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Article

Experimental exploration of nano-phase change material composites for thermal management in Lithium-ion batteries

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Abstract: The present study reports an experimental investigation carried out for the thermal management of cylindrical lithium-ion battery simulator using aluminium oxide (nano particle)-eicosane (phase change material) composites. The experiment involves varying the power input from 4 to 10 W in 2 W increments and adjusting the weight percentage of nanoparticles (wt %) from 0.5 to 0.9 in 0.2 wt % intervals. The examination of battery temperature evolutions in response to heating power, a comprehensive heat transfer analysis incorporating Nusselt number, determination of maximum temperature difference, thermal resistance analysis, and exploration of temperature variations in the absence of Phase Change Material (PCM) are considered. The results show that increase in weight percentage of alumina nanoparticles in phase change material cannot always improve the thermal performance. The results of the present study give a guideline for designing battery thermal management system. The power levels used in the experiment vary from 4 W to 10 W in steps of 2 W. For a power level of 4 W, the heat flux is 1.088 kW/m^2, and for a power level of 10 W, the heat flux is 2.72 kW/m^2.

Keywords: nanomaterials; thermal management; phase change materials; mock up battery

1. Introduction

The need for power storage and sources has been steadily increasing due to technological advancements, with approximately 80% of this energy being sourced from fossil fuels. However, it's crucial to shift focus towards renewable, sustainable, and environmentally friendly energy sources. Billions of batteries are manufactured annually, highlighting their significance in various comfort and safety applications. Enhancing the power performance and lifespan of batteries is imperative. Factors such as round trip efficiency, electrochemistry, reliability, power, energy capability, and cycle life are greatly influenced by high temperatures. Efficient thermal management systems are necessary for effective heat dissipation during battery charging and discharging processes. Integrating phase change materials (PCM) into battery thermal management systems (BTMS) can offer an efficient, reliable, and eco-friendly solution, particularly for fluctuating discharge systems. Recent research has investigated the heat transfer properties of base fluids using suspended nanometer-sized solid particles, or nanoparticles, which enhance heat transfer efficiency. Combining PCM with nanoparticles holds promise in maintaining battery temperatures within safe limits. Khlissa et al. [1] explored the critical role of PCMs in thermal energy storage, essential for reducing building energy consumption and achieving net-zero energy goals. Despite their growing importance, PCMs faced limitations due to subpar heat conductivity. The study extensively categorized and examined PCM features, highlighting recent advancements in nanoencapsulated PCM techniques and thermal energy storage technology. The assessment underscored researchers' efforts in enhancing PCMs, particularly through nano-enhanced PCMs.

Many researchers have experimentally studied about the thermal management of LIB using phase change material. Arshad et al. [2] explored the thermal performance enhancement in pin-fin heat sinks by employing n-eicosane as a PCM. Various configurations with different volumetric fractions of n-eicosane and aluminum thermal conductivity enhancers (TCEs) were investigated to optimize cooling efficiency. Results indicated that a 2 mm thick pin-fin heat sink with n-eicosane provided the most effective thermal performance, ensuring reliable temperature control for electronic packages. In their study, Wang et al. [3] investigated the thermal performance of cylindrical batteries using a composite paraffin and fin structure. They found that the composite fin structure effectively managed higher heat densities with minimal thermal resistance and extended operational durations. Experiments were conducted with both pure PCM and finned cases, revealing that the composite PCM-fin system displayed superior thermal performance compared to pure PCM.

Similarly, Hussain et al. [4] conducted experiments employing graphene-coated nickel foam saturated with PCM to analyze thermal management in lithium-ion batteries (LIBs). Their findings indicated a significant improvement in the thermal conductivity of pure paraffin wax by 23 times after infiltration into GcN foam. Moreover, the freezing and melting temperatures of graphene-coated nickel foam saturated with paraffin wax were altered compared to pure paraffin. In a separate study, Babapoor et al. [5] investigated the thermal performance of a LIB simulator in the presence of PCM. Their results demonstrated that a mixture of PCM with 2 mm long carbon fibers at a mass percentage of 0.46% exhibited the most favorable thermal performance, leading to a reduction in temperature rise within the battery simulator by nearly 45%.

