

Studies on Lithium Electrode Coated with Li_2CO_3 in Lithium Rechargeable Batteries

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The demand for Li secondary batteries used as portable devices has increased [1]. Much effort has been expended to produce lithium secondary batteries with lithium metal anodes because of their high energy density compared with Li-ion batteries. However, the poor recharge-ability of the lithium anode prevents the practical use of such batteries. This is caused by the formation of lithium dendrites during the charging process. Such dendritic lithium leads to internal short circuits through the separator to touch the cathode material. Many researchers are currently studying to suppress the dendritic formation of lithium during the charging process, but good results have not yet been obtained [2].

The formation of dendritic lithium is related to the interface state and reactivity between lithium and the electrolyte. Several methods have been suggested to control the passivating layer on the surface of lithium. These methods are as follows; (i) electrochemical deposition method of lithium metal in nonaqueous electrolyte containing organic or inorganic additives, (ii) temperature control method of electrolyte, and (iii) pretreatment method of lithium anode by polymer or solid electrolyte materials. These approaches were somewhat shown to enhance cycling performance of Li secondary batteries.

Our previous work reported on the use of lithium carbonate (Li_2CO_3) and lithium phosphorus oxynitride (LiPON) to make a good SEI layer on the surface of carbon for Li-ion batteries. The SEI layer formed using these materials was enough to suppress the initial irreversible capacity associated with solvent decomposition. Especially, the SEI layer composed of Li_2CO_3 has good properties to protect

carbon anode surface from electrolytic decomposition reaction. Therefore, the Li_2CO_3 is also expected as good SEI layer to prevent lithium anode surface of Li secondary batteries.

In this work, the lithium electrode coated with Li_2CO_3 was prepared as a means of controlling the SEI layer when lithium is immersed in electrolyte consisting of 1.0 M LiPF_6 in the mixed solvent of ethylene carbonate (EC) and diethyl carbonate (DEC). The effects of Li_2CO_3 were examined using chronopotentiometry (CP), impedance spectroscopy, and chronoamperometry (CA).

Fig. 1 shows the voltage-time profiles of lithium electrodes at a rate of 10 mA/cm^2 in 1 M $\text{LiPF}_6/\text{EC}:\text{DEC}$ (1:1, volume ratio). The cycling experiment indicates that the polarization during dissolution (over 0.0 V) and deposition (under 0.0 V) is different for both the uncoated and the Li_2CO_3 -coated electrodes. The Li_2CO_3 -coated electrode exhibited far less polarization. As shown in Fig. 1, the Li_2CO_3 -coated electrode was polarized to $\pm 0.35 \text{ V}$ while the uncoated electrode was polarized to irregular voltage over $\pm 0.35 \text{ V}$. One reason for this result can be associated with the solid electrolyte interface (SEI) layer. The surface of the uncoated lithium electrode is may covered with the products of electrolyte decomposition reaction. The SEI is composed of the various layers, which are Li_2O , LiOH , LiF , and Li_2CO_3 layers. The surface of the Li_2CO_3 -coated electrode, however, is may covered with only Li_2CO_3 . This is an important point of difference between two systems.

References

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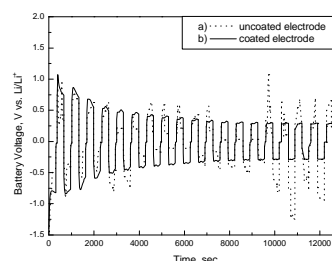


Fig. 1. Voltage-time profiles of lithium electrodes at a rate of 10 mA/cm^2 in 1 M $\text{LiPF}_6/\text{EC}:\text{DEC}$

