

## CHARACTERIZATION, DEVELOPMENT AND EXPERIMENTAL VALIDATION OF A DYNAMIC MODEL OF AN OPEN CATHODE PEM FUEL CELL

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**RESUMEN:** *La distribución del agua en una PEMFC afecta su comportamiento y durabilidad, por tanto la comprensión de los mecanismos fundamentales del transporte del agua es imperativa y pueden ser fuertemente influenciados mediante estrategias apropiadas de control. Se ha desarrollado un modelo dinámico, no isotérmico, bidimensional de una PEMFC autohumidificada y de cátodo abierto. El modelo consta de tres submodelos usando el balance de energía, el balance de flujos y el balance másico de agua en el sistema, referidos al ánodo, cátodo, capas de difusión y la MEA. El modelo ha sido desarrollado para ser utilizado en la simulación y el estudio de los efectos del transporte y la distribución del agua en la pila y su influencia en el funcionamiento del sistema a fin de desarrollar nuevas estrategias de control. El modelo ha sido validado a partir de datos obtenidos directamente de ensayos experimentales.*

**ABSTRACT:** *Water distribution in a fuel cell affects performance and durability, thus fundamental understanding of water transport mechanisms are imperative and can be strongly influenced through proper control strategies. A two-dimensional, non-isothermal, dynamic model of an open cathode, self-humidified PEMFC was developed. The model consists of three sub models using on energy, momentum and water mass balance of the system with respect to an anode, cathode channel, diffusion layers and MEA. The model is intended to be used to simulate and study the effects of water transport/distribution and its influence on the system performance to develop new water management control strategies. Essential coefficients for water transport, namely the water diffusion, EOD, and bulk specific heat capacity coefficients, have been determined experimentally. The effective water diffusion coefficient was determined to be  $3.3 \times 10^{-8} \text{ m}^2 \text{ s}^{-1}$  at  $30^\circ\text{C}$  and increases by  $3 \times 10^{-10} \text{ m}^2 \text{ s}^{-1}$  per degree until  $50^\circ\text{C}$ . The EOD coefficient was found to be 0.47 to 0.48 water molecules per proton at stack currents from 0.044 to  $0.133 \text{ A cm}^{-2}$ . The bulk specific heat capacity for one cell unit was determined to be  $1260 \text{ J kg}^{-1} \text{ K}^{-1}$ . The model has been validated by using experimental data directly obtained from laboratory tests.*

**Keywords:** PEMFC, Water transport, EOD, Diffusion, non-isothermal

### 1. INTRODUCTION

Proton exchange membrane fuel cells (PEMFC) are gaining attention as viable energy conversion devices for a wide range of applications from automotive, stationary to portable systems. For the past 20 years astonishing progress in terms of PEMFC materials, component design, production, and system power density improvements have been achieved. However, there is still a lot to be done in the field of fuel cell controls, which makes it essential to understand the different physical phenomena within a fuel cell system and how they need to be controlled in order to improve efficiency, operating range and durability. If the water movement within a PEMFC could be controlled quickly and thus keeping optimal membrane water content and high membrane conductivity, the efficiency could be improved significantly. In order to characterize, understand and manipulate the water transport mechanisms, experimental work is needed as well as a mathematical model that describes the physical phenomena [1]. This article describes the developed dynamic model, as well as the performed experimental work and the model validation of an open cathode, self-humidified PEMFC.

### 2. SPECIFIC STACK CHARACTERISTICS

This work treats the modeling of the water transfer fluxes of the commercially available 100W PEMFC system H-100 from Horizon Fuel Cells. This open cathode system is self humidified and air cooled. It includes a cooling fan directly attached to the fuel cell housing, which removes heat from the stack by forced convection and at the same time provides oxygen to the cathode, electro-magnetic valves for hydrogen inlet and purge and a short circuit unit for applying water and heat to the cathode catalyst. Since the anode is dead-ended, a periodical hydrogen purge removes water that has crossed over from the cathode that would otherwise hinder the transport of reactant gas to the catalyst layers. With the manufactures' controller the interval of the hydrogen purge and the short circuit is independent of the stack conditions. This means that even if the stack does not require a purge or a short circuit, the system performs it anyway, which reduces efficiency. In order to increase efficiency, without reducing the robustness or operating range of the system, a broad understanding of the water transport inside the fuel cell is necessary.

