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Passive control of temperature excursion and uniformity in high-energy Li-ion battery packs at high current and ambient temperature

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ABSTRACT

A strategy for portable high-power applications with a controlled thermal environment has been developed and has demonstrated the advantage of using the novel phase change material (PCM) thermal management systems over conventional active cooling systems. A passive thermal management system using PCM for Li-ion batteries is tested for extreme conditions, such as ambient temperature of 45 °C and discharge rate of 2.08*C*-rate (10 A). Contrary to Li-ion packs without thermal management system, high-energy packs with PCM are discharged safely at high currents and degrading rate of capacity of the Li-ion packs lowered by half. Moreover, the compactness of the packs not only decreases the volume occupied by the packs and its associated complex cooling system, but also decreases the total weight for large power application.

1. Introduction

As is well-known, lithium-ion battery packs have very strong advantages over other types of advanced batteries for high-energy/high-power applications such as electric-powered transportation [1]. However, the need for safe containment and management of appreciable heat effects associated with lithium-ion batteries in high-power applications remains a challenge to be met before wide-spread commercialization can occur [2].

A novel tool which can significantly simplify the solution to this problem is the use of phase change materials (PCM) in cooling Li-ion battery packs. The concept of using PCM, as well as the material details of the technology that makes the application practically possible have been introduced and illustrated in various earlier papers [3] and patents [4,5]. In particular, detailed experimental and modeling results have been presented at various conferences [6,7].

In this work, we present examples of a strategy for using PCM in portable high-power applications with a controlled thermal environment and demonstrate a passive thermal management system using PCM for high-energy Li-ion packs for both normal and stressed operating conditions such as high current drain at high ambient temperature. We confirm the validity of this strategy based on modeling predictions by experimental data. The aim of this comparison is three-fold:

(1) Illustrate the effectiveness of achieving temperature uniformity in a module (or pack) by using PCM.
(2) Demonstrating that cycle-life of a module (or pack) is directly correlated with temperature uniformity as well as low average temperature of the module – thereby, simultaneously, enhancing safety.
(3) Illustrating the flexibility of using PCM by design of the PCM matrix and choice of the PCM materials and composition.

The detail of the work is listed below.

(a) Comparison of temperature increase in 8S2P packs w/wo PCM and temperature uniformity along the packs operating at room temperature and *C*-discharge rate (4.4 A).
(b) Comparison of temperature increase and the capacity degrading in 4S4P (four cells in series and four strings in parallel) packs w/wo PCM at room temperature and *C*-discharge rate (8.8 A).
(c) Electrochemical and thermal response of 7S2P pack without PCM
    (i) at normal conditions, i.e. room temperature and 1.0*C*-discharge rate,

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Table 1
Specifications of the commercial cells used in this study

Operating voltage	3.0–4.2 V
Cell type	Type 18650
Cell capacity	
Type A (HE)	2.2 Ah
Type B (HE)	2.4 Ah
Nominal voltage	3.7 V

Table 2
Specification of battery packs

Pack operating voltage	3.0–4.2 V cell^{-1}
Cell capacity/pack capacity	
4S4P	2.2 Ah/8.8 Ah
8S2P	2.2 Ah/4.4 Ah
7S2P	2.4 Ah/4.8 Ah
Total weight of pack	
4S4P, 8S2P	~2 lbs
7S2P	~2 lbs
Spacing between cells	
4S4P	3 mm
8S2P	3 mm
7S2P	2 mm

(ii) at stressed conditions, i.e. at 45 °C, at 2.08*C*-discharge rate (10 A).

Test results conducted in this work clearly demonstrated the advantage of using the PCM thermal management system over conventional active cooling systems. The compactness of the packs not only decreased the volume occupied by the packs and its associated complex cooling system, but also decreased the total weight for large power application. It is worth to mention that the packs studied in this work are only approximately optimized. The main purpose here is a proof of principle under severe ambient temperature.

