engines. This transition has been marked by a notable increase in EV adoption, with over 10 million electric vehicles projected to be in operation globally by the close of 2020. This trend is reflected in a 41% increase in the number of electric vehicle registrations in 2020, even amidst a global downturn in the automotive sector which saw a 16% reduction in sales[1]. Electrified vehicles, including Hybrid Electric Vehicles (HEVs) and Electric Vehicles (EVs), rely on electrochemical batteries as their primary energy

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storage medium. Lithium-ion Batteries (LIBs) are the most widely adopted among these technologies, owing to their high energy density, elevated voltage, prolonged cycle life, and cost-effectiveness[2]. Despite the many advantages of using batteries in electrified vehicles, their degradation rate is often expedited by exposure to high temperatures[3]. There is currently a wide range of battery options that vary greatly in terms of cell chemistry and design. Nevertheless, the battery produces heat during charging and discharging at the cellular level, thermal management poses a serious problem for battery use in automotive applications. Battery performance and lifespan are known to be compromised by temperatures above 40°C. According to a scientific study by Song *et al.*, the optimal operating range for lithium-ion batteries is generally considered to be between -15°C and 40°C. When discharging the battery, transitioning from a lower C rate to a higher C rate leads to increased temperatures on the primary surface of the battery. In scientific terms, elevated temperatures usually have adverse effects on key performance indicators of batteries, such as electrochemical behaviour, round-trip efficiency, power and energy output, reliability, and cycling stability[4]. Hence, a specialized Battery Thermal Management System (BTMS) is necessary to control the temperature of batteries during high-rate charge and discharge, due to the significant impact of temperature on battery performance and safety. Such a system can optimize battery operation, increase durability, and reduce the risk of overheating or thermal runaway[5]. Based on the findings of the literature review, it can be inferred that the utilization of three distinct cooling methodologies, namely liquid cooling, air cooling, and Phase Change Material (PCM), can potentially enhance the performance of batteries[6-8]. The most widely used cooling method is PCM cooling, mainly due to its compact size and ability to maintain an isothermal state while being charged or discharged[9,10]. As a result, several researchers have pointed out that the low thermal conductivity of Phase Change Materials (PCMs) considerably hinders their thermal performance. Consequently, enhancing the effective thermal conductivity of PCM through heat transfer enhancement techniques is crucial[11,12]. NePCM refers to adding nanoparticles to phase-change materials to increase thermal conductivity, which can decrease the latent heat of PCM. Adding too many nanoparticles can adversely affect performance since the high latent heat of PCMs is essential for maintaining system temperature while the PCM's temperature remains constant[13]. In addition to the techniques mentioned previously, incorporating metal fins within the PCM is also an effective heat transfer enhancement method[14]. To the best of our knowledge, the implementation of fin-enhanced PCM systems for the thermal management of lithium-ion batteries has been limited. As a result, it would be beneficial to conduct more comprehensive studies on the efficiency of PCM-Fin-based BTMS, which could offer valuable insights for practical implementation. This study involves a numerical investigation into the thermal behaviour of a cylindrical battery that incorporates a composite of Nano-enhanced Phase Change Material (NePCM) and fin structures. A novel Battery Thermal Management System (BTMS) is proposed in which a combination of fins, specifically designed for improved thermal enhancement, are affixed to the battery and submerged within the NePCM material. The results of this study have implications for the development of advanced battery thermal management systems, as they can inform the design of more efficient and effective cooling mechanisms to regulate battery temperature during operation. In order to improve thermal conductivity, the addition of carbon-based nanoparticles to phase-change materials has been found to be more effective than adding metallic-based nanoparticles.

2.0 Methodology

A model for this study is a transient state heat flow from an 18650 li ion cell to a layer of Phase Change Materials enclosed around it. A paraffin wax (organic PCM)) is the base material for PCM and the optimized longitudinal fin configuration with the cylindrical ring is employed based on experimental results published in literature[15,16].

Nanoparticles are incorporated into the PCM and fins to improve the PCM's thermal conductivity. The illustration as shown in Figure 1 The aim of this numerical study is to conduct a comparative analysis of metal-based and carbon-based nanoparticles.

