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Energy Engineering

PRODUCTION COST OPTIMIZATION OF PROTON EXCHANGE MEMBRANE (PEM) FUEL CELLS APPLIED ON HYDROGEN FUEL CELLS ELECTRIC VEHICLES (HFCEVs) BASED ON REDUCTION OF PLATINUM LOADING

Research work for the Bachelor Degree of Energy Engineering

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The Research work

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ABSTRACT

The transport sector is one of the most responsible for GHG emissions as light-duty vehicles generate one-third of global CO_2 emissions. One way of reducing the emissions in the transportation sector would be by replacing current fuels vehicles with lower carbon fuels or with zero-carbon vehicles. A sustainable alternative to the vehicles with an internal combustion engine (ICEVs) are the fuel cell electric vehicles (FCEVs) as long as the hydrogen is generated from renewable energy sources.

In the past few years, there have been significant improvements on the development of FCEVs; however, further development of this technology is needed to be as efficient and cost effective as ICEVs. In order to achieve a massive penetration of these type of vehicles into the market, key improvements on cost reduction using less expensive materials, especially in the catalyst area, are needed.

Catalyst cost is projected to be the largest contributor to overall system level costs at high volume production. Therefore, by reducing the platinum group metal (PGM) content with an alternative catalyst, the overall cost of the system could be reduced. The present research work is a review of literature of the methods and techniques currently used for the reduction of platinum loading and the use of non-platinum group metals at the catalyst layers of the Membrane Electrode Assembly (MEA) for production cost optimization of Proton Exchange Membrane (PEM) fuel cells for electric vehicles fueled by hydrogen.

RESUMEN

El sector transporte es uno de los más responsables de las emisiones de GEI, ya que los vehículos ligeros generan un tercio de las emisiones globales de CO_2. Una forma de reducir las emisiones en el sector del transporte sería reemplazando los vehículos de combustibles actuales con combustibles que emitan menos gases o con vehículos con cero emisiones. Una alternativa sostenible a los vehículos con un motor de combustión interna son los vehículos eléctricos con pila de combustible, siempre y cuando el hidrógeno utilizado como combustible sea generado a partir de fuentes de energía renovables.

En los últimos años, han habido mejoras significativas en el desarrollo de los vehículos eléctricos impulsados por hidrógeno; sin embargo, es necesario un mayor desarrollo de esta tecnología para que sea tan eficiente y rentable como los vehículos convencionales. Para lograr una penetración masiva de este tipo de vehículos en el mercado, se necesitan mejorar ciertos aspectos como la reducción de costos en los materiales y especialmente en el área del catalizador.

Se proyecta que el costo del catalizador es el mayor contribuyente a los costos generales a nivel de sistema en un alto volumen de producción. Por lo tanto, al reducir el contenido de metal del grupo del platino con un catalizador alternativo, se podría reducir el costo general del sistema. El presente trabajo de investigación es una revisión de la literatura de los métodos y técnicas actualmente utilizados para la reducción de la carga de platino y el uso de metales del grupo que no son platino en las capas de catalizador del Ensamble de Electrodo de Membrana para la optimización del costo de producción de la pila de combustible del tipo de Membrana de Intercambio de Protones (PEM) utilizada en vehículos eléctricos alimentados por hidrógeno.

KEY WORDS:

Hydrogen fuel cells; Membrane electrode assembly; Proton Exchange membrane; Platinum loading.

CHAPTER I

PROBLEM STATEMENT

All over the world, there is a growing awareness about global warming and there is an increasing concern on reducing the emissions of CO_2 and other greenhouse gases. Therefore, the human civilization is facing one of the greatest challenges of the century: Controlling Greenhouse gas emissions for minimizing the impact on the environment [1]. Based on it, in 1997, the Kyoto Protocol promoted by the United Nations Framework Convention on Climate Change, established the main goal of stabilizing the greenhouse gas concentrations present on the atmosphere avoiding dangerous anthropogenic interference with the climate system [2].

As shown in Figure 1, the energy sector, in the whole world, is responsible for more than the 60% of GHG emissions [3]. We own alternative technologies that can allow us to replace the current technology used on the energy sector. On one hand, renewable energies avoid CO_2 emissions by replacing fossil fuels on the power sector, mainly in the conventional thermal power plants. On the other hand, electric vehicles avoid the tailpipe emissions of internal combustion engines of the conventional vehicles [4].

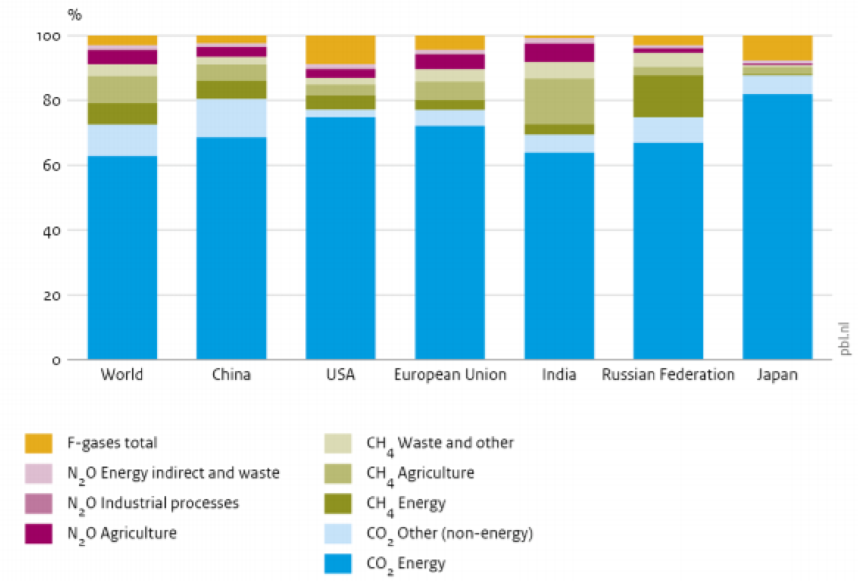

Figure 1. Contribution to 2016 GHG emissions per emission category

Source: IEA, 2017 [3].

1.1 Contribution of the transport sector to the GHGs emissions

The energy sector is very complex and plays a significant role on our daily life. Worldwide, the CO_2 emissions generated only by the transport sector, as a subsector of the energy sector, represents one of the most significant amounts of emissions. For example, in 2016 the transport sector contributed 24 % of total EU-28 greenhouse gas emissions [5]. The Figure 2 shows that fuel combustion used for the transport sector is the second most important source of GHG emissions [5].

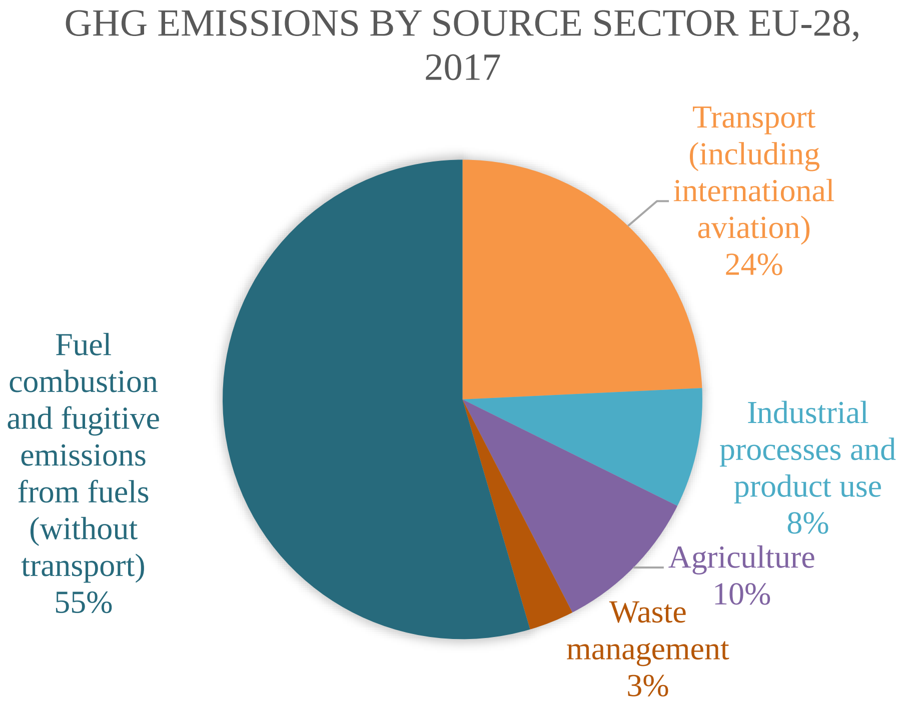

Figure 2. GHG emissions by source sector, EU-28

Source: 2017 [5].

The same happens in the U.S., transportation accounted for the largest portion (28%) of total U.S. GHG emissions in 2017; according to the *Inventory of U.S. Greenhouse Gas Emissions and Sinks 1990-2017* (The national inventory that the U.S. prepares annually under the United Nations Framework Convention on Climate Change) [6]. In Figure 3 is shown the U.S. GHG Emissions by Sector for the 2017. Furthermore, in the same year, the CO_2 emissions across sector, transport accounted for 15% of Asian emissions [7].

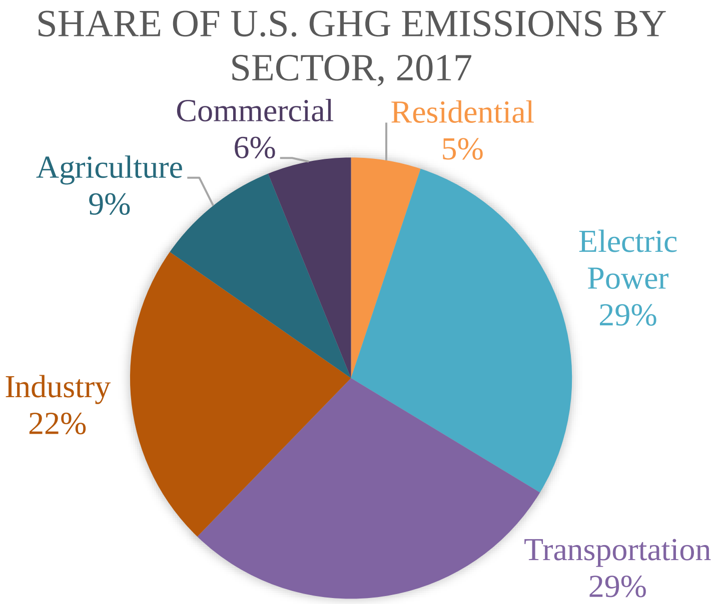

Figure 3. Share of U.S.GHG Emissions by Sector, 2017

Source: Environmental Protection Agency [6].

