

Experimental investigation of thermal characteristics of lithium ion battery using phase change materials combined with metallic foams and fins

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Abstract. Phase change materials are of great interest in energy storage and energy management applications due to their high latent heat and excellent cycling stability. In this paper, the thermal characteristics of phase change materials (PCM) for thermal management of cylindrical 18650 lithium-ion battery (LIB) were experimentally investigated. A commercial paraffin wax with a melting temperature range between 47 – 53.8°C was used in this study. A metal cylinder with a heater was used to emulate the heat generation from a battery, which was surrounded with the paraffin PCM and contained in a metal housing. The experiment was conducted in an environmental test chamber with controlled ambient temperatures and power inputs. Both the battery temperature and the housing wall temperature were measured during steady-state heating and cyclic heating conditions. Since PCM has low thermal conductivity, thermal enhancement techniques were investigated by adding metal foams (MFs) or combining metallic foam and fins into the PCM to enhance the thermal conductivity. The battery temperatures were measured for all the cases and the results were analyzed and discussed.

1. Introduction

Lithium-ion batteries generate a lot of heat during discharge, which can raise their temperature and affect their operational performance [1]. High temperatures could cause thermal runaway, which is a catastrophic self-accelerated degradation of the Li-ion battery as described in [2], leading to spontaneous combustion or even explosion. Due to the high latent heat and chemical stability, paraffin wax based phase change material (PCM) can be used to maintain the battery at the appropriate temperature level [3-5]. When the heating temperature reaches the melting point of the PCM during the discharge stage of high C-rates (the discharge current to the nominal current ratio [6]), PCM can absorb the heat and maintain temperature of battery at a constant level, reducing the temperature rise of the battery itself as well as temperature nonuniformity among the cells. When the battery is at idle state or operating at low work load, the absorbed heat by the PCM can be released during the solidification process, which helps to maintain the battery temperature at a smaller range of temperature swing. Among the available PCMs, paraffin has been found with the advantages of high

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latent heat, small volume change during melting, good chemical stability, low cost and least subcooling phenomenon, which is the most suitable PCM candidate for the thermal management of lithium ion batteries.

In the past, the phase change material used in electric vehicles (EV) battery thermal management has been studied [7]. However, the shortcoming of low thermal conductivity inherent in paraffin affects its heat transfer performance. The improvement of the thermal conductivity for paraffin can be categorized into different types such as adding fins [8-9], graphite[10-11], metallic foams[12] and thermally conductive fillers[13], etc. Metallic foam with a large number of randomly distributed but similar-sized pores form a high thermal conductivity matrix and conduct the heat to the PCM filled in the matrix effectively. In addition, the porosity of open-cell foam metal reach more than 95%, which does not reduce the PCM amount and thus the heat storage capacity significantly [14]. Zhou et al.[15]experimentally investigated the heat transfer performance of paraffin wax with open-cell metallic foams and compared with the case of embedded expanded graphite. The results indicated that both foam materials improved the heat transfer performance of PCM, and metallic foams can provide better heat transfer performance than expanded graphite due to the continuous inter-connected structures. Duan et al. [16] implemented PCM surrounding the cylindrical heater which was used to simulate the heat source of power battery at low heating power. The experimental results showed that the addition of phase change material, in comparison with natural convection cooling, can significantly reduce the temperature of the heat source. Khateeb et. al[17]applied paraffin wax with embedded aluminum foam to the battery pack in a scooter and measured the temperature rise of battery under low discharge C-rates (1/5-1C). In comparison with the case of PCM only, the addition of aluminum foam further reduced the battery temperature rise. However, the aluminum foam in [17] had a low porosity (porosity80%) which may affect the heat storage capacity of the PCM. As will be shown in this paper, the highly porous copper foam (97.5%) will be used in combination with fin structure, which would be appropriate for high discharge rate applications.

In this paper, the thermal characteristics of the battery cell with PCM and thermal enhancements were examined experimentally at a high power dissipation level. The mock-up battery was made of aluminium cylinder (18mm in diameter, 65mm in length) with inserted heater to simulate the heat generation of 18650-size lithium-ion battery. The cases with pure PCM, PCM combined with metallic foam, and PCM combined with fins and metallic foams were designed and tested. The heating power was set to 6.6W, 8.8W, 13.2W, respectively, corresponding to high discharge rates 8-11C [18], which is to represent the high power dissipation from power batteries during conditions such as uphill and acceleration. The cycling operation conditions were also tested to examine the responses of the battery. Finally, the heat transfer characteristic of the battery by combining the metallic foams and fins in the PCM was also investigated.

