Journal of Power Sources 252 (2014) 298–304

Contents lists available at ScienceDirect

Journal of Power Sources

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

Measurement of anisotropic thermophysical properties of cylindrical Li-ion cells

S.J. Drake [a], D.A. Wetz [b], J.K. Ostanek [c], S.P. Miller [c], J.M. Heinzel [c], A. Jain [a,*]

[a] Mechanical and Aerospace Engineering Department, University of Texas at Arlington, 500 W First St, Rm 211, Arlington, TX 76019, USA
[b] Electrical Engineering Department, University of Texas at Arlington, 416 Yates St, Arlington, TX 76019, USA
[c] Naval Surface Warfare Center, Carderock Division, U.S. Navy, 1000 Kitty Hawk Ave., Building 77L, Philadelphia, PA 19112, USA

HIGHLIGHTS

- Presents a new method for measuring thermal properties of cylindrical Li-ion cells.
- Results are in excellent agreement with finite-element simulation models.
- Measurements provide cell-level thermal conductivity and heat capacity values.
- Measurements indicate strong anisotropy in thermal conduction.
- Measurements indicate poor radial thermal conductivity.

ARTICLE INFO

Article history:
Received 16 August 2013
Received in revised form
27 November 2013
Accepted 29 November 2013
Available online 11 December 2013

Keywords:
Lithium-ion batteries
Thermal conduction anisotropy
Thermal conductivity
Heat capacity

ABSTRACT

Cylindrical Li-ion cells have demonstrated among the highest power density of all Li-ion cell types and typically employ a spiral electrode assembly. This spiral assembly is expected to cause large anisotropy in thermal conductance between the radial and axial directions due to the large number of interfaces between electrode and electrolyte layers in the radial conduction path, which are absent in the axial direction. This paper describes a novel experimental technique to measure the anisotropic thermal conductivity and heat capacity of Li-ion cells using adiabatic unsteady heating. Analytical modeling of the method is presented and is shown to agree well with finite-element simulation models. Experimental measurements indicate that radial thermal conductivity is two orders of magnitude lower than axial thermal conductivity for cylindrical 26650 and 18650 $LiFePO_4$ cells. Due to the strong influence of temperature on cell performance and behavior, accounting for this strong anisotropy is critical when modeling battery behavior and designing battery cooling systems. This work improves the understanding of thermal transport in Li-ion cells, and presents a simple method for measuring anisotropic thermal transport properties in cylindrical cells.

1. Introduction

Several recent incidents, including fire in a Li-ion battery pack aboard an aircraft [1] have highlighted the importance of thermal design of Li-ion batteries. Fundamental studies of thermal phenomena occurring within Li-ion batteries are essential for developing a basic understanding of these technological challenges, and to design cooling systems prevent thermal runaway during high power operation. It is also critical to measure and understand the fundamental thermal transport properties of a Li-ion cell for accurate system-level modeling and design. Due to the inherently high energy content in Li-ion batteries, thermal phenomena play an important role in performance as well as safe battery operation, and in most cases, contribute to operating limits [2–5]. While electrical and electrochemical phenomena in a Li-ion cell have been widely studied [3,6–10], relatively lesser literature exists on thermal transport in a Li-ion cell. Operation at higher temperatures is known to decompose the electrolyte and thus degrade cell performance and lifetime. If the heat exceeds a critical threshold, which varies upon chemistry type, electrolyte release and fire, and ultimately a cathode thermal runaway situation may arise [11]. Capacity and power reduction has also been found to occur at high operating temperature [12], although lowered impedance and thus increased voltage may offer better output at slightly elevated temperatures.

* Corresponding author. Tel.: +1 817 272 9338; fax: +1 817 272 2952.
E-mail address: jaina@uta.edu (A. Jain).

0378-7753/$ – see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.jpowsour.2013.11.107

Fig. 1 shows a top view cutout of a typical Li-ion cylindrical cell. A composite layered material made up of an anode, cathode, separator and current collectors is soaked in electrolyte and rolled in a Swiss roll fashion. Electrode tabs are present at the ends of the cell to collect and conduct electric current to the external cell terminals.

Due to this construction, the nature of radial thermal transport is expected to be significantly different from axial thermal transport. While in the radial direction, heat flow is expected to be impeded by several thermal contact resistances between the thin layers of various, in the axial direction, thermal conduction is expected to occur primarily along the current collector materials, which often run continuously in the axial direction. Thermal contacts between materials in microsystems often present significant thermal resistance, sometimes even greater than the material thermal resistance itself [13]. Due to this expected anisotropy of thermal transport, it is important to experimentally measure the radial and axial thermal conductivities of a cylindrical Li-ion cell, since the assumption of isotropic thermal transport properties in Li-ion cells design will either underpredict or overpredict the temperature field, both of which are undesirable.