2. Experimental work

2.1. Preparation of battery packs

Graphite matrixes filled with commercial PCM was prepared in various dimensions and details can be found elsewhere [8]. The matrix was drilled with holes of 18.2 mm diameter and commercial Type 18650 Li-ion cells with 2.2 Ah or 2.4 Ah capacities were placed. Details of the cells used in this study are listed in Table 1. The module was integrated with safety circuits in order to regulate cells voltage and prevent over-charge. All of the strings in the module were connected in 7S × 2P configuration (seven cells in series and two strings in parallel), 8S2P or 4S4P with a safety circuit that was rated at required discharge current and potential. Mostly, safety circuits are used in order to keep safe operating conditions, which is 1*C*-rate. Therefore, the packs can be discharge within given test conditions up to 2*C*-rate. The specifications of the packs and ambient conditions are summarized in Table 2 and the specifications of PCM/graphite matrix is summarized in Table 3 [8]. The configurations for the packs were selected based on the nominal voltage of the packs which provides 25–30 V.

Table 3
Thermo-physical properties of PCM/graphite composite [8]

Thermal conductivity	16.6 W (m K)$^{-1}$
Latent heat	185 kJ kg^{-1}
Specific heat	1.98 kJ (kg K)$^{-1}$
Bulk density of composites	789 kg m^{-3}

The PCM used in this test has a particular melting range, carbon/PCM ratio, composite density, and thermal conductivity, which may be varied case by case in a more systematic optimization. Thus, the patented PCM/graphite composite material makes Li-ion packs operable in enlarged or shortened operating windows with safer environment. The generic optimization used in the tests reported here were fine-tuned depending on current load pattern and cell thermal characteristics. Also, the effectiveness of PCM is not limited to a particular type of chemistry. Roth studied thermal characterization of Type 18650 Li-ion cells with calorimetric techniques and showed abrupt exothermic spikes between 105 °C and 135 °C [9]. In this study, the safety limit for the cells was considered as 85–90 °C on the body of the cells so that there will be enough room before the cells go into thermal runaway.

2.2. Testing of high-power packs

The battery packs with and without PCM were assembled and tested at room temperature and/or 45 °C. Two K-type thermocouples were placed inside the pack; one at the center and the other one at the corner, to measure the temperature response at the two extreme locations in the pack. The battery pack was charged first in galvanostatic mode at either 0.7*C*-rate or 0.333*C*-rate with a voltage cut-off limit of 4.2 V per cell and then in a potentiostatic mode until the current drops to 50 mA per cell. An hour resting period was then followed by either 1*C* or 2.08*C* discharge rate until the voltage drops to 3.0 V per cell after which a 2-h discharge-resting period completed one full cycle. The schematic of the loads is shown in Table 4 and a schematic of the experimental setup is shown in Fig. 1.

The battery packs in 7S2P configuration were also tested at high discharge rates, i.e. 8.0 A and 10 A, to study the effect of various discharge rates on the battery safety and performance.

3. Results and discussions

3.1. PCM effect on overall temperature and temperature uniformity along the pack

Each cell used in the battery packs were tested initially for the observance of rated capacity so that the cells that were used in the packs were in good condition. Therefore, the cells were charged at *C*/3 and discharged at *C*-rates. The cells showed high capacities even at relatively high discharge rates. The average discharge capacities of tested cells at *C*/1 discharge rate were corresponding to 92.7% and 93.4% of the nominal capacities for 2.4 Ah and 2.2 Ah cells, respectively. The cells that were above 90% of the nominal capacity were chosen for the packs.

