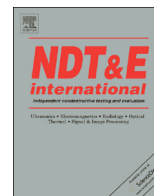




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Lithium-ion battery electrode inspection using pulse thermography

Nathan Sharp^{a,*}, Peter O'Regan^b, Douglas Adams^a, James Caruthers^b, Anand David^b, Mark Suchomel^b

^a School of Mechanical Engineering, Purdue University, West Lafayette, IN, 47906, USA

^b School of Chemical Engineering, Purdue University, West Lafayette, IN, 47906, USA

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ABSTRACT

Pulse thermography was used to experimentally evaluate lithium-ion battery electrode quality. The camera data was processed to improve high frequency capabilities. Lab manufactured electrodes with gross defects, thickness variations, and composition variations were all detectable with this method. A thickness difference of 4 μm (4 percent of total) was detectable with the method. Lab electrodes were compared with commercial electrodes with comparable results. Both types of electrodes showed a significant thickness oscillation that has not previously been reported in lithium-ion battery electrodes. The results suggest that pulse thermography can be used as an effective tool for online quality control.

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1. Introduction

The strong push towards the increased use of renewable energy drives the need for energy storage that is inexpensive, light, and durable. Rechargeable lithium ion batteries have become very popular because they have a high energy density but are currently being held back by their high price [1,2]. They operate via an ion-shuttling mechanism: an applied load causes lithium ions to migrate between the negative electrode (anode) and positive electrode (cathode). The electrode materials permit lithium ion intercalation; the distribution of lithium ions between the electrodes determines the cell potential. Lithium ion batteries are constructed by assembling several layers of anode–cathode pairs to achieve the desired capacity.

The amount of charge that can be stored in an electrode surface is determined by its material properties and dimensions. If these properties are homogeneous along the direction of the electrode surface then the electrochemical potential from the lithium ions will be constant between the electrode surfaces and the only gradient in potential will occur in the direction that is normal to the surface. However, if there are areas on the electrode that vary in thickness or composition, potential and current gradients will develop parallel to the electrode surface. These undesired potential gradients can create regions that have a voltage potential that is higher than the overall voltage of the battery. If the voltage exceeds a critical value then undesirable and irreversible reactions can occur which eventually can cause battery failure. Electrode

surfaces degrade through normal use regardless of the initial quality of those surfaces. However, the greater the inhomogeneity that is introduced into the electrode surface during manufacturing, the more likely it is that the battery will actually fail before its design life is reached [3]. Also, failure on only a small part of one electrode surface can cause failure of the entire battery if it generates an internal short circuit. Because there are so many anode–cathode layers stacked in each battery cell, even a very small percentage of defects can adversely affect battery life.

2. Materials and methods

Sample anodes and cathodes were used in the experimental thermography testing. It was desirable to use electrodes that were as similar as possible to electrodes used in commercial batteries, so the primary goal in the selection of electrode materials was to imitate commercial electrodes. In order to perform the necessary experiments, it was required to manufacture the electrodes for testing instead of purchasing them commercially.

2.1. Material choice, composition, dimensions

Lithium ion electrodes are typically made up of three components: an active material, which allows lithium ions to intercalate and forms an electrochemical potential; carbon black, which improves the electrical conductivity of the electrode; a binder (polyvinylidene fluoride, or PVDF), which helps keep the electrode from becoming brittle and cracking. PVDF is a useful binder for battery electrodes because it is inert and inexpensive. The exact

* Corresponding author. Tel.: +1 602 573 8734.

E-mail address: nsharp@purdue.edu (N. Sharp).

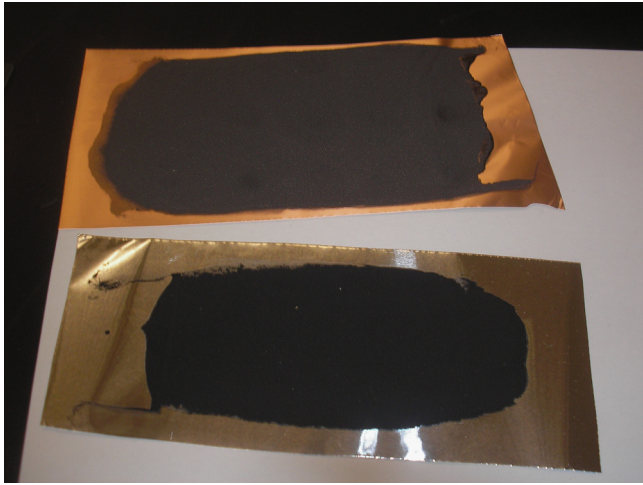


Fig. 1. Samples of finished electrodes.

