A hybrid thermal management system for lithium ion batteries combining phase change materials with forced-air cooling

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12 **Abstract**

13 Passive thermal management systems using phase change materials (PCMs)
14 provides an effective solution to the overheating of lithium ion batteries. But this
15 study shows heat accumulation in PCMs caused by the inefficient cooling of air
16 natural convection leads to thermal management system failures: The temperature in a
17 battery pack operating continuously outranges the safety limit of 60 °C after two
18 cycles with discharge rate of 1.5C and 2 C. Here a hybrid system that integrates
19 PCMs with forced air convection is presented. This combined system successfully
20 prevents heat accumulation and maintains the maximum temperature under 50 °C in
21 all cycles, even with 7 °C rise in the ambient temperature. Study on airspeed effects
22 reveals that thermo-physical properties of PCMs dictate the maximum temperature
23 rise and temperature uniformity in the battery pack, while forced air convection plays
24 a critical role in recovering thermal energy storage capacity of PCMs. A numerical
25 study is also carried out and validated with experiment data, which gives theoretic
26 insights on thermo-physical changes in this hybrid battery thermal management
27 system.

28 **Key words:** phase change material; forced air convection; thermal management;

29 Li-ion battery; passive cooling; hybrid system.

30

1. Introduction

Li-ion batteries are considered as excellent power sources for hybrid or electric vehicles (HEVs/EVs) due to high energy and power density [1]. However, the remarkable deterioration in the performance of Li-ion batteries at elevated temperatures is hampering their practice applications [2-4]. It has been reported that the optimum operating temperatures for Li-ion batteries should range from 20 to 50 °C, and the maximum temperature difference in a battery pack should be kept within 5 °C [5]. Consequently, efficient thermal management systems are highly needed for Li-ion batteries to remove the massive amounts of heat generated in the charge/discharge process.

Forced air convection is a routine solution to thermal management of electronic components. However, forced air convection brings about nonuniform temperature distribution in a battery pack, leading to different degrading rates for each cell [6, 7]; As a result, the cycle life of the whole pack would be shortened. Although optimizing the flow ducts for air can improve the temperature uniformity, system complexity is also increased [8-11].

Recently, passive thermal management system using phase change materials

48 (PCMs) have been developed as an alternative to active cooling. Taking advantage of
49 high latent heat, PCMs can absorb the massive amounts of heat generated by Li-ion
50 batteries and keep the temperatures of the batteries within the melting range of the
51 PCMs [12, 13], thereby reducing both the maximum temperature and the temperature
52 difference in the battery pack [14-16]. Furthermore, the performance of the passive
53 thermal management systems can be improved via the enhancement in the thermal
54 conductivity of the PCMs by adding some thermal conductive materials like metal
55 foams [17, 18], graphene [19], or expanded graphite (EG) [20-23]. Mills et al. [20]
56 reported that PCMs decreased the capacity loss rate of Li-ion batteries by more than
57 50%.

58 In spite of the high efficiency, PCMs can only absorb heat passively. Running out
59 of the available latent heat under extreme conditions - such as high heat density
60 during the high-current charge or discharge, or high ambient temperature - may cause
61 the thermal management systems [24]. Even under mild conditions (ambient
62 temperature $< 30\text{ }^{\circ}\text{C}$ and discharge rate $< 2\text{ C}$), lack of external cooling may also lead
63 to thermal management system failures. A typical charge-discharge protocol for
64 batteries includes three steps: (1) constant-current charging; (2) constant-voltage

65 charging; (3) constant-current discharging. In continuous operations, most heat is
66 generated by batteries during discharging (Step 3), which is absorbed by PCMs. The
67 heat stored in PCMs can only be dissipated to the ambient during charging (Step 1
68 and Step 2). If natural convection is not sufficient to cool batteries and PCMs down
69 before the next discharge starts, accumulated consumption of latent heat will result in
70 uncontrollable battery temperature rise. Therefore, passive thermal management
71 systems using PCMs are needed to combine with more efficient active cooling to
72 avoid potential failures and improve its reliability. However, few attention has been
73 paid on hybrid active–passive thermal management systems by now, except Hassan
74 introduced forced air convection to improve the performance of a passive thermal
75 management system at high ambient temperatures [25].