The effect of PCM matrix was tested on the 8S2P packs with and without PCM. It is clearly seen from the results that the surface temperature of the cells were kept at lower temperatures when PCM was used (Fig. 2). The reason for this behavior was due to the absorption of the dissipated heat by the PCM. The wax used in this test had a melting *range* of 42–45 °C and the calculated ratio of wax volume to pack volume was 80%. The temperature of the cells in the PCM matrix showed constant increase until the pack temperature reached the *melting range* of the PCM. The result shows that the rate of temperature increase with PCM was slower in this region than the one without PCM. This is because a continuous matrix was achieved when the gaps between the cells filled with the PCM. In other words, the rate of poor heat convection was replaced with high heat conduction. The enhancement of thermal conductivity was studied by our group and the detailed work can be found elsewhere [8,10]. When the temperature of the pack reached the *melting range*, a knee like curve in Fig. 2 explained the behavior of PCM as

Table 4
Schematics of load cycles

	Test 1 4S4P	Test 2 8S2P	Test 3 7S2P T_{amb} = 25 °C	Test 4 7S2P T_{amb} = 45 °C
Charge	0.3*C*-rate (2.93 A)	0.3*C*-rate (1.46 A)	0.7*C*-rate (3.36 A)	0.7*C*-rate (3.36 A)
Rest	1 h	1 h	1 h	1 h
Discharge	1*C*-rate (8.8 A)	1*C*-rate (4.4 A)	1*C*-rate (3.36 A)	2.08*C*-rate (10 A)
Rest	2 h	2 h	2 h	2 h

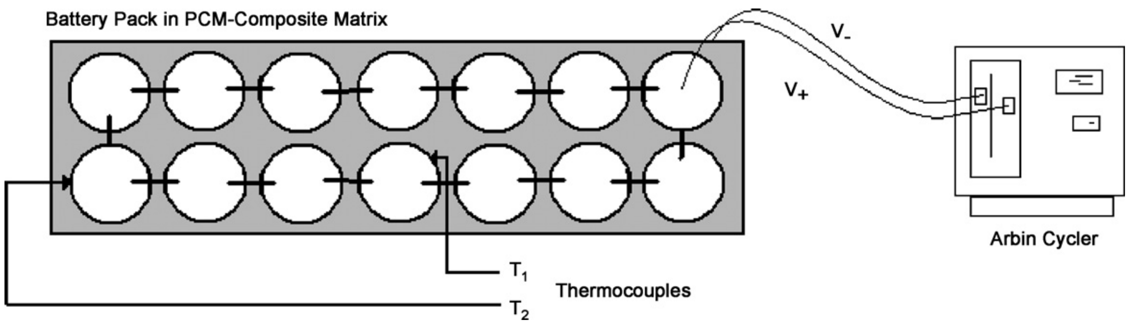

Fig. 1. Schematic of experimental setup (not scaled).

the change of slopes of the temperature–time curve happens during melting of the PCM. This is because of the utilization of the latent heat of fusion of the PCM by the Li-ion pack.

Another remarkable point was the temperature difference between the cells. When no PCM was used, there was near 10 °C difference between the surface temperature of the cell in the center of the pack and the surface temperature of the cell at the corner of the pack. Amine et al. showed the effect of temperature on the capacity fade for single prismatic cells and indicated that the interfacial impedance of the graphite electrode increased significantly [11]. Based on our experimental results, it shows that each cell in a pack will have different thermal environment if it is cycled without any thermal management system. Therefore, cells will undergo capacity fading at different rates due to the different surrounding temperature and eventually will result in lower capacity utilization and faster capacity fading. On the contrary, the temperature difference along the pack with PCM was only 4 °C. Further optimization of the PCM/graphite can result in more temperature uniformity in the pack with PCM. Impregnation of PCM into graphite matrix is under investigation and so far we have achieved 95% v/v PCM/pack ratio. The results will be published soon. Moreover, the pack with PCM was kept below 45 °C at all times which is preferred for Li-ion cells.

Fig. 2 depicts the slow cooling for the pack with PCM. It took longer time for the pack to reach its initial state. However, this did not create any safety concern as the maximum temperature of the pack was already below 45 °C. An increase in temperature during the charge period ($t > 150$ min) was a result of charge at constant current due to higher ohmic behavior. This behavior ended during charge at constant voltage as the current gradually drops down to 50 mA per cell.

