



Enhancing utilization of lithium metal electrodes in all-solid-state batteries by interface modification with gold thin films



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HIGHLIGHTS

- Bulk-type all-solid-state Li symmetric cells are fabricated by vacuum-evaporation.
- A high utilization of 40% is achieved for Li metal electrode in the cells.
- The reversibility of Li utilization is improved by interface modification with Au.

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ABSTRACT

To evaluate ideal interfaces between Li metal and $\text{Li}_2\text{S-P}_2\text{S}_5$ solid electrolytes, bulk-type cells were fabricated using thin Li metal films prepared by vacuum-evaporation. The symmetric cell of $\text{Li}/75\text{Li}_2\text{S}\cdot 25\text{P}_2\text{S}_5/\text{Li}$ showed a high utilization of Li metal (about 40%), though abrupt decrease of the utilization was observed after 5 cycles. A high utilization of Li metal (about 25%) was retained for 5 cycles by inserting Au thin films at the interface between Li metal and $\text{Li}_2\text{S-P}_2\text{S}_5$ solid electrolytes. Morphology of Li metal after dissolution/deposition cycles for the cell with Au thin films became more uniform compared to the cell without Au thin films.

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1. Introduction

Li ion batteries have been widely used as electric power sources for mobile phones, laptops and other devices. However, the highest energy storage which the Li ion batteries can deliver is not enough to meet the increasing demands of markets such as electric vehicles with long-distance drive. To achieve higher energy density, Li metal is the most attractive material as a negative electrode because of its extremely high theoretical capacity (3861 mAh g^{-1}) and the lowest negative electrochemical potential (-3.04 V vs. SHE) [1]. However, dendrite growth and low coulombic efficiency during charging and discharging are fatal problems for Li metal batteries using conventional organic liquid electrolytes. Dendrite growth leads to short-circuiting and increases the risk of fire [2]. The use of inorganic solid electrolytes in Li metal batteries is expected to solve the problems.

Li metal batteries using inorganic solid electrolytes are categorized to thin-film or bulk-type batteries. Thin-film batteries are prepared by accumulating solid electrolytes and electrode active materials on a plate by RF magnetron sputtering or vacuum-evaporation. These processes construct favorable interfaces between solid electrolytes and electrode active materials, including Li metal. Li metal batteries have achieved excellent cell performance in thin-film batteries [3–8]. Thin-film batteries using lithium phosphorous oxynitride (LiPON) as solid electrolytes show long cycle life over 40,000 times without the capacity fading [3]. However, energy storage in thin-film batteries is small because by using sputtering and vacuum-evaporation processes, it is difficult to accumulate a large amount of electrode active materials. On the other hand, bulk-type batteries are prepared by stacking solid electrolyte powders and composite electrode powders of electrode active materials and solid electrolytes by pressing [9]. Metal foils in place of the composite electrodes are used as metal electrodes such as Li electrodes. The bulk-type batteries are expected to have larger energy storage due to the addition of larger amounts of active

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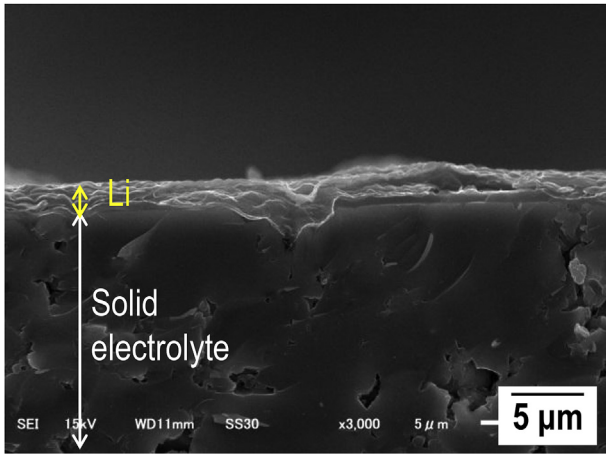


Fig. 1. Cross-sectional SEM image of the interface between a Li thin film and $\text{Li}_2\text{S-P}_2\text{S}_5$ solid electrolytes.

materials to the batteries. However, at least three points should be improved to realize bulk-type Li metal batteries; the first improvement is the Li ion conductivity of solid electrolytes, the second one is the chemical stability of solid electrolytes against Li metal electrode, and the third one is the interface condition between Li metal and solid electrolytes. For bulk-type batteries, it is difficult to form favorable interfaces between Li metal and solid electrolytes because grain boundaries and voids remain on the surface of solid electrolytes due to compression of the powders. Inhomogeneous interfaces are a serious issue for bulk-type batteries using Li metal electrode, forming inhomogeneous electrodeposition of Li metal during repeated charge-discharge cycles.

