A review of modeling pem fuel cells for monitoring applications

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Abstract: *Low operating temperature, compactness, high efficiency as well as low to zero emissions are features that cause large interest in Proton Exchange Membrane (PEM) fuel cells and are reasons that application of this technology is considered in many areas. However, for a massive deployment of the PEM fuel cell technology to the market, good control and monitoring are mandatory to increase efficiency and durability. For the control and monitoring of PEM fuel cell systems, appropriate system models are required. In this study, a review of modeling approaches to the PEM fuel cell systems is considered.*

***Keywords**: PROTON EXCHANGE MEMBRANE, FUEL CELL, HYDROGEN, MODELING*

1 Introduction

A fuel cell is a device that uses an electrochemical reaction to produce electrical energy from the chemical energy of fuel. In some aspects, a fuel cell is similar to a battery. Both a fuel cell and a battery have positive and negative electrodes accompanied by an electrolyte and both generate electricity through electrochemical reactions. Unlike a battery, a fuel cell requires an auxiliary supply of fuel and oxidant. Also, a battery can only function as long as the electrodes' material is not depleted. Then, it can be either replaced or recharged while a fuel cell cannot be discharged as long as reactants are supplied [1][2]. Typically hydrogen and oxygen are used for fuel cells but neither has to be in its pure form. Hydrogen may be also present as a mixture with other gases. It has been considered as the best effective fuel for quite a long time, as it features a higher electrochemical reactiveness compared to other fuels (hydrocarbons, alcohols) [3].

Fuel cells have a long history, dating back to 1839 when Sir William Robert Grove demonstrated the first fuel cell showing that electricity and water can be generated by mixing hydrogen and oxygen in the presence of an electrolyte. Another remarkable moment marking the fuel cell history is when the first practical fuel cell was developed by General Electric Company in 1962 [1][4]. Despite the long period of fuel cell analysis and examinations, there is still an ongoing introduction to the energy market because of the challenges the technology is facing. The major obstacles are cost and durability. A causality problem appears with the fuel cell price. One of the reasons the fuel cells are expensive is because they are not being mass-produced while their limiting production is due to their expensive price. Thankfully, the cost of fuel cells customized for fuel cell electric vehicles went to a reduction due to the continuous decrease in the platinum loading in PEM stacks [2].

In recent years, fuel cell technology is being used in various areas such as transportation, back-up power, portable devices, stationary devices, etc. Mostly, the technology has been present in the transportation area so that numerous models of vehicles have been produced. According to the International Energy Agency (IEA), most of the sales continue to be Toyota Mirai cars in California, following by Japan, Korea, and Germany. The increase in the number of sales contributes to expansion in the refueling infrastructure as well. At the end of 2018, 376 hydrogen refueling stations were in operation and leading countries have announced a target to build a total of 1000 hydrogen refueling stations during the years of 2025 - 2030 [5]. Without a doubt, the technology for the production and supply of high-purity hydrogen along with the infrastructure is essential for the success of PEM fuel cells in many application fields. A broad review and evaluation of hydrogen production methods for better sustainability are made in [6].

Modeling plays a significant role in the fuel cell design and development process, as it helps the designer narrow down designs to fabricate and test. The test performed on the final designs can result in either a final prototype or an iteration of additional designs for improvement. Using fuel cell modeling as a successful design tool requires the model to be accurate, robust, and able to provide useful answers quickly. Accuracy can be improved by using the correct assumptions, properties, and other numerical input parameters, predicting the correct objective, and being able to match the modeling results with experimental data. But, enhancing model robustness and accuracy can lead to bigger complexity and longer computational time. That's why the designer needs to select a model that balances robustness, accuracy, and computational effort.

Fuel cells can be divided into several categories, characterized primarily by the fuel, type of electrolyte, and operating temperature [1][3]. There are six main different types of fuel cells:

- Alkaline fuel cells (AFC);
- Proton Exchange Membrane fuel cells (PEMFC);
- Direct Methanol fuel cells (DMFC);
- Phosphoric Acid fuel cells (PAFC);
- Molten Carbonate fuel cells (MCFC) and
- Solid Oxide fuel cells (SOFC).

