



Contribution ID: 58

Type: not specified

## Probing Structures, Dynamics and Reactions in Electrolytes and Energy Systems by complementary neutron scattering techniques across extended scales and conditions

Wednesday 5 June 2024 09:00 (40 minutes)

Energy conversion and storage devices as proton-exchange membrane fuel cells (PEMFC) or Li-ion batteries (LIB) are complex electrochemical cells composed of an electrolyte sandwiched by two electrodes. The layered geometry allows ion fluxes through the electrolyte from one electrode to another. Ions also diffuse inside the porous structures of conductive layers (e.g. catalytic layers in PEMFC, positive/negative microstructured electrodes in LIB) where electrochemical reactions take place (oxygen reduction in a fuel cell and conversion/intercalation reaction in a LIB cathode, typically). While the fuel cell converts the chemical energy into electrical current by using a continuous flux of reactants, the battery stores the chemical energy in the host structures, with ions shuttling back and forth during charges/discharges cycles along the battery lifetime. In both energy devices, understanding the basic working and degradation mechanisms across an extended range of length and time scales is key to optimize performance and durability.

Neutron techniques are beneficial to non-destructively study the bulk properties of active material and/or individual components, as well as their interfaces, and also monitor the dynamics in the operating/cycling systems. The so-called operando experiments are enabling to measure a quantity of interest and its transformations/evolutions during PEMFC or LIB function –for instance, host material structures & nanostructures, species states and distribution, etc [1]. These quantities can be measured in representative conditions using custom operando cells, or even commercial cells in some cases, in fresh and also in aged systems, hence enabling to understand the origins of performance loss.

In this talk we will emphasize recent developments of neutron imaging and microbeam small angle scattering methods that are employed, including correlatively, to study water management in PEMFC [2,3] and lithiation heterogeneities in LIBs [4]. We will also show the advances in multi-resolution QENS to uncover the characteristics of ion mobility in polymer systems capable to conduct protons, hydroxides, hydrides or lithium ions [5,6].

[1] D. Atkins, et al. Accelerating Battery Characterization Using Neutron and Synchrotron Techniques: Toward a Multi-Modal and Multi-Scale Standardized Experimental Workflow. *Advanced Energy Materials*, 12, (17), 2102694, 2022.

[2] Martinez, N. et al. Combined Operando High Resolution SANS and Neutron Imaging Reveals in-Situ Local Water Distribution in an Operating Fuel Cell. *ACS Appl. Ener. Mat.* 2, 8425–8433, 2019.

[3] Lee, J. et al. Neutron imaging of operando proton exchange membrane fuel cell with novel membrane. *J Power Sources* 496, 2021.

[4] E. Lübke, et al. The Origins of Critical Deformations in Cylindrical Silicon Based Li-Ion Batteries. *Energy & Envir. Science*, 2024, in review.

[5] F. Foglia et al, Decoupling polymer, water and ion transport dynamics in ion-selective membranes for fuel cell applications. *Journal of Non-Crystalline Solids*: X 13 100073, 2022

[6] Takeiri, F. et al. Hydride-ion-conducting K<sub>2</sub>NiF<sub>4</sub>-type Ba–Li oxyhydride solid electrolyte. *Nature Materials*, 21, 325–330, 2022.

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