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Proceedings of Symposium on Ultrasonic Electronics, Vol. 37 (2016)
16-18 November, 2016

Mechanical properties of lithium-ion battery electrode
リチウムイオン電池電極の機械的特性

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1. Introduction

Recently, the lithium ion battery (LiB) has been used as a power supply of electric cars [1-3], but there are many problems such as short lifetime and low power density on the battery as yet. In general, the LiB composed of positive and negative electrodes, electrolyte and separator, is considered to start ageing from its electrodes. In this study, we focus our attentions to changes of mechanical properties of positive electrode of the LiB. The mechanical properties are studied by measuring resonance frequency and internal friction of the electrode at various temperature and charge/discharge cycles. The results indicated that the charge/discharge cycling causes decrease in the resonance frequency and increase in the internal friction of the LiB electrode.

2. Experimental

The positive electrode was composed of the active materials of $LiMn_2O_4$ and $LiNiCoAlO_2$, the conducting filler of carbon black, the binder of polyvinylidene difluoride (PVdF) and the collector of Al foil. A vibrating reed method was used to measure both resonance frequency and internal friction of the electrode. The measurement system of the mechanical properties of the electrode reeds is shown in Fig. 1. The reed sample was set in a copper box equipped in a vacuum chamber. Displacement of the free end of the reed sample was measured using a laser/CCD camera system. The temperature of the reed sample was changed from 100 K to 400 K with a rate of 0.2 K/min.

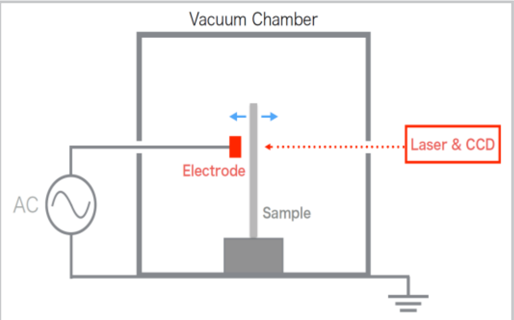

Fig. 1 Schematic diagram of measurement system.

3. Results and discussion

Figure 2 shows the resonance frequencies of the reed samples with lengths of 27, 29, 31 and 39 mm as a function of temperature. The positive electrode used was not enclosed into the battery, namely, the electrode did not react with electrolyte. As seen in

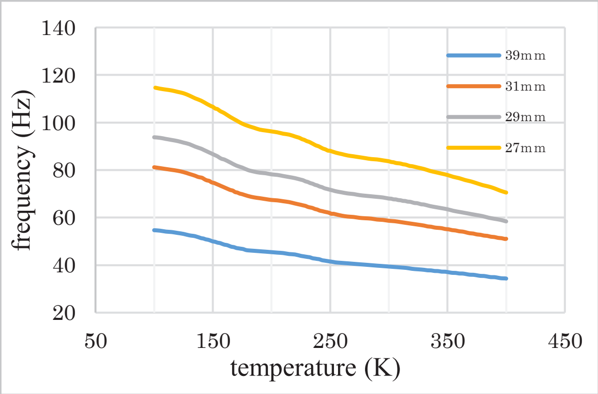

Fig. 2 Resonance frequencies of the reed samples of the pristine electrode as a function of temperature.

this figure that resonance frequencies decrease with increasing temperature and there are two sharp decreases on the frequencies at ~150 K and ~240 K. In general, a sharp decrease in the temperature-dependent resonance frequency will result in a peak on temperature-dependent internal friction [4].

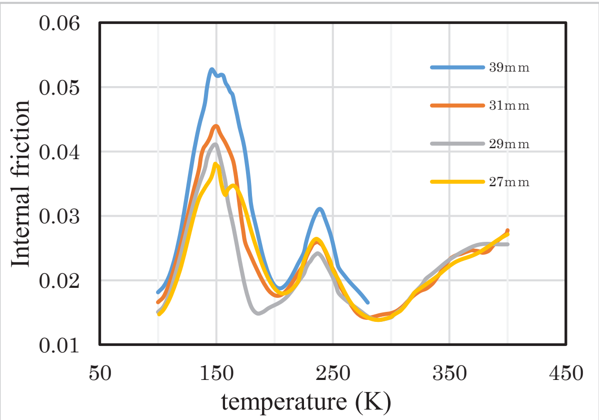

Fig. 3 Internal friction of the reed samples of the pristine electrode as a function of temperature.

