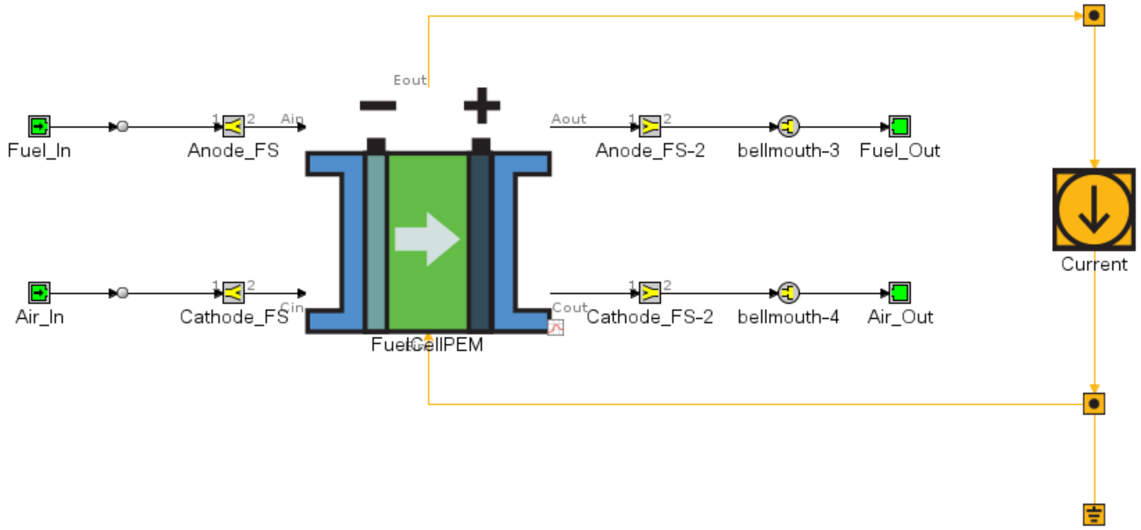

Degradation Analysis of Fuel Cells for Heavy-Duty Vehicle Applications

Bachelor thesis in Mechanics and Maritime Sciences

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Chalmers University of Technology
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Cover:
GT-Suite fuel cell model[1].

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Abstract

Climate change is one of the most pressing issues of today, and the emissions from fossil fuel-powered vehicles are major contributors. A potential solution to reduce the use of fossil fuels in vehicles is the implementation of fuel cells. However, the main challenge of using fuel cells is degradation, which results in a loss of performance. Therefore, on behalf of Volvo trucks, this project aims to model, analyze and draw conclusions about the performance and durability of a fuel cell used in trucks, with a special focus on the degradation in the catalyst layer caused by Ostwald ripening. The degradation is studied by modeling in Matlab and GT-Suite. The Matlab model studies the Ostwald ripening phenomena for two different drive cycles and outputs electrochemical surface area dependent on cell runtime. The data are then used to analyze the performance of a fuel cell stack in GT-Suite. The GT-Suite simulation calculates power output as well as polarization curves for the cell, which was done for each data point provided by the Matlab analysis. In relation to the Ostwald ripening phenomenon, a steady voltage displays a 17% loss of electrical power at 10 000 hours, in contrast, fluctuating voltage shows a 17% loss already at 6000 hours. It reveals a clear dependence on whether the voltage is steady or fluctuating. The study concludes that Ostwald ripening is a significant contributor to fuel cell degradation and needs to be taken into account for further development of the technology. Temperature and humidity have also been shown to ha an effect on fuel cell performance and need to be studied further in order for fuel cells to become a potential long-term solution for heavy-duty vehicles.

Keywords: Fuel cell, heavy-duty, degradation, Ostwald ripening phenomena.

Sammandrag

Klimatförändringen är ett av dagens mest akuta problem och utsläppen från fordon som drivs av fossila bränslen är en stor bidragande faktor. En potentiell lösning för att minska användningen av fossila bränslen i fordon är implementering av bränsleceller. Största utmaningen med bränsleceller är nedbrytning över tid, vilket leder till försämrad prestanda. Detta projektet utförs på uppdrag av Volvo Trucks och målet är att modellera, analysera och dra slutsatser om prestanda och livslängd för bränsleceller som används i lastbilar. Särskilt fokus riktas mot nedbrytning i katalysatorskiktet till följd av Ostwald ripening. Nedbrytningen studeras genom modellering i Matlab version 2022b och GT-Suite version 2022. Matlabmodellen studerar Ostwald ripeningfenomenet för två olika körcyklar och ger utdata för elektrokemisk yta beroende på bränslecellens driftstid. Datan används sedan för att analysera prestandan för en stack i GT-Suite. GT-Suitesimuleringen beräknar både effekten och polarisationskurvor för cellen, vilket gjordes för varje datapunkt som tillhandahölls av Matlab. I fallet med stabil spänning blev förlusten av elektrisk effekt 17% vid 10 000 timmar, och i fallet med växlande spänning blev förlusten 17% redan vid 6000 timmar. Detta visar på ett tydligt beroende på om spänningen är stabil eller växlande. Efter studien dras slutsatsen att ytterligare utveckling och forskning är nödvändig för att bränsleceller ska bli en potentiell långsiktig lösning för tunga fordon.

Nyckelord: Bränslecell, tunga fordon, nedbrytning, Ostwald ripening.

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Emil Nyström, Emma Olger, Stjepana Sever and Emelie Stewall

Gothenburg, May 2023

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1 Introduction

In a world increasingly affected by climate change and global warming, there is still heavy dependence on fossil fuels for transportation. According to the UN, fossil fuels remain the biggest contributors to climate change [4]. Emissions from vehicles amounted to 20.2% of all global CO_2 emissions [5]. Heavy-duty trucks are responsible for a significant proportion of transportation emissions, accounting for a staggering 23 % [6] of total emissions despite making up only 2 % of all vehicles on the road. Finding alternative solutions to combustion engines is therefore essential to build a more sustainable future.

With the economic and political pressure to reduce dependence on fossil fuels, the electric vehicle market has grown substantially in the past decade and continues to evolve. Today, nearly all electric vehicles rely on rechargeable lithium-ion batteries to power electric motors. However, the cost, weight, and long recharging times of batteries limit their use in heavy-duty vehicles such as trucks. As a result, fuel cells are being evaluated by several different actors in the industry. Fuel cells effectively convert hydrogen gas into electricity through a chemical reaction while producing heat and water vapor. They are faster to refuel, lighter, and have a higher energy density than conventional batteries[7]. Their range is up to 1000 km and recharging takes only 15 minutes, making them ideal for heavy-duty vehicles, especially in countries and regions where recharging a battery is not a possibility because of the lacking electrical infrastructure [8]. However, the degradation of the cell poses a problem and needs to be studied further in order for fuel cells to be an environmentally-friendly energy solution for heavy-duty vehicles[9].

To address this issue, Volvo Trucks, hereafter Volvo, is sponsoring this capstone project aimed at modeling, analyzing and drawing conclusions about the performance and durability of a fuel cell stack in a truck under varying operating conditions. To achieve this, the project team is composed of four students from Chalmers University of Technology, hereafter Chalmers, and five students from Pennsylvania State University, hereafter, Penn State, with backgrounds in mechanical engineering, energy engineering and chemical engineering with engineering physics. The project will be limited to a literature study to gain insight into the underlying mechanism, as well as simulations to comprehend and visualize the degradation process over time.

1.1 Background

According to their homepage, "Volvo team is committed to driving the transition to sustainable, safe, and more productive transport and infrastructure solutions while making the journey to become a net-zero society" [10]. One way to do that is through the implementation of hydrogen fuel cells, which has the possibility to not produce any pollutants or greenhouse gases [11] but only water and heat. This makes fuel cells an effective solution to achieve the goal of more sustainable transport and infrastructure solutions with zero pollution.

Since heavy-duty trucks account for a significant portion of transportation emissions, it is crucial to explore new green technologies. In 2019, the EU introduced a regulation that mandates reductions in emissions from heavy-duty vehicles (HDV). By 2030, emissions have to be reduced by 15% by 2025 and 30% by 2030 [8]. Volvo Trucks plans to introduce fuel cell-driven trucks to meet these standards and have together with Daimer Truck AG created a joint effort to produce fuel cell systems capable of powering larger scale vehicles. The effort, known as Cell-Centric, aims to supply both companies with fuel cell engines to install in their commercial-grade trucks [12].

Introducing fuel cells in heavy-duty offers an advantage beyond zero-emission technology. Fuel cell engines are more efficient than combustion engines, which is especially important for heavy-duty vehicles that require a lot of power to transport heavy loads[13]. However, it is crucial to consider the total operating lifespan of these trucks. This means that the fuel cell technology used in the engine must be resilient enough to sustain this repeated heavy use that they will be subjected to.

1.1.1 Mechanisms of the fuel cells

A fuel cell is a device that converts chemical energy into electrical energy by utilizing an electrochemical reaction between a fuel (hydrogen) and an oxidant (oxygen) to generate electricity without combustion, as illustrated in Figure 1. The Proton Exchange Membrane (PEM) fuel cell, also known as the Polymer Electrolyte Membrane fuel cell, is the type of fuel cell that is studied in this project. The PEM fuel cell is distinctive because it uses a solid polymer electrolyte and the electrodes are made out of carbon, containing platinum as the catalyst. These cells are often used in transportation applications such as heavy-duty trucks due to their low operating temperature (60°C – 80°C), quick start and low density compared to other fuel cells types [14].

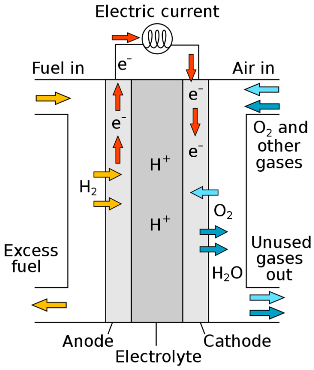


Figure 1: Schematic representation of a fuel cell.

The PEM fuel cell operates by supplying hydrogen gas (H_2) to the anode, where it is converted to two hydrogen ions (protons) and two electrons via a catalyst, as shown in Equation (1a). The electrons are then used to power an electrical device such as a motor or charge a battery. The protons diffuse through an electrolyte membrane that blocks the flow of electrons and then arrive at the cathode. At the cathode, the atmospheric oxygen (O_2) is catalytically split into ions. The ions then react with two protons through an exothermic reaction to produce water, as seen in Equation (1b). The water is later transported out of the cell using airflow.[15]. This reaction is continuous, and the power output can be regulated by adjusting the concentration of hydrogen and oxygen in the cell. The voltage output is generated by the potential difference between the cathode and anode and can be increased by stacking numerous cells together, [16]. The byproduct of fuel cells is water and heat, making it an attractive alternative to other clean energy sources. The overall reaction is represented by Equation (1c).

$$\begin{cases} \text{Anode reaction: } H_2(g) \rightarrow 2H^+ + 2e^- & (1a) \\ \text{Cathode reaction: } \frac{1}{2}O_2(g) + 2H^+ + 2e^- \rightarrow H_20(l) + \text{heat} & (1b) \\ \text{Overall reaction: } H_2(g) + \frac{1}{2}O_2(g) \rightarrow H_20(l) + \text{heat} & (1c) \end{cases}$$

1.1.2 Components of the fuel cell

PEM fuel cell consists of several key components, each of which plays a critical role in the overall operation of the cell. As illustrated in Figure2 and 3, these components are:

1. Bipolar plates. Located on each end there are bipolar plates. These plates are responsible for separating the anode and cathode, conducting electrical current, distributing the fuels, absorbing and removing water, and providing mechanical stability[17].
2. Gasket. Inside the plates, there is a gasket. This part is responsible for sealing the cell, preventing fuel from leaking out, and preventing air and water from coming in[17].
3. Gas diffusion layer. Surrounding the membrane and inside of the gaskets there is a gas diffusion layer (GDL). Responsible for the transport of gases to and from the electrode and for distributing the fuel and oxidant gases evenly over the surface of the electrode. Therefore the GDL is generally made from a porous material that provides a high surface area for gas[17].
4. Polymer Electrolyte Membrane (PEM) and the Catalyst Layer (CL). Located in the middle of the cell and is responsible for the electrochemical reaction that generates electricity. The following components are found in this region[17]:
    - Polymer Electrolyte Membrane- The main task of the membrane is to let the protons pass through to the cathode whilst hindering the electrons from passing through. It is generally made of perfluorosulfonic acid (PFSA) polymer, also called Nafion, as they are highly conductive to protons and highly selective to hydrogen ions, making them an ideal material for use in fuel cells[17].
    - Catalyst layers- The catalyst layer speeds up the reactions, the more active the surface area that is exposed to the gases the more efficient will the process be. It is coated on the surface of the cathode and the anode and is usually made of platinum or platinum alloy[17].
        - Anode- Fuel is oxidized which releases electrons. The anode is often made of porous carbon material, coated with the catalyst[17].
        - Cathode- Oxygen is reduced here and as a consequence, the ions attract electrons and the residual product will be water. The cathode is also made out of porous carbon material and coated with the catalyst[17].

