Water management in membrane electrolysis and options for advanced plants

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Abstract

The development of polymer electrolyte membrane electrolysis (PEMEL) is driven by increasing performance to decrease the costs of electrolysis systems. One option for increasing power density is decreasing the Ohmic losses within the cell. This can be enabled by using thinner membranes, although the disadvantage of thin membranes is their lower diffusion resistivity for water, hydrogen and oxygen what influences the efficiency and the operating conditions. In this paper the water transport and the Ohmic resistance of catalyst coated membranes with different thickness are analyzed. The disadvantage of high water permeability in thin membranes can be used to change the feed configuration in stacks and systems. It is possible to feed the electrolysis only from the cathode, which simplifies the mass transport (single phase) in the anode's porous transport layer and reducing stack and system dimensions, as well as costs.

Keywords: PEM electrolysis; Water management; Membrane resistance; Electrolyte; System setup

Introduction

In proton exchange membranes the water management plays a crucial role to achieve high power densities and a lot of effort is made especially in fuel cells to ensure a proper humidity in the membrane. This article will focus on water management of PEM electrolysis and will analyze its impact on system's setup. In PEM water electrolysis, the water is typically supplied anodically or anodically and cathodically and split by means of an electrical current at the anode electrode into oxygen, protons and electrons, while the hydrogen is produced cathodically [1]. Fundamental research has been performed on the development of electrochemically-active components with a power of a few Watt, in strong contrast to large-scale systems (30 kW to a few MWs) available on the market. The advantages of PEM electrolysis systems over other techniques are the relative simple system setup and the high power densities that can be achieved in PEM based cells and stacks. With regard to component development, catalyst-coated membranes (CCMs) are manufactured by different companies, with research groups putting great effort into the implementation of new catalyst materials and the reduction of catalyst loading [2]. As an electrolyte material, sulfonated tetrafluoroethylene-based fluoropolymers like Nafion are typically used. The properties of Nafion and the mechanisms of water and ion transport in Nafion are important for this study and a broad overview of these values is given by Mauritz and Moore [3]. All studies assume a good humidification of the membrane because of the liquid media supply. Due to the high currents the specific ionic conductivity has a crucial impact on the CCM's performance. An extensive study of the function of hydrated polymers was performed by McBrierty [4], while ionic conductivity independent of the water content was studied by Silva [5]. Additionally, Tsampas investigated the conductivity of fully hydrated Nafion membranes by means of impedance spectroscopy [6]. Suermann studied the influence of operating pressure on electrochemical cell performance and determines electrochemical parameter [7]. An important parameter in terms of the water management is the electro osmotic drag. Pivovar [8] gives an overview of electro-osmosis in fuel cell polymer electrolytes and methods to measure the drag coefficient. Zawodzinski et al. have found that the drag coefficient in PSFA membranes has a strong dependence if liquid water or vapor is used in the cells media supply [9,10]. Bussel describes the water transport mechanisms in PEM fuel cells [11] and describes the water diffusion in dependence of the water content in the membrane at 80 °C.

Water, oxygen and hydrogen transport in membranes was discussed by Schalenbach [12–14], with the influence of membrane thickness under particular consideration. Bensmann, meanwhile, has developed a method to measure the hydrogen crossover in an electrolysis cell operated with differential pressure [15]. Divisek, in turn, looked at water uptake and diffusion in Nafion membranes [16]. Beside the membrane the porous transport layer (PTL) is influencing the mass transport and the performance of the cell. Several authors studied the effect of the used materials, the structure and the porosity of the PTL on the performance of the cell [17,18]. We are extensively studying the water transport in porous transport layers, by in operando synchrotron X-ray radiography and by pressure gradient measurements at PTLs with different porosities [19]. There we have found that the water and gas transport in anodic layers with low porosities limits the cells performance. Therefore we will show an alternative configuration without multiphase flow at the anode with water supply from the cathode. A similar approach is shown in two papers related to modeling [20,21], here the limitations of the media supply from the cathode are discussed. As an overarching summary of all these effects and a discussion of major development issues Babic et al. [22] can be seen.

