Neutron Computed Tomography Imaging of Lithium Concentration in $\text{LiNi}_{0.33}\text{Mn}_{0.33}\text{Co}_{0.33}\text{O}_2$ Cathode

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Abstract

$\text{LiNi}_{0.33}\text{Mn}_{0.33}\text{Co}_{0.33}\text{O}_2$ (NMC) is a promising substitute for LiCoO_2 because of its good thermal stability and high energy density. In this work, the lithium concentration distributions in an NMC cathode using neutron computed tomography technique at Oak Ridge National Laboratory's High Flux Isotope Reactor (HFIR) CG-1D Cold Neutron Imaging Facility. Samples with four different state of charge (SOC) were prepared for neutron imaging: 70% SOC, 100% SOC, 105% SOC, and 110% SOC. The neutron tomographic reconstruction of NMC cathode reveals the information of electrochemical transport and spatial Li distribution inside the cathode. The experimental results were explained by a diffusion numerical model which maps the Li concentration evolution during the electrochemical reactions. The study demonstrates that neutron imaging technique can be a very powerful tool to understand the lithium concentrations and evaluate its state of conditions, thus providing information for design of safe lithium ion batteries and estimating their lives.

Keywords: Neutron scattering, Lithium ion battery; Concentration; Computed tomography; Modeling

1. Introduction

Lithium ion batteries have excellent performance of combined high energy and power density, making it desirable energy storage devices for portable electronics, power tools, and electric vehicles [1, 2]. During the operation of lithium ion batteries, lithium ions insert or extract into and from the host materials. Hence, the amount of lithium ions in the host material is a good indicator for evaluating the available energy stored.

In order to understand the lithium concentration evolution during the battery operations, physics based models including electrochemical reactions and mass transport have been developed [3-6]. These models can all be benefited by measurement of lithium concentration for model validation. However, direct and nondestructive measurement of lithium concentration evolution is very limited [7, 8].

Recently, neutron radiography technique was successfully used to observe the lithium growth in carbon [9]. In neutron radiography, the transmission of neutrons are captured by a scintillator and recorded as a gray scale radiograph. The imaging contrast comes from the differences in attenuation coefficients of the elements. The interactions of neutrons with elements can be described by the Beer-Lambert law [10],

$$I = I_0 \exp(-\sum_i \mu_i x_i) \quad (1)$$

where I and I_0 are the transmitted and incident neutron beam intensity. μ is the total attenuation coefficient of element i . x is the effective thickness of the element. The attenuation coefficient μ is described by

$$\mu = \sigma_{tot,i} N_A \mu_i \quad (2)$$

where $\sigma_{tot,i}$ is total neutron cross section and μ is concentration. The advantage of neutron imaging over other X-ray computational tomography is that the neutron primarily interacts with a material via the nucleus while X-ray interacts with electrons. This interaction is particularly strong for hydrogen and lithium, while being much weaker for many metallic elements. Note that the interaction of neutrons with hydrogen and lithium is of totally different origin. Hydrogen is scattering neutrons via incoherent scattering. Lithium is absorbing neutrons via a resonance process and transmuted.

For NMC electrode, the total cross section of Li is a magnitude higher than Ni, Mn and Co according to the NIST (National Institute of Standard and Technology) database [11]. Table 1 shows the NIST's mass attenuation coefficient values of Li, Ni, Mn, and Co [12]. As shown in the table, Li has the lowest mass attenuation coefficient. This leads to a huge transmission difference of Li with respect to Ni, Mn and Co, and makes measurement of lithium concentration possible.

Table 1: Values of the mass attenuation coefficient, μ/ρ (cm^2/g), at 1 MeV [12]

Li	5.503×10^{-2}
Ni	6.160×10^{-2}
Mn	5.852×10^{-2}
Co	5.906×10^{-2}

In this work, the lithium concentration of NMC electrode will be studied by neutron radiography technique. The paper will be organized as follows: In section 2, the experimental details are given. In section 3, the lithium concentration of the NMC electrode is studied and compared with the model. Conclusions are given in section 4.

2. Experimental

2.1 NMC sample preparation

The commercial cells used in this study is LG 18650 cells (LGDBHE41865). The cathode is $\text{LiNi}_{0.33}\text{Mn}_{0.33}\text{Co}_{0.33}\text{O}_2$ (NMC), and the anode is mesocarbon microbeads (MCMB) graphite. The normal operating potential is between 2.0 V (0% state of charge) and 4.2 V (100% state of charge). The nominal capacity is 2500 mAh. Samples with four different state of charge (SOC) were prepared for neutron imaging: 70% SOC, 100% SOC, 105% SOC, and 110% SOC.

Over-charge tests were carried out using the BT-2000 32- channel cycler (Arbin Instruments, TX). Galvanostatic tests were performed on the LG 18650 NMC cells with a nominal 2500 mAh capacity. The cells were charged to 4.20 V at 1 C rate (constant current of 2.50 A) first; then, the charging was continued at a constant voltage of 4.20 V until the current was less than 0.02 A, which is regarded as a 100% state of charge. The over-charge test conditions were carried out at 1 C rate (constant current, 2.50A) and based on the Coulombs charged into the cell; namely, the discharge step was terminated when the cells reach 105% SOC (63 min., 2550 mAh) and 110% SOC (66 min., 2750 mAh).

The cells were carefully disassembled in an argon filled glove box after charging the cells. The cathodes were cut off from the disassembled cells for neutron imaging.

