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Reversible Fuel Cells Combined with Hydrogen Storage as a Tool to Improve the Electric Grid's Reliability and Resiliency

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ABSTRACT

Based on an ongoing Lawrence Berkeley National Laboratory (LBNL) Project, this Thesis focused on two main objectives: developing a set of Technical Targets (both at cell/stack and at system level) and a Parametric Cost Study for Reversible Fuel Cells, a promising and flexible technology to store excess electricity from Renewable Energy Sources (RES) into hydrogen, hence improving the electricity grid resiliency and reliability. Specifically, the focus is on Unitized systems, in which a single stack can operate in both FC mode and EC modes as this configuration carries some potential cost benefits, since it deals with only one stack and a simpler BOP.

The Technical Targets were developed for both Low Temperature Reversible Fuel Cells (R-PEM) and High Temperature Reversible Fuel Cells (R-SOFC), with the aim of assisting future research towards the most critical Research and Development (R&D) needs, to upgrade performance and durability, and hopefully providing a path towards greater competitiveness with other energy storage technologies. To set these targets, an in-depth literature review on fuel cells, electrolyzer and RFCs was performed, followed by discussions with experts in order to vet the metrics and their values.

The Parametric Cost Study was carried out with the aim of providing researchers and investors with a tool to help them in future Research, Development and Deployment (R&DD) of the technology. Due to the project timing, the costing for stack and BOP components was mainly drawn from existing cost studies. Starting from some key assumptions, the cost study was carried on by producing a set of functional specification, by defining the system design and main BOP components, by costing the stack and the BOP, finally by parametrizing the cost study depending on some key input metrics. The parametric cost study has been set up to calculate the Levelized Cost of Storage (LCOS), too, for an easier comparison with other Energy Storage Systems (ESS). This cost study for R-PEM finds, for a 250 kW fuel cell based system at 100 units/year, a capital cost of $4,557/kWh (including installation costs and margin profits), capital cost that can be pushed, at higher production volume (10,000 units/year), to $2,971/kWh.

THESIS OUTLINE

List of Tables

List of Figures

Figure 22. *Disaggregation of the system by relative percentage of overall system cost for a 250 kW (FC mode) system*

1. INTRODUCTION

This Master's Thesis has been developed during my period at *Lawrence Berkeley National Laboratory* (LBNL) as an affiliate of Max Wei, Research Scientist in the Energy Analysis and Environmental Impacts Division, and under the supervision of Massimo Santarelli, Professor at *Politecnico di Torino*.

The Thesis is based on a Project part of the DOE's (Department of Energy) Grid Modernization Initiative (GMI); specifically, the Fuel Cell Technology Office (FCTO) – under the Beyond Batteries effort – focuses on the concept of Reversible Fuel Cells (RFCs) as a tool to improve the electric grid resiliency. The Project has been carried out also with the collaboration of Ahmad Mayyas, a researcher engineer at *National Renewable Energy Laboratory* (NREL).

1.1. Structure of the Thesis

In the *Introduction*, we started with a brief mention of the operation principles and state-of-the-art of Fuel Cells and Electrolyzer to better delineate the context of the analysis. Then, in the *Aim of the Project* section, we have clarified in detail the purpose of the Project on which this Thesis is based.

The next chapter focuses on unitized *Reversible Fuel Cells* technology (the combination in a single stack of fuel cells and electrolyzer technologies) and analyses its state-of-the-art configuration, with a discussion of its advantages and a brief mention of the most critical Research and Development (R&D) needs.

The following chapter faces the topic of identifying the most significant and effective metrics to describe the performance of RFCs in order to develop a set of technical targets, useful to guide future research and development on the matter. This analysis is carried out for both R-PEM and R-SOFC.

The next chapter works out a system design for UR-PEM, with the aim of determining the main components and cost factors for the stack and the balance of plant, thus producing a parametric cost study.

The last chapter reports the *Conclusions* and closing comments on the results of this research.

