Ammonium Salts as an Electrolyte Additive for Lithium Metal Anode

Kazuki Yoshii, Hikari Sakaebe
Research Institute of Electrochemical Energy, Department of Energy and Environment,
National Institute of Advanced Industrial Science and Technology (AIST),
1-8-31 Midorigaoka, Ikeda, Osaka 563-8577, Japan

E-mail: k.yoshii@aist.go.jp

Lithium metal has been considered an attractive candidate of the anode material for the next-generation secondary batteries because of its high theoretical capacity and low redox potential. Suppression of the dendritic growth of lithium during charging and reversible deposition and dissolution of lithium are still the problems to be tackled for the utilization of lithium metal as an anode. Among various strategies in order to solve the problems reported before, an addition of additives for the electrolytes is one of the simplest way and some additives have been suggested as a useful candidate[1-3]. In the present paper, we examined the effect of ammonium salt as an additive for an organic electrolyte toward total performance of electrodeposition and dissolution of lithium.

Tetrabutylammonium hexafluorophosphate ($N_{4444}PF_6$) was mixed with 1.0 M of $LiPF_6$ in propylene carbonate (1.0 M $LiPF_6$_PC) in the Ar-filled glovebox. Electrodeposition and dissolution cycling test of lithium on Ni foil was performed at 25 °C using two-electrode cell prepared in the Ar-filled glovebox. Lithium foil was used as a counter electrode.

Figure 1 shows change of the voltage during galvanostatic electrodeposition and dissolution of lithium using 1.0 M $LiPF_6$_PC with and without 50 mM of $N_{4444}PF_6$. 5 mAh cm^{-2} of lithium was used for the cycling test after 10 mAh cm^{-2} of lithium was deposited at first charge. The cycle stability was significantly improved by addition of $N_{4444}PF_6$, suggesting that stable SEI was probably formed. The electrodeposition of lithium was also conducted with two-electrode cell without separator. The morphology of the deposited lithium obtained in 1.0 M $LiPF_6$_PC with 50 mM of $N_{4444}PF_6$ was smooth and dense in contrast to the heterogeneous electrodeposit obtained in 1.0 M $LiPF_6$_PC. These results indicate that $N_{4444}PF_6$ is likely to be candidate as an additive for the electrolyte toward lithium metal anode.

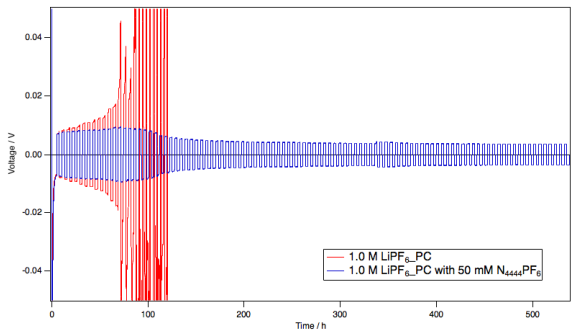

Figure 1. Change of the voltage during galvanostatic electrodeposition and dissolution of lithium. Electrolyte: 1.0 M $LiPF_6$_PC with and without 50 mM of $N_{4444}PF_6$. Current density: 2.0 mA cm^{-2}.

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References:

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