



Contribution ID: 51

Type: Oral Presentation

Direct measurement of nanoscale lithium diffusion in solid battery materials using radioactive tracer of ^8Li

Thursday, 14 May 2015 16:50 (20 minutes)

The diffusion coefficient of lithium in solid materials used in secondary Li-ion batteries is one of key factors that determine the rate at which a battery can be charged and discharged. Nevertheless, reported lithium diffusion coefficients in Li battery materials measured with various methods are largely scattered over several orders of magnitudes, for example, 6 orders for LiCoO_2 and 8 orders for LiMn_2O_4 , which are commercially used as electrodes. The traditional radioactive tracer method with a serial sectioning technique can provide the most accurate diffusion coefficient in a direct way, but it cannot be applied for Li battery materials because of no availability of radioactive Li isotopes with a half-life suitable for such offline applications. We developed an in situ lithium diffusion tracing method using a short-lived radioactive ion beam of ^8Li , with the half-life of 0.84 s, which immediately decays into two alpha particles. Tracing the time evolution of the changes in the energies of the alpha particles from diffusing ^8Li , which is primary implanted into a sample of interest, we can extract Li diffusion coefficient directly. The method has been successfully applied to measure diffusion coefficients in Li ionic conductors (e.g., [1]). The range of measurable lithium diffusion coefficient by the current method is from 10^{-6} down to 10^{-10} cm^2/s . For measurements of Li diffusion coefficients in battery electrodes such as LiCoO_2 , it is required to improve the lower limit of the detection by this method.

We have proposed a new method by detecting alpha particles emitted from decaying ^8Li at a small angle (10 degree) relative to a sample surface that is irradiated with a low-energy (8 keV) ^8Li beam. The new method has been successfully applied to measuring Li diffusion coefficients for an amorphous $\text{Li}_4\text{SiO}_4\text{-Li}_3\text{VO}_4$ thin film, demonstrating that the new method is sensitive to diffusion coefficients down on the order of 10^{-12} cm^2/s , corresponding with nanoscale Li diffusion of several 10 nm/s [2]. Using the new method, measurements of Li diffusion coefficients in electrode materials such as LiMn_2O_4 are in progress. In this presentation, we will introduce recent experimental results of our newly developed nanoscale Li diffusion measurement.

[1] S. C. Jeong, et al., *Solid State Ionics* (2009) 180, 626.

[2] H. Ishiyama, et al., *Jpn. J. Appl. Phys.* (2014) 53, 110303.

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Session Classification: Session 15