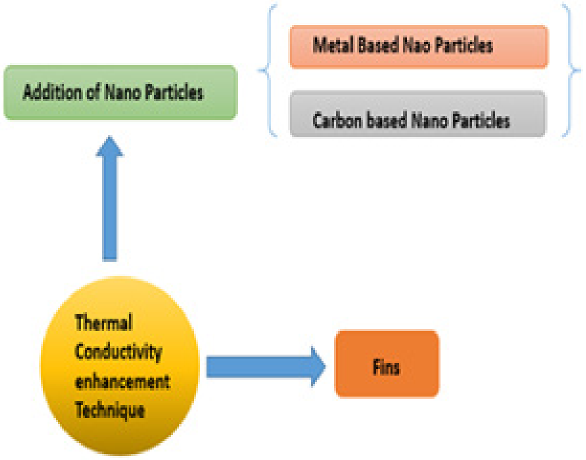


Figure 1. The illustration of thermal conductivity enhancement technique.

2.1 Physical Model and Theory

The aim of the study was to investigate the effect of incorporating nano-additives on heat transfer enhancement in an LHTES (Latent Heat Thermal Energy Storage) system. The outcomes of the study indicated that incorporating a small quantity of nano-materials (less than 5.0 vol%) in the PCM (Phase Change Material) resulted in acceptable conformity between experimental correlations and mixture models. Moreover, using the mixing model, simple equations accurately predicted the results[17]. In the present study, the mixing model was employed to ascertain the specific heat (Cp) and density (ρ). Additionally, the simplified version of the mixing model was utilized to compute the latent heat (L) and thermal expansion (β) as follows:

$$\mu_{NePCM} = \left(1 - \frac{\varphi}{\varphi_{\max}}\right)^{-A\varphi_{\max}} \mu_{PCM} \tag{1}$$

$$\rho_{\text{NePCM}} = (1-\varphi)\rho_{\text{PCM}} + \varphi\rho_{\text{NM}} \tag{2}$$

$$(\rho C_p)_{\text{NePCM}} = (1-\varphi)(\rho C_p)_{PCM} + \varphi(\rho C_p)_{\text{NM}} \tag{3}$$

$$(\rho L)_{NePCM} = (1-\varphi)(\rho L)_{PCM} \tag{4}$$

$$(\rho\beta)_{NePCM} = (1-\varphi)(\rho\beta)_{PCM} \tag{5}$$

The modified Krieger-Dougherty model is used to calculate the effective dynamic viscosity for NePCMs containing other nanomaterials:

$$\mu_{NePCM} = \left(1 - \frac{\varphi}{\varphi_{\max}}\right)^{-A\varphi_{\max}} \mu_{PCM} \tag{6}$$

Where the volume proportion of nanomaterials is denoted by φ the shapes of nanomaterials affect the values of A (intrinsic viscosity) and $\varphi_{\max}$ (maximum packing factor).

In this study, A = 2.5, 9.25 and $\varphi_{\max} = 0.632, 0.268$ respectively for metal based metal oxide Nano particles are considered. In addition, as a result of the structural variations of the nanomaterials, two distinct models were employed to estimate the thermal conductivity of the NePCM.

$$k_{NePCM} = k_{PCM}\left[\frac{k_{NP} + 2k_{PCM} - 2\varphi(k_{PCM} - k_{NP})}{k_{NP} + 2k_{PCM} + \varphi(k_{PCM} - k_{NP})}\right] \tag{7}$$

$$k_{NePCM} = k_{PCM}\left[\frac{1 - \varphi + 2\varphi\frac{k_{CNT}}{k_{CNT}-k_{PCM}}\ln\left(\frac{k_{CNT}+k_{PCM}}{2k_{PCM}}\right)}{1 - \varphi + 2\varphi\frac{k_{CNT}-k_{PCM}}{k_{PCM}}\ln\left(\frac{k_{CNT}+k_{PCM}}{2k_{PCM}}\right)}\right] \tag{8}$$

