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Article

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Research paper

Cold temperature performance of phase change material based battery thermal management systems

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ABSTRACT

This study evaluates performance of passive thermal management system of high power Li-ion batteries in a cold environment. Two different phase change materials (PCM) consisting of pure paraffin and paraffin-graphite composites are considered and compared with a battery module in absence of PCM material. Battery modules are partially discharged for a period and then subjected to different cold time periods representing short and long vehicle stops during winter. Battery performance parameters such as capacity, power, and temperature along with thermophysical properties of each module are recorded, calculated, and compared for different scenarios. Thermal management systems moderate temperature rise during discharge and slow the temperature loss during cold stops but also slow the temperature rise after cold stops. Results show that after a short (10 min) cold stop the paraffin wax offers no advantage by keeping the module warm. Both paraffin and no PCM modules had comparable energy values. On the other hand, during long (2 hours) cold stops a significant detriment (15% reduced energy vs 6% reduced energy) was observed in PCM modules by delaying battery warm up when compared to modules with no PCM. Commercial PCM modules with graphite/paraffin composites performed in-between PCM and no PCM modules (9% reduced energy) after long cold soaks but had no advantage for short cold soaks (2.5% energy reduction) due to higher heat dissipation.

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1. Introduction

The depletion of non-renewable energy sources and the environmental pollution accompanying fossil fuel use has become a critical issue across the globe. Using electric vehicles (EVs) instead of traditional vehicles can reduce greenhouse gas emissions up to 40% when the electric power is produced by renewable energy (Rao and Wang, 2011).

Improving the performance and cycle life of Li-ion batteries is a key factor for EV viability. Li-ion batteries need to operate in a temperature range of 20 °C to 40 °C in order to prevent thermal runaway and poor life cycling at high temperatures as well as avoid high resistance at low temperatures. Moreover, uneven distribution of temperature in a battery pack reduces its performance and life (Pesaran, 2002). Given that batteries generate a significant amount of heat at high charge and discharge rates, thermal management systems (TMS) in battery packs in EVs are critical to avoid diverse temperature impacts. Conventional TMS rely on air or liquid, but are too expensive, heavy, and complex (Pesaran, 2001; Hallaj and Selman, 2000).

Another option is to use phase change materials (PCM) that absorb heat through their latent heat during melting. A desirable PCM for a battery thermal management system possesses a melting point coincident with the operating temperature of battery operation, large latent heat of fusion per unit mass, high specific heat, high thermal conductivity, high density, chemical stability, and low cost (Agyenim et al., 2010; Alrashdan et al., 2010). The large latent heat per unit mass requires less PCM to be used, high thermal conductivity allows fast dissipation of heat, and high specific heat allows for greater thermal storage outside of the phase change temperature region. A proper figure of merit can be introduced as the result of multiplying thermal conductivity, specific heat, latent heat and dividing by volume change and density.

TMS using PCM were first developed by Hallaj and Selman and have many advantages over conventional TMS (Sabbah et al., 2008; Hallaj and Selman, 2002). Several studies have examined the suitability of PCM for battery TMS in hybrid and plug-in hybrid electric vehicle systems in stressful conditions such as high power draw at high temperature (Kizilel et al., 2008, 2009; Kim et al., 2008). Other research in this area focused on the material properties such as thermal conductivity to improve performance efficiency of PCM (Alrashdan et al., 2010; Goli et al., 2014).

Nearly all research has been directed towards the study of PCM at high temperatures while their benefits and weaknesses at low

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temperatures have not been as thoroughly investigated. Song et al. (2012) emphasized that battery thermal management is necessary not only at high temperatures but it also significantly affects the battery performance and life in low ambient temperatures. There have been only a few studies in battery thermal management at low temperatures (Song et al., 2012; Shidore and Bohn, 2008; Pesaran et al., 2003). None of these studies have addressed the fundamental question of whether or not PCM materials could be a net detriment to EV battery performance by delaying battery warming after a long cold soak and affecting capacity, temperature and power of battery.