Therefore, the transport sector is one of the greatest responsible of GHG emissions all over the world, and only light-duty vehicles, in the U.S. for example, generate one-third of total CO_2 emissions from the country [8]. As it is effectively impossible to capture CO_2 emissions from individual vehicles, the only way of reducing the emissions in the transportation sector is by replacing current fuels with lower carbon fuels or even better, with zero-carbon vehicles [8]. A sustainable alternative to the vehicles with an internal combustion engine, are the battery and fuel cell electric vehicles (Hereafter BEVs/FCEVs), as long as the electricity and hydrogen are generated from renewable energy sources [4]. EVs are already been developed and play an important role in the energy transition. Their massive implementation will allow us to achieve the reduction of global CO_2 emissions.

1.2 Batteries versus Hydrogen Fuel Cells Electric Vehicles

Nowadays, a potential EV driver can choose between a battery-powered electric vehicle and a hydrogen powered fuel cell electric vehicle. Each of these technologies has its own advantages and disadvantages regarding cost, range, performance, and infrastructure, as shown in Table 1, those are some of the differences between BEVs and FCEVs.

	BEV	**FCEV**
Recharging/Refueling infrastructure	80% charging could be at home, chargers are easier to install	Too complex the implementation of a hydrogen recharging network
Recharging/Refueling speed	Low	Fast as ICV
Range	Limited by power density of batteries	As good as ICVs
Energy demand	18-25 kWh of electricity per 100 km	60 kWh Electricity-H_2-electricity
Flexibility to follow intermittent Renewable Energy Systems (RES)	Has to be connected to the power grid in times of high RES generation	H_2 can be generated any time and be easily storage
Round-trip efficiency	>70%	40-60%

Table 1. Differences between BEV and FCEV

Source: Own elaboration based on the following references [4][9][10].

Currently there is no clear answer as to which one could dominate the future low carbon vehicle market. However, there are various technical, economic and infrastructural barriers that limit the large scale adoption of both the FCEVs and BEVs [11]. Both types of vehicles share many of the same components because they operate with the same electric system inside the EV; they both have an electric motor and power controller or inverters; however, their main energy source, their input, is very different. HFCVs use a fuel cell to convert the hydrogen energy into electricity and BEVs use batteries to storage the energy [12].

In the future, both of these technologies will probably coexist, while the BEVs are more suitable for short range and small vehicles, the HFCEVs are mostly to be applied in medium and long range vehicles [13]. By 2050, BEVs and FCEVs could become less expensive than the advanced ICEVs. Putting aside the complexity of developing a hydrogen infrastructure, fuel cell vehicles are the most promising alternative to ICEVs mainly because they are not subject to the limitations of battery vehicles [8].

1.2.1 Need of improvements on FCEVs

In the past few years there have been significant improvements in the development of FCEVs, however further development of this technology has to be done because still aren't as efficient and cost effective as ICEVs are. According to The National Research Council, one of the main technology challenges for the HFCV are making the fuel cell system as durable and cost-effective as today's gasoline internal combustion engine vehicle [8]. In addition, the Committee on Transitions to Alternative Vehicles and Fuels states that the research done on advanced materials and battery concepts will be critical for the success of electric drive vehicles. It also recommends that the following research area will have the greatest impact: New catalyst structures that increase and maintain the effective surface area of chemically active materials and reduce the use of precious metals like platinum: fuel cell stack and batteries [14].

In order to achieve a massive penetration of these type of vehicles into the market, they need to be as cost effective as today's ICEVs so that they can be more competitive and widely commercialized. Based on the research that has been done so far, still more research has to be done in order to achieve a cost reduction in materials, especially in the catalyst area. The last years, the researchers have been focused on reduction of platinum group metal (PGM) content, platinum alloys, novel support structures, and non-PGM catalysts. Given that the catalyst cost is projected to be the largest contributor to overall system level costs at high volume production, if we achieve substituting the platinum with an alternative material that still works as a catalyst, then the overall cost of the system could be reduced [15].

1.3 Justification and Motivation

Based on the previously stated information, this thesis will be developed with the aim of contributing to the exploration on the literature of alternative materials for the fuel cell stack, methods and techniques with the objective of making this technology more cost effective. Moreover, the motivation of this research is the achievement of scaling-up the use of FCEVs in the market and subsequently the accomplishment of the reduction of CO_2 emissions worldwide. In the following paragraphs, there are three more detailed reasons that justify the motivation of this work.

First, the production cost a hydrogen fuel cell vehicle is still too high for being a competitive technology as internal combustion engines vehicles. On 2017, a team from the Department of Energy, Argonne National Laboratory, made an analysis on the cost of direct hydrogen fuel cell vehicles. They found that the estimated system cost to date for 100,000 and 500,000 units per year, result in a total system cost of 50$/kWnet and 45$/kWnet, respectively. For 2025, there is a target of a total system cost of 40$/kWnet and the ultimate goal is to reduce the overall HFCEVs system to 30$/kWnet in order to be on a essentially cost parity with ICEVs [16].

Second, there has been less research and technology improvements for FCEVs than for BEVS. Over the past decade, there had been very significant cost reductions and performance improvements of Li-ion battery packs for EVs, making the BEVs a more competitive technology available in the market [17]. As shown in Figure 4, between 2010 and 2016 the battery costs had been reduced on a 77%, being now near to reach the $125–$150 target that makes EVs competitive with conventional gasoline vehicles [18]. On the other hand, as shown in Figure 5, between the same period of time (2010-2016), the costs of fuel cell systems had been reduced only on a 17%, being now still far from the target of 30$/kW for being as competitive as ICEVs [19].

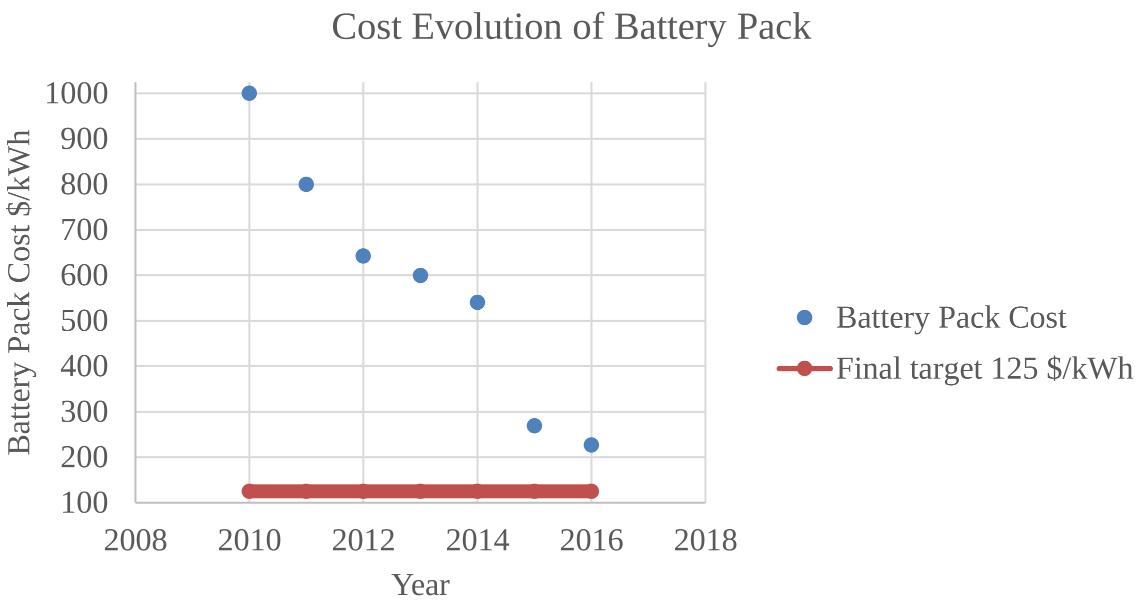

Figure 4. Cost Evolution of Battery Pack for EVs (2010-2016)

Source: [18][20].

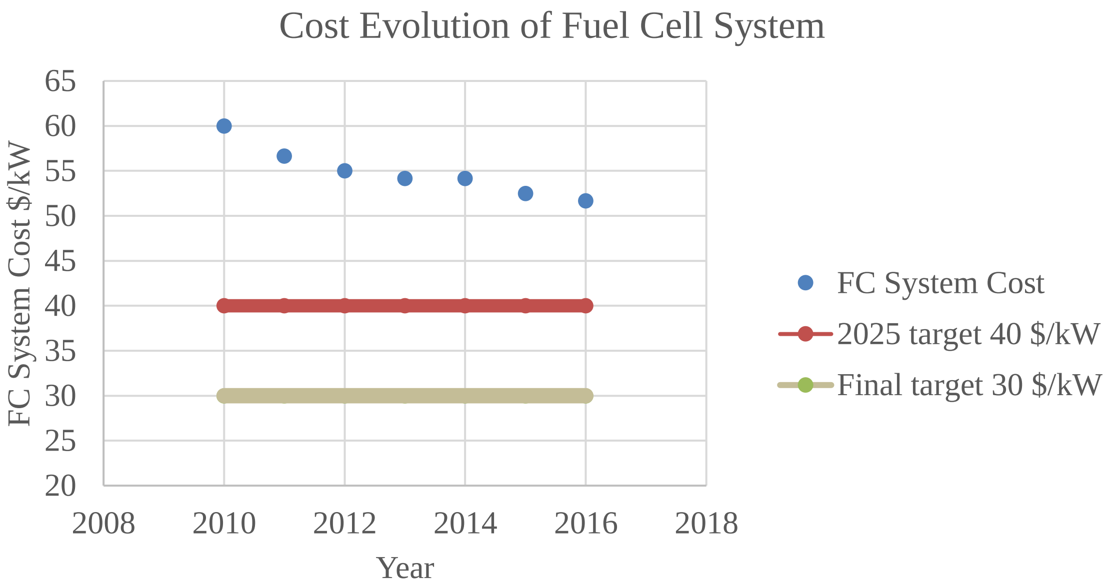

Figure 5. Cost Evolution of Fuel Cell System for EVs (2010-2016)

Source: National Renewable Energy Laboratory [19].