1.1.3 Fuel cell electric vehicle

As mentioned in the previous section, hydrogen is stored onboard fuel cell powered vehicles and used to power the fuel cell stack. Due to the low power output from a single fuel cell, a number of

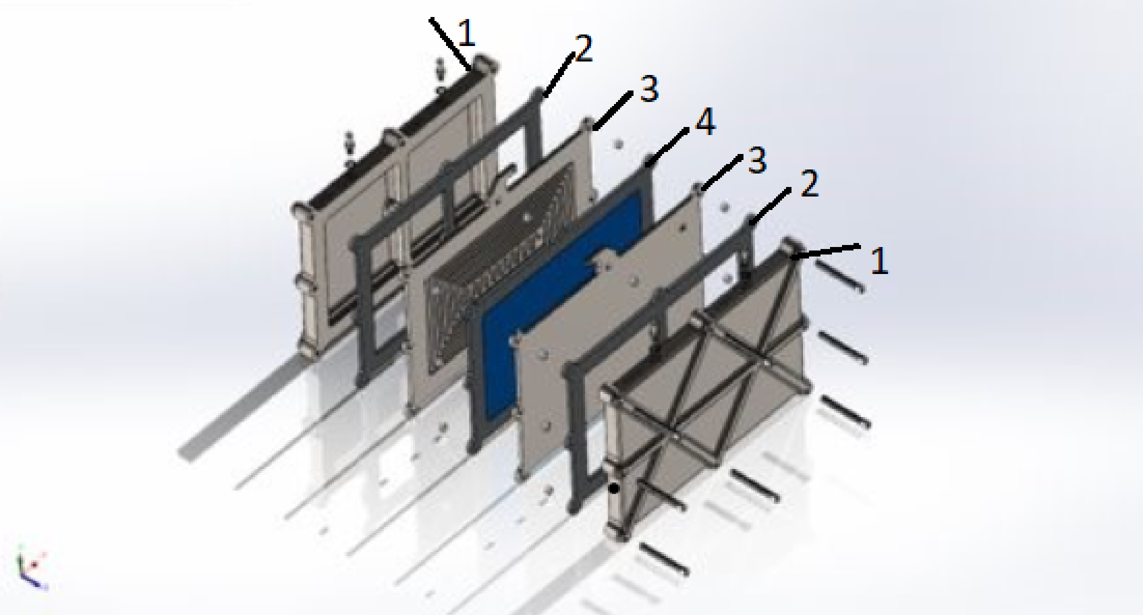


Figure 2: SolidWorks model of the Hydrogen Fuel Cell System, where the blue layer is the PEM and the adjacent layers are the anode and cathode. 1.End plate/Biopolar plate, 2.Gasket, 3.Gas diffusion layers and 4. PEM + catalyst coating.

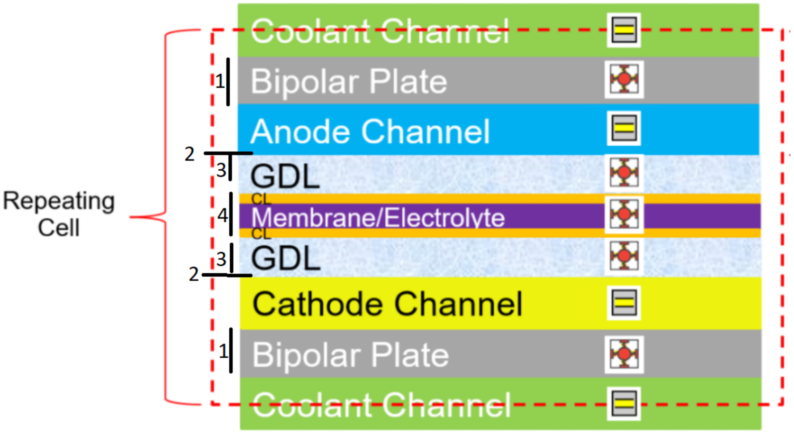


Figure 3: Repeating cell from GT-Suite[1] with the same numbering as Figure 2.

cells are often combined into a stack. In heavy-duty vehicles, a fuel cell stack can contain hundreds of cells [18]. Independent of the number of fuel cells in the stack, the electric power is managed by a power electronics controller [19]. The idea is to make the power delivery from the fuel cell as idle as possible and in cases where the power demand is low the fuel cell is turned off. The power electronics controller also manages a high voltage battery pack in order to control the speed of an electric traction motor that is used to drive the wheels of the fuel cell electric vehicle (FCEV). Transmission is needed in order to transfer mechanical power from the electric traction motor to the wheels [19]. Apart from storing electric energy from the fuel cell and powering the electric traction motor, the battery pack is used to recapture breaking energy and provide extra power during accelerations [19]. The energy to start the vehicle and provide accessories is not provided by the battery pack. Instead, the energy comes from an auxiliary low-voltage battery. A DC/DC converter is needed to convert the voltage between the different batteries. In order to keep the temperature in an appropriate range for the fuel cell, battery and other components in the FCEV, a thermal system is needed as well [19], and in order to use air in the inlet, a compressor and humidifier is needed for the cell to operate [18].

1.2 Degradation in a fuel cell

A fuel cell will degrade which means the performance and lifespan will be reduced over time. Components of the cell degrade in different ways and have different impacts on the function of the cell, with most of the mechanisms impacting performance. The most critical part of the fuel cell is the part where all the electrochemical processes take place since that is where chemical energy is converted to electricity: the membrane electrode assembly (MEA) [20]. MEA consists of PEM, GDL and the CL. The durability of MEA, especially the PEM and CL, has therefore a huge impact on the overall performance and longevity of the cell [20].

The electrolyte membrane in a fuel cell is as stated before a critical component that is responsible for proton conductivity and the electrochemical reaction. One of the primary factors that can degrade the membrane is water management[21]. If the water management is insufficient, the membrane can become dehydrated, causing it to become brittle and crack. A crack in the membrane will cause unwanted substances to leak through and the cell stops working. On the other hand, if there is excessive water, the membrane can swell, leading to increased ionic resistance which hurts its performance. As the membrane needs some moisture to operate correctly, freezing temperatures becomes a problem and can cause the water to destroy the membrane. High temperatures can also expose the membrane to a critical breakdown as well as a risk for local drying. Too much water in the MEA affects the CL as well: water blocks the reaction gasses from reaching the active point which affects the fuel cell's power output, and in some extreme cases it even causes permanent damage to the cell due to oxygen starvation [20].

The gas diffusion layer is also subject to some degradation due to temperature changes, especially at the startup of the cell. This process can cause numerous damage over time if the cell is not properly preheated before a cold start[21].

The catalyst layer and the electrodes are also exposed to degradation due to harsh environments. High temperatures, low pH, and dynamic load are primary factors that cause particle growth and corrosion that decreases membrane stability and conductivity. Platinum particle degradation (and in turn the degradation of the catalyst layer) is mostly due to four degradation mechanisms: the Ostwald ripening effect, migration, agglomeration and loss of platina particles [20]. All of these mechanisms result in diminishing active surface area and in turn, worsen the fuel cell's performance over time. Dissolution/loss of platinum particles can lead to platinum particles fastening on the PEM's surface and blocking the passage of protons which further decreases its power output.

1.2.1 The Ostwald ripening Phenomenon

One of the aforementioned degradation mechanisms that affect the catalyst layer performance is called Ostwald ripening. It is a phenomenon in which small particles within a material increase in size over the course of its lifespan[22]. Figure 4 shows an example of this process in a rolled steel product.

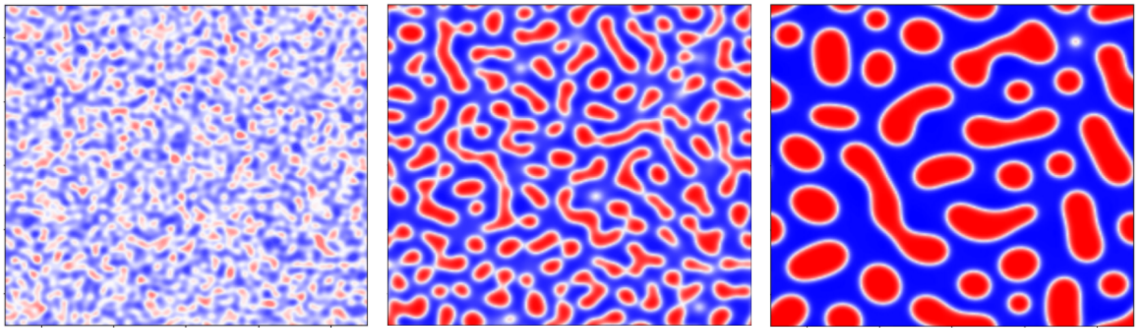


Figure 4: Particle growth, also known as Ostwald ripening, on steel product. This results in a smaller active surface area which in turn lowers the fuel cell's power output over time.[2].

The Ostwald ripening phenomenon also occurs within the CL during operation[23]. The catalyst layer of a fuel cell unit consists of small platinum particles attached to a larger support material, typically carbon support and Nafion[23]. During fuel cell operation (particularly in the cathode side of the cell), the platinum atoms lose electrons from the reaction chemistry and become platinum ions. When this happens, they are subject to oxidation by oxygen ions entering the cathode side through the gas diffusion layer.

Platinum oxide forms and breaks off from the larger platinum structure. Eventually, this platinum oxide molecule is broken apart into a platinum ion and water with the presence of two hydrogen ions passing through the proton exchange membrane. The platinum ion can then neutralize and re-attach itself to a platinum structure within the CL [23]. The continuous motion of platinum oxide and platinum ions throughout the CL results in the Ostwald ripening phenomenon depicted in Figure 4. Larger platinum particle sizes lead to degradation in fuel cell performance because the surface area to volume ratio of the platinum decreases. In other words, there is less platinum available to contact hydrogen ions passing through the CL. Lower catalyst interaction leads to a decrease in performance[23]. The amount of available platinum surface area available for the electrochemical reaction is known as the electrochemical surface area (ECSA)[23].

1.3 Client needs

This capstone project is undertaken on behalf of Volvo and is therefore adjusted to meet their requirements and visions. Moreover, it will mainly focus on improving software simulation skills and analyzing the output rather than developing the existing product. Therefore, the following requirements have been established by the client:

- Utilize GT-Suite for one-dimensional modeling.

