

MICROSTRUCTURE & MORPHOLOGICAL EVOLUTION OF SOLID-STATE BATTERIES DURING CYCLING

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All-solid-state batteries have been presented as the ideal solution to address i) the safety limitations of conventional Li-ion batteries by suppressing the flammable organic electrolytes and ii) the problem of insufficient energy densities if coupled to Li metal. To date, two types of solid Li-ion electrolytes have been mainly studied, namely, sulfur-based and ceramic-based materials. As they are easy to process and they offer a high lithium ion conductivity in the range of 1-20 mS/cm, sulfide-based electrolytes such as e.g. thio-LISICONs and argyrodites are therefore regarded as suitable candidates to be used in lithium all-solid-state batteries.

Despite the progress in the development of superionic conductor, many aspects regarding their chemical and mechanical issues remain unsolved especially during electrochemical activities where the electrolyte is heavily decomposed. If the electrode engineering i.e. composite electrode (mixture of electroactive material, conductive agent and solid electrolyte) is under intense investigation, the role of the solid electrolyte used as separator is so far poorly studied. Quite often, the electrochemical fading mechanisms observed with ageing are solely ascribed to the conventional fading parameters (cycling, temperature, interfacial reaction, chemo-mechanical degradation), but it might be that most of those parameters are happening prior any electrochemical activities during the sintering process. As an example, if the sintering (even cold sintering) is not properly performed, voids will appear in the solid electrolyte pellet, and during charge/discharge, the same voids could then evolve and propagate into cracks causing mechanical failure.

In this work, two different microstructure & morphological evolution will be monitored.

- The first one will focus on the relationship between the sintering parameters (time/pressure) of the sulfide-based solid electrolyte as a function of the electrochemical impedance spectroscopy. The goal is thus to establish a correlation between sintering and grain boundaries evolution.
- The second one will be based on an in-depth operando investigation of the microstructure evolution of sulfide-based solid electrolyte during electrochemical cycling.

In both case, X-ray tomography and X-ray diffraction computed tomography were selected as advanced techniques of choice to establish the correlation between electrochemistry, microstructure, morphology, and electrochemical fading. The results obtained here should serve as a preliminary basis to develop better solid-state batteries using sulfide-based electrolyte.