

Cycling stability of all-solid-state lithium symmetric cells using Li₃PS₄-LiI electrolytes

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For the fabrication of high energy and high power density batteries, a lithium metal negative electrode has been widely studied since it has extremely high theoretical capacity (3861 mAh g⁻¹) and the lowest negative electrochemical potential (-3.045 V vs. SHE). However, the growth of lithium metal dendrite during lithium dissolution and deposition, which leads to short-circuiting and raises safety concerns, is a fatal problem. Inorganic solid electrolytes are one of the most attractive materials to solve the safety issues. Recently, the behavior of lithium dissolution and deposition at high temperature has been studied. All-solid-state Li/Li₇La₃Zr₂O₁₂/Li cells showed a good cycling performance at 100°C [1,2]. Our group has investigated all-solid-state cells using Li₂S-P₂S₅ glass electrolytes. Li₂S-P₂S₅ glass electrolytes are a good candidate to fabricate lithium metal batteries because of their high ionic conductivities (>10⁻⁴ S cm⁻¹) and good formability to be densified just by cold pressing [3,4]. Furthermore, in order to improve their performance, the addition of a third component to Li₂S-P₂S₅ glass electrolytes has been studied. Mercier *et al.*, has reported that the ionic conductivity increase to about 10⁻³ S cm⁻¹ by adding LiI to Li₂S-P₂S₅ glass electrolytes [5]. Based on the theoretical calculation, LiI is stable to lithium metal [6]. Therefore, the addition of LiI is expected to improve not only the ionic conductivity but also chemical stability to lithium metal. In this study, Li₃PS₄-LiI solid electrolytes were prepared by mechanical milling. All-solid-state lithium symmetric cells using Li₃PS₄-LiI solid electrolytes were fabricated and their behaviors of lithium dissolution and deposition were investigated.

Galvanostatic cycling tests were carried out at 100°C under 1.25 mA cm⁻² for 4 hours in half a cycle. As shown in Fig. 1, the cell using the 54Li₃PS₄·46LiI (mol%) glass electrolyte did not short-circuit even after cycling for more than 3400 hours and showed a high cycling stability. After the test, the Li/54Li₃PS₄·46LiI interface was evaluated by SEM observation. Compositional contrast and cracks were not observed at the interface, indicating that good contacts of the interface were maintained even after cycling more than 3400 hours.

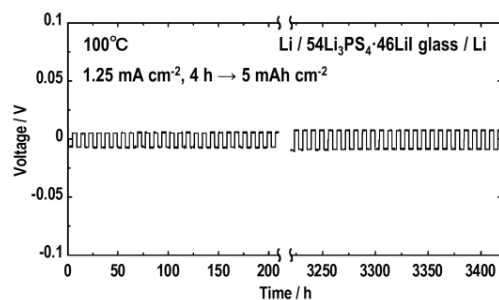


Fig. 1 Long-term galvanostatic cycling test of the Li/54Li₃PS₄·46LiI/Li cell.

References:

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