### 3. MODEL DESCRIPTION

#### 3.1. Modeling objective and scheme

The developed model is used to simulate and study the effects of the dynamic control mechanisms for water management on the fuel cell performance. As water distribution and transport is dependent on temperature the model has to include not only the mass balance but also the energy balance. Since the primary objective of the model is to describe the effects of water transport, the air mass flow through the channel plays an important role. Therefore a 2D partial cross-section of the fuel cell parallel to the cathode gas flow channels is modeled. Fig. 1 shows the model geometry. The modeled sub-domains for the simulation of water propagation and distribution are the cathode flow channel, the cathode GDL and the anode GDL. To simulate the heat transfer within the cell, the Grafoil gasket and the anode bipolar plate also have to be considered, due to conductive heat transfer. The membrane is included in the anode GDL, which is denoted by the dashed line in Fig. 1. Thus, in terms of water transport and generation the more important cathode catalyst boundary still remains. Since the material of the membrane is unknown, only a bulk diffusion coefficient for the whole MEA and diffusion layers was determined experimentally.

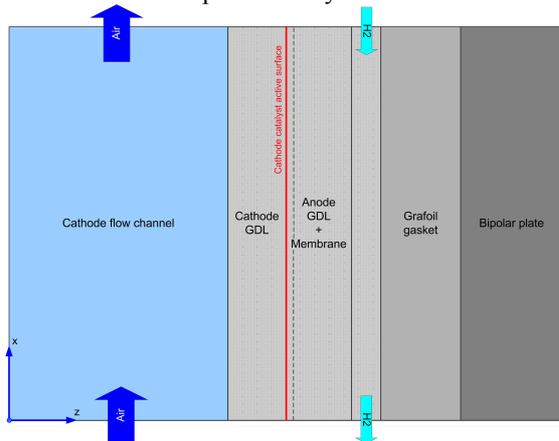


Fig. 1. Geometry of the modeled PEMFC cross section

Fig. 2 shows a schematic of the different physical phenomena that occur within the fuel cell, how they are coupled to each other and how they are treated in the model. The heart of the model is the water transport sub model, which describes the distribution of water vapor concentration in the MEA and the flow channels. In order to describe convective transport, the water transport sub model is coupled to the momentum transport sub models of anode and cathode, because the density of the reactant gases is dependent on the amount of water in the gas. Since density is also dependent on temperature, the energy transport sub model has to be included, as well. The diffusion coefficient is also a function of temperature, which links the diffusive mass transport to the temperature distribution. The water transport from anode to cathode due to the

electro-osmotic drag effect is a function of the stack current, which is set by the external load. The energy transport model, which describes the temperature distribution within the cell not only includes conductive heat transfer through the MEA and the bipolar plates, but also the convective cooling by the fan. Therefore it is coupled to the velocity field in the cathode channel, obtained by the momentum transport sub model.

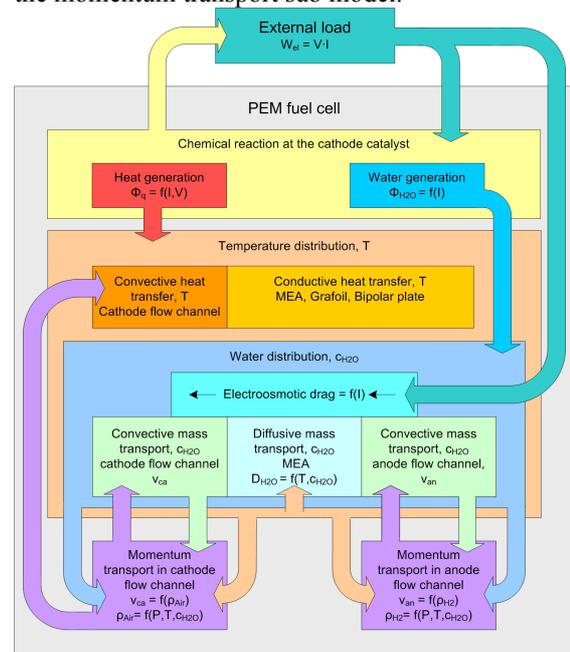


Fig. 2. Schematic of the sub models' interactions

Depending on the external load, a certain amount of heat and water is generated at the cathode catalyst surface due to the chemical reaction. This generation can be described by the stack current and voltage that is set by the external load. The resulting fluxes of water and heat are treated as an input to the water and energy transport sub model, respectively. The different sub models are explained in the following sections.