- Observe the fuel cell's function for the first half of a truck's operating time, in this case, the first 10 000 hours.
- Partially analyze the degradation of the fuel cell with varying operating conditions.

1.4 Aim

The aim is to model, analyze and draw conclusions about the performance and durability of a fuel cell stack in a truck. A particular focus will be the degradation of the catalyst layer due to Ostwald ripening. This will be accomplished by making a one-dimensional model of a fuel cell and monitoring its power output with decreasing ECSA to investigate whether or not such a fuel cell stack can meet the requirements established by the client.

To reach the aim of the project and to meet the client's needs the project needs to include the following tasks:

- Investigate the mechanism of a fuel cell and how it is integrated into the full stack of fuel cells.
- Construct and design two different one-dimensional fuel cell models, whereof a more complex and precise model in GT-Suite and a simpler model in Matlab to study the Ostwald ripening over a longer period of time.
- Evaluate the performance of the one-dimensional model and specifically study how the output is affected by the decreasing ECSA.
- Analyze the degradation of the ECSA and develop an understanding of the mechanism.
- Organizing and working in a globally distributed team.

1.5 Limitations

As stated above, the aim of this project is to simulate a fuel cell's functionality over time to show the effects of the Ostwald ripening. Since buying and testing a fuel cell is both time-consuming and expensive, the project will be limited to literature studies and computer simulations in GT-Suite[1] and Matlab [24]. The project's main focus will be the degradation of the catalyst layer due to the Ostwald ripening.

Since the objective is to learn more about the influence of the Ostwald ripening on the life of a fuel cell and the resulting degradation, there will be no design or recommendation of improvement on the existing auxiliary functions.

Even though multiple fuel cell stacks are usually used to power vehicles, the simulations are going to be made on only one fuel cell stack for the sake of simplicity since multiple fuel cells can easily be coupled together. Considering the fuel cells that will be used in a truck, it is a requirement for them to handle heavy-duty conditions. Volvo's request is that the fuel cell remains functional for at least 10 000 hours mid-life. Due to confidentiality, Volvo has not been able to provide any numbers defining the fuel cell simulation and therefore all in-data parameters to the simulations have to be estimated. The functionality of the fuel cell is going to be defined in terms of output power.

Moreover, the project is conducted over a time span of 20 weeks, from January to May. The Penn State students have a time span of about 15 weeks, i.e. the team from Penn State has their final deliverables at the beginning of May.

1.6 Ethical and environmental aspects

Even though fuel cells have great potential to be a great sustainable alternative to lithium-ion batteries, currently there are several ethical and environmental concerns regarding their widespread use. In order for a project to be ethically justified, by Chalmers[25], it is important that it is of benefit to the people. In this case, a fuel cell's catalyst layer and its corresponding degradation are investigated. A highly efficient fuel cell without the degradation problems of today would be of benefit to society since fuel cell trucks could replace diesel and gasoline-driven trucks, which would reduce global greenhouse gas emissions and improve air quality.

Another problem with the increased use of fuel cells is that companies and people in general view fuel cells as a highly environmentally friendly and fossil fuel-free solution. However, it depends on the whole process; the electrolytic product within the fuel cell will exclusively be water, meaning that the fuel cell itself does not contribute to any hazardous greenhouse gases. Nevertheless, hydrogen itself may be derived from fossil fuels. Today, most hydrogen is produced from fossil fuels [26]. Whenever hydrogen is generated from renewable sources such as wind, solar, biomass or hydropower, only then the fuel cell will result in zero emissions. Companies can take advantage of this and utilize it as greenwashing by labeling it as more sustainable than it is compared to combustion engines, to increase their own competitiveness.

The fuel cell is driven by oxygen and hydrogen, which are sources that are readily available in the atmosphere and thus more sustainable than fossil fuels. Nevertheless, if the technology development is driven forward and fuel cell vehicles become more common there will be an increased demand for hydrogen and metals used in the fuel cell. Fuel cells have platinum plates in the layer next to the membrane. Platinum must be used there since it is the only metal that is capable of surviving in such an acidic environment [17], but platinum has a high carbon footprint; 20 600 tons of carbon dioxide are emitted per ton of produced platinum [27]. If fuel cells become more widespread, this could cause serious consequences for the environment.

Efficiency in a fuel cell is also high compared with combustion engines [26], as the potential energy within the chemical bonds are converted into electricity with fewer losses, therefore less fuel is needed which is also beneficial for sustainability. In conclusion, the hydrogen fuel cell may not be the best alternative today seen from an environmental perspective. Still, with further research and optimizations, it has a high potential to surpass traditional power sources.

2 Theory

In order to accurately model and analyze a PEM fuel cell, a deeper understanding of its degradation mechanisms and the equations used to implement them is necessary. To gain insight into the overall performance of the cell, one needs to understand the polarization curves and the loss of voltage that contributes to the decreasing cell potential. Equations used to implement this model are presented in this section. This provides the information that will enable the simulation and prediction of real-time fuel cell performance is presented.

2.1 Voltage Losses

Like all other electrical and mechanical systems, fuel cells experience losses that affect their performance. Therefore it was necessary to understand the losses affecting the fuel cell in order to draw conclusions on the performance over time. There are different kinds of losses and there are many underlying reasons for losses to occur, e.g. the fact that heat is created as the fuel cell is working. Since the functionality of the cell was defined in terms of output power, losses affecting the cell voltage are considered and explained in this section. The total loss of voltage is mainly dependent on four factors: open circuit losses, activation losses, ohmic losses, and concentration losses. All of these will contribute to a decrease in the efficiency and performance of the fuel cell.

The cell voltage $\mathrm{V_{cell}}$ in the GT-Suite simulation is calculated according to [15]:

$$V_{\text{cell}} = V_{\text{OC}} - V_{\text{act}} - V_{\text{mt}} - V_{\text{ohm}} - V_{\text{delOC}} \tag{2}$$

where $\mathrm{V_{OC}}$ is the open circuit voltage, $\mathrm{V_{act}}$ is the activation loss, $\mathrm{V_{mt}}$ is the mass transport/concentration loss, $\mathrm{V_{ohm}}$ is the ohmic loss and $\mathrm{V_{delOC}}$ is the internal voltage loss. The internal voltage loss is an input parameter to the GT-Suite simulation and this value will be subtracted from the Nernst potential of the cell as a constant value [15], where the Nernst equation, which according to Amphlett et. al [28] describes the thermodynamic potential of the overall chemical reaction of the fuel cell:

$$E = E^{\circ} - \frac{RT}{nF} \ln[p^{*}_{h_2}(p^{*}_{o_2})^{0.5}] \tag{3}$$

where E° is a reference potential.The rest of the losses are explained below.

2.1.1 Open circuit losses

The open circuit voltage as well as the internal voltage losses are used to compute the cell voltage in Equation (2). The open cell voltage is in GT-Suite [15] defined by Gibbs free energy [1] of formation of the overall chemical reaction of the fuel cell described in Equation (1c). According to the fact sheet in GT-Suite, the implemented open-cell voltage can be described by:

---

[1] Gibbs free energy is a combination of the enthalpy h and entropy s of the chemical reaction [29] according to $\Delta g = \Delta h - T\Delta s$ where T is the absolute temperature. The correlation is based on assumptions of constant temperature and pressure throughout the reaction. Gibbs free energy tells how spontaneous a reaction is.

$$V_{\mathrm{OC}} = \frac{-\Delta \bar{g}_{\mathrm{f}}}{2\mathrm{F}} \tag{4}$$

where $-\Delta \bar{g}_{\mathrm{f}}$ is the difference in Gibbs free energy over the reaction and F is Faraday's constant. According to the software, the difference in Gibbs free energy can be described as the Gibbs free energy of water subtracted by the Gibbs free energy of water and half the Gibbs free energy of oxygen according to [15]

$$-\Delta \bar{g}_{\mathrm{f}} = \left(\bar{g}_{\mathrm{f}}\right)_{\mathrm{H_2O}} - \left(\bar{g}_{\mathrm{f}}\right)_{\mathrm{H_2}} - \left(\bar{g}_{\mathrm{f}}\right)_{\mathrm{O_2}} \tag{5}$$

Thus in total, the open cell voltage can be expressed as:

$$V_{\mathrm{OC}} = \frac{\left(\bar{g}_{\mathrm{f}}\right)_{\mathrm{H_2O}} - \left(\bar{g}_{\mathrm{f}}\right)_{\mathrm{H_2}} - \left(\bar{g}_{\mathrm{f}}\right)_{\mathrm{O_2}}}{2\mathrm{F}} \tag{6}$$

The internal voltage loss is calculated when the fuel cell is an open circuit, i.e. when there is no current flowing through the cell [15]. This parameter should be specified as input to the simulation. According to the GT-Suite fact sheet it can alternatively be calculated based on the numerical polarization curve fit. In the simulation the open cell voltage loss is specified.

2.1.2 Activation loss

Activation losses take place in the cell's catalyst layer and they occur due to the slowness of the reactions that take place on the electrode's surface; oxygen reduction on the cathode and hydrogen oxidation on the anode. Losses due to oxidation are often neglected since oxidation is much faster than oxygen reduction. Activation losses in a PEM fuel cell are described by the Tafel Equation [15]:

$$V_{\mathrm{act}} = \frac{\mathrm{R_{gas}} \cdot T}{2 \cdot \alpha \cdot \mathrm{F}} \cdot \ln\left(\frac{i}{i_0}\right) \tag{7}$$

where $\mathrm{R_{gas}}$ is the universal gas constant, T is the absolute temperature, F is Faradays constant and α is the Charge transfer coefficient, which is a parameter that is involved in the change of the rate of the electrochemical reaction [15] described in Equation (1). Furthermore, i is the current density and i_0 is the exchange current density, i.e. the current density at which the activation losses start becoming non-zero [15]. The exchange current density is in GT-Suite evaluated at every time step using the formula

$$i_0 = i_0^{\mathrm{ref}} \mathrm{a_c} \mathrm{L_c} \left(\frac{P_{\mathrm{O_2}}}{\mathrm{P_{ref}}}\right)^{\gamma} \exp\left(\frac{\mathrm{E}}{\mathrm{R}}\left(\frac{1}{\mathrm{T_{ref}}} - \frac{1}{\mathrm{T_{ref}}}\right)\right) \tag{8}$$

where $P_{\mathrm{ref}} = 1$ atm and $T_{\mathrm{ref}} = 298.15$ K. i_0^{ref}, a_{c}, L_{c} and E are input parameters for the reference exchange current density, catalyst-specific surface area, catalyst loading, and activation energy respectively. i_0 is evaluated at each timestep based on the current oxygen pressure $P_{\mathrm{O_2}}$ and cell temperature T_{cell}.

2.1.3 Ohmic loss

Ohmic losses are due to the electronic and ionic conduction in the different components of the fuel cell. This will lead to thermal energy losses, and as a consequence, the efficiency of the cell is decreasing. Ohms law is describing this phenomenon [15]:

$$V_{\text{ohm}} = I \cdot R \tag{9}$$

The Ohmic resistance is evaluated at each timestep according to:

$$R = t\sigma \tag{10}$$

Where,

$$\sigma = (b_{11}\lambda_m - b_{12})\{\left(b_2\left(\tfrac{1}{303} - \tfrac{1}{\mathrm{T_{cell}}}\right)\right)\} \tag{11}$$

where b_{11}, b_{12} and b_2 are input parameters to the simulation. λ is a measure of the water content of the membrane and T_{cell} is the absolute cell temperature.

