Demonstration of a new quantitative imaging technique using an energy dependence of an epithermal neutron transmission spectrum for application to Li-ion batteries

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1. Introduction

A quantitative imaging of lithium in a Li-ion battery during charging/discharging (C/D) process is required for improvement of its performance and safety. Neutron imaging techniques like neutron radiography (NR) and neutron resonance transmission imaging (NRTI) may be the candidates. In NR, quantitative imaging for one element with its preobtained calibration curve has been performed against the sample from which the neutron transmission of the target element only can be obtained. A Li-ion battery includes various kinds of elements. If the quantity distribution of the elements except lithium are held during C/D process, the transmission of lithium can be obtained, and the quantification of lithium is possible by NR. However, that is impossible because the almost elements move during the process. In NRTI, enhanced images for several elements have been obtained without the limitation like the NR. However, most of light elements are not available due to the absence of resonance in generally available neutron energy region. Therefore, conventional neutron imaging techniques cannot perform a quantitative imaging of lithium in a Li-ion battery.

For quantitative imaging of lithium in a Li-ion battery, we have developed a new technique. The new technique utilizes the energy dependence of a transmission spectrum. The energy dependence of neutron transmission spectrum is different in dependence of kinds of element, and experimental neutron transmission spectra reasonably agree with the spectra which are calculated from the evaluated nuclear data in the energy range of epithermal neutrons. Particularly, lithium has the large energy dependence even in the energy range of epithermal neutrons because it has a large absorption cross-section. Therefore, this technique has a potential to perform quantitative imaging of lithium in a Li-ion battery. In our previous study, this technique was applied to a test sample composed of iron and carbon elements at the Hokkaido University Neutron Source. Quantification of areal densities of the two elements was succeeded [1]. However, evaluation of lithium and imaging measurements have not been performed yet. Therefore, we carried out an experiment of spectroscopic transmission imaging of lithium at the J-PARC, which can produce a high L/D pulsed neutron beam, for the quantitative imaging of lithium using the proposed technique.

2. Experiment

We carried out a spectroscopic transmission imaging experiment at the NOBORU instrument located at the beam port 10 of pulsed neutron source of the Materials and Life Science Experimental Facility. As a demonstration sample, we prepared a stepped-wedge aluminum-lithium alloy (composition: Al 95 wt. %, Li 2.5 wt. %, Cu 1.5 wt. %, Mg 1.0 wt. %). The stepped-wedge sample has 5 steps of the thicknesses; 1.6 mm, 3.2 mm, 4.8 mm, 6.4 mm and 8.0 mm. The areal density of each element is proportional to the thickness of the step, and the actual areal densities of those in each step were calculated from the composition, weight and dimensions of the sample. In this experiment, we measured position-dependent neutron transmission spectra of the sample by a gas electron multiplier (GEM) detector [2] which is a two-dimensional time-of-flight (TOF) detector. The distance between the detector and the sample was one meter to remove

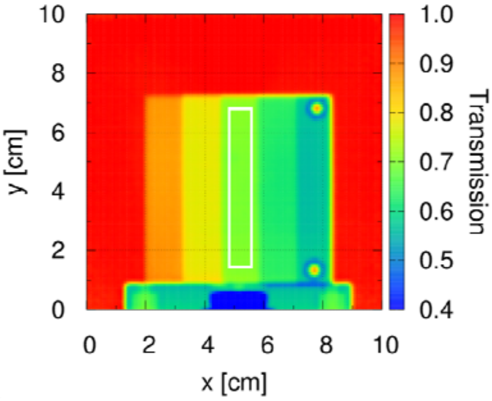

Fig. 1. Neutron transmission image of the aluminum-lithium stepped-wedge sample, which is obtained by neutron counts integrated in all neutron energy range.

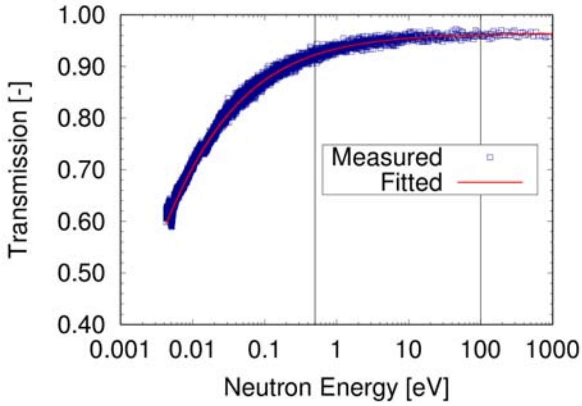

Fig. 2. An example of the fitting analysis. The plots show the measured neutron transmission spectrum in the step of thickness of 4.8 mm, and the curve shows the spectrum fitted in the energy range of 0.5 -100 eV.

scattered neutrons from the sample, and a collimator of the diameter of 3.16 mm was used for reducing the divergence of the neutron beam. The positions of the collimator, the detector and the sample were 8.0 m, 13.46 m and 14.46 m from the moderator surface, respectively.

The measured transmission spectrum of each step was fitted by a theoretical neutron transmission spectrum calculated from the evaluated nuclear data of lithium and aluminum, and the areal densities of those were obtained in each step. The copper and magnesium were ignored in this analysis because the neutron transmission spectrum was little affected by these elements of which cross-sections and the quantities are far smaller than the others. For confirming the accuracy of the technique, we compared the deduced areal densities with the actual ones.

3. Results

Fig. 1 shows the neutron transmission image of the stepped-wedge sample integrated in all neutron energy range. Transmission values varied in accordance with each step, and those in each step were almost uniform. Fig. 2 shows an example of the fitting analysis. In the figure, the measured neutron transmission spectrum was obtained from the sum of neutron counts in the area surrounded by a white line in Fig. 1. The energy range of the analysis was 0.5 - 100 eV. From the results of the fitting, it was confirmed that the obtained areal densities of lithium and aluminum in all steps were estimated within the differences of 10%, compared with the actual densities.

4. Conclusion

To realize a quantitative imaging of lithium in a Li-ion battery, we have developed a new neutron imaging technique which utilizes the energy-dependence of a neutron transmission spectrum in the epithermal neutron energy range. For the demonstration of the proposed technique, we carried out a spectroscopic transmission imaging experiment of a stepped-wedge sample which was composed of aluminum-lithium alloy at the NOBORU instrument. From the result of the analysis, the areal densities of lithium and aluminum in each step of the sample were obtained within the differences of 10% compared with the actual areal densities. We are now analyzing neutron transmission spectra in each pixel for the quantitative imaging in higher spatial resolution.

[1] H. Ishikawa, H. Sato and T. Kamiyama, Physica B (in press).
[2] S. Uno et al, Phys. Procedia 26 (2012) 142-152.