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Measurement of in-plane thermal conductivity and heat capacity of separator in Li-ion cells using a transient DC heating method

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HIGHLIGHTS

- Presents measurement of in-plane thermal conductivity and heat capacity of separator.
- Experimental data are in excellent agreement with analytical model.
- Measurements indicate very low thermal conductivity of the separator.
- Measurements indicate weak temperature dependence of thermal properties.
- Measurements presented here may facilitate high performance and safety.

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ABSTRACT

The separator is a critical, multi-functional component of a Li-ion cell that plays a key role in performance and safety during energy conversion and storage processes. Heat flow through the separator is important for minimizing cell temperature and avoiding thermal runaway. Despite the critical nature of thermal conduction through the separator, very little research has been reported on understanding and measuring the thermal conductivity and heat capacity of the separator. This paper presents first-ever measurements of thermal conductivity and heat capacity of the separator material. These measurements are based on thermal response to an imposed DC heating within a time period during which an assumption of a thermally semi-infinite domain is valid. Experimental data are in excellent agreement with the analytical model. Comparison between the two results in measurement of the in-plane thermal conductivity and heat capacity of the separator. Results indicate very low thermal conductivity of the separator. Measurements at an elevated temperature indicate that thermal conductivity and heat capacity do not change much with increasing temperature. Experimental measurements of previously unavailable thermal properties reported here may facilitate a better fundamental understanding of thermal transport in a Li-ion cell, and enhanced safety due to more accurate thermal prediction.

1. Introduction

Thin flexible substrates are frequently used in engineering applications such as flexible electronics [1,2], separators for Li-ion cells [3–5], organic semiconductors [6], flexible displays [7–9], etc. Compared to a thick rigid substrate, a flexible substrate offers reduced weight, increased design flexibility, bendability, etc. [10]. In addition, thin substrates also often provide valuable functionality, such as controlled ionic conductance through thin separators in Li-ion cells [3]. The thin, flexible nature of the separator in a Li-ion cell also makes it possible to roll the electrode-separator assembly and compactly package it inside a high energy density cell [3]. Fig. 1 shows an image of the electrode-separator roll from a Li-ion cell, and a schematic of various layers in the assembly. The positive electrode is typically made of a transition metal oxide, whereas the negative electrode is typically graphite. Transfer of Li ions from one electrode to another enables charging or discharging of the cell. The two electrodes are typically separated by the separator, which is typically based on an electrically insulating porous material and is about a few tens of microns thick. The separator material plays a multi-functional role [5,11].

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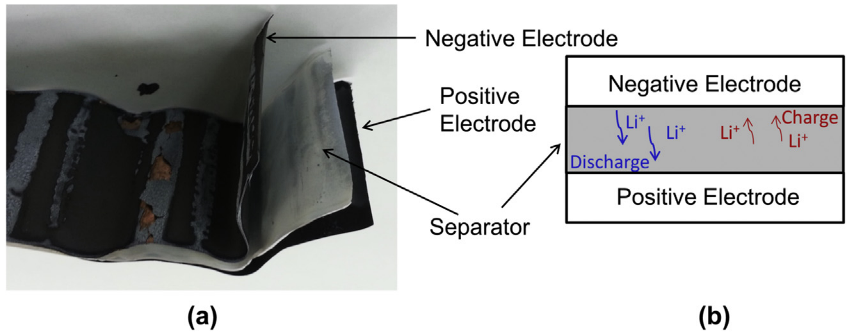

Fig. 1. (a) Image of the electrode-separator roll in a Li-ion cell; (b) Schematic of various layers in the assembly.