On the other hand the stack has to be implemented in a system, which is necessary for the media and current supply. Millet summarizes the developments from catalysis to stacks [23] and Tjarks shows the influence of MEA properties on the efficiency of whole systems [24]. Large scale PEM systems in the MW power size are developed by different companies using the standard systems setup. Some of these systems are operated with balanced pressure at anode and cathode like the company Siemens does [25–27] systems from other companies are operated at differential pressure like Protononsite, ITM, Giner and Hydrogenics [28–31] which can be advantageous in terms of systems efficiency but also means a higher mechanical stress in the membrane. The commercial systems are designed rather conservatively with regard to the set power density. Typical values are in the range of 4 W/cm^2 or lower. From the company Giner and Threadwell Cooperation oxygen generators are available that use PEM electrolysis stacks with water supply from the cathode [32].

A lot of research and development work is driven by decreasing costs. The research activities of Proton On-Site towards low cost PEM electrolysis are shown by Ayers et al. [33]. They have found that the share of the catalyst coated membrane (CCM) on the total costs is only 13% whereas the balance of plant and the PTLs have a share of 57% of the total system's costs. If it will be possible to reduce the costs of the system components or of the PTLs the impact on the total costs shall be higher than by further reducing catalyst loading of the MEA. The price is today very high (>1000 € kW^{-1}), but further cost reduction due to scaling up the manufacturing technique and of the system's size is expected. The manufactures expect a cost breakdown to about 500 € kW^{-1} in the year 2030 [34].

Here, we present a study of ion conductivity and diffusive mass transport in the membrane during real electrolysis operation at ambient pressure. In the experimental part a classic electrochemical setup that measures polarization in dependence of the current density is used to determine resistivity and water transport in the membrane. Ultimately, we introduce an alternative system design based on oxygen generating systems that allows cost reductions and system simplification, suitable to any electrolysis systems dimension.

Experimental

Membrane thickness and its influence on performance

To study the influence of the membrane thickness on cell performance and understand the effect on water transport in catalyst-coated membranes (CCMs) with different thicknesses, four types of CCMs with an active area of 17.64 cm^2 were fabricated with the following composition:

- Anode: IrO_2 with a loading of 2.25 mg cm^{-2} ± 0.25, ionomer 0.9 mg cm^{-2}
- Cathode: Pt/C with a loading of 0.8 mg cm^{-2} ± 0.1, ionomer 0.3 mg cm^{-2}

These electrodes were then hot-pressed onto different Nafion membranes by applying a defined pressure (0.5 kN cm^{-2}), at 130 °C for the thick membranes and 150 °C for the thin Nafion NR-212. Nafion 117 was chosen as the base material and, additionally, thinner membranes with thicknesses in the range of 22–127 µm were also tested. The aim of using thin membranes like Nafion NR-212 or Nafion HP was to reduce the Ohmic resistance and consequently improve the power density. In a systematic (Table 1) way, these CCMs were tested in combination with Toray paper (TGP-H-120), which has a reproducible quality, low contact resistance and good mass transport properties [35]. The Toray paper was ideal for short term testing, as the measurements are not influenced by passivation degradation processes when using Ti porous transport layers (PTLs). However, Toray paper is not suitable for durability testing due to carbon oxidation on the anode side.

Table 1 Specification of the CCMs used in the experiments, thickness is given for 23 °C and 50% relative humidity.

alt-text: Table 1

Label	Material	Thickness [μm]	Data	Anode	Cathode
Type 1	Nafion 117	183	[36]	2.25 mg cm⁻² IrO_2	0.8 mg cm⁻² Pt/C
Type 2	Nafion 115	127	[36]		
Type 3	Nafion NR-212	50.8	[37]		
Type 4	Nafion HP	22	[38]		