Situ et al. [6] proposed a double copper mesh-phase change material plate (DCM-PCMP) method for heat dissipation, specifically tailored for rectangular battery modules. This approach effectively increased thermal conductivity and, when combined with air-cooling, significantly improved thermal management, reducing internal battery temperature and power consumption. Karimi et al. [7] investigated thermal management in cylindrical lithium-ion batteries (LIBs) using different phase change material (PCM) composites. By mixing pure PCM with Ag, Cu, and Fe_3O_4 nanoparticles and metal matrix, they enhanced thermal conductivity. Ag nanoparticle-PCM composites exhibited superior performance, while metal matrix-PCM composites loaded with PCMs achieved the lowest battery body temperatures. Pan and Lai [8] studied the thermal performance of LIBs using copper/paraffin composites phase change material (CPCM). They tested four heat dissipation methods under various discharging currents and found that CPCM effectively absorbed heat, reducing battery temperature by about 50% compared to natural wind cooling, with uniform temperature distribution.

Wang et al. [9] explored the use of paraffin and paraffin/aluminum foam composites PCM for LIB thermal management. They observed improved melting processes and temperature uniformity with the addition of aluminum foam, with the composite PCM exhibiting significantly higher effective thermal conductivity compared to pure paraffin. Ling et al. [10] investigated two PCM composites with different thermal conductivities and their impact on battery thermal performance. They found that RT44 HC/EG composite outperformed RT44 HC/fumed silica composite, reducing maximum temperature difference by up to 60 ℃. Hussain et al. [11] developed a nickel foam-paraffin wax composite for thermal management of high-power LIBs, demonstrating a 31% and 24% reduction in temperature compared to natural cooling and pure PCM, respectively, at discharge rates less than 2 ℃.

Bai et al. [12] proposed a LIB module with a phase change material/water cooling plate, effectively cooling the battery's heat generation area and preventing thermal runaway. The PCM between adjacent batteries improved temperature uniformity. Rao et al. [13] utilized a battery module with six cells along flow channels to study the effects of aluminum block length and velocity on thermal performance, finding that maximum temperature remained under 40 ℃ with specific inlet conditions.

Wu et al. [14] developed a paraffin/expanded graphite composites-based battery module (PCM module) and a two-dimensional thermal model, recommending a weight fraction of 15%–20% for expanded graphite for optimal thermal management. Alipanah and Li [15] investigated the thermal management system of LIBs using pure octadecane, pure gallium, and octadecane-Al foam composite materials, observing improved discharge time and reduced battery surface temperature with the use of composite materials. Jiang et al. [16] developed a tube-shell LIB pack with a PCM passive thermal management system coupled with forced air cooling, ensuring uniform temperature distribution and high heat dissipation efficiency.

Zhao et al. [17] optimized a steel cell with a PCM thermal management system using numerical simulation, achieving uniform temperature distribution and increased energy density. Lazrak et al. [18] studied thermal conductivity enhancement and PCM melting temperature effects using integrated BTMS, developing a prototype based on numerical studies. Babu Sanker and Baby [19] provided a review on the thermal performance of various thermal conductivity enhancers in PCM-based thermal management of LIBs, summarizing advancements in the field.

The research delves into the characteristics of nano-enhanced PCMs. The study focuses on investigating the unique properties and performance improvements resulting from the integration of nanomaterials in PCMs. Insights gained from these studies contribute to advancing the understanding and potential applications of nano-enhanced PCMs in various thermal management systems. Liu et al. [20] studied the impact of optical properties with the use of glazing structures incorporated with PCMs. The study revealed that the refractive index of the PCM had a limited effect on various parameters, while the extinction coefficient significantly influenced temperature characteristics and optical properties. The research suggested caution in using liquid PCM with a large extinction coefficient for optimal light utilization but deemed a high extinction coefficient in solid PCM acceptable for efficient energy utilization. Li et al. [21] investigated the thermo-physical parameters of PCM on the thermal performance of a PCM-filled double glazing unit. Results indicated that increased density, latent heat, and melting temperature of the PCM effectively improved thermal performance, resulting in an elevated temperature time lag and reduced temperature decrement factor. However, the study found that enhancing thermal conductivity and specific heat capacity became ineffective beyond certain thresholds, specifically when thermal conductivity exceeded 2.1 W/(m K) and specific heat capacity was less than 4460 J/(kg K).