76 In the current work, a novel thermal management system that integrates the PCM
77 and forced air convection is presented for a 5S4P Li-ion battery pack that charges and
78 discharges continuously without rest. Its thermal management performance is
79 compared with the passive thermal management system using paraffin/EG composite.
80 This hybrid system is supposed to boost the reliability of PCM-based thermal
81 management system without sacrificing its simple structure. Besides the experimental

investigation, a numerical study based on the combination of thermal models for batteries [26-28] and the enthalpy-based models for PCMs [29] is also carried out and validated with the experiment data, which gives theoretic insights into thermo-physical changes in this hybrid thermal management systems.

2. Experimental Section

2.1. Battery pack

Twenty 18650 Li-ion batteries (Samsung, ICR18650-26FM, 2.6Ah) were connected in the way of 5S4P (five cells in series and four cells in parallel). Specifications of this battery module are listed in Table 1. The spacing between centers of two neighboring cells was 30 mm. As shown in Figure. 1(a), six K-type thermocouples with error less than 0.5 °C were attached to the battery surface (marked in green), to monitor the temperature evolution of batteries. Thermal management performance is judged by two indices: (1) the maximum temperature $T_{\max}$ and (2) the maximum temperature difference ΔT in this pack. In any cases, lower $T_{\max}$ and ΔT are desirable.

The battery pack was charged and discharged by a battery cycler CT2001D, manufactured by Wuhan LAND electronics Co., Ltd, China, current and voltage

99 accuracy of which were $\pm 0.1\%$. The charge process of the battery module was the
100 same, which first charge the pack in galvanostatic mode at 1C rate with a voltage
101 cut-off limit of 4.25V per cell followed by a potentiostatic mode until the current
102 drops to 50 mA per cell. The fully charged battery module was then discharged at
103 constant C-rates - 1C, 1.5C or 2C - with a voltage cut-off limit of 2.75V per cell. The
104 charge-discharge cycle went on for five times, but would be manually stopped to
105 prevent damages to batteries if the maximum temperature of battery exceeded 60 °C.

106 **2.2. Thermal management systems**

107 Battery performance was tested under two different thermal management
108 systems: (1) completely passive thermal management system; (2) hybrid thermal
109 management system that combined PCMs with force air convection.

110 In the passive thermal management system, batteries were surrounded by
111 RT44HC/EG composite PCMs. RT44HC (purchased from Ruhr Energy Technology
112 Co., Ltd, Hangzhou, China) was chosen as the basic PCM, due to its proper melting
113 point and high specific phase change enthalpy. RT44HC/EG composites were
114 prepared in the same way as mentioned in [30]. Thermo-physical properties of
115 RT44HC/EG composite are listed in Table 2. Passive thermal management was tested

116 at the ambient temperature around 25 °C.

117 In the system combining PCM with forced air convection, the battery module
118 that had inserted into RT44HC/EG composites was put into an air tunnel with a
119 rectangular cross section, whose area was 200×200 mm². The full structure also
120 included: a 500 mm long contraction with the inlet cross section area of 400×400
121 mm²; two valves that controlled air flow rate; and an exhaust fan that drew in air, as
122 shown in Figure 1(b). Before flow through the battery module, airspeed was measured
123 by an anemometer (KA33, provided by KANOMAX) with an accuracy of ±0.1m/s.
124 The inlet temperature of air ranged from 30-34 °C. The airspeed was firstly fixed to 5
125 m s⁻¹, but then set to 3 m s⁻¹, 1 m s⁻¹ to further study the effect of airspeed on thermal
126 management performance.

127 In this study, charge efficiency reached over 95% and discharge efficiency
128 ranged from 80% to 95%, depending on discharge rates. Difference of
129 charge/discharge efficiency in two different thermal management systems was less
130 than 5% unless it was manually stopped, because thermal management systems has a
131 small impact on battery cycle performance in only five cycles. We focus on battery
132 thermal characteristics in different thermal management systems, rather than its

133 electrochemical performance.

