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Current Status of Operando-μ^+SR for Battery Materials at J-PARC

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Ion transport in solids is a key feature for the operation of ion batteries. There are two parameters for describing ion transport in battery materials; one is a self-diffusion coefficient (D^J) and the other is a chemical diffusion coefficient (D^C). The former diffusion is caused by thermally activated fluctuation of ions, while the latter diffusion is caused by a flow due to a concentration gradient of ions. Majority of work concerning battery materials, D^C has been measured with an electrochemical technique under a concentration gradient of the ion in a half-cell. D^C is then estimated using the relationship: $D^C = \Theta D^J$, in which Θ denotes a thermodynamic factor.
According to the Cottrell equation, the time evolution of the current of the planer electrode in the half-cell under an ion-concentration-gradient has a relation, $I(t) \propto A_{re}\sqrt{D^C}C$, where A_{re} and C denote the reactive surface area of the electrode and the concentration of the ion. Thus, the obtained value from the electrochemical measurement is not D^C but $D^C A_{re}^2$. Because the correct A_{re} in liquid or solid electrolytes is unknown, it is very difficult to determine D^C. We have thus initiated series of experiments to measure intrinsic D^J of battery materials with μ^+SR [1]. Due to the change in the crystal structure and occupancy of a regular Li site with SOC, D^J is predicted to depend on SOC [2]. Therefore, it is highly desirable to measure D^J as a function of SOC under working condition, namely, an operando μ^+SR. We are attempting to establish such technique in J-PARC, and show the current status.

[1] For example, J. Sugiyama et al., Phys. Rev. Lett. 103, 147601 (2009).
[2] A. Van der Ven and G. Ceder, Electorchem Solid-State Lett. 3, 301 (2000).

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