2. Experimental setup

Three PCM configurations were designed to surround the battery in the metal housing to examine the thermal characteristics of the battery. The schematic diagram for the three configurations is given in Figure 1. The housing was made of aluminium with the inside diameter of 31mm and outside diameter of 41mm. In the first configuration, pure paraffin wax approximately 25 grams was put into the space between the battery and the housing, as shown in Figure 1.(a), Figure 1. (b) shows the second configuration, the copper foams with pore density of 20 PPI and porosity of 97.5% was first put into the housing contained with the battery, and then the paraffin of the same mass of 25 grams was filled into the foam. In the third design, four aluminium fins with the size of $65 \times 6.8 \times 1\text{mm}^3$ were first mounted into the pre-fabricated grooves on the surface of the mock-up battery, and then filled with the melted PCM and the metallic foam as shown in Figure 1.(c). Care must be taken to minimize voids in filling the PCM into the metallic foam.

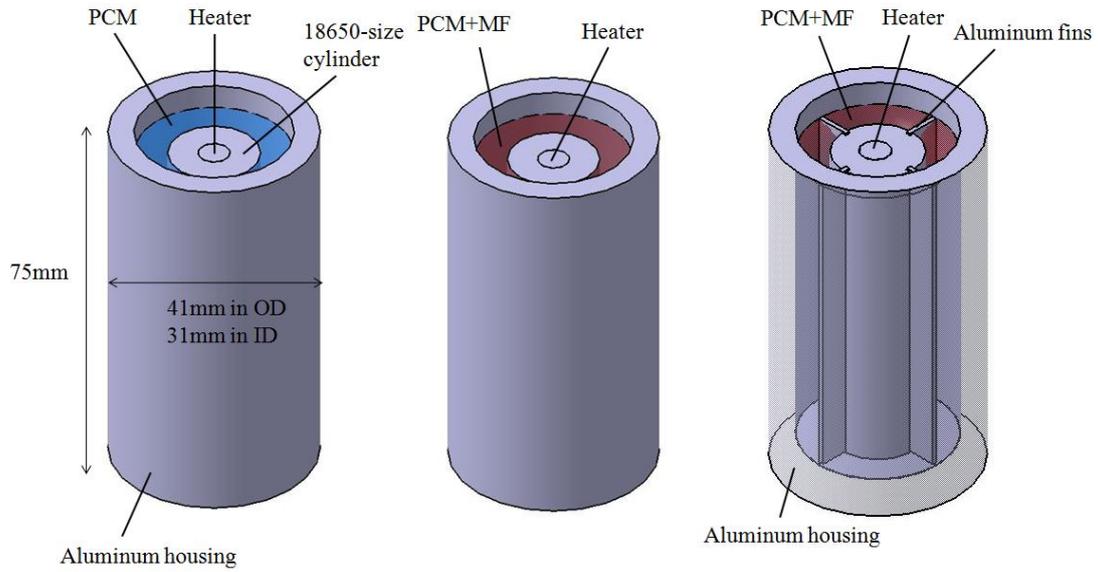


Figure 1. (a) Pure PCM

Figure 1. (b) PCM-MF

Figure 1. (c) PCM-MF-Fins

Figure 1. Three PCM configurations for battery thermal management in the present experiment