Several papers have reported computational and experimental investigation of temperature distribution at the cell-level and at the pack-level [2,3,5,14–17]. Accurate information about underlying thermal properties such as thermal conductivity and heat capacity is critical for the accuracy of such models. However, only a limited set of papers have reported work on thermal property measurement of Li-ion cells. Bandhauer, *et al.* provide a review of thermal phenomena associated with Li-ion cells and also note the lack of thermal property measurement data [18]. Heat capacity measurements of a few batteries using calorimetry have been reported [19]. However, this method does not provide thermal conductivity measurements. A few papers have reported core-to-outside lumped thermal resistance of a cell [14,20], but such a parameter is not particularly useful since not all the heat is generated at the core of a Li-ion cell. Due to the distributed nature of heat generation within a Li-ion cell, it is more appropriate to model the cell using effective thermal conductivity and heat capacity. Maleki, *et al.* have reported measurements of thermal conductivity and heat capacity of Li-ion cells using xenon flash technology (XFT) and steady-state measurements [21]. However, this approach requires two different experiments for measuring thermal conductivity and heat capacity, and more importantly, does not provide thermal conductivity of the cell assembly as a whole. The XFT method is also cumbersome and expensive, and is usually not readily available. Most of the previous work neglects the anisotropy of thermal conduction within a Li-ion cell. It is desirable to experimentally measure anisotropic components of thermal transport properties in a Li-ion cell. Such work will enhance the accuracy of thermal computation for system-level thermal design, and will also help in developing a fundamental understanding of heat flow within a Li-ion cell.

Fig. 1. Top view cutout of a typical Li-ion cylindrical cell.

This paper describes a new method for measurement of effective heat capacity and anisotropic thermal conductivity of cylindrical Li-ion cells. The method described here is relatively simple, and provides rapid measurement of axial and radial thermal conductivity in addition to heat capacity. The method utilizes the thermal response of the cell to axial or radial heating in an adiabatic configuration. An analytical heat transfer model is developed for modeling temperature distribution during such adiabatic heating. It is shown that simultaneous measurement of thermal conductivity and heat capacity can be obtained in a single experiment. Experimental data are in excellent agreement with the analytical model and indicate strong anisotropy in thermal conduction in 26650 and 18650 cells. Results indicate a significant underprediction of peak temperature in using only the axial thermal conductivity compared to a case where the measured anisotropy is accounted for. Results presented in this manuscript will enable more accurate performance models of Li-ion batteries and better designs for battery cooling systems.

2. Experimental method

An experimental technique for determining the heat capacity and anisotropic thermal conductivity of a cell is developed in this work. The technique yields thermal conductivity and heat capacity in a single experiment. In order to do so, the cell is subjected to adiabatic heating in the radial or axial direction, and its temperature rise is measured as a function of time. A flexible Kapton heater is attached to either the curved or top circular surface of the cell, to heat either in the radial or axial direction, respectively. T-type thermocouples are attached at various locations along the cell outer wall to measure temperature. When making radial measurements, thermocouples are attached on the outside surface at mid-cell height. For axial measurements, thermocouples are attached in the center at the two circular ends of the cell. Fiberglass insulation tape is wrapped around the cell to minimize heat loss. All experiments are carried out in a vacuum chamber at −75 kPa (gage) vacuum to further reduce heat loss. The cells are electrically characterized by performing electrochemical impedance spectroscopy (EIS) using a potentiostat, which determines the individual charge and discharge profile of each cell. Using this information, the cells are fully charged prior to testing to avoid variations in lithiation state of the electrode materials and degeneracy in lithium distribution, which might otherwise influence thermal properties. The cell under test is suspended on thin paper arms to minimize thermal conduction loss through surface area contact. To characterize the amount of heat loss through the insulation, one thermocouple is placed outside the insulating tape, and another is suspended in vacuum a small distance from the heater. Fig. 2(a) and (b) shows images of the cell with the flexible heater and thermocouples in place inside the vacuum chamber. A Keithley 2612A sourcemeter is used for supplying heating current and measuring voltage. The thermocouple output is sampled at 2 Hz using a National Instruments (NI) 9213 24-bit thermocouple module within an NI cDAQ-9171. Data acquisition is controlled using NI LabVIEW software. Electrical resistance of Kapton heaters used in this work is measured at temperatures between 25 °C and 50 °C. There is less than 0.1% change in resistance, which shows that the temperature coefficient of resistivity is negligible. This ensures constant heat flux into the battery even as the temperature rises. Each