The application of PCM involves harnessing its ability to absorb and release thermal energy during phase transitions. This technology is widely used in various industries, such as building construction and electronics, to enhance thermal performance and energy efficiency. Salih et al. [22] baffles attached alternately to the upper and lower walls. The channel contained PCM in the semi-cylinders, heated at a constant temperature, with cold air inducing convective and conductive heat transfers, fluid-structure interaction, and PCM melting. The findings, obtained through solving normalized mathematical equations with the finite element method and ALE scheme, revealed that increasing Reynolds number and decreasing the elasticity modulus of flexible baffles retarded the melting volume fraction, with notable effects on Nusselt number and pressure drop along the channel. Chen et al. [23] used PCM to cool lithium-ion batteries in electric cars was examined. A cylindrical battery submerged in a PCM-filled chamber with varying fin configurations aimed to optimize cooling during the discharging process. Utilizing COMSOL Multiphysics software, simulations revealed that a battery with 15 fins exhibited optimal PCM melting performance initially, while the lowest maximum temperature and maximum liquid volume fraction occurred with 9 fins, providing valuable insights for effective battery cooling strategies. Ruhani et al. [24] simulated the cooling system of a two-dimensional LIB pack with nine cells, considering airflow at Reynolds numbers ranging from 80 to 140. The study, employing the finite element method, individually examined the temperature of each battery cell, assessed pressure drop, and analyzed the cooling system temperature. Results indicated that an increase in Reynolds number lowered the maximum temperature of the battery pack, while variations in intake size affected the maximum temperature of battery cells and intensified pressure drop in the cooling system. Rashid et al. [25] highlighted the efficacy of latent heat energy storage using PCM to reduce energy consumption. Despite the fluctuating price of paraffin wax, a common PCM derived from fossil fuels, interest grew in bio-based PCMs for sustainable thermal energy storage. Bio-based PCMs, derived from readily available and biocompatible sources such as animal fat combinations and oils, were explored as a renewable alternative with reduced oxidation risk compared to paraffin wax. The study reviewed and discussed the selection, phase change mechanisms, combinations, preparation, and applications of bio-based PCMs, proposing improvements for diverse utilization in thermal energy storage applications. Kholsi et al. [26] conducted a investigation PCM energy storage tank utilizing carbon nanotube (CNT)–water nanofluid under actual climatic conditions in Ha'il, Saudi Arabia. Two configurations, with and without conductive baffles, were examined. The tank, filled with encapsulated paraffin wax as PCM and circulating CNT–water nanofluid, aimed to raise PCM temperature to 70 °C for thermal energy storage during night and cloudy weather. Finite element method simulations, based on measured weather conditions over three months (December, March, and July), revealed that CNT–nanofluid usage reduced charging time and enhanced performance, with the best results observed in July due to high solar irradiation. Additionally, baffles showed no beneficial effects on the melting process.

From the preceding discussions, it is clear that several investigations have been carried out to study the thermal management of cylindrical LIB. But experimental investigations using nanofluids is scarce. Thus, aim of the present work is to

1) To perform experiments in order to determine the effect of the following parameters on the heat transfer performance.
    - Different concentrations of nanofluid—0.5 wt %, 0.7 wt % and 0.9 wt %.
    - Various input power levels—4 to 10 W in steps of 2 W.
2) To compare the performance of battery without phase change material.

In the present study thermal management of lithium-ion battery (LIB) using nano Phase Change Material Composites in different concentrations (Case 1, Case 2, Case 3) are done. Case 2 demonstrates optimal battery temperature reduction, enhanced heat transfer, and the lowest thermal resistance. Nusselt number enhancement of 20% is observed, with a remarkable 70% reduction in battery temperature. Case 2 limits the temperature difference to 50 ℃ and cools the battery below 40 ℃ within 200 seconds at a 4 W heat input. These findings highlight the effectiveness of nano-based PCM composites for improved battery thermal management.