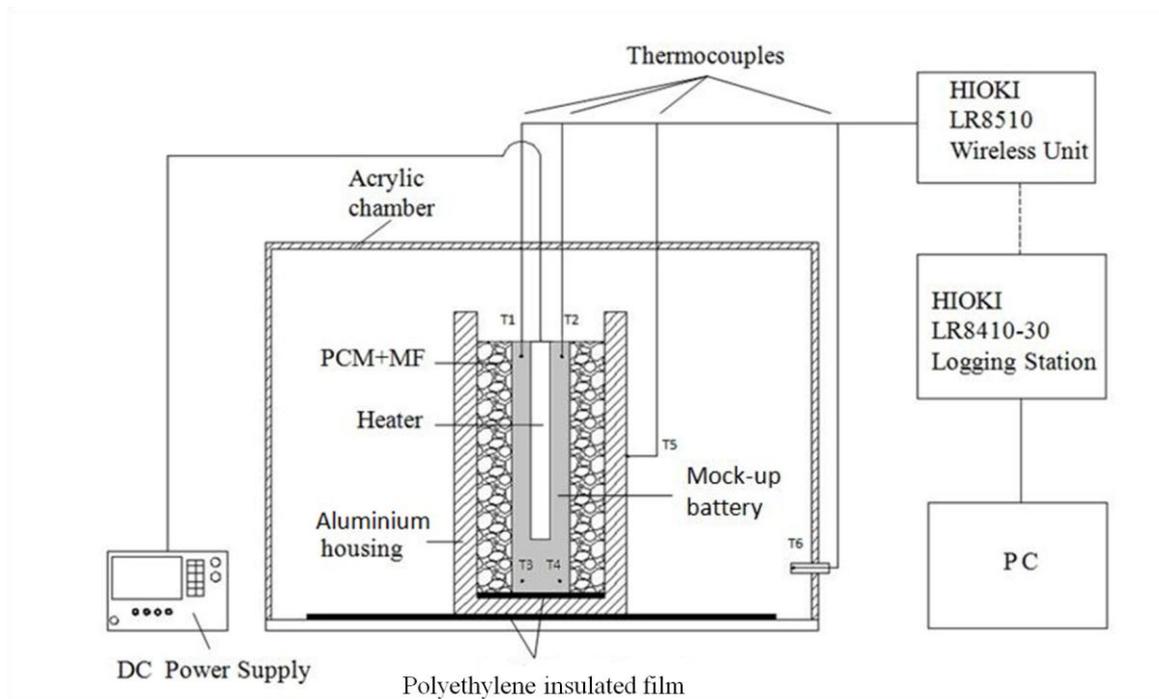


Figure 2. Schematic diagram of the experimental system

All the experiments were tested in an acrylic chamber with the outer dimension of 410mm×410mm×410mm and wall thickness of 5mm to avoid the adverse effects of ambient temperature fluctuations. The whole testing system was placed in the environmental test chamber and the chamber temperature was set to 25°C. The experimental setup is schematically showed in Fig.2. In this experiment, the phase change material was supplied as paraffin sections from Shanghai Yi Yang

Instrument Co.Ltd. Differential Scanning Calorimeter (DSC) was used to obtain the melting and solidification temperatures and heat release of PCM during phase transition. The onset melting temperature was taken as the intersection of the heat flow line w/o phase change and the tangent line drawn at the turning point near the peak temperature. According to the above method, the pure paraffin wax started melting at the intersecting temperature of 47°C, and completed at the peak temperature 53.8°C. The calculated latent heat during melting is 204.5 J/g based on the DSC test. The porosity of copper foam can be calculated as follows [14].

$$\varepsilon = \left(1 - \frac{M}{\rho_1 V}\right) \times 100 \quad (1)$$

Where, M is the mass of the composite sample; ρ_1 is the density of copper $\rho_1=8.92\text{g/cm}^3$, V is the volume of the composite sample.

The porosity was experimentally found to be 97.5%, and the pore density is 20 PPI (pore per inch) which was given by the supplier. The conductivity of paraffin wax with embedded copper foams was measured using LFA457MicroFlash system, which has the measurement range 0.1-2000W/mK, accuracy $\pm 5\%$, repeatability $\pm 3\%$. Due to the limitation of instrument sample holder, composite material sample was prepared in the size of $6.1 \times 14.07 \times 13.9\text{mm}^3$. The specific preparation procedure is described as follows. The copper foam was cut into specified size, and then put into a polyethylene (PE) bag. The melted paraffin was poured directly into copper foam in PE bag and compressed to the desired shape. The whole sample was cut into specific size after it was solidified, and the surface was polished with fine sandpaper to improve the accuracy in the measurement of the thermal diffusion. Thus, the thermal diffusion rate was measured by the flash method. The value should be transferred into thermal conductivity by the following formula [19]:

$$k = \alpha \rho C_p \quad (2)$$

Where, α is the thermal diffusivity which was measured by flash method $\alpha = 1.2 \text{mm}^2/\text{s}$, ρ is the density of sample, C_p is the specific heat of composite sample which can be calculated as

$$C_p = \frac{\varepsilon V C_{p2} \rho_2 + (1-\varepsilon) V C_{p1} \rho_1}{M} \quad (3)$$

where, ε is the porosity of copper foam $\varepsilon=97.5\%$, V is the volume of composite sample, C_{p2} is the specific heat of paraffin wax $C_{p2}=2.1\text{J/gK}$, ρ_2 is the density of paraffin $\rho_2=0.85\text{g/cm}^3$, C_{p1} is the specific heat of copper, M is the mass of composite sample.

