Capacity Fading Mechanism and Side Reaction Research on Graphite/ LiNi_{0.5}Mn_{1.5}O₄ Full Cell Yanbao Fu, Xiangyun Song, Paul Ridgeway, Gao Liu, Vincent Battaglia 70R0108B, Lawrence Berkeley National Laboratory

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The spinel lithium nickel manganese oxide LiNi_{0.5}Mn_{1.5}O₄ (LNMO) material offers high energy density performance with its high, flat, voltage plateau of ca. 4.7V vs. Li/Li⁺ and its theoretical specific capacity of 148 mAh g⁻¹ [1]. LNMO is considered one of the promising and attractive cathode materials for high power lithium ion batteries. In the past several years, the research of LNMO focused on synthesis of different morphology, the relationship of its crystal type with its capacity and cycle performance, doping with other metal elements. However, LNMO has not been utilized in the production of lithium ion batteries as the full cell with LNMO cathode and graphite anode shows poor cycle performance, especially in the initial several cycles capacity faded a lot. Till now, there are few research reported on capacity fading mechanism research of graphite/LNMO full cell [2]. In this research, we will discuss the mechanism of capacity fading and side reaction in Graphite/LNMO full cells.

LNMO (obtained from NEI) and graphite (CGP-G8, purchased from Coconphilips) composite electrode laminate were prepared by Hydro-Quebec. 2325 coin cells were assembled for electrochemical measurements.

The cycle performance of LNMO coupled with CGP-G8 is shown in Fig.1. The columbic efficiency of the 1st cycle is 76.6%, which is lower than other common used cathode materials couple with graphite. The low CE is attributed to several factors: SEI formation of anode and cathode (side reaction happened on both anode and cathode, and the irreversible capacity of full cell is the sum of cathode and anode), capacity matching of cathode and anode (Charge process is limited by cathode, while discharge is controlled by anode), capacity loss of cathode at 4.0 V. In the later 4 cycles of formation, the capacity continued to fade. From EIS we observed that the semicircle corresponding to film impedance on anode and cathode surfaces grew gradually with cycling, which indicated that SEI formation on both anode and cathode sides is another source of capacity loss of CGP-G8/LNMO full cell. We researched the side reaction rate and found that side reaction rate is almost the same at different C-rate for this cell, which can also explain that capacity faded more seriously at C/20 than other rates.

We also analyzed the charge and discharge curve in depth, just shown in Fig. 4. Capacity loss is from 1^{st} plateau of cathode, or second plateau of cathode, or anode side (mainly from 1^{st} and 2^{nd} capacity plateaus). The detailed information will be discussed in the meeting.

Reference

- Q. Zhong, A. Bonakdarpour, M. Zhang, Y. Gao, and J. R. Dahn, J. Electrochem. Soc. 144, 205 (1997).
- [2] Y. Fu, X. Song, V. Battaglia et.al, Abstract #964, PRiME 2012 ECS meeting, 2012, Honolulu.



Fig.1. Cycle performance of LNMO/CGP-G8 cell



Fig. 2. Capacity-voltage profile of LNMO/CGP-G8 cell.



Fig.3. EIS of cathode and anode side for initial 5 cycles obtained from 3-electrode cell.



Fig.4. Adjusted charge curves (bump corresponding to $Ni^{3+} \rightarrow Ni^{4+}$ was stacked together).