Some of their characteristics are summarized in **Грешка! Източникът на препратката не е намерен.**, while a more detailed description can be found in [1],[2], and [7]. This study focuses on PEM fuel cells.

Table 1. *Basic parameters of fuel cells* [8].

Type	Operating temperature (°C)	Generated voltage (V)	Power	Used fuel
AFC	65-220	1,1-1,2	to 20kW	$H_2 + O_2$
PEMFC	50-120	1,1	kW	H_2/methanol + O_2/air
DMFC	130	1,1	to 10kW	methanol/ethanol + O_2/air
PAFC	150-210	1,1	hundreds kW	H_2/hydrogenous gas + air
MCFC	600-700	0,7-1,0	MW	H_2/hydrogenous gas/CO + air
SOFC	650-1000	0,8-1,0	to 10 MW	H_2/hydrogenous gas/CO + air

2 PEM fuel cells basics

The core component of a PEM fuel cell is the polymer membrane which acts as an electrolyte. It is impermeable to gases but it conducts protons. The membrane becomes proton conductive when it is well hydrated while remaining insulated to gas transport [9]. A common electrolyte material is Nafion [10]. As technology is evolving, research is being done on every aspect of the fuel cells. Recent approaches for improving Nafion performance are evaluated in [11].

On both sides of the membrane, there are two porous, electrically conductive electrodes made usually out of carbon cloth or carbon fiber paper [12]. At the interfaces of the electrodes and the polymer membrane, there are layers with catalyst particles called the catalyst layers. Technically, the catalyst layer may be a part of the porous electrode or a part of the membrane, depending on the manufacturing process [1]. The catalyst layer consists of three phrases. Fine catalyst particles of platinum are supported on complex structures of carbon particles that provide electronic conduction. This carbon-supported platinum catalyst creates a large surface catalyst area so that the reaction can proceed at a feasible rate [10]. The electrodes must be porous so the reactant gases reach the catalyst layers where the electrochemical reactions take place. Reactants spread from gas channels to catalyst layers through the gas diffusion layers. The gas diffusion layers are made from a wet-proof and carbon-based material, controlling the heat, mass, and electricity transport while providing robust mechanical support and protection for the delicate catalyst layer and membrane through the process of assembly and operation [13].

The membrane, the catalyst layers, and the gas diffusion layers form the membrane electrode assembly (MEA). The MEA is then inserted between bipolar plates, also known as collector plates, which are essential for multi-cell configuration by connecting the anode of one cell to the cathode of the adjacent cell. A multi-cell configuration is also known as a 'stack'. The bipolar plates perform important functions, providing the structural support for the stack and separating the gases in adjacent cells, which implies they must be porous to gases [14]. They have been made from graphite and various metals to have the desired resistance to corrosion [10]. The structure of a PEM fuel cell is presented in Figure 1.

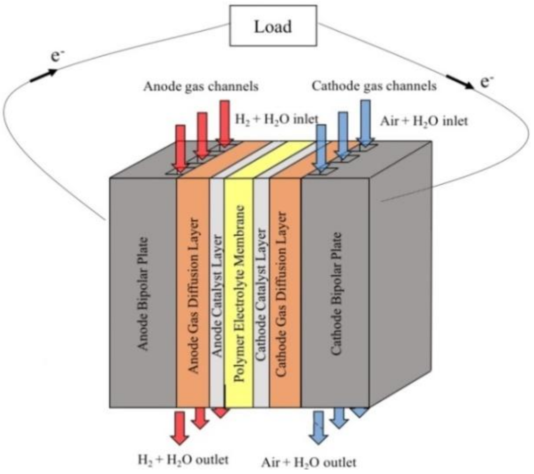

Figure 1. *Structure of a PEM fuel cell* [12].

