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# Electrode engineering for ASSB: Challenges and prospects

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All-solid-state batteries (ASSBs) have emerged as a promising technology to meet the growing demands for energy storage, offering significant advantages in terms of energy density, durability and safety. However, even if the properties of novel solid electrolyte materials have been widely investigated, few studies focused on the electrode engineering needed for solid-state batteries (no wetting possible). Indeed, the electrode engineering relies on the optimization of ionic and electronic conductivities as well as a proper interfacial contact between the electroactive materials and the solid electrolyte.

We will first address the fundamental challenges associated with the selection of electroactive material, focusing on the electrochemical properties, chemical stability, volumetric expansion, rate capability and cycle life. Next, we will look at the electrode formulation, especially the ratio between the components to ensure both ionic and electronic pathways. We will address key considerations such as viscosity, particle dispersion, thickness, and adhesion, and discuss advanced formulation techniques to improve the homogeneity and reproducibility of the slurry. Finally, management of electrode-electrolyte interfaces in ASSBs will also be discussed, such as undesirable interfacial reactions, limited ion diffusion phenomena, and mechanical stresses induced by variations in the volume of electroactive materials. Based on these parameters, we will demonstrate that it is possible to achieve excellent electrochemical performance of solid-state batteries assembled with  $\text{LiFePO}_4$  as a positive electrode material and a polymer-based electrolyte, all cycled at 25°C and 40°C.

Furthermore, we will investigate this system by means of advanced characterization techniques such as X-ray tomography and *operando wide-angle X-ray scattering (WAXS)* (ID31 beamline at ESRF) as can be seen in Figure 1. With the former technique, we successfully performed *operando* diffraction in the vertical-profile of the electrode showing the electronic limitation of the composite electrode while the latter technique allowed us to demonstrate that there is still some porosity near the interface between the solid electrolyte and the electrode materials, which may lead to an increase in cell polarization.

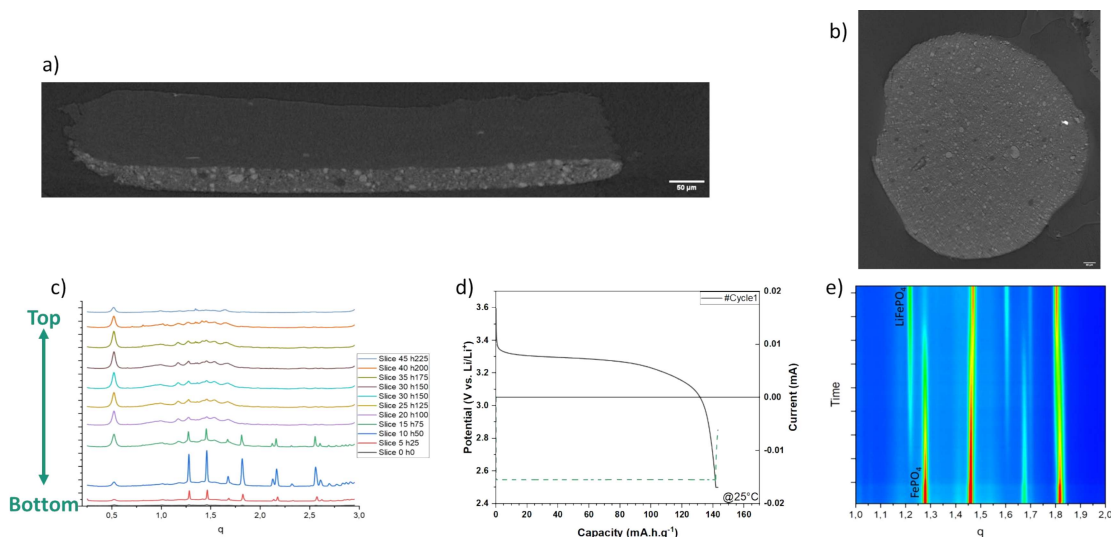


Figure 1: Advanced characterization of solid-state batteries. *a,b*) X-ray tomography of electrode/electrolyte stack along vertical and planar axes. *c,d,e*) Operando WAXS measurement (ID31, ESRF) on solid polymer electrolyte, showing vertical scan diffractograms in order to localize electrode area, electrochemistry curves resulting from cycling at C/20, as well as corresponding heat map of the middle of electrode revealing FP-LFP transition via classic biphasic mechanism.