Third, as shown in Figure *6*, the greatest cost of the fuel cell stack is represented by the MEAs, and inside of it is the catalyst component that represents 41% of the overall cost. Finally, based on the three reasons stated above, is justified the research topic of this thesis project. The development of this research work will contribute in some way to the target of reducing the system cost to $40/kWnet, by evaluating alternative materials for the most expensive component of the fuel cell stack, the MEA, mainly by substituting the catalyst. In addition, it will contribute to the elaboration of more scientific research for fuel cells, a topic in which still has not been developed an exhausted research all over the world.

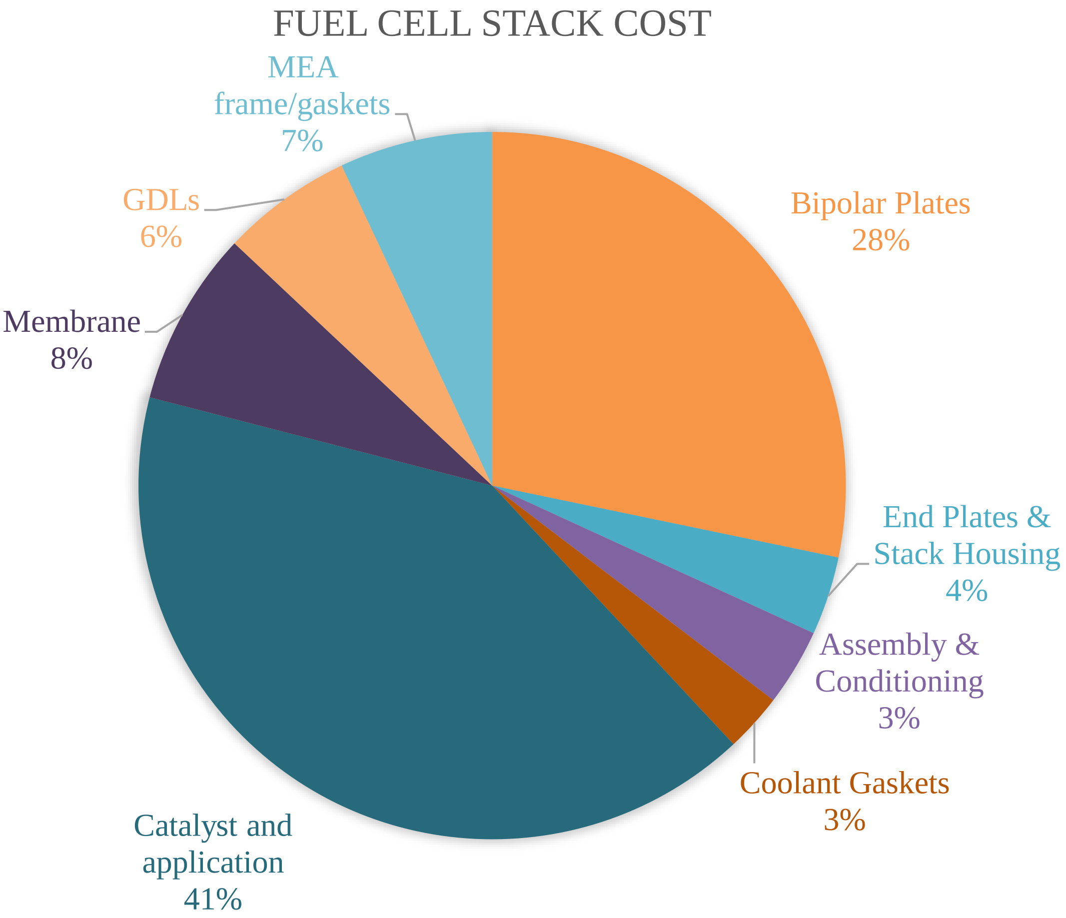

Figure 6. Component cost breakdown at production volume of 5M units/year for the FC stack

Source: [16].

1.4 General Objective

Evaluate the reduction of platinum loading and the use of non-platinum group metals at the catalyst layers of the Membrane Electrode Assembly (Hereafter MEA) for production cost optimization of Proton Exchange Membrane (Hereafter PEM) fuel cells for electric vehicles fuelled by hydrogen.

1.5 Specific Objectives

- Investigate and identify the properties of PGM-free catalysts with the potential of being used at MEA, as well as the possible reductions of platinum loading on the electrodes.
- Perform a review of literature about the applications of reduction of platinum loading, as well as the use of PGM-free on electrodes of the PEM fuel cells.
- Investigate about experimental validations of the performance of the fuel cell with alternative materials to platinum catalyst and alterations on conventional catalysts.

CHAPTER II

THEORETICAL FRAMEWORK

In the late 1900s, the concept of the fuel cell emerged as a new electrochemical power device. Since the many scientists and engineers believe that fuel cells hold promise as an alternative energy source to help offset our traditional reliance on coal, oil, and natural gas [21]. However, it was in 1839, the first discovery of the fuel cell principle by William Grove, a lawyer and physicist from Great Britain. [22]. Nevertheless, it was until 1960s, in the Apollo space program, that the fuel cells where used to power the onboard electrical systems of the Apollo spacecraft. They were used because no batteries could last long enough for a flight to the moon; the system provided both, electricity and drinking water for the astronauts. This energy device supplied 1.5kW of continuous electrical power, over ten thousand hours of operation, without a single in-flight incident [23].

Besides that, the performance of fuel cells that NASA deployed was exemplary, the cost per kilowatt was astronomical because the FC were hand-built and used exotic materials. Therefore, other types of technologies emerged with a higher commercial potential, and research of FC continued at a low funding level [22]. It was recently at the beginning of 1980s, when governments agencies in the US, Canada and Japan, significantly increased their efforts on the development of fuel cell technology [24].

2.1 Fuel Cell Technology

According to the U.S. Department of Energy, a fuel cell is an electrochemical device that converts the chemical energy of hydrogen or another fuel to electrical energy in a cleanly and efficiently way [16]. Unlike batteries, the energy supply in a fuel cell is external and can be supplied essentially indefinitely by refueling the external tank, the same as in an internal combustion engine. Generally, fuel cells use gaseous or liquid fuels, such us natural gas, methanol, etc., but hydrogen fuel cells uses hydrogen as fuel and the oxidant for a fuel cell either is usually oxygen in air [25]. In comparison to heat engines, the energy conversion in fuel cells is direct and simple and it is not limited by thermodynamic limitations like Carnot efficiency [26].

Fuel cells are a promising technology for the transportation, power generation and even as an energy storage device because of their high energy density and efficiency and low environmental impact [27]. They are very versatile because can be used as very small devices producing only a few watts of electricity, or as large power plants producing megawatts. Fuel cells are suitable for different applications because there are different types. These are classified according to the nature of the electrolyte they use, each type require particular materials and uses different fuels and reactants [28]. As shown in Table 2, fuel cells have advantages and disadvantages when compared to internal combustion engines and batteries. Those disadvantages are the reason why still this technology cannot massively replace other technologies with lower efficiency and higher impact on the environment, however these limitations eventually will be solved by the development of research and engineering solutions [29].

Advantages	**Disadvantages**
Higher efficiency: direct production of electric energy (40-50%)	Lack of hydrogen infrastructure, is difficult to produce and store hydrogen
Low chemical, acoustic and thermal emissions (have no big moving parts and GHG emissions are low if hydrogen is used as fuel)	Emerging technology that still needs to accomplish reductions in cost, weigh, size, and increase in reliability and durability.
Fuel flexibility, can be used hydrogen, natural gas, propane, and even anaerobic digester gas	Require relatively a pure fuel, without contaminants that can deactivate the fuel cell catalysts. Therefore, is some cases is needed a reformer on the fuel cell system.
Compactness, higher energy density and energy storage capacity. Fuel cell system is lighter than batteries.	Fuel cell system is still a little heavier than internal combustion engine systems
Exhibit good load-following characteristics because are solid-state devices that react chemically and instantly to changes into load.	
Don’t need recharging rather, must be refueled, which is faster	
Modularity, allow independent scaling between power and capacity. The fuel cell size can be adapted by simply changing the number of fuel cells stacks.	

Table 2. Advantages and disadvantages of fuel cell technology

Source: Own elaboration based on the following references [24][30][29].

2.1.1 Basic operation principles

A fuel cell consists mainly of an anode, an electrolyte, and a cathode. As shown in Figure 7, on the anode side, the fuel is oxidized electrochemically and releases positively charged ions; on the cathode side, oxygen molecules are reduced to oxide or hydroxide ions [31]. The electrolyte is the medium through which either the positively charged ions travel from anode to cathode or the negatively charged ions travel from cathode to anode. The only byproduct that goes out of the system is water vapor. A catalyst is often used to speed up the electrochemical reactions at occur on the electrodes [28].