2.1.4 Concentration loss

For fuel cells, concentration losses are voltage losses due to gas transport [30]. According to the GT-Suite fact sheet [15] concentration losses are voltage losses as a result of the change in concentration of the reactants at the electrode surfaces. The concentration loss in the simulation is computed as

$$V_{\text{mt}} = -C \cdot \ln\left(1 - \frac{i}{i_l}\right) \tag{12}$$

where C is the mass transport loss coefficient, i is the current density and i_l is the limiting current density. The mass transport loss coefficient is, according to the GT-Suite documentation fact sheet [15], for a PEM fuel cell different between the anode and cathode and can be expressed as

$$C = \begin{cases} \frac{\mathrm{R_{gas}}T}{2\mathrm{F}} & \text{Anode side} \\ \frac{\mathrm{R_{gas}}T}{4\mathrm{F}} & \text{Cathode side} \end{cases} \tag{13}$$

where $\mathrm{R_{gas}}$ is the universal gas constant, T is the absolute fuel cell operating temperature and F is Faraday's constant. From the fact sheet [15] also follows that the simulation uses a single coefficient for the simulation. This coefficient is specified as input to the simulation. Equation (13) and the order of the ingoing constants is used to estimate the value of the constant. The gas constant is of order 10^0, the temperature of order 10^2 and Faraday's constant is of order 10^5. Therefore the constant should be of order $10^2 \cdot 10^2/10^5 = 0.001$.

The current density of the fuel cell is the current per active fuel cell area unit. The limiting current density is according to the GT-Suite simulation fact sheet [15] the current density at which the fuel is consumed at its maximum supply rate. Independent of the limiting current density at one of the electrodes, when the limiting current density is reached at the other electrode the fuel cell voltage drops to zero [15]. The limiting current density is, like the mass transport coefficient, specified as a data parameter to the simulation.

2.1.5 Identifying losses in a polarization curve

A polarization curve displays the voltage output of the fuel cell for a given current density loading and is one of the most common methods of testing fuel cells and estimating the performance. The losses listed above can be identified in a polarization curve as shown in Figure 5.

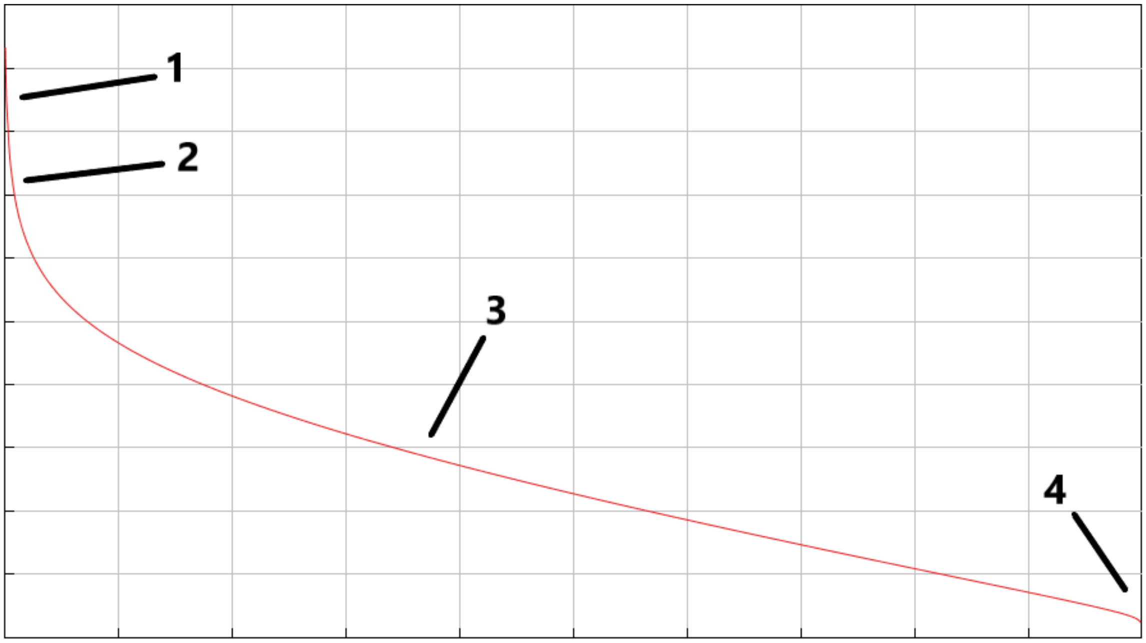


Figure 5: Losses in a polarization curve. 1: Open circuit losses due to fuel crossover. 2: Rapid drop due to activation losses. 3: Linear drop due to ohmic losses. 4: Concentration loss at high current densities[3].

2.2 Catalyst layer degradation and the Ostwald ripening

The losses presented in the previous sections are necessary in order to understand the fuel cell performance, but in order to study the performance over time other degradation mechanisms must be studied as well. As previously stated, there are many different degradation mechanisms that can cause a decrease in the performance of a fuel cell. Some of the most prevalent degradation modes include Ostwald ripening in the catalyst layer and corrosion of the PEM. In this section, the degradation of the catalyst layer in terms of Ostwald ripening is explained.

At the beginning of a catalyst layer's lifespan, its properties can be observed under a microscope. In particular, the average platinum particle size throughout the layer can be determined. A typical initial platinum particle radius is around 2.6 nm[23]. Typical models that try to predict the growth of these particles over time consider dozens of different particle teams normally distributed about this size. These models are computationally expensive because every set of particles has multiple state variables. To simplify this model, in the paper *Prognostic-oriented Fuel Cell Catalyst Aging Modeling and Its Application to Health-Monitoring and Prognostics of a PEM Fuel Cell*, Xian Zhang and Pierluigi Pisu developed and proposed a model that only considers two particle sizes: one with a radius greater than the average and one with a radius less than the average[23]. The model then predicts the change in the platinum surface area along with the current platinum oxide coverage at the surface of the platinum sites. These two parameters are the state variables of the model. The input parameters of the model together with their physical meanings are shown in Table 1.

Table 1: Ostwald Ripening Parameters.

Parameter Name	**Physical Meaning**	**Units**
V_{pt}	Total Volume of Platinum in Fuel Cell	cm^3
A_{fc}	Fuel Cell Cross-Sectional Area	cm^2
α_{r}	Constant Relating the Decrease in the Smaller Particle Radius as a Multiple of the Larger Particle Radius Increase	Unitless
β_{r}	Constant Modeling the Change in Larger Particle Radius Relative to the Current Average Particle Radius	Unitless

Because there are two system states, two differential equations and two initial conditions are needed for simulation. The governing differential equations were derived in 3D and are shown below.

$$\frac{\mathrm{d}A_{\mathrm{geo}}}{\mathrm{d}t} = -4k_1 \cdot \frac{u_c}{u_\theta}\theta_{\mathrm{vac}} \cdot \frac{M_{\mathrm{pt}}}{\rho_{\mathrm{pt}}} \cdot \frac{F\alpha_1}{RT} e^{\left(\frac{F\alpha_1 A_{\mathrm{geo}}}{3V_{\mathrm{pt}}\mathrm{R}T}\right)} \cdot \frac{A_{\mathrm{geo}}^3}{9V_{\mathrm{pt}}^2} \cdot \frac{\alpha_r \beta_r^2}{(1+\beta_r)(1-\alpha_r\beta_r)} \tag{14}$$

$$\frac{\mathrm{d}\theta_{\mathrm{PtO}}}{\mathrm{d}t} = \frac{v_2}{\Gamma_{\mathrm{max}}} - \frac{2\theta_{\mathrm{PtO}}}{\bar{r_a}} \cdot \frac{\mathrm{d}\bar{r_a}}{\mathrm{d}t} \tag{15}$$

The input to the system is the variable u_c, which is an Arrhenius equation given by:

$$u_c = \exp\left(\tfrac{\mathrm{F}V}{\mathrm{R}T}\right) \tag{16}$$

Voltage V is the instantaneous operating voltage of the fuel cell. The constant terms of the open cell voltage (see section 2.1) are teamed together into a single parameter denoted by ε. The values of all these terms can be found in Appendix A. For the parameters defined in the appendix, the value of ε is $-4.94 \cdot 10^{-26}$. Equation (14) can then be written as Equation (17). The functions g_1 and g_2 are defined in Equations (18) and (19), respectively.

$$\frac{\mathrm{d}A_{\mathrm{geo}}}{\mathrm{d}t} = \varepsilon \cdot g_1(V, \theta_{\mathrm{PtO}} \cdot g_2(A_{\mathrm{geo}}) \tag{17}$$

$$g_1(V, \theta_{\mathrm{PtO}}) = u_c \theta_{\mathrm{vac}} = \exp\left(\frac{FV}{RT}\right) \cdot \theta_{\mathrm{vac}} \tag{18}$$

$$g_2(A_{\mathrm{geo}}) = e^{\left(\frac{F\alpha_1 \mathrm{A_{geo}}}{3V_{\mathrm{pt}}RT}\right)} \cdot A_{\mathrm{geo}}^3 \tag{19}$$

The remaining parameters in these equations are functions of the two state variables. These terms are all shown in Table 2. Equation (20) shows the definitions of θ_{vac} and $\bar{\mathrm{r_a}}$.

Table 2: Ostwald Ripening States.

Variable Name	**Physical Meaning**	**Units**
A_{geo}	Total Surface area of Platinum Particles	cm^2
θ_{PtO}	Fraction of Surface Area Covered by Platinum Oxide	Unitless
θ_{vac}	Fraction of Surface Area Available for Reaction	Unitless
$\bar{\mathrm{r_a}}$	Average Particle Radius	cm

$$\begin{aligned} \theta_{\mathrm{vac}} &= \max(0, 1 - \theta_{\mathrm{PtO}}) \\ \bar{r}_a &= \frac{3V_{\mathrm{pt}}}{A_{\mathrm{geo}}} \end{aligned} \tag{20}$$

The initial conditions for the two states are calculated by using Equation 21.

$$A_{\mathrm{geo}}(t=0) = \frac{3V_{\mathrm{pt}}}{\bar{r}_a(t=0)} = \frac{3V_{\mathrm{pt}}}{2.6 \cdot 10^{-6}} \theta_{\mathrm{PtO}} \tag{21}$$

The electrochemical surface area (ECSA) is then related to the geometric area of the platinum by Equation (22) [23].

$$ECSA \approx 0.67 A_{\mathrm{geo}} \tag{22}$$

For this study, a catalyst layer can be considered at its end of life (EOL) when its ECSA has reached 25% of its original value[31].

The input to the two-state model is the voltage of the fuel cell. In the context of a FCEV, this voltage input may vary depending on the power demand of the traction motor at any given time. Ideally, a fuel cell should operate at a constant voltage, producing a constant current. Rapid changes in voltage could cause faster degradation of the fuel cell[32]. For this reason, many FECVs feature an auxiliary battery that can supply additional power when needed to the traction motor.

3 Methodology

The project included a literature study and simulations in Matlab and GT-Suite. These were performed in order to understand fuel cell degradation mechanisms and how they affect the durability and performance of the fuel cell. Moreover, the literature study was used to understand the simulations and find suitable initial values. The project setup, literature study, simulations and project management are explained in the following sections.

3.1 Project overview

Initially, a planning phase was conducted in order to structure the project and ensure that the fuel cell performance and degradation are investigated within the limited time of the project. As illustrated in Figure 6, a planning phase was followed by research in order to understand the fuel cell and modeling. The research included studies of literature and contact with experts within the field. To understand more about the fuel cell performance and degradation two simulations were built and run, followed by analysis of the results. During the analysis phase, the performance of the current model was evaluated. Sometimes the model did not run or behaved strangely and then the process started over from the research phase. Iterations between research, simulation and analysis were performed in order to make the model run as well as possible given the prerequisites of the team members and time frame. Lastly, the results of the final simulation were analyzed and discussed in order to draw conclusions about the fuel cell performance and degradation over time. Documentation and report writing have been written continuously throughout the project.

