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Electric Potential Distribution Across the $LiMn_2O_4$/Solid-Electrolyte Interface Observed by Electron Holography

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In recent years, as the power source, lithium ion rechargeable batteries are used for portable electric devices because of high energy density and light weight. A flammable liquid electrolyte is generally used for the electrolyte because ions can move fast in liquids. However, they have suffered from the issue of leakage and combustion of the liquids. Therefore, the development of all-solid-state lithium ion batteries which uses the nonflammable solid electrolyte are expected to be one of the main power sources for hybrid and fully electric vehicles for safety and high power density. However, there is a high interfacial-resistance of Li ions transfer between the electrode and the solid electrolyte. Observing the behavior of Li ions around the electrode/solid-electrolyte interface is important to clarify the mechanism of the interfacial reaction.

Electron holography is a powerful technique for directly observing the electric field in a transmission electron microscope (TEM) [1]. The electric potential resulting from Li ions can be detected as a phase shift of an electron wave passing through the LIB sample [2]. We show the dynamical change of electric potential resulting from Li ion movement during the battery charge in the TEM.

We prepared a model sample of the all-solid-state LIB illustrated in Fig. 1 (a). The $LiCoO_2$ and the $LiMn_2O_4$ were deposited on the solid electrolyte ($Li_{1+x+y}+Al_x(Ti,Ge)_{2-x}Si_yP_{3-y}O_{12}$) using pulse laser deposition, and they were used as positive and negative electrodes, respectively. Voltage was applied to the sample in the TEM and the electron holography was performed to observe the potential distribution around the $LiMn_2O_4$/solid-electrolyte interface. Figure 2 (a) and 2 (b) show the TEM image and the electric potential image across the interface at the charged state (1.2 V), respectively. The flat potential distribution was observed in the $LiMn_2O_4$, and the gradual potential slope was formed in the solid electrolyte. This shows that the electric double layer was formed by charging Li ions near the interface and spread for 1.4 μm.

References

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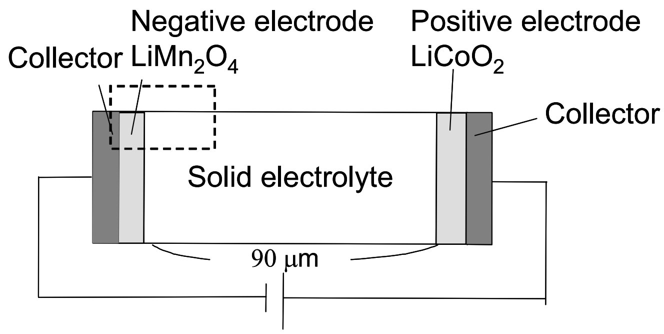

FIG. 1. A model sample of all solid state lithium battery.

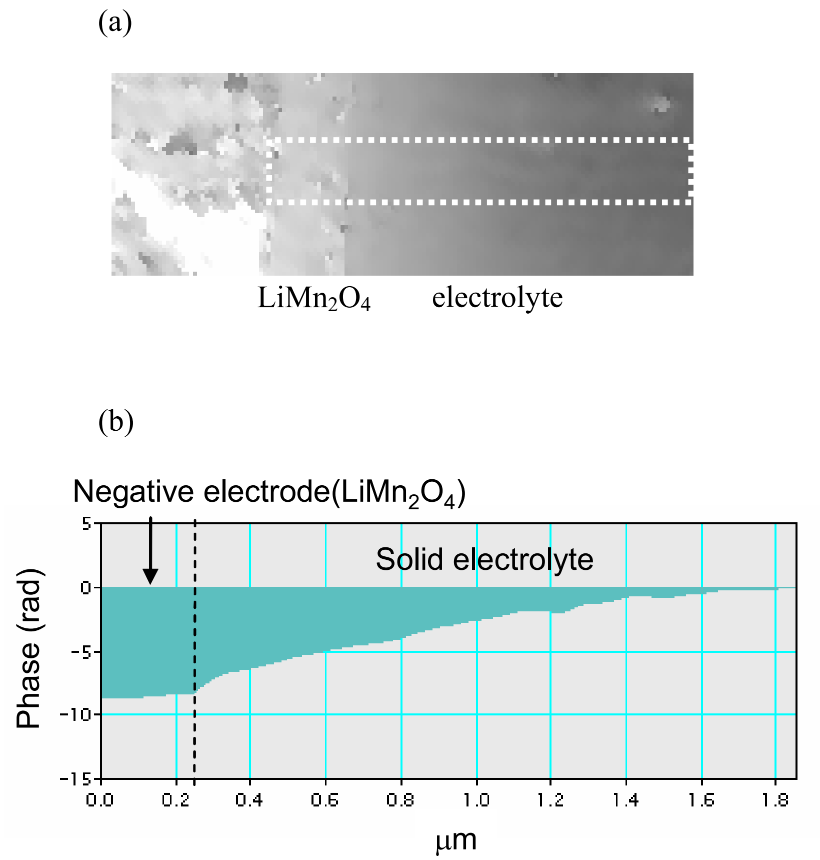

FIG. 2. Electric potential distribution around the negative-electrode/solid-electrolyte interface at the charged state (1.2V). (a) Reconstructed phase image. (b) Line profile surrounded by the dotted lines in (a).

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