2. Experimental setup

The test setup comprised a simulated battery heater enclosed within a metal casing and filled with phase change material (PCM) in between. The mock battery, constructed from aluminum, measured 18 mm in diameter and 65 mm in height. It featured a concentric hole of 6 mm diameter at the center to accommodate a heater (3.56 cm in diameter and 5 cm in length), replicating the heat generation of a commercial 18,650 lithium-ion battery (LIB). The battery, with or without aluminum oxide nanoparticles, was positioned vertically within an aluminum housing with an inner diameter of 36 mm and a wall thickness of 10 mm. To insulate the battery from the metal housing, the sides and bottom were shielded with a 25 mm layer of nylon. With slight modifications of procedures described by Radhakrishnan et al. [27] and Kothari et al. [28], nanoparticles is mixed with n-eicosane PCM. Initially, the required weight percentage of nanoparticle was carefully measured and added to heated n-eicosane using a Rotek Magnetic Stirrer cum hot plate. After melting, the nanoparticle-infused PCM underwent sonication at 50 ℃ for 2 h using an Analab sonicator. To prevent sedimentation, the mixture was further homogenized for 30 minutes with a Stirrer. The same procedure is followed for all weight percentages of nanoparticles. The PCM, eicosane used in this study was supplied by M/s Sigma Aldrich.

Nanoparticles with different weight fractions, namely, 0.50 wt %, 0.70 wt % and 0.90 wt % are used in conjunction with PCM and the case without nanoparticles (pure PCM) is used as baseline for comparison. The battery temperature was measured by nine calibrated K-type thermocouples. The experiment was started by turning on the data acquisition system (Keysight 34972A) and power supply. **Figure 1** shows the photograph the experimental setup used in the present study.

Figure 1. A photograph of the experimental setup used.

2.1. Model assembly

The mock-up battery, metal housing, insulation cavity, heater and acrylic plate are assembled to form the model assembly. **Figure 2** represents the design of the model assembly.

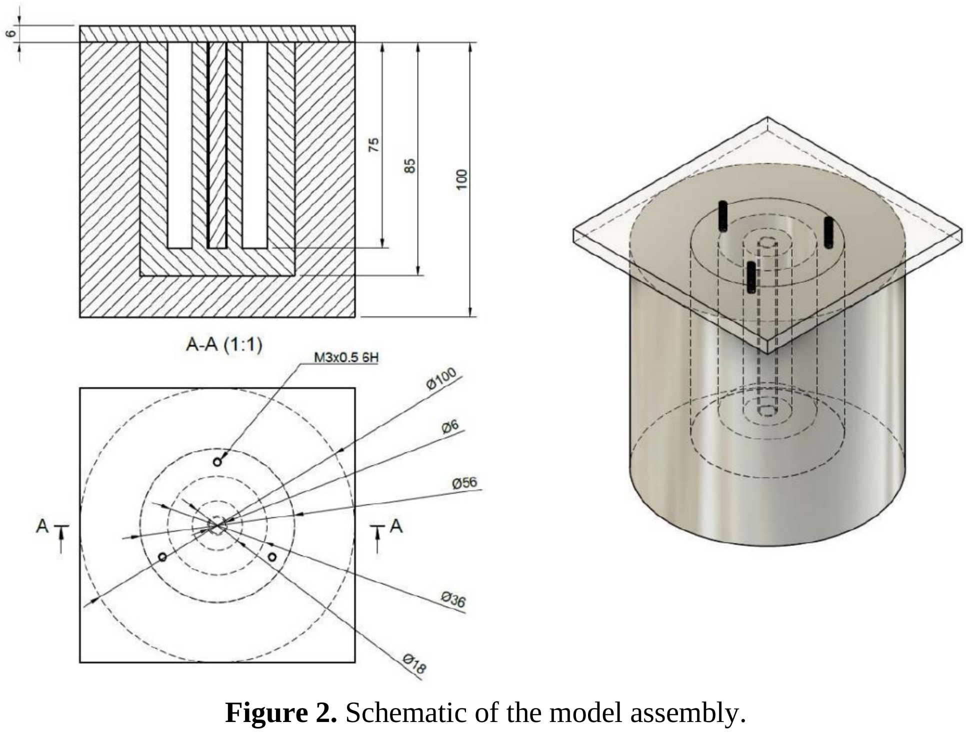

Figure 2. Schematic of the model assembly.