134 **3. Mathematical model**

135 **3.1. Battery model**

136 Two models can describe the heat generation in batteries: 1) First-principle
137 models that track the progress of electrochemical reactions, part of which comprises
138 the heat generation term [31-34]; 2) Lump-capacitance model that focuses on battery
139 thermal behaviors [8, 26-28].

140 The lump-capacitance model is used in this work, because of its smaller
141 computational cost. Lump-capacitance model considers physical properties in the
142 battery cell homogeneous, and calculates heat generation rate with the measured the
143 current-voltage characteristics of batteries.

144 The energy conservation equation in a battery cell is specified as

$$\rho_b c_{p,b} \frac{\partial T}{\partial t} = k_b \nabla^2 T + q \quad (1)$$

145 Where ρ_b , $c_{p,b}$ and k_b represent density, specific heat and thermal conductivity of
146 battery, which is listed in Table 2. T and t represent the local temperature and time, q
147 is the battery heat generation rate per unit volume, which is approximated by the sum
148 of the irreversible Joule heat q_{irr} and reversible entropic heat q_{rev}

149 q_{irr} is caused by the battery internal electrical resistance, which can be
150 determined by the product of current I and the difference between terminal voltage U
151 and equilibrium voltage E:

$$q_{irr} = \frac{I(U - E)}{V} \quad (2)$$

152 where V is the volume of each battery cell. U for every single cell was recorded
153 during charge and discharge cycles by the battery cycler. E was approximated by the
154 voltage during the charge and discharge of one battery at a low C-rate: 0.05 C.
155 Although the current was small enough, the voltage during a charge differentiated
156 from that in discharge. Therefore, correlations between E and DOD were provided for
157 charge and discharge separately:

158 $E = -5.50 \times DOD^4 + 8.65 \times DOD^3 - 3.69 \times DOD^2 - 0.45 \times DOD + 4.12$, (3-1) for discharge

159 $E = -28.67 \times DOD^6 + 73.93 \times DOD^5 - 73.96 \times DOD^4 + 35.05 \times DOD^3 - 6.95 \times DOD^2 -$
160 $0.50 \times DOD^4 + 4.23$, (3-2) for charge

161 The reversible heat represents the entropic loss ΔS and is given as

$$q_{rev} = -\frac{T\Delta S}{V} \quad (4)$$

162 The entropy change was an function of DOD and I, based on the data in [27], a
163 correlation is given by:

$$\Delta S = (110.8 \times \text{DOD}^3 - 229.1 \times \text{DOD}^2 + 64.3 \times \text{DOD} - 11.4) I/F \quad (5)$$

In which F is the Faraday constant, 96 485.3 C mol⁻¹.

3.2. PCM model

Enthalpy method was used to simulate the heat transfer in PCMs, in the same way as described [30]. Thermo-physical properties are listed in Table 3 and energy equations come as follows:

$$\rho_{PCM} \frac{\partial H}{\partial t} = k_{PCM} \nabla^2 T \quad (6)$$

$$H = h + \Delta H \quad (7)$$

$$h = \int_{T_0}^T c_{p,PCM} dT \quad (8)$$

$$\Delta H = \beta \gamma \quad (9)$$

$$\beta = \begin{cases} 0 & T < T_m \\ 1 & T > T_m \end{cases} \quad (10)$$

ρ_{PCM} — packing density of PCM, k_{PCM} — thermal conductivity of PCM, H — enthalpy of PCM, h — sensible heat of PCM, ΔH — enthalpy of melted PCM, β — liquid fraction of melted PCM, γ — specific phase change enthalpy.

3.3. Initial conditions and boundary conditions

The initial state is specified as:

$t=0$;

176 $T(x,y,z)=T_0;$

177 Boundary condition at the interface between battery and PCM relies on the
178 energy conservation:

$$-k_b \frac{\partial T}{\partial n} = -k_{PCM} \frac{\partial T}{\partial n} \quad (11)$$

179

180 $\frac{\partial T}{\partial n}$ represents temperature gradient.