Based on above equations, the conductivity of the composite PCM can be obtained as $k=2.2\text{W/mK}$. The relative deviation was found to be -2.3% compared with the two-dimensional model of the periodic hexagonal structure of metallic foam matrix given in Calmidi's work [20]. The aluminium cylinder with the diameter of 18mm, and the length of 65mm inserted with heater inserted was fabricated to simulate the lithium-ion battery heating for heat transfer study. A Watlow cartridge 1/4'' heater was used to connect to a DC power supply. The aluminium foil was wrapped tightly to the heater to strengthen the heat transfer between heater and aluminium cylinder. HIOKI LR8410-30 thermometer was used to measure the temperature. Four thermocouples (T1, T2, T3, T4) were inserted into the corresponding holes on the top and bottom surface of the battery. In order to improve the temperature measuring accuracy, aluminium foils were used to fix the four K type thermocouples, securing the thermocouples in good contact on the bottom of the holes. Thermocouple T5 was fixed by Kapton tape on the surface of aluminium housing in the middle height. The ambient temperature inside the acrylic chamber was measured by thermocouple T6 which was held with a sleeve tube connected to the measurement unit and filled with an insulating material to prevent from outside influences. The relative error of temperature measurement is estimated to be 2%. The error of the heating amount can be calculated based on error propagation formula $\Delta P/P = \Delta I/I + \Delta U/U$, which is within 2%. Overall the error is estimated around 4%. After completion of experimental apparatus connection, the temperature inside the test chamber in Fig.2 was set to 25°C till steady-state was

achieved. In the experiments, the heating power was set to 6.6W, 8.8W, 13.2W, and the heating time for each corresponding powers was controlled so that the housing temperature reached the peak temperature of 53.8°C. The temperature evolution for the mock-up battery was obtained by averaging four thermocouples, whereas the housing temperature was recorded at each heating power, and the temperature profiles were plotted and analyzed.

3. Results and discussion

3.1. Temperature reduction of mock-up battery using metallic foam-PCM composite materials

In this paper, heating powers of 6.6W, 8.8W, 13.2W were implemented to examine the limit state of the battery cooling capacity for the three PCM cases illustrated in Figure 1. Figure 3 shows the comparison of temperatures for the battery and housing for the case of pure paraffin and the composite materials with paraffin embedded copper foam at the heating power of 6.6W. The temperature rise of the mock-up battery can be divided into three stages. The first stage corresponds to the sensible heating stage 0 ~ 380s, and the battery temperature (T_b) reached 47°C at 380s. At this stage, the paraffin remained unmelted and the PCM heat transfer was through conduction only, thus the profile of temperature rise was steep. The second stage was the melting process which started at 380s and ended at 2700s. At this stage, the battery surface temperature went up beyond 47°C which corresponded to the onset of melting as obtained by the DSC test. Paraffin near to the battery surface started to melt, absorbed the heat and slowed down the temperature rise of battery. When the temperature of housing (T_w) reached 53.8°C at around 2700s, the melting was considered completed. The temperature plateau was observed obviously and the battery temperature kept almost unchanged. This could be explained by the effect of natural convection induced inside the melting PCM, which maximized the heat transfer rate from battery to the PCM melting interface. The third stage started at 2700s, the temperature of the aluminium housing reached 53.8°C, the PCM was considered fully melted, and the battery temperature started to rise rapidly again after the completion of PCM melting.

In the next experiment, metallic foam was added to increase the effective thermal conductivity of paraffin. Heat generated from battery could be conducted to the paraffin by copper foam and battery temperature and internal temperature gradient within paraffin were reduced. But on the other hand, higher flow resistance posed by metallic foams suppresses natural convection of PCM [21]. Thus, there is no clear-cut temperature plateau as shown in Fig. 3. The temperature rise of battery gently increased during the melting process. However, the improvement of heat conduction overwhelmed the adverse effect on natural convection in paraffin. The distinction of each stage was not as obvious as the case of pure paraffin, the temperature difference between battery and the housing was narrowed, and the battery temperature was lower than the case of paraffin without copper foam. When the temperature of housing reached 53.8°C, the battery temperature was measured 65.9°C which dropped by 3.4°C compared to pure paraffin as shown in Fig.3 at 6.6W heat input. The similar battery temperature decreased with the addition of copper foam can be found in Fig. 4, Fig. 5 at increased heating power of 8.8W, 13.2W, respectively. On one hand, the battery temperature and the temperature difference between battery and housing further increased with the increasing heating power. On the other hand, the composite of paraffin combined with copper foam showed better performance than the pure PCM case. The larger the heat input was, the larger the battery temperature drop was between the composite PCM and pure PCM cases. The battery temperatures upon the completion of the melting are listed in Table 1 for the different heating powers.