The mock up battery screwed in acrylic plate and bolted top of metal housing. The cartridge heater was inserted into the hole drilled at the centre of the mock-up battery. The thermocouple fixed at various positions and PCM composites melted and poured into the metal housing. The thermocouple wires are taken out through the holes made at the acrylic plate.

2.2. Equations used in the present study

The PCM (eicosane) based alumina nano composite with mass concentrations of 0.5, 0.7 and 0.9 wt % and pure PCM was used to investigate the thermal management of cylindrical LIB. The temperature drops between battery temperature at the effective thermal control point (T_c) and maximum melting temperature of PCM (T_m) is of particular interest to evaluate its thermal performance. The overall thermal resistance of the system is calculated from:

$$R_{cm} = \frac{T_c - T_m}{Q} \tag{1}$$

where Q is the heat input. In the present heat transfer analysis, the instantaneous Nusselt number is defined with the instantaneous battery to the housing temperature difference, namely,

$$N_u = \frac{QW}{(Tb - Tw)A_b\, k_f} \tag{2}$$

The Nusselt number shows the quality of heat transfer.

2.3. Uncertainty analysis

The results of the uncertainty analysis of the experiments are reported in **Table 1**. The final uncertainty in power was estimated to be ±2.4% for a power level of 4 W.

Table 1. Results of the uncertainty analysis.

Sl. no.	Quantity measured	Uncertainty
1	Temperature	±0.2 ℃
2	Voltage	±0.1 V
3	Current	±0.01 A
4	Power input	±2.4%

3. Results and discussion

The experiment involved utilizing power levels ranging from 4 W to 10 W in increments of 2 W. At 4 W, the heat flux measured 1.088 kW/m^2, while at 10 W, it reached 2.72 kW/m^2. The objective of this investigation is to analyze the thermal properties of the composite PCM-nanoparticle-based thermal management system. **Table 2** provides the nomenclature list for the samples.

Table 2. List of nomenclatures of samples.

Sample	Label
Pure PCM	Case 1
0.50 wt % Nano -PCM composites	Case 2
0.70 wt % Nano -PCM composites	Case 3
0.90 wt % Nano- PCM composites	Case 4

3.1. Battery temperature evolutions with heating power

Experiments were carried out for the battery contained in the housing for various cases are described in this section. The detailed battery temperature graph for 8 W is described in **Figure 3**. Case 2 shows the lower battery temperature curve. As compared to the case 1 about 4 ℃ to 6 ℃ difference is shown by case 2. All the nano based PCM composites shows better heat transfer performance than case 1. The nanoparticle addition in pure PCM increases the thermal conductivity of PCM composites and it helps uniform heat distribution inside the PCM composites.

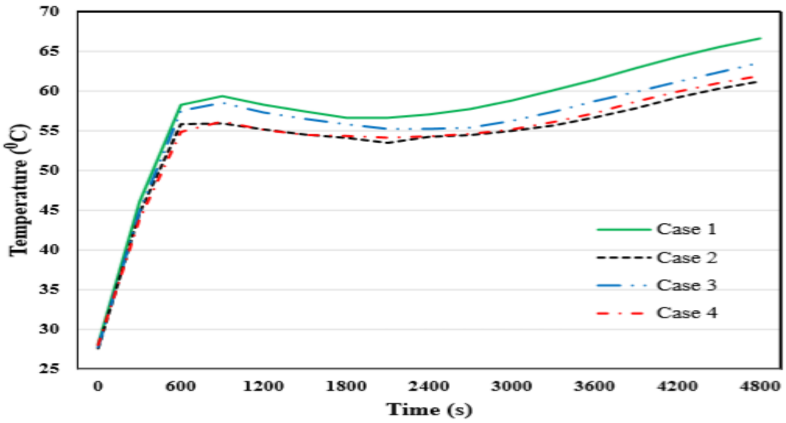

Figure 3. Battery temperature–time response for a heat input 8 W.