181 The contact surface between PCM and the aluminum container shares similar
182 boundary condition:

$$-k_{PCM} \frac{\partial T}{\partial n} = -k_{Al} \frac{\partial T}{\partial n} \quad (12)$$

183 The energy equation at the boundary between heat sink and the ambient is given
184 by:

$$-k_{Al} \frac{\partial T}{\partial n} = \alpha(T - T_{amb}) \quad (13)$$

185 Measuring the convective heat transfer coefficient α is a tough task, so we
186 undertook a preliminary CFD study with commercial software Fluent 6.3 to determine
187 α on average. Battery pack was simplified to six walls with a constant temperature of
188 42 °C (phase change temperature), over which air flowed at different rates, as shown
189 in Figure. 2. Heat transfer between the six surfaces and air was simulated, and heat

190 transfer coefficients at different air flowrates are listed in Table 4.

191 After assigning initial conditions and boundary conditions, battery model and
192 PCM model were solved simultaneously by Fluent 6.3.

193 **4. Results and Discussion**

194 **4.1. Comparison on Performance of Passive and Active** 195 **Thermal Management Systems**

196 Figure 3 (a) compares the maximum temperature ($T_{\max}$) history in the battery
197 pack equipped with two different thermal management systems, discharging at
198 1C-rate. Both the completely passive thermal management system (black lines) and
199 the enhanced system that combines PCM with forced air convection (red lines)
200 successfully kept battery temperature under 45 °C in all five cycles. Peak temperature
201 in each cycle was similar between two thermal management systems, but $T_{\max}$ at the
202 end of a charge in the combined system was lower than the completely passive
203 system— even though the ambient temperature was nearly 7 °C higher.

204 With higher discharge rates such as 1.5C and 2C, the completely passive
205 systems only succeeded in controlling battery temperature under 45 °C in the first two
206 cycles, but failed at the third discharge: $T_{\max}$ exceeded 60 °C, as shown in upper black

207 lines Figure. 3(b) and Figure. 3(c).

208 The poor heat transfer rate between PCMs and the ambient should be responsible
209 for the failure of passive thermal management system. When discharging at small
210 currents, small amounts of heat produced by batteries are stored in PCMs primarily as
211 sensible heat, which can be easily dissipated to the ambient environment via the low
212 efficient air natural convection. However, with high-current discharge, much more
213 heat is generated and stored in PCM primarily as latent heat. Although at lower
214 ambient temperature, the inefficient air natural convection is unable to fully release
215 the latent heat before next discharge starts, which: 1) reduces heat storage capacity; 2)
216 increases the initial temperature for next discharge. After several discharges, the
217 shrunk thermal energy storage capacity fails to deal with the high heat flux from
218 batteries— latent heat is run out in very short time and battery temperature rises
219 beyond the safety limit.

220 The hybrid system provides a more reliable thermal management performance.
221 Auxiliary forced air convection cools PCM down below its phase change temperature
222 during the charge, which prevents heat accumulation. Full recovery of latent heat
223 helps maximize the thermal energy storage capacity of PCM and keep $T_{\max}$ under 46

224 °C in all cycles, as show in red lines in Figure. 3(b) and Figure. 3(c). Forced air
225 convection lowers the battery temperature and accelerates the latent heat release,
226 preventing the failure of the PCM-based passive thermal management system.

227 Temperature distribution in both the completely passive or combined thermal
228 management system remains uniform, thanks to the high thermal conductivity and
229 heat capacity of RT44HC/EG. In all cycles discharging at any rates, the average
230 maximum temperature difference in the module is kept below 3 °C, as show in Figure
231 4.

232 **4.2. Airspeed effect**

233 Figure. 5 compares $T_{\max}$ histories in the hybrid systems with different airspeeds,
234 showing airspeed higher than 3 m s⁻¹ is sufficient to prevent heat accumulation to keep
235 battery temperature within safety range throughout all cycles.

236 However, the maximum temperature in the first cycle was kept below 46 °C -
237 close to the phase change temperature of RT44HC - in both the completely passive
238 system and hybrid system with airspeed of 5 m s⁻¹. The PCM with the maximum
239 thermal energy storage capacity dominates the maximum battery temperature rise.
240 Controlling battery temperature within the optimum range requires PCMs with proper

241 phase change temperature and sufficient amount of available latent heat.