The primary role of the separator is to provide a pathway for Li ions to migrate from one electrode to the other while blocking electron transport [12]. The separator also provides mechanical strength without deterioration at high temperature [13,14] and contributes to conductance of heat generated inside the cell. A number of studies have been carried out for understanding and quantifying ionic conductance through the separator material [5,11]. A more limited number of studies have investigated mechanical stresses in the separator that may occur during cell operation [13–15]. Despite the importance of thermal transport within the cell, however, there is a lack of literature on measurement, modeling and optimization of thermal transport properties of the separator. Heat generated throughout the cell must conduct through the electrode roll to the outer surface, where it is eventually dissipated to the surroundings. This makes it important to understand the nature of thermal conduction with the cell [16], and rate-limiting steps that determine the effectiveness of this process [17,18]. Among all materials in the electrode roll including positive electrode ($LiFePO_4$, $LiCoO_2$, etc.), negative electrode (graphite), current collectors (metal) and separator, the separator is expected to have the lowest thermal conductivity, and hence must be investigated in detail. In addition, thermal contact resistances between the separator and electrodes may also be important. A number of papers have addressed thermal modeling within a Li-ion cell [19–22]. The accuracy of temperature fields predicted by such models depends critically on the accuracy of underlying thermal properties of constituent materials. There is a lack of experimental data on thermal properties of the separator, and most past work on thermal modeling [19–22] appears to use assumed values for thermal properties of the separator. Given the importance of accurate temperature prediction on battery safety, it is clearly very desirable to experimentally measure these properties. Such a measurement will contribute towards the thermal engineering, and hence operational safety of Li-ion cells.

The fundamental governing energy equation, the solution of which determines the temperature distribution within a thermal system is given by Ref. [23]

$$k\nabla^2 T + Q''' = \rho C_p \frac{\partial T}{\partial t} \tag{1}$$

where T is the temperature field and Q''' is volumetric heat generation rate. The two fundamental thermophysical properties that appear in this equation, and that play a key role in determining the nature of thermal transport through the separator material are its thermal conductivity, k (W m^{-1} K^{-1}) and specific heat capacity, C_p (J kg^{-1} K^{-1}) [24]. While k determines the rate of thermal conduction through the separator, C_p characterizes the extent of heat storage within the material. The quantity $k/\rho C_p$ is often referred to as the thermal diffusivity, α (m^2 s^{-1}). Note that equation (1) assumes that k is an isotropic property, although in some materials, k may be different in different directions [24].

A number of experimental techniques have been reported in the past for measurement of thermophysical properties of substrates [25–29]. In general, the temperature rise in the material of interest in response to a known heat flux is measured and compared with a theoretical model to determine k and C_p. Heat flux is imposed by either Joule heating due to an electric current, or optically through a laser. Methods based on constant, time-varying and periodic heat flux have been used in the past [25]. Two separate experiments are often required to measure both, although two measurements within the same experiment have also been used [27]. A vast amount of literature exists on the measurement of thermophysical properties of thick, rigid substrates, typically a few mm or thicker [25]. In addition, thin films, a few μm or thinner, deposited on a thick, rigid substrate have also been thermally characterized [30]. On the other hand, not much research has been reported on measurement of thermophysical properties of thin, flexible substrates. Neither of the approaches outlined above for rigid substrates or thin films will work for a substrate that is a few tens of μm, such as a typical Li-ion separator. This necessitates a new approach for thermophysical property measurement. A typical separator in a Li-ion cell is a few tens of μm thick [11,15], which presents a challenge in measurement of thermophysical properties.

An additional challenge in the measurement of thermophysical properties of a flexible substrate is in the microfabrication of heater and sensor elements. While microfabrication is carried out commonly on rigid substrates such as Silicon wafers and glass slides [31], fabrication of metal features on a thin flexible substrate is not as well developed. The mechanical stiffness of a typical separator of a Li-ion cell is even less than that of typical substrates used for flexible electronics [1,2].

This paper presents a novel experimental method for measurement of in-plane thermal conductivity and heat capacity of a Li-ion cell separator. The method is based on measurement of temperature rise in two parallel metal lines during a short time following DC heating in one of the lines. This method is capable of measurement on substrates for which experimental methods for neither thick rigid substrates, nor thin films are applicable. Measurements are in excellent agreement with an analytical model based on the assumption of a semi-infinite domain. Both in-plane thermal conductivity and heat capacity are measured at room temperature and at an elevated temperature. Results from this work are expected to contribute towards better thermal understanding and safety of Li-ion cells. Analytical modeling and experimental method are presented in next two sections, followed by a discussion of results.