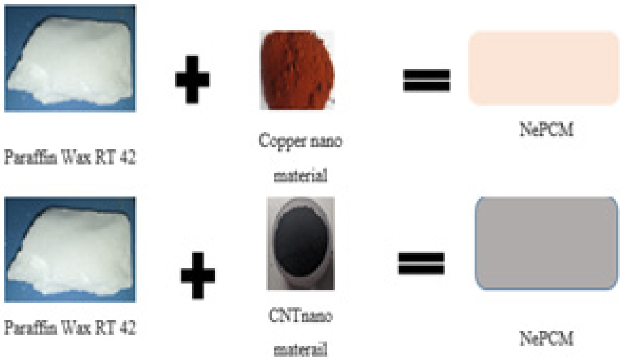


Figure 2. Preparation Process of NePCM.

2.2 Geometric Model

In the ongoing numerical study, a specific arrangement comprising 18,650 Lithium-ion cells and a Phase Change Material (PCM) is utilized. Additionally, a new arrangement of fins with a cylindrical ring attached to them is chosen based on a previously published experimental study[15].

Table 1. Properties of Nanomaterial[17]

Nano material	**c_p [J/kg*K]**	**ρ [kg/m^3]**	**k [W/m*K]**
Copper (spherical)	385	8933	400
CNT (1Dimension)	425	2600	6600

In the above model, cylindrical cell having with 4 rectangular fin which is cylindrical ring. The illustration as shown in Figure 3.

2.3 Governing Equation

The enthalpy porosity method is frequently used in ANSYS FLUENT to model solidification and melting phenomena. This method is grounded on a set of fundamental assumptions[9,10]an extensive review of a Battery Thermal Management Systems (BTMSs) (BTMSs which include the application of Boussinesq approximation for density due to the low ratio of (Δρ)/(ρ) for PCM, the homogeneity and isotropy of PCM's solid and liquid phases, the predominance of conduction for heat transfer in PCM, and the neglect of radiation in the heat transfer process. Additionally, it is assumed that the flow is laminar, transient, and incompressible. Based on these premises, the governing equations are formulated, and the continuity equation is one such equation. The assumptions outlined above serve as the basis for defining the governing equations.

Continuity equation

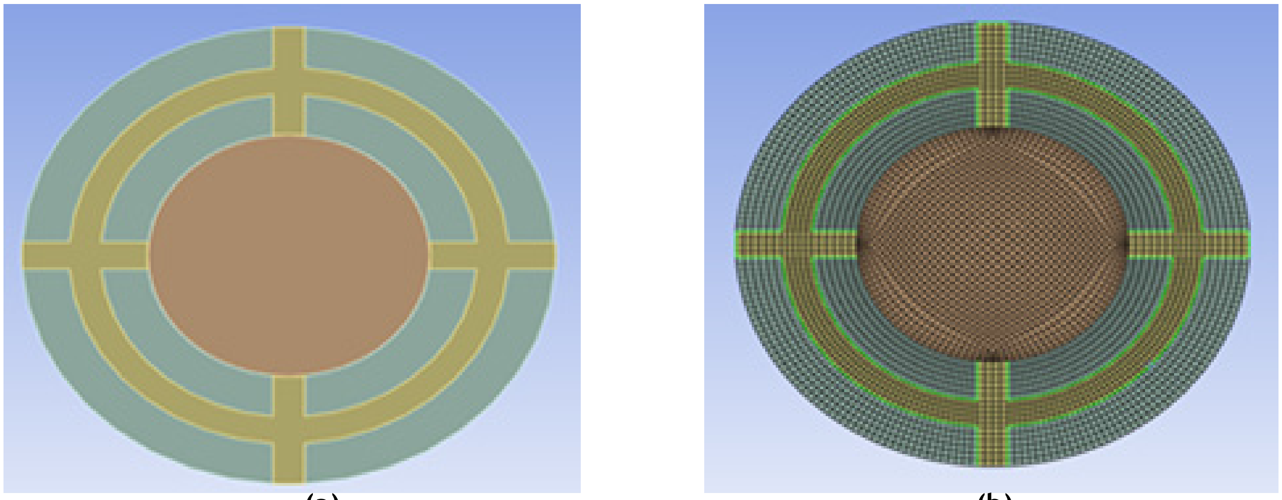
$$\frac{d\left(\rho_{pcm}\right)}{dt}+\frac{d\left(\rho_{pcm}u\right)}{dx}+\frac{d\left(\rho_{pcm}v\right)}{dy}=0 \tag{9}$$