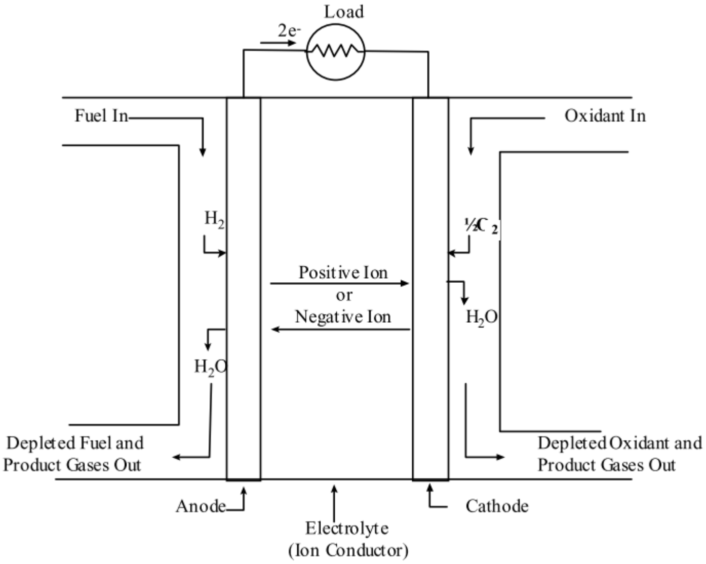

Figure 7. Basic schematic fuel cell operation

Source: [32].

The reactants of fuel cells can be classified based on the properties of the components that donate the electron (oxidant) or accept the electron (reductant). Oxidants mainly include pure oxygen and the reductants, which are also known as the fuels, include pure hydrogen or gases that contain hydrogen like ethanol, methanol, natural gas, etc [33]. The most basic reaction that occur inside the fuel cell consists of the combustion of hydrogen fuel into water, and this process is split into two electrochemical reactions: the oxidation reaction (1) that occurs on the anode and the reduction reaction (2) that occurs on the cathode. These are the two half reactions and the overall combustion reaction is given by the combination of the two half reactions (3) [34].

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

$$½O_2 + 2H^+ + 2e^- \rightarrow H_2O \tag{2}$$

$$½O_2 + H_2 \rightarrow H_2O \tag{3}$$

The role of the electrolyte is to contain the two half reactions electrically while allowing the movement of ions: the protons produced at the anode are moved to the cathode side where they will combine with the oxygen and form a molecule of water [26]. Therefore, electrolytes should be both good proton conductors and excellent electric insulators. There is a third requirement for electrolytes, they need to be impermeable to gases so that the anodic and the cathodic compartments remain separated, in order to prevent gas crossover [28]. Through the electrolyte, the flow of electrons is hindered; therefore, it is forced to go through another way. There should be an external electrical circuit through which the electrodes flow and that allows fuel cells to provide DC power by the direct collection of electricity [35].

In the case of the electrodes, a high surface area is an important feature in order to maximize each half reaction zone, for this reason they are relatively porous compounds [36]. According to the application of fuel cell technologies, the materials of the whole systems depends on each type of fuel cell and they are describe on the section below. Every type of fuel cell is characterized by its own particular dimensions, reactants and materials; however the basic structure of fuel cells consists of an electrolyte, two electrodes, and requires a fuel and a oxidant for producing electricity [37].

2.1.2 Types of Fuel Cells

Nowadays, there are different types of fuel cells that are currently under development and are classified primarily based on the electrolyte they use. The electrolyte is an essential part of the fuel cells, and it determines the operating parameters, such as the type of catalysts, the electrochemical reactions, the operating temperature, the reactants that can be used and therefore the applications for which these cells are most suitable [38]. The most promising types of fuel cells, with their characteristics, are shown in Table 3. The schematic fuel cell operation of each type can be found on the Annex *1*, Annex *2*, Annex *3*, Annex *4*, Annex *5*, Annex *6*.

Fuel cell type **Operation temperature** **Efficiency**	**Common Electrolyte** **Charge carrier** **Fuel** **Electrode catalyst**	**Applications**
Polymer electrolyte membrane (PEMFCs) 50-100 °C 60%	Perfluorosulfonic acid H+ Hydrogen Platinum based	Distributed generation Portable power Transportation Ideal for vehicles
Direct methanol (DMFCs) 60-130 °C 60%	Perfluorosulfonic acid H+ Methanol Pt-ruthenium	Portable power Early market applications
Alkaline electrolyte (AFCs) 50-100°C 60%	Alkaline polymer, aqueous KOH OH^- Pure hydrogen Nickel	Portable power Backup power Space shuttles
Phosphoric acid (PAFCs) 150-250 °C 40%	H_3PO_4 H^+ Hydrogen Pt catalyst dispersed on carbon	Distributed power
Molten carbonate (MCFCs) 500-700 °C 45-50%	$(Li, K, Na)_2CO_3$ CO_3^{2-} Hydrocarbon fuels (methane) Non-precious metal	Distributed power Electric utility
Solid oxide (SOFCs) 600-1000 °C 60%	Stabilized Zirconia Oxides O^{2-} Hydrocarbon fuels (methane) Non-precious metal	Distributed power Electric utility APUs

Table 3. Types and characteristics of fuel cells

Source: Own elaboration based on the following references [25][26][27][28][29][39][40].

2.1.3 Application of fuel cells on transportation

One of the most common application of fuel cell technology is on the transport sector. In addition to the environmental advantages offered by fuel cells, they have several properties that make their use suitable for many transport applications, including for scooters, passenger cars, busses, and even space shuttles [37]. FCEVs are considered low emission vehicles and the HFCEVs zero emission, these vehicles have greater efficiency than ICEVs and BEVs [41]. This type of vehicle propulsion system has short startup times, high dynamic load demand and requires to operate at low temperatures, therefore, PEMFCs are the technology most widely used for this application [30].

The transportation application of FCs is mainly concentrated on automobiles, buses and niche transport applications [42]. The development and application of PEMFC in the transport sector is competitive and promising due to the desire of depleting fossil fuels and the need of zero emission vehicles. Currently most of the governments (like USA and Japan) and the companies car makers are actively engaged on the development of this technology and are working on the onboard integration of fuel cell systems and electric energy storage devices with an energy management system [29].

The main components and the basic operation of a hydrogen fuel cell vehicle are shown on Figure *8* where the letter A represents the electric motor, the letter B the fuel cell stack, C is the battery and D is the high-pressure hydrogen tank. On the first step, the oxygen present in the air gets into the system. Then on step 2, the oxygen and hydrogen supplied to fuel cell stack. The third step is the generation of electricity and water, through the electrochemical reaction produced inside the fuel cell stack. On step 4, the electricity generated feed the electric motor and then, step 5, the motor is activated and powers the vehicle system, enabling it to move. The last step, step 6, is the water expulsion of the outside the vehicle as tailpipe emission [43].

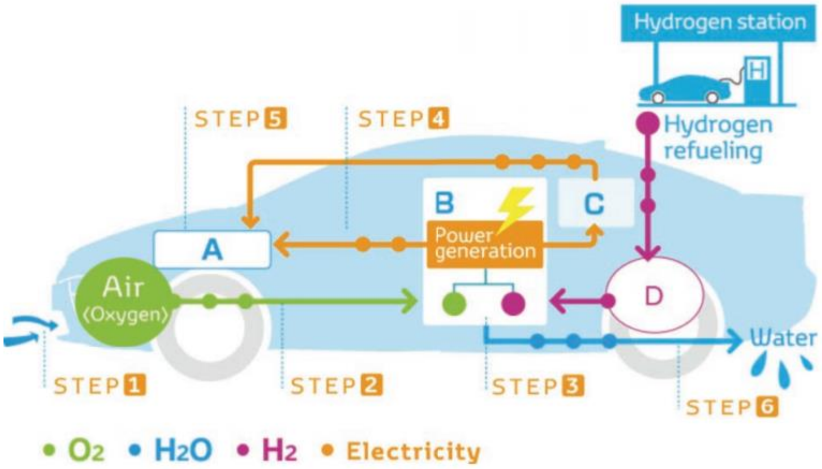

Figure 8. HFCEV operation diagram

Source: [43].

2.1.4 Evolution of hydrogen fuel cell electric vehicles technology

Today, passenger cars powered by fuel cells have demonstrated about 3000 hours of operations (depending on speed: 150-300 thousands kms), but still durability issues, like start-stop operation and steep transient load cycling, have to be solved. However, the major problem are the high costs despite the significant cost reduction achieved over the last years [44].

The automobile manufacturers that are currently leading the development of HFCEVs, in the United States, are Daimler and Ford; there is also the Automotive Fuel Cell Cooperation (AFCC) with Ballard [45]. On 2013, the companies Daimler AG, Ford Motor Company and Nissan Motor Co. signed an agreement where the three parties agreed for a joint development of common fuel cell system to speed up the development of the technology, furthermore increase the availability of zero-emission technology and significantly reduce costs [46]. In Japan, the largest auto manufacturers are Toyota, Honda and Hyundai [34].

Despite all the efforts of the governments and companies putted on the development of FCEVs, today still these type of vehicles are not massively available on the market, many of them are still prototypes units [30]. Nevertheless, in the last 5 years, fuel cell cars started to come out of demonstration stage and are becoming commercials, by today, companies like Toyota, Hyundai and Honda have started initial commercial sales of their fuel cell cars. In 2013 the first fuel cell vehicle became commercially available and since then until 2017, 6,364 hydrogen fuel cell vehicles were sold globally [47].

The most advance fuel cell vehicle with the world's best fuel cell efficiency and with a driving range of 666 km is the NEXO Fuel Cell SUV. This vehicle can reach a maximum speed of 179 km/h, a maximum output motor of 120 kW, the total output of the fuel cell systems is 135 kW, the stack is conformed of 440 cells (250-450V) [48]. The evolution of the FCEVs of Hyundai is shown on Fig. 9.

1st Generation 2000 - 2004	2nd Generation 2005 - 2008	3rd Generation 2009 - 2011	4th Generation 2012 - 2016	5th Generation 2017
Core Technology Development	System Technology Development & Fleet Operation	Semi-Automatic Production	Pre-Commercial Production (pilot-scale)	Commercial Production
	Stack Bipolar Plate – Graphite	Stack Bipolar Plate – Metal		Full Automated Production line
Several FCEVs	Tens of FCEVs/year	> 100 FCEVs	1000 FCEVs	Several thousand FCEVs

Figure 9. Evolution of FCEVs manufactured by Hyundai

Source: [49].

