In Situ Detection of the Solid-Electrolyte Interphase Formed in Bis(fluorosulfonyl)amide Ionic Liquids

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The solid-electrolyte interphase (SEI) is formed by the decomposition of electrolytes and plays an important role in the cycling performance of anode materials for rechargeable lithium batteries. However, the properties of the SEI have not been elucidated because the SEI is considered to be a thin layer ranging from a few to hundreds of nanometers and the in situ analysis of the SEI is difficult. The ex situ analysis of the SEI suggested that the SEI is composed of some inorganic and organic components. However, the original nature of the SEI cannot be studied from the samples after treatments such as washing and drying. We have reported that the formation and breakdown of the SEI formed in some ionic liquid electrolytes can be evaluated in situ by monitoring the outersphere electron transfer redox reactions. The rate of the redox reaction depends on the tunneling distance between an electrode surface and the redox species. The detectable thickness of the SEI is considered to be a few nanometers, indicating that the initial stage of SEI formation can be monitored by this method. In addition, the breakdown or disappearance of the SEI under open circuit can be monitored by simple electrochemical measurement such as cyclic voltammetry. In the present study, the formation and breakdown of the SEI formed in some bis(fluorosulfonyl)amide (FSA-) ionic liquids containing LiFSA were investigated at room temperature.

The redox reactions of some metallocenes were used as redox probes. The formation of SEI on a platinum electrode under the potentiostatic cathodic reduction was detected by a shift in the peak for anodic oxidation of the metallocene in the ionic liquid electrolytes by cyclic voltammetry. The formation and growth of the SEI depended on the organic cation. The formation of the homogeneous SEI covering the entire electrode surface was suggested by the peak shift. In addition, the recovery of the anodic peak shifted by the SEI formation indicated that the SEI was soluble or dispersible in the electrolytes. Therefore, it was suggested that the SEI formed in the ionic liquids was a gelled and/or viscous phase formed by the dissolution and/or dispersion of the degradation products in the electrolytes.

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References

1. Kato S., Serizawa S., Katayama Y., *J. Electrochem. Soc.* **2022**, *169*, 076509.
2. Okazaki S., Serizawa N., Katayama Y., *Electrochemistry* **2024**, *92*, 043006.

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