



The Settling Behavior of the Carbon Based Gas Diffusion Layer in PEM Fuel Cells

Examining Gas Diffusion Layers Ex-situ through Cyclic Mechanical Compression

Bachelor thesis in Chemical Engineering, 180 hp

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Cover: A SEM image of GDL A, close up on carbon fibers and MPL in an inhomogenous arrangement.

Abstract

This thesis investigate how the Gas diffusion layer in Proton Exchange Membrane Fuel Cells settle when exposed to mechanical compression with cyclic load of 4.5 MPa and 7 MPa. A Zwick Roell material tester was used to carry out the mechanical testing. 3 different state of the art gas diffusion layers was examined, they are referred to as GDL "A", "B" and "C" in this report. Three different tests were carried out, the first with 10 cycle loading, with the load hold time of 60 s. The second test was a 10 cycle loading test as well, but with the load hold time of 60 min. The third test was a 50 cycle loading, with 60 s hold. Images in a Sweeping Electron Microscope was also taken, to evaluate the fiber structure in the GDLs. The initial reason was to evaluate the fiber structure before and after compression, this method was abandoned, but the images still presented us with some valuable information about the different structures of the GDLs and could be used to discuss the results. The results imply that the load hold time effects the settling, and also confirms previous research about the so called hysteresis effect where the first loading cycle is the most damaging to the GDL structure. Further on, the 50 cycle testing show that the settling of the GDLs continues after 10 cycles and more load testing, both cyclic and static, is of interest in future research.

Keywords: PEM fuel cells, gas diffusion layer, cyclic compression, cyclic load, mechanical load, settling behaviour, scanning electron microscope.

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List of Acronyms

Below is the list of acronyms that have been used throughout this thesis listed in alphabetical order:

Battery electric vehicle
Catalyst Coated Membrane
Fuel Cell
Gas Diffusion Layer
Micro Porous Layer
Proton Exchange Membrane
Scanning Electron Microscope

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Introduction

1.1 Background

Since the 19th century the world's primary energy sources has been coal, oil and natural gas. It has gradually become clear that the emissions caused by combustion of these raw materials has led to severe negative environmental impacts. These are, higher global temperature, acidification of land and water courses and unclean air with a high concentration of harmful particles toxic to the respiratory tract in animals and humans. A conversion from fossil fuels to renewable energy sources is needed to turn this negative trend. In the last decades, focus on green energy has therefore been at focus, such as wind, hydro and solar power and biofuels. Rapid development has occured for electrical motors to vehicles, driven by charged batteries. This area is in big need of continued development and perfection, and all solutions are far from found, however, the potential is infinite in what solutions the high technology of today can bring.

One of the new, high technology energy solutions is fuel cells, powered by hydrogen gas. Fuel cells were presented as long as a century ago, but has only been a subject of commercial development since the 1980's, because of it's highly complex design and need of advanced technology. There is a number of different fuel cells, but the most popular is the PEM fuel cell. The heart of the PEM fuel cell, is the proton exchange membrane, acronymed PEM, sandwiched between electrodes made out of micropourous layers. This sandwich is called the MEA (Membrande Electrode Assembly) and is where all the electrochemical reactions take place. The MEA is supported by two bipolar plates, and this constellation is a fuel cell. In order to increase the power from fuel cells, they can be stacked on each other, this is called a fuel cell stack. Depending on what set-up the stacks will operate in and how much power that must be generated, the number of stacks is increased or decreased. The energy supply span between 5 KW to several MW and can be used in off- and on-road vehicles, maritime and aviation. The fuel cells are also a subject for a new generation of power plants, and can be used stationary to support other energy systems as well due to their potential of achieving high effects in power.

Batteries are the leading energy supplier of today's electrical vehicles. Car owners can charge their car at home, and a growing number of gas stations and parking lots offer electrical charging solutions as well. The batteries are easy to charge, and the electrical motor of a Battery Electric Vehicle (BEV) is highly functional, with a stable torque and high efficiency. The downside of electricity in general and BEV in particular is the poor ability for energy storage. Prior to charging, energy losses are inevitable during transportation and storage from power plant to socket. Here, hydrogen fuel cells have the upper hand. Hydrogen gas, H_2 , can be compressed and stored in tanks with almost zero loss. It takes not much longer time to fill up a hydrogen vehicle with H_2 compared to fossil fuels such as diesel and petrol. A hydrogen fuel cell car on the market today is filled up within 6 minutes and the only emission is liquid water. Hydrogen tanks take up a lot of space though, and a large amount of energy is needed to compress and fill the tanks with hydrogen gas. Battery electrical solutions and fuel cell solutions do not replace each other but would rather compliment each other. The problems with energy storage would for example be solved if electrical batteries could be charged by fuel cells. The electricity would not need to be stored, since the fuel cell would charge the batteries with electricity created from hydrogen that easily can be stored in tanks with minimal losses. The fuel cell industry is important for future energy storage solutions, and more research is needed in this fast growing field. Fuel cells are still young and the development is rapid, where solutions for further improvements are waiting just around the corner.

