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Typ: **Invited talk**

Neutrons as key technique for a better understanding of lithium and post-lithium batteries

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Li-ion batteries are key players for the energy transition and help decrease greenhouse emission owing to their storage ability. But batteries are also very complex electrochemical systems that deserve an in depth characterisations to understand their limitation during cycling. Indeed, batteries are constantly under further development and employing new type of chemistry and materials that needs to be investigated during operation i.e., in operando mode.

Neutron powder diffraction (NPD) is a technique of choice to investigate structural changes, especially for light elements like lithium. Developing an electrochemical cell dedicated to operando neutron diffraction measurements is challenging due to the large amount of electroactive materials needed and due to the incoherent neutron scattering with hydrogen, highly contributing to the background.

We report here an optimal cylindrical cell based on 18650 design1-3: i) the casing is made of transparent TiZr alloy4 and ii) several grams of active materials per electrodes. Additionally, we modified several set-up on D19 neutron diffraction beamline at ILL enabling the investigation of the electrochemical cell during relaxation processes giving information about diffusion. NCM811 vs. graphite was selected as promising full-cell battery due to the high energy density expected with this electrode couple. Figure 1 represents the neutron diffraction patterns evolution of both phases along the first (de)lithiation. As can be seen both the stages in graphite lithiation (peak (002)) and the insertion reaction of NCM811 (peak (003)) can be easily followed and analysed by mean of Rietveld refinement.

In another example, we used neutron imaging to follow the Li diffusion processes by using Li isotopes ^6Li and ^7Li . By monitoring fast charge behaviour in Li-rich cathode materials vs. Li metal, we demonstrated that the electrode engineering is a key parameter to optimise to allow fast charging protocol. Coupling experimental data to modelling reveals the hindrance of several Li transport pathway for electrocoupling fast charge and high-power features. Similar examples will be presented on solid state batteries.

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