Inside a PEM fuel cell, a few processes are carried out, such as gas flow through the channels, electrochemical reactions, proton transport, electron conduction, water transport, etc. The design of the components and properties of materials must be adapted for these processes to take place with minimum obstructions and losses. Although a fuel cell system seems like a very simple device, numerous processes happen simultaneously, and for that reason, it is very important to understand those processes and their dependence on component design and material properties. It is also crucial to know that changing only one parameter in a fuel cell is not possible because a change of one parameter causes a change in at least two other parameters and at least one of them has the opposite effect of the one we expect to see.

2.1 Applications

Fuel cells have a promising potential of becoming competitive players in different areas as a result of their ability to generate power from a chunk of a watt to hundreds of kilowatts. Such areas are transportation, stationary, and portable power. Fuel cell system design is not inevitably the same for each of these applications. On the contrary, each application has its specific requirements, such as efficiency, water balance, quick startup, size, weight, and fuel supply.

Among the application areas, transportation is the most promising and competitive. This is because the transportation industry is responsible for 17% of global gas emissions every year and fuel cells offer near-zero emissions without having to compromise the efficiency of the vehicle's propulsion system. A broad investigation of hydrogen fuel cell vehicles is done in [15]. Some car manufactures have already produced their fuel cell vehicle models, for example, Toyota, Honda, Hyundai, and Chevrolet. The United States alone currently has over 10000 fuel cell vehicles and over 30 hydrogen stations. Germany possesses around 100 hydrogen gas stations, while Japan has a roadmap of 300000 fuel cell electric vehicles by the year of 2030. China's presence in fuel cell electric vehicles expanded significantly in 2018, with up to 2000 small trucks produced. However, as these vehicles wait for the corresponding refueling infrastructure, just 400 were registered for road use in 2018 [4]. While the deployment of fuel cell electrical vehicles is low compared to plug-in hybrids, several countries have announced ambitious targets towards 2030, amounting to 2.5 million fuel cell electric vehicles [5]. When compared to other fuel cell-powered vehicles, buses provide more flexible design, more flexible weight, and size constraints for hydrogen storage system, and less complex hydrogen infrastructure requirements since bus routes are usually fixed. Figure 2 shows the main components in a typical fuel cell-powered bus based on the Mercedes-Benz Citaro Fuel EcoBus. The bus uses the space flexibility on the roof, front, and back of the bus.

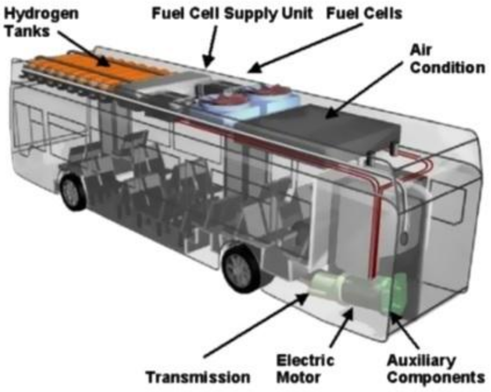

Figure 2. *Main components in a typical fuel cell bus based on the Mercedes-Benz Citaro Fuel Cell EcoBus [8].*

Other vehicles, including scooters, personal wheelchairs, airport tugs, bicycles, golf carts, in addition to material handling vehicles, including forklifts and pallet trucks, can be comprised of fuel cell systems as well [2]. Most forklifts use rechargeable batteries or combustion engines. However, fuel cells have many advantages over the current forklifts energy systems. Fuel cells require only 2-5 minutes for refueling from a fueling station unlike recharging or changing batteries which takes 15-30 minutes. As being exhaustively used in the warehousing and distribution industry, their operation time is very important and fuel cells have longer operation cycles in contrast to battery cycles which often last less than 8 hours [16]. With having high efficiency too, they can potentially replace conventional forklifts. Around 1300 fuel cell-powered forklifts are operative in the US market today. The biggest player in the fuel cell forklift market is Plug Power [2]. In regards to vehicle fuel cell approaches, more details about fuel cell-powered forklifts, fuel cell-powered electric wheelchairs, and electric bicycles are demonstrated in [17], [18], and [19] respectively.

