In-situ Li Ion Battery Imaging of Charge-Discharge Using Stray Field NMR

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Introduction

Electrochemical processes typically involve ion conductive pathways in order to achieve inter-conversion between electric energy and chemical energy for energy storage. During the charge/discharge cycling, many inter-exchangeable structural phases are developed in the electrodes. Some of the structural phases become irreversible, leading to capacity decay, low energy efficiency, and a short cycle life. The microscopic structural information at the atomic level is very important, particularly under in situ conditions, for understanding electrochemical processes (such as ionic transport properties, interfacial properties, chemical reaction, electrochemical stability, and structural transformation) of the electrode materials. Imaging with sufficient resolution of an electrochemical cell is an effective non-destructive tool for characterizing ion motions as well as the structural and dynamic changes upon the composite motion of the ions during electrochemical processes, thereby providing expansion of microscopic structural information to the atomic level - a very important step to understand the electrochemical processes.

Experimental

The stray-field-imaging (STRAFI) experiments on lithium-ion-batteries have been carried out on the ultra-narrow bore 830 MHz (19.6 T) magnet at NHMFL [1]. The ^{7}Li STRAFI profiles have been obtained to monitor non-destructively charge-discharge cycles with a resolution of 39 µm [2]. We performed real-time STRAFI measurements on $LiCoO_2$ and $LiFePO_4$ half-cell batteries, i.e. monitoring real time lithium ion transfers when the charge-discharge process is applied, allowing one to see how the lithium population distribution changes as a function of charge-discharge time.

Results and Discussion

Fig. 1 shows the 1D profile of the $LiCoO_2$||Li half-cell battery as a function of charge/discharge time. The two sharp peaks in the left side are attributed to skin effects on the metal's surface. As the battery is charged, the Li content of the cathode depletes as the anode surface increases. However, there exist some residual Li-ions when charged. In case of the $LiFePO_4$||Li half-cell as shown in Fig. 2, Li ions completely move out of the $FePO_4$ olivine structure [2]. The Li ions transfer in charge-discharge processes of $LiCoO_2$ and $LiFePO_4$ half-cells has been well demonstrated.

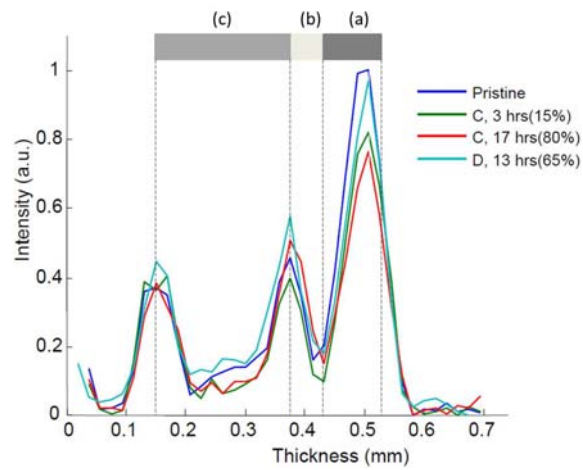

Fig 1. $LiCoO_2$ half-cell, (a) $LiCoO_2$ electrode, (b) separator layers consisting of two Celgard 3501 membranes, (c) Li foil

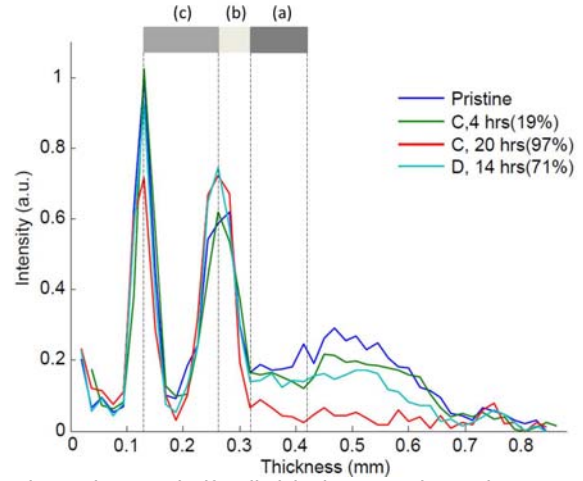

Fig 2. $LiFePO_4$ half-cell, (a) $LiFePO_4$ electrode, (b) separator layers, (c) Li foil

Conclusions

The high resolution STRAFI is a very useful method for in situ studies of charge-discharge processes of Li ion battery cells.

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References

[1] Tang, J.A., *et al*., Journal of Magnetic Resonance, **225**, 93-101 (2012).
[2] Tang, J.A., *et al.,* Scientific Reports, **3**, 2596 (2013).