3.2. Effect on long-term performance (capacity fading)

The initial performance of each pack was very important to see the long-term effect of PCM. Fig. 3 depicts the initial discharge capacities for 4S4P packs with and without PCM when the packs were discharged at *C*/1-rate. At the beginning of the long-term test, the pack without PCM showed a higher discharge capacity. Amine et al. showed that electrolyte conductivity and electrode wetting properties increase at high temperature at the expense of higher degradation rate [12]. Due to the larger thermal mass of the pack PCM and the temperature control by PCM, the operating temperature of the pack with PCM was lower than the pack without PCM, therefore, resulting in lower initial capacity. At the end of the first cycle, initial capacities of the packs were measured as 94% and 88%

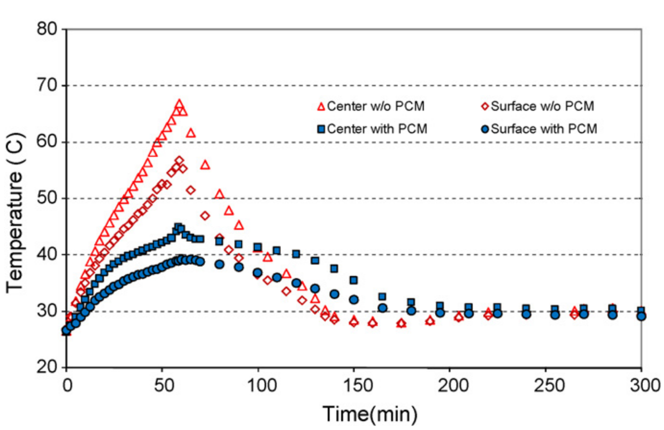

Fig. 2. The effect of PCM on the thermal response of the 8S2P battery packs.

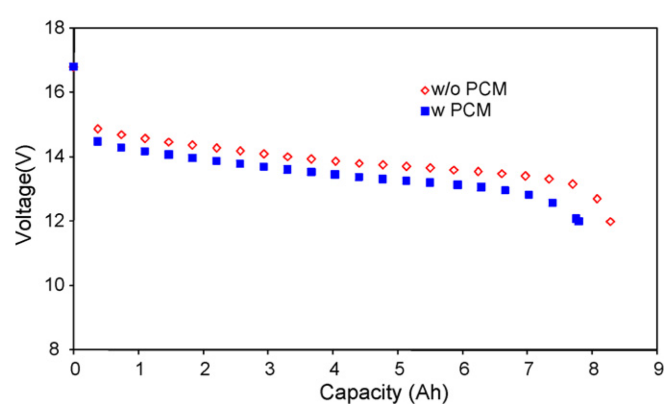

Fig. 3. Initial discharge capacities of 4S4P battery packs *with and w/o* PCM at room temperature and *C*-discharge rate (8.8 A).

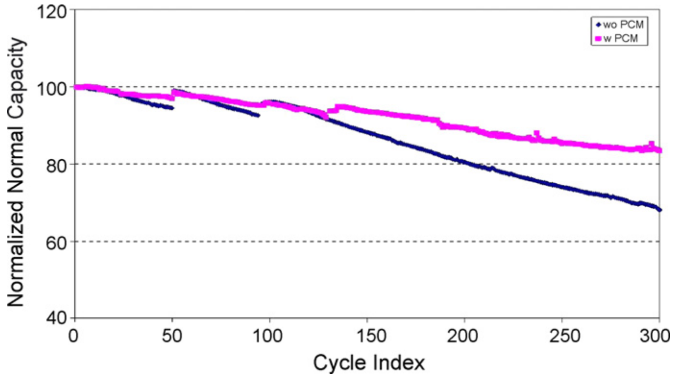

Fig. 4. Capacity fading of 4S4P battery packs *with and w/o* PCM (Ah) (charge: 0.3*C*-rate/2.93 A, 1*C*-rate/8.8 A, T_{amb}: 25 °C, capacity: 8.8 Ah).

of the nominal capacity (8.8 Ah) for the packs without PCM and with PCM, respectively.