1.2. Fuel Cells Basics

Fuel cells have a long history: the first ones to be invented - in the first half of the 19th century, in Europe – were the Low Temperature Fuel Cells, but no practical use was found for another century. Only in the 1950s General Electric Company started developing fuel cells (e.g. Proton Exchange Membrane), mostly for space applications (e.g. Gemini space mission in the 1960s), even though funding in research and development were sparse until the last thirty years, due to the high cost of the materials involved (e.g. platinum catalyst) [1].

Briefly, a fuel cell is an electrochemical cell, that is able to produce electric power through a direct chemical reaction. There are different types of fuel cells depending on the stack composition and the corresponding temperature limits of operation. We concentrate our attention on two main types:

- *Proton Exchange Membrane (PEM)* fuel cell, which works at low temperature because the electrolyte has a good ionic conductivity only if it is fully hydrated, which is why water needs to be at liquid state (i.e. under 100 °C, usually between 50-80 °C)
- *Solid Oxide Fuel Cell (SOFC)*, which works at high temperature (i.e. above 100 °C, usually between 600-800 °C) and consequently needs more strong materials, able to sustain the high temperature and thermal cycles.

The characteristic of operation of a fuel cell is described by the so-called *Polarization Curve*, a graph reporting on the x-axis the current density (usually in A/cm^2) and on the y-axis the cell voltage (in V). The following is an example of polarization curve:

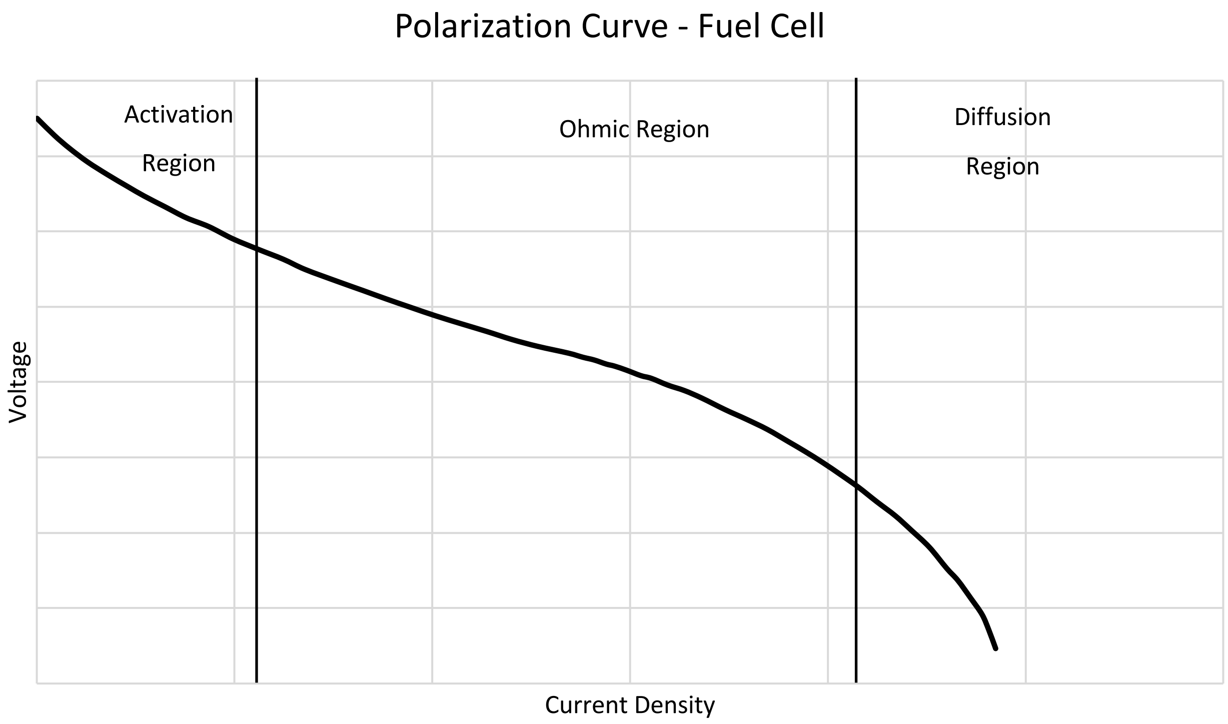


Figure 1. Typical polarization curve for a fuel cell, with Activation, Ohmic and Diffusion Regions

As shown, in a fuel cell a current density increment corresponds to a voltage decrease, caused by different kinds of overvoltage; in particular the graph is divided in three regions based on the corresponding prevailing overvoltage: activation, ohmic and diffusion respectively, each with its peculiar slope.

