Modification of Grain-Boundary for Prevention of Lithium Growth through Garnet-type Solid Electrolytes

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Garnet-based lithium ion conducting solid electrolyte is a promising solid electrolyte for all solid-state batteries [1]. However, lithium dendrite growth during lithium deposition on the negative electrode causes short-circuit with increasing current density [2, 3]. In this work, we prepared garnet-based Li\textsubscript{3}La\textsubscript{3}Zr\textsubscript{1.5}Ta\textsubscript{0.5}O\textsubscript{12} (LLZT) pellets, with different synthesis conditions, and investigated correlation between the pellet structure and short-circuit prevention. LLZT powder and pellets were prepared using a solid-state reaction and spark plasma sintering technique, respectively, as reported in our previous work [2]. The prepared pellets are designated as LLZT-air (conventional method), LLZT-w/o-air (without exposing to air), and LLZT-2calc (calcined twice with addition of LiOH). A combination of structural and chemical characterization techniques, such as SEM, STEM and FT-IR revealed presence of LiOH and Li\textsubscript{2}CO\textsubscript{3} on the LLZT powder and effects of excessive lithium salt on the microstructure of the pellets. To investigate the short-circuit prevention, symmetric cells of Li | LLZT | Li were cycled at various current densities at 25°C, which were gradually increased until the cells showed voltage drop [2, 3]. As shown in Figure 1, the critical current density, at which voltage drop occurred, depended on the specimens. LLZT-2calc exhibited the highest, while LLZT prepared without exposing to air showed low short-circuit prevention. This suggests that Li\textsubscript{2}CO\textsubscript{3} and/or LiOH on surface of starting LLZT powder is effective to improve density of grain boundaries in the pellets to suppress the short circuit.

\begin{figure}[h]
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\includegraphics[width=\textwidth]{figure.png}
\caption{DC polarization curves of Li | LLZT | Li cells using LLZT pellets prepared with different condition.}
\end{figure}

References: