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Structural change of carbon anode in a lithium-ion battery product associated with charging process observed by neutron transmission Bragg-edge imaging

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Abstract

Spectroscopic neutron Bragg-edge imaging was performed to study a lithium-ion battery (LIB) product. This non-destructive neutron imaging method is suitable for the evaluation of industrial products, but presents some difficulties for application to multicomponent products. The LIB includes a strong neutron scatterer and an absorber, and is thus a suitable test case for the use of neutron imaging in actual product measurement. In this study, we analyzed the variation of the graphite anode structure with changes in the battery charge level. The experiments were carried out using the compact neutron source at the Hokkaido University neutron source facility (HUNS). To eliminate the effect of scattered neutron contamination, we first determined the distance between the sample and detector required to reduce this effect to under 1 %. Using this separation, the charge level dependence of the anode structure was measured. The graphite {002} Bragg-edge could be recognized on the neutron transmission spectra. The Bragg-edge was shifted and broadened with increasing battery charge. The edge was consistent with the existence of multiple graphite structural stages. The layer spacing distribution images for different charge levels showed the inhomogeneous fluctuation on the LIB lattice plane. Based on the images the fraction of the graphite structural stages were analyzed. The ratio of each stage varied with the charge level, and the ideal intercalation structure, in which the graphite layers are stuffed with Li-ions, was found to be minor in the final charging state.

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Keywords: Bragg-edge transmission; lithium-ion battery product; Strong scatterer; Strong absorber; Intercalation

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1. Introduction

Non-destructive test methods to evaluate products are important in many industrial fields. Radiography techniques are suitable for this purpose, because the transmission image obtained is directly related to the real subject at actual scale. In particular, neutron radiography has been attracting attention as a useful test method, since neutrons have deep penetrating ability. Recently, energy-selective neutron radiography has been developed (E. H. Lehmann, et. al., 2009). This technique gives us a set of images that differs from those offered by conventional radiography. The energy-selective images show characteristic contrast, which are affected by the energy-dependent total cross-section. Analyzing the details of the energy-dependence is called spectroscopic transmission imaging, which can analyze the distribution of crystalline microstructures (S. Nagashima, et. al., 2014). This means the method can non-destructively evaluate a sample's microstructural characteristics and the distribution of material functions within it, both of which make this method attractive for analyzing industrial products.

In this study we selected a lithium-ion battery (LIB) as an industrial product test case (V. Etacheri, et. al., 2011). In the battery, lithium ions move between anode and cathode according to the charging stages. When the battery is charged, the lithium ions intercalate into the carbon crystal lattice, which is used as the anode material. When this process is repeated, the crystal structure of the anode gradually collapses, eventually causing a decline in battery performance. The structure variation of the graphite anode in a LIB product during the charge-discharge cycle is important in determining the performance of the battery. The spectroscopic neutron transmission technique is suitable to analyze the crystalline structure of the functional materials within the product, the carbon anode in this case, because of its non-destructive evaluation ability.

During LIB charging the crystal lattice spacing of the graphite {002} becomes larger than that in the LIB's discharged state, because the lithium ions intercalate into the graphite layers of the anode. The lattice spacing of graphite can thus be related to the charge level of the LIB. Using pulsed neutron Bragg-edge imaging, which is one analysis mode of the neutron transmission spectrum, we may obtain the spatial distribution of the charge level in a LIB product. However, it is not clear that Bragg-edge analysis can be applied to the LIB, which consists of some materials having large neutron cross sections. For an example, hydrogen in the battery electrolyte, which has a large scattering cross section, makes the transmission measurement difficult by creating a large measurement background. In addition, the measurements are affected by lithium ions due to their large absorption cross section. Therefore, at first we examined the feasibility of applying the Bragg-edge transmission method to a LIB product. We then performed transmission imaging of the materials in a LIB product, and studied the variation of the anode crystal structure and real-space distribution with changes in the charge level. For such industrial applications a compact accelerator-driven neutron source is very useful for continuous study, so the effectiveness of such a neutron source was also validated in this study.

2. Experimental

The LIB sample under study is shown in Fig. 1. It was a lamination-type product. Each battery cell consisted of a set of four layers: the negative electrode, a separator, the positive electrode, and a second separator. The negative electrode was graphite with 110 μm thickness, and the positive electrode was $LiFePO_4$ 116 μm thick. The separators were 20 μm thick microporous polyethylene. The whole battery was composed of a stack of 19 cells within an organic liquid electrolyte.

The experiments were performed at the Hokkaido University neutron source (HUNS) facility (M. Furusaka, et. al., 2014). The beam line had a cold source, which was equipped a mesitylene moderator maintained at around 40 K at the time. The energy resolution of the Bragg-edge region was approximately 3 % with the moderator system. We used a GEM detector having 128 x 128 pixels with a pixel size of 0.8 mm x 0.8 mm (S. Uno, et. al., 2012). The detector was placed at a position 6 m from the moderator surface, and we used the time-of-flight technique for the neutron energy determination. In order to evaluate the effect of the scattered neutrons, the distance between the sample and the detector was varied. This scattering contamination effect was also simulated using the Monte Carlo code PHITS (T. Sato, et. al., 2013).