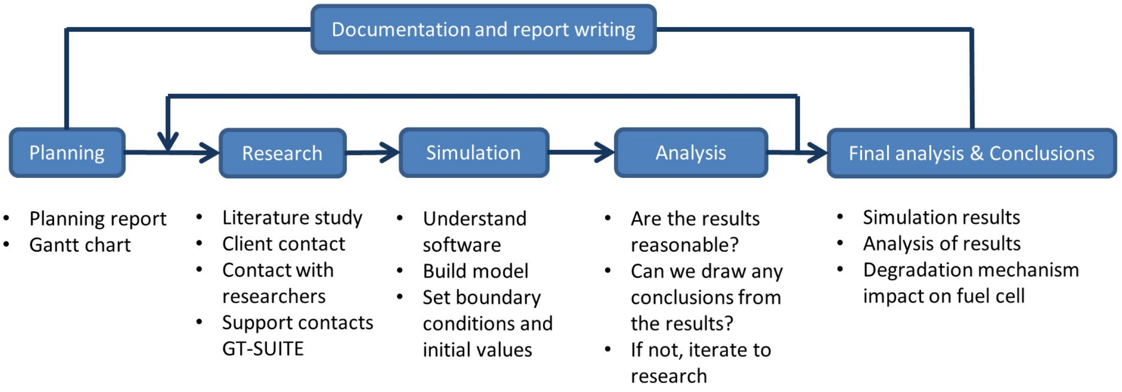


Figure 6: Flow chart representing the process of the project.

3.2 Literature overview

The research needed to understand the mechanisms of the fuel cell and how it degrades was found by consulting experts at Chalmers and Volvo, and by searching for research papers online using Google Scholar and other search engines provided by Chalmers Library, e.g. Scopus. Searching for research papers, the team looked at the publication date and a number of quotations in order to find considerable references that were up-to-date and thereby relevant.

In the early stages of the project, the literature study mainly focused on how a fuel cell works and which degradation mechanisms exist. The parts and external factors that are most critical for the fuel cell performance were identified. An effort was also put into finding ways of controlling these factors. After the initial stage, both teams entered the simulation phase. Then the research focused on finding appropriate modeling methods and boundary conditions, which then was conducted continuously in parallel with the modeling. In this stage, the main part of the information on the in GT-Suite simulated mechanisms was found in the GT-Suite help fact sheets. The fact sheets were used in order to understand what was implemented and thereby also the limitations and possible improvements of the model.

3.3 Simulation and analysis

For the simulation of the CL, two different software Matlab and GT-Suite were used. The Penn State team focused on a Matlab simulation of the Ostwald ripening phenomena while the Chalmers team focused on the GT-Suite simulation.

3.3.1 Matlab implementation of Ostwald ripening

For the implementation of the Ostwald ripening, Matlab version r2022b was used. The Matlab implementation of the Ostwald ripening included only the simplified model using the two particle sizes described in section 2.2. The aim of the model is to study the decay of the platinum in the CL on the cathode side over the mid-lifetime of a heavy-duty vehicle. The input parameters of the model are shown in Table 1. The typical average initial particle radius of 2.6 nm defined in section 2.2 has been employed for the simulation.

To investigate how operating voltage impacts degradation, two simulations have been run. The first simulation was run at a constant operating voltage of 0.8 volts which is meant to mimic an FCEV operating at ideal conditions throughout its entire run. The second voltage load tested in the model was a cyclic load with a period of 300 seconds. The load oscillates between 0.8 volts and 0.93 volts, see Figure 7. This load is meant to mimic an FCEV operating at non-ideal conditions.

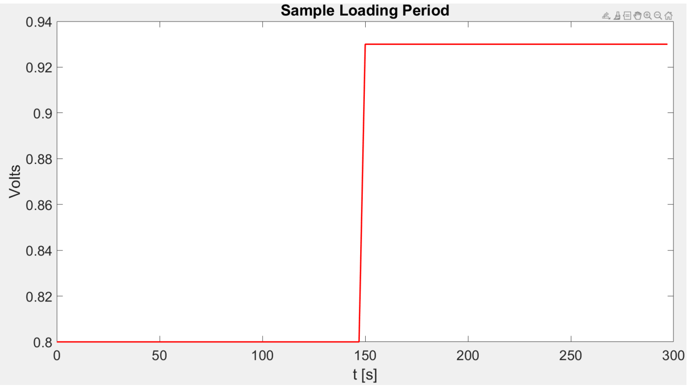


Figure 7: Sample period for a second load test.

Lastly, concerning the Ostwald ripening simulations, the CL area reduction over time was compiled in a table. These values were then used as input parameters of the catalyst-specific surface area used to compute the exchange current density which is present in the computation of the activation loss in GT-Suite according to Equations (7) and (8).

3.3.2 GT-Suite

Since it is difficult to accurately model a fuel cell in Matlab, the output of the Matlab simulation was put into a GT-Suite simulation. GT-Suite is a multi-physics software used for creating and simulating models primarily in the automotive industry. The software is used by the client, but no input data or industrially used models were provided. Instead, the team developed a standalone model from a preexisting template model using estimated data from literature studies and professional advice from the client and experts at Chalmers. The workflow of the simulations is visualized in Figure 8 and further detail on the data used can be found in Appendix B.

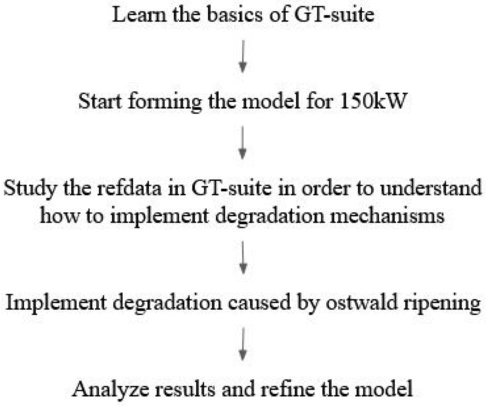


Figure 8: Workflow during modeling.

The model aims to simulate a stack with 600 cells optimized to generate 150kW of electrical power and a screenshot of the model is presented in Figure 9. The simulations with the different catalyst-specific surface area from the Matlab simulation was run as multiple cases. Each case corresponded to the surface area scaling factor from the Matlab Ostwald ripening simulation. The GT-Suite simulation considered, amongst other parameters, the electrical power, temperature and humidity. During the simulation, the flow and pressure properties in the in- and outlets of the cell were also taken into account. However, none of these were changed and they were not considered during the construction of the model.

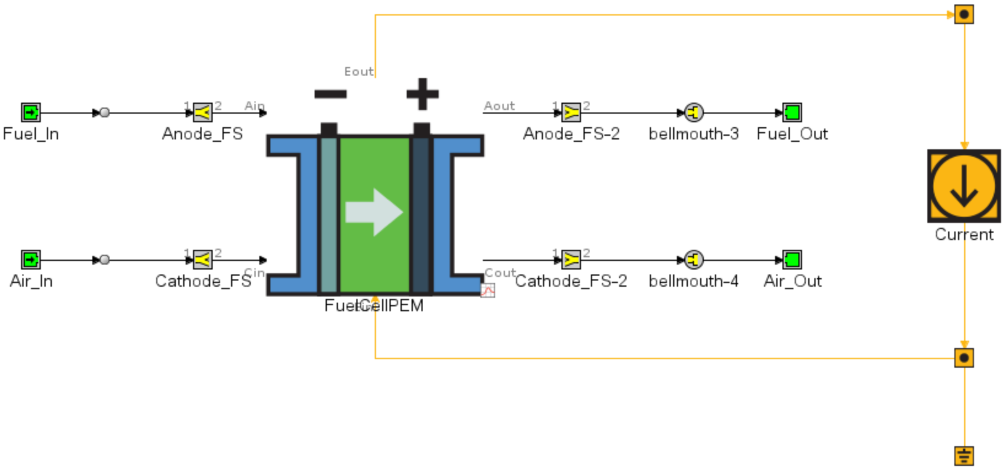


Figure 9: A screenshot of the GT-suite model.

3.4 Project Management

In order to organize the project, distinct tasks were assigned to the different teams. Due to client requirements and the inaccessibility of the GT-Suite software to the Penn State team, the Chalmers team was assigned with simulating the fuel cell by using the GT-Suite software. However, due to unfamiliarity with the software, a learning process was required in order to gain a comprehensive understanding of the simulation tool and to be able to end up with a cohesive model. This also involved research for reasonable parameters since the client was unable to provide this information.

In contrast, The Penn State students worked on developing a more specific model that focused on the Ostwald ripening phenomena. As the GT-Suite model was computationally intensive, the teams adopted an iterative process to obtain the results from each other and to integrate these values in each model. The Penn State team drew inspiration from the team's polarization curve and created a Matlab code to predict the Ostwald ripening phenomena and provide the Chalmers team with surface area scaling factors that could be incorporated into the GT-Suite model.

Throughout the 20-week duration of the project, the two teams had two weekly meetings, whereof one with the supervisors and another with the client. These meetings provided updates on both teams' progress and enabled collaboration and data exchange, as well as guidance from the client to ensure that the project remained in line with their requests.

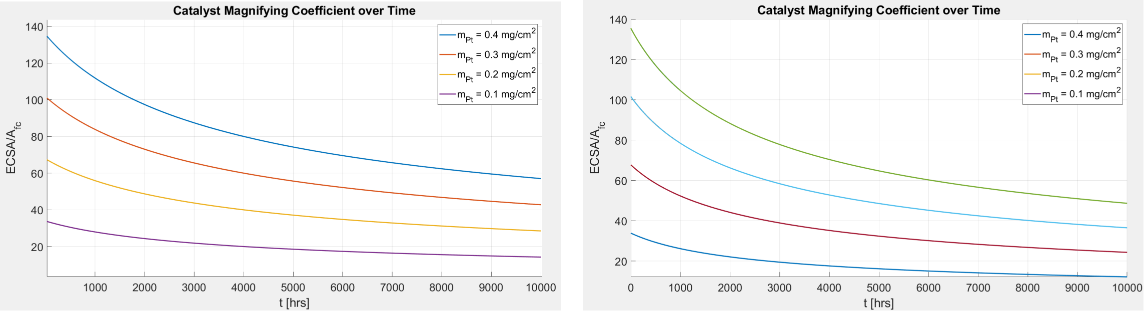
4 Results

Since the result aims to investigate and illustrate the performance of a fuel cell stack in a truck, the effect of Ostwald ripening has been investigated using two different software programs Matlab and GT-Suite. The Matlab simulations are used to induce decay for ESCA, and the resulting data is then analyzed using GT-Suite simulations to investigate power output.

4.1 Catalyst layer degradation due to Ostwald ripening

For the constant 0.8 volts test, the normalized ECSA vs time graph for this input is shown in Figure 10a. The constant input of 0.8 volts is, as previously stated meant to mimic an ideal drive cycle of an electric vehicle. The different graphs correspond to different initial surface areas: the first graph corresponds to the first column and the second graph to the second column in Table 3.

In Figure 10b the corresponding graph of the ECSA over time for the cyclic load meant to mimic more real conditions, from Figure 7, is presented. The graphs correspond to the same platinum densities as in Figure 10a. From the different figures follow that the decay of ECSA is much faster in the non-ideal case. In Table 3, the total degradation of the platinum ECSA of the catalyst layer in percent over time is presented. The degradation percentages are based on the Ostwald ripening simulations in Matlab.



(a) ECSA decay for a constant 0.8V input.

(b) ECSA decay for the square wave input from Figure 7.

Figure 10: Results from the Matlab Ostwald ripening simulations.

Table 3: ECSA reduction over cell life time in percent.