1.2.1. Proton Exchange Membrane (PEM) Fuel Cells

A single *cell* is composed by three main components: two electrodes (the anode and the cathode, both porous to create a path for the fuel and the oxidant to the point of reaction), and – in between – the electrolyte, whose property is to allow ionic mobility but not electronic one. In actuality, the membrane is not perfect and suffers from hydrogen molecules/electrons transport, a phenomenon called *cross-over effect*. The cross-over effect reduces the performance of the cell in terms of delivered voltage and it is inversely proportional to the thickness of the membrane.
In order to have an overview of the structure, a schematic of a PEM fuel cell is shown below:

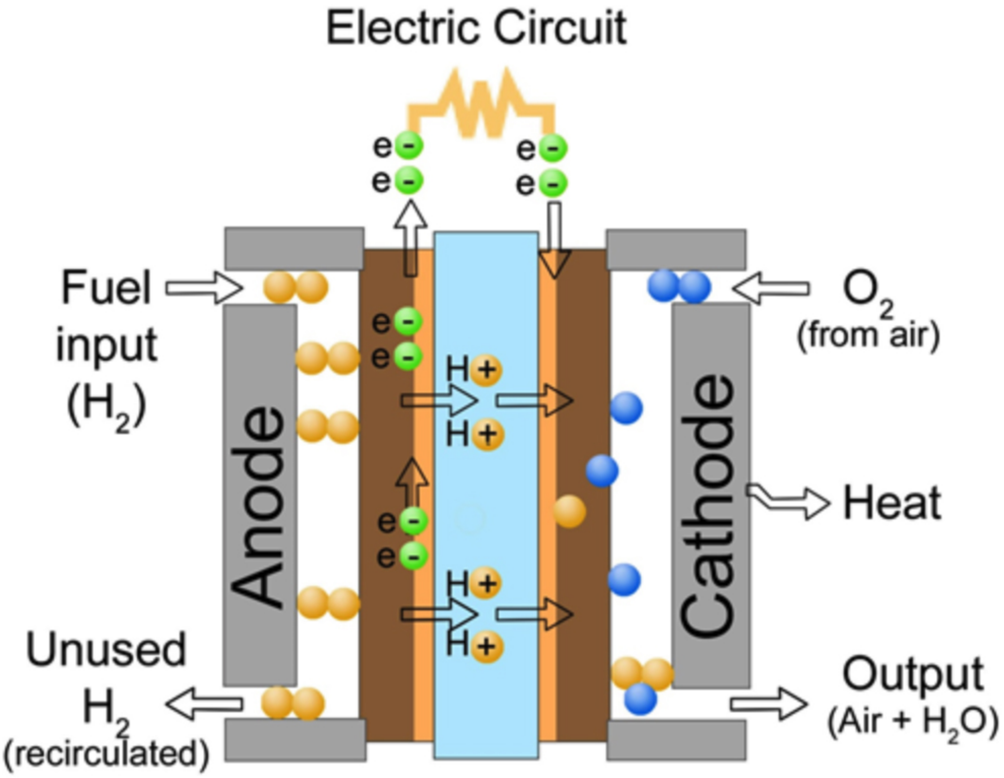


Figure 2. Proton Exchange Membrane Fuel Cell (PEM-FC) schematic [2]

The *electrodes* are composed by a *Gas Diffusion Layer* (GDL), with the function of diffusing the fuel and the oxidant to the point of reaction (called *Three Phase Boundary*, TPB), and by a *Catalyst Layer* (CL); both are made of porous carbon, but the catalyst layer presents a catalyst dispersed at the interface with the electrolyte. In a PEM the two electrodes are identical and made of the same material.