The effect of temperature on the packs can be seen in the long-term; therefore the battery packs were cycled for 300 cycles. Fig. 4 shows the cycle-life of the battery packs. The discontinuous behavior of the packs at near cycles 50, 96, and 135 was due to power failure which cause interrupt of the cycling experiment. After each relaxation the capacity fading for the pack without PCM was significantly higher than the one with PCM. Table 4 shows the corresponding fading rates for each pack after each stop. The fading for the pack with PCM was always lower than 6 mAh cycle^{-1} whereas it was around 12 mAh cycle^{-1} for the one without PCM. As explained above, this was due to the lower operating temperature around the cells in the pack with PCM. A detailed study on Li-ion cells at elevated temperatures was published by Ramadass et al. and as they increased the operating temperature from 45 °C to 50 °C, they observed that the fading rate doubled for commercial 18650 cells [13]. In our study, the operating temperature for the pack with PCM was kept at below 45 °C at all times with a gain of considerably low capacity fading (Table 5).

3.3. *Effect of PCM on the packs under stressed conditions*

Next, the battery packs were tested under stressed conditions. Fresh packs were built in the configuration of 7S2P which had smaller gaps between cells (2 mm). The reason for decreasing the gap by 50% is to see the behavior of PCM/graphite composite below and above the *melting range* of the wax under excessive load. Fig. 5 depicts the temperature profile of the battery packs without PCM at different discharge rates. When the pack was discharged at a *C*/1 rate, the battery packs ran for about an hour and 93% of its nominal capacity was utilized. It was clear that the heat generated at 1*C* during discharge was high enough to cause safety concerns at this condition. On the other hand, since the body temperature of the cell at the center of the pack reached the limit of 85 °C, the tests were immediately stopped at 20 min and 12.7 min, when the discharge rates were 1.67*C* (8 A) and 2.08*C* (10 A), respectively. In both

Table 5
Capacity fading for 4S4P packs

Cycles	With PCM (mAh cycle^{-1})	Without PCM (mAh cycle^{-1})
1–50	5.5	10.7
51–96	5.3	13.4
97–300	5.7	12.2

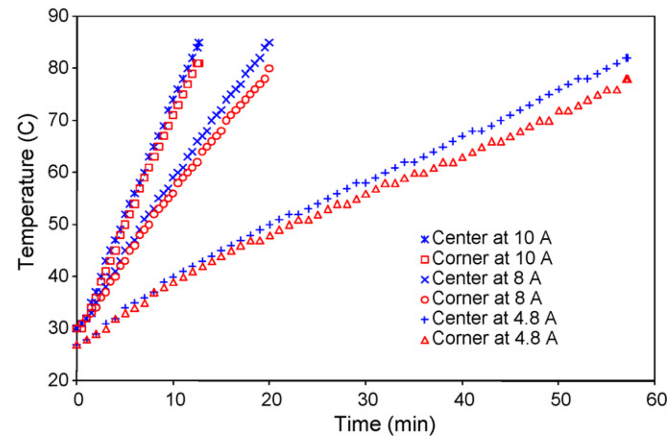

Fig. 5. Temperature profiles of packs without PCM at different discharge rates (T_{amb} = 27–30 °C).