Momentum equation

$$\frac{d\left(\rho_{pcm}u\right)}{dt}+\frac{d\left(\rho_{pcm}u^{2}\right)}{dx}+\frac{d\left(\rho_{pcm}uv\right)}{dy}=\frac{dp}{dx}+\frac{d}{dx}\left(\mu\frac{du}{dx}\right)+\frac{d}{dy}\left(\mu\frac{du}{dy}\right)+\rho g+S \tag{10}$$

Table 2. Thermophysical Properties

Sl. No.	**Properties**	**RT-42[18]**	**Li-ion[19]**	**Aluminium**	**Acrylic**
1	Density, (kg/m^3)	820	2720	2719	1215
2	Mass, (g)		44.5		
3	Heat capacity, (J/kg*K)	2,000	300	871	1300
4	Thermal conductivity (W/m* K)	0.2	3	202.4	0.17
5	Dynamic Viscosity (kg*m^{-1}*s^{-1})	0.02			
6	Solidus Temperature, (K)	311.15			
7	Liquidus Temperature, (K)	316.15			
8	Melting Heat (kJ/kg)	165			
9	Thermal expansion coefficient (/K)	0.0001			
10	Dimension (mm)	--	18 × 65		



(a) **(b)**

Figure 3. (a) Proposed model. (b) Mesh generation of Proposed model.

Energy equation

$$\frac{d(\rho_{pcm}h_e)}{dt}+\frac{d(\rho_{pcm}uh_e)}{dx}+\frac{d(\rho_{pcm}vh_e)}{dy}=\frac{d}{dx}\left(k_{pcm}\frac{dT}{dx}\right)+\frac{d}{dy}\left(k_{pcm}\frac{dT}{dy}\right) \tag{11}$$

The enthalpy porosity method, which is commonly used for simulating Phase Change Material (PCMs), involves a set of energy equations[20]. These equations can be expressed as follows:

$$H = h_e + \Delta H \tag{12}$$

$$h_e = h_{ref} + \int_{T_{ref}}^{T} C_\rho \, dT \tag{13}$$

The variable "h_e" denotes the sensible heat, while "h_{ref}" represents the reference enthalpy at temperature "T_{ref}". As the Phase Change Material (PCM) melts at varying temperatures, its latent heat can be expressed as follows:

Where β is the liquid fraction, is given by

$$\Delta H=\beta L \tag{14}$$

$\beta=0$ if $T<T_s$

$$\frac{T-T_s}{T_l-T_s} \, if \; T_s < T < T_l$$

1 if $T>T_i$

The temperatures at which the substance transitions from a solid to a liquid state are denoted by T_s and T_i where T_s is the solidus temperature and is the liquids temperature.

2.4 Boundary Condition

In the numerical simulation conducted for this study, the initial temperature of the system at time t = 0 was set to T_i = 300 K. Heat generation from a source was then applied to the cell at various current rates, specifically 1C, 2C, and 3C, which corresponded to heat rates of 10.44 kW/m^3, 41.78 kW/m^3, and 94.02 kW/m^3, respectively. To account for natural convection, the simulation utilized an experimentally determined heat transfer coefficient (h) of 5.7 W/m^{2}K for the acrylic shell[21].

2.5 Sensitivity Analysis

To optimize computational efficiency, grid independence was assessed for all designed configurations to determine the most effective grid size. A study of time step independence was also conducted, using 5093 elements and time steps of 0.1, 0.5, and 1s. No variation in liquid fraction was observed with adjustments to the time step. The current computational task utilizes 5093 elements and a time step of 0.5s.