3.2. Heat transfer analysis with Nusselt number

In the case of Nusselt number (Nu) for heat input 8 W, case 2 and case 4 have superior Nusselt number values. Instantaneous Nusselt number for 8W heat input is shown in the **Figure 4**.

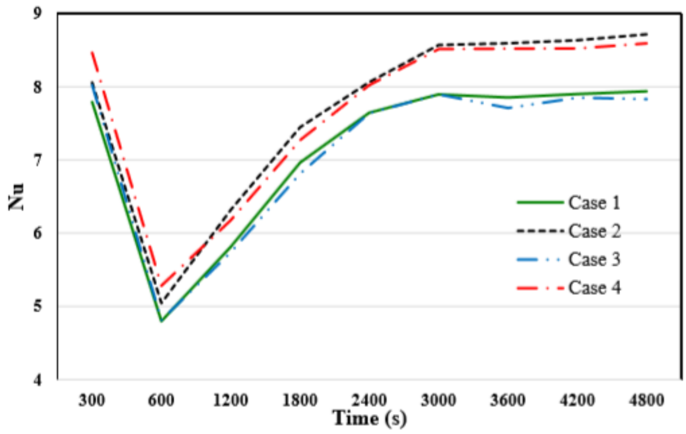

Figure 4. Instantaneous Nusselt number for heat input 8 W.

3.3. Maximum temperature difference

Battery thermal management has some main objectives including decreasing battery body temperature, increasing effective thermal conductivity of the cooling medium, and maintain the temperature uniformity inside the system by minimize the temperature difference between battery and cooling medium. The presence of nanoparticles in PCM can provide a better heat transfer path through the cooling medium. The maximum temperature difference at 4800 s for 4 W power is shown in the **Figure 5**. Case 1 shows the greater maximum temperature difference of 13 °C. Case 2 having the lower maximum temperature difference of 3 °C. Around 10 °C temperature difference between case 1 and case 2. Case 4 shows higher maximum temperature difference among the three nano-PCM composites.

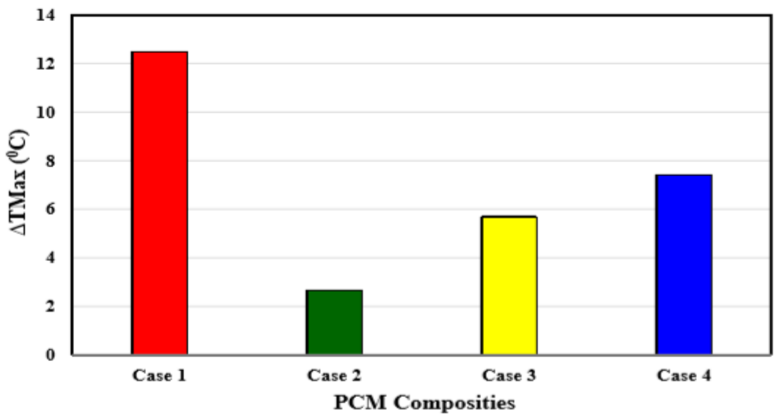

Figure 5. Maximum temperature difference at 4800 s for 4 W heat input.

3.4. Thermal resistance analysis

The thermal resistance is obtained from Equation (3)

$$R_{cm} = \frac{T_c - T_m}{q} \tag{3}$$

The obtained thermal resistance results for different nano based PCM composites are discussed below. It is seen that the thermal resistance does decrease with the increase in percentage weight of nano particles in phase change material. The improved heat transfer causes the decrease in thermal resistance within the thermal management system. **Figure 6** indicates the thermal resistance at 8 W power level. Case 1 gives high thermal resistance and case 2 gives low thermal resistance and is suitable for better thermal management inside the system. Case 3 shows much higher thermal resistance than case 2 and 4.

