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Measurements of heat generation in prismatic Li-ion batteries

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HIGHLIGHTS

- Developed a calorimeter for heat generation measurement of prismatic batteries.
- Measured battery heat generation from −10 °C to 40 °C and 0.25C–3C discharge rates.
- Observed endothermic heat flow at low discharge rates and 30 °C–40 °C temperatures.
- Observed non-negligible heat of mixing at discharge rates as low as 0.25C.
- Observed a double plateau in battery discharge curve for 30 °C–40 °C temperatures.

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ABSTRACT

An accurate understanding of the characteristics of battery heat generation is essential to the development and success of thermal management systems for electric vehicles. In this study, a calorimeter capable of measuring the heat generation rates of a prismatic battery is developed and verified by using a controllable electric heater. The heat generation rates of a prismatic A123 $LiFePO_4$ battery is measured for discharge rates ranging from 0.25C to 3C and operating temperature ranging from −10 °C to 40 °C. At low rates of discharge the heat generation is not significant, even becoming endothermic at the battery operating temperatures of 30 °C and 40 °C. Heat of mixing is observed to be a non-negligible component of total heat generation at discharge rates as low as 0.25C for all tested battery operating temperatures. A double plateau in battery discharge curve is observed for operating temperatures of 30 °C and 40 °C. The developed experimental facility can be used for the characterization of heat generation for any prismatic battery, regardless of chemistries.

1. Introduction

Rechargeable Lithium ion (Li-ion) batteries are commonly used as an energy storage medium for electric and hybrid electric vehicles (EVs and HEVs) due to their superior power density (>240 Wh kg^{-1}) and non-existent memory effects [1–3]. However, low temperature performance and high temperature capacity degradation are major limitations of the technology [4–7]. Batteries need to be maintained within an optimal temperature range to minimize degradation and avoid thermal runaway [8], irregardless of the ambient temperature that the EV or HEV is operating in. The design of a thermal management system capable of this requires knowledge of Li-ion battery heat generation rates over a wide range of operating temperatures and discharge conditions.

Heat generation in Li-ion batteries includes two main components: reversible heat generation due to entropic changes in the battery, and irreversible heat generation due to ohmic losses, charge-transfer overpotentials, and mass transfer limitations [9]. High current densities at the current collectors also create additional Joule heating, particularly in large form batteries such as the prismatic type used in EVs and HEVs [10–13]. These forms of heat generation are temperature dependent and, due to the complexity of the system, must be measured experimentally for specific batteries.

Heat generation rates of Li-ion batteries have been measured using two methods: accelerated-rate calorimetry (ARC) and iso thermal heat conduction calorimetry (IHC).

The ARC method determines the heat generation rate of the battery by measuring the rise in battery temperature over time and the amount of heat expelled from the battery to the

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surroundings. Li-ion batteries have a layered internal structure, and hence it is difficult to measure its heat capacity accurately. Since the battery is an electrochemical device, changes in the battery temperature during testing could lead to inaccurate heat generation characterization [15]. Hong et al. used a commercially available accelerated rate calorimeter (ARC2000, Columbia Scientific Industries) to measure the heat generation rates of Sony type US18650 1.35 Ah cylindrical Li-ion battery at discharge rates of 0.33C, 0.5C, and 1C at 308 K, with the maximum measured heat generation rates of 1.63 WL^{-1} [16]. Al Hallaj et al. made measurements with the same experimental apparatus on 18650 Li-ion batteries from AT&T and Panasonic, and found maximum heat generation rates of 0.26 WL^{-1} for discharge rates of less than 0.1C and charge rates of 0.33C [17].

The IHC method utilizes a large heat sink in contact with the surface of the battery to keep the battery at isothermal operation during measurements. This technique confines the measurements to low battery discharge rates, since fast discharge of the battery leads to higher rates of heat generation which the heat sink cannot extract, causing a temperature gradient within the battery [1]. Kim et al. utilized a commercially available micro-calorimeter (IMC, CSC4400, Calorimetry Science Corp.) to classify the heat generation rates of Li-ion coin type (size 2016) batteries. The heat generation rate of the battery was measured using temperature sensors placed between the battery and an aluminium heat sink and for discharge rates of 0.1C, 0.2C, 0.33C, and 1C from 300 K to 330 K. The corresponding maximum heat generation rates were reported as 0.82 WL^{-1} for discharge rates between 0.1C and 0.2C, 0.97 WL^{-1} for discharge rates of 0.2C–0.5C, and 3.21 WL^{-1} for discharge rates of 0.5C–1C [18]. Kobayashi et al. measured the heat generation rates of cylindrical Sony US18650 Li-ion batteries using a Calvet-type conduction micro-calorimeter (MMC5111-U), which has an isothermal aluminium vessel in contact with the test battery, and a thermomodule was used to measure the amount of heat transfer from the battery to the heat sink. The battery was discharged at 1/50C and 1/10C at an ambient temperature of 300 K and 330 K, and a maximum heat generation rate of 0.97 WL^{-1} was measured for discharge rates between 0.1C and 0.2C [19]. Onda et al. measured the heat generation rates of small cylindrical Sony US18650 Li-ion batteries using a thermal bath as the constant temperature heat sink. The test battery was wrapped in a thin film for electrical insulation, and the temperature of the battery was recorded using a type K thermocouple. The battery was discharged at 0.1C, 0.5C, and 1C, with a corresponding maximum measured heat generation rate of 11.0 WL^{-1}, 27.5 WL^{-1} m and 84.5 WL^{-1} [20]. Bang et al. used the same equipment as Kim et al. to perform in situ heat generation rates of a $LiMn_2O_4$ coin type Li-ion cell. Measurements were performed at discharge rates of 0.1C, 0.14C, 0.33C, and 1C at battery temperatures between 300 K and 330 K. The results show a maximum heat generation rate of 0.63 WL^{-1} for discharge rates of 0.1C–0.2C, 2.65 WL^{-1} for discharge rates of 0.2C–0.5C, and 7.51 WL^{-1} for discharge rates of 0.5C–1C [21].

