

Identifying degradation mechanisms in lithium-ion batteries with coating defects at the cathode[☆]



Lamuel David^a, Rose E. Ruth^a, Debasish Mohanty^a, Harry M. Meyer III^b, Yangping Sheng^a, Sergiy Kalnaus^c, Claus Daniel^{a,d}, David L. Wood III^{a,d,*}

^a Energy and Transportation Science Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831-6083, USA

^b Material Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831-6083, USA

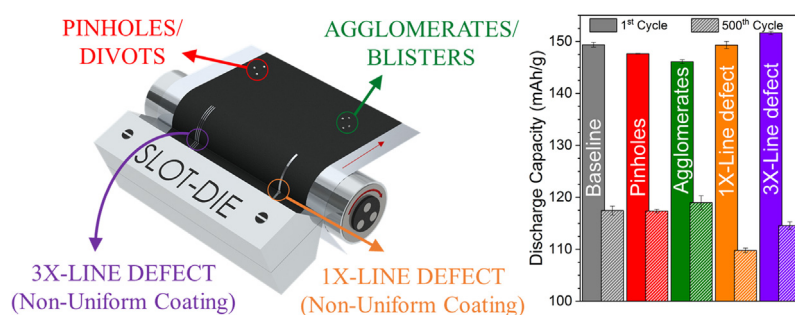
^c Computational Sciences and Engineering Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831-6083, USA

^d Bredeben Center for Interdisciplinary Research and Graduate Education, University of Tennessee, Knoxville, TN 37996, USA

HIGHLIGHTS

- The impact of electrode coating defects on cell performance is evaluated.
- Interactions between cathode and anode cause degradation in cells with defects.
- Computational analysis validates findings from chemical characterization.
- Electrodes with certain coating defects can be repurposed for less demanding uses.
- Repurposing electrodes will reduce scrap rates and lower manufacturing cost.

GRAPHICAL ABSTRACT



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ABSTRACT

Understanding the effect of electrode manufacturing defects on lithium-ion battery (LIB) performance is key to reducing the scrap rate and cost during cell manufacturing. In this regard, it is necessary to quantify the impact of various defects that are generated during the electrode coating process. To this end, we have tested large-format 0.5 Ah LiNi_{0.5}Mn_{0.3}Co_{0.2}O₂/graphite pouch cells with defects intentionally introduced into the cathode coating. Different types of coating defects were tested including agglomerates, pinholes, and non-uniform coating. Electrodes with larger non-coated surface had greater capacity fade than baseline electrodes, while pinholes and agglomerates did not affect performance adversely. Post cycle analysis of electrodes showed that the anode facing the defective region in the cathode was clearly impacted by the defect. Further characterization using Raman spectroscopy, X-ray photoelectron spectroscopy, and X-ray diffraction provided evidence for a proposed mechanism for material degradation related to the most detrimental type of coating defect.

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* Corresponding author at: 2370 Cherahala Boulevard, Knoxville, TN 37932-6479, USA.

E-mail address: wooddl@ornl.gov (D.L. Wood).

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1. Introduction

Lithium-ion batteries (LIBs) have been successfully commercialized in portable electronic devices [1,2]. However, batteries for electric vehicles (EVs), hybrid electric vehicles (HEVs), and plug-in hybrid electric vehicles (PHEVs) [3] have stricter requirements like lighter weight, longer range, improved safety, and longer cycle life [4]. EV batteries need to be cost-effective as well. The latest EV cell cost target from the US Department of Energy (DOE) is \$80/kWh with a useable energy density of 750 Wh/L and a peak power density of 1500 W/L by 2020. This is a very aggressive target, since the current cells for electric vehicles cost around \$245/kWh and have an energy density around 285 Wh/L [5]. The cost of expensive metals like cobalt, nickel, and lithium is a major component of the final price of LIBs [6]. On this subject, there is growing research aimed towards lowering the cost of materials in LIBs [7]. However, any change in the battery materials has to undergo a rigorous testing phase to address strict safety requirements and to exhibit equivalent or improved battery performance [8]. Therefore, new material technologies require long periods for implementation. To tackle the immediate need for cost reduction, there is a growing demand to reduce the manufacturing costs to offset high material costs [9].

There are several methods to reduce electrode manufacturing cost through advanced material processing and material handling technologies. These include adapting cheaper water-based solvents, developing solvent-free coating [10], or implementing spray/electrostatic coating methods [11]. However, changing processing technologies at an existing battery manufacturing plant requires a high capital cost. Another method to reduce cost without large capital investment is to improve quality control practices to reduce scrap rates. The cost of raw materials in the electrode is high, and coating defects are one of the primary sources of waste in battery manufacturing. The current quality control process involves discarding defective coatings regardless of the type of defect and using only ideal electrode coatings, potentially contributing to excessive waste. The quality of the electrode coating depends on uniform thickness, porosity [12], material distribution (areal weight), and adhesion to the current collector [13]. Any inhomogeneity of these properties results in defects [14] that lead to local aging of the electrode with loss in capacity and cycle life [15].

