HIGHLANDER

HIGH performing uLtrA-durable membraNe electroDe assEmblies for tRucks

HIGH PERFORMING uLTRA-DURABLE MEMBRANE ELECTRODE ASSEMBLIES FOR TRUCKS

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DELIVERABLE REPORT

D2.1: BASIC VERSION OF PERFORMANCE-DEGRADATION MODELLING FRAMEWORK FINISHED AND IMPLEMENTED INTO OPEN ACCESS MODELLING PLATFORM		
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DISSEMINATION LEVEL		
PU	*Public*	***x***
SEN	*Sensitive, limited under the conditions of the Grant Agreement;*	
NATURE OF THE DELIVERABLE		
R	*Document, report*	
DEM	*Prototype demonstrator*	
DEC	*Website*	
DMP	*Data management plan*	
OTHER	*Software, algorithms, models*	***x***

SUMMARY	
Keywords	*Degradation Model, Software, Documentation*
Abstract	*A first version of a hierarchical degradation modelling framework was formulated and implemented as software code. The code was documented and is made publicly available in an open access modelling platform. With this implementation, the conceptual workflow of the modelling work is set up and demonstrated. In the next steps, the model can be refined, extended, and parameterized. Furthermore, model-based diagnostic tools and test protocols can be built.*
Public abstract for confidential deliverables	*For a confidential deliverable, write here a public abstract to be uploaded on the project website*

REVISIONS			
Version	**Date**	**Changed by**	**Comments**
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D2.1 BASIC MODELLING FRAMEWORK IMPLEMENTATION

CONTENTS

1 INTRODUCTION

HIGHLANDER aims at developing an integrated model framework for the prediction of performance over the lifetime of a fuel cell stack. Currently, performance models exist to characterise components or cells suitably at the beginning of life. However, to fully optimise a cell for its operation over the targeted lifetime and, thereby, minimise the TCO, the performance must be monitored and analysed over the whole lifetime. Hence, HIGHLANDER targets the development of a coupled performance and degradation model. The coupled model integrates physicochemical descriptions of structure-dependent processes on all scales, i.e., it connects macroscale transport with microscopic local reaction conditions, mechanisms and rates at interfaces. The physical nature of the model ensures its predictive capabilities. With those predictive capabilities, the model enables the streamlining of materials design and screening efforts. Additionally, it will be used to link in situ observables to state-of-health indicators, which are not directly measurable, thus enabling model-based health monitoring of FC stacks and components.

2 SCOPE

The aim of WP2 is to develop a holistic, physics-based model framework for the prediction of performance over the lifetime of a fuel cell stack. This model will be provided to enable improvements in design and health monitoring of FC stack and system components. For this purpose, in D2.1. a basic version of the model, based on existing published mathematical models and their connection, was implemented as executable computer code. The computer code was made open source available, editable and extendible via a Git repository. The code was documented; example data, tutorials and manuals make it easily accessible and operable for unexperienced users.

3 DISCUSSION

The basic modelling framework is depicted in Figure 1. It consists of two nested self-consistency loops. The inner loop (black arrows) connects degradation models on the level of a single catalyst particle (described in section 3.1) with a model of the ensemble of all catalyst particles on the electrode level (Section 3.2) and a porous electrode model that describes electrode performance as well as the local reaction environment (concentrations, potential, pressure, temperature etc.) of the single particle (Section 3.3). The electrode performance feeds into the outer loop (red arrows) that assembles electrode performance and other component's losses (ohmic loss, crossover losses, thermodynamic losses) into cell performance metrics (i.e. the cell polarization curve), which then connect to diagnostic tools (ECSA loss and polarization curve fitting routines, Section 3.4).

Models on the level of catalyst particles describe different catalyst degradation mechanisms, namely dissolution and redeposition of catalyst material (so called Ostwald ripening), aggregation and sintering of particles, and inactivation of particles, e.g., due to losing electric or ionic contact or being poisoned by impurities.

On the electrode level, a particle population balance module tracks the evolution of all catalyst particles. Here, the main property in the basic model is the particle size distribution (PSD). From moments of the PSD, other relevant properties like total particle number, mean catalyst particle radius, total catalyst surface area (ECSA, electrocatalytically active surface area) and total Pt mass are calculated [Rindaldo2010,2012,2014; Urchaga2015].

