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Interface growth and lithium concentration homogeneity in all solid state batteries with LLZO-LCO cathodes by neutron depth profiling

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All-solid-state lithium batteries (ASSLIBs) offer promising advantages in terms of energy and power density as well as safety over lithium-ion batteries (LIBs) with liquid electrolytes. However, they require solid electrolytes with high ionic conductivity comparable to established liquid-based systems. Urgent questions in the application of ASSLIBs are tied to the three key properties, (i) chemical stability during cell manufacturing and (ii) cycling, which can cause interface, and (iii) lithium gradient growth between the different solid phases [1, 2].

However, direct measurement and quantification of the Li distribution in LIBs and ASSLIBs is difficult or impossible with conventional methods, although it would be crucial for understanding the processes governing the key properties, stability, ionic conductivity and homogeneity of lithium distribution. Two of the most promising techniques for these investigations are neutron beam-based depth profiling (NDP) and ion beam-based particle induced x-ray (PIXE) and gamma ray emission (PIGE) spectroscopy. The non-destructive NDP method proved to be successful to investigate liquid systems [3, 4] at the FRM II neutron source in Garching. Other neutron sources have applied NDP for studying interfaces or for thin film cell investigations [5, 6]. Our partner group at IEK-1 produces oxide based ASSLIBs that show one of the best performing solid electrolytes and are thoroughly investigated using continuum modelling [7].

We study the distribution of lithium in our ceramic-based ASSLIB cells with LLZ ($\text{Li}_{6.6}\text{La}_3\text{Zr}_{1.6}\text{Ta}_{0.4}\text{O}_{12}$) as the electrolyte and separator, and LiCoO_2 as cathode active material by using both NDP and PIXE/PIGE measurements. The NDP data measured at the neutron source in NPI CAS Rez suggest that a thin interface is formed on the surface of LLZO.

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Primary author: Dr SEIDLMEYER, Stefan

Co-authors: CANNAVO, Antonino (Nuclear Physics Institute of the CAS Nuclear Physics Institute CAS (UJF)); CEC-CIO, Giovanni (Nuclear Physics Institute of the CAS Nuclear Physics Institute CAS (UJF)); SEOK, Ah-Ram (IEK-1 FZJ); FINSTERBUSCH, Martin (IEK-1 FZJ); VACÍK, Jiří (Nuclear Physics Institute of the CAS Nuclear Physics Institute CAS (UJF)); GILLES, Ralph

Presenter: Dr SEIDLMEYER, Stefan

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