242 In addition, the average temperature difference in the battery pack is also
243 independent of airspeeds, as shown in Figure.6. Great heat capacity and thermal
244 conductivity of the PCM/EG composite dictates a uniform temperature distribution.

245 Thermo-physical properties including the phase change temperature,
246 sensible/latent heat capacity and thermal conductivity play an important role in
247 PCM-based thermal management systems. Active cooling plays a more important role
248 in cooling PCM down to unleash PCM's potential in thermal energy storage.

249 **4.3. Model validation**

250 Temperature history of Cell 2 and Cell 5 obtained from simulation is compared
251 with experiment data, shown in Figure. 7(a~d). In the system cooled by air natural
252 convection, numerical result matches well with experiment well.

253 In the system cooled by forced air convection, numerical model underpredicts
254 battery temperature. Neglecting thermal resistance between battery and PCMs or
255 prediction errors of convective heat transfer rate in our preliminary CFD study may
256 contribute to these deviation. Simulation also reports a slower temperature drop near
257 42 °C but faster near 37 °C. Due to the limit of Fluent solver, we assumed that PCM

258 only had one endothermic peak and one exothermic peak during phase change. But
259 the composite PCM actually has two endothermic peaks and two exothermic peaks, as
260 shown in the DSC curve in Fig. 8. One-peak assumption makes sense in the melting
261 process, because of the narrow difference between two peaks ($<2\text{ }^{\circ}\text{C}$). But as the the
262 second exothermic peak temperature ($36.3\text{ }^{\circ}\text{C}$) is much lower than the first peak
263 (43.3°C). This assumption to put two peaks into one causes two problems: (1)
264 increases the latent heat of first exothermic peak, which slows down the temperature
265 drop at $42\text{ }^{\circ}\text{C}$; 2) neglect the second phase change, which accelerates the temperature
266 drop at $37\text{ }^{\circ}\text{C}$ — which explains the discrepancy between model and experiment. But
267 overall, numerical results match experiment data well: the average temperature
268 difference between numerical model and experiment is less than $2.3\text{ }^{\circ}\text{C}$.

269 With the help of numerical model, we can give an insight of the impact of heat
270 accumulation. Figure 9 demonstrates melting fraction of PCMs along with time, in
271 systems with different airspeed. In the system cooled by air natural convection and
272 forced convection with airspeed $\leq 1\text{ m s}^{-1}$, melting fraction rises from 50% and 35%
273 at the end of first cycle, to 90% and 78% at the end of fourth cycle, which is a sign of
274 ongoing reduction of latent heat. For systems cooled by strong convection (air

275 speed $\geq 3\text{ m s}^{-1}$), melting fraction at the end of each cycle drops down to zero due to
276 the efficient heat transfer between heat sink and the ambient. A full recovery of
277 thermal energy storage capacity helps control battery temperature under safety limit.

278 **5. Conclusions**

279 This study aims at raising attentions on potential failures of thermal management
280 systems using PCMs for Li-ion batteries under stressful operations and the
281 significance of effective external cooling for PCMs.

282 We find that heat accumulation in PCMs caused by the inefficient natural
283 convection of air will lead to the final failure of the passive thermal management
284 system. Although the PCM-based thermal management system effectively maintains
285 battery temperature within safety range in a single charge-discharge cycle, the
286 maximum temperature in the 5S4P battery pack exceeds the safety limit of $60\text{ }^{\circ}\text{C}$ after
287 two cycles, with charge rate of 1C and discharge rate of $1.5\text{C}/2\text{C}$, at room temperature
288 of $25\text{ }^{\circ}\text{C}$.

289 Integrating PCMs with the more efficient active cooling improves the reliability
290 of passive thermal management system. The hybrid system presented in this paper
291 that combines forced air convection successfully controlled the maximum temperature

292 of the battery pack below 50 °C in cycles at any rates lower than 2C, even with a 7 °C
293 increase in ambient temperature.

294 The introduction of forced air convection will not affect the uniform temperature
295 distribution provided by PCMs. The maximum temperature difference in the hybrid
296 thermal management system was less than 3 °C.

