Chemical and electrochemical degradation in metal anode/solid-electrolyte interfaces for solid state batteries

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Solid state batteries are posed to be the next generation of safer batteries that aims to substitute the state-of-the-art liquid flammable electrolytes-based batteries. The further integration of Li or Na metal anodes opens the door to higher energy densities and novel battery electrochemistries. The development of these systems has been however, so far hindered by the lack of understanding of the dynamic metal/solid interfaces that leads to chemical, electrochemical and mechanical degradation during cell operation.

Two of the best solid state alkaline-ion conductors are derivatives of $Li_7La_3Zr_2O_{12}$ Garnet-type structures for Li and $Na_3Zr_2Si_2PO_{12}$ NaSICON type structures for Na able to reach values of 10-1 mS/cm at RT. In this work, we focus on the study of Na/NaSICON and Li/Garnet interfaces paying particular attention to the effect that different processing conditions can have on the microstructure, local chemical composition of bulk, surfaces and grain boundaries (Figure 1). All these parameters have a direct impact on the Li and Na-ion dynamics leading to significant implications on the cell performance in terms of power density and cycle life due to different degradation issues such as SEI and dendrite formation.

Our main results highlight the complexity of these systems and propose new *in situ* techniques to gain a deeper understanding of the processes taking place. In this regard, we introduce a novel and beyond state-of-the-art dual secondary ion mass spectrometer (SIMS) aimed at innovative 3D chemical and microstructural analysis with *in operando* capabilities.

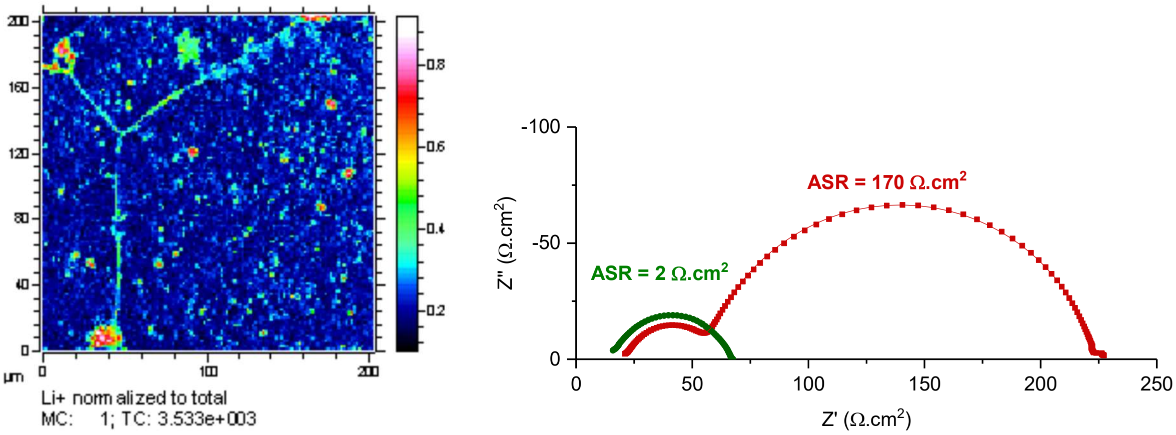

Figure 1. Visualisation of induced local chemical inhomogeneities and their correlation to electrochemical performance