Time [hrs]	**Area Reduction 0.8V**	**Area Reduction 0.8-0.93V**
0	0%	0%
1 000	17%	25%
2 000	29%	37%
3 000	35%	44%
4 000	42%	50%
5 000	45%	55%
6 000	48%	57%
7 000	51%	59%
8 000	54%	61%
9 000	56%	64%
10 000	58%	66%

4.2 GT-Suite simulations

The results from simulating the fuel cell in GT-Suite are divided into two parts in order of importance. One compares the electrical output to cell runtime using the data in table3 and the second displays polarization curves for the cell independent of the Ostwald ripening effect.

4.2.1 Electrical output

Figure 11 shows the power output decaying during the cell lifetime. Each data point corresponds to a value from Table 3. The impact of unsteady voltage on power output is more significant than that of constant voltage. However, it should be noted that the two curves appear to be almost parallel between 2000 and 8000 hours, which indicates that the degradation occurs with equal speed. The most noticeable differences occurred before 2000 hours. Regarding unsteady voltage at 9000 and 10000 hours, the simulation data for power output is not available due to the unreliability of the simulation at these values. Comparing the two cases one can see that the power output at 10 000 hours has been reduced by 17% when the voltage is steady, and the same loss of performance is recorded at 6000 hours when the voltage is fluctuating.

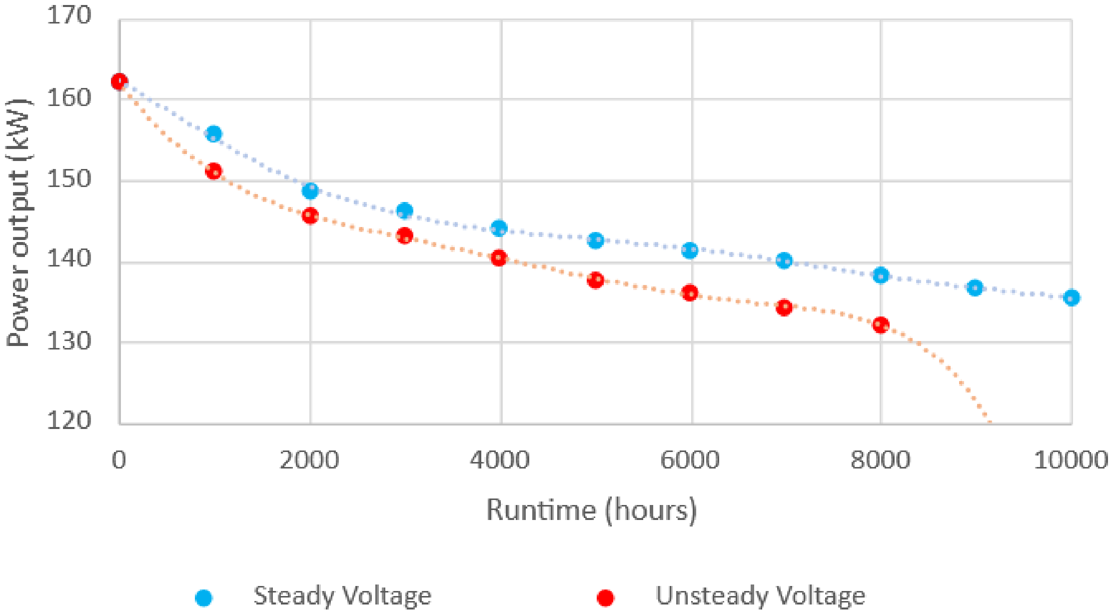
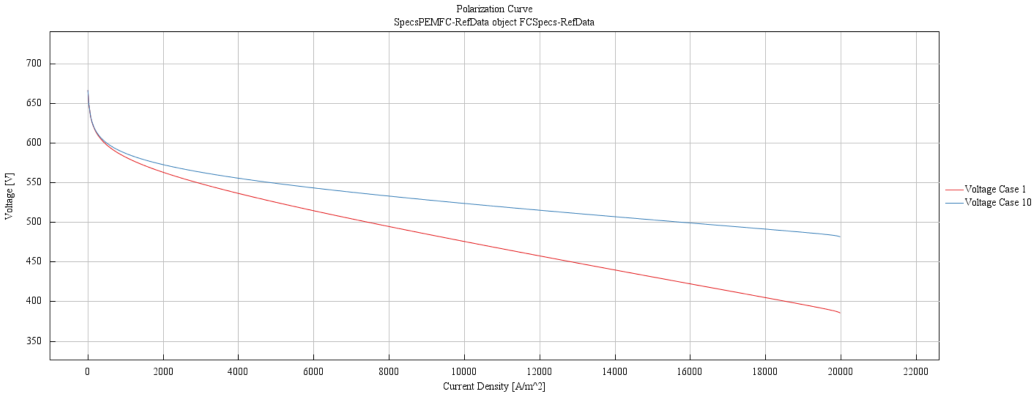


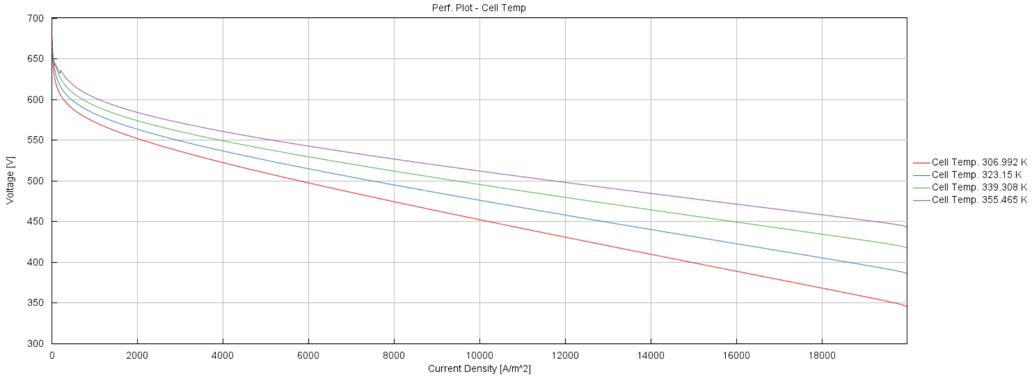
Figure 11: Power output of the fuel cell stack for steady and unsteady voltage at different parts during cell lifetime.

4.2.2 Polarization curves

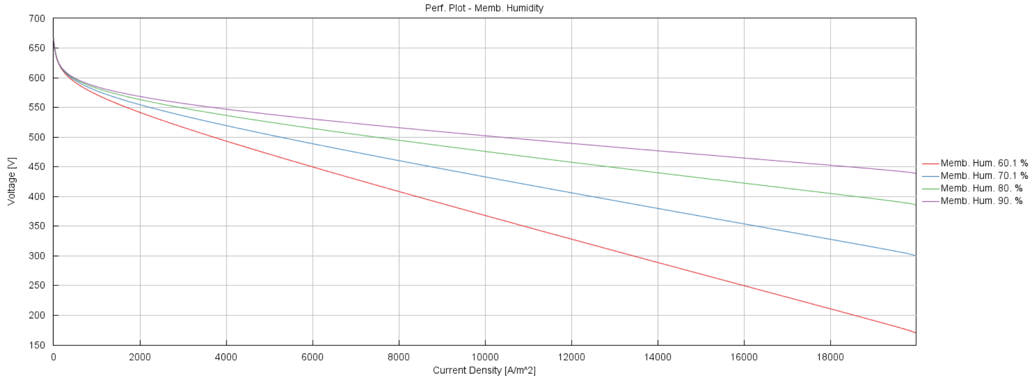
In Figure 12a the linear drop is more significant at case 1 (0 hours) with the rapid drop at higher voltages being similar. Figure 12b displays a lower temperature giving a resulting lower voltage. The curves are quite similar in linear drop, and they are mostly defined by the drop at the start. Figure 12c displays how a lower humidity in the membrane causes the voltage to be lower. Between 0 and 500 A/m^2 the lines are quite similar but with a noticeable difference in linear drop after for values above 500 A/m^2.



(a) Polarization curves at 0 (case 1) and 10 000 hours (case 10).



(b) Polarization curves with no area degradation showing varying operation temperatures



(c) Polarization curves with no area degradation comparing membrane humidity

Figure 12: Polarization curves from GT-Suite.

5 Discussion

In order to be able to draw conclusions regarding the fuel cell stack performance and durability, the results must be considered. The results of the Ostwald ripening simulation and the GT-Suite simulation are discussed. Then the connection between the models is investigated followed by a re-connection to the aim and client needs. Furthermore, the reliability of the results is discussed along with possible error sources. Lastly, future developments are investigated.

5.1 Catalyst layer degradation due to Ostwald ripening

The graphs in Figure 11 show a steady downward trend for power output, which relates to the performance of the CL and the ECSA reduction. It is clear from the graph that unsteady voltage puts an extra toll on the fuel cell and results in worse power output, as expected. At about 8000 hours the simulation becomes unreliable and does not show any results at that point. This could mean that degradation of ECSA becomes too great at that point that it is no longer possible to simulate power output or that there are other unknown problems with the simulation. However, continuing the linear trend proposed earlier, the degradation after 10 000 can be estimated to be 22%. The fact that the power output is larger and more steady in the case of steady voltage in Figure 11 is what could be expected since the steady voltage is meant to mimic an ideal drive-cycle while the unsteady voltage is meant to mimic a more realistic scenario.

In this project, EOL for the catalyst layer was defined as ECSA reaching 25% of the original surface area. After 10 000 hours in ideal conditions, it reached only 58% which means that the fuel cell could definitely be used for longer than that and still be considered functional. However, the case with unsteady voltage is more true to real-life conditions and therefore more relevant to the client. In the case of varying voltage, the area reduction reached 66%, which is larger but still far from the EOL defined by 25%. Though further investigation is needed in order to determine whether the catalyst layer can be considered functional during the full lifetime of a heavy-duty vehicle.

However, even when considering the non-ideal conditions, that is still an optimistic estimate of its mid-life. Since only the Ostwald ripening was modeled and no other Platinum related degradation mechanisms, such as platinum accumulation on the PEM that limits the passage of protons, this mid-life is still higher than in reality. The reason that Ostwald ripening was in particular chosen for this project was mostly because it was fairly easy to simulate given the teams' limited knowledge of GT-Suite since it only required changing the ECSA. It would definitely be interesting to further investigate how these other mechanisms, in combination with Ostwald ripening, degrade the cell given a better understanding of the software.

It is also important to note that the Matlab model is simplified in regards to particle size; it only considers two particle sizes, slightly smaller than average and slightly larger than average. In reality, there is an entire spectrum of different sizes which would also make the computations much more difficult and long. Particles bigger than the biggest size in this model definitely occur in real life, which would mean an even bigger decrease in ECSA and therefore even shorter lifespan for the fuel cell. Authors of the paper that the two-particle model was taken from concluded that the model is still pretty accurate compared to experimental data. Those small differences could still make an impact on the GT-Suite model and should therefore still be taken into consideration.

5.2 Polarization curves

The polarization curves in Figure 12 display cell voltage under different operating conditions and at different current densities. Independent of curve and case there is a voltage drop at the beginning of the curve, which is reasonable since initial losses are expected as described in section 2.1.

What distinguishes the different curves is Figure 10 is instead the gradient. In Figure 12a the linear drop in voltage is greater for the curve representing the initial state than the one representing 10 000 hours of run time. This is a reasonable outcome as the linear drop represents ohmic losses, which are due to the electronic and ionic conduction. If the area where the electrochemical reaction occurs is larger the electronic and ionic conduction are going to be greater resulting in higher losses. Form Figure 12b follows that there are fewer losses for higher cell operating temperatures. The figure shows that the most significant effect of a lower temperature is at the rapid drop that ours at low current densities (0-1000 A/m^2). Temperature can also be seen to affect the linear drop but not as dramatically. This tells us that temperature has the most effect on activation losses and some effect on ohmic losses. Similar results were attained for the humidity in Figure 12c with higher humidity decreasing voltage loss. The curves are similar initially, showing no effect on activation losses, but the difference in the linear drop is significant signaling humidity primarily affects the ohmic voltage losses.