1.2 Aim of study and limitations

The purpose of this study is to understand the settling behaviour of Gas Diffusion Layers (GDLs) in Proton Exchange Membranes (PEMs) better. The experimental work will consist of mechanical cyclic loading to compress and decompress samples of GDLs in cycles. Three different, state of the art GDLs will be evaluated during this work. After the GDLs have been exerted to mechanical load, they will be analysed in a Scanning Electron Microscope (SEM). The SEM produces high resolution images of the analyzed material and hopefully the pictures will contribute to the understanding of the mechanisms behind the settling behaviour. Aspects of interest is how the fibers deform and if the connection points between the fibers increase or decrease. The results provided aim to support the definition of a method to predict the settling behaviour of the GDL and hopefully contribute to the ongoing research and work of mechanical characterisation of the GDL. This study is limited to exclusively testing of the Gas Diffusion layer, hence no other components of the MEA. Also no other fuel cell types than the PEMFC will be explained. Neither will the settling behaviour of the GDL impact on transportation and use of medium in the fuel cell be studied more that to create a bigger picture for broader understanding of the topic.

2

Theory

This work will handle the Gas Diffusion Layer, a to the size small, but functionally a crucial part of the Membrane Electrode Assembly (MEA). The GDL is a macroporous layer that enables gas transport between the bipolar plates to the Catalyst Coated Membrane (CCM). It also acts as a mechanical support to the MEA. The GDLs are between 100-400 µm thick depending on supplier[13], and the fibers are 10 um in diameter. Apart from the fibers, the GDL also contain binder, usually PTFE (Poly Tetra Flouride Ethylene) that connects the fibers, creating binding points. The GDL is an elastic material but during high stress and cyclic load it loses a lot of its elastic properties and become more and more plastic. The behaviour of when the GDL looses its elasticity is called that the materials settles. The cause of the settling behavior, and how the properties change during settling is still quite unknown. More research is needed to fully understand the GDL and its settling behavior. Most of the research made up to todays date has focused on how GDLs reacts to the first cycle of cyclic load, where most of the settling occurs according to several studies. It is less known how the GDL settle during continued cyclic loading, when most of the deformation of the fibers already has occured. This topic is important when designing and building fuel cell stacks. Since the fuel cells operate during changing environment due to temperature, relative humidity- and pressure increases and decreases, the cells are always exerted with forces that affect the load the MEA is exposed to. When assembling fuel cell stacks, an important step is to seal the stacks to preclude the gas in the cells. This procedure is called the assembling load, and exerts a big compressive load on the fuel cells. This is viewed as the first compression cycle of the fuel cell. When the fuel cell stack later is operating, in, lets say a car, the continuing cycling occurs, certainly not as big as the first compression cycle, but big enough to affect the thickness and efficiency of the GDL during lifetime operation. This has been observed from in situ life cycle testing of fuel cell stacks. More research handling the settling behaviour after the first loading cycle is hence sought-after for further understanding of this phenomenon.

2.1 Fuel Cells

2.1.1 Fuel cells compared to batteries and diesel generators

A fuel cell converts chemical energy from hydrogen or other fuels into clean electricity via electrochemical reactions. Fuel cells are unique due to the variety of their potential applications, from providing power to Watt-systems in smart phones to Maritime MegaWatts-power stations. Another beneficial property of fuel cells is the wide range of fuels they use [8]. Most fuel cells are fueled by hydrogen, either directly fed to the fuel cell or indirectly via hydrogen rich fuels such as methanol, ethanol and hydrocarbon fuel cells [9]. There are a several different fuel cell technologies. DMFCs (Direct Methanol fuel cells), AFC (Alkaline fuel cells), PAFCs (Phosphoric acidic fuel cells) and MCFCs (Molten carbonate fuel cells) are a number of them. The biggest difference between different fuel cell technologies is what electrolyte and catalyst that is used. This also affect what conditions the operation need, such as temperature, pressure and level of purity in the fuel. This thesis will solely focus on Proton Exchange membrane fuel cells (PEMFC). Fuel cells are similar to a battery in some aspects [2]. It has a positive and a negative electrode and an electrolyte. The fuel cell generates DC electricity through electrochemical reactions. But, unlike a battery, a fuel cell require constant supply of fuel and oxidant (hydrogen respectively oxygen), and the electrodes does not undergo any chemical changes. A battery generates electricity by the electrochemical reactions that involve the materials already in the battery. The battery will therefore at some point discharge, and this happens when all the materials that take part in the electrochemical reactions are depleted. A fuel cell does not discharge as long as the reactants (hydrogen and oxygen) are supplied. An even better comparison could be a diesel generator. A diesel generator produce electricity while combusting diesel, while a fuel cell produce electricity with hydrogen as fuel. The difference is that a fuel cell converts the energy of the hydrogen through electrochemical reactions with the only bi-product of water. There is no combustion inside a fuel cell, the fuel cell operates in temperatures of around 80 degrees C. Hence there is no emissions of CO, CO_2 or other greenhouse gases while operation. Nevertheless to say, the production of hydrogen is not as environmentally sustainable (See section 2.2).



2.1.2 Proton Exchange Membrane Fuel Cells

Figure 2.1: Parts of a PEMFC [10].

