

## THE CHARACTERISTIC THICKNESS OF POLYMER ELECTROLYTE MEMBRANE AND THE EFFICIENCY OF FUEL CELL

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### ABSTRACT

We propose a simple diffusion model of a PEM fuel cell and perform its thermodynamic analysis. Our description is based on a set of two mass balance equations involving water and proton transport through the membrane coupled with two reaction equations describing the electrochemical reactions at the electrodes. Equations for the water and proton flux densities are constructed in a linearized form suitable for the analysis from the point of view of irreversible thermodynamics. In terms of our simplified model, relations for the characteristic thickness of a PEM membrane is derived, and the maximum efficiency of a fuel cell is evaluated, both as functions of the transport properties of the PEM material.

### INTRODUCTION

Fuel cells and hydrogen technology in general are receiving an increasing attention nowadays, and not only from the scientific community and power industry, because the vision of clean, safe, and independent energy source can certainly help to win some political points as well. Although the basic principles of the fuel cell operation were discovered more than 150 years ago, there are still many open questions to be answered in this field. Our contribution presented in this article tries to apply the general irreversible-thermodynamics [3] treatment to the transport processes inside an operating fuel cell. We therefore create a simplified thermodynamic model of the fuel cell operation and perform its thorough analysis.

We tried to balance out the complexity of our model with its accuracy. The model was constructed so that it is easily (analytically) solvable, yet it is relevant to the transport processes inside the fuel cell. The crucial simplification lies in the fact that we treat the whole membrane-electrodes assembly (MEA) of the fuel cell as a single homogeneous one-dimensional electrochemical reactor, which has the transport properties (water diffusivity, proton conductivity) of a polymer-electrolyte membrane (PEM), usually Nafion<sup>TM</sup>, and also has the catalytic capabilities of both the anode and cathode layer of an ordinary fuel cell. This way our model becomes analytically solvable, and the general relations coming from irreversible thermodynamics can be applied.

There are more elaborate models of the fuel cell operation available in the literature that perform a thorough 3D modelling of a fuel cell, and which we do not intend to compete with, see e.g. [1, 2, 5, 8, 10], or a comprehensive review [13]. We hope that our contribution will help to understand what are the important factors of a successful fuel cell operation.

### NOMENCLATURE

$c$	[mol/m <sup>3</sup> ]	molar concentration
$D$	[m <sup>2</sup> /s]	diffusion coefficient
$F$	[C/mol]	Faraday constant (96485.3383)
$i$	[A/m <sup>2</sup> ]	electric current density
$j$	[mol/m <sup>2</sup> s]	molar flux density
$k$	[m <sup>-1</sup> ]	perturbation wave number
$k^{\pm}$	[m <sup>3i</sup> /mol <sup>i</sup> s]	forward/backward reaction rate, $i \in \{0, 1, \frac{3}{2}\}$
$l$	[m]	characteristic length
$L_{ij}$	[mol <sup>2</sup> /msJ]	phenomenological coefficients
$n_d$		electroosmotic drag coefficient
$q$		degree of coupling
$R$	[J/molK]	universal gas constant (8.31447215)
$t$	[s]	time variable
$T$	[K]	temperature
$w$	[mol/m <sup>3</sup> s]	reaction rate
$x$		spatial coordinate (proton flow direction)
$z$		ion number

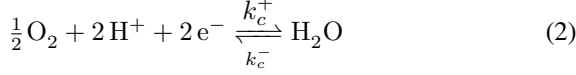
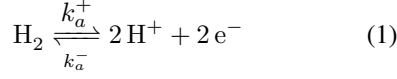
### Greek symbols

$\eta$		efficiency
$\lambda$		water content
$\mu$	[J/mol]	chemical potential
$\sigma$	[S/m]	proton conductivity
$\phi$	[V]	electrical potential
$\omega$	[s <sup>-1</sup> ]	perturbation wave frequency

