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Electrode/Electrolyte Interfacial Contact Analysis for Solid-State Li Batteries by means of *in situ* X-ray tomography

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1. Introduction

A polymer composite solid electrolyte is prevalent in current all solid-state Li battery technology owing to its excellent flexibility, good ionic conductivity, and, most importantly, its notable energy density achieved with a metallic Li anode [1]. However, interfacial issues associated with the use of a Li anode obstruct the development of a Li metal based solid-state battery. Li dendrite growth within the electrolyte after repeated cycling reduces critical current density and promotes cell shorting. In addition to initial poor electrode/electrolyte contact, thick SEI layer formation and Li stripping during cycling further deteriorate the interfacial contact and reduce the electrochemical performance of the cell (Figure 1). Imaging is significant to analyse the interfacial failure mechanism and assure uniform current distribution at the interface over prolonged cycling [2]. The use of noninvasive in-situ X-ray tomography, unlike other ex-situ imaging techniques, remains to be explored widely in this field, and appears critical for in-depth understanding of interfacial changes. Thus, the present research work focuses on the design of a symmetric cell and the 4D imaging of interfacial events between the polymer composite solid electrolyte and the Li anode during cycling using in situ X-ray tomography.

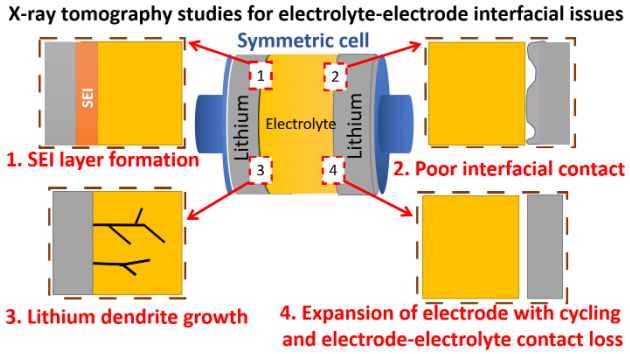

Figure 1. Schematic presenting application of X-ray tomography to understand the interfacial contact phenomena in solid-state Li metal batteries.

2. Materials and Methods

Firstly, a polymer composite solid electrolyte was synthesized. This electrolyte was stacked between two thin strips of metallic Li anode under pressure to guarantee contact. The stack was then mounted in a Swagelok type cell. The assembly of the symmetric cell took place inside a glove box under an inert argon atmosphere to avoid oxidation. Electrochemical Impedance Spectroscopy (EIS) was carried out on the symmetric cell to determine its interfacial resistance. Next, galvanostatic Li stripping/plating was performed at different current densities until failure of the system was observed. At regular intervals the electrolyte-electrode interfacial region was scanned with X-ray tomography. Post-mortem, the interfacial region was analyzed with SEM.

3. Results and Conclusion

As will be shown at the conference, the X-ray scans enable to visualize the interfacial changes responsible for the degradation of the cell, despite the limited spatial resolution of the scans (µm-range). Nevertheless, the degradation follows the increase in interfacial resistance. The higher-resolution SEM images provide complementary data to the X-ray data and together provide the clues to further improve the cycle performance.

This work demonstrates the potential influencing factors at the interface, paving the way for further research into how to improve cycling performance.

4. Acknowledgements

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5. References

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[2] S.M Bak et al, DOI 10.1038/s41427-018-0056-z