5.3 Model integration

Since the GT-Suite model integrates values from the Matlab simulation the results from the GT-Suite simulation are heavily dependent on the validity of the Ostwald model. One advantage of this method is that the effect of more advanced degradation phenomena could be simulated with GT-Suite even with the limited prerequisites about using the software and timeline for the project. If all simulations of degradation were made in GT-Suite, the simulation time would be heavily increased and a lot of computational power would be spent on simulations of the flow fluid dynamics, which is not the main focus of the project. On the other hand, the fact that the simulations are integrated gives small errors in the Matlab simulation the risk of creating larger errors in the GT-Suite simulation, this is unlikely because of the simplicity of the integration, but the risk should not be overlooked.

5.4 Evaluation of aim & client needs

The fuel cell and the catalyst layer have been modeled in Matlab and GT-Suite for 10 000 hours and analyzed. Graphs showing clear deterioration of the fuel cell's power output with time have been produced and discussed. Therefore, for this project, the aim and client needs have been satisfied.

The client wanted to observe the fuel cell studied over at least 10 000 hours. From Figure 11 follows that the performance of the cell decreases over time, but still keeps about 83% of the original performance after 10 000 hours in the case of steady voltage. In the case of unsteady voltage, there is a huge voltage drop after about 8 000 hours since the results become unreliable. Table 3 also follows that the area reduction is much more significant in the case of unsteady voltage than in the case of steady voltage, which could be expected due to the mimicking of ideal and non-ideal drive cycles. Great area reductions are undesired and therefore it might be the case that the performance of the fuel cell becomes too poor after 8 000 hours in the case of unsteady voltage. This means that the fuel cell, in the case with unsteady voltage, most probably does not meet the requirements of the client for the 10 000 hours mid-life. As stated above, the sharp decline in power output in Figure 11 could be due to another phenomenon than major degradation of the ECSA, there might still be the hope of achieving the 10 000-hour mid-life. Further tests and simulations are, however, needed to confirm that.

5.5 Reliability and error sources

Firstly, it is important to realize that a simulation is a simplification of reality and should be viewed as a prediction. The model may not capture all physical and chemical reactions that are happening within the fuel cell or in its surrounding environment. With more accurate data and more advanced modeling, the model will be more computationally expensive and difficult to validate when more parameters need to be taken into account. To strike the right balance, one can compare the model´s data to experimental data and verify if the result aligns with the real-time experiment.

Secondly, apart from the fact that a simulation is a simplification of reality and thereby may not capture all effects in the surrounding environment. The time frame of the project is relatively short and the team had no prerequisites for GT-Suite modeling, which limited the refinement of the model. For clarity, it is also worth mentioning that the team lost the GT-Suite license for ten days during a critical time in the project, which due to the time frame affected the possibility to create and refine the model.

Thirdly, the model's input values were estimated from a literature study of existing data, see Appendix B. Volvo could not provide precise data due to trade secrets, which makes the model less reliable. This discrepancy can have a significant impact on the result if the values differ in the same direction, and less impact if that is not the case, making it a black box. Even the input values of the boundary conditions can have a huge impact on the final model compared to the conditions that the real-time fuel cell will be exposed to. To reduce this error and make more reliable results, a sensitivity analysis can be calculated and integrated into the model.

Finally, the possibility of erroneous input data is not only present in the GT-Suite model, but in the Matlab model as well. Like in the case of the GT model a sensitivity analysis could be made on the Matlab model in order to check the impact of the results. If the simulations prove to be sensible, one must make sure to include the sensitivity of the area reduction in the sensitivity analysis of the GT-Suite simulation. Since no sensitivity analysis has been made and no data has been provided from the client, more verification is needed in order to draw accurate and applicable conclusions from the results.

5.6 Future development

The research carried out before modeling started showed that there are numerous different ways to degrade a fuel cell and given the timetable of the project, there was no opportunity to model them all. Therefore the model built has examined some different mechanisms that affect the performance of the fuel cell stack, with Ostwald ripening being the most suited. This gives an idea of what one mechanism can do to the cell and a hint at what you could expect when modeling a different mechanism. For future research, the first step would be to further improve the current model to observe if it is possible to obtain smaller losses by refining the data and parameters of the simulated cell. It would also be of interest to model the cell to give 150kW after 10 000 hours of degradation, instead of having it give the required power only when not exposed to any degradation.

Implementing more mechanisms with higher detail would be the final goal of further research as the model then could be compared with a real fuel cell which could solidify the theory and methods used in the simulation.

To continue the research, Chalmers needs to update the GT-Suite software to version 2023 or newer in order to access the new and more refined solvers and templates. This would provide the team with more opportunities to assist Volvo and explore further possibilities in the detailed modeling of degradation mechanisms. External software, such as Matlab, might not be required, allowing the project to be conducted within a single software environment.

6 Conclusions

In summary, the degradation of fuel cells in heavy-duty applications has been shown to depend on whether the voltage is steady or unsteady when investigating the Ostwald ripening phenomenon. An unsteady voltage results in a more significant reduction of the effective catalyst surface area (ECSA) and degradation over time, resulting in a loss of performance. The simulations indicate that, in this case of unsteady voltage, there is no data for power output after 8 000 hours. Therefore further refinement of the model is required in order to make it work for the 10 000 hours. In contrast, the steady voltage curve does not have his problem and shows a result for the full 10 000 hour time span. Therefore, a fuel cell that works under those conditions can have a 10 000 hour mid-life during real-time conditions.

Since the Ostwald ripening phenomenon is a specific degradation mechanism, numerous other mechanisms should be investigated to gain more knowledge about the overall performance of fuel cells. Although membrane humidity and cell temperature have been studied in this project and shown to have an impact on the performance, more underlying mechanisms that affect the degradation of the fuel cell should be investigated to obtain more accurate results. Taking this into consideration, this study concludes the necessity of further development and research for the fuel cell to become a potential long-term solution for heavy-duty vehicles.

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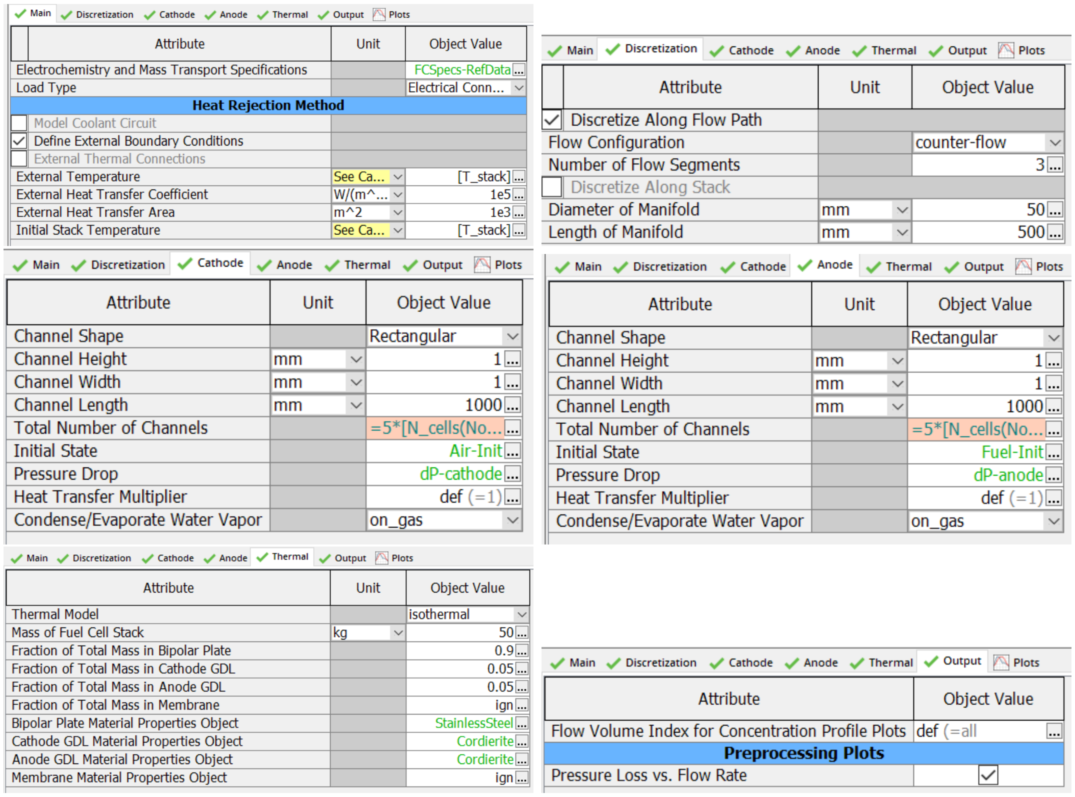
A Ostwald ripening Model

Parameter	**Physical Meaning**	**Value**
u_θ	Platinum Dissolution Reference Rate Constant	2.753e17
M_{pt}	Platinum Molar Mass	195084 mg/mol
ρ_{pt}	Platinum Density	21540 mg/cm^3
β_r	Constant Modeling the Change in Larger Particle Radius Relative to the Current Average Particle Radius	0.0038
α_r	Constant Relating the Decrease in the Smaller Particle Radius as a Multiple of the Larger Particle Radius Increase	1.1
k_1	Platinum Dissolution Reaction Rate Coefficient	1e-10
Γ_{max}	Moles of Active Sites per Unit of Platinum Area	2.18e-9 mol/cm^2

B GT-Suite setup and parameters

All values within parenthesis are shown in Figure 15. An explicit solver was used and the Active Surface area scaling factor (ASASF) differs between each case. Additional setup windows that are now shown here were kept as default for the example model used as the base (Figure 17).