Different tests were performed with these CCMs. An initial electrochemical performance characterization was carried out at 80 °C, with high water flow rates at both the anode (25 ml min^{-1}) and cathode (25 ml min^{-1}) noted. The cell temperature obviously has a crucial impact on performance, and so the water was preheated before it entered the cell so that a constant cell temperature of 80 °C was ensured. To compensate temperature fluctuations affected by the electrochemical reactions, the cell temperature was also controlled by an electric heater that was integrated into the endplates. In these measurements, only current densities of up to 4 A cm^{-2} were achieved due to limitations in the power supply used. For all tests ambient pressure is used; this is the most critical condition in relation to the multiphase flow because in this case the gas fraction has its highest volume. In large scale electrolysis systems usually pressures in a range between 5 and 30 bar where used, but the higher pressure does not affect the polarization plots that much, when considering a pressure in the range of 20 bar the maximum effect on the cell voltage is an increase in the range of 60 mV [24]. When analyzing the permeation of gas (oxygen and hydrogen) the use of very thin membranes like Nafion HP is critical because of the high permeability and the risk of the formation of explosive gases [13] but when operating at ambient pressure the hydrogen concentration at the anode is always lower than 2%, this is one reason why we have used atmospheric pressure. Fig. 1 shows the polarization curves and demonstrates how strong the membrane thickness and therewith the resistance of the ion transfer influences the performance of the cell. In Fig. 2 the specific power and the efficiencies in relation to the lower heating value (LHV) of hydrogen are shown. The lower heating value (Δh_{H2}: 241.82 kJ mol^{-1}) is chosen to have comparable results to other efficiency considerations that are usually based on the LHV. The calculation is done according to the following equation, with the measured voltage U_{cell} and the Faraday constant F: 96485 As mol^{-1}:

$$\eta = \frac{\dot{n}_{H2}\,{}'h_{H2}}{I\cdot U_{cell}} = \frac{{}'h_{H2}}{2F\cdot U_{cell}} \tag{1}$$

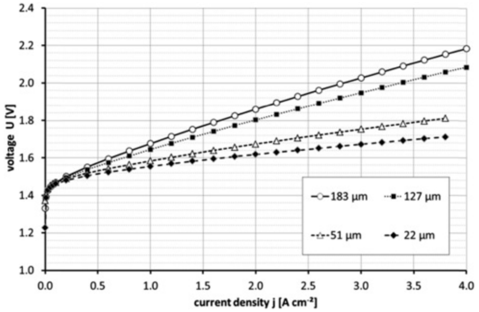


Fig. 1 Voltage (U) over current density (j) plots of CCMs with Nafion-based membranes of different thickness from 22 to 183 μm (cell area 17.64 cm^2, cell temperature 80 °C, water flow rates V_a: 25 ml min^{-1}, V_c: 25 ml min^{-1}).

alt-text: Fig. 1

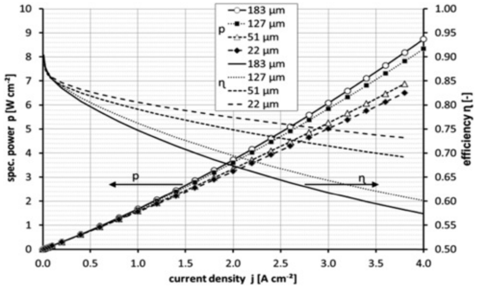


Fig. 2 Current density (j) specific power (p) and efficiency (η) plots of CCMs with Nafion-based membranes of different thicknesses from 22 to 183 μm (cell area 17.64 cm^2, cell temperature 80 °C, water flow rates V_a: 25 ml min^{-1}, V_c: 25 ml min^{-1}).

alt-text: Fig. 2

Notable power densities of up to 4 A cm^{-2} were achieved when using different Nafion membranes (see Fig. 2). At a current density of 3 A cm^{-2}, by changing the PEM thickness from 183 µm (Nafion 117) to 22 µm (Nafion HP), the efficiency of the cell can be improved by more than 10% points due to the lower resistance. The efficiency was calculated, in relation to the lower heating value, to be comparable to that of other energy converters (e.g., fuel cells, internal combustion engines, heaters, ...) against which this value is often compared. Gas crossover was neglected when operating at high current densities and ambient pressure, but further studies have shown that at low current densities (<0.5 A cm^{-2}) or high differential pressures, the hydrogen crossover that critically affects faradaic efficiency and safety operation must be considered [15,39].

CCMs fabricated with thin membranes (<50 µm) show much better performance characteristics than standard CCMs using Nafion 117 membranes. The reason for this can be better understood by a look into Table 2, in which the fit parameters of the polarization plot are shown. We used a simple linear fitting on the experimental data, resulting in the offset voltage U_0 in volts and an Ohmic resistance R in Ω cm^2. The offset voltage is in all measurements in the same range, which indicates the reproducibility of the measurements and the Ohmic resistance is linear decreasing with the membrane thickness.