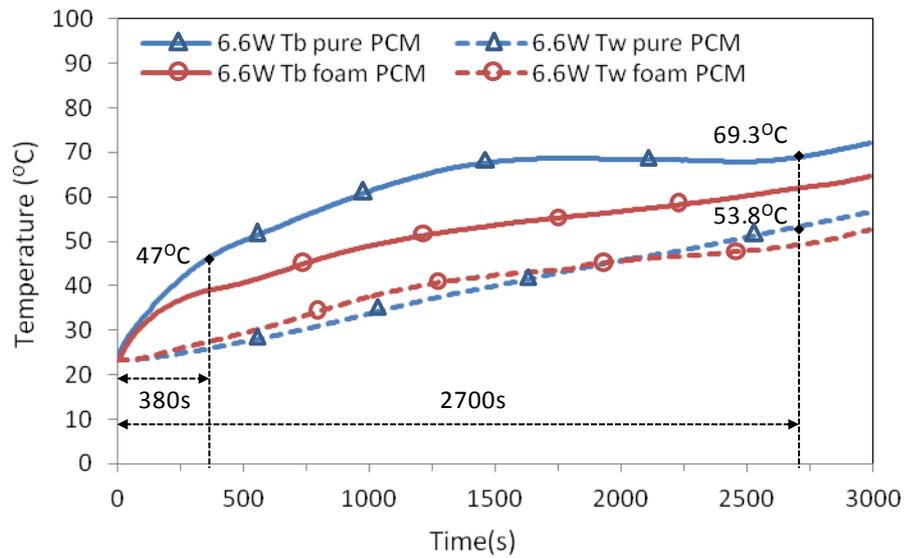


Figure 3. Temperature profiles for battery and housing wall for pure PCM and PCM-MF cases at heating power of 6.6W

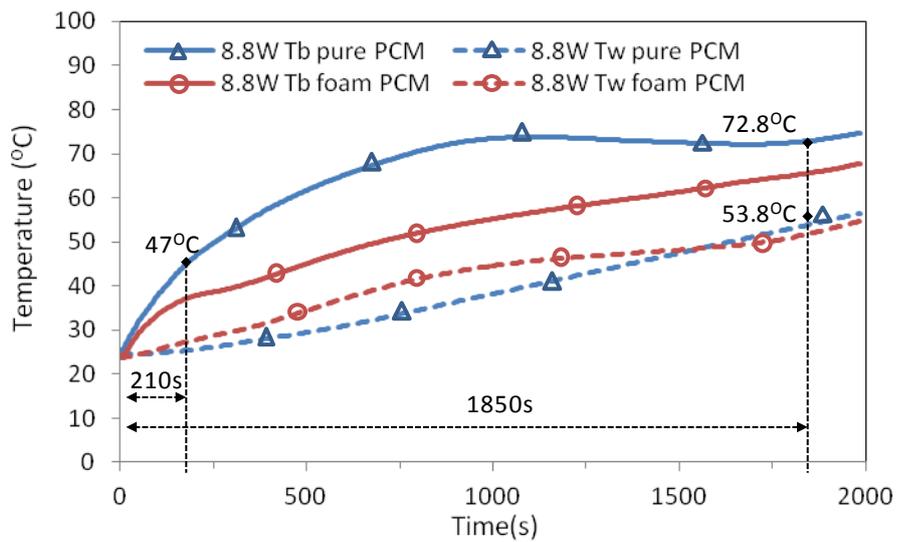


Figure 4. Temperature profiles for battery and housing wall for the pure PCM and PCM-MF cases at heating power of 8.8W

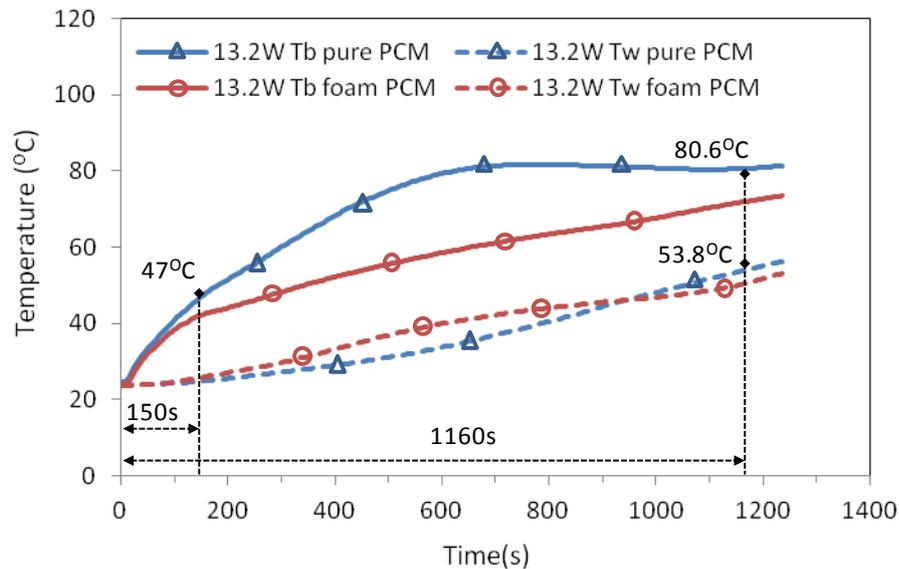


Figure 5. Temperature profiles for battery and housing wall for pure PCM and PCM-MF cases at heating power of 13.2W.