297 Study on airspeed effects shows the maximum temperature rise and the
298 maximum temperature difference in the battery pack is independent on air speed only
299 if PCM has full thermal energy storage capacity. Therefore, PCMs and forced air
300 convection are considered to play different roles in battery thermal management–
301 PCMs control the maximum temperature and the maximum temperature difference in
302 battery pack, while forced air convection cools PCMs down in intervals between two
303 discharges, which helps recover the latent heat of PCMs.

304 Numerical model shows reasonable agreement with experiment, with average
305 temperature deviation less than 2.3 °C. It is easy to observe from numerical results
306 that melting fraction of PCMs at the end of each cycle keeps rising in the completely
307 passive thermal management system, but remains zero in the hybrid thermal
308 management system, which helps explain the heat accumulation in PCMs.

309 The passive thermal management system combined with forced air convection
310 has a simple structure, high efficiency and reliability. Our further studies will focus on
311 taking advantage of the higher cooling efficiency of active cooling to cut the
312 excessive weight of PCMs.

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414 Figure Captions

415 Fig. 1 Schematic of experiment: (a) A 5S4P battery pack, temperature of six cells

416 (marked in green) was measured through K-type thermo-couples; (b) structure

417 of the air channel

418 Fig. 2 Illustration of the simplified model for preliminary CFD study which

419 determines the surface convective heat transfer rate (temperature of six walls of

420 inside box treated as constant 42 °C)

421 Fig. 3 Comparison of the maximum temperature in the battery pack between the

422 completely passive thermal management system and the hybrid system that

423 combines PCMs and forced air convection with discharge rate: (a) 1C; (b) 1.5C;

424 (c) 2C

425 Fig. 4 Comparison of historic average maximum temperature difference in passive

426 and hybrid systems

427 Fig. 5 History of the battery maximum temperature in the hybrid thermal

428 management system with different airspeeds (discharge rate: 1.5C)

429 Fig. 6 Comparison of historic average maximum temperature difference in the

430 hybrid system with different airspeeds (discharge rate: 1.5C).

431 Fig. 7 Comparison between numerical results and experiment in the cycling with

432 discharge rate of 1.5C: temperature of (a) Battery 5# and (b) Battery 2# in the

433 passive thermal management system; temperature of (c) Battery 5# and Battery

434 2# in the hybrid system with airspeed of 5 ms^{-1}

435 Fig. 8 DSC curves for RT44HC (80 wt%)/EG composites

436 Fig. 9 History of the PCM melting fraction in the hybrid thermal management

437 systems with different airspeeds (pack discharge rate: 1.5C)

438

439 **Table**

440 Table 1 Specification of Li-ion battery module

Property	Specification
Cell type	Type 18650
Module capacity	10.4 Ah
Pack operating voltage	13.75V-21.25V
Max. discharge rate	2 C-rate (20.8A)
Max. Charge rate	1 C-rate (10.4A)
Operating temperature	20-40 °C

441 Table 2 Thermo-physical properties of battery cell

Property	Value
Density (kg m^{-3})	2700
Thermal conductivity ($\text{W m}^{-1} \text{K}^{-1}$)	7.14
Specific heat ($\text{kJ kg}^{-1} \text{°C}^{-1}$)	0.9

442 Table 3 Thermo-physical properties of RT44HC/EG composite

Property	Value
EG mass fraction (%)	20%

Bulk density (kg m ⁻³)	714
Specific heat (kJ kg ⁻¹ °C ⁻¹)	2.5
Latent heat (kJ kg ⁻¹)	181
Thermal conductivity (W m ⁻¹ K ⁻¹)	7.85

443

Table 4 Heat transfer coefficient for each surface and the total heat transfer rate

		5m s ⁻¹	3m s ⁻¹	1m s ⁻¹	Natural convection
Surface heat transfer coefficient (W m ⁻² K ⁻¹)	Downwind	28	19	13	9.9
	Bottom	34	16	11	8.8
	Upwind	58	27	25	9.9
	Side	46	21	14	11.6
	Top	36	19	11	9.9

444

Figure1

Figure2

Figure3a

Figure3b

Figure3c

Figure4

Figure5

Figure6

Figure7a

Figure7b

Figure7c

Figure7d

Figure8

Figure 9