Neutron-Diffraction: Elucidating Diffusion Pathways and Activation Barriers in Lithium-Ion Conductors

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Sensors, electrolytes, energy storage systems: the fields of application for solid-state ion conductors are diverse. Scientific, as well as economic, interest in them is still on the rise. High-temperature neutron diffraction gives access to averaged positions of the atomic nuclei and their precise displacements, even when dealing with notoriously elusive chemical species like lithium ions. Notably, the high cost and ethical implications of such experiments, which are predominantly conducted at nuclear research reactors, demand the best possible evaluation (*cf.* Fig. 1).

If they are of adequate quality, data acquired from single-crystal or powder diffraction permit modeling of anharmonic thermally activated displacement. This enables the visualization of diffusion pathways and the determination of associated migration barriers *via* evaluation of probability-density function (PDF) and effective one-particle potential (OPP) [1, 2].

But what about low-quality datasets or model failure? In these cases, an examination of the scattering-length density reconstructed via maximum-entropy methods (MEM) may at least yield semiquantitative results [3]. As a supplement, heuristic means, or with difficult data, topological analyses are helpful. Within them, the framework of static species is searched for voids that are accessible for mobile ions. Representations of the procrystal-void surface provide quick insight into possible pathways, whereas Voronoi-Dirichlet partitioning (VDP) allows classifying them according to their suitability for certain ions [4].

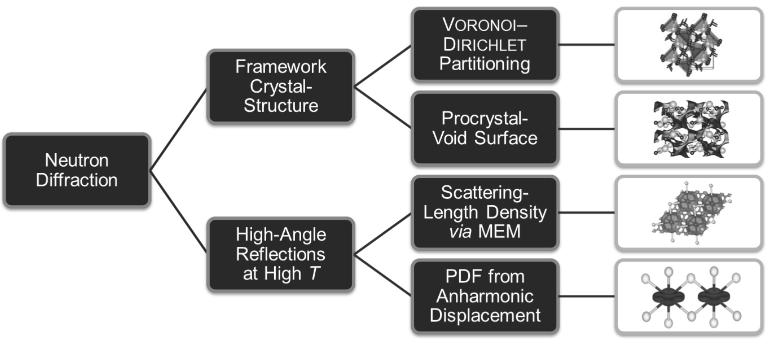

Figure 1: Methods for evaluation of neutron-diffraction data with different quality demands.

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