Table 1. Comparison of battery temperature between the three investigated cases at completion of melting at the three investigated heating powers

P (W)	Pure PCM		PCM - MF		PCM-MF-Fins	
	Tb (°C)	Tw (°C)	Tb (°C)	Tw (°C)	Tb (°C)	Tw (°C)
6.6	69.3	53.8	65.9	53.8	60.8	53.8
8.8	72.8	53.8	68.7	53.8	62.2	53.8
13.2	80.6	53.8	74	53.8	65.1	53.8

3.2. The temperature evolution for battery with foam-paraffin at heating and cooling cycles

The above study compared the experiment case of pure paraffin and paraffin combined with copper foam under steady-state heating conditions. To further examine the effectiveness of the configuration of paraffin with copper foam, the cyclic heating test was conducted by switching on and off the power supply to simulate the dynamic electric vehicle operation. The ambient temperature was set to 25°C, and the heating power was set to 4.4W. Both cases were heated in 3000s and cooled in 3000s under natural convection, and then the same cycles were repeated. The temperature curves of battery for cycles were plotted in Fig.6. The battery temperature with pure paraffin reached 60°C at the end of second heating, beyond the allowable working temperature for lithium ion battery which was within the range of 0-55°C. In contrast, the battery temperature was maintained below 53°C for the case of paraffin combined with copper foam during heating period. The temperature rise was obviously lower than the case of pure paraffin, indicated that battery surrounded with the copper foam-paraffin composite materials was more effectively maintained within the proper operating temperature at the cyclic heating condition.

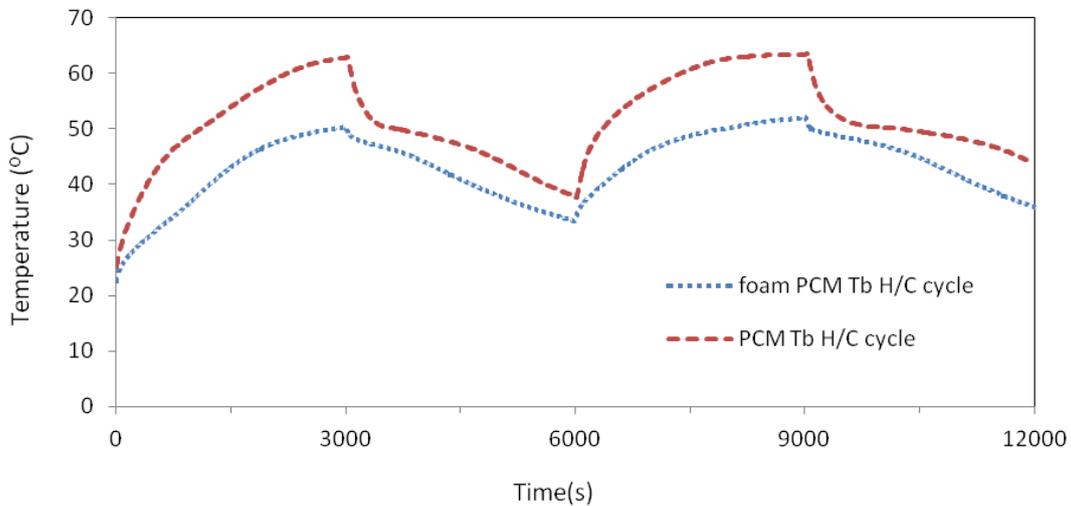


Figure 6. Comparison of effect on battery temperature for pure paraffin and foam-paraffin in heating and cooling cycles

3.3. Effect of fin inserts into foam-paraffin composite materials

In this section, the combination of fins and copper foam in the PCM was examined as the thermal enhancement technique. Four aluminium fins of 1mm thick were symmetrically inserted into foam-paraffin composite materials as indicated in Figure. 1(c). The experimental condition was controlled in the same way as mentioned above. The temperature rise of battery and housing are shown in Figures.7-9 for the different heating power. The shape of curves of battery and housing temperature evolution for the foam-paraffin-fins was similar to the case in foam-paraffin, but both the absolute temperature and the battery to housing temperature difference in the former case were lower than the latter case. Adding fins, on the one hand, improved the effective conductivity in paraffin and, on the other hand, reduced the thermal contact resistance by extending the contact area between battery and the copper foam, and enhanced the heat transfer performance of metallic foam [22]. At the heating power of 6.6W, battery temperature reached 60.8°C when the housing temperature reached 53.8°C, dropped by 5.1°C compared to the case of foam-paraffin, 8.5°C compared to the pure paraffin. The temperature drop was more significant for the higher heating power. The battery temperature of three experimental cases at the fully melted time at different heating power of 6.6W, 8.8W, 13.2W were listed in Table 1.