From the above literature review, it is clear that the previous works on the heat generation measurement of Li-ion batteries have exhibited a wide range of results, even for batteries of the same chemistry and form [1]; and they are limited to: (*i*) small sized cylindrical or coin type batteries which are not applicable for HEV and EV use; (*ii*) low rates of discharge ($\leq$1C) which are not representative of the electrical needs of the EV; (*iii*) battery operation near room temperatures, which do not reflect the wide range of operating temperatures of EVs. Therefore, the objective of the present study is to measure the heat generation rates for large prismatic type of Li-ion batteries for a wide range of discharge rates and operating temperatures, as encountered by EVs and HEVs. In this work, an experimental technique that can accurately measure the heat generation rates of prismatic Li-ion batteries is developed, and the heat generation rates of an A123 prismatic $LiFePO_4$ battery with a 20 Ah capacity for use in automotive applications is measured and verified for various discharge and operating conditions.

2. Experimental

2.1. Experimental setup

In the present study, the calorimeter is constructed by surrounding the battery with a material of known thermal properties (hereafter referred to as calorimeter material) such that a temperature profile within the material can be deduced for any battery heat generation rate. Prismatic batteries have a high surface area to thickness ratio, which promotes heat transfer from its front and back faces. Hence, the calorimeter is in the form of two identical slabs attached to the front and back faces of the test battery. The heat generated by the battery is conducted through the slabs into a constant temperature heat sink. Measured temperature change within the calorimeter material due to the heat generated by the battery can be used to infer the unknown heat generation rate of the battery. For the calorimeter design process, heat generated by the battery is estimated to be in the range of 10 W, and simulations are performed to estimate the temperature rise in the different calorimeter materials of various thickness due to the heat generation rate. High density polyethylene (HDPE) is selected as the calorimeter material due to its stable thermal properties at the planned test temperatures and its high temperature rise due to the estimated heat generation rate of the battery.

An exploded view of the calorimeter is shown in Fig. 1. The Li-ion battery is placed between two HDPE slabs, both of which are five times the thickness of the battery. A coating of thermal grease is applied to the surface of the battery to minimize contact resistance between the battery and HDPE surfaces. The HDPE slabs with the battery are placed between two aluminium slabs. This prevents deformation of the softer HDPE material during assembly to ensure good surface contact between the battery face and the HDPE. The calorimeter is bolted together in eight locations, tightened in a criss-cross pattern to ensure even tightening, and is immersed in a constant temperature bath (ThermoFisher A25B, accuracy of $\pm$0.1 °C). The working fluid within the thermal bath is a 50-50 mixture of water-ethylene glycol, which allows measurements at sub-zero temperatures. Two high accuracy thermocouples (with accuracy of $\pm$0.1 °C) are embedded 4 mm away from the battery contact surface vertically into the HDPE material (one in each slab), centred on the battery. The placement of the thermocouple at the centre of the battery minimizes the edge effects of heat transfer from the HDPE to the surroundings. Two additional thermocouples are placed at the surface of the battery to monitor the battery temperature throughout testing (not shown in Fig. 1). An insulating cover is placed over the assembly such that the battery terminals are exposed to air, which minimizes heat transfer from both the bath and the top of the calorimeter to the ambient. A schematic of the experimental apparatus is shown in Fig. 2.

The heat generation rate of a commercially available $LiFePO_4$ prismatic Li-ion battery from A123 with a nominal capacity of 20 Ah is measured, although any battery (Li-ion or otherwise) of prismatic shape can be used in the calorimeter. The battery measures 23 cm $\times$ 16 cm $\times$ 0.7 cm, and is controlled via Greenlight Innovations G12-200 multichannel battery test station, which has an accuracy of $\pm$0.2 A in current source control, $\pm$0.05 V in voltage