In this study we undertake to answer these questions by assessing the performance of batteries in cold ambient temperature with and without PCM based TMS. Advantages, such as the PCM keeping the battery package warm during short stops, will be compared with drawbacks such as delaying battery warming after a long cold soak.

2. Experimental methods

A Ford Escape Hybrid car can consist of 68 battery modules. Each module contains 20 commercially available 1.5 Ah Type 18650 high power Li-ion cells (Sabbah et al., 2008). A scaled down experiment was employed to induce the required temperature and battery load to the battery module. In this test the battery module consisted of seven high power 2.2 Ah Li-ion battery cells (Interstate Batteries, model 18650, Dallas, TX) assembled in series configuration and connected to a DC power supply (Model 224 Programmable current source, Keithley Instruments Inc, Cleveland, OH), an electronic load (6060B 300 W electronic load, Hewlett Packard, Palo Alto, CA) and a data acquisition system (Model 2000 Digital Multimeter, Keithley Instruments Inc, Cleveland, OH). LabVIEW (National Instruments Corporation, Austin, TX) was used as a communication interface between the battery hardware components and the software used to record temperature, voltage, and power at two second intervals. Battery cell specifications are listed in Table 1.

The individual battery cells were arranged in a hexagonal packing for battery testing (see Fig. 1). To avoid the influence of slight temperature fluctuations between testing on different days the modules were fitted into a waterproof container and placed in a large water bath set to 23 °C. Temperature within the battery modules was monitored via K type thermocouple probes located at the center of the battery module (see Fig. 1). The two Keithley multimeters were used with an error percent of 6% in acquiring temperature. Data acquisition system collects capacity, and power with 5% error. The battery module was charged at 0.1C rate and then discharged at C/1 rate. The C/1 discharge rate represents the normal maximum continuous discharge rate of the 18650 Li-ion cells recommended by the manufacturers in order to prevent overheating.

The battery module was tested with three thermal management conditions; (i) no TMS, (ii) a simple paraffin wax PCM based TMS, and (iii) a commercial paraffin wax and expanded graphite composite PCM based TMS (AllCell Technologies, Chicago, IL). The commercial PCM as discussed by Mills and Alrashdan is made by compacting expanded graphite followed by impregnating it with PCM material such as paraffin wax. This is done in order to more rapidly disseminate heat via the graphite component of the composite. Commercial PCMs have a thermal conductivity of about 30 times of that of the paraffin alone (Alrashdan et al., 2010).

For all three TMS conditions, the battery module was discharged galvanostatically for half an hour at 23 °C before being placed in a cold environment at a temperature of −17 °C to −18 °C for a time period between 10 min and 240 min to represent vehicle stops during winter conditions. Batteries were not discharged during

Table 1

Battery module specification.

Operating voltage	3–4.2V
Cell type	18650
Nominal voltage	3.7V
Operating temperature range	Charge 0 °C to 45 °C Discharge −10 °C to 60 °C
Weight	45g

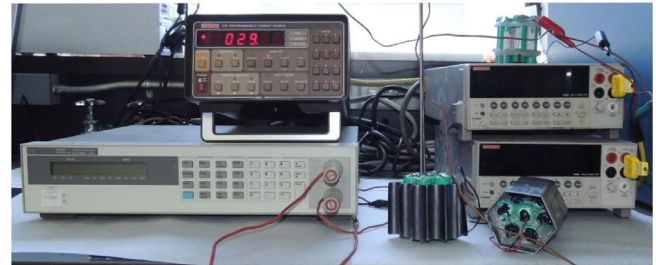


Fig. 1. Illustration of battery test setup.

these cold wait periods and the time durations were selected based on statistical average short stops provided by Shahidinejad et al. (2012). After the wait period had elapsed, the battery module was returned to the water bath at 23 °C and was immediately discharged to 21 V. We justify discharging at 23 °C because even in cold winter climates electric vehicles are equipped with an active thermal management system to keep cabin temperature and ambient temperature around battery modules near room temperature (Pesaran, 2001). The mass of PCM required for passive thermal management of this battery module was calculated using a lumped system analysis where heat discharged by the battery is set equal to the sensible heat of the PCM plus latent heat of the PCM as described in Eq. (1).