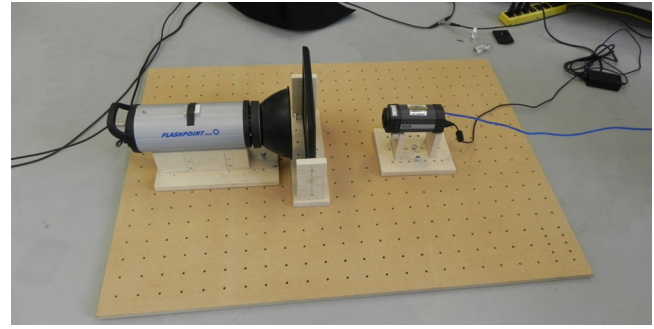


Fig. 2. Test setup.

composition of electrodes in commercial batteries varies by manufacturer and is generally kept as a trade secret, but research on the subject is widely available [4–6]. Electrode compositions and percentages were chosen based on the literature. Lithium cobalt oxide (LiCoO_2) was used for the active cathode material and graphite was used for the active anode material. Their compositions by weight were 94 percent active material, 3 percent carbon black, and 3 percent binder.

A slurry for coating the electrode materials was formed by combining the above compounds with enough *n*-methyl-2-pyrrolidinone (NMP) to create a proper viscous flow [7,8]. The ratio depended on the percentage of active material; for 94 percent active material the ratio was 1.365 g NMP for 1 g solids (more NMP is needed if the percent active material goes down). Using a Gardco doctor blade, the anode and cathode slurries were applied with a 100 μm thickness to 9 μm copper and 13 μm aluminum substrates, respectively. The NMP evaporates during the drying process, leaving porous films with thicknesses of roughly 60 percent of their coated amount.

All sample electrodes tested were manufactured at Purdue University unless otherwise noted. Fig. 1 shows a finished cathode and anode film such as the ones used in the pulse thermography testing. The electrodes were cut into squares with lengths between 50 and 150 mm.

A FLIR Systems A325 infrared camera was used to measure the infrared radiation from the specimens at 60 frames per second with a 320 by 240 pixel resolution while a Flashpoint 2420 A flash lamp with a xenon bulb was used to provide a pulse of heat to the current collector surface. The pulse provides a short burst of light (roughly 1 ms) and infrared radiation which quickly heats the copper or aluminum current collector. The camera uses microbolometers to measure the amount of infrared radiation emitted from the electrode surface in the spectral range of 7.5–13 μm . The flash bulb, electrode, and camera were mounted onto a peg board, shown in Fig. 2, to facilitate consistency in the experimental test setup.

3. Theory and calculation

The spectral hemispherical emissive power, I^b , which is emitted by a black body outside its surface, can be modeled using Planck's law, shown in Eq. (1), where C_1 and C_2 are universal constants, λ is the wavelength, and T is the temperature in Kelvin.

$$I^b(\lambda) = \frac{C_1}{\lambda^5 (e^{C_2/\lambda T} - 1)} \quad (1)$$

Since few real objects behave like a black body, a new term called the spectral emissivity coefficient, defined in Eq. (2), is introduced. Using the spectral emissivity coefficient, Eq. (1) can be rewritten for real bodies by simply multiplying by ε , shown in Eq. (3).

$$\varepsilon(\lambda) = I(\lambda)/I^b(\lambda) \quad (2)$$

$$I(\lambda) = \varepsilon(\lambda) \frac{C_1}{\lambda^5 (e^{C_2/\lambda T} - 1)} \quad (3)$$

For the purpose of this research, the wavelength dependence of the emissivity constant will be neglected and all emissivity values reported will be a single value which acts as an effective emissivity constant. Prior to purchase of the camera, FLIR performed a calibration of the camera to convert the intensity values read in by the microbolometers into temperature measurements.

3.1. Temperature calibration

Manual calibrations were performed to check the electrode emissivity and ensure that the camera gave readings which were linear with temperature. Camera measurements were recorded for five steady state surface temperatures. The measurements were averaged across several pixels and several frames to reduce noise effects.

Electrodes were made with three different compositions to observe the effects of a difference in composition on the emissivity of the material. The active material was varied between 88 percent and 96 percent, a much more extreme variation than would be expected in a real electrode, to develop an understanding of the sensitivity of the emissivity to the composition. Fig. 3 shows the camera reading as a function of thermocouple temperature for three different compositions and indicates that the emissivity is essentially identical for all three composition cases. Based on these results, the emissivity was assumed to be homogeneous for all tests. The readings also confirmed that the camera readings were linear with temperature.

3.2. Signal processing

While the camera readings are based on radiation from the surface of the electrode, the heat transfer inside the electrode is dominated by heat conduction, which is modeled by the heat conduction equation, shown in Eq. (4). If heat is transmitted onto one surface of a system, the time it takes for the opposite surface to reach 1/2 the equilibrium temperature can be solved from Eq. (4) as a function of the thickness of the system, L , and the thermal diffusivity, α . The equation for the $t_{1/2}$ rise time is shown in Eq. (5). Chen et al. [7] reported the thermal diffusivity of a LiCo_2 electrode is $5.35e-3 \text{ cm}^2/\text{s}$. By entering the thickness of the electrode (100 μm) and the thermal diffusivity into Eq. (5), the half rise time can be estimated at approximately 2.8 ms.

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