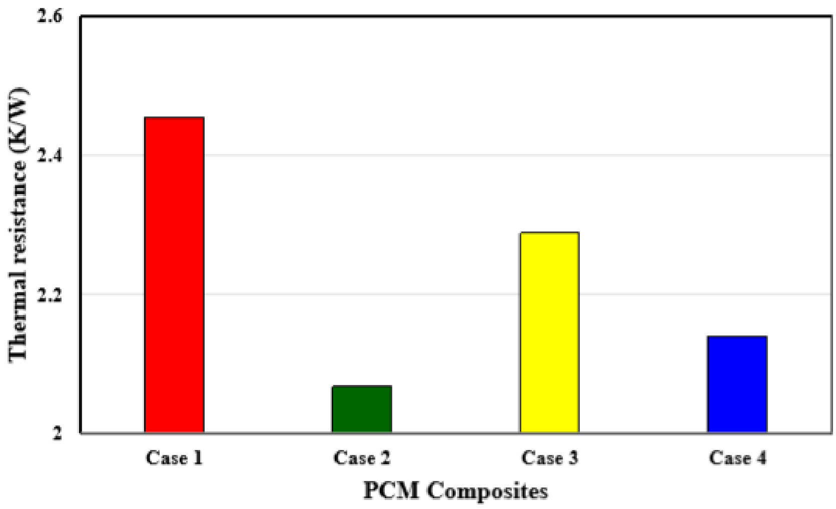

Figure 6. Thermal resistance for 8 W heat input.

3.5. Temperature variations without PCM

Battery temperature variations and maximum temperature difference without PCM are discussed in this section. The battery temperature can limit within a desired range using PCM composites. **Figure 7** shows the battery temperature variation for different heat inputs without PCM. For 4 W, 6 W, 8 W and 10 W, heat input temperature reached at 4800 s are118 °C, 152 °C, 180 °C and 212 °C respectively. **Table 3** shows the battery temperatures at 4800 s with PCM composites and without PCM. The effectiveness of the PCM composites in the temperature control of battery temperature can get directly from the values in **Table 3**. The PCM composite reduces the battery temperature around 60 to 70 percentage at various power levels.

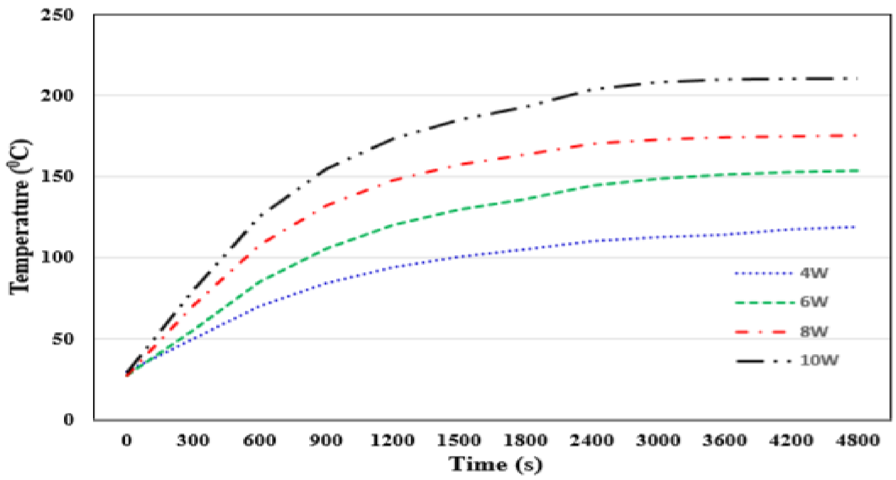

Figure 7. Battery temperature variation without PCM.

Table 3. Battery temperatures at 4800 s with PCM composites and without PCM.