Proton Exchange Membrane Fuel Cells (PEMFC) is one of the most commerzialised fuel cells today [2]. A PEMFC consist of a polymer membrane sandwitched between two electrodes consisting of a catalyst layer (CL), and a gas diffusion layer (GDL). This constellation is called the Membrane electrode assembly (MEA) and the MEA is in its turn sandwiched between two flow field plates, see figure 2.1 [6]. All of this together, is one fuel cell unit. The design of the cells is identical and therefore they are modular. Just like batteries can be stacked to achieve higher voltage and power, fuel cells can be stacked. This assembly of cells is called a fuel cell stack and is graphically described in figure 2.2. By adding more cells to the stack, more power may be generated.





Why PEM fuel cells are widely popular today is because of its desirable properties [2][18]. They operate at relatively low temperatures, not higher than 60-80 degrees celcius. This makes them easier to contain and reduce thermal losses. They are also small in volume and light weighted, making them suitable for automotive and portable applications. The popularity also depends on the fact that PEM Fuel Cells has been demonstrated in almost any conceivable application. They are on the

other hand expensive in production. A fuel cell operating at low temperatures will be more sensitive to contaminated hydrogen [14]. Generally, the higher temperature the fuel cell operates in, the less is the importance of a pure fuel and noble metals. The cleaner the hydrogen, and the more noble the catalyst metals are, the more expensive is the fuel cell. The efficiency is therefore an important aspect. A fuel cell operating during low temperatures does not need much additional power to function, making it cheap in operation and effective. On the other hand, the fuel cell is in need of noble metals, such as platinum for the catalyst, and it is of importance that the hydrogen is clean [9]. Unclean hydrogen, where hydrocarbon particles and sulfur still exist after reformation, easily poison the cells by carbon monoxide since the CO binds to the Pt catalyst site. This is degrading the operation efficiency.



2.1.3 How does a fuel cell function?

Figure 2.3: A PEM fuel cell in action. The left side of the fuel cell is the anode side, and the right side is the cathode. The heart of the fuel cell is the Proton Exhange Membrane, purple in the figure [19].

Hydrogen flows in at the anode side of the cell, as can be seen visualized in figure 2.3 [32]. The hydrogen gas travels through the porous electrodes and at the catalyst site it reforms in to protons (H⁺) and electrons (e⁻). The electrochemical reaction taking place at the anode can be seen in eq. 1. The electrons travel through the external electrical circuit. The membrane is not permeable for gases but it is proton conductive, meaning the protons travel through the membrane to the cathode side. The hydrogen gas that doesn't react at the anode catalyst site does not travel through the membrane, it is instead recirculated at the anode. When the protons reach the cathode side of the cell, oxygen (O₂) from the air inlet reacts with the protons (H⁺) at the cathode catalyst site, and water is formed. Water is a byproduct

of the cell, but it has another important function as well. To enable permeation and proton conductivity the membrane must be humid. All of the water formed in the reaction at the cathode side (see eq. 2) is therefor not let out as a byproduct, but reused to create the humid environment inside the fuel cell.

$$2H_2 \rightarrow 4H^+ + 4e^-$$

$$O_2 + 4H^+ + 4e^- \rightarrow 2H_2O$$
 2

2.2 Hydrogen as a fuel

It has been estimated that around 90 % of the atoms in the universe is hydrogen atoms, but they do not exist in nature by themself [12]. To produce hydrogen gas, H_2 , the atoms need to be decoupled from other elements such as water, H_2O , or methane, CH₃; compounds found in water, plants and fossil fuels. How the production is carried out, and how the decoupling is done determines the environmental sustainability of the hydrogen energy. Hydrogen energy is separated into three categories, gray, blue and green hydrogen. Today, the majority of H_2 produced is gray hydrogen. It is made out of the process called steamed methane reforming, where a catalyst reacts to methane and high temperature steam, resulting in hydrogen and carbon dioxide. This process can be used with other hydrogen rich compounds such as propane, gasoline and coal as well. The steamed methane reforming process is powered by fossil fuels and produce byproducts resulting in 830 million metric tons of CO_2 emission each year, equal to the CO_2 emissions of The United Kingdom and Indonesia combined [5]. If the CO_2 from the steamed methane reforming process is captured and stored instead of emitted, the hydrogen produced will be labeled as blue hydrogen. Blue hydrogen therefore still leave a negative environmental footprint. When the hydrogen is produced by electrolysis of water fueled by renewable energy such as solar or wind power, it is called green hydrogen. The only byproduct is oxygen, and therefore this method is non-pollutant. Electrolysis is a technique were an otherwise non-spontaneous reaction takes place with the help of electricity. Electrolysis of water (H_20) results in two hydrogen gas molecules $(2H_2)$ and one diatomic oxygen molecule O_2 . Water Electrolysis is a very expensive method and today less than 1 % of the H₂ production results in green hydrogen. According to the International Energy Agency, 0.1~% of the hydrogen production came from water electrolysis in 2019 [24].