$$Q_{\text{discharge}} = m_{\text{PCM}} C_p (T_m - T_i) + m_{\text{PCM}} \lambda \quad (1)$$

where, m_{PCM} , C_p , and λ represent mass, specific heat, and latent heat of the PCM, respectively. T_m and T_i are PCM melting point and initial temperature and $Q_{\text{discharge}}$ is the Li-ion battery discharge heat. For commercial PCM ratio of wax volume to pack volume was 80%.

Thermophysical properties such as melting point, latent heat and specific heat was measured using differential scanning calorimetry, (DSC, Netzsch Sirius 3500). Measurements were conducted at a scan rate of 1 K/min at temperature range of 20 °C to 100 °C in a nitrogen atmosphere. Specific heat measurement method based on ASTM E1269 – 11 was selected to analyze the data (ASTM, 2005).

Table 2 summarizes the thermophysical properties of the paraffin wax and graphite composite PCM used in this study. Laser flash analysis (LFA, 457 Netzsch) was used to measure the thermal diffusivity of paraffin wax.

3. Results and discussion

3.1. Battery performance after short and long cold soak

In this section the advantages, such as the PCM keeping the battery package warm during short stops, and drawbacks, such as delaying battery warming after a long cold soak, will be quantitatively discussed. From time $t = 0$ s up until $t = 1800$ s the samples are all initially discharged in the water bath and then placed into the cold environment for a specified time. From time $t = 1800$ s up until $t = 3600$ s the samples are removed from the cold environment and allowed to continue discharging in the water

Table 2
Thermophysical properties of paraffin wax and paraffin/graphite composite PCM.

PCM type	ρ (g/cm ³)	C_p (J/gK)	α (mm ² /s)	κ (W/mK)	T_M (°C)
Paraffin wax	0.88	1.98	0.29	0.50	46–58
Commercial PCM	0.78	2.31	10.74	16.6	46–58

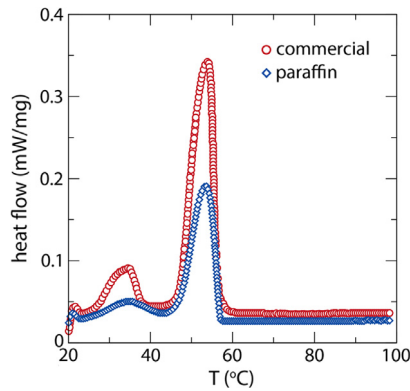


Fig. 2. Comparison of heat capacity values for paraffin wax and commercial composite PCM materials.

bath. Battery performance metrics were calculated as a function of cold soak time and module type. Power drop was calculated by evaluating power ($P = IV$) at $t = 2000$ s and assessing the percent change between modules that were discharged with no cold soak as opposed to those that experienced a cold soak. Warming rate was calculated as the slope of temperature–time curve after cold vehicle stop (Fig. 3), assuming a constant warming rate, by calculating the slope of line connecting initial and final point.

Fig. 3a shows the battery cell temperature inside the module versus time during a one hour discharge (C/1) with and without a paraffin wax PCM and with commercial PCM. As expected, when the battery modules are discharged continuously in the water bath with no cold soak the PCM helps keep the battery module temperature lower. The commercial PCM keeps the temperature lower than the paraffin PCM, consistent with the additional heat absorbance of commercial PCMs as measured by DSC in Fig. 2. These trends which are observed from time $t = 0$ s up until $t = 1800$ s are consistent for all experiments.