	Attribute	Unit	Object Value
	Electrochemistry and Mass Transport Specifications		FCSpecs-RefData
	Load Type		Electrical Conn...
	Heat Rejection Method		
☐	Model Coolant Circuit		
☑	Define External Boundary Conditions		
☐	External Thermal Connections		
	External Temperature	See Ca...	[T_stack]
	External Heat Transfer Coefficient	W/(m^...	1e5
	External Heat Transfer Area	m^2	1e3
	Initial Stack Temperature	See Ca...	[T_stack]

	Attribute	Unit	Object Value
☑	Discretize Along Flow Path		
	Flow Configuration		counter-flow
	Number of Flow Segments		3
☐	Discretize Along Stack		
	Diameter of Manifold	mm	50
	Length of Manifold	mm	500

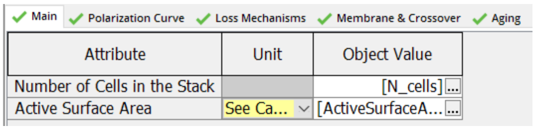
Attribute	Unit	Object Value
Channel Shape		Rectangular
Channel Height	mm	1
Channel Width	mm	1
Channel Length	mm	1000
Total Number of Channels		=5*[N_cells(No...
Initial State		Air-Init
Pressure Drop		dP-cathode
Heat Transfer Multiplier		def (=1)
Condense/Evaporate Water Vapor		on_gas

Attribute	Unit	Object Value
Channel Shape		Rectangular
Channel Height	mm	1
Channel Width	mm	1
Channel Length	mm	1000
Total Number of Channels		=5*[N_cells(No...
Initial State		Fuel-Init
Pressure Drop		dP-anode
Heat Transfer Multiplier		def (=1)
Condense/Evaporate Water Vapor		on_gas

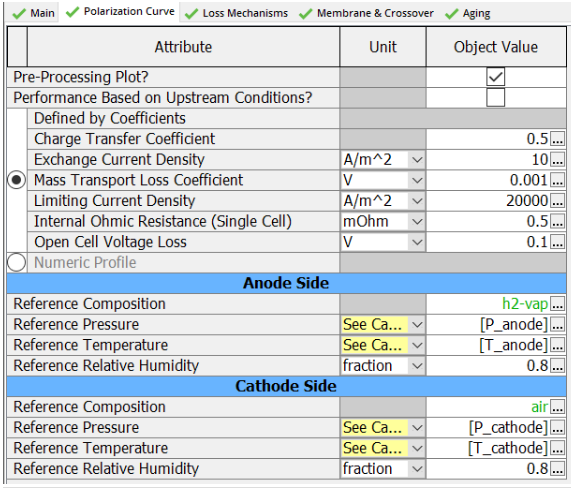
Attribute	Unit	Object Value
Thermal Model		isothermal
Mass of Fuel Cell Stack	kg	50
Fraction of Total Mass in Bipolar Plate		0.9
Fraction of Total Mass in Cathode GDL		0.05
Fraction of Total Mass in Anode GDL		0.05
Fraction of Total Mass in Membrane		ign
Bipolar Plate Material Properties Object		StainlessSteel
Cathode GDL Material Properties Object		Cordierite
Anode GDL Material Properties Object		Cordierite
Membrane Material Properties Object		ign

Attribute	Object Value
Flow Volume Index for Concentration Profile Plots	def (=all
Preprocessing Plots	
Pressure Loss vs. Flow Rate	☑

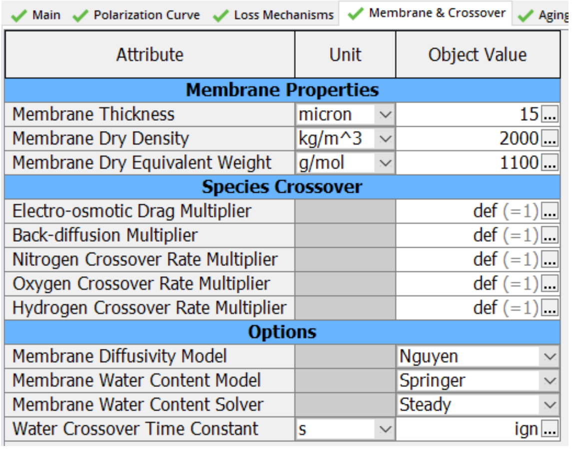
Figure 13: Fuel cell template setup



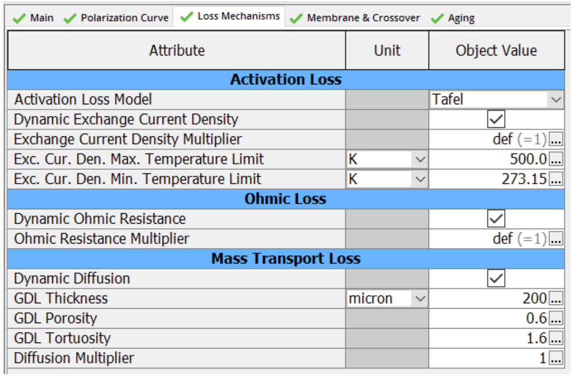
Attribute	Unit	Object Value
Number of Cells in the Stack		[N_cells]
Active Surface Area	See Ca...	[ActiveSurfaceA...



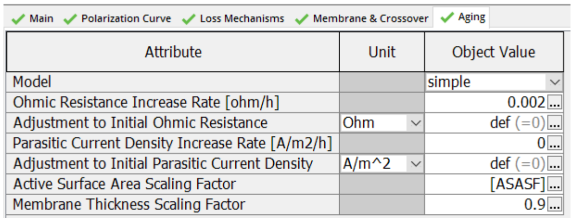
	Attribute	Unit	Object Value
	Pre-Processing Plot?		☑
	Performance Based on Upstream Conditions?		☐
	Defined by Coefficients		
	Charge Transfer Coefficient		0.5
	Exchange Current Density	A/m^2	10
●	Mass Transport Loss Coefficient	V	0.001
	Limiting Current Density	A/m^2	20000
	Internal Ohmic Resistance (Single Cell)	mOhm	0.5
	Open Cell Voltage Loss	V	0.1
○	Numeric Profile		
	Anode Side		
	Reference Composition		h2-vap
	Reference Pressure	See Ca...	[P_anode]
	Reference Temperature	See Ca...	[T_anode]
	Reference Relative Humidity	fraction	0.8
	Cathode Side		
	Reference Composition		air
	Reference Pressure	See Ca...	[P_cathode]
	Reference Temperature	See Ca...	[T_cathode]
	Reference Relative Humidity	fraction	0.8



Attribute	Unit	Object Value
Membrane Properties		
Membrane Thickness	micron	15
Membrane Dry Density	kg/m^3	2000
Membrane Dry Equivalent Weight	g/mol	1100
Species Crossover		
Electro-osmotic Drag Multiplier		def (=1)
Back-diffusion Multiplier		def (=1)
Nitrogen Crossover Rate Multiplier		def (=1)
Oxygen Crossover Rate Multiplier		def (=1)
Hydrogen Crossover Rate Multiplier		def (=1)
Options		
Membrane Diffusivity Model		Nguyen
Membrane Water Content Model		Springer
Membrane Water Content Solver		Steady
Water Crossover Time Constant	s	ign

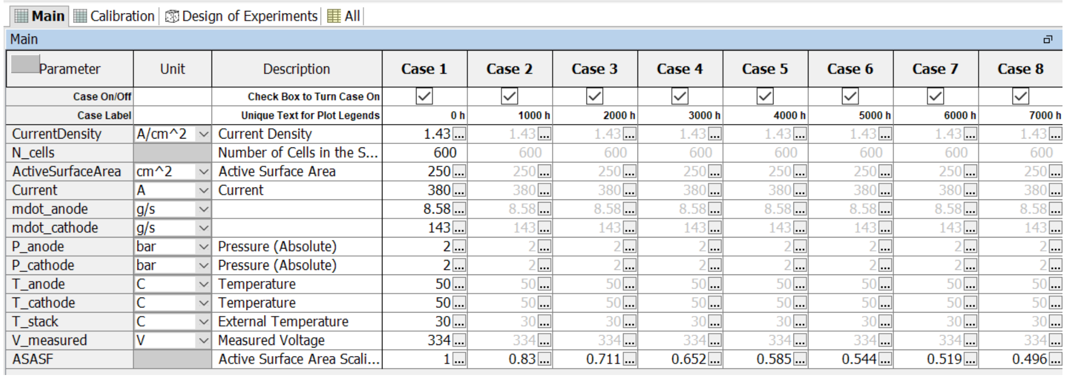


Attribute	Unit	Object Value
Activation Loss		
Activation Loss Model		Tafel
Dynamic Exchange Current Density		☑
Exchange Current Density Multiplier		def (=1)
Exc. Cur. Den. Max. Temperature Limit	K	500.0
Exc. Cur. Den. Min. Temperature Limit	K	273.15
Ohmic Loss		
Dynamic Ohmic Resistance		☑
Ohmic Resistance Multiplier		def (=1)
Mass Transport Loss		
Dynamic Diffusion		☑
GDL Thickness	micron	200
GDL Porosity		0.6
GDL Tortuosity		1.6
Diffusion Multiplier		1



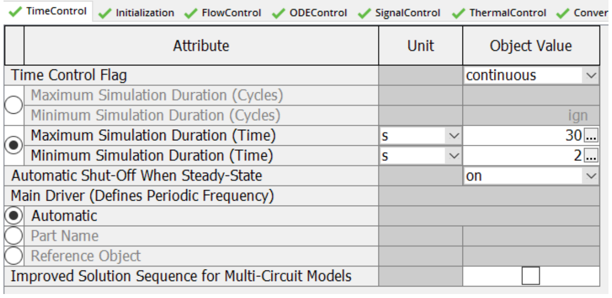
Attribute	Unit	Object Value
Model		simple
Ohmic Resistance Increase Rate [ohm/h]		0.002
Adjustment to Initial Ohmic Resistance	Ohm	def (=0)
Parasitic Current Density Increase Rate [A/m2/h]		0
Adjustment to Initial Parasitic Current Density	A/m^2	def (=0)
Active Surface Area Scaling Factor		[ASASF]
Membrane Thickness Scaling Factor		0.9

Figure 14: Reference data setup



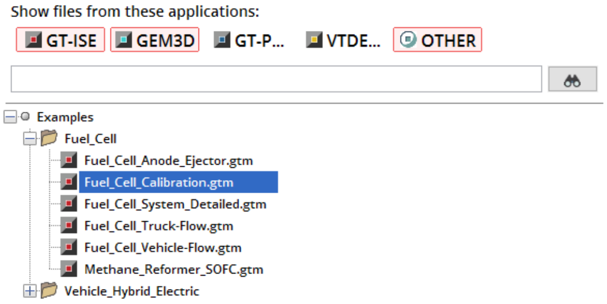
Parameter	Unit	Description	Case 1	Case 2	Case 3	Case 4	Case 5	Case 6	Case 7	Case 8
Case On/Off		Check Box to Turn Case On	☑	☑	☑	☑	☑	☑	☑	☑
Case Label		Unique Text for Plot Legends	0 h	1000 h	2000 h	3000 h	4000 h	5000 h	6000 h	7000 h
CurrentDensity	A/cm^2	Current Density	1.43	1.43	1.43	1.43	1.43	1.43	1.43	1.43
N_cells		Number of Cells in the S...	600	600	600	600	600	600	600	600
ActiveSurfaceArea	cm^2	Active Surface Area	250	250	250	250	250	250	250	250
Current	A	Current	380	380	380	380	380	380	380	380
mdot_anode	g/s		8.58	8.58	8.58	8.58	8.58	8.58	8.58	8.58
mdot_cathode	g/s		143	143	143	143	143	143	143	143
P_anode	bar	Pressure (Absolute)	2	2	2	2	2	2	2	2
P_cathode	bar	Pressure (Absolute)	2	2	2	2	2	2	2	2
T_anode	C	Temperature	50	50	50	50	50	50	50	50
T_cathode	C	Temperature	50	50	50	50	50	50	50	50
T_stack	C	External Temperature	30	30	30	30	30	30	30	30
V_measured	V	Measured Voltage	334	334	334	334	334	334	334	334
ASASF		Active Surface Area Scali...	1	0.83	0.711	0.652	0.585	0.544	0.519	0.496

Figure 15: Case Setup



	Attribute	Unit	Object Value
Time Control Flag			continuous
○	Maximum Simulation Duration (Cycles)		
	Minimum Simulation Duration (Cycles)		ign
●	Maximum Simulation Duration (Time)	s	30
	Minimum Simulation Duration (Time)	s	2
Automatic Shut-Off When Steady-State			on
Main Driver (Defines Periodic Frequency)			
●	Automatic		
○	Part Name		
○	Reference Object		
Improved Solution Sequence for Multi-Circuit Models			☐

Figure 16: Simulation time settings


Show files from these applications:

- Examples
  - Fuel_Cell
    - Fuel_Cell_Anode_Ejector.gtm
    - Fuel_Cell_Calibration.gtm
    - Fuel_Cell_System_Detailed.gtm
    - Fuel_Cell_Truck-Flow.gtm
    - Fuel_Cell_Vehicle-Flow.gtm
    - Methane_Reformer_SOFC.gtm
  - Vehicle_Hybrid_Electric

Figure 17: Where to find example model used as starting point

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