2.6 Computational Procedure

The finite volume technique was employed to solve the energy, momentum, and mass equations, and the ANSYS FLUENT CFD tool was utilized to conduct simulations. To address this issue, the pressure-velocity coupling was handled using the SIMPLEC algorithm, while the pressure correction equations in transient state computations employed the PRESTO! Pressure interpolation scheme. Under-relaxation factors were incorporated, and the resolution of the energy and momentum equations involved second-order upwind discretization. The numerical investigation was conducted at an ambient temperature of 300K, with melt fraction, thermal energy, density, and momentum values of 0.9, 0.7, 1, and 0.7 respectively. The pressure correction factor was set at 0.3.

3.0 Result and Discussions

3.1 Thermal Response of Lithium-ion Cell to Various C-rates in Absence of Phase Change Material (PCM)

The performance of the cell was evaluated under current rates of 1C, 2C, and 3C without any phase change material (PCM). Internal heat generation was calculated to be 10.44 kW/m^3, 41.78 kW/m^3, and 94.02 kW/m^3 at 1C, 2C, and 3C respectively. At a lower current rates up to 2C the temperature of a lithium-ion cell was observed to be below 40°C (313.15 K), which is considered a safe temperature range for batteries. However, as the current rate increased to 2C and 3C, the temperature of the battery also increased and exceeded the safe temperature range, as shown in Figure 4.

The determination of the temperature distribution within a solid cylinder containing an internal region is contingent upon the imposition of appropriate boundary conditions.

$$Tr \ = T_a + q_g R/2h + q_g/k\,(R^2 - r^2) \tag{15}$$

Where T$_a$ is ambient temperature, q$_g$ is internal heat generation heat transfer coefficient is radius of solid cylinder.

By substituting the appropriate values at r = 0, the maximal temperature can be determined for 1C, 2C, and 3C, as depicted in Figure 4.

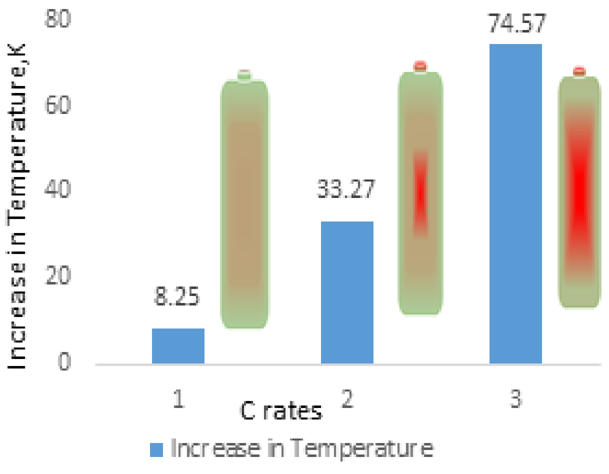


Figure 4. Temperature variation in Li ion cell at different C rates.

As previously discussed, the utilization of metal fins has the potential to extend the thermal boundary, facilitating superior heat transfer by enabling the heat source to penetrate the PCM. As a result, the numerical battery module has been modified to include fin application, which will be elaborated upon in subsequent sections.

3.2 Combined Effect of Position of Cylindrical Ring Attached to the Fin and Nano Additives in PCM Module

For the study of proposed BTMS, in the simulation, a cylindrical ring was placed in contact with the battery, and its position was fixed. This section evaluates the thermal management performance of the BTMS by conducting simulations to investigate the impact of the cylindrical ring attached to the fins. Illustration is shown in Figure 5.