Fig. 3b shows that paraffin is beneficial in keeping the battery warm during short stops. The battery module with paraffin PCM only has 55% of the temperature change compared to that without PCM. Despite having reversibly absorbed more heat, the commercial PCM has also dropped in temperature comparable to the no PCM module and this is likely due to the expanded graphite additives that increase the thermal conductivity of the commercial PCM. The paraffin PCM module warms up more slowly (\approx half the rate) such that at $t = 3600$ s the modules have reached the same temperature. On the other hand, after a long 240 min stop the temperature change for both modules is nearly the same which is expected when thermal equilibrium is established. The influence of temperature (Fig. 3) on battery performance (power drop, temperature drop, warming rate) in these cold soak conditions is detailed in Table 3. Percent reduction in power (evaluated at $t = 2000$ s) after 10 min and 20 min cold soaks was small in all modules with values ranging between 0.1% and 2.3%. Paraffin PCM modules performed slightly better than no PCM and commercial PCM, consistent with their smaller temperature drop. After long cold soaks the power drop (evaluated at $t = 2000$ s) was 10.6% for no PCM module and 11.3% and 11.6% for paraffin and commercial PCM modules, respectively suggesting a very slight advantage for non PCM modules.

These results for temperature as a function of time and warming rates after cold soak both provide supporting evidence for our original contention that PCM based TMS will provide thermal damping that can be both potentially beneficial (keeping module warm) and detrimental (delay warm up time) in cold environments depending on the time duration of the cold soak. However, the percent change in power evaluated at a single temperature does not capture fully the affect of the cold soak. For example, consider the voltage profile for the different modules as they are plotted against time for different cold soak times in Fig. 4. The drop in voltage following the cold soak becomes increasingly severe with cold soak duration. However, as the module is returned to the water bath and battery discharge is resumed the module begins to warm up causing the voltage to rise, but this is counteracted by the diminishing battery capacitance producing the observed voltage–time profiles.

Temperature is an important factor in determining electrochemical behavior of Li ion batteries. At equilibrium condition, temperature can affect Gibbs free energy of formation and consequently open circuit potential (OCV) of the cell by direct correlation. In addition, it can indirectly effect cell potential by changing enthalpy and entropy of reactions as provided by following equations:

$$\Delta \bar{G} = \Delta \bar{H} - T \Delta \bar{S} \quad (2)$$

$$E_{OCV} = -\Delta \bar{G}/nF \quad (3)$$

where $\Delta \bar{G}$ is the Gibbs free energy of formation, $\Delta \bar{H}$ is the change in enthalpy of formation, $\Delta \bar{S}$ is the change in entropy of formation, E_{OCV} is the open circuit voltage, n is the number of electrons in the redox reaction, and F is Faraday's constant.

Under normal operation the condition of the cell and its potential will be influenced by kinetics of reaction, charge and mass transfer as well as ionic and electronic conductivity of cell components such as the electrolyte conductivity, in particular. Thus, operating cell potential will be lower than OCV due to several potential drop factors. This can be shown by following equation:

$$E = E_{OCV} - \eta_c - \eta_a - iR \quad (4)$$

where η_c and η_a are the potential drops due to concentration and activation polarization, i is the operating current of cell on load, and R is the ohmic internal resistance of the cell.

Ji et al. (2013) conducted a systematic study to investigate effect of low temperature on electrochemical performance of high power Li-ion batteries using 2.2 Ah 18650 battery cell. Discharging the cells at 1C, they showed that by decreasing temperature of environment from 25 °C to −20 °C the voltage and capacity of cell decreased 86% and 66%, respectively. Thus, the observation featuring in Figs. 3 and 4 showing correlation of voltage and temperature are in accordance with literature suggesting that Gibbs free energy of formation decreases with decreasing temperature. In addition, by decreasing the temperature under normal operation charge transfer resistance, ohmic resistance, and concentration polarization all contribute to lower potential.