Speaking of stationary power, the desires for both stationary and transportation markets are similar: high efficiency and low emissions. The system design is also similar, differing in some features. Size and weight requirements are more important in the transportation area, but not so significant in the stationary area. The acceptable noise level is lower in stationary applications, especially if the device is planned for indoor use. Important notice is that the noise does not arise from the fuel cell itself but from the air and fluid handling devices [1]. Starting time is crucial in the transportation area whereas time limits do not exist in the stationary area unless the system is operated as a back-up. Besides, while the car operating time is expected to be 3000 – 5000 hours and slightly longer for a bus, stationary fuel cell-powered systems are expected to have a lifetime of 40000 – 80000 hours (5 to 10 years) [1]. To power the off-grid radio base stations, Telekom Italy provided a reliable solution of an integrated system including three different energy sources: photovoltaic panels, battery packages, and fuel cells. This is a potential solution replacing the currently Diesel generators powering their off-grid radio base stations [20].

Portable applications for fuel cells are mainly divided into two categories: portable power generators and military devices. The military market is particularly attractive because most of the time it is adopter for new technologies, willing to accept higher prices and limited performance if the main strict requirements are met [1]. The development of small fuel cell systems for portable power applications has resulted in a wide variety of stack configurations. Some systems are miniaturized replicas of the larger automotive or stationary power fuel cells with the same components.

2.2 *Review of main physical processes and their governing equations*

A fuel cell is provided with simultaneous supply of fuel gas and oxidant. Hydrogen is supplied in the form of H_2 molecules on the anode side and oxygen is supplied in the form of O_2 molecules on the cathode side. The electrochemical reactions also take place simultaneously at the catalyst layer surfaces on both sides of the fuel cell. In general, an electrochemical reaction involves either oxidation or reduction of the gases.

After hydrogen is supplied, it spreads through the anode gas diffusion layer to the anode catalyst layer. The contact of hydrogen molecules with the platinum catalyzer causes a reaction where hydrogen molecules break into individual atoms of H first and further to protons and electrons, as shown in (1). The reaction on the anode side is called hydrogen oxidation.

$$2H_2 \rightarrow 4H^+ + 4e^- \tag{1}$$

After oxidation, protons move through the proton conductive membrane, while electrons are subject to the external electric load and they are received at the catalyst surface on the cathode side through the bipolar plates. While moving through the membrane protons attach onto water molecules forming hydronium complexes H_3O^+ that move through the membrane from the anode to the cathode. This process is called electro-osmotic drag. On the cathode side, supplied oxygen diffuses to the catalyst layer. Once the hydrogen proton has passed through the membrane, it is electrochemically combined with oxygen and electrons to form water, as follows:

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

The reaction on the cathode side is called oxygen reduction. Water that travels from the cathode to the anode due to a large concentration gradient across the membrane is called back-diffusion transport. These simultaneous reactions result in current of electrons through an external circuit. The whole fuel cell operation scheme is shown in Figure 3.

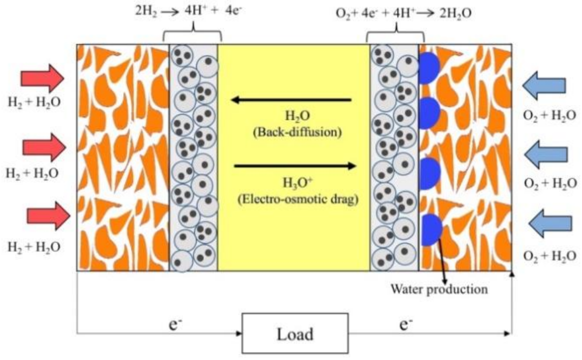

Figure 3. *PEM fuel cell operation scheme* [12].