We have developed sulfide-based solid electrolytes and reported that $\text{Li}_2\text{S-P}_2\text{S}_5$ solid electrolytes have high ionic conductivities ($>10^{-4} \text{ S cm}^{-1}$) and chemical stability against Li metal [10,11]. $\text{Li}_2\text{S-P}_2\text{S}_5$ solid electrolytes are a good candidate to realize bulk-type Li metal batteries. Meanwhile, the interface condition between Li metal and solid electrolytes is still insufficient in spite of less grain boundaries and voids of the sulfide electrolyte powders pressed at

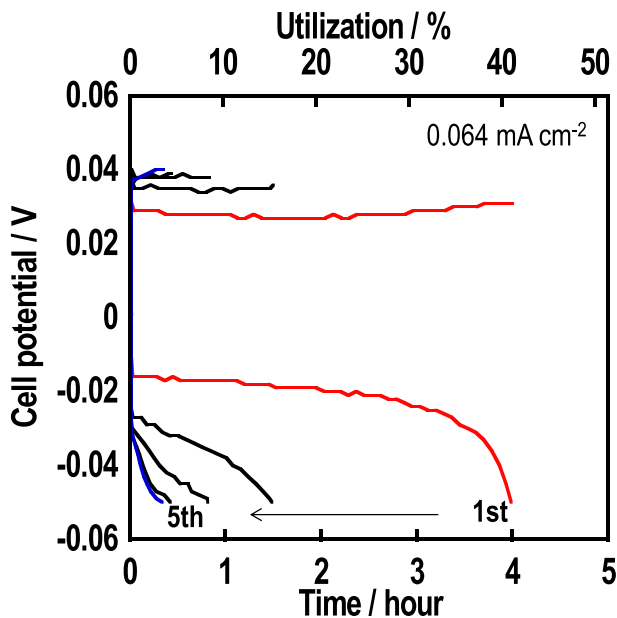


Fig. 2. Galvanostatic cycling tests for Li symmetric cells.

ambient temperature [12]. In our previous reports about bulk-type Li metal batteries, an abrupt potential change was observed for the batteries using a Li foil because of inhomogeneous interfaces between a Li foil and the electrolyte [13]. Our group also reported that favorable interfaces prepared by vacuum-evaporation of Li metal improved the reversibility of Li dissolution and deposition in bulk-type batteries. In addition, we and other researchers have suggested that insertion of a Li-alloy thin layer at the interface between Li electrode and solid electrolyte layers brought about a stable Li dissolution and deposition in the all-solid-state Li metal cells [14–16]. These interface modifications are effective in establishing homogeneous interfaces between Li metal and solid electrolytes.

To achieve high energy density in bulk-type Li metal batteries, investigations about utilization of Li metal are essential. Thin-film batteries such as Li/LiPON/LiCoO₂ cells have shown excellent cycle performance. Because Li/LiPON/LiCoO₂ cells use the lithiated positive electrode, the utilization of Li metal is not high. Bulk-type cells using metallic thin Li films prepared by vacuum-evaporation instead of Li foils are suitable to evaluate utilization of Li metal because ideal interfaces between Li metal and solid electrolytes are formed. The bulk-type cells with Li thin films are not suitable for practical application because of the smaller amount of energy

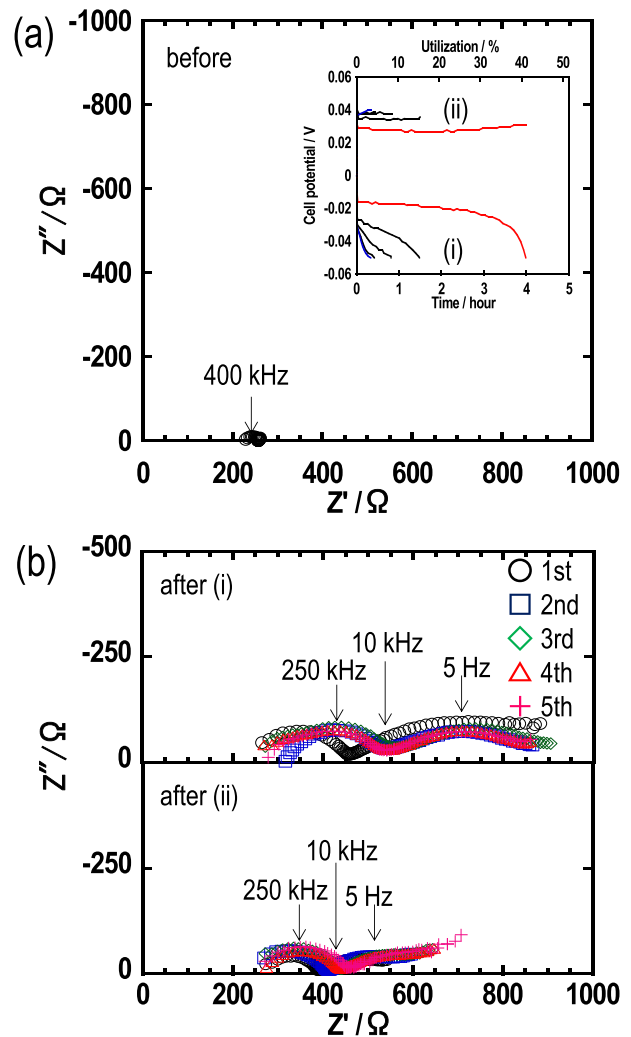


Fig. 3. Impedance plots of Li symmetric cells (a) before galvanostatic tests, and (b) after the tests at the negative potential direction (i) and at the positive potential direction (ii).

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