Electrochemical behavior of Li-ion full cell battery employing Si-graphene negative electrode and $LiMO_2$ positive electrode during cycling

KwangSup Eom*[a], Inhwan Do[b], Thomas F. Fuller[a]

[a]School of Chemical & Bimolecular Engineering
Center for Innovative Fuel Cell and Battery Technologies
Georgia Institute of Technology, Atlanta, Georgia 30332

[b]XG Sciences, Inc.
3101 Grand Oak Drive Lansing, MI 48911

The development of new electrode materials with higher energy density is one of the most significant challenges for Li-ion cells. Silicon is an attractive negative electrode material due to its high theoretical specific energy. Unfortunately, Si expands by a factor or three during Li intercalation, leading to a large irreversible capacity loss (ICL). In order to mitigate this capacity fading, Si composites with buffer materials such as graphenes have been used. Graphene has received much attention because it has high electrical conductivity as well as good mechanical properties, leading to improved rate capability and cycle life for the Si electrode. To take advantage of the two positive effects of graphene, the size and composition of Si and graphene should be optimized. In this study, we used a Si-graphene composite fabricated with optimized conditions employing nano-sized Si and nano-sized graphene particles, which showed good cyclability in half-cell tests. Even though there are many reports on Si-graphene negative electrode, most of data are from half-cell tests. A half cell has obviously different electrochemical behavior from that of a full cell. Causes for these differences are attributed to, limited supply of cyclable lithium, dissolution of electrolyte and metals from the positive electrode, *e.g*. Hence, in order to estimate the feasibility for a practical use of a new electrode, a full-cell test is needed.

In this study, we designed the Li-ion full cell using a new Si-graphene negative electrode with a $LiMO_2$ positive electrode and explored its electrochemical and microstructural behaviors during battery cycling. Fig.1 (a) presents the charge/discharge curves of a full cell at C/15. The full cells showed no capacity fading between 1st and 5th cycles except for the large ICL related with the first charge. The initial discharge capacity based on weight of positive electrode is 133 mAh g^{-1}, which is lower than $LiMO_2$ half cell (169 mAh g^{-1}) although capacity of negative electrode is designed 1.13 times more than positive electrode based on actual initial capacities of each electrode. It is probably due to an increase in overall resistance through the cell. Fig. 2(b) shows dC/dV behaviors with cycling. It is notable that the 1st lithiation behavior is different from that of subsequent cycles. During the succeeding charge, a broad peak presented at 3.9 V decreased dramatically and disappeared after the 2nd cycle, and the second peak at 4.1 V decreased after the 1st cycle, and a new broad peak appeared at 3.4 V and remained during cycling. It is suggested that this behavior is attributed to the phase transformation of Si from crystalline to amorphous. Fig. 2 (a) shows capacity fading of this full cell. With initial cycling, the capacity decreased very slightly and the Coulombic efficiency increased, stabilizing at 98.9~99.5% after ten cycles. This excellent performance shows the possibility of new practical Li-ion battery with high specific energy density. Nevertheless, the capacity decreased gradually by about 20 mAh g^{-1} at C/2 from 10th to 50th cycle. It is probably due to continuous formation of SEI layer by Si pulverization during cycling. This hypothesis is supported by changes in the EIS data of Fig.2 (b). From 5th to 50th cycles, the SEI resistance of first semi-circle increased by 27 %, whereas the other resistances were unchanged. For more improvement of the cycling performance of this full cell, a study on the electrolyte additive is presently being performed.

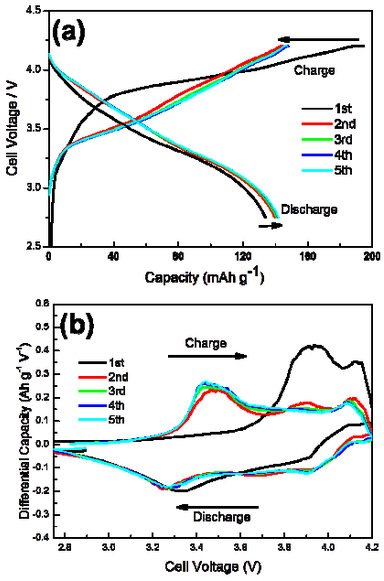

Fig. 1 Charge/discharge curves (a) and differential capacity curves (b) of full cells employing Si-graphene negative electrode and $LiMO_2$ positive electrode for the first 5 cycles at C/15 between 2.75 and 4.2 V.

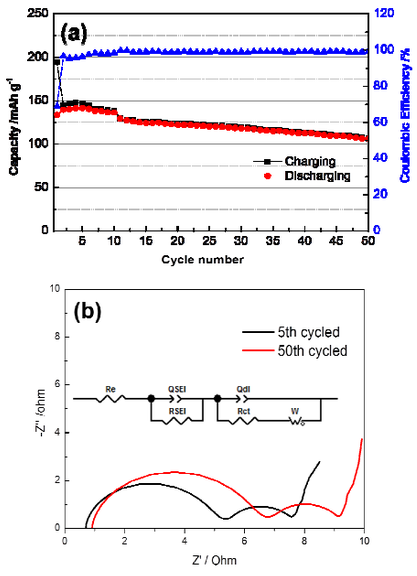

Fig. 2 (a) Capacity fading of a full cell using a Si-graphene negative electrode and $LiMO_2$ positive electrode at C-rates of C/15 in 1st ~ 5th, C/5 in 5th ~10th, and C/2 in 10th ~50th. (b) EIS curves of the full cell after the 5th and 50th cycle.

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