Table 2 Comparison of CCMs made of different membrane materials at a temperature of 80 °C, offset voltage and Ohmic resistance calculated from linear fitting of polarization plots.

alt-text: Table 2

Label	Material	Thickness [µm] (23C, rH: 50%)	Offset Voltage U_0 [V]	Ohmic Resistance R [mΩ cm^2]
Type 1	Nafion 117	183	1.5253	166
Type 2	Nafion 115	127	1.5123	144.2
Type 3	Nafion NR-212	50.8	1.5131	79
Type 4	Nafion HP	22	1.5073	55.4

In Fig. 5, triangular symbols, the Ohmic resistance R is plotted against electrolyte thickness. These values were then fitted again linearly and the intersection with the y-axis section established as the minimum Ohmic resistance that can be reached with an infinitely thin membrane. All Ohmic losses in this case affected by the electrode and the interface to the electrode.

Water transport in catalyst coated membranes (CCMs)

Water transport in the membranes from the anode to cathode (known as electroosmotic drag; drag coefficient n_{drag}: H_2O/H^+) and in the opposite direction is also of interest when estimating the water management in a cell or a whole system. To determine the water transport through the membrane, the cell was not only operated in the conventional way, with water supply from the anode, but with water only supplied from the cathode side (see Fig. 3). To understand the resulting influence of the water management, this was investigated by running polarization curves of the different CCMs that were operated in this mode.

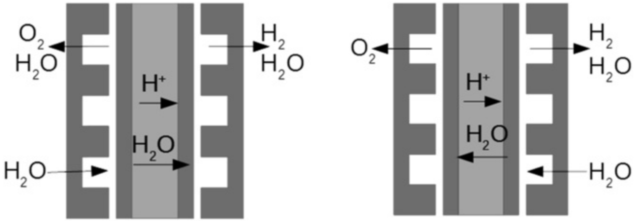


Fig. 3 Sketch of a cross-section of the setup used in test cells. Water transport in the membrane at different flow regimes is indicated by black arrows. Left: water supply at the anode; right: water supply at the cathode.

alt-text: Fig. 3

With this type of media supply, the water diffuses from the channels through the porous cathodic transport layer and membrane to the anode. If a current is applied, a certain amount of water, which can be calculated by the Faraday Law, is consumed at the anode. By further increasing the current, a strong voltage increase can be obtained when not enough water is available at the anode, known as mass transport limitation (see Fig. 4). During these measurements no liquid water at the outlets of the anode was observed.

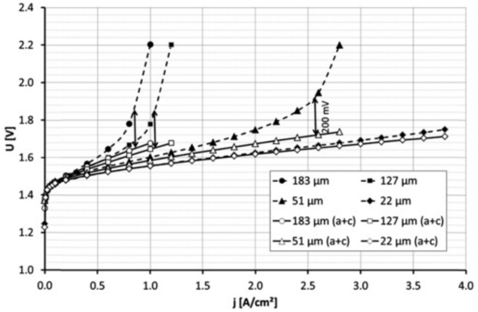


Fig. 4 Cell voltage (U) of CCMs with different membrane thickness (22–183 µm) over current density (j) plots. Filled symbols: operation without anodic but with cathodic media supply (80 °C, V_a: 0 ml min^{-1}, V_c: 25 ml min^{-1}). Unfilled symbols: cell voltage of CCM with different thickness at 80 °C and both sides fed with water (a+c) (V_a: 25 ml min^{-1}, V_c: 25 ml min^{-1}).

alt-text: Fig. 4

Afterwards, the mass transport was determined by analyzing the limiting current. To determine the limiting current in an objective manner, the polarization plots with no mass transport limitation (a+c) are used as baseline and we have defined a voltage increase of 200 mV where the mass transport limits the cell. It can also see that both polarization plots with cathodic and those with anodic and cathodic media supply have in the range of low current densities nearly the same behavior.

Thus we use the specific Ohmic resistance obtained in the low current densities regime to generate the slope of the baseline to determine the limiting current densities where we have reached mass transport problems. The limiting current in all cases is defined when an additional overvoltage of more than 200 mV is exceeded (see Fig. 4). Due to the setup we have used only limiting currents at values lower than 4 A cm^{-2} can be observed. For all measurements, these mass transport currents are listed in Table 3.