### 3.2. Energy transport sub model

The objective of the energy sub model is to predict the heat propagation and temperature distribution within the cell, which has a strong influence on the water transport model, and therefore has to be solved first. Heat is transferred by two mechanisms within the fuel cell. The cathode flow channel is dominated by forced convection due to the fan, whereas the heat transfer through the MEA, diffusion layers and bipolar plates is mainly by conduction. The fundamental law describing heat transfer is the first law of thermodynamics. Based on this law, the heat equation that describes conduction and convection within the fuel cell results in:

$$\left(\rho C_p\right) \frac{\delta T}{\delta t} + \left(\rho C_p\right) \vec{v} \cdot \nabla T = \nabla(k \cdot \nabla T) + S_e \quad (1)$$

The first term on the left-hand side describes the rate of heat generation and the second term describes the heat flux due to convection. The first term on the right-hand side accounts for conductive heat flux through the media with the thermal conductivity  $k$ , which is described by Fourier's law. The source term  $S_e$  represents heat flux due to other heat sources or sinks, which in this case is the heat flux through the catalyst surface. The velocity vector  $v$  is obtained by solving the Navier-Stokes equation for momentum transport within the cathode flow channel. Because the air density is needed to determine the velocity field and the convective heat transport, it is first calculated within the momentum transport model and then included in the heat transfer equations. This is possible because the momentum transport model of the cathode flow channel is coupled to the energy transport model. The energy model is coupled with every other sub-model because temperature has a significant effect on the fluid properties, such as densities of the reactant gases or their dynamic viscosities. Furthermore, the diffusivity of water through the MEA is a strong function of temperature as shown in the experiments of [2]. The temperature dependence of the studied fuel cell stack diffusivity has been determined experimentally (see section 4).

### 3.3 Momentum transport sub models

The objective of the momentum sub models is to provide the velocity fields in the anode and cathode flow channel, respectively, for the energy and the water transport sub model, which are used to describe convective mass and energy transport. As the air flow through the cathode channels is laminar and the pressure difference along the channels is very small, the Navier-Stokes equation for an incompressible fluid can be used to model the momentum transfer through the cathode flow channels:

$$\rho \left( \frac{\partial \vec{v}}{\partial t} + \vec{v} \cdot \nabla \vec{v} \right) = \nabla \left\{ -P \vec{I} + \mu \left[ \nabla \vec{v} + \left( \nabla \vec{v} \right)^T \right] \right\} \quad (2)$$

By solving this equation numerically, the velocity field in the cathode flow channel can be obtained, which serves to calculate convective water mass transport, and convective heat transfer through the channel, as well as convective heat transfer from the GDL surface into the channel.

The pressure driven convective flux of hydrogen through the anode GDL, which removes product water from the GDL and the catalyst layer can be described by Darcy's law:

$$\vec{v} = \frac{-\kappa}{\mu} \nabla P \quad (3)$$

where  $\kappa$  denotes the hydraulic permeability of the porous medium and  $\mu$  the dynamic viscosity of the fluid.

### 3.4 Water transport model

Since water in a fuel cell is transported by convection, diffusion and is also generated within the cell, the different equations for each transport mechanisms have been combined in a mass balance equation:

$$\frac{\partial c_{H_2O}}{\partial t} + \nabla \left( -D_{H_2O} \nabla c_{H_2O} \right) = \nabla J_{H_2O} - \vec{v} \cdot \nabla c_{H_2O} \quad (4)$$

The first term on the left-hand side of the equation corresponds to the accumulation of water in the system. The second term accounts for the diffusive transport within the MEA, described by Fick's law. The first term on the right-hand side represents a source flux of water due to the chemical reaction and also the electro-osmotic drag (EOD). Finally, the second term on the right-hand side accounts for the convective transport due to a velocity field. The EOD coefficient as well as the diffusivity of water vapor from anode to cathode is determined experimentally.

