Master's Thesis / Bachelor's Thesis for Experimental Investigations of Thermal Diffusion in Hydrophilic Gas Diffusion Layers

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0.1 Project Description

In Polymer Electrolyte Membrane Fuel Cells (PEMFC), the gas diffusion layer (GDL) is crucial for controlling the water flow away from the catalyst layer. There needs to be a driving force for the water to move across the GDL from the electrolyte/electrode interface. The temperature gradient is one of the driving forces for water movement. [1]. Water existing in a hydrophilic structure can be considered to be in a relatively ordered state rather than water existing outside the structure. Therefore, water transport from outside of the structure's cold side to the hot side leads to an increase in total entropy and is thus thermodynamically favored [2]. In contrast, the water flow direction is from hot side to the cold side in hydrophobic structure. This difference of the movement arises from thermal diffusion. It can be defined as the flow of water from the hot side to the cold side or vice versa when both surfaces of a structure are at different temperatures [3].
The water must be evacuated from the GDL to prevent flooding. Hence, the free void volume for the diffusive flow of the gases of GDL must maintain. The GDL material must be hydrophobic to facilitate gas flow through the pores of the GDL, preventing condensation of water in the pores. In contrast, liquid water will condense in the hydrophilic GDL, which will prevent oxygen from diffusing to the cathode/electrolyte interface. This process was shown [3] in the presence of thermal diffusion flux through the hydrophilic/hydrophobic GDLs. It is shown that the thermal diffusion fluxes through the hydrophobic GDLs are obviously higher than the fluxes through the hydrophilic GDLs. However, this significant difference in fluxes is largely due to the microporous layer (MPL), which also provides hydrophobicity to the structure. This is because MPL contains 23% PTFE and smaller pore size rather than non-MPL GDLs. This smaller pore size causes high void space, high capillary pressure, and low liquid saturation. These properties make easier water vapor diffusion. In non-MPL and hydrophilic GDL, liquid water will condense in the GDL and restrict water vapor diffusion.
The other difference between hydrophobic and hydrophilic structure is the thermal diffusion coefficient's sign. A plot of thermal diffusion fluxes and temperature gradient gives then the thermal diffusion coefficients. The thermal diffusion studies, although limited, seem to agree on a conclusion that the thermal diffusion coefficient is positive for hydrophobic while it is negative for hydrophilic porous materials [3]. Furthermore, there are interesting observations in hydrophilic GDLs [4,5]. One of the observations in SGL10AA is that the water flow is highest during the initial stages of GDL which

is dynamic state, and as water saturation increases the water flow to the cold side reduces over time to a constant value reaching a steady-state state [4]. The other observation likewise demonstrated a comparable decrease in flow rate for SGL10AA and SGL10BA. In addition, it has also been observed that the flow first occurs from the hot side to the cold side, then gradually slows down, changes direction, and is eventually kept constant. Nevertheless, this last flow is significantly smaller than the flow at the initial conditions [5]. It is explicitly observed experimentally that first the dynamic state and the reaching to the steady-state state afterwards in this study as well. The decrease in this flow rate is due to the condensation of water in the macropores and then the limitation of water vapor diffusion because of the blocking of the pores. Regardless, it is reasonable to suppose that the process will vary depending on the type of GDL. Although the behavior of the thermal diffusion flow in the steady state has been studied, the flow behavior in the dynamic state in hydrophilic layers has not been studied. There is no clear finding regarding at what point and why the direction of thermal diffusion flux changes on the hydrophilic GDLs. This study aims whether the moment of the flow direction alteration and the time for reaching a steady state will change as conditions change, in other words, to investigate the dynamic state and saturation process of hydrophilic GDLs with different PTFE ratios at different temperatures and temperature differences. For this purpose, the experiments of this study will be conducted at Eskisehir Technical University, Turkey. Afterward, in the view of these experimental results, the transport and saturation processes in the hydrophilic gas diffusion layers will be explained mathematically by means of thermal diffusion coefficients at the University of Stuttgart. The mathematical model will be a continuation of [3] work and will be developed using MATLAB/Python.