Table 3 Measured limiting current densities at different membrane thicknesses (22–183 µm) and temperatures (60–90 °C).

alt-text: Table 3

Label	Material	Thickness [µm]	Max. Current [A cm^{-2}] @ 0.2 V mass transport over potential			
			90 °C	80 °C	70 °C	60 °C
Type 1	Nafion 117	183	0.9	0.8	0.7	0.6
Type 2	Nafion 115	127	1.2	1.1	0.9	0.8
Type 3	Nafion NR-212	50.8	2.3	2.6	2.2	2.2
Type 4	Nafion HP	22	>3.8	>3.8	>3.8	>3.8

Specific resistance in catalyst coated membranes (CCMs)

Polarization plots with cathodic media supply only were analyzed and the values for the U_0 and R were fitted, similar to what was previously shown in Table 2. The values for specific resistance (R) over membrane thickness are shown in Fig. 5. The values with exclusively cathodic media supply (c) are marked with unfilled symbols, while the values shown in the first Table 2 with the supply on both sides (a+c) are marked with filled symbols.

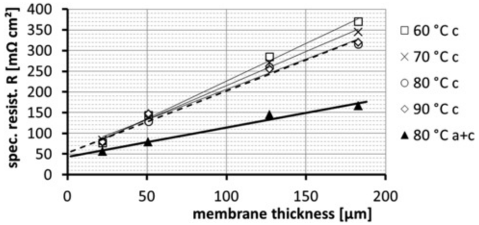


Fig. 5 Specific Ohmic resistance in dependence of membrane thickness (c: fed only by cathode, a+c: water feed at the anode and cathode) at different cell temperatures derived from current voltage plots.

alt-text: Fig. 5

Amongst the Nafion membranes tested, the lowest specific resistance in the polarization plots is in the range of 50 mΩ cm^2, with both electrodes fed with water measured at 80 °C. When comparing the resistance of the Nafion HP (thickness 22 µm) membrane with the “ideal” membrane (zero thickness) it is obvious that a further decrease in the membrane thickness will not significantly improve cell performance. The specific resistance in case of membrane thickness approaching zero is only affected by the electrode and the interface between membrane and electrode.

The so-called Ohmic losses in Fig. 5 also include losses due to the reaction kinetics at the electrodes, because they are determined by the slope of the polarization characteristics, and not by impedance spectroscopy. If the cells are only fed by the cathode, the specific resistance is 1.467 mΩ $cm^2 µm^{-1}$ at 80 °C; if both sides were fed with water, this value reduces by a factor of two, to 0.709 mΩ $cm^2 µm^{-1}$. When only feeding the membrane through the cathode, the water content in the anode electrode and the membrane is probably lower, this increases the Ohmic losses. As these measurements are done with electrodes that are optimized for a liquid-fed anode, a modification of the electrode structure regarding the water transport and design could increase performance.

Discussion

We have characterized catalyst coated membranes with different thicknesses at various temperatures with an inverse water supply, and shown the possibility of getting much more information from CCMs than with current voltage plots at only one set of operating conditions. In the following, the mass transport during electrolysis is explained and discussed. Fig. 6 (left) shows the mass flow in the cell. We have a supply with liquid water in the cathode inlet ($H_2O'_{c,liquid}$) and when the cell is operated, we generate oxygen and hydrogen by splitting the water. At the cathode's outlet, we obtain an excess of liquid water, gaseous hydrogen and water vapor at the saturation pressure. At the anode, we obtain no liquid water, but oxygen that is saturated with water vapor. The question is what happens in the membrane. There, diffusive water transport from the cathode to the anode ($H_2O_{mem.}$) occurs, driven by the water concentration gradient. On the other hand, we have an electro-osmotic driven water drag (H_2O_{drag}) from the anode to the cathode that depends on the drag factor and is associated with the proton transport (H^+) from the anode to the cathode through the membrane. Fig. 6 (right) shows a schema of the different water transport regimes in the membrane.

Fig. 6 Mass flow in the electrolysis cell with cathodic media supply (left), water flux in the membrane indicted by arrows (right).

alt-text: Fig. 6

Using Faraday's Law, the amount of water that is consumed at the anode can be calculated. The water that is transported via water drag can be considered by adding the drag coefficient to the equation:

$$\dot{m}_{a,H2O,consum} = \frac{j \cdot (1 + n_{drag})}{2F} M_{H2O} \tag{2}$$

where j is the current that is applied to the cell, n_{drag} is the water drag coefficient and M_{H2O} is 18 g mol^{-1} and F is the Faraday Constant, 96485 As mol^{-1}.