3 *Modeling approaches*

Modeling is necessary to describe all the fundamental phenomena even though the complicated processes make the modeling task particularly challenging. Models are also used to predict fuel cell performance under different operating conditions. Because of the complexity, assumptions are needed to simplify the model and focus on a certain mechanism or a limiting case. It is important to understand the assumptions in order to understand the model's limitations and accurately interpret its results.

There are different types of PEM fuel cell models depending on the purpose they serve. One group of models focuses on a single cell or specific parts of the fuel cell, such as the gas channel, the membrane, the catalyst layer, or the gas diffusion layer. Stack-level models consider the arrangements of more than one cell. System-level models are focused on the entire fuel cell stack and the auxiliary components that form a complete fuel cell system. Models can be also classified depending on the dimensionality: one, two, or three dimensional. Two-phase flow models or single-phase flow models can also be differentiated, whether liquid water formation within the cell is a phenomenon of interest [12].

3.1 *Single-cell models*

Single-cell models describe the electrochemical and transport processes in each fuel cell component. In [21], PEM fuel cell model is developed using a recurrent neural network. It is shown that the nonlinear dynamics of the PEM fuel cell can be effectively modeled using a two-layer recurrent neural network for later analysis of the fuel cell behavior. Rakesh et al. [22] presented a one-dimensional single-phase model of diffusion in a gas diffusion layer of PEM fuel cell. During the operation of a fuel cell, gas, water vapor, and liquid water get transported at different rates through the gas diffusion layers, and the authors aim is to build a more realistic model considering tortuosity and Knudsen diffusion mechanism for a deeper understanding of the process happening in the GDLs. Authors in [23] focus specifically on a one-dimensional model predicting the cell performance based on oxygen diffusion at the cathode catalyst layer. They conclude that PEM fuel cell performance increases by increasing cathode catalyst layer pore size and pore volume.

One-dimensional models are used in stack-level modeling as well. Gao et al. [24] developed a one-dimensional model where the fuel cell stack is considered as a cell-level structure, each cell is then divided into a layer level, and each level is modeled as an independent control volume.

However, one-dimensional fuel cell models ignore spatial physical characteristics such as reactants pressure drop or non-uniform current density distribution. The development of a multidimensional model is very valuable, but additional spatial information means more computational burden. In [25], a two-dimensional model development process is explained while presenting and improving three numerical algorithms and making performance comparison. Authors in [26] develop a three-dimensional numerical model employing the volume of the fluid method to simulate the two-phase flow in the cathode gas channel with the electrochemical reactions and water balance in the membrane. They observe that lower performances can be predicted at high currents showing that the model can compute the effects of water flooding. Speaking of two-phase models, Grötsch et al. [27] present a reduced two-phase model based on lower computational time with reasonable accuracies for more suitable process control purposes. An important three-dimensional, two-phase model was developed by Tao et al. to perform parameter sensitivity examination [28].