2.2.1 Storage of hydrogen

Hydrogen gas is the lightest molecule existing, hence it has very low density. 1 kg of hydrogen gas occupies more than 11 m^3 at room temperature and atmospheric pressure [1]. The storage of hydrogen is therefore challenging, and for it to be economically viable it needs to be stored with increased density. Several methods to store hydrogen at increased density exists with one disadvantage; all the methods

require energy input in some form of work or heat, something that is neither energynor cost effective. Today, most hydrogen is stored in gaseous or liquid form, see figure 2.4. Hydrogen stored in this way is pure and in its molecular form. In some applications such as PEM fuel cells, the purity of the hydrogen is of outmost importance while in other applications the purity is less important, for instance when the hydrogen is to be combusted with air. This opens up for a variation of other storing possibilites, such as methods were molecular hydrogen held by van der Waals bonds are absorbed onto or into a material; and atomic hydrogen can be chemically bonded to either metal hydrides or chemical hydrogen, nitrogen and oxygen.



Figure 2.4: Vertical hydrogen storage tanks.

2.2.2 Replacing fuel sources in a near future

The supply of climate friendly hydrogen fuel is believed to increase since it can be produced by excess electricity from wind turbines and solar panels [24]. This would mean that hydrogen fuel cells could solve the giant energy storage problem there is today. The demand for hydrogen fuel and fuel cell solutions is expected to increase, since it is a realistic alternative replacing fossil fuels in heavy duty vehicles, aviation and maritime in contrast to batteries [16]. Also for industrial processes, such as production of steel, cement and chemicals where it is hard or impossible to change from fossil fuels to electricity [17].

PEM fuel cells are on the verge of achieving commercialization, but are held back by concerns regarding durability [26]. Carbon-based support materials have been proposed as catalyst support for PEMFCs. They are meant to maximize the utilization of the catalytic Pt nanoparticles. However, the oxygen reduction reaction where water is formed is sluggish, and this leads to a large over-potential during the start-up and shut-down operation. Together with fuel starvation and reversal decay of electrodes, it will contribute to electrochemical oxidation of the carbon support, meaning the carbon support will start to corrode. When the carbon support start to corrode, the catalyst layer will start to decay and Pt catalyst particles will separate from the catalyst layer surface. When this happens, the Pt particles become electronically isolated and increase in size, degrading their performance. Other support materials have therefore been tested, such as inorganic based ones. These materials have shown to possess high stability toward the corrosion, but they still suffer from insufficient porosity, is less electronically conductive and offers low thermal stability, all to a higher cost. Therefore, more research is needed to develop the carbon-based supporting materials and minimize the carbon corrosion for proper utilization of catalyst and increased durability of the fuel cells.

2.3 The Membrane Electrode Assembly

The Membrane Electrode Assembly (MEA) consist of a polymer electrolyte membrane (PEM), and two porous electrodes made out of a catalyst layer (CL) and a Gas diffusion layer (GDL). The PEM is the heart of the fuel cell. It is made from polymer and has a thickness of around 500 µm. It's most important function is as a proton conductor, meaning that it must be good at transporting protons. The membrane must be chemically and mechanically stable in the fuel cell environment. The best known membrane material is Nafion, made by Dupont [27]. It is made of perfluorosulfonic acid (PFSA). On each side of the PEM, is a porous electrode. They consist of a catalyst layer, a gas diffusion layer and often but not always a micro porous layer situated between them. The MPL act as a protective layer between the GDL and the CL and enhance the electrical and thermal contact of the components. The microporous layer (MPL) is often implied as a part of the GDL, since it is coated on the surface of the GDL. By adding bipolar plates on each side of the MEA, a fuel cell is created. The electrodes is, as stated above, of porous structure and consist of the catalyst layer and the GDL. These materials are constituted of pores and solid matrix. The pores enable gas and mass transportation from the outside of the MEA, to the interface between the electrode and the membrane where all the electrochemical reactions take place, the Catalyst layer. The solid matrix ensures removal of heat and transportation of electrons and protons through the electrodes. At the catalyst layer the hydrogen molecules are split into protons and electrons. The protons are transported through the membrane whereas the electrons take an external path to the electrode on the other side of the membrane. This is where electricity is generated [2].

2.3.1 The Gas Diffusion Layer

The Gas Diffusion layer is a carbon based material, around 100-300 µm thick with carbon fibers in the sizes of 5-10 µm in diameter spread in a inhomogenous structure [25]. The two most common structures of GDLs is as cloth or on paperform. GDLs are typically of soft and brittle nature, they are elastoplastic and binders are added to the fibrous structure to increase the connection points and make the structure more stable. The GDL enable gas transport from the gas inlets at the anode and cathode and distribute reactants to the cathalyst site where the electrochemical reactions take place [22]. It also contributes as a transporter of electrons from the catalyst site to the external electrical circuit. The GDL also provide a pathway

for liquid water removal [21]. GDLs are often treated with PTFE, a hydrophobic teflon based agent. This makes the GDL more hydrophobic which is beneficial for the water transport as it prevents flooding. Flooding is the phenomenon where the water removal management is malfunctioning and the pores of the GDL get clogged with water, leading to hampering of the mass transport of gases to and from the CL through the electrodes. By treating the GDL with PTFE, the hydrophobicity of the material is enchanced.

Gas Diffusion Layers are produced comercially by several manufacturers worldwide today. The GDLs have different properties when it comes to PTFE- and binder treatment, structure, thickness, fiber diameter resulting in materials with different brittleness, elasticity and compressive behaviour.



Figure 2.5: SEM pictures of two different GDL structures. (a) Paper form, the most common structure and (b) Cloth form.