Besides the water consumption in the reaction and the water drag, additional water is consumed when taking the water vapor transport $\dot{m}_{H2O,a,gas}$ affected by the oxygen flow $\dot{m}_{O2,a}$ into account. This mechanism is affected by the gas temperature and the operating pressure. In our measurements oxygen pressure is ambient pressure and the temperature is the cell temperature, and the mass flow of water vapor can be calculated as follows:

$$\dot{m}_{O2,a} = \frac{\mathrm{j}}{4F} M_{O2} \tag{3}$$

$$\dot{m}_{H2O,a,gas} = \dot{m}_{O2,a} x \tag{4}$$

$$x = \frac{M_{H2O}}{M_{O2}} \frac{p_{s,H2O}}{p - p_{s,H2O}} \tag{5}$$

where M_{H2O} is 18 g mol^{-1} and oxygen M_{O2} has 32 g mol^{-1}. The values of x for ideal gas vapor mixtures are listed in the following table (see Table 4).

Table 4 Temperature T, saturation pressure of water $p_{s,H2O}$ and water loading x in relation to dry oxygen.

alt-text: Table 4

T [°C]	$P_{s,H2O}$ [kPa]	x
60	19.94	0.140
70	31.19	0.255
80	47.39	0.507
90	70.14	1.321

Fig. 7 shows how the water transport is depending on membrane thickness at different temperatures. The assumption is that at the maximum current, j_{max}, at the anode water is consumed for the electrochemical reaction and the minimal drag coefficient, n_{drag}, is close to 1 because we have a vapor equilibrated membrane. The drag factor of one is given by Ref. [9] and it is used in the same manor by Myles [20] for the analysis of the water transport in oxygen generator operated at elevated pressures. Also Bussel et al. have shown that the water drag is one when we assume a membrane water content (H_2O/SO_3) from 1 up to 9 [11]. If this assumption is not valid, this means the diffusive driven water transport is higher, therefor diffusive water flows at drag coefficients from 1.5, 2.0 and 2.5 are also drawn into this diagram as dotted lines. For liquid equilibrated membranes the drag coefficient can raise up to 2.5 (see Ref. [9]).

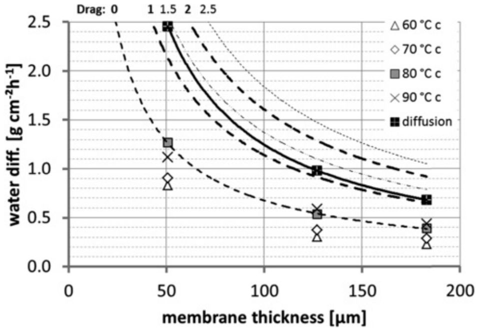


Fig. 7 Water transport in Nafion membranes over the membrane thickness in dependence of temperature (60 °C, 70 °C, 80 °C, 90 °C) and of the drag factor (1.0; 1.5; 2.0; 2.5). The black line (diffusion) shows results of the calculated diffusive water transport at 80 °C with a diffusion coefficient of 3.5 10^{-6} cm^2 s^{-1}.

alt-text: Fig. 7

When comparing the results, we found that the influence of an increase in temperature on diffusivity is relatively low, but a change of membrane thickness has a considerable impact on the water transport.

By varying the membrane thickness, it is also possible to identify the maximum potential performance of the electrodes with an ideal membrane without Ohmic losses (membrane thickness 0) and to understand the membrane's influence on the water management. Beyond the characteristic curve functional maps of CCMs can be determined that can be incorporated into larger system models.

The weak point in the mass transport analysis is that we can't determine the water transport driven by the water drag, which is in the opposite direction of the diffusive water flow. The amount of water transport affected by the water drag is important for the dimensioning of systems, because in conventional systems the drag water has to be transferred back from the cathode to the anode. We have assumed a drag coefficient of one, but unfortunately at this stage, final evidence cannot be demonstrated.

To crosscheck whether the assumptions are reasonable we calculate the water flux driven by diffusion in Nafion membranes by using Eq. (6).

$$\dot{n}_{H2O,dif} = A \cdot D \cdot \frac{'c}{'s} \quad (6)$$

The water concentration c at the liquid fed cathode is 0.055 mol cm^{-3} and at the anode we assume a water vapor pressure of 47.39 kPa (saturation pressure at 80 °C). The diffusion path length Δs is the thickness of the membranes (see Table 1). Typical diffusion coefficients at 80 °C are in the range from 2 10^{-6} cm^2 s^{-1} to 7 10^{-6} cm^2 s^{-1} depending on the membrane's water content [11]. We assume a diffusion coefficient of 3.5 10^{-6} cm^2 s^{-1} and the resulting diffusive water flux is also plotted in Fig. 7. It can be seen that the available water amount is in the range of the curve with a drag coefficient of one. So we can't determine the drag coefficient to one with absolute certainty but these results fit very well to the assumptions we have made and support the ideas we have presented.