### MODEL

Our simple model of the fuel cell operation is presented in this section. The model fulfills two requirements, being analyti-

cally solvable on one hand, and suitable for the application of the irreversible-thermodynamic treatment [3] on the other hand. The whole MEA, as already mentioned, is modelled as a homogeneous 1D electrochemical reactor into which hydrogen and oxygen are fed and where water and electric energy are produced. We allow the following two chemical reactions to take place within the reactor



where the subscripts  $a$  and  $c$  might imply that we describe the *anode* and *cathode* reaction of an ordinary hydrogen fuel cell. In our description, however, these two reactions take place over the whole reactor area. The corresponding reaction rates can be written according to the mass action law as

$$w_a = k_a^+ c_{\text{H}_2} - k_a^- c_{\text{H}^+}^2 \quad (3)$$

$$w_c = k_c^+ c_{\text{O}_2}^{\frac{1}{2}} c_{\text{H}^+}^2 - k_c^- c_{\text{H}_2\text{O}} \quad (4)$$

where, to further simplify the description, the backward reaction rates  $k_a^-$  and  $k_c^-$  are taken as zero.

The concentrations of hydrogen and oxygen are kept constant throughout the reactor cell,  $c_{\text{H}_2} = \bar{c}_{\text{H}_2}$ ,  $c_{\text{O}_2} = \bar{c}_{\text{O}_2}$ . The concentrations of water  $c_{\text{H}_2\text{O}}$  and concentration of protons  $c_{\text{H}^+}$  are allowed to vary across the reactor due to the chemical reactions (1, 2) and diffusion. We can write down the respective mass balance equations for water molecules and protons within the reactor

$$\frac{\partial c_{\text{H}_2\text{O}}}{\partial t} = w_c - \nabla \cdot \mathbf{j}_{\text{H}_2\text{O}} \quad (5)$$

$$\frac{\partial c_{\text{H}^+}}{\partial t} = 2w_a - 2w_c - \nabla \cdot \mathbf{j}_{\text{H}^+} \quad (6)$$

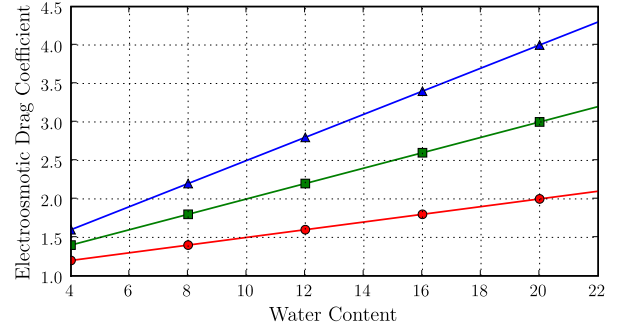
where  $\mathbf{j}_{\text{H}_2\text{O}}$  and  $\mathbf{j}_{\text{H}^+}$  are the fluxes of water molecules and protons. To be able to account for the thermodynamic coupling between the transport of water and protons within the reactor cell, we write the fluxes in the form

$$\mathbf{j}_{\text{H}_2\text{O}} = -D_{\text{H}_2\text{O}} \nabla c_{\text{H}_2\text{O}} - L_1 \nabla \phi \quad (7)$$

$$\mathbf{j}_{\text{H}^+} = -L_2 \nabla c_{\text{H}_2\text{O}} - \frac{\sigma}{F} \nabla \phi \quad (8)$$

where  $D_{\text{H}_2\text{O}}$  is the diffusion coefficient of water diffusing through the membrane,  $\sigma$  is the membrane proton conductivity, and  $F$  is the Faraday number. In the case we take  $L_1 = 0$ , Eq. (7) simplifies to the Fick's law of diffusion of water molecules. By setting  $L_2 = 0$ , Eq. (8) gives the Ohm's law for protons. The reason for including the additional parameters  $L_1 \neq 0 \wedge L_2 \neq 0$  is the possibility to take into account the coupling between the diffusion process and the electroosmotic drag of water and protons.