Using the ECSA in a macroscopic electrode model, described with porous electrode theory [Eikerling1998,2004,2007,2014; Kulikovsy2014; Olbrich2023_1], allows calculating the overall performance of the cell over its lifetime. The model's capability of keeping track of structure, properties and performance allows developing diagnostic tools and protocols to parameterize the model [Bernhard2023].

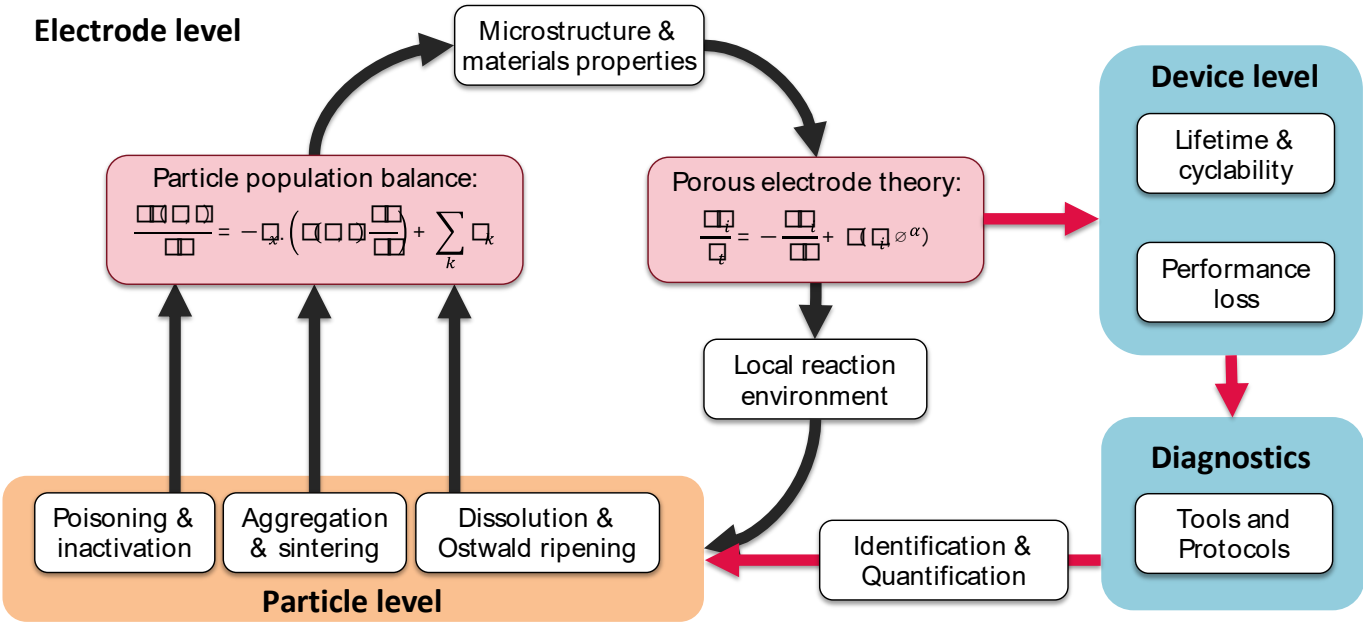

Figure 1: Basic Modelling Framework.

3.1 Particle level models

On the particle level, the basic model focusses on the Ostwald ripening mechanism, i.e. the dissolution of catalyst particles and the redeposition of dissolved particles [Rinaldo2010]. Since small particles are less stable, they dissolve faster. Large particles grow due to their enhanced rate of redeposition. Overall, the surface excess energy drives particle growth towards larger radii, which reduces the overall ECSA.

3.2 Particle population balance model

The particle population balance in the basic model focusses on the most relevant particle property: the particle radius [Rindaldo2010,2012,2014; Urchaga2015; Bernhard2023]. In principle, the particle population balance can be extended to include other particle properties, like surface tension, oxidation state, or the surface area fractions of particles due to different crystalline facets. This allows to include other or more detailed degradation mechanisms into the model if this becomes necessary in the project, e.g. due to experimental observations made, or insufficient accuracy or predictive capabilities of the model. The main material property that is calculated on the electrode level is the electrochemically active surface area, ECSA.