0.2 Experimental Setup and Procedure

The test cell was designed to measure the thermal diffusion flow rate and flow direction at the desired temperatures (Fig. 0.1). The GDL and membrane were sandwiched between the flow channels of the test cell. The temperature of each half-cell including channels and deionized water, used during the experiments, will be kept constant at the desired temperature with the help of water circulating in the heating/cooling chamber located just behind the flow channels. While one side of the cell will be set to hot, another side will be set to the cold side. The water flow, whose flow characteristics vary depending on different parameters, is observed based on measurements on the scales connected to the tubes on both sides. Unused GDLs and membrane will be placed between the cell in Figure 0.1. The weight of the flowing water will be measured with the help of scales that will be positioned and connected to the tubes on both sides of the cell. Since the measurement will be made instantaneously, it will be possible to observe when the flow changes direction and the decrease/increase in the flow rate depending on the weight. Thermal diffusion water flux measurements within the scope of the study will be conducted with Nafion 117 membranes, and PTFE-free carbon paper, 5%, and 10% PTFE-containing carbon papers will be used as GDLs to evaluate the effect of the PTFE content for hydrophobicity. Also, the single GDL will be placed without a membrane to understand the behavior of GDL. In addition, after one experiment, the GDL will be changed to a non-used one, but the membrane will stay as a used one to see if the behavior will start from the beginning (dynamic state) or steady state. Besides, this experiment will be done by changing the membrane and not changing GDL. By this means, it may be understood the behavior of discrete single layers. Temperatures of 60 and 75ºC, and 3, 5 and 10ºC temperature differences will be planned to be done. The planned operating parameters are summarized Table 1.

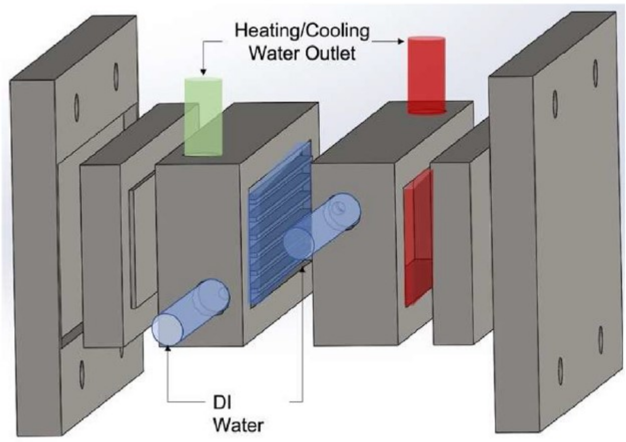

Figure 0.1: The test cell

Table 0.1: Materials investigated and conditions used

Solution	Membrane	Gas Diffusion Layer	Temperature	ΔT
Deionized water	Nafion 117	PTFE-free carbon paper	60 ºC	3 ºC
		5% PTFE-containing carbon paper		
	used Nafion 117	10% PTFE-containing carbon paper	75 ºC	5 ºC
		used PTFE-free carbon paper		10 ºC

0.3 References

[**1**] J. Benziger, J. Nehlsen, D. Blackwell, T. Brennan, J. Itescu, Water flow in the gas diffusion layer of PEM fuel cells, *Journal of Membrane Science* 261 (2005) 98–106, doi:10.1016/j.memsci.2005.03.049.
[**2**] S. Kim, M.M. Mench, Investigation of temperature-driven water transport in polymer electrolyte fuel cell: Thermo-osmosis in membranes, *Journal of Membrane Science* 328 (2009) 113–120, doi:10.1016/j.memsci.2008.11.043.
[**3**] K. Bozkurt, L. Akyalcin, S. Kjelstrup, The thermal diffusion coefficient of membrane-electrode assemblies relevant to polymer electrolyte membrane fuel cells, *International Journal of Hydrogen Energy* 48 (2023) 1501-1513, doi.org/10.1016/j.ijhydene.2022.09.302.
[**4**] S. Kim, M.M. Mench, Investigation of Temperature-Driven Water Transport in Polymer Electrolyte Fuel Cell: Phase-Change-Induced Flow, *Journal of The Electrochemical Society*, 156 (3) B353-B362 (2009)
[**5**] K. Bozkurt, Polimer Elektrolit Membran Yakit Hücrelerindeki Farkli Membran Ve Gaz Difüzyon Tabakalarinin Termo-Osmoz Ve Hidrolik Geçirgenlik Ölçümleri Ve Modellenmesi, MSc Dissertation, Eskisehir, Turkey