## 4. EXPERIMENTAL DETERMINATION OF COEFFICIENTS

The diffusion coefficient of water vapor through the MEA and gas diffusion layers is dependent on temperature and water content. In order to develop a mathematical relation between diffusion, temperature and water content, water diffusion has to be separated from the other water transport mechanisms, namely back diffusion and hydraulic permeation. As the experiments of Husar et al. [2] have shown, water transfer due to hydraulic permeation is at least an order of magnitude lower than that due to the two other transport mechanisms, and therefore can be neglected. To separate diffusion from the EOD, the fuel cell is disconnected from the external circuit and nitrogen is used instead of hydrogen, which also guarantees that no water can be generated due to possible leakage of the anode.

Fig. 3 shows the membrane diffusivity of water vapor at different temperatures. Compared to the work of Springer et al. [3] the experimentally obtained diffusion coefficient is smaller. This might be due to the different membrane type and thickness used in this work. However, similar results were found by McKay & Stefanopoulou [4], who also performed in-situ measurements of the diffusion coefficient with a fuel cell stack.

In order to determine the EOD coefficient experimentally, diffusive water transport through the membrane has to be minimized. This can be obtained by keeping the water concentration on both sides equal, which is achieved by setting the same

dew point temperature for the anode and cathode inlets.

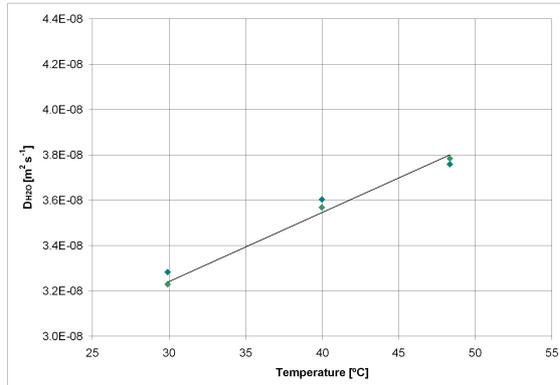


Fig. 3. Membrane Diffusivity as a function of stack temperature

The measured EOD coefficient slightly increases from 0.47 to 0.48 by increasing the stack current from 1 to 3 A, which is in accordance with the data of [2], but their test was performed at higher current densities and using a thicker membrane. To determine a specific heat capacity for the stack, a constant current is drawn for a short period of time, and the stack temperature evolution is measured. The test resulted in a specific heat capacity of  $1260 \text{ J kg}^{-1} \text{ K}^{-1}$ . A similar value is also used in the heat transfer model of He et al. [5].

### 5. MODEL VALIDATION

The model has been compared to dynamic and static experiments performed with the studied PEMFC stack. These tests have shown that the model is able to give a proper steady state and dynamic representation of the actual stack behavior.

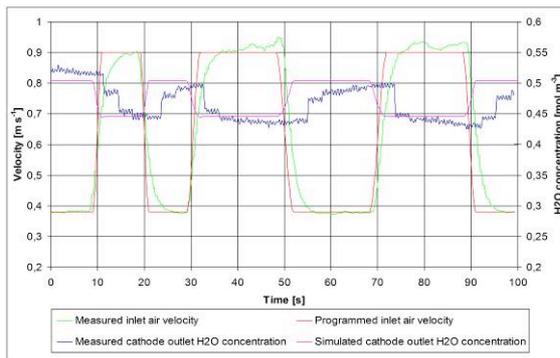


Fig. 4. Dynamic validation – cathode water concentrations at various fan velocities

Fig. 4 shows the comparison of the simulated cathode outlet water concentration with the measurement data, when changing the cooling fan velocity. The test results show that there is a delay in the cathode outlet dew point measurement, which is due to the response time of the dew point sensor used in this experiment. However, disregarding this unavoidable measurement error, the simulated

cathode outlet water concentration gives a good representation of the experimental results.

### 6. CONCLUSION

A two-dimensional, non-isothermal, dynamic model of an open cathode, self-humidified PEMFC system has been developed with respect to water and heat transport. The model is kept simple and is capable of representing system specific control mechanisms for water and heat management. As it combines most of the physical phenomena that occur within a PEMFC, it permits a comprehensive study of the effects of these mechanisms. It is based on experimental data for EOD and water diffusion directly obtained from laboratory tests with the investigated fuel cell. The model has been validated by comparing simulation results to experimental data, which has shown that the model predictions match the experimental data well. The model is easy to handle by the user-friendly CFD software COMSOL Multiphysics, and can be easily extended. However, the model can still be improved by including two phase flow characteristics and condensation/evaporation driven water transport.

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