Because of the high costs and extensive knowledge required for the development of this technology, many companies are joining their forces in order to achieve the massive commercialization of cost-efficient HFCVs [48]. *"The partnership between Hyundai Motor Group and Audi will leverage collective R&D capabilities in fuel cell technology to elevate their presence in the FCEV market. Therefore, the agreement also includes mutual access to fuel cell components. As a first step, Hyundai Motor Group will grant its counterpart the access to parts that are based on Hyundai's know-how accumulated from the development of ix35 Fuel Cell as well as NEXO. Audi – responsible for the development of fuel cell* *technology within the Volkswagen Group – will also be able to take full advantage of Hyundai's FCEV parts supply chain. Hyundai Motor Company, the world's first mass-producer of fuel cell vehicles, has been offering SUV-Class FCEVs since 2013, and currently sells them in 18 countries around the world*" [50].

Despite the achievements that have been accomplished so far, there are still two major challenges to HFCEVs commercialization, reducing cost and improving durability [51]. Ongoing research is mainly focused on the development of new materials for the membranes, catalysts, bipolar plates and membrane electrode assemblies in order to reduce the cost and extend the life of fuel cell stack components [40].

2.2 Proton Exchange Membrane Fuel Cells

The proton exchange membrane fuel cell use an electrolyte that conducts the protons from the anode to the cathode, these electrolyte is composed of a solid polymer film of acidified Teflon [30]. This is the most suitable type of fuel cell for transportation applications. The characteristics that make this type of fuel cell the most widely used on vehicles are the low operating pressures (15-30 psig) which increases the safety of the system and low temperatures (<100°C) that allows relatively short start-up times and the "load following" that means that it responds almost instantaneously to changing power demands [22].

A PEM hydrogen fuel cell generates approximately 0.7 V DC under no-load conditions. In order to obtain higher voltages, multiple PEM fuel cells can be connected in series. A series-connected set of fuel cells technically forms a battery, but engineers call it a stack [52]. The PEM cell stacks have already been made compact and powerful enough, and they provide power and acceleration equal to, or even better than, internal combustion engines; for this reason, since 1994 FCEV are available in the market been offered by companies like Daimler, Toyota, Nissan, Honda, Hyundai, General Motors, Ford, etc [53].

This technology has drawn the attention because of its simplicity, viability, suitability, and quick start-up and because it has been even proposed as a promising power source for zero emission vehicles [36]. PEMFC systems are suitable for different vehicle applications since they do not require the use of hazardous fluids, and they enjoy high power densities while maintaining low operating temperatures [29]. It is shown in Table 4 the advantages and disadvantages of this specific type of fuel cell.

Advantages	Disadvantages
• Tolerant to carbon dioxide • Operate at low temperatures and pressures • Use solid, dry and non-corrosive electrolyte • High voltage, current and power density • Use stable materials • Relatively simple mechanical design • Compact and rugged	• Only tolerate 50 ppm CO • Tolerate few ppm of total sulfur • Use an expensive platinum catalyst • Is difficult to work with the membrane, water management problems

Table 4. Advantages and disadvantages of PEMFCs

Source: Own elaboration based on the following references [30][29][26].

2.2.1 Working principles

Inside a PEM fuel cell, two electrochemical reactions take place and make up the total redox reaction, at the anode an oxidation reaction, loss of electrons, (1) and at the cathode a reduction reaction, gain of electrons, (2) [23]. On the reduction reaction the H^+ is drawn through the electrolyte from the anode to the cathode the reactive attraction of hydrogen to oxygen, while the electrons are conducted through an external DC circuit [28]. The overall cell electrochemical reaction (3) is obtained combining the anode and cathode reactions. The product of the whole reaction is water; this must be continually removed of the system to facilitate the continuity of further reactions. All of this process is better shown on Fig. 10. Schematic operation of PEMFCs [40].

$H_2 \rightarrow 2H^+ + 2e^-$ (1)

$\frac{1}{2}O_2 + 2H^+ + 2e^- \rightarrow H_2O$ (2)

$\frac{1}{2}O_2 + H_2 \rightarrow H_2O$ (3)

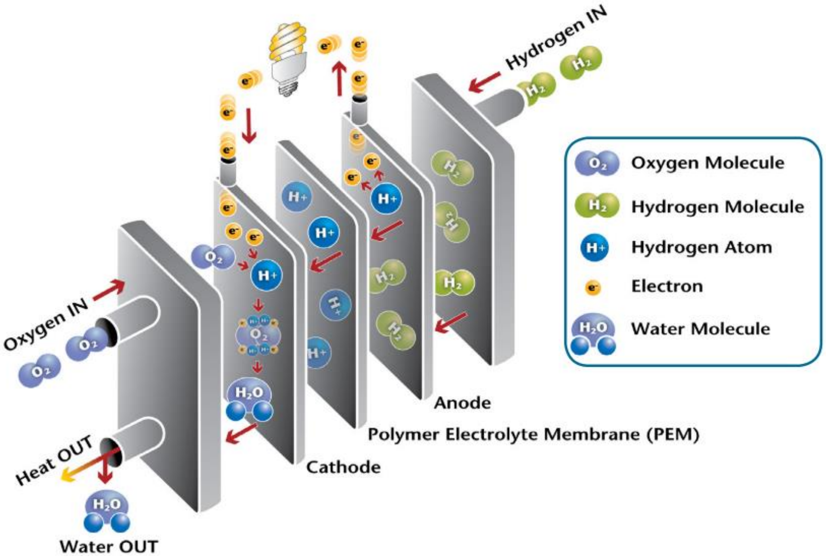

Figure 10. Schematic of a Polymer Electrolyte Membrane fuel cell

Source: [40].

2.2.2 Structure and Components

The main component of a PEM fuel cell is the membrane electrode assembly (Hereafter MEA), which is composed by the membrane, the catalyst layers, and gas diffusion layers (Hereafter GDLs). As shown in Figure 11, the MEA components are the GDLs, the catalyst layers (Hereafter CLs), and the proton exchange membrane. On one side is an electron-conducting anode consisting of porous GDLs as an electrode and an anodic catalyst layer; on the other side is an electron-conducting cathode consisting of a cathodic catalyst layer and again, a porous GDL as an electrode and in the middle there is a proton-conducting electrolyte, a hydrated solid membrane [55].

Besides the MEA, there are the hardware components that incorporate the MEA into a fuel cell and enable an effective operation. These includes gaskets that provide a seal, preventing leakage of gases, they are usually made of rubbery polymer like Teflon (PTFE) [24]. There are also bipolar plates that assemble individual PEMFCs into a whole fuel cell stack and incorporate channels for the fluids, they are usually made of metal, carbon or composite and provide electrical conduction and physical strength to the stack [54]. Finally, outside the cell there are current collectors with the reactant gas flow fields and take the electrons to an external circuit, providing a DC current [38].

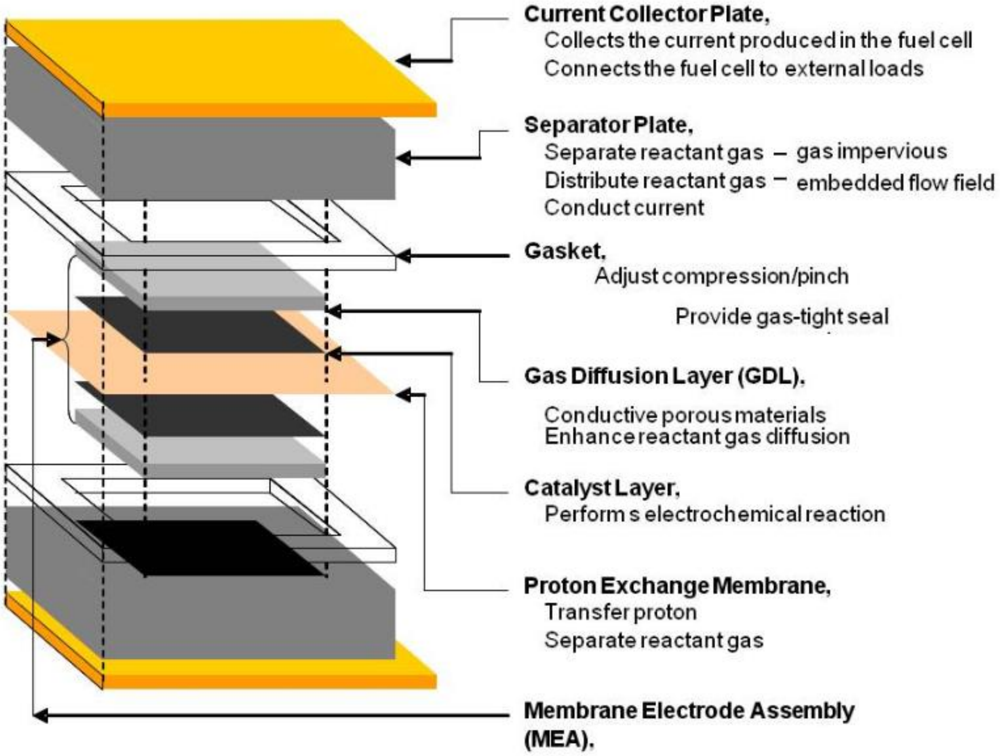

Figure 11. Basic structure and components of the PEMFC

Source: [55].

The most common membrane used at PEMFC is Nafion (DuPont), a sulphonated polymer, its layer has a thickness between 35 and 170 μm, while thinner is the membrane the cell will have a higher conductivity because there will be less resistance, nonetheless this implies water management problems [29]. The main function of the GDLs is to act as a gas diffuser, which means to provide mechanical support as an electrical pathway for electrons and as a channel for the evacuation of the byproduct, water. Typically are constructed from carbon paper with thickness between 100 and 300 μm [56].