2.4 Performance of PEM Fuel Cells

2.4.1 An operating fuel cell

An operating fuel cell is always in humid state, to enable permeability over the membrane and thereby the proton conductivity from the anode to the cathode [2]. The protons are conducted over the membrane via the water molecules that are absorbed in the humid membrane. If the membrane would dry out, there would be both chemical and mechanical degrading effects. The conductivity would cease and protons would not be able to reach the cathode, therefore there would be no permeability in the cell. When the membrane dries it will start to crack, and will thereby drastically loose its low resistance abilities. Even if a dried membrane would become humid, and the permeability would emerge, the stressed membrane would have loose some of its permeability characteristics. To obtain a humid membrane, the main contributors are the diffusion of water produced as a biproduct in the cathode and humidifiers in the anode and cathode [4]. Humidifiers are expensive, and many fuel cells only contains one, usually at the cathode site. During operation, the conditions will change. Temperature, pressure and relative humidity to name some parameters will fluctuate and thereby the water content in the membrane. The membrane will because of this, so called "breath", meaning changing its thickness depending on the surrounding conditions. Only a few studies up to date have been conducted on this matter, but numbers show that a swelled membrane is $3 \mu m$ thicker than in its dried, pristing form. This may not look like a big number, but since a stack of fuel cells, say a 14 MW system, consist of around 400 cells packed together, every µm needs to be taken in account when assessing the fuel cell design.

2.4.2 The Polarization Curve

The polarization curve, also known as the IE curve, is the most fundamental and standard in situ diagnostic technique for evaluating the performance of PEM fuel cells [31]. Voltage is plotted against the current density and is recorded under steady state or dynamic conditions [3]. Polarization curves provide information on the performance losses of the fuel cell during operating conditions. There are three notable polarization losses; activation losses, ohmic losses, and mass transport losses.



Figure 2.6: a) An operating PEM fuel cell where the different compartments of a PEMFC also is shown. The electrolyte consists of the catalyst covered membrane, the anode and cathode consist of the gas diffusion layer. Fuel and Air travels in the channels of the bipolar plates. b) The polarisation curve for fuel cells [23].

The activation losses occur when the chemical processes in the cell initially have not started, and activation energy is necessary to start the reactions toward the formation of electricity and water. The Activation losses are dominant in the low current density region and arises majorly from the slow kinetics of oxygen reduction at the cathode [7]. The electrochemical reactions of a fuel cell take place at the catalyst sites. The better the catalyst, the less activation energy is required to initiate the reaction.

The ohmic losses arise of the combined resistances of the different components of the fuel cell. This includes the resistance of the ions and/or electron flow in the GDL, the membrane and the various interconnections. The voltage losses at intermediate current densities are due to ohmic losses. All ohmic losses are directly proportional to the current. This makes them the major source of loss in both the low and high temperature fuel cells.

The final type of loss is the mass transport loss. This loss occurs in both low and high temperature fuel cells, but only frequently at high electrical current densities. It is the result of transport limitation, or when the fuel cell is using fuel or oxygen to a greater extent than it can be supplied. During operation there is also a build up of water at the cathode, particularly at high electrical currents. The catalyst sites can become clogged due to this, and the inflow of oxygen is hampered. Therefore it is of importance to remove excess water, otherwise the mass transport losses will be higher.

The use of polarization curves are valuable for the systematic assessment of fuel cell operating parameters, and will support the prediction of fuel cell performance out of parameters such as flow rate of the reactant gases, relative humidity, temperature, pressure and stochiometry in the cell [3]. The curves are used when designing the fuel cell and is a guidance in selecting suitable catalysts, membranes and GDLs to obtain high PEM fuel cell performance.

Membrane Catalyst Layer R D E Gas Flow Field

2.4.3 Mechanical Degradation

Figure 2.7: The different compartments of one side of a MEA. The "teeth" mentioned in section 2.5.2 can be seen in the bottom of the figure in the Gas Flow Field. When load is applied, the teeth will start to dig into the GDL, this phenomenon is called intrusion.

Fuel cells need to endure a lot of stress during lifetime operation, both chemical and mechanical stress. The focus of this thesis will be that of mechanical stress, leading to mechanical degradation. There is two main phenomenon's occurring that contribute to the mechanical stress. First, there is the assembly stress [30]. When assembling a fuel cell stack, a assembling load of around 35 KN is applied to the stack, and the bipolar plates that encloses the MEAs push inside to the MEA. It is an inhomogeneous loading since the gas and water channels are void spaces and the landings are like teeth. This inhomogeneous loading is called intrusion, see fig. 2.7 [20]. The assembly pressure is the most damaging to the GDL structure during its lifetime. Secondly, there is the phenomenon known as the breathing of the membrane. Depending on operation parameters as pressure, humidity and temperature the membrane fluctuates, or, "breathe". When the membrane breathes, loading is exerted on the GDL, and the loading is cyclic. The mechanical stress leads to intrusion and breaking of fibers. This leads further to decrease of pore size and increase in mass transfer resistance, increased electrical resistance, flooding and clogging of pores [28][15]. All of these consequences contribute to the ohmic- and mass transport losses, (see fig. 2.6 b) leading to degradation of the fuel cell [31].