Approach for alternative electrolysis systems setup

The setup used for the characterization of the CCMs can also be used to build up whole electrolysis systems. The cathodic water supply is used today in oxygen generators but not in hydrogen generators due to mass transport limitations when using thick standard membranes like Nafion 117 (183 µm). But with thin membranes the idea of cathodic media supply makes also sense for hydrogen generating systems and will definitely lead to further simplifications of the electrolyzer systems.

Function and set-up

The task of the classical systems components in the anodic compartment is to supply the electrolysis stack with water and transport the oxygen produced, as well as the phase separation of gaseous oxygen and liquid water (Fig. 8 left). In the cathodic compartment, the structure looks similar to the anode; a water supply is not necessary, however, as normally the water in the membrane is pulled by electro osmotic drag and in a separator, this water must be separated from the hydrogen produced [40]. Thus even in anode feed system a gas separator is necessary at the cathode to separate hydrogen and drag-water. In large systems the water is usually transferred back to the anode. The stacks in such systems need flow structures at the anode and cathode for the efficient water supply and the gas transport. This increases the production effort and drives up the costs of stacks. The analysis of the media transport in the membrane has shown that a water supply at the anode can be canceled; as water is drawn by diffusion through the membrane from the cathode to the anode. An advanced setup which takes into account this diffusive transport of water in the membrane is shown on the right side of Fig. 8. In this system, only one cathodic separator is still necessary to separate the hydrogen generated from water.

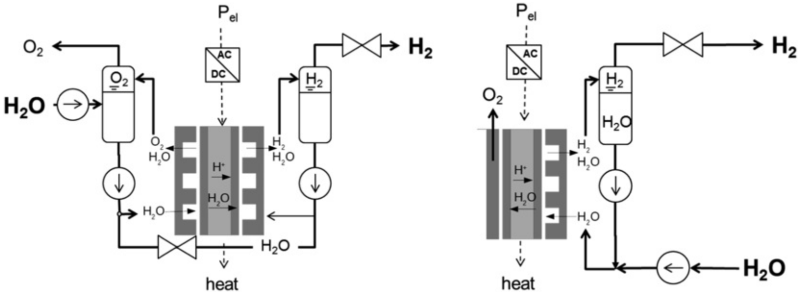


Fig. 8 Conventional system setup (left) and advanced system with feed through the cathode (right). Conventional system: stack, 2 circulation pumps, 1 water supply pump, 2 gas separators, 2 pressure regulating valves. Advanced system: stack, 1 circulation pump, 1 water supply pump, 1 gas separator, 1 pressure regulating valve.

alt-text: Fig. 8

Currently, electrolyzer stacks have an anodic and cathodic media room used for the supply of water and removal of the reaction products. The number of components required for the media supply can be significantly reduced by the present approach. Moreover, in the stack separators able to be operated with multi-phase flow are typically used on the anode and cathode sides. In the case of the design shown here, these are only necessary on the cathode side. At the anode, the multiphase flow is replaced by a single phase flow, which leads to reduced thickness of the cells and is also advantageous in case of corrosion resistance at the anode. In terms of CCM development, the further optimization of the electrodes is necessary. It should be possible to improve the anode and reach a similar specific resistance, as in the conventional-fed electrolysis.

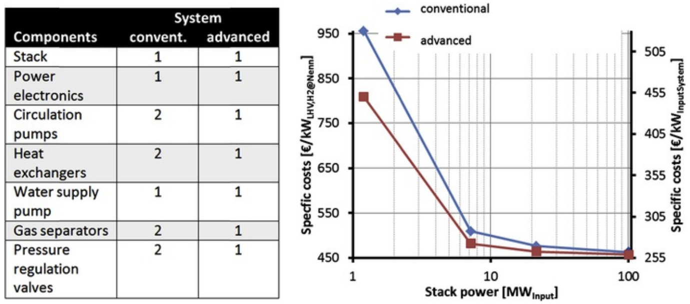
Costs

The cost distribution over components of conventional PEM electrolysis systems is given by Ayers et al. [33] and Luca et al. [34], where the stack corresponds to 53–60% and the balance of plant components to 15–32% of the total costs; 48–51% of stack's costs are mainly affected by the bipolar plates.