2.2.3 Catalyst Layers

The catalyst layers are where the electrochemical reactions take place; they transport the electrons from the GDLs to reaction site, as well as the protons from the membrane to the reaction site. Usually these layers are made of platinum and the typical thickness of the CLs is between 1 to 20 μm [57]. The nanoparticles of platinum are dispersed on a high surface area supported by carbon for later being mixed with an ion-conducting polymer, usually Nafion [54].

2.2.3.1 Materials

The catalyst layer, is a porous electrode containing carbon-supported platinum mixed with a polymer electrolyte, given that he electrochemical reaction occurs here, the material and structure of the CLs have a major influence on the overall performance of the fuel cell [58]. In order to facilitate the electrochemical reactions, the CLs should meet the following three properties: electronic conductivity, protonic conductivity and gas diffusivity as shown on Figure 12.

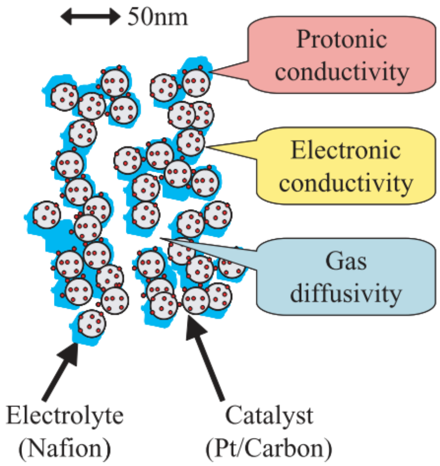

Figure 12. Schematic illustration of a catalyst layer

Source: [58].

On one hand, platinum is a rare metal and one of the most expensive metals on Earth. It is known for its allure in engagement rings, and because it works as an excellent catalyst, however the availability of this metal on Earth is limited. Without a cheaper substitute for platinum, these clean energy technology (PEMFC used in hydrogen vehicles) won’t be able to compete against means of transportation that burn fossil fuels [59]. On the other hand, the activated carbon is a common material used for supporting the Pt catalyst layers, thanks to its stability in both acid and basic media; in addition the fact that the carbon can be burnt off, allowing an economical and ecological effective recovery of the precious metal platinum [60]. Nowadays, there is a growing awareness of the need to study the surface chemistry of carbon-supported material to achieve an improvement on the catalyst performance. On way of modifying the catalytic behavior of activated carbon-supported noble metal catalysts can be done by increasing the active area through oxidation treatments of the support prior to metal loading [61].

2.2.3.2 Cost Analysis

A key factor that will determine the successful commercialization of the fuel cell technology is its cost competitiveness. Now, the predominant cost driver of a HFC vehicle is the amount of precious metal (Platinum) used on the PEMFC, that is the reason why researchers are striving to reduce the amount of Pt required for the cell by designing better materials [62]. As shown in Figure 13, the highest cost of a HFC vehicle is on the fuel cell stack with a 41% of the overall system cost, and if we go back to Chapter I, to Figure 7, at the breakdown costs of fuel cell stacks, the catalyst is the most expensive component of the system.

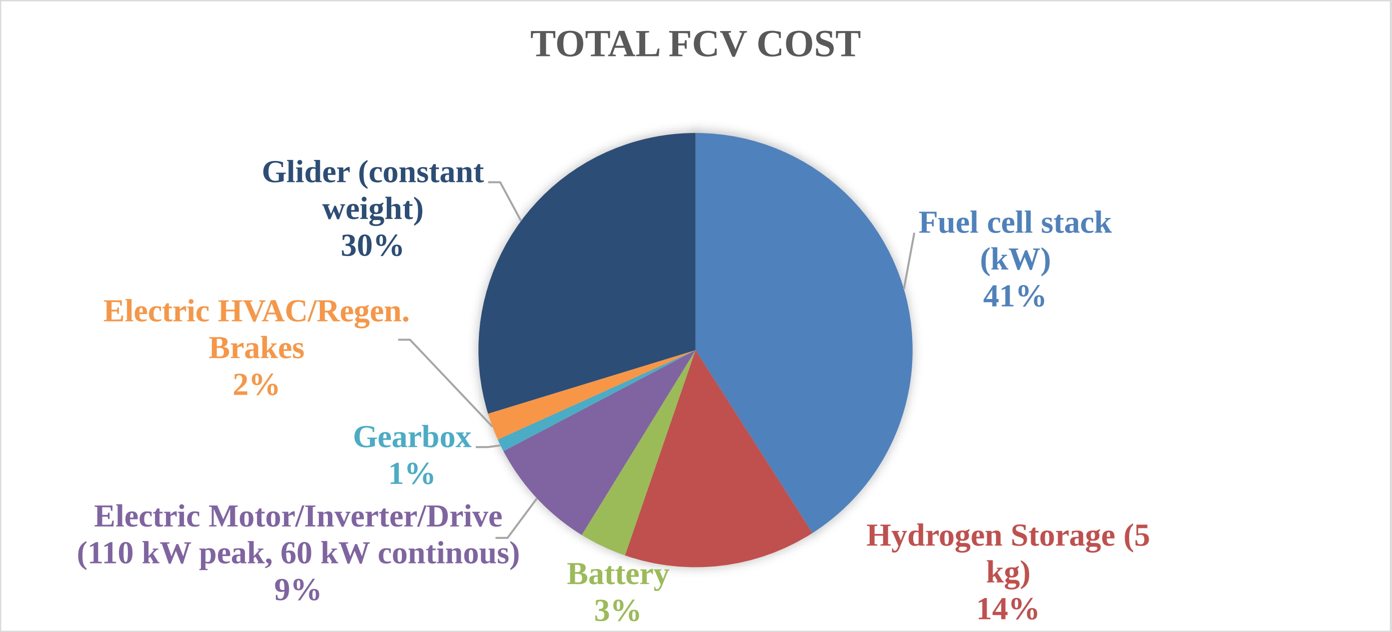

Figure 13. Estimates of breakdown FCV costs, 2016

Source: Own elaboration based on the following reference [45].

As already mentioned, HFCEVs cannot be commercialized yet on a large scale because there is a significant technical-cost barrier, the high amount of Pt required on the CLs. Therefore, many research centers, universities, and government agencies are working on either reducing the platinum loading or developing non-platinum group metal catalysts (Hereafter PGM-free) [41].

2.2.3.3 Current Cost Optimization Methods

One of the problems on the objective of using less platinum as a catalyst is that these expensive catalyst is the most efficient alternative so it is important to make sure that the cheaper alternatives are not only less expensive but also at least as efficient [63]. Researchers at Stanford University have found a technique to increase the energy efficiency and performance of the platinum catalyst meaning it may be cost-effective in the end. Their developed method consists of a thin material (lithium cobalt oxide) that can strain a platinum lattice, both compressed and stretched out, and the catalytic activity is then doubled [64].

The design of electrodes of polymer fibers is a method that support the catalyst because the surface area where reaction can occur is greater, which means that less Pt is needed. Scientists at the Vanderbilt University have found this fiber electrode design that also significantly boosts the fuel cell performance [65]. Another method of cost optimization is the development of non-precious metal catalyst, for example, researchers at the Washington State University have found a catalyst made of a nanomaterial called aerogel, consisting of about 92% air. That catalyst owns a high porosity that increases the surface are, therefore allows to reduce the need of Pt [66].

There are a lot of methods for cost optimization of PEMFC that are being discovered, research on-going in this field and improvements on electrodes and fuel cell catalysts are being done all the time. The improvements that have been described above are in an early stage of development; therefore, there is more work that needs to be done [67].

2.2.4 Performance and durability targets

The two primary metrics for analyzing the activity of a PGM catalyst are: specific activity (A/cm2), and mass activity (A/mg_{PGM}) [68]. Due to the lingering kinetics of the oxygen reduction reaction, which is more less 5 orders of magnitude slower than hydrogen oxidation kinetics, the greater portion of the PGMs are required at the cathode side [69]. Researchers on PEMFC are aware of this challenge and focus their research on improving the catalysts used for the hydrogen oxidation at the cathode. As shown in Table 5 the cathode has a higher Pt loading than the anode. Nonetheless, the reduction of Pt loading is expected to be reduced on both sides, anode and cathode [70]. As shown in Table 6, the U.S. DOE had established performance and durability targets for the 2020 and in the long term, for the PGM-free and PGM cathodes used at the MEAs.

Case	**Anode Areal Loading mg_{PGM}/cm^2**	**Cathode Areal Loading mg_{PGM}/cm^2**	**Anode PGM Mass (g)**	**Cathode PGM Mass (g)**	**Normalized PGM Content g/kW (rated, gross)**
State-of-art	0.05	0.2	4.5	18	0.25
2020 DOE Target	0.025	0.1	2.3	9	0.125
Stretch Target	0.0125	0.05	1.1	4.5	0.0625

Table 5. Relationship between PGM real loading and absolute/Normalized Mass (90kW, $1W/cm^2$).

Source: U.S. DOE [70].

Characteristic	**Units**	**2015 Status**	**2020 Targets**
Platinum group metal total content (both electrodes)	g/kW (rated, gross) at 150 kPa (abs)	0.16	0.125
Platinum group metal (PGM) total loading (both electrodes)	mg_{PGM}/cm^2 (electrode area)	0.13	0.125
Mass activity	A/mg_{PGM} at 0.9 $V_{ir\text{-}free}$	>0.5	0.44
Loss in initial catalytic activity	% mass activity loss	66	<40
Loss in performance at 0.8 A/cm^2	mV	13	<30
Electro catalyst support stability	% mass activity loss	41	<40
Loss in performance at 1.5 A/cm^2	mV	65	<30
PGM-free catalyst activity	A/cm^2 at 0.9 $V_{ir\text{-}free}$	0.016	>0.044

Table 6. Technical targets: Electro catalysts for transportation applications.

Source: Own elaboration based on the following references [68][71][72][73][74].