Figure 2.8: A high resolution picture taken in a scanning electron microscope (SEM). The intrusion is the phenomenon where the edges of the channels (also referred as to "teeth") of the bipolar plates press inside the GDL material. In the picture the teeth is seen in the lower left and right corners. Since the channel is a void space between the edges, the loading is inhomogenous as seen in the figure [28].

2.4.4 Previous research on the subject

This subject is not well investigated but studies have been conducted on the settling behavior of GDLs. All research indicate that the first loading cycle when doing cyclic loading testing are the most damaging to the fibers and deforms the GDL the most. During the first cycle the material loose the majority of its elastic properties. Fig. 2.9 shows the stress-strain curve for a GDL. The phenomenon of the first cycle being the most damaging to the material is called "The hysteresis effect"[28][11]. Gigos et al. [11] carried out cyclic load on the GDL, where the load range of the first 10 cycles was 0–7 MPa and the last 10 cycles was 7–12.6 MPa. As shown in Fig. 2.9, there is a hysteresis between loading cyclic and the irreversible strain increases with load range. The authors mentioned that the irreversible strain increases with the maximal load applied to the GDL and that the shapes of the two limit curves are quite similar, but they failed to explain.



Figure 2.9: The stress-strain curve with stress on the y-axis and strain on the x-axis. As can be seen, the most settling of the GDL occurs during the first cycle. Note how the space between the unloading-lines decrease (x-axis)[11].

Method

3.0.1 Equipment and materials

The mechanical loading was carried out in a testing machine from Zwick/Roell, a Z010. Picture of samples were taken in a Scanning electron microscope (SEM) and software used for this thesis was TestXpert 3 from Zwick/Roell, Excell and MatLab.

3.0.1.1 Apparatus

Zwick/Roell Z010 testing machine, Scanning Electron Microscope (SEM)

3.0.1.2 Materials

Fume Cupboard, ruler, scalpel and the three following GDLs named as A,B,C in this thesis;

State of Art GDLs					
Properties	А	В	С		
Thickness	0.25 μm	0.25 μm	180 µm		
Cross sec. Area	38 mm2	38 mm2	38 mm2		
Fiber geometry	paper	paper	paper		
Type	sheet	roll	sheet		
PTFE treated	yes	yes	yes		
MPL	yes, on one side	yes, on one side	yes, on one side		

3.0.1.3 Software

TestXpert 3 from Zwick/Roell, Excell, Matlab

3.1 Procedure

3.1.1 Preparing samples for testing

The GDLs are distributed to the site in either sheet or roll-up form. The GDLs are cut into circular samples with the help of a in house-made cutting form suitable for cutting out up to 4 samples at a time, and a hydraulic press. The more brittle the GDL is, the bigger the risk of breaking the structure in the press, therefore it is important to adjust the hydraulic pressing force in relation to what GDL is prepared, and to cut the samples one by one, instead of 4 that is more suitable for the flexible GDLs that has a more stable structure. Hence, the preparation time for the brittle GDLs are longer.

3.1.2 Test 1: 10 cycles with a hold time of 60 seconds at every compression

The three GDLs are cut into circles with an area of 38 mm². The sample is then placed between two compressive plates in a Zwick/Roell Z010 and a load of 7 MPa is exerted on the sample. A starting pressure of 0.025 MPa is applied and then the machine load up to 7 MPa at 60 µm/s. The sample is hold for 60 sec before being relaxed at 120 µm/min to the pressure of 4.5 MPa. The sample is then loaded again to 7 MPa, hold for 60 sec and relaxed to 4.5 MPa. This is performed 9 times before relaxing the sample to 0.025 MPa right before ending the test.



3.1.3 Test 2: 10 cycles with a hold time of 60 minutes at every compression

This is a 10 cycles long test, almost identical with the 10 cycles test, the difference in a hold time of 60 minutes at 7 MPa and 4.5 MPa instead of 1 minute, as in the 10 cycle-test.



3.1.4 Test 3: 50 cycles with a hold time of 60 seconds

To understand more of the settling behaviour during life time operation it is of interest to compress the GDL in many cycles ex situ. The standard test procedure is 10 cycles in this report, but to get a more understanding if the GDL show a continuing settling behaviour after 10 cycles the cycles are increased from 10 to 50. The sample is still cycled between 4.5 MPa and 7 MPa, with a hold time of 60 seconds.

3.1.5 Preparing samples for the SEM

One sample from each test is prepared for the SEM. The circular samples is cut into a rectangular shape, approximately 10x20 mm to be easier to handle during the cryo breakage procedure and to fit the SEM sample holder. The samples are placed in an sample holder specially designed for this purpose. The device is lowered down in a wide beaker inside a fume cupboard. The beaker gets filled with liquid nitrogen til most of the sample is covered in liquid. The Nitrogen gets to settle for 10 seconds, or until the worst boiling and splashing is over. The sample is then cut through with a clinical scalpel. The device is then placed in room temperature to warm up, the screws have frozen and need to thaw before the device can be handled and the sample can be collected. The side with the incision made in the cryo breaking procedure was in this case marked as an angled cut in the corner of the opposite side to easy distinguish the incision when later inserting the sample in the SEM. A total of 11 samples was prepared for the SEM. The SEM pictures is taken in the cross section of the samples. This for looking if the fibers deform, break or rearrange. This will not give any information about the fiber connection points.