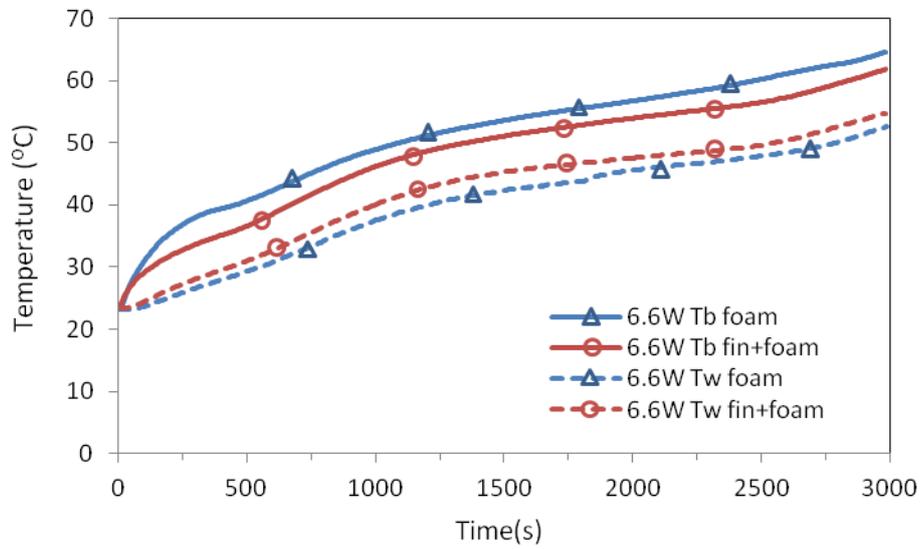


Figure 7. Temperature profiles on battery and housing for the cases of PCM-MF and PCM-MF-fins at the heating power of 6.6W

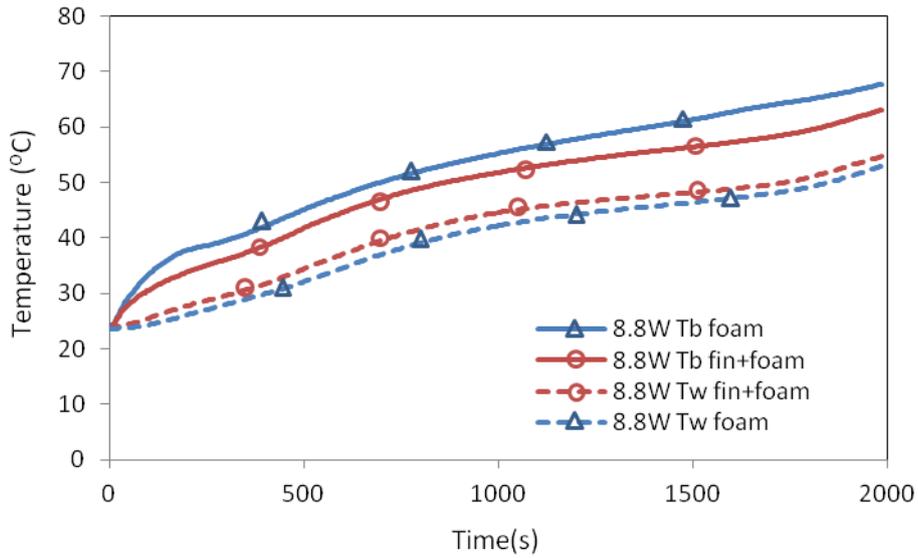


Figure 8. Temperature profiles on battery and housing for the cases of PCM-MF and PCM-MF-fins at the heating power of 8.8W