Sl No	Power(W)	PCM composite	Without PCM
1	4	46 °C	118 °C
2	6	52 °C	152 °C
3	8	61 °C	180 °C
4	10	68 °C	212 °C

4. Conclusions

The present work comprises experimental investigations of thermal behaviour of a composite structure thermal management system. The following parameters were analysed:

- Varying heat inputs (4 to 10 W in steps of 2 W).
- Varying nano-particles weight percentage in the PCM (0.50 wt %, 0.70 wt % and 0.90 wt % of PCM).

Nano based phase change material composite structure was studied by examining the representative time points, effect of heat input and different weight percentage of nano particle PCM composites. To control the temperature and to maintain the temperature uniformity inside the BTMSs system temperature difference between battery and cooling medium was decreased. The different trends and results from the analysis is discussed below.

- Battery temperature evolution was studied for various heat inputs, revealing optimal performance with a 4 W heat input in case 4.
- Case 2 consistently demonstrated lower battery temperatures than other nano-based PCM composite structures and the pure PCM case for heat inputs of 6 W, 8 W, and 10 W. Even at a high heat input of 10 W, case 2 limited the temperature below 70 °C.

- Nano particle PCM composites consistently exhibited lower battery temperatures than the pure PCM case across all power levels, attributed to increased thermal conductivity from the addition of nano particles.
- Instantaneous Nusselt number analysis indicated that case 2 outperformed other composites at heat inputs of 6 W, 8 W, and 10 W, with a 20% increase compared to other cases. Improved Nusselt numbers signify enhanced heat transfer within the cooling medium due to nano particle addition to paraffin.
- The superior Nusselt number in case 2 contributed to an improved temperature distribution in the BTMS.
- Nano-based PCM composites consistently demonstrated lower temperature differences between the battery and cooling medium for all heat inputs, indicating a more uniform temperature distribution within the BTMS. Pure PCM exhibited higher temperature differences due to its lower thermal conductivity.
- Case 2 exhibited the least temperature difference between the battery and PCM medium for all heat inputs, limiting the difference to 50 ℃ compared to other cases.
- Thermal resistance in the cooling medium, affecting heat transfer, was reduced by the addition of nano particles, resulting in lower thermal resistance for PCM composite structures compared to the pure PCM case. Case 2 had the lowest thermal resistance, decreasing by 25% compared to other cases.
- In comparison to a case without PCM, the nano-based PCM composite structure demonstrated a remarkable reduction in battery temperature by up to 70%.
- The maximum temperature difference between the battery and cooling medium decreased significantly, reaching a reduction of up to 37 ℃. These findings underscore the efficacy of battery thermal management through the utilization of nano-based PCM composite structures.

To summarize the experimental investigations, Case 2 nano-PCM composite structure has better battery temperature reduction, improved heat transfer within the medium, lower thermal resistance and faster battery cooling. Thermal resistance in this case was found to be the lowest and a reduction of 25% was observed when compared with case 1. Nusselt number enhancement of 20% was also observed in case 2. Temperature difference between battery and cooling medium obtained for case 2 was maximum 5 ℃ and cooled down the battery below 40 ℃ within 200 s for 4 W heat input.

Author contributions: Conceptualization, VM and AB; methodology, VM; software, VM; validation, VM, AB and RB; investigation, VM; resources, VM; data curation, VM; writing—original draft preparation, VM; writing—review and editing, AB and RB; visualization, VM; supervision, AB and RB; project administration, AB and RB. All authors have read and agreed to the published version of the manuscript.

Conflict of interest: The authors declare no conflict of interest.

Nomenclature

A_b	Surface area through which heat is transferred

BTMS	Battery thermal management systems
CPCM	Composites phase change material
DCM-PCMP	Double copper mesh-phase change material plate
K_f	Thermal conductivity of the fluid
LIB	Lithium-ion battery
Nu	Nusselt number
PCM	Phase change material
Q	Heat input
R_{cm}	Overall thermal resistance
T_b	Bulk temperature of the fluid
T_c	Thermal control point
T_m	Maximum melting temperature of PCM
TMS	Thermal management system
T_w	Wall temperature of the surface
w	Width of the surface through which heat is transferred

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