Temperature effect on Li wettability of garnet structured $Li_7La_3Zr_2O_{12}$

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All solid-state battery has been attracted much attention especially in its improved safety on e-mobility applications. Nevertheless, its popularization would rely on the using of metallic Li as anode so that its energy density is comparable or more than nowadays Li-ion batteries. Recent researches found that the using the garnet structured $Li_7La_3Zr_2O_{12}$ (LLZ) as solid-state electrolyte and Li as anode often results in a short circuit due to the formation of Li dendrite. Extensive works have been reported to solve or relief the dendrite problem by modifying the interface in between metallic Li and LLZ using thin metal layer[1] or gel polymer [2], adding second phase additive to the grain boundaries[3], changing LLZ surface roughness [4] and elaborately removing surface contaminants of LLZ[5]. The applied methods are in aim of either reducing the contact resistance in between LLZ and Li or obstructing the dendrite percolation path. However, little attention was paid to the heat treatment temperature, varied from 60 ^{o}C to 300 ^{o}C in different reports, of the used symmetric cell after Li was attached to the LLZ pellets.

In this study, Ta-substituted LLZ pellets were prepared from solid state reaction with a relative density of ~93%. Pellets were fine polished with SiC sandpaper and plasma etched in an Ar-filled glove-box to remove possible surface contaminants before Li foils were attached to form Li//LLZ//Li symmetric cells. It was observed that the interface resistance can be dramatically reduced from 250 $\Omega \cdot cm^2$ to 15 $\Omega \cdot cm^2$ with increasing heat treatment temperature from 25 ^{o}C to 310 ^{o}C. The galvanostatic cycling of Li//LLZ//Li symmetric cells shows a long term stability when using a current density of 0.2 mA cm^{-2} at 25 ^{o}C. Furthermore, the low interface resistance was observed to be unaffected by applying an Au interlayer when heat treated the symmetric cells beyond the Li melting temperature which suggested the high temperature heat treatment has major effect on increasing the wettability of Li to LLZ for reducing the interface resistance.

References:
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