A measurable property that describes the above-introduced coupling phenomena is the *electroosmotic drag coefficient*  $n_d$  [12], [7], [6] which gives us the number of water molecules that are dragged along with one proton as it travels through the



**Figure 1:** Three different trial functions of the electroosmotic drag coefficient  $n_d$  that are used in our subsequent calculations (Figs. 2, 3) to illustrate the effect of varying  $n_d$ .

reactor cell. A direct consequence of this definition is that the cross-coefficient  $L_1$  takes the form

$$L_1 = \frac{\sigma}{F} n_d \quad (9)$$

describing the transport of water molecules due to the coupling with proton flow caused by the electric field  $\nabla \phi$ . The inverse effect of dragging protons along with the diffusion of water in its concentration-gradient field is introduced by taking

$$L_2 = \frac{D_{\text{H}_2\text{O}}}{\lambda} n_d \quad (10)$$

where the parameter  $\lambda = \frac{c_{\text{H}_2\text{O}}}{c_{\text{H}^+}}$  is usually called the *water content* in the literature. This definition of the water content is not very handy in our case as it does not give a constant value over the whole reactor area. The concentration of protons  $c_{\text{H}^+}$  is in our model constant, but the water concentration on the contrary is not constant as we will see later. A more suitable definition of the water content is given later in the text by Eq. (30).

The experimental data of the drag coefficient  $n_d$  available in the literature are somewhat ambiguous. Constant values around unity independent of  $\lambda$  were reported [7]. A linear dependency of  $n_d$  on  $\lambda$  (for  $\lambda > 9$ ) was reported as well [12]. A slightly different definition of the drag coefficient [6] gives even negative values of  $n_d$ . We therefore refrain from using particular experimental data of  $n_d$  and create a set of simple trial functions for use in our subsequent calculations. These trial functions depend linearly on  $\lambda$ , they differ in the slope of the linear dependency, and are plotted in Fig. 1.

Dependencies of the water diffusion coefficient  $D_{\text{H}_2\text{O}}$  and the proton specific conductivity  $\sigma$  are taken from [12].

## CHARACTERISTIC LENGTH

Let us turn our attention to the solution of the model presented in the preceding section and to its stability. The initial step towards a solution is recalling the charge conservation law which can be written as  $\nabla \cdot \mathbf{j}_{\text{H}^+} = 0$  and which assures the continuity of the proton current. Using Eq. (8) we find that

$$\Delta \phi = -\frac{FL_2}{\sigma} \Delta c_{\text{H}_2\text{O}} \quad (11)$$

Eqs. (5, 6) now possess a stationary solution (i.e.  $\frac{\partial c_{\text{H}_2\text{O}}}{\partial t} = 0$  and  $\frac{\partial c_{\text{H}^+}}{\partial t} = 0$ ) which takes the form

$$\bar{c}_{\text{H}_2\text{O}} = -\frac{k_a^+ c_{\text{H}_2}}{2D_{\text{H}_2\text{O}} \left(1 - \frac{n_d^2}{\lambda}\right)} x^2 + C_1 x + C_2 \quad (12)$$

$$\bar{c}_{\text{H}^+} = \sqrt{\frac{k_a^+ c_{\text{H}_2}}{k_c^+ c_{\text{O}_2}^{\frac{1}{2}}}} \quad (13)$$

We see that the stationary proton concentration  $\bar{c}_{\text{H}^+}$  is constant throughout the reactor cell, being a function of the reaction rates and the reactant concentrations (which are constant as well). The water concentration in the stationary case  $\bar{c}_{\text{H}_2\text{O}}$  is a polynomial function of  $x$  which is the spatial coordinate spanning from the anode side of the reactor ( $x = 0$ ) to the cathode side ( $x = l$ ). The integration constants  $C_1, C_2$  can be evaluated according to the boundary conditions  $\bar{c}_{\text{H}_2\text{O}}|_{x=0} = c_{\text{H}_2\text{O}}^a, \bar{c}_{\text{H}_2\text{O}}|_{x=l} = c_{\text{H}_2\text{O}}^c$  which give