3.2 *Stack and system-level models*

Usually, stack and system-level models are more frequent. They are lumped parameter models used to evaluate fuel cell performance under different operating conditions. A lumped system is a system where the variable of interest is a function of time [29]. Pukrushpan et al. [30] developed a system-level model that includes the fuel cell stack, the hydrogen supply system where hydrogen is supplied from high-pressure tank, the air supply system, the cooling system, and the humidification system. The stack temperature is considered constant because of the slow dynamics of this variable compared to the transient dynamics included in the model. There is another model from these authors, consisting of an integrated fuel cell stack and a fuel processor system [31]. Because hydrogen is not a cost-free fuel, exhausting without recirculation would bring disadvantages in the context of cost-efficiency. For that matter, authors in [32] propose a model of a fuel cell system with recirculation of both fuel and oxidant exhaust. Some stack and system-level models are developed with a certain focus. A numerically model was developed in [33] to study the dynamic behavior of a PEM fuel cell stack subjected to load changes. On the other hand, Deng et al. [34] focus strictly on the PEM fuel cell stack thermal system. In [35], the authors develop a one-dimensional model to study the dynamic behavior of PEM fuel cell stack operating in dead-end mode to explain the different performances between a "fresh" and "aged" stack. Simulations indicated that the liquid water accumulation is at the origin of the performances decrease with aging, due to its effect on decreasing the actual gas diffusion layer porosity that in turn causes the starving of the active layer with oxygen. Ondrejička et al. [8] focus on analyzing temperature effects on the performance and efficiency of a PEM fuel cell stack. Their model consists of three interconnected subsystems that are responsible for simulating electrochemical, thermodynamic, and mass transport effects that occur within the fuel cell stack. The diagram of the model is presented in Figure 4. The analysis shows that an increase in the temperature influences the activation losses the most, significantly increasing their value. On the contrary, ohmic losses are reduced which in turn causes an increase of nominal power output of the stack.

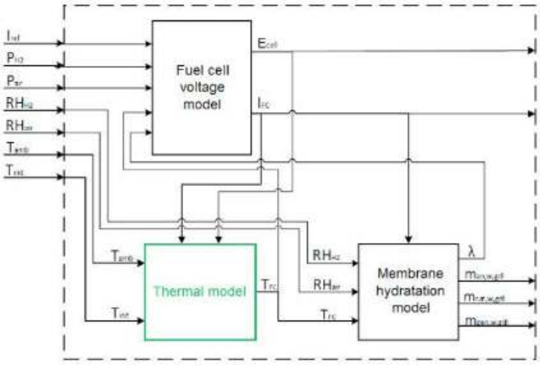

Figure 4. *Topological diagram of PEM fuel cell model* [8].

Many models that are made based on the 1.2 kW Nexa fuel cell system which contains Ballard fuel cell stack, shown in Figure 5. This module is considered as a benchmark and is widely used because of its simplicity compared to complex electrochemical models while still allowing prediction of the oxygen excess ratio (λ_{O2}), which is critical in fuel cell control design [36]. Authors in [36] develop a model that may help in evaluating the effects of the load dynamics on the PEM fuel cell system and improving its efficiency by using optimal operation analysis based on the λ_{O2} trajectory. Similarly, Saengrung et al. [37] achieved successful voltage and current prediction by using only two input variables, airflow and stack temperature. They have shown that back-propagation and radial basis function networks are capable of predicting the performance of a particular fuel cell system with satisfactory accuracy in a very short period of time.

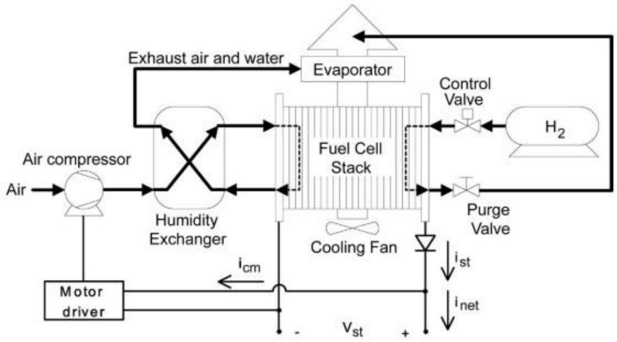

Figure 5. *1.2 kW Ballard Nexa module* [36].