2.2 Neutron computed tomography imaging

Neutron computed tomography imaging was performed at the CG-1D beamline at the High Flux Isotope Reactor (HFIR), Oak Ridge National Laboratory. The CG-1D Cold Neutron Imaging Facility beam uses a polychromatic neutron beam in the cold range for neutron radiography measurements. CG-1D has high sensitivity to lithium to track the lithium ions. The schematic of neutron radiography set up is shown in Fig. 1. The neutrons are generated by fission reaction in the 85 MW reactor core. The thermal neutrons scatter from the beryllium reflector and reach a supercritical hydrogen moderator held at a temperature of 17 K. This provides a source of cold neutrons with increased flux and with wavelengths between 4 and 12 Å. The neutrons are collimated using a series of beam scrapers. Apertures are used at the entrance of the helium-filled flight path to allow variations of the figure of merit L/D , where L is the distance between the aperture and the detector where the image is produced. L/D of 800 is used in this study. A $50\text{ }\mu\text{m}^6$ LiF/ZnS scintillator converts the transmitted neutron into light that is recorded by a charge-coupled device (CCD) camera. A gray scale 2D-projection, based on neutron transmission intensity, is saved as the raw data. The sample is mounted on a rotation stage that rotates through 180° to collect over a thousand of radiographs. In this work, each projection was taken in rotation steps of 0.1° with 60 s exposure time. For post-processing, the projected images were reconstructed by filtered-back-projection (FBP) method using Octopus imaging software (XRE NV, Belgium, <https://octopusimaging.eu/>).

(Fig. 1: Schematic of CG-1D neutron imaging set up [13].)

3. Results and discussion

The normalized gray scale images of NMC samples are shown in Fig. 2. The texture in the images comes from the aluminum foil which wraps around the NMC samples. Using the case SOC=70% as the baseline and normalizing its grayscale to [0, 1], all the other three SOC's images were normalized using the same scaling factor. The normalized grey scales for SOC=100%, 105%, and 110%, are [-0.25, 1.09], [-0.26, 1.20], and [-0.28, 1.09], respectively.

From Fig. 2, it can be observed that with the increase of SOC, the neutron images of the NMC electrode become lighter, which is consistent with the lower end of the grey scale analysis. This is because lithium ions are extracted from the NMC cathode during the charge process. When lithium concentration is low, more neutrons will be transmitted. The experimental lithium ion concentration under different charge conditions were obtained by averaging the neutron transmission intensity at different locations. Then the transmission intensity was converted to lithium ion concentration according to the Beer-Lambert law (see equations 1 and 2). The obtained lithium ion concentrations are shown in Fig. 3.

(Fig. 2: Normalized gray scale neutron scattering images of NMC samples which are wrapped in aluminum foil. (a) SOC=70%, (b) SOC=100%, (c) SOC=105%, (d) SOC=110%.)

In order to compare the experimental results, a 1D diffusion model was used. At any time, the mass transport of lithium ions should follow Fick's second law, which is given by,

$$\frac{\partial c(x,t)}{\partial t} = D \frac{\partial^2 c(x,t)}{\partial x^2} \quad (3)$$

where c is lithium ion concentration, x is the coordinate along electrode thickness and D is diffusion coefficient. The initial and boundary conditions are:

- (1) Initial lithium ion concentration is constant throughout the thickness of the NMC electrode.
- (2) Zero lithium ions flux at the electrode-current collector interface.
- (3) Constant lithium ions flux at the electrode-electrolyte interface.

The above conditions are given by:

$$c(x, t = 0) = c_0 \quad (4)$$

$$-D \frac{\partial c(x=0,t)}{\partial x} = 0 \quad (5)$$

$$-D \frac{\partial c(x=L,t)}{\partial x} = \frac{I}{FA} \quad (6)$$

where c_0 is the initial lithium concentration, I is applied current, F is Faraday constant, A is surface area and L is the thickness of the NMC electrode. In the model, the initial lithium concentration was chosen as 48990 mol/m^3 [14]. The diffusion coefficient of lithium was $7.6 \times 10^{-13} \text{ m}^2/\text{s}$ [15]. The thickness of the NMC electrode was estimated around $250 \text{ }\mu\text{m}$. The simulated lithium ion concentration at different SOC were averaged for comparison.

As shown in Fig. 3, experimental lithium ion concentrations are compared with the simulation results. A strong linear correlation exists between the lithium concentration and the

SOC: Lithium concentration (mol/m^3) = $-38370 \text{ SOC} + 48061$ ($R^2 = 1$). It can be seen that the simulation data are consistent with the experimental measurements. The difference may be caused by the assumption of the constant diffusion coefficient and simplified reaction kinetics.

(Fig. 3: Comparison of simulated lithium ion concentration and experimental results.)

4. Conclusions

1. In this work, neutron imaging technique was successfully applied to study the lithium ion concentration inside an NMC electrode with different state of charge. It demonstrates that neutron imaging technique can be a very powerful tool to study the lithium concentrations in lithium ion batterie and evaluate its state of conditions, thus providing information for design of safe lithium ion batteries and estimating their lives.

2. The lithium ion concentrations obtained by experimental measurements are consistent with the 1D diffusion model. Both the experiment and the model show a strong linear correlation between the lithium concentration and the SOC: Lithium concentration (mol/m^3)= $-38370 \text{ SOC} + 48061$ ($R^2 = 1$).

Acknowledgement

This research used resources at the High Flux Isotope Reactor, a DOE Office of Science User Facility operated by the Oak Ridge National Laboratory. This manuscript is based upon work

supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences
under contract number DE-AC05-00OR22725.

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Figure captions

Fig. 1: Schematic of CG-1D neutron imaging set up [12].

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Fig. 3: Comparison of simulated lithium ion concentration and experimental results.

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(a)

(b)

(c)

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