Figure 3 shows the internal friction of the same reed samples with lengths of 27, 29, 31 and 39 mm as a function of temperature. The internal friction changes strongly with temperature in the range of 100~400 K. Two peaks are observed at ~150 K and ~240 K on the curves. The peak at ~150 K may be due to a surface-related relaxation process as well as the peak at ~240 K is due to a relaxation process of β-crystalline phase in the PVdF binder [5].

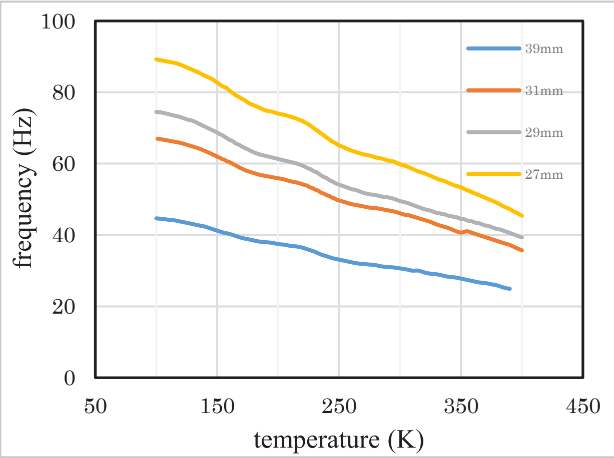

Fig. 4 Resonance frequencies of the electrode for the charge/discharge cycling of only once as a function of temperature.

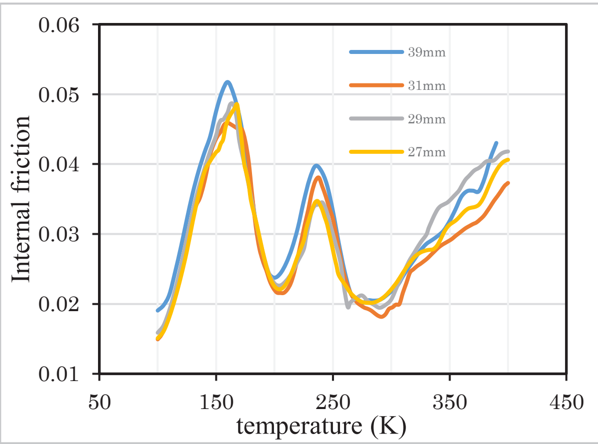

Fig. 5 Internal friction of the electrode for the charge/discharge cycling of only once as a function of temperature.

Figure 4 shows the resonance frequencies of the reed samples of the positive electrode with lengths of 27, 29, 31 and 39 mm as a function of temperature, where the electrode was obtained from the battery charged and discharged only once. Like the results shown in Fig. 2, the frequencies decrease with increasing temperature but their values decreased about 20 Hz despite of the charge/discharge cycling of only once.

Figure 5 shows the internal friction of the reed samples with lengths of 27, 29, 31 and 39 mm as a function of temperature, where the positive electrode was charged and discharged only once. The internal friction changes with temperature in the range of 100~400 K. No significant shift on the internal friction peak, and also compared with the results in Fig. 3, is observed.

4. Conclusions

Mechanical parameters of resonance frequency and internal friction of the positive-electrode of the LiBs were measured at temperatures from 100 K to 400 K. Two thermally-activated relaxation processes were observed at ~150 K and ~240 K. They were related to the relaxation processes occurred in the PVdF binder of the positive electrode. The internal friction was due to a surface relaxation for the peak at ~150 K and a phase transition of the β crystalline phase in the PVdF binder at ~240 K, respectively. Significant internal friction processes were not observed in the active materials of $LiMn_2O_4$ and the current collector of Al foil [6].

The mechanical parameters can be anticipated as effective indicators for characterizing ageing behaviors of the LiB electrode as a whole. By using this measurement technique, it becomes possible to enhance the cost-performance of the LiBs by matching each life times of positive and negative electrodes, separator and electrolyte.

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