Electrochemical Impedance Spectroscopy Study on Interface Characterization of Lithium Ion Rechargeable Batteries during Charge and Discharge Cycle

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It is important to control the electrode/electrolyte interfaces to improve the performance of lithium-ion rechargeable batteries. For example, it is well known that a stable film is formed on the graphite electrode by the decomposition of the electrolyte solution during the first charge cycle in EC electrolyte solution. This film protects the electrolyte solution from its further decomposition and is called as a solid electrolyte interphase (SEI). It is possible to investigate the conditions inside the battery without its destruction by an electrochemical impedance spectroscopy (EIS). In EIS measurement, the time stability of the electrode, which means no variation of electrode reaction rate during the measurement of the electrochemical impedance spectrum, is required in the reaction system. Stoynov and Savoa-Stoynov proposed the compensation method by using 3-dimensional complex plot, whose axes are real, imaginary parts and time, for the deviation of low frequency components of the impedance without time stability.<sup>1</sup> Itagaki et al. applied this method to the investigation of active dissolution mechanisms of metallic electrode.<sup>2</sup> Since the electrode reaction in lithium-ion rechargeable batteries does not satisfy the time stability during the charge and discharge cycle, it is supposed that low frequency components of impedance contains significant errors due to the time variation. In the present paper, the impedance for the negative and positive electrodes of lithium-ion rechargeable battery during charge and discharge cycle is corrected by the above-mentioned compensation, and the variations of electrode/electrolyte interfaces are analyzed. In all previous reports regarding impedance analysis on lithium-ion rechargeable batteries, the charge-discharge was stopped to measure the impedance spectrum.<sup>3</sup> The first purpose of the present paper is to develop the new electrochemical method in which the impedance spectra are measured simultaneously with the charge- discharge sequence of rechargeable batteries.

The second purpose is to characterize the electrode/electrolyte interfaces by the new impedance method. This method was applied to the investigation of the negative and positive electrodes of lithium-ion rechargeable batteries. In the investigation of the negative electrode, the resistance  $R_{sei}$  and capacitance  $C_{sei}$  for the SEI film were determined, and the formation mechanisms of SEI film and the influence of the additives were discussed.

References

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