Journal of Power Sources 364 (2017) 432–436

Contents lists available at ScienceDirect

Journal of Power Sources

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

Short communication

Fragmentation of copper current collectors in Li-ion batteries during spherical indentation

Hsin Wang*, Thomas R. Watkins, Srdjan Simunovic, Philip R. Bingham, Srikanth Allu, John A. Turner

Oak Ridge National Laboratory, Oak Ridge, TN, USA

HIGHLIGHTS

- Copper current collectors fractured during spherical indentation.
- The fragmentation was hidden and detectable by optical and electron microscopy.
- X-ray computed tomography (XCT) showed the hidden fractures of current collectors.

ARTICLE INFO

Article history:
Received 17 July 2017
Received in revised form
14 August 2017
Accepted 17 August 2017

Keywords:
X-ray tomography
Li-ion battery
Spherical indentation
Internal short circuit
Fragmentation

ABSTRACT

Large, areal, brittle fracture of copper current collector foils has been observed by 3D x-ray computed tomography (XCT) of a spherically indented Li-ion cell. This fracture is hidden and non-catastrophic to a degree because the graphite layers deform plastically, and hold the materials together so that the cracks in the foils cannot be seen under optical and electron microscopy. The cracking of copper foils could not be immediately confirmed when the cell is opened for post-mortem examination. However, 3D XCT on the indented cell reveals "mud cracks" within the copper layer and an X-ray radiograph on a single foil of the Cu anode shows clearly that the copper foil has broken into multiple pieces. This failure mode of anodes in Li-ion cell has very important implications on the behavior of Li-ion cells under mechanical abuse conditions. The fragmentation of current collectors in the anode must be taken into consideration for the electrochemical responses which may lead to capacity loss and affect thermal runaway behavior of the cells.

1. Introduction

Mechanical abuse and subsequent electrical failure of Li-ion batteries have become a research focus recently [1–4]. This is largely due to the increased deployment of electrical vehicles (EV) worldwide since 2011 [5]. Typically, 300–500 large-format Li-ion cells (capacity > 15 Ah each) are used in a battery pack of electric vehicles, which must pass the same crashworthiness tests as traditional vehicles with internal combustion engines. Although the battery protection from mechanical damage is an integral part of EV design, deformation of the cells can still occur in vehicle accidents, so that an understanding of how each battery component responds under mechanical deformation is important. It has also been observed that materials behave differently in planar and cylindrical jellyroll configurations [6]. Considering that the cells are filled with electrolyte that influences the mechanical response, testing mechanical responses of full-sized Li-ion batteries is more relevant than testing dry components or stacked layers [7]. In typical abuse tests, the cells are charged and mechanically deformed until the onset of internal electrical short circuit. In some cases, thermal runaway is induced, and the cells are consumed by fire that leaves nothing to exam. In other cases, even though the cells survive the short circuit event, the localized heating melts the separators and eliminates any details that could provide important from post-mortem inspection. In order to understand the materials responses in a cell under mechanical deformation, we have carried out studies using progressive indentation [8]. The collective movement of the layers and materials experiencing deformation are used to establish mechanical and electrochemical models of the batteries and modules.

* Corresponding author.
E-mail address: wangh2@ornl.gov (H. Wang).

http://dx.doi.org/10.1016/j.jpowsour.2017.08.068

Our on-going study is focused on the mechanical response of materials at the on-set of the internal short circuit which can be detected by a sudden open-circuit voltage drop. In order to preserve the materials after the internal short circuit event, the cells were discharged to <10% stage of charge (SOC). Even without much charge, the cell voltage often bounced back above 3.2 V during an internal short. The current during such a short circuit is small so that no significant heating occurs to damage the separator and neighboring materials. However, a typical post-mortem exam requires opening of the cell and peeling apart the layers, which undoubtedly changes/disturbs the critical damage site(s). In this study, we employed 3D XCT technique to probe the internal structure of the battery non-destructively. During the study we discovered, for the first time, how the anodes, especially the copper current collectors, fail under indentation. The observed fragmentation of copper foils was not expected, and more importantly, it could not be seen by routine optical and electron microscopy. This hidden foil fragmentation mode occurs in a layered structure and full-sized cells filled with an electrolyte. In this paper, we focus on the discovery of hidden fragmentation of the current collectors. In addition to 3D x-ray tomography, optical microcopy and scanning electron microscopy (SEM), x-ray radiography and post-mortem exam were carried out to confirm this observation.