We have developed a complex cost model to rate the costs of all system's components and the influence of operating conditions on system's costs. When assuming the conventional setup (water supplied to anode and cathode) in our cost model the stack and the process engineering components like stack, pumps, separators and heat exchangers have a share of only 42% of the total costs. 58% of the costs are affected by the transformer and the power electronics.

For whole systems water supply only at anode or cathode means that only one circulation pump is needed but the remaining gas separator and pump have to cope with higher flow rates; so more powerful and therefore more expensive components have to be chosen. In case of the popular anodic water supply also at the cathode a gas separator is still needed to get rid of the drag water at the cathode. In systems with cathodic media supply only no gas separator at the anode is needed which means that we have in the whole system only one separator.

For the cost calculation the components shown in Fig. 8 and additional heat exchangers were considered. Fig. 9 (left) lists the components needed in conventional and advanced systems setup. Fig. 9 (right) illustrates the specific costs of the produced Hydrogen on the left y-axis and the specific costs for the installed electric power at the right y-axis over the power of the system. Thus, if the alternative setup is used the total systems costs can be reduced by 15% in case of a 1.2 MW system compared with a system where we considered media supply at anode and cathode. In larger systems this cost decrease is lower, but in case of small systems it can be even higher. Cathodic fed systems are better suited for small systems in the kW power range and at low hydrogen pressures in relation to the costs.



	System	
Components	convent.	advanced
Stack	1	1
Power electronics	1	1
Circulation pumps	2	1
Heat exchangers	2	1
Water supply pump	1	1
Gas separators	2	1
Pressure regulation valves	2	1

Fig. 9 Components that are considered in the cost calculation of system setups (left). Specific costs of conventional and advanced PEM electrolysis system over systems power (right).

alt-text: Fig. 9

In our assumptions we have neglected the additional cost decrease affected by thinner anodic structures due to the simplified one phase media transport. Also, the effects of lower corrosion problems at the anode are not considered, so a further decrease in the costs is possible when taking this into account as well. Furthermore, the systems power density will arise and therewith, the footprint of the system will be decreased, which will also lead to reduced investment (CAPEX).

A detailed analysis of the costs, potentials and drawbacks of such advanced electrolysis systems will be in the focus of another study that is currently under preparation.

Conclusions

In the present manuscript, an experimental method for measuring Ohmic losses and water transport in PEM electrolysis is discussed. The main advantage of this approach is its simple setup and the possibility of using conventional components that are already available in common electrochemical test stations. The challenge with this method are the opposite media flow directions in the porous cathodic transport layer (PTL). In these measurements, the water diffusion in the membrane was investigated by varying the membranes thickness, where a clear correlation between the membrane thickness and water transport was found. We also observed that a further reduction of membrane thickness does not significantly improve the overall performance. The lowest Ohmic resistance lies in the range of 50 mΩ cm^2 and further performance optimization can only be realized by improving the structure of the electrodes.

Another important finding is the concept of an alternative electrolysis system setup by implementing a water management based on cathodic media supply. In the conventional approach, flow field structures and corresponding manifolds are integrated into cells and stacks on both the anode and cathode sides. In the alternative concept the anodic flow structure can be simplified, due to single phase flow. With a modified water supply, the system's investment can be reduced by more than 15%, due to the lower number of components. In addition, the complexity is also reduced, which will consequently reduce the number of possible failures in the system.

By showing the limits to improving the cell performance by reducing the membrane thickness, an alternative system design was developed and studied, which will aid the electrolysis community in reaching ambitious low cost levels and in turn achieving consistent market penetration. In additional studies the durability of systems with such thin membranes should be investigated, may the thin membranes show a higher degradation due to less mechanical strength?

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Highlights

- The diffusive water transport in Nafion membranes was electrochemically assessed.
- The performance of catalyst coated membranes was improved by reducing Ohmic resistance.
- An alternative and simplified electrolysis system design for hydrogen production is presented.
- The advanced system can be around 15% cheaper than systems with cathodic and anodic media supply.

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