$$C_1 = \frac{c_{\text{H}_2\text{O}}^c - c_{\text{H}_2\text{O}}^a}{l} + \frac{k_a^+ c_{\text{H}_2} l}{2D_{\text{H}_2\text{O}} \left(1 - \frac{n_d^2}{\lambda}\right)} \quad (14)$$

$$C_2 = c_{\text{H}_2\text{O}}^a \quad (15)$$

Let us now investigate the stability of the above presented stationary solution. We introduce small perturbations into the stationary solution, i.e. we write the solution in the form

$$c_{\text{H}_2\text{O}} = \bar{c}_{\text{H}_2\text{O}} + c_{\text{H}_2\text{O}}^0 \exp(kx - \omega t) \quad (16)$$

$$c_{\text{H}^+} = \bar{c}_{\text{H}^+} + c_{\text{H}^+}^0 \exp(kx - \omega t) \quad (17)$$

where  $c_{\text{H}_2\text{O}}^0$  and  $c_{\text{H}^+}^0$  are the amplitudes of the perturbations,  $x, t$  are the spatial and temporal variable. The unknown wave number  $k$  and frequency  $\omega$  will be investigated further.

The perturbed concentrations can be installed into Eqs. (5, 6), whose form simplifies thereafter to

$$0 = \left( D_{\text{H}_2\text{O}} \left(1 - \frac{n_d^2}{\lambda}\right) k^2 + \omega \right) c_{\text{H}_2\text{O}}^0 + 2k_c^+ c_{\text{O}_2}^{\frac{1}{2}} \bar{c}_{\text{H}^+} c_{\text{H}^+}^0 \quad (18)$$

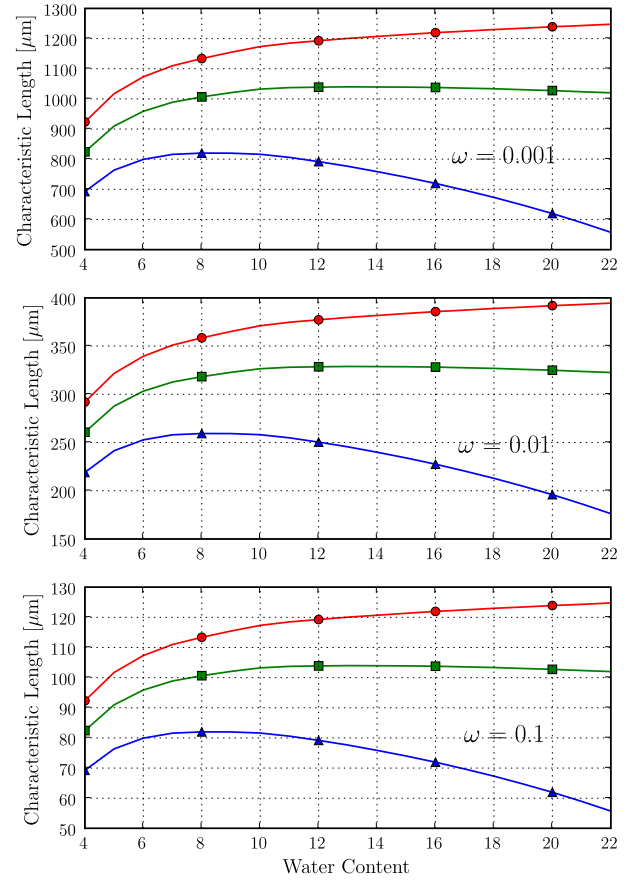
$$0 = \left( -4k_c^+ c_{\text{O}_2}^{\frac{1}{2}} \bar{c}_{\text{H}^+} + \omega \right) c_{\text{H}^+}^0 \quad (19)$$