Cylindrical rings are attached to the fins in PCM-based BTMS to improve the heat transfer efficiency between the fins and the PCM. The rings create a gap between the fins and the PCM, allowing for better airflow and reducing the boundary layer effect that exists between the two surfaces. This results in improved heat transfer rates and more effective regulation of temperature fluctuations in the battery. The rings also provide a mechanical buffer that helps prevent damage to the fins and the PCM due to thermal expansion and contraction during the heating and cooling cycles. Additionally, the use of cylindrical rings helps maintain a consistent spacing between the

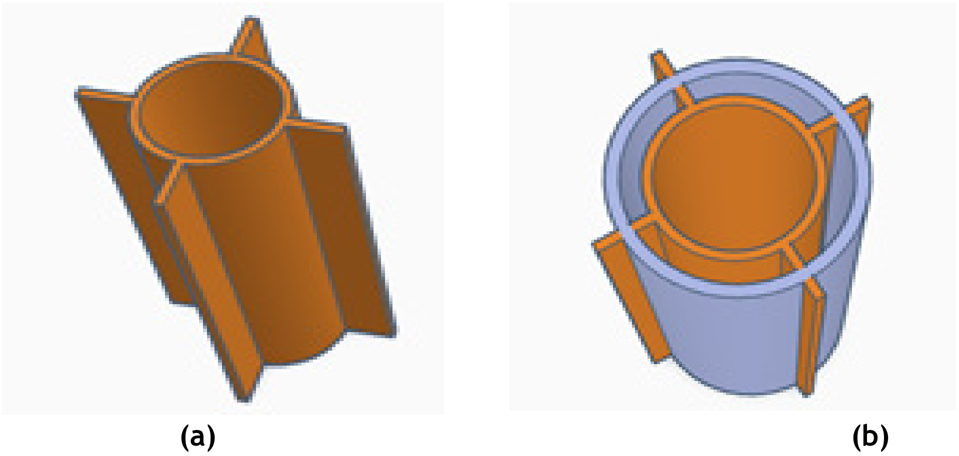


Figure 5. (a) Only Fin. (b) Cylindrical ring attached to the 4 Fins.

fins and the PCM, leading to more uniform heat transfer and better overall performance of the BTMS. Overall, the addition of cylindrical rings to PCM-based BTMS represents a promising approach for improving heat transfer properties and the efficiency of thermal energy storage systems.

The position of the ring is indicated by the dimensionless Distance (D*) is a parameter used to indicate the position of a cylindrical ring, which is defined as the ratio of the radial distance of the ring to the diameter of the battery. To achieve a greater heat storage capacity and heat dissipation capability, it is necessary to optimize the quantity of cylindrical rings. The current study, takes into account the volume of the 18650 battery and previous literature on the subject[15].

Nanomaterial are added to Phase Change Materials (PCMs) to improve their thermal properties, including their melting and solidification rates, thermal conductivity, and stability. In BTMS fins are commonly used to increase heat transfer, but the addition of nano composites can further enhance the heat transfer properties of the PCM. This can lead to more efficient and effective BTMS systems that better regulate temperature fluctuations in battery module.

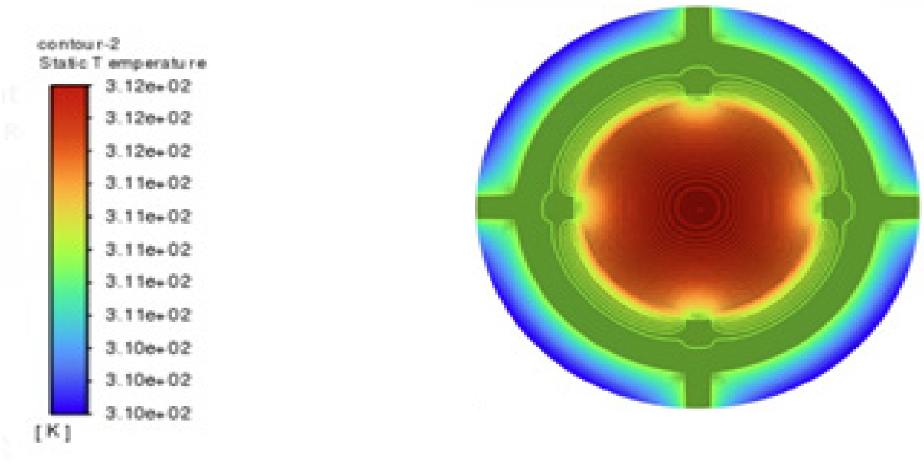
In the current investigation, a NePCM module was utilized to examine the thermal behaviour of two distinct Nano enhanced PCMs, specifically those containing metal-based and carbon-based nanoparticles. The module contained a cylindrical ring featuring four rectangular fins that were affixed at the centre.