2.3 Methods and techniques for reduction of platinum loading at the catalyst layers

During the past decade, researchers had being developing different catalyst with the objective of reducing the PGM present in the MEA and therefore reducing the production costs of the PEMFC technology, they had been focused on the development promising ORR catalysts, because of its higher use of Pt [67]. These catalysts can be categorized as (1) Pt/C, (2) Pt and Pt alloy/de-alloy, (3) core-shell, (4) nonprecious metal catalysts (PGM-free), (5) shape-controlled Nano crystals, and (6) Nano frames, as shown in Figure 14, currently the catalyst used is the platinum catalyst supported by carbon particles (1), and the others (2, 3, 4, 5, 6) are being under development.

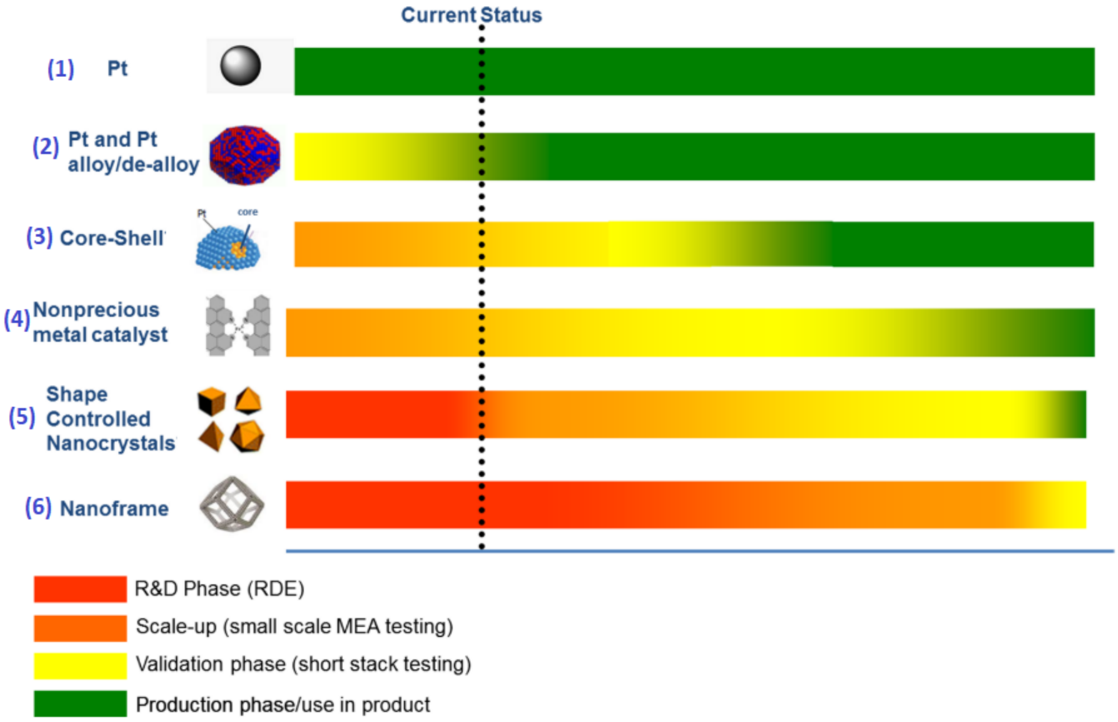

Figure 14. Development timelines for Pt, Pt alloy/de-alloy, core−shell, nonprecious metal, shape-controlled, and Nano frame ORR electro catalysts

Source: [75].

2.3.1 Direct reduction of Pt loading

The Pt/C is the simplest and most commonly type used as catalyst of PEMFC because when limiting the design of the catalyst to a single element, in this case platinum, there are no many options for improving the activity and durability of the cell. In fact, further improvements in activity and durability with conventional platinum supported by carbon catalyst rely on advances in “catalyst−support” interactions and modifications, nor in single material treatments [76]. The development of this Pt optimization method has reported enhancements on both activity and durability of PGM- based hydrogen oxidation at the cathode side. However, the several methods that follow this objective will not be able to meet long term mass activity requirements using conventional nanoparticles [77]. Nonetheless, if further PGM reduction is successfully achieved using this method(<6gPGM/vehicle), the cost of PGM itself will become a smaller fraction of the total fuel cell cost, making it unnecessary to entirely remove PGM [75].

2.3.2 Pt alloy electro catalyst

The second most developed method for reducing the platinum loading of the CLs are the Pt-alloys, like PtCo and PtNi catalysts. These materials are becoming the new baseline catalyst at the commercial level because they are able to achieve better durability than Pt/C while allowing high mass activities [78]. This almost mature optimization method has already been used on the catalyst layer of the Toyota Mirai, 2018 as shown on Figure 15.

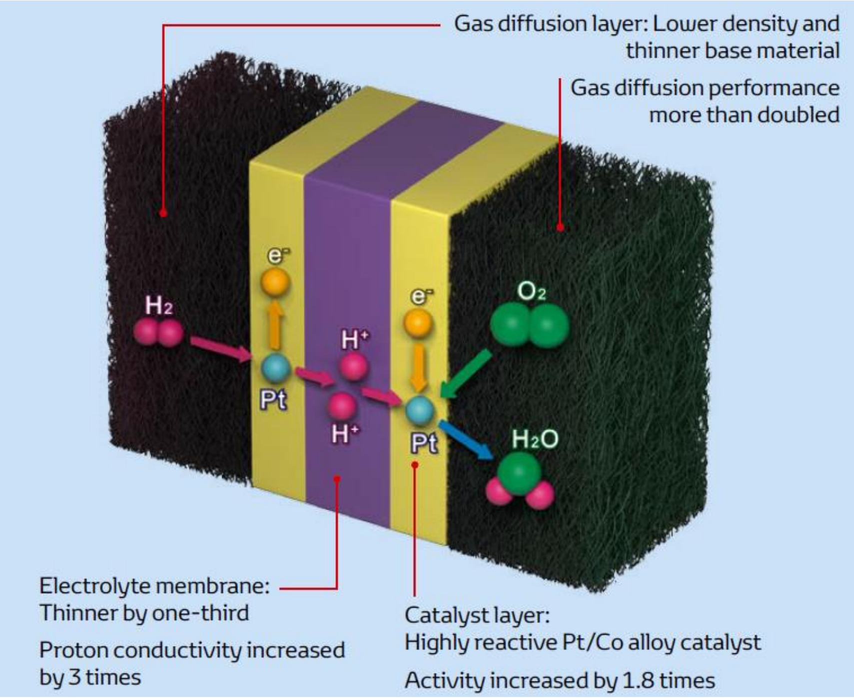

Figure 15. Electrode innovations Toyota Mirai, 2018

Source: [16].

Carbon supported binary and ternary alloys have demonstrated up to three times higher mass activity than Pt/C, these improved electrocatalytic activity of Pt-alloys (Pt with other metals like cobalt, nickel, iron, titanium, aluminum, silver, among others) has been attributed to [26]:

- Smaller platinum bond distances.
- Structure sensitive inhibiting effect of OH ads.

Despite the high mass activity presented, improvements on the stability and durability of these catalysts have to be done. Further work on Pt-alloys has to be performed with the objective of removing the base metal poorly alloyed to the platinum [80]. There are some treatments that can be applied to the Pt-alloy by either acid and heat treatment, inhibiting a improved stability and activity, due to the formation of a richer platinum surface [80]. If these methods are further developed and proof a successful improvement on the durability of alloy catalysts, this type of catalyst could substitute the Pt/C at the MEA, and contribute to the cost optimization of PEMFC [75].

2.3.3 Core shell nanoparticles

During the past years, another type of material achieved an important progress; this is the core-shell nanoparticles, which is shown in Figure 16. This method relies on dispersing Pt particles only on the surface of the catalyst nanoparticle because this is the only active ORR catalyst, while another metal, like palladium, can make up the bulk optimizing the surface area of Pt. This method allows the highest possible platinum utilization, being an attractive cost effective method [81]. Perhaps the reduction of the cost achieved with this type of catalyst, on the cathode side is almost impossible to achieve to achieve PGM loadings <0.1 mg/cm2 with it [82].

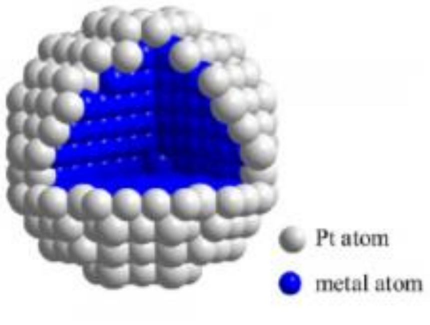

Figure 16. Basic concept of core−shell nanoparticles

Source: [83].

2.3.4 Shape Controlled Nano crystals (Pt-based nanoparticles)

There is another method similar to the core-shell nanoparticles called shape-controlled catalysts, where the structure of the particles are modified in a Nano scale, achieving a high specific activity by generating high mass activity with those nanostructure modifications [84]. At Georgia Institute of Technology, a group of researchers has developed a 9 nm $Pt_{2.5}Ni$ catalyst; despite the low Pt utilization afforded by the large 9 nm particles. In this study, a mass activity of 3.3 A/mg was achieved [85]. This achievement was accomplished through maintaining the ideal $Pt_{2.5}Ni$ crystal structure at a Nano scale; despite the great accomplishment still it has been obtain at a RDE level only, and more research has to be done so that it can be proved in industry [36].

2.3.5 Use of non-Pt group metals at CLs

As the ultimate goal on the development of PEMFC is to eliminate the PGM from the catalyst in order to reduce the production costs, there are some solutions with an approach of zero-Pt that are gaining attention. This approach consists of packing as many active sites as possible into a catalyst based on carbon and nitrogen, allowing a breakthrough in ORR activity [68]. However, non-PGM catalysts in the actual state of the art, are very unstable and this cause a poor performance at high power densities, due to mass transport limitations. This last aspect needs to be improved because it is required to provide acceptable ORR activity [86].