A non-trivial solution  $c_{\text{H}_2\text{O}}^0, c_{\text{H}^+}^0$  of Eqs. (18, 19), i.e. non-zero perturbations, can be achieved only when the following two conditions are satisfied

$$\omega = 4\sqrt{k_a^+ k_c^+ c_{\text{O}_2}^{\frac{1}{2}} c_{\text{H}_2}} \quad (20)$$

$$k = \pm i \sqrt{\frac{\omega}{D_{\text{H}_2\text{O}} \left(1 - \frac{n_d^2}{\lambda}\right)}} \quad (21)$$

As a result, we see that the perturbations of our stationary solution decay with time (as  $\omega > 0$ ) and they oscillate in space, and they do not, therefore, destroy the stability of the fuel cell operation. To evaluate the decay frequency  $\omega$  (20) the knowledge of the concentrations of reactant gases and the reaction rates is required. As these are not specified in our model let us just



**Figure 2:** Characteristic length  $l$  of polymer-electrolyte membrane (22) as a function of the membrane water content  $\lambda$  evaluated for three different trial functions of the electroosmotic drag coefficient  $n_d$  (according to Fig. 1) and three different values of the perturbation decay frequency  $\omega = (0.001, 0.01, 0.1) \text{ s}^{-1}$ .

shortly elucidate the nature of  $\omega$  instead of exactly evaluating it. The frequency  $\omega$  is inversely proportional to the *damping time*  $\tau$ , i.e.  $\tau = 1/\omega$ , which gives us the time period after which the initial perturbation is damped to  $1/e$  of its initial value. This way we can find out (or, on the other hand, prescribe) on what time scale are the perturbations in our reactor damped.

It is the wave length  $4l$  of the perturbation oscillation which can be given an interesting physical meaning. The wave length is inversely proportional to the imaginary part of the wave number  $k$ , i.e.  $\text{Im}(k) = \frac{2\pi}{4l}$ , that by using Eqs. (20, 21) gives

$$l = \frac{\pi}{2} \sqrt{\frac{D_{\text{H}_2\text{O}} \left(1 - \frac{n_d^2}{\lambda}\right)}{\omega}} \quad (22)$$

which we shall call the *characteristic length* of the reactor cell.

The reason for defining the characteristic length as one fourth of the perturbation wave length is obvious as a perturbation reaches zero after travelling one fourth of its wave length. By setting the reactor size to its characteristic length we assure that, for example, a perturbation in the water pressure on the cathode will not be transferred through the membrane to the anode, but, on the contrary, such a perturbation will be effectively damped. The characteristic length of the membrane is shown in Fig. 2

as a function of the electroosmotic drag coefficient  $n_d$  and the decay frequency  $\omega$ .

## COUPLING AND EFFICIENCY

A second step in our thermodynamic treatment concerns the coupling between the transport phenomena inside the reactor cell. We therefore consider the entropy production due to the transport processes under consideration, i.e. diffusion flows and electric currents

$$\sigma(s) = - \sum_k \left( \mathbf{j}_{D,k} \cdot \nabla \left( \frac{\mu_k}{T} \right) + \frac{\mathbf{i}_k}{F} \cdot \nabla \left( \frac{z_k F \phi}{T} \right) \right) \quad (23)$$

which has the general form of a product of generalized fluxes  $(\mathbf{j}_{D,k}, \frac{\mathbf{i}_k}{F})$  and generalized forces  $(\nabla(\frac{\mu_k}{T}), \nabla(\frac{z_k F \phi}{T}))$ . According to the standard irreversible-thermodynamics procedure [11] we construct a set of linearized equations for the mass fluxes (phenomenological relations)

$$\mathbf{j}_{\text{H}_2\text{O}} = -L_{11} \nabla \mu_{\text{H}_2\text{O}} - L_{12} \nabla (F\phi) \quad (24)$$

$$\mathbf{j}_{\text{H}^+} = -L_{21} \nabla \mu_{\text{H}_2\text{O}} - L_{22} \nabla (F\phi) \quad (25)$$

where the phenomenological coefficients  $L_{ij}$  need to be further specified. Considering the chemical potential in the form  $\mu_i = \mu_{i0} + RT \ln c_i$  and comparing Eqs. (24, 25) with Eqs. (7, 8) we find that the phenomenological coefficients take the form

$$L_{11} = \frac{D_{\text{H}_2\text{O}} c_{\text{H}_2\text{O}}}{RT} \quad L_{12} = \frac{\sigma}{F^2} n_d \quad (26)$$

$$L_{21} = \frac{D_{\text{H}_2\text{O}} c_{\text{H}^+}}{RT} n_d \quad L_{22} = \frac{\sigma}{F^2} \quad (27)$$