3.2. Energy analysis

In order to better capture the effect that short and long cold temperature soaks have on battery modules it is necessary to consider the instantaneous change in voltage as this relates to

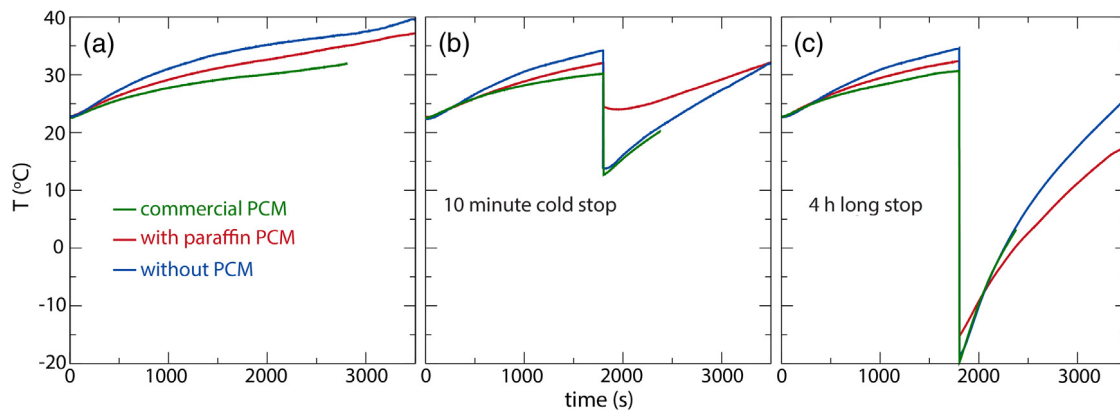


Fig. 3. Temperature variation versus time at the center of the Li-ion battery module for three different modules with (a) one hour discharge in water bath, (b) one hour discharge in water bath interrupted by a 10 min cold soak in the middle of discharge, (c) one hour discharge in water bath interrupted by a 4 h long cold soak in the middle of discharge.

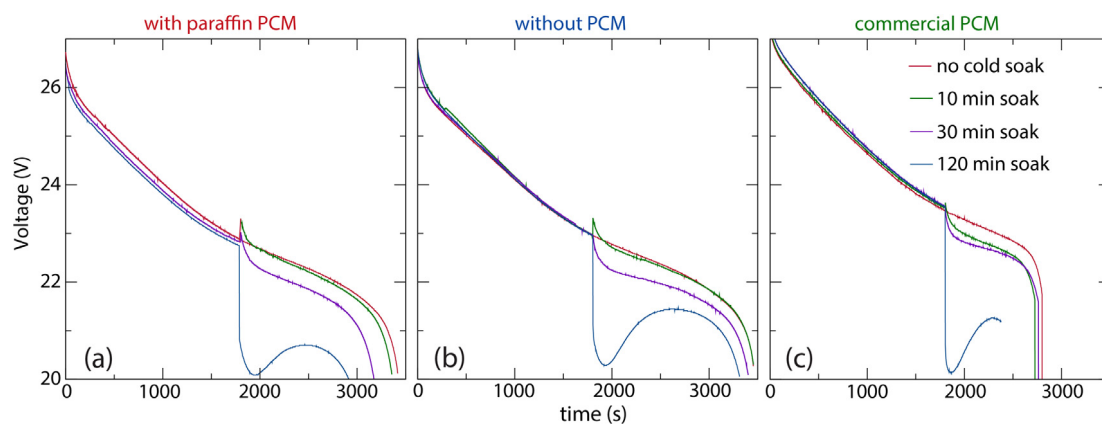


Fig. 4. Voltage variation versus time and cold soak duration for three different Li-ion battery modules featuring (a) paraffin PCM, (b) no PCM, and (c) commercial PCM.

Table 3

Parametric comparison of result for battery module in absence of PCM system and in present of PCM system of paraffin and paraffin/graphite composite.