3.3 Porous electrode model

For the description of the cell performance, an extension of the analytical PEMFC model of Kulikovsky 2014 is used. While the analytical description of ion and oxygen transport processes are used as in the original model, the description of the electrochemical reaction is extended to include the change of the catalyst activity over the lifetime. This is achieved by modifying the exchange current density (the descriptor of catalytic activity in the model) with the ECSA calculated in the particle population model. This coupling of the degradation model with the performance model allows calculating a polarization curve (i.e., a curve describing the cell voltage as a function of cell current density), which is a fundamental descriptor of cell performance.

3.4 Fitting tools

In addition to the basic modelling framework, a first fitting tool was implemented that allows the fitting of degradation parameters (particle dissolution rate, redeposition rate, and surface tension of the particles) from measured ECSA loss curves. ECSA can be accessed in-situ experimentally, by measuring cyclic voltammograms and extracting the ECSA from the amount of underdeposited hydrogen in the low potential region of the voltammogram. With this fit, the model allows the prediction of polarization curves throughout the lifetime of the cell. Furthermore, the population balance model predicts the evolution of the particle radius distribution, a structural information that is difficult and expensive to measure (TEM imaging).
Additionally, a second fitting routine for the polarization curves was implemented, which allows for model validation and accuracy assessment, or to gain insight into the degradation mechanisms without ECSA loss information available.

3.5 Model dissemination and documentation

The model is implemented in a decentralized version control system (GitLab), which allows to monitor and document changes in the code, allows multiple people to work on the code together simultaneously, as well as creating different versions of the code (e.g., to incorporate different levels of detail in the degradation model). The GitLab repository is implemented on servers of FZJ with controlled access. The repository is open access, with a need to create a user account. This allows tracking of downloads of the code or attributing code changes to the respective developer.
The model is coded in a modular fashion, which easily allows to extend the code (e.g. new degradation models, new model based diagnostic tools or fitting routines), while the core code of the model framework remains intact. This modularisation allows keeping single parts of the code under restricted access (e.g. newly developed models, or specific values of materials parameters that partners do not wish to disseminate). Additionally, it is possible to use different licenses (different open acces or closed access licenses) for individual modules.
The model is documented with comments in the source code itself, as well as using GitLab's built-in documentation system. The model implementation is part of the open access modelling platform "JuMPER – Juelich open access Modelling Platform for Electrochemistry Research" of the Forschungszentrum Jülich GmbH. The whole modelling platform is accessible through a central internet link:

https://go.fzj.de/jumper

This central link allows moving or even changing the modelling platform if necessary without losing its published access point. It is also possible to point this link to a "landing page", i.e. a welcoming internet website that then guides the visitor to the GitLab repository.

4 CONCLUSIONS AND FUTURE WORK

Now that the fundamental workflow of the model is established and implemented, as well as the model dissemination path is set up and tested, the model can be extended using the methods described in Section 3 (modular architecture, distributed version control system, collaborative programming environment, licensing model). The focus of detailed modelling work is now to include the role of the ionomer in the catalyst layer (whereas the basic version of the degradation module only treats the catalyst particle size distribution explicitly). This work will introduce a detailed microstructure module in Figure 1 [Eikerling1998,2004,2014], which will also include a description of the wettability distribution in the catalyst layer [Olbrich2022,2023_2].
On the software side, the population balance is currently solved with a finite volume discretization scheme. However, other mathematical procedures, called method of moments are available, which can solve the population balance computationally more efficient and accurate. With this update, future optimization, analysis and data fitting routines will work faster and use less computational resources (e.g., storage, bandwidth, CPU/GPU cores). Additionally, more complex models can be solved, which is necessary in the future when including more degradation mechanisms or more detailed descriptions of mechanisms. Thus, we are implementing and testing these numerical algorithms in the next step.

5 PUBLICATIONS

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7 APPENDIX

Please find all model codes, examples, and model documentation in the Juelich Open Access Modeling Platform for Electrochemistry Research (JuMPER) at the following link:

https://go.fzj.de/jumper