Moreover, the cross-coefficients  $L_{12}$  and  $L_{21}$  have to obey the reciprocity relation  $L_{12} = L_{21}$  which results into

$$c_{\text{H}^+} = \frac{RT}{F^2} \frac{\sigma}{D_{\text{H}_2\text{O}}} \quad (28)$$

giving us the proton concentration in the membrane as a function of the transport properties of the PEM membrane, as opposed to Eq. (13) which expresses the proton concentration in terms of the reactant concentration and the catalyst efficiency.

Now we are able to evaluate the *degree of coupling*  $q$ , a property that characterizes the thermodynamic coupling between the transport phenomena in our model. A general definition of the degree of coupling is [11]

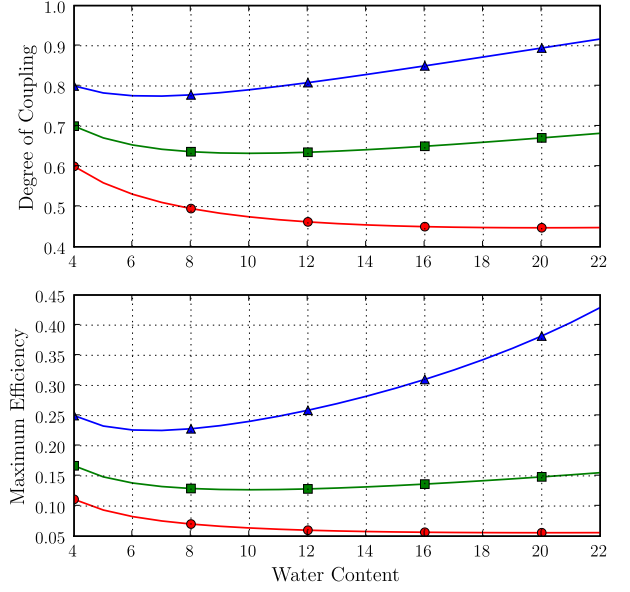
$$q = \frac{L_{21}}{\sqrt{L_{11} L_{22}}} \quad (29)$$

A non-zero coupling is a necessary requirement for the conversion of energy of one process into another and, therefore, a condition for a successful fuel cell operation in our case. If we define the water content, introduced in Eq. (10), as

$$\lambda = \frac{L_{11}}{L_{21}} n_d \quad (30)$$

then the degree of coupling takes the simple form

$$q = \frac{n_d}{\sqrt{\lambda}} \quad (31)$$



**Figure 3:** Degree of coupling  $q$  (31) as a function of the membrane water content  $\lambda$  (top graph) for three different trial functions of the electroosmotic coefficient  $n_d$  (see Fig. 1). Maximum efficiency  $\eta_{max}$  of the fuel cell operation (35) as a function of the membrane water content  $\lambda$  (bottom graph).

The definition of the water content  $\lambda$  (30) is correct from the theoretical point of view as it involves phenomenological coefficients (and the drag coefficient) that are constant throughout the reactor cell. All these coefficients depend on  $\lambda$ , and Eq. (30) therefore defines the water content in an implicit manner.