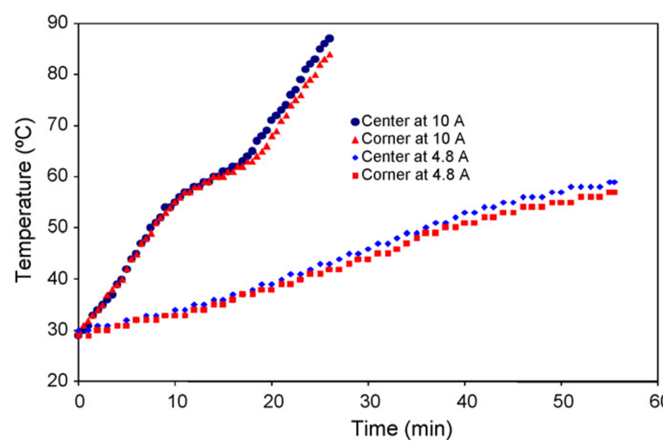

Fig. 6. Temperature response of the pack with PCM at different rates (T_{amb} = 27–30 °C).

cases the experiments were stopped due to safety concern before nominal capacity was utilized. Therefore, it is clear that optimum operating temperature of the batteries is an important aspect of any thermal management system in order to gain the full capacity of the battery packs.

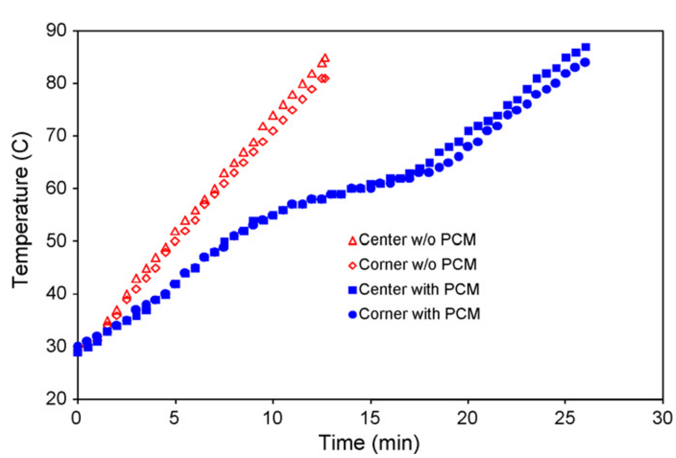

Fig. 7. Temperature profile of packs with and w/o PCM during discharge at 10 A and 30 °C.

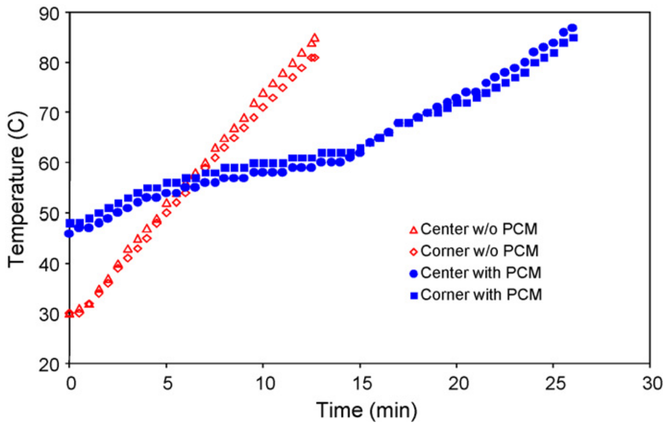

Fig. 8. Temperature profile for the pack at 10 A discharge current, at 45 °C with PCM compared to 30 °C w/o PCM.

Fig. 6 shows temperature profile for a 7S2P pack with PCM (melting range: 52–55 °C). Temperature uniformity along the pack was achieved and the maximum temperature difference along the pack was only 2 °C. Results also depict that near 90% of the standard capacity was utilized at 2.1*C*-rate during discharge.

The comparison of the temperature profiles of the battery packs with and without PCM at 10 A discharge current are shown in Fig. 7. The pack with PCM was able to discharge completely and utilized 90% of the nominal capacity. On the other hand the pack without PCM utilized near 50% of the nominal capacity before the experiment was stopped for safety concerns. The results clearly demonstrate that by using PCM it is possible to utilize the full capacity of the battery at high discharge rates even at elevated temperatures.