CONCLUSIONS

In the last decades has been proved the benefits that the technology of fuel cells has for the environment and the energy sector. The applications of hydrogen fuel cells range from industrial backup power systems to means of transport like light duty vehicles, buses, trains and heavy-duty trucks. However, this technology still cannot be massively implemented mainly because of one hurdle, the high manufacturing costs. Researchers and developers have identified the use of platinum on the catalyst area of the fuel cell as the major cost of this technology. In order to promote the large-scale commercialization of the fuel cell technology and ensure the transition to a clean transport sector, catalyst developments need to be done.

1. Recent catalyst developments have been done by reducing the platinum loading on the catalyst layers. There are different methods and techniques that are being tested at the stage of research and development, therefore, still it has not been achieved the improvements necessary on the fabrication of PEM fuel cells, which are the type of fuel cells mainly used on the transportation applications.
2. Challenges remain at the catalyst layers, the advances made in the platinum group metals are more advanced than in the development of non-platinum group metals. The development of Pt catalyst supported by carbon is the most advanced method for Pt reduction however still is needed to improve durability and performance through innovative catalyst layer designs that can be modify by different fabrication techniques. Moreover, this is the area of research that has to be explored now.

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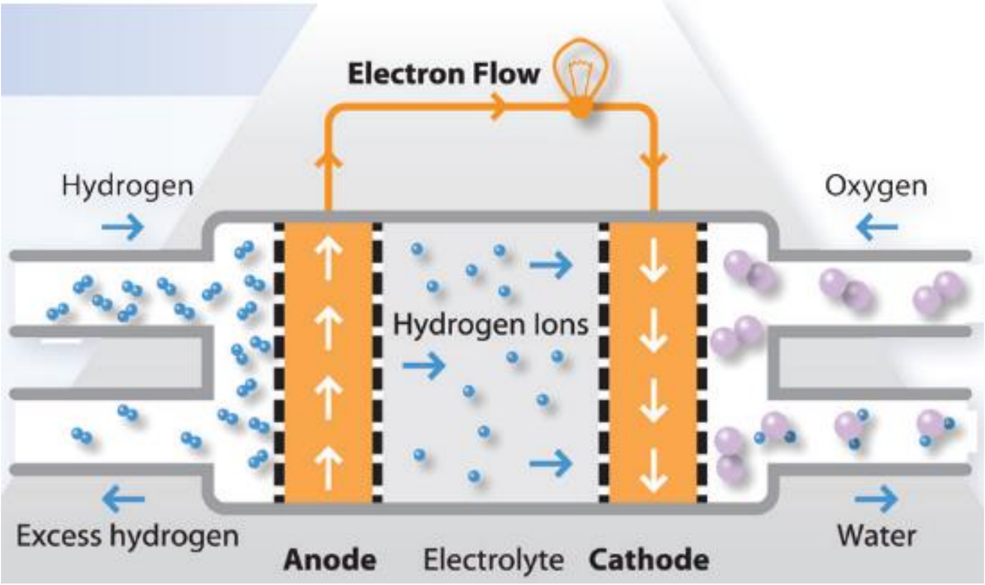
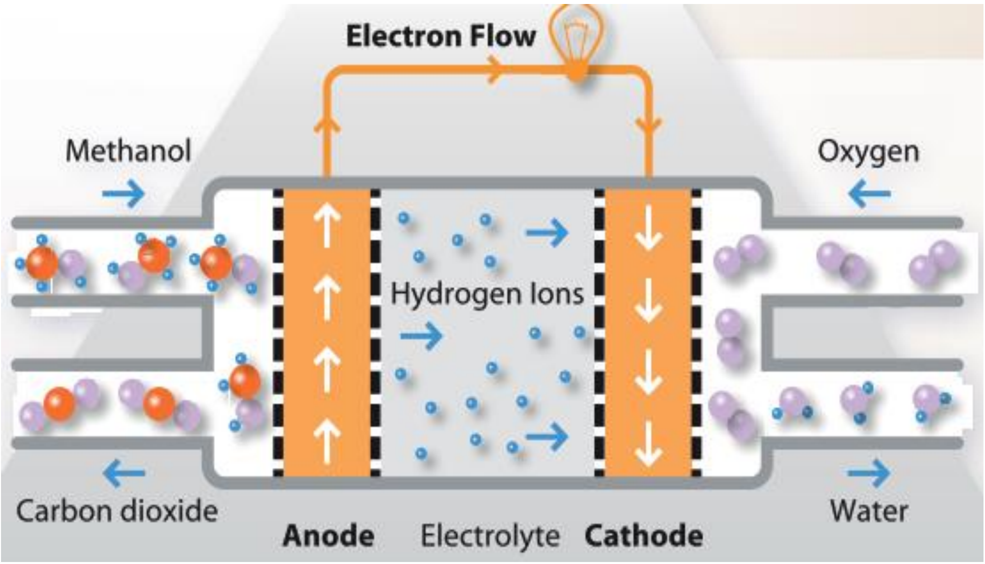
ANNEXES

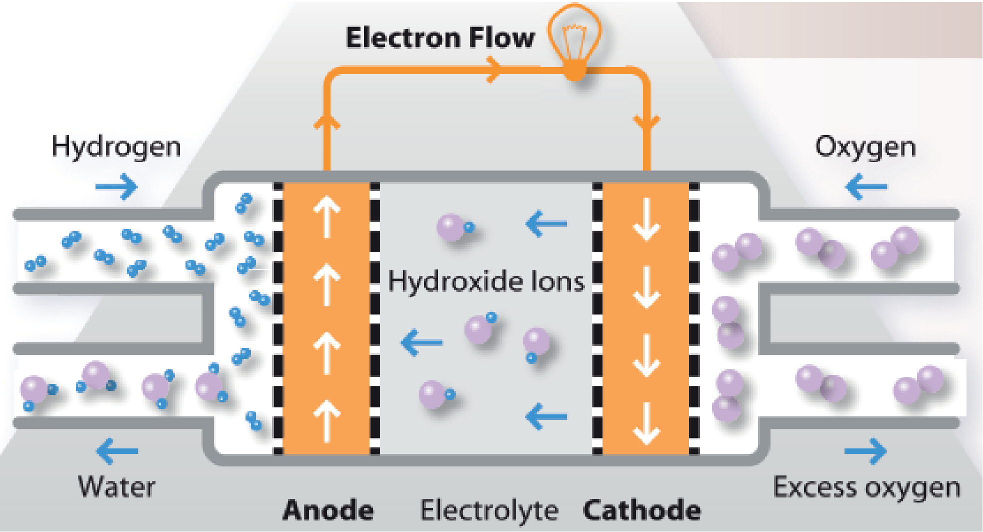
Annex 1 Polymer electrolyte membrane (PEMFCs)



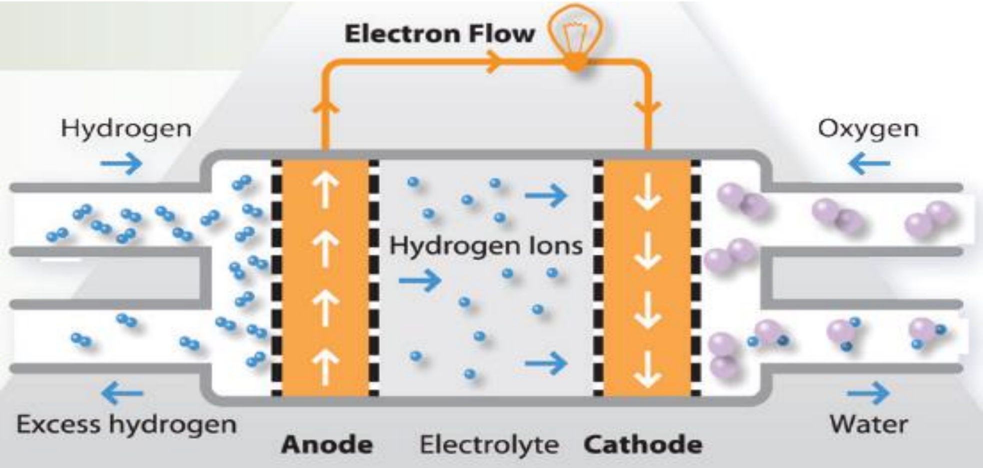
Annex 2 Direct Methanol (DMFCs)



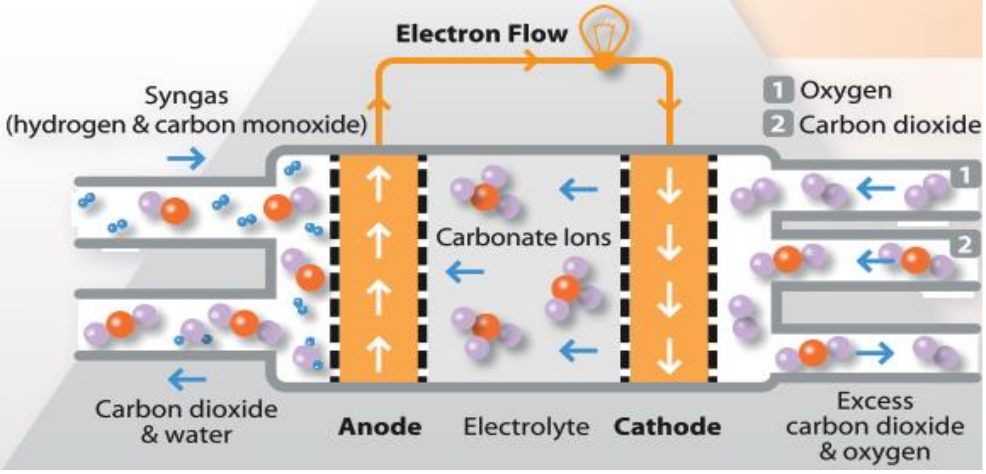
Annex 3 Alkaline electrolyte (AFCs)



Annex 4 Phosphoric acid (PAFCs)



Annex 5 Molten carbonate (MCFCs)



Annex 6 Solid oxide (SOFCs)