Energy 144 (2018) 977–983

Contents lists available at ScienceDirect

Energy

journal homepage: www.elsevier.com/locate/energy

Thermal management performance of phase change materials with different thermal conductivities for Li-ion battery packs operated at low temperatures

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ARTICLE INFO

Article history:

Keywords:
Li-ion battery
Thermal management
Phase change material
Low temperature
Thermal conductivity
Uniformity

ABSTRACT

Thermal management performance of two composite phase change materials (PCMs)—a highly thermally conductive 60 wt% RT44HC/expanded graphite (EG) composite and a 60 wt% RT44HC/fumed silica composite with a lower thermal conductivity—is studied for a 20-cell Li-ion battery pack working at 5 and −10 °C. The temperature and voltage distributions in each battery pack are measured during single-discharge tests at 0.5C, 1C, 1.5C, and 2C and over 20 charge-discharge cycles that simulate battery operation in an electric vehicle. By comparing these systems with the PCM-free battery pack, we aim to find an appropriate material to improve the low-temperature performance of Li-ion cells. The results indicate that the low thermal conductivity of the RT44HC/fumed silica composite induces an even higher temperature difference over the battery pack than the PCM-free case, causing an uneven voltage distribution and consequently an early end to charging and discharging. However, the highly thermally conductive 60 wt% RT44HC/EG composite PCM can narrow the temperature variation among the cells and hence help to reduce the voltage differences. The high thermal conductivity of the PCM plays an essential role in achieving a uniform temperature distribution to improve the consistency of the battery performance.

1. Introduction

Li-ion batteries are widely used in mobile phones, laptops, and hybrid energy and pure electric vehicles (HEVs and EVs, respectively) because of their low self-discharge rates, high specific powers and energy densities, and long cycle lives [1,2]. However, when the temperature drops to below 0 °C, Li-ion batteries substantially lose both energy and power [3]. The degradation of the electrode and electrolyte properties is responsible for the poor battery performance observed at low temperatures [4]. The limited diffusivity of lithium ions on graphite anode surfaces [5,6] and low electrolyte ionic conductivity [7], which are linked to the formation of solid electrolyte interface (SEI) films [8], have been found to dramatically increase the charge-transfer resistance (R_{ct}) of Li-ion batteries at subzero temperatures [9]. The increased resistance not only reduces the battery voltage, lowering its output power, but also prematurely ends charging and discharging, leading to a decline in capacity [10].

In addition to developing electrodes [11–14] and electrolytes [7,15,16] that work over wide temperature ranges, heating is an easier and more efficient solution to improve the low-temperature battery performance [17,18]. Zhang et al. [19] demonstrated that preheating Li-ion batteries with a sinusoidal alternating current (AC) with an amplitude of 7 A (2.25C) and a frequency of 1 Hz can heat the batteries from −20 to 5 °C within 15 min. Ruan et al. [20] optimized the frequency of the sinusoidal AC to achieve a rapid temperature increase. Ji et al. [3] showed that convective heating using a warm air/liquid system effectively aids battery thermal management. Wang et al. [21] developed a three-electrode battery that can switch to a fast-heating mode by connecting the cathode to a nickel anode. Although effective, heating systems are complicated and expensive; in addition, three-electrode cells are yet to be applied in commercialized battery systems. Moreover, all these methods consume energy to heat the battery, which sacrifices the battery capacity.

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https://doi.org/10.1016/j.energy.2017.12.098

Phase change materials (PCMs) can store and release significant amounts of heat during their solid–liquid phase changes [22]. Thus, PCMs have been widely used to store the excess heat generated by batteries to prevent overheating [23–25]. If we can make use of the heat stored in the PCM by returning it back to the battery, then the battery can be used at low temperatures without consuming its stored energy. Rao et al. [26] and Sasmito et al. [27] proved that PCMs slow the drop in the battery temperature. Our previous work [28] showed that a PCM with low conductivity successfully increased the average temperature of a single 18650 Li-ion battery cell over continuous cycling, which raised its power, discharge voltage, and cycling life.

However, batteries are commonly connected in series and parallel into a pack, which is much more complicated than just a single cell. Cells at different temperatures have uneven internal resistances or electrochemical reaction rates, leading to differences in the terminal voltage and discharge/charge capacity. The voltage/capacity variations between cells continuously increase as the battery pack operates. Ultimately, some cells reach the cut-off voltage earlier than others, which prematurely forces the whole pack to stop charging/discharging, incurring a capacity loss [29]. Therefore, battery pack thermal management not only requires that each battery stays within the optimal temperature range, but also that the temperature differences between the cells should be small. Ling et al. [30] indicated that the high thermal conductivities of PCMs narrow the temperature differences between the cells. In addition, the thermal insulation properties of PCMs become less important in a battery pack as significantly more heat is generated than that generated with one cell and the battery temperature rises faster. Instead of the PCM/fumed silica composite we used in our previous work that considered one cell [28], a PCM with a higher thermal conductivity is a better choice for the thermal management of battery packs operating at low temperatures.

