IN SITU STUDY OF THE PHASE TRANSFORMATIONS IN THE GRAPHITE ANODES BY RIETVELD METHOD

A. Missyul[a,b], I. Bolshakov[a], R. Shpanchenko[a]
[a]*Samsung SDI, Suwon, South Korea*
[b]*ALBA Synchrotron Light Source, Cerdanyola del Valles, Barcelona, Spain*

Laboratory XRD remains the most used technique for structural analysis of battery materials due to equipment simplicity and accessibility. Analysis of the X-ray diffraction data for cathode materials is usually carried out with Rietveld method. A situation with analysis of the XRD data for graphite anode materials is entirely different. Layered nature of graphite results in a very high preferred orientation and a large number of the structural defects produce different types of strong distortions of the diffraction peaks profiles. As a result, typically analysis of the anode diffraction data is limited by the comparison of the positions and intensities of the 002 graphite peak and corresponding peaks of the charged phases. This limitation might lead to the incorrect conclusions about the chemical composition of the charged anodes, especially when both LiC_{12} and LiC_{18} phases exist simultaneously.

We developed a reproducible procedure for the Rietveld refinement of the mixtures of the most important intermediate phases during the anode charging procedure, namely, LiC_{12}, LiC_{18} and LiC_{30} based on the structure description provided in [1,2]. This, combined with the well-known LiC_6 structure, allows performing the detailed analysis of the graphite anode in the SOC range 20-100%. As an example of the importance of the full profile analysis we provide the results of the *in situ* investigation of the high-density graphite anode showing the variation of the degree of the preferred orientation during cycling and within each cycle. This variation is described for the first time and its possible effects on the charging process and the battery performance are discussed.

[1] Billaud D., Henry F.X., Lelaurain M., Willmann P. *J. Phys. Chem. Solids*, 1996, 57, 775.
[2] Billaud D., Henry F. *Solid State Commun*., 2002, 124, 299.