At this point, we are able to draw some conclusions about the *efficiency*  $\eta$  of a fuel cell operation as modeled by our simplified approach. The usual formula for the fuel cell efficiency<sup>1</sup> reads  $\eta = \frac{W}{\Delta G}$ , where the electric work  $W$  the fuel cell is able to perform is compared to the Gibbs energy  $\Delta G$  destroyed during the electrochemical reaction within the cell. Such a relation can be written in terms of the above-presented thermodynamic treatment as

$$\eta = \frac{\mathbf{j}_{\text{H}^+} \cdot F \nabla \phi}{-\mathbf{j}_{\text{H}_2\text{O}} \cdot \nabla \mu_{\text{H}_2\text{O}}} \quad (32)$$

where the numerator accounts for the electric work our reactor cell performs per unit length, and the denominator stands for the chemical energy that is spent per unit length of our reactor cell. By introducing the property  $y = \frac{F \nabla \phi}{\nabla \mu_{\text{H}_2\text{O}}}$ , Eq. (32) takes the form

$$\eta = - \frac{L_{21} y + L_{22} y^2}{L_{11} + L_{12} y} \quad (33)$$

and we are able to evaluate the maximum value of the efficiency by solving  $\frac{d\eta}{dy} = 0$ . The choice of  $y$  is now obvious as it frees us from dealing with the actual values of the cell voltage and the Gibbs energy of reaction, and enables us to handle the efficiency as a function of the phenomenological coefficients  $L_{ij}$  only. The

<sup>1</sup>Usually denoted as the second law efficiency, or the voltage efficiency in the literature [4].

extremal values of  $\eta(y)$  are found for

$$y_{1,2} = -\frac{L_{11}}{L_{21}} \left( 1 \pm \sqrt{1 - q^2} \right) \quad (34)$$

from which the greater one is the maximum we searched for. We can now evaluate the maximum efficiency  $\eta_{max} = \eta(y)|_{y=-\frac{L_{11}}{L_{21}}(1-\sqrt{1-q^2})}$  which gives

$$\eta_{max} = \left( \frac{1 - \sqrt{1 - q^2}}{q} \right)^2 \quad (35)$$

and we find that it is a simple function of the degree of coupling  $q$  only. The dependency of the maximum efficiency is shown in Fig. 3 as a function of the water content  $\lambda$ . Our calculated maximum efficiency  $\eta_{max}$  is somewhat lower than the maximum theoretical efficiency of a PEM fuel cell (e.g.  $\eta_{th} \sim 0.8$  at 80 °C [4]) which is, however, not equivalent to our definition of efficiency. The maximum theoretical efficiency (also known as the thermodynamic efficiency [9]) expresses the theoretical maximum electric work a fuel cell is able to provide divided by the formation enthalpy of water or, in other words, the maximum fraction of heat (that would be gained by burning the reactants) that can be converted to electrical work by the fuel cell. This efficiency was constructed in order to provide a comparison between fuel cells efficiency and Carnot cycle efficiency. However, the definition of efficiency we use above in Eq. (32) gives us the fraction of the *real* electrical work the fuel cell (as described by our simplified model) is able to provide and the maximum theoretical electrical work of the fuel cell that is given by the Gibbs free energy of formation of the product water. The real electrical work is influenced by the transport processes (and their coupling) inside the fuel cell that our model accounts for, i.e. the diffusion of water through the membrane, the membrane proton conductivity, and the drag coefficient. Precise experimental data of these thermodynamic properties and an experimentally measured efficiency of a working fuel cell are necessary to confirm that our simplified model is accurate enough in terms of efficiency, and they will be, therefore, subject of our future investigation.

## CONCLUSION

The results of our thermodynamic analysis based on a simplified model revealed some interesting aspects of a PEM fuel cell operation. First, our characteristic length (22), that theoretically assures a stable fuel cell operation, roughly coincides with the thickness of commonly-used polymer electrolyte membranes used in fuel cells. Second, we found the importance of the water content  $\lambda$ . This parameter, defined by Eq. (30), is a function of the transport properties of the membrane, and it was shown that it is related to the two parameters of the fuel cell operation – the degree of coupling  $q$ , and the maximum efficiency  $\eta_{max}$ . This way the empirical knowledge, that the fuel cell membrane has to be well-hydrated in order to obtain reasonable performance, is theoretically confirmed.

## ACKNOWLEDGEMENT

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