2. Experimental

Commercially available lithium cobalt oxide (LCO) cells were used. The cells have a jellyroll configuration within an aluminum enclosure with dimensions of 30 mm × 40 mm x 4.5 mm. About 20 initial conditioning cycles were carried out to verify each cell's capacity and performance. The cycling used 0.5 A constant current (about 0.7C) discharge to 3.0 V follow by 0.5 A constant current charging to 4.1 V. The cell was held at 4.1 V constant voltage until current was less than 0.05 A. The cell capacity was stable at 700 mAh after 3–4 cycles. Mechanical indentation was carried out on the cells after the final discharge. The cell was put under a servo-motor driven mechanical load frame and a 12.7 mm diameter spherical stainless steel indenter was used to compress the cell in a direction perpendicular to the flat sides, until a 0.1 V was detected, which was indicative of the occurrence of an internal short circuit. The loading speed was 0.25 mm per minute and the maximum compressive load was about 4 kN. At that point, the load was released. The indented cell with a short-circuit was examined with 3D x-ray tomography. The tomography data was collected with a Zeiss Versa 520 X-ray Computed Tomography (XCT) system using a tungsten source running at 140 kV/64.4 μA max (9 W). Two magnifications were used, 0.4X and 4X. The data is collected and reconstructed with Zeiss' Scout and Scan software [9] with further analysis conducted on TXM3DViewer [10]. The full cell was sliced into three pieces after the initial tomography scan. The middle slice contained the indentation, and x-ray tomography was repeated to confirm no artifacts were caused by extra cell materials. The middle slice was then completely disassembled and the jellyroll was opened to expose each layer. A single copper anode layer with a small puncture was scanned by x-ray radiography and examined using a Hitachi model 3400 scanning electron microscope.

3. Results and discussion

The indented cell was examined by 3D XCT after an initial short

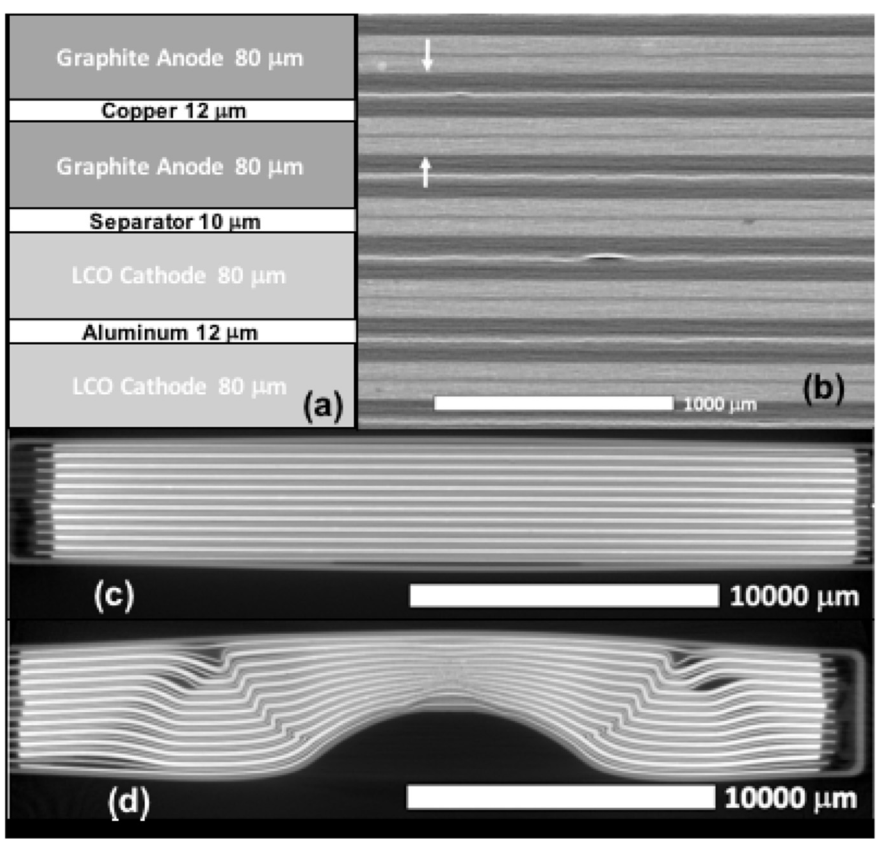

Fig. 1. (a) Schematic of measured thicknesses of layers in a jellyroll, (b) x-ray tomography of a cross section of the jellyroll, (c) x-ray tomography scan of an un-deformed cell and (d) x-ray tomography scan of an indented cell.