4 Discussion

Performance, durability, and cost are the most important challenges for PEM fuel cells. Cost reduction efforts are being made within research fields that cover construction and assembly methods, component materials as well as improved fuel production, storage, and transport. On the other hand, the performance and durability of PEM fuel cells depend on the operating conditions, component materials, and inadequately captured challenges while modeling. Several challenges should be captured while modeling a PEM fuel cell:

- Fuel or oxidant starvation – Poor distribution of reactants can affect their flow which can result in fuel cell degradation and cell voltage drop. Authors in [38] specifically study the fuel cell behavior under oxidant starvation, showing that it highly affects the current distribution. To avoid this problem, proper reactant distribution is critical, monitored by the system controlling sensors and indicators.
- Thermal management – Thermal management is extremely important, especially when the stack is operated at extreme temperatures, like below 0°C or above 80°C. Several studies have shown that improper thermal management can lead to membrane or catalyst degradation. Automotive fuel cells must survive and operate in extreme temperatures from -40°C to +40°C. This requirement has a huge effect on system design. Survival and startups in extremely cold temperatures require specific engineering solutions, like the use of antifreeze coolants or water management. However, water cannot be completely eliminated because water is essential for the membrane proton conductivity.
- Water management – Water management is one of the most complex phenomena to maintain and for that reason is one of the most studied issues in PEM fuel cell technology. Proper water management requires meeting two conflicting requirements: adequate membrane hydration and avoidance of water flooding. Flooding can occur both at the anode and the cathode, but it is more likely to happen at the cathode since water is being produced at that side of the fuel cell. Flooding at the anode side is less frequent, but both have serious consequences in performance and degradation. On the other side, a shortage of water is more likely to occur at the anode side causing higher membrane proton resistance and voltage drop. In [39], authors have proposed diagnostics and prevention techniques, using two sensing electrodes at the fuel inlet and fuel outlet. The response is given before the cell voltage is affected which enables early diagnostics of those failure modes. Also, authors in [40] combine the pressure drop and cell resistance measurements to detect flooding or drying. They claim that a pressure drop increase is a reliable sign of increased water content in a fuel cell, while an increase in cell resistance may lead to cell drying.
- Cell exposure to impurities – Impurities in both fuel and oxidant intake have a significant effect on fuel cell performance and durability. It has been demonstrated that even very small amounts of impurities present in the fuel or air streams or fuel cell system components can severely poison the

anode, membrane, and cathode, particularly at low-temperature operation. The contaminants can strongly adsorb on the catalyst surface to block the reaction sites, enter the membrane to reduce proton conductivity, and cross over the membrane to affect the other side of the MEA [41]. In the automotive industry, even though fuel cells can meet the actual car operating time expectations, numerous startups and shutdowns, operation in various ambient conditions, impurities in fuel and air may have a dramatic effect on fuel cell operating lifetime.

- Degradation – During the lifetime of the PEM fuel cell stack, all the components are prone to degradation and their performances decrease. It is important to point out that degradation processes on each component do not affect the operation and the lifetime of the PEM fuel cell equally. Out of all components, the membrane is the most critical one. Three different types of degradation can happen on the membrane: thermal, mechanical, and chemical degradation. The second most critical components are the electrodes. According to [42], degradation processes on the electrodes can be divided into two categories: the catalyst layer and the carbon support degradation. There is also a problem with corrosion of catalyst carbon support during startups and shutdowns. It can occur because of the uneven distribution of the fuel and crossover of oxygen through the membrane.

Based on this review, with a comprehensive understanding of all these challenges, more accurate modeling can be established which will successfully detect them for further control.

5 Conclusion

The PEM fuel cell technology is significantly powerful to be compared to technologies used especially in the transport and energetic industries. Unlike similar classic technologies, PEM fuel cell technology ensures higher efficiency of conversion of chemical energy into electrical energy, followed by near-zero emissions and long life. This study focuses on the basics of the PEM fuel cell technology, as well as the modeling approaches. A review of various modeling methodologies is proposed. Generally, to create a model the application field is of primary interest, as it dictates the main focus and aim of the model. This is important to know especially because some assumptions have to be made. To do that, a clear understanding of the complex phenomena inside a PEM fuel cell is necessary to enable correct assumptions and satisfactory results. It can be observed that there are many modeling approaches that can be seen as an additional aid in the future of this study. Certainly, such a system as a PEM fuel cell system can be better understood by crossing several modeling approaches.

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