Module type	Cold soak condition	Temperature drop inside the module (%)	Power drop (%)	Warming rate Kmin ⁻¹
(i) No PCM	No cold soak	0	0	0.29
	10 min	55	−0.3	0.65
	20 min	93	−2.3	0.95
	240 min	151	−10.6	1.91
(ii) Paraffin PCM	No cold soak	0	0	0.25
	10 min	24	−0.1	0.27
	20 min	45	−1.8	0.42
	240 min	147	−11.3	1.36
(iii) Commercial PCM	No cold soak	0	0	0.20
	10 min	57	−1.3	0.72
	20 min	86	−2	0.99
	240 min	163	−11.6	1.95

energy. The amount of chemical energy, E , that the battery has produced can be evaluated by integrating from initial time, $t_{initial}$, to final time, t_{final} , the time dependent voltage multiplied by the current as shown in the following equation

$$E = \int_{t_{initial}}^{t_{final}} VI dt \quad (5)$$

where V is the voltage, and I is the current which was maintained at 1 A during the galvanostatic battery discharge.

The voltage–time profile for all modules changes with cold soak duration. Therefore, the total available energy of the battery module is influenced by cold soak time as well. To accurately determine this energy for each battery module and cold soak time

the voltage–time plot was numerically integrated. An example of one such integration used to determine E is shown in Fig. 5. Once E was calculated for both paraffin PCM and no PCM modules for all cold soak durations it was possible to compare the battery's energy as a function of cold soak duration. Fig. 6 shows the percent change in total energy as a function of cold soak time for both paraffin PCM and no PCM modules.

When we compare the percent change in energy for paraffin PCM modules vs no PCM modules vs commercial modules we find definitive evidence to support the hypothesis of this work. PCM modules may perhaps offer a very slight advantage over no PCM modules (+0.35% vs +0.1%, within statistical error) after a 10 min cold soak by keeping the battery module warm and thereby maintaining a slightly higher voltage. However for 30 min and

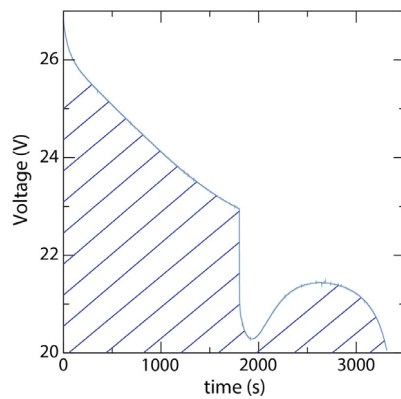


Fig. 5. Example of energy, E , calculation by numerical integration over time, and multiplying by current.

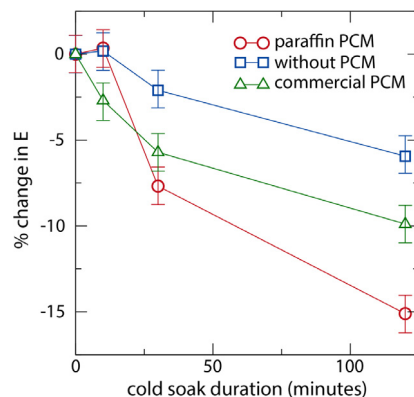


Fig. 6. Percent change in calculated energy, E , as a function of cold soak duration for paraffin PCM vs no PCM battery modules.

120 min cold soaks the delayed warming greatly diminishes battery module energy. After 120 min the PCM modules loses 15% of its energy whereas the no PCM module only loses 6% of its energy. Commercial PCM modules also lose energy after long cold soak (9%) though not as severe as PCM modules. However, we do not see a benefit after short cold soaks with commercial PCM modules. This is likely due to the fact that the commercial PCM modules dissipate heat very effectively and as a result are unsuccessful in keeping battery modules warm.

4. Conclusions

Cold temperature performance of passive thermal management systems for high power batteries of electric vehicles was studied. Results showed that the low thermal conductivity of paraffin wax, normally considered a detriment at high temperatures when thermal management systems seek to prevent overheating, may actually provide a slight advantage during short (10 min) cold

soaks by keeping the module warm and thereby improving energy retention. On the other hand, paraffin wax becomes detrimental after a long cold soak. The warming rate of paraffin PCM compared to commercial PCM or modules with no PCM is 29% slower which causes a significant energy loss (15% compared to 6% for no PCM modules).

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