In this work, we prepare two kinds of composite PCM: (1) a 60 wt% RT44HC/expanded graphite (EG) composite with a high thermal conductivity, and (2) a 60 wt% RT44HC/fumed silica composite with a low thermal conductivity. We study their impact on the thermal response and electrochemical performance of 20-cell Li-ion battery packs operated at −10 and 5 °C. We intend to determine the PCM with the properties that are most appropriate for the thermal management of battery packs operated at low temperatures.

2. Experimental

2.1. Battery pack configurations

Each Li-ion battery pack system consisted of 20 commercially available 2.6 Ah 18650 cylindrical cells (Samsung), connected in a 4P × 5S configuration by nickel sheets. The PCMs (580 g, with a density of 527 kg/m^3) were used to fill the gaps between the cells, forming 150 × 120 × 65 mm modules. K-type thermocouples with an accuracy of ±0.1 K were attached on the outside surfaces, in the middle along the height of the batteries, with thermally conductive epoxies (OB101, Omega Engineering). Ten thermocouples were used in the first and second rows to monitor the temperature distribution and history of each battery pack. An infrared thermometer was also used to capture 2D temperature distributions. Fig. 1 shows the battery pack with (a) the RT44HC/EG composite, (b) the RT44HC/fumed silica composite, and (c) no PCM.

2.2. Cooling tests

Cooling tests were carried to test how long the battery could be kept warm with/without PCMs, which studied the ability of the PCM in preventing battery getting cold too fast, when an electric vehicle suddenly stopped after running for a while. Since the battery temperature can rise above the phase change temperature of the PCMs after some time under high discharge current, all three battery pack systems were first heated to 47 °C. Then the battery packs were placed in a severely cold chamber at −10 °C, to test the PCM performance at an extreme condition. The temperature drops with time were recorded and compared to investigate the thermal insulation effects of the different systems. The battery can be kept in a warm condition for longer period, the battery can be better kept away from the harm of too cold.

2.3. Single-discharge tests

To study the short-term influence of the PCMs on low-temperature battery operations, the battery packs were first charged at 25 °C. Each pack was initially charged galvanostatically at 1C per cell until the 4.25 V cut-off voltage was reached for one column, and then the whole pack was held at 21.25 V until the current dropped below 0.05C per cell. The battery packs were then placed in a low-temperature environment at 5 or −10 °C for 2 h and discharged at 0.5C, 1C, 1.5C, or 2C until the voltage of one cell column dropped to 2.75 V. The batteries were charged and discharged by a Land Battery Test System. The voltage of each column in parallel, total voltage, and pack current were recorded by the battery test system, for which the accuracy of the voltage and current measurements were within 0.1%.

2.4. Charge–discharge tests over 20 cycles

To study the effect of the PCMs on the battery packs under realistic operating conditions, we adopted the urban dynamometer driving schedule (UDDS) US06 to simulate a vehicle driving mode, and transformed the vehicle velocity profile into a current history for the battery pack based on the model presented in Ref. [31]. However, according to the specification of the batteries used, the maximum discharge C-rate was limited to 2C. After four US06 tests, each battery pack underwent a full charge to complete a single cycle. This procedure was repeated 20 times at 5 and −10 °C.

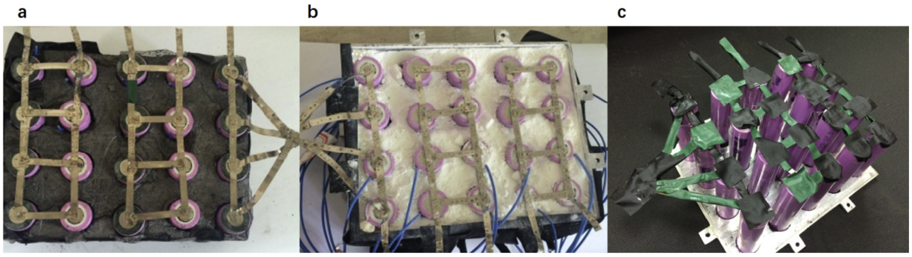

Fig. 1. Photographs of the Li-ion battery packs with different PCMs: (a) the 60 wt% RT44HC/EG composite; (b) the 60 wt% RT44HC/fumed silica composite; (c) no PCM.