Results and Discussion

4.1 Results from mechanical load testing in Zwick

4.1.1 10 cycles testing between forces 4.5 MPa and 7.0 MPa with 60 seconds hold time



Figure 4.1: The settling behaviour of GDL A, B and C respectively. The data boxes with blue colour show values of the sample when the force is 0.025 MPa. Green data boxes are for the force 4.5 MPa and red is 7.0 MPa.

GDL	Start µm End µm		total loss, ∆µm	total loss, $\Delta\%$
Α	A 174,952 ±3,91 136,498 ±2,43		38,454	21,98
в	193,418 ±1,95	114,605 ±3,21	78,813	40,75
с	180,033 ±3,04	146,852 ±2,32	33,181	18,43

Figure 4.2: Table of the start and end thickness of the different GDLs, and the total loss of thickness in both μm and %.

Figures 4.1 and 4.3 show that when relaxing the force from 7 MPa to 4.5 MPa, the thickness is regained. Meaning, the sample relaxes back as soon as the force is decreased. This together with the end thickness that is relaxed back 80 % for A and C and 60 % for B is proof that the GDL is a elastic material. The material do settle, with every compression cycle. For example, in figure 4.1, GDL B is decreasing in thickness at every 7 MPa point (red) and also when relaxed to 4.5 MPa (green) the thickness is decreasing, supporting that it settles with every compression cycle. The same goes with GDL A and C. However, the settling rate subsides after only a few cycles.

4.1.2 10 cycles testing between forces 4.5 and 7.0 MPa with 60 minutes hold time

For the test with 60 minutes hold time only testing with GDL B was conceivable since the tight time schedule, hence there are no results for GDL A and C.



Figure 4.3: The settling behavior of GDL B when load hold time is 60 minutes. 0.025 MPa have blue data lables, 4.5 MPa are green and 7.0 MPa are red.



Figure 4.4: Comparison of the start and end thickness for GDL B for 10 cycles with 60 s hold (blue) and 10 cycles with 60 min hold (red).

GDL	Hold time	Start µm	End µm	total loss, ∆µm	total loss, $\Delta\%$
в	60 s	193,418 ±1,95	114,605 ±3,21	78,813	40,75
в	60 min	193,418 ±1,95	106,603	86,815	44,88

Figure 4.5: Table of comparison of the start and end thickness of GDL B, in µm and percent. For two different tests, 10 cycles 60 s hold and 10 cycles 60 min hold.

The results show that when compressing the GDL during a longer time, it settles more (fig. 4.4). This give an insight in the rearrangement of the fibers, its a process rather than static change. The first question to ask is if the fibers break or rearrange, or both. This could possibly be due to the binder material being damaged, or mechanical chain reactions through the fiber structure. It could also be because of the powderish materials distributing along the rearrangement that contributes to more settling.

4.1.3 Change in thickness: 10 cycles compared to 50 cycles

For this test only testing with GDL C was conceivable since the tight time schedule, hence there are no results for GDL A and B.



Figure 4.6: Comparison of the start and end thickness of GDL C for 10 cycle with 60 s hold (red) to 50 cycles with 60 s hold (blue).

GDL	Cycles	Start µm	End µm	total loss, ∆µm	total loss, ∆%
с	10	180,033 ±3,04	146,852 ±2,32	33,181	18,43
с	50	180,033 ±3,04	141,629 ±0,85	38,404	21,33

Figure 4.7: Table of the comparison of the start and end thickness of GDL C for 10 cycle with 60 s hold and 50 cycles 60 s hold.

The 50 cycles test (Figure 4.6) shows that the GDL still settles after 50 cycles, even though the rate is extremely low. In future work it would be possible to determine at what point the settling should be considered complete and a method to predict the settling behaviour for different GDLs. The results obtained give the thickness of the samples directly after the compression cycles. Since the GDLs show a constant ability to regain part of their thickness when relaxed, it would be interesting to know what will happen with samples allowed to relax for a longer time. This is a interesting aspect for future work.

Conclusion wise, the results obtained and presented in section 4.1 imply that more load testing is of interest, both cyclic but also static loading. This would give a broader understanding of the settling behavior of the GDLs. The test results show (fig. 4.5) that during the 60 seconds testing, GDL B lost $\tilde{4}1$ % of thickness in total. Meanwhile, the 60 minute hold time test resulted in a total loss of $\tilde{4}5$ % in thickness, hence why static load testing would be of interest. These results should, as stated before, be supported by further testing results but they give a hint that the hold time affects the settling of the GDLs and therefor it should be investigated more.

4.2 Scanning Electron Microscope Images

The main purpose of the SEM images was from the beginning to compare pristine samples with compressed ones, to see how the fibers rearrange or get damaged from compression. Due to an inconsequent method when taking the images, such as the difference in magnification, sharpness and brightness the images are hard to compare. What can be distinguished from the images are mostly the physical attribute of the fibers and an overview of the material composition.



Figure 4.8: Cross section image of GDL A



Figure 4.9: Cross section image of GDL B



Figure 4.10: Cross section image of GDL C. The black boxes show the thickness of the sample at two points. This was calculated in the SEM.