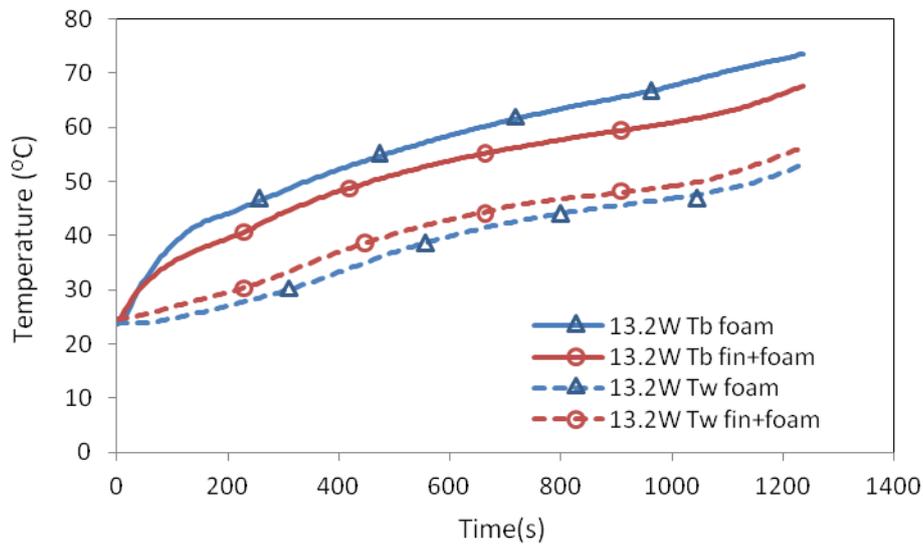


Figure 9. Temperature profiles on battery and housing for the cases of PCM-MF and PCM-MF-fins at the heating power of 13.2W

The temperature difference between battery and the aluminium housing at different time steps for different heating powers were listed in Table 2 to compare the thermal performance of the three PCM cases. Better thermal performances were achieved in both the foam-paraffin and foam-paraffin-fin cases than that the pure paraffin case. Among them, the case of foam-paraffin-fin exhibited the lowest battery to housing temperature difference with time evolution. Similar trend was found for the cases of heating power 8.8W and 13.2W. Nonetheless, adding fins can slightly affect the capacity of heat storage of the composite materials since the volume of four fins in the present study is more significant than the metallic foam, which is the unfavourable factor. The increase in volume by adding four fins of 1mm is 1.56cm^3 , whereas the actual volume of the copper foam is 0.735cm^3 . In contrast, adding fins could reduce volume fraction of PCM by 5.3%, which may adversely affect the capacity of the heat storage.

Table 2. Comparison of the battery to housing temperature differences for the three cases, PCM, MF-PCM and MF-PCM-fin at the different heating power.

P(W)	Time(s)	Tb – Tw [°C]		
		PCM	MF-PCM	MF-PCM-fins
6.6	600	23.8	11.5	5.8
	1100	28.0	11.2	6
	1600	27.3	11.5	5.9
	2500	17	12.3	6.9
8.8	600	34	13.1	7.5
	1100	33.9	13.2	7.3
	1600	23.2	15.5	8.3
	1900	18.6	15.4	8.3
13.2	600	45.5	18.6	10.9
	900	37.3	19.8	11.4
	1100	29	21.7	11.7

4. Conclusions

In this paper, an experimental study of the thermal characteristics of mock-up LIB was conducted with PCM and the thermal enhancement techniques. The conclusions can be drawn as follows.

- In the PCM case, the battery temperature evolution can be divided into three phases depending on the melting condition of the pure PCM: temperature rise with unmelted PCM, temperature plateau with partially melted PCM, and temperature rise with fully melted PCM.
- The addition of metallic foam can enhance the heat conduction and reduce the battery temperature obviously. The battery temperature at the time PCM fully melted is 3.4-6.6°C lower than that with pure PCM for heating power 6.6W-13.2W.
- Under cycling condition, the temperature swing with metallic foam and PCM can be reduced by $\sim 10^\circ\text{C}$ in comparison with the case with the PCM only.
- The addition of four fins can further reduce the T_b by 4.1-8.9°C compared with the case with metallic foam and PCM for heating power 6.6W-13.2W.
- The addition of fins can further reduce the battery temperature, which also reduce the PCM volume and may have an adverse effect on capacity of heat storage.

Acknowledgements

We acknowledge the financial supports from the Eastern Scholar program by Shanghai Higher Education Institution and the project 14520501100 granted by Shanghai Scientific and Technological Commission.

A. 1

Nomenclature

- C_p specific heat capacity of sample (J/gK)
 C_{p1} specific heat capacity of copper (J/gK)
 C_{p2} specific heat capacity of paraffin (J/gK)
 I current (A)
 k thermal conductivity (W/mK)
 M mass of sample (g)
 P heating power (W)
 T_b battery temperature ($^\circ\text{C}$)
 T_w housing wall temperature ($^\circ\text{C}$)
 U voltage (V)
 V volume of sample (cm^3)
 α thermal diffusivity (cm^2/s)
 ε porosity
 ρ_1 density of copper (g/cm^3)
 ρ_2 density of paraffin (g/cm^3)

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