The pack with PCM was also tested at elevated temperature, i.e. T = 45 °C. Fig. 8 shows the comparison between the packs with PCM at 45 °C and 10 A discharge current and the pack without PCM at 30 °C at the same 10 A discharge current. The pack without PCM utilized only 42% of nominal capacity before the experiment stopped while the one with PCM was completely discharged safely and utilized 90% of the nominal capacity even at elevated temperature of 45 °C. The small temperature difference (2–3 °C) between the center and the corner confirms the uniform distribution of PCM in the matrix and that the heat is conducted efficiently throughout the graphite matrix. The amount of PCM and its melting temperature had a significant effect on keeping the temperature of the battery pack within safety limits. The results also showed that with PCM thermal management about 90% of the nominal capacity was utilized repeatedly even at extreme conditions with high discharge rate and high ambient temperature (Fig. 9a and b). The recovery of full capacity after stressed operation is an important evidence of flexibility of PCM as a thermal management tool.

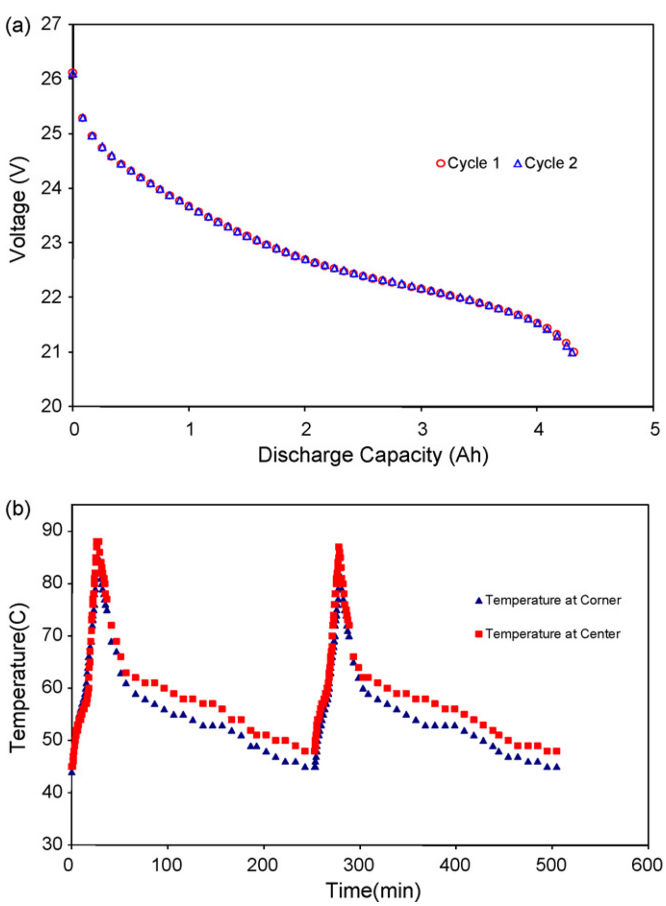

Fig. 9. (a) Discharge capacity of 7S2P pack under stressed conditions (pack rated capacity: 4.8 Ah). (b) Temperature profiles of 7S2P pack with PCM (T_{amb}=45 °C, discharge rate=2.1*C*-rate/10 A).

4. Conclusions

The advantage of using the novel PCM thermal management systems over conventional active cooling systems was successfully demonstrated in this study. Compared to complex cooling systems, the packs with PCM provide compactness to various power applications and reduce the weight of the large power systems. Discussed results show that PCM passive control may make active control, if necessary at all, complementary and/or secondary in function, and therefore leads to a much simplified and more economic design.

It was also demonstrated that the thermal stability of the pack requires controlled thermal management during fast discharge and high temperature applications for high-energy Li-ion cells (i.e. 10 A discharge and 45 °C temperature). To our best knowledge, this is the first time that such accomplishment was achieved using a passive thermal management system under such extreme conditions.

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