

Investigation of solid state batteries by operando neutron imaging

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The continuous demand for electrical energy storage devices challenges battery developers to create devices with ever increasing energy and power density. In this context, performance and safety are the key parameters to master, however incidents still continue to happen with new systems. All-solid-state lithium-ion batteries promise to enhance safety by suppressing the organic electrolyte. Thanks to intensive research on sulfide-based superionic conductors, the power density of these batteries is expected to be on par or even exceeding the one of conventional lithium-ion batteries. Despite some progress in the development of superionic conductor, some aspects related to transport properties inside the solid electrolyte and/or in the composite electrode still remain non-elucidated.

In order to get a better insight about lithium transport upon cycling, the development of a reliable electrochemical cell is thus of a prime importance when studying a solid state battery in *operando* mode. This is never an easy task, since the design of such cells has to be adequate for the analysis technique chosen, in our case neutron imaging. Neutron imaging requires low contrast materials (e.g. aluminum) for the casing of the cell, a good trade-off between the spatial resolution and the field of view, as well as finding materials which concentration changes significantly affect neutron attenuation. Once a proper design is achieved, several parameters such as lithium transport (diffusion and migration) and lithium concentration can be investigated in order to improve battery engineering/performance. Herein we present a reliable cell design developed for *operando* neutron imaging investigations of a solid state battery. The main advantage of the neutron imaging is the high contrast of Li especially when ⁶Li is employed. As can be seen in Figure 1, already at OCP there is an isotopic exchange between the counter electrode made of ⁶Li-In and the solid electrolyte made of natural Li. Upon cycling, this exchange is even more pronounced since we can even notice a gradient (in the grey scale) of ⁶Li/natural Li. Cell design and further transport mechanisms will be discussed in this presentation.

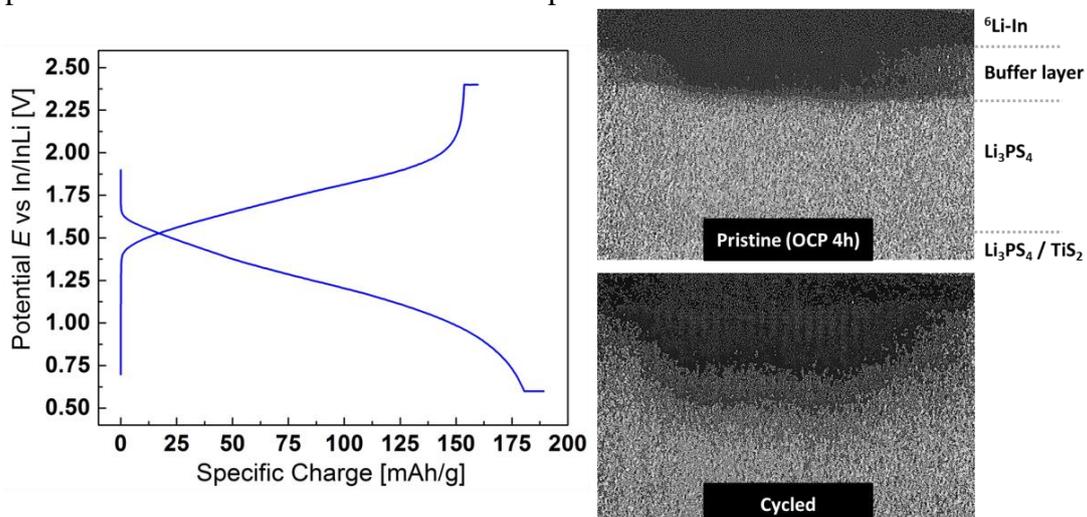


Figure 1. (left) 1st Galvanostatic cycle of ⁶Li-In|Li₃PS₄|TiS₂:Li₃PS₄ cycled at C/10 rate in the special cell developed from the neutron imaging measurement; (right) Neutron radiography image of the cell at OCV after 4 h and after the first cycle .