

Bridging PEFC Catalyst Microstructure and Macroscopic Lifetime-Performance in Modeling and Experimental Parameterization

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Electrochemical energy devices like fuel cells, electrolyzers or batteries harness porous electrodes with complex heterogeneous microstructures to optimize the interplay between the transport processes and electrochemical reaction. The use of nano-to-microstructured electrodes increases performance as well as catalyst utilization. However, such electrode structures are also more susceptible to degradation and performance decline. Statistical-physical models for microstructure degradation must be interlinked to macrohomogeneous performance models. This talk outlines the challenges in establishing this link, as depicted in Figure 1, and discusses possible solutions.

The catalyst layer of a PEFC is a complex composite electrode of Pt-based catalyst nanoparticles on a carbon substrate, ionically connected by ionomer and water as the active reaction medium. During operation, catalyst nanoparticles undergo changes due to dissolution, re-deposition of dissolved catalyst (i.e., Ostwald ripening), coagulation or inactivation of catalyst particles. The carbon substrate can undergo corrosion and the ionomer can undergo restructuring, which could affect the water retention behavior of the electrode. The changes in catalyst particle properties, especially the particle radius distribution, lead to a decrease of the overall catalyst surface available for the reaction, and thus to a performance decline. At a first glance, the coupling between microstructure and performance could be achieved by using the catalyst surface area as a scaling factor for the exchange current density. However, difficulties arise when incorporating degradation mechanisms that alter structural properties and conditions other than the catalyst particle size. For example, dealloying of catalysts like PtCo or PtNi changes both particles size and materials composition. When deconvoluting both effects in accelerated stress tests on automotive cells, we found that the change in catalyst activity, described by Tafel slope and exchange current density, correlates with the change in active surface area [1]. Changes in the ionomer morphology can lead to altered wetting behavior and liquid water retention, leaving the catalyst layer more prone to flooding [2]. Additionally, proton conductivity changes, but its description needs to take structural features of the ionomer morphology into account [3]. Overall, linking structural degradation effects to device-level performance requires finding and tailoring appropriate descriptors and structure property relationships.

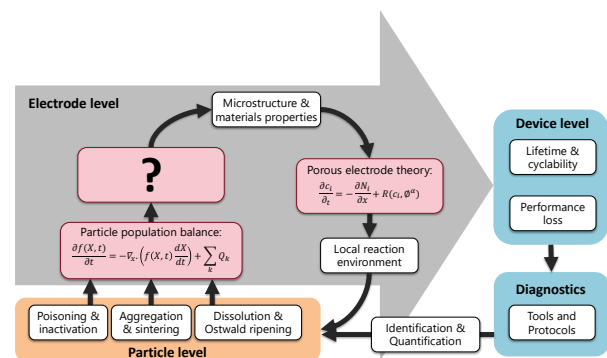


Figure 1: Hierarchical modelling framework for the degradation of particle-based electrodes. The coupling between nano-to-microscopic structure and device-level performance and lifetime is discussed in this talk.

References

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