

초청강연

SEI Formation on Graphite Negative Electrodes of Lithium-Ion Batteries

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In rechargeable lithium batteries, carbonaceous materials are used as negative electrodes. It is generally recognized that a kind of passivating film, called solid electrolyte interface (SEI), is formed at the interface between the carbon electrode and electrolyte solutions in the initial stage of charging. The formation of SEI prevents further solvent decomposition and contributes to the enhancement of their safety and cycleability. On the other hand, the SEI formation causes an irreversible capacity. It is very important to understand the nature of SEI formation and suppress the consumption of charge consumed for SEI formation in order to increase cell capacity and to design electrolyte solution. Although a large number of studies are focusing on the elucidation of SEI these several years, reactions resulting in the formation of SEI and its properties have not been fully understood. We have so far studied initial steps of SEI formation on graphite in some electrolyte systems employing mainly in situ STM and AFM.

Intercalation of solvated lithium ion is an initial stage of the SEI formation at around 0.8 V vs. Li⁺/Li. The intercalated solvent molecules are partly decomposed between graphite layers under reduced potential of the graphite and leave immobile products there. The remaining solvent molecules are further decomposed at negative potential and exfoliation of graphite layer at the surface follows the decomposition and the decomposition products lead to the formation of SEI directly or after reaction with electrolyte solution. The potential where co-intercalated solvent is decomposed is dependent of the solvent species. Reaction kinetics and decomposition potential of solvent give a major influence on SEI formation together with the expansion of graphite layer by the co-intercalation of [Li(solv)_n]. When 12-crown-4 is added to electrolyte solution, complex of Li⁺ with 12-crown-4 preferentially intercalates into graphite and the ligand 12-crown-4 is more stable against reductive decomposition between graphite layers than carbonate solvent.

Although it has been pronounced that inorganic products such as LiF, Li₂CO₃ play a major role in SEI, we believe that some organic species like lithium carbonate are also important including lithium alkoxide.