The cross section images of GDL A, B and C contribute to a clear insight how the material is composed. The fibers of GDL A and B (fig. 4.8 and 4.9 respectively) are straight and stiff. The fibers of GDL C (fig. 4.10) on the other hand, have a curly appearance, therefore they seem more elastic and bendable. The powder on top of GDL A and B is the MPL. In GDL C the MPL seems to be more evenly distributed through the material. PTFE binder is another powderish material inside the GDLs. It does not seem to be distinguished from the MPL, hence it is difficult to draw any conclusions. What can be distinguished is though; GDL A seem to have most MPL as a coated layer on top, the same goes with GDL B, but GDL B seem to have more powder structures surrounding the fibers, making it more porous than A. This powder could be MPL or PTFE, or a mixture of both. It could be possible that the powder also contribute as an attenuator, or a soft foundation for the fibers during stress. It may protect the fibers from damaging each other by crossing over and break when loading is exerted on them. Another analytical instrument would be needed to clarify this. Concluded can though be, that GDL C does not have a MPL coated on top, but evenly spread out in the matrix. The elastic and bendable nature of C may be the factor of why there is no MPL coated on top. One of the main purposes of the MPL is to hinder fibers of the GDL from poking. Poking will mainly occur when fibers break and the risk of them breaking in GDL C seems lower than for A and B since C's fibers probably rearrange and bend rather than break when exposed to stress. The MPL is still needed for GDL C though, since other purposes of it are to enhance the electrical and thermal contact of the GDL and catalyst layer.



Figure 4.11: The fibers of GDL A (image to the left) and GDL B (right)



Figure 4.12: The fibers seem to be more elastic and bendable in GDL C.

Another source of error is found in the sample preparation for the SEM with the cryo cutting procedure. The samples are thin, small and fragile and hard to handle with care. To place a sample in the sample holder without damaging it is tricky. After pouring liquid nitrogen over the sample the cutting procedure takes place with an scalpel. This is another risk moment since the precision and action of cutting must be perfect to not damage the fiber structure and hence not getting a good incision. Figure 4.11 is a perfect example of poor sample preparation, the incision is not sharp and it looks like the structure has collapsed. Since the procedure was performed with a small set of samples and under time pressure there was not much room for mistakes nor improvement of the method. The images have on the other hand contributed with information of how the fibers look and how they are positioned in the material. GDL A and B (figure 4.11) have stiff and straight fibers in contrary to GDL C. When examining to the cyclic load test results, this becomes interesting.

The test results for 10 cycles with 60 s hold show that GDL B loose almost 41 % of its thickness whereas GDL A and C loose around 20 %. From the SEM images, the fibers of A and B are straight and stiff. They also hold more MPL and/or PTFE than C, especially B. B is also the thickest GDL, and together with loosing a lot of its thickness, a theory is that it is a very porous GDL that easily loose its thickness. GDL A and C looses almost 22 and 18,5 µm respectively, in contrast to GDL B this is not much, which is wanted when designing MEAs. The less the GDL settles, the less it will contribute to the mechanical degradation of the fuel cell. A theory could be that the reason GDL C settles the least, is because of its more soft nature. Instead of the fibers breaking during stress, they bend and rearrange, forming and adapting to their neighboring fibers. As they don't break, this could mean that the elasticity of the GDL would be retained to a greater extent. GDL C does not have a clear MPL coating as the other GDLs. The MPL is rather believed to be dispersed through the matrix. The powder content in C is overall less than A and B, from the images speaking. The inhomogenous form of the fibers in C probably fill up the void spaces in the matrix more effectively than A and B, and does not need powder as an attenuator like A and B probably does.

4. Results and Discussion

Conclusion

The SEM Images obtained did not generate the information needed to answer the question at issue. The question of how mechanical load affects the fiber structure in different GDLs, such as rearrangement or breaking of fibers, increase or decrease in connection points et cetera remains unanswered. This is mainly because of the images being taken in cross section. The cross section give information about the diameter of the fibers, thickness of sample, and possibly an insight in how the fibers break, but tell nothing about the connection points and give minimal information about the rearrangement of the fibers. Further on the images should be taken consequently, with the same magnitude and resolution. However, as stated before the images still generate valuable information such as bigger insight of the material's construction, how the MPL and PTFE forms around the fibers, and how different GDL's fibers distribute and look in µm size. They also seem to contribute some what to the understanding of how the different GDLs looses their thickness and how they settle.

Further on, conclusions can be drawn regarding the fiber structure and its influence on the thickness, settling behaviour and the relaxation behaviour of gas diffusion layers. From the results, GDL C settles the least of the three GDLs, but it is not as elastical as GDL A and B. GDL C does not relax back as much as A and B, but the hysteresis effect is also not as damaging to the thickness of C compared to A and B. Even though GDL C seems to be less elastic it's different fibrous composition seem to affect the settling behaviour in a positive manner. Whether how much the fiber composition versus the PTFE and MPL layer affect the GDL's behavior and properties needs further research to answer.

For future work the aspects of the relaxation behaviour, and the settling behaviour, and how they depend on each other needs more focus to fully understand the life cycle of the gas diffusion layer and thereof how to use it to its full potential in fuel cell operation.

5. Conclusion

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