In this study, we evaluate the effect of electrode inhomogeneities on the electrochemical behavior of lithium-ion batteries. We analyzed the electrochemical properties of three types of coating defects in cathodes: (a) pinholes, (b) agglomerates, and (c) line defects. In our previous study, we used coin cells for analysis which showed that defects in electrode coating significantly impacted electrochemical performance [16]. However, the study was influenced by cell-to-cell variations, and the fraction of defective area relative to the total electrode area was large, which exaggerated the influence of the defect. To overcome these limitations, we continued our studies using large-format multilayer pouch cells, which closely resembled industrial battery manufacturing conditions. To our knowledge this is the first study to evaluate the impact of different types of coating defects on the electrochemical performance of lithium-ion batteries under conditions that replicate state-of-the-art electrode coating and cell manufacturing. Cell testing was performed using a rigorous protocol to accelerate the degradation process. Further, chemical characterization using Raman spectroscopy, X-ray photoelectron spectroscopy (XPS), and X-ray diffraction (XRD) was performed on the harvested electrodes to determine the mechanism of degradation and the extent to which it influences electrochemical properties. As in our previous study [16], the electrode defects were specifically generated in a controlled manner to standardize the experiment. By better understanding how cells with coating defects behave, we are able to determine if these coatings are suitable for other applications. For example, electrodes with some defects could possibly be used in low-power or low-energy applications like grid-storage or backup power storage devices instead of becoming waste.

2. Experimentation and methods

2.1. Materials and electrode fabrication

The electrodes were fabricated at the U.S. Department of Energy (DOE) Battery Manufacturing R&D Facility at Oak Ridge National Laboratory. Anode and cathode slurries were prepared by dispersing the active material, binder, and conductive additives in *N*-Methyl-2-pyrrolidone with a planetary mixer (Ross PDM-1/2). The cathode consisted of $\text{LiNi}_{0.5}\text{Mn}_{0.3}\text{Co}_{0.2}\text{O}_2$ powder (NMC532, Toda America Inc., 90 wt%), polyvinylidene fluoride (PVDF, Solvay 5130, 5 wt%), and carbon black (Denka, 5 wt%). The anode contained natural graphite (ConocoPhillips, A12, 92 wt%), PVDF (Kureha 9300, 6 wt%), and Super P Li (Timcal, 2 wt%). The slurries were applied with a slot-die coater (Frontier Industrial Technology, Inc.) to one side of a foil current collector (15 μm thick Al for the cathode and 9 μm thick Cu for the anode). The active material loadings for the cathode and anode were 15.0 mg/cm² (2.4 mAh/cm²) and 8.0 mg/cm² (2.88 mAh/cm²), respectively, yielding a negative to positive capacity ratio (n/p ratio) of 1.2. Electrodes were calendared to 35% porosity.

2.2. Pouch cell assembly

Full pouch cells with 500 mAh capacity were built using the electrodes that were dried in a vacuum oven for 24 h. The pouch cells consisted of six anodes and six cathodes stacked alternately with a porous polymer separator (Celgard 2325). The electrolyte was a 1.2 M solution of LiPF_6 in ethylene carbonate (EC) and ethyl methyl carbonate (EMC) (3:7 ratio by weight) (SoulBrain, MI). The electrolyte fill factor, defined as the ratio of the electrolyte volume to the total pore volume in the cell, was 1.6.

2.3. Electrochemical testing

Electrochemical testing was carried out using a Maccor battery cycler. Cells were cycled within a voltage window of 3.0–4.2 V. Cells first underwent 4 formation cycles at C/10 charge and discharge rate followed by 500 aging cycles at 1 C rate (1 C = 160 mA/g_{NMC}). The cell voltage was held at 4.2 V for 3 h at the end of each charge during the aging cycling. Every 50 cycles the cells underwent a deep discharge with a constant voltage hold until the current dropped to C/20. A DC resistance test, hybrid pulse power characterization (HPPC), was performed after formation cycling and after every 50 cycles during discharge. The HPPC protocol included one full cycle at C/10 rate followed by one full cycle at C/3 rate to prepare the cell for characterization. The cell was recharged to 4.2 V at C/3 rate and the HPPC was performed during discharge at C/3 rate at every 10% state of charge (SOC). The HPPC protocol starts with a one-hour rest step followed by a discharge pulse at 2 C for 10 s and a regeneration pulse at 1.5 C for 10 s. There is a rest step for 40 s before and after the regeneration pulse. Three cells were cycled for each type of defect to standardize the results. After cycling, the cells were disassembled in an argon-filled glove box, and the cycled electrodes were soaked in DMC solvent for 1 min to remove residual electrolyte prior to post-mortem characterization.

2.4. Microstructural characterization

2.4.1. FTIR

Fourier-transform infrared spectra were collected in attenuated total reflectance (ATR) mode with a germanium crystal. The FTIR instrument (Bruker Alpha) was housed in an argon-filled glove box to prevent air exposure.

2.4.2. Raman spectroscopy

For Raman spectroscopy, anodes were sealed under glass inside an

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