3.2.1 Metal Based Nano Additives in Proposed Module

The Proposed PCM modules with 4 fins for BTMS have shown promising results when using copper nanoparticles. Copper nanoparticles have improved the PCM's thermal conductivity and rate of heat transfer, allowing for more effective cooling and heating of buildings. By increasing the surface area and promoting better air flow, the fins in the module further enhance the heat transfer performance. Figure 6(a) shows the temperature distribution, where the highest temperature on the cell reaches 312 K at 3C rate. Additionally, the liquid fraction profile shown in Figure 6(b) indicates that the maximum liquid fraction occurs near the cell with a value of 0.038 (about to melting), while the minimum liquid fraction is observed away from the cell with a value of 0 (solid).

3.2.2 Carbon Based Nano Additived in Praposed Module

In this study we are used SWCNT (Single Walled Carnon Nanotube), have a much smaller diameter and higher surface area compared to MWCNTs (Multi Walled Carnon Nanotube), which can improve the interfacial contact between the SWCNTs and the PCM, leading to improved thermal conductivity. A PCM's thermal behaviour can be improved by adding SWCNTs by increasing thermal conductivity, lowering melting point, and increasing latent heat storage capability. A number of variables, including the concentration and size of the



(a)

Figure 6. (a) Temperature contour for Cu based NePCM at 3C rate.

(b)

Figure 6. (b) Liquid fraction contour for Cu based NePCM at 3C rate.

SWCNTs, their dispersion within the PCM, and the thermal characteristics of the PCM itself, may affect the precise impact of SWCNTs in a given PCM, such as RT42. Figure 7(a) depicts the temperature distribution, with the cell's highest temperature reaching 312 K at a 3C rate and remaining nearly constant throughout. Furthermore, Figure 7(b)'s liquid fraction profile shows that the maximum liquid fraction, which is 0.0738 (about to melt), occurs close to the cell, while the minimum liquid fraction, which is 0.0508 (started to melt), indicates that heat transfer is occurring at NePCM.

4.0 Conclusion

The present research article details a numerical study conducted an 18,650 Li-Ion battery, aimed at analysing its performance during a 1200s discharge at a 3C rate while considering the impact of different Crates and further analysis to ensure its safe and efficient operation within established operational limits using different nano additives. As a result, it can be inferred that

- To optimize the battery's performance and temperature control, an assessment of the optimal fin number and PCM thickness was carried out based on prior research findings. The study involved a calculation of the battery's temperature at different C rate.

- The proposed module can create a thermal conductive network within the NePCM, which can quickly transfer heat produced in the battery to the NePCM and significantly extend battery life.
- Copper nano particle can improve the thermal conductivity of the PCM due to its high surface area and the ability to form a stable network within the PCM. However, the addition of copper nano particle can also lead to an increase in viscosity and a decrease in the latent heat storage capacity of the PCM.
- On the other hand, SWCNT can also enhance the thermal behaviour of PCMs used in BTMS by increasing their thermal conductivity, reducing their melting point, and improving their latent heat storage capacity. The smaller size and high aspect ratio of SWCNTs allow for a larger interfacial area with the PCM, resulting in greater heat transfer. However, proper dispersion techniques must be employed to ensure optimal performance of SWCNT-enhanced PCM.
- Ultimately, the most suitable nanomaterial for BTMS depends on the specific application requirements and the properties of the PCM being used. Further experimental and optimization studies are needed to fully understand the impact of these nanomaterials and their optimal conditions for use in BTMS.

5.0 Acknowledgments

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Nomenclature

k	thermal conductivity (W/m-K)
h	heat transfer coefficient (W/m^2-K)
H	enthalpy (J/kg-K)
ρ	density (kg/m^3)
Cp	specific heat (J/kg-K)
T	Temperature (K)
t	Time (s)
Subscripts	
PCM	phase change material
b	battery
a	ambient
s	solid
l	liquid
m	melting
ref	reference