The electrodes have multiple functions, such as:

- to allow an electronic connection with the bipolar plates (explained below)
- to distribute uniformly fuel and oxidant at the TPB
- to mechanically support the Membrane Electrodes Assembly (MEA), i.e. the portion of the cell including the two electrodes and the electrolyte membrane.

The type of *catalyst* depends on the kinetic of reaction, therefore a PEM fuel cell, working at low temperature, needs a very good catalyst such as platinum (or platinum alloy), while SOFC can use a less expensive catalyst such as nickel.

Bipolar plates perform many roles in a fuel cell, from taking care of fuel and oxidant distribution to cooling management, separating single cells in a stack, collecting the current, carrying away the produced water.

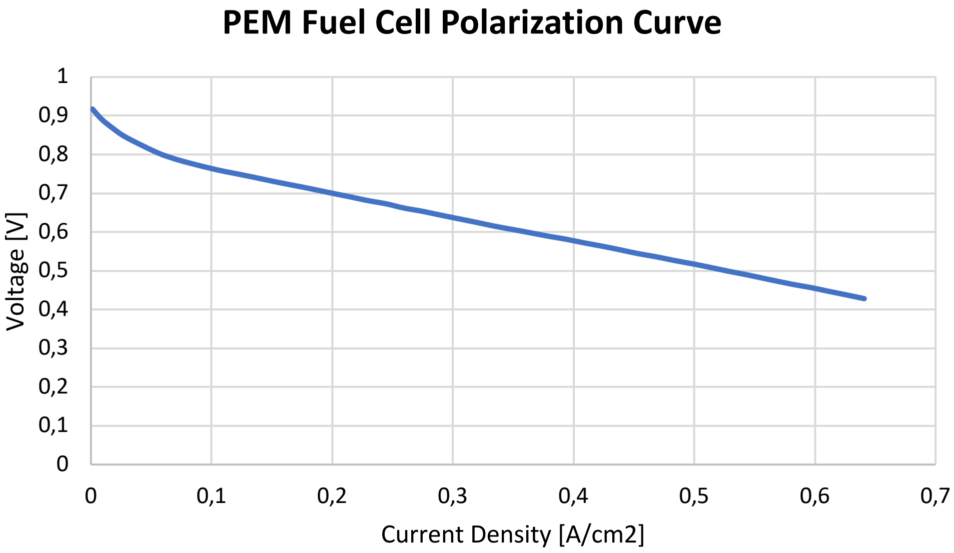


Figure 3. Polarization curve for a PEM fuel cell [3]

The fuel (hydrogen) is fed to the anode side, where the Hydrogen Oxidation Reaction (HOR) takes place, while air (or pure oxygen) is fed to the cathode side, where the Oxygen Reduction Reaction (ORR) occurs. We describe here the operation of a PEM fuel cell, whose reaction equations are reported below:

Anode (HOR) $H_2 \longrightarrow 2H^+ + 2e^-$

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

Complete reaction $H_2 + O_2 \longrightarrow H_2O$

After the HOR, the cations move to the cathode side through the electrolyte, while the electrons move through an external circuit to recombine at the cathode with the cations, creating water. This way, a current of electrons flow across the load.

1.2.2. Solid Oxide Fuel Cells (SOFCs)

The structure of a SOFC is equivalent to the one of a PEM fuel cell, however, a SOFC presents several differences in the operation and in the materials utilized:

- temperature of operation (high temperature, 600-800 °C, over low temperature, 50-80 °C)
- type of catalyst (higher temperature means higher kinetic of reaction, therefore a less expensive and active catalyst is required)
- type of ion transported across the electrolyte (anions instead of cations)
- anode and cathode are non-symmetric and differ in size and materials used (a popular configuration is the anode-supported cell, because it offers high Three Phase Boundary (TPB) where the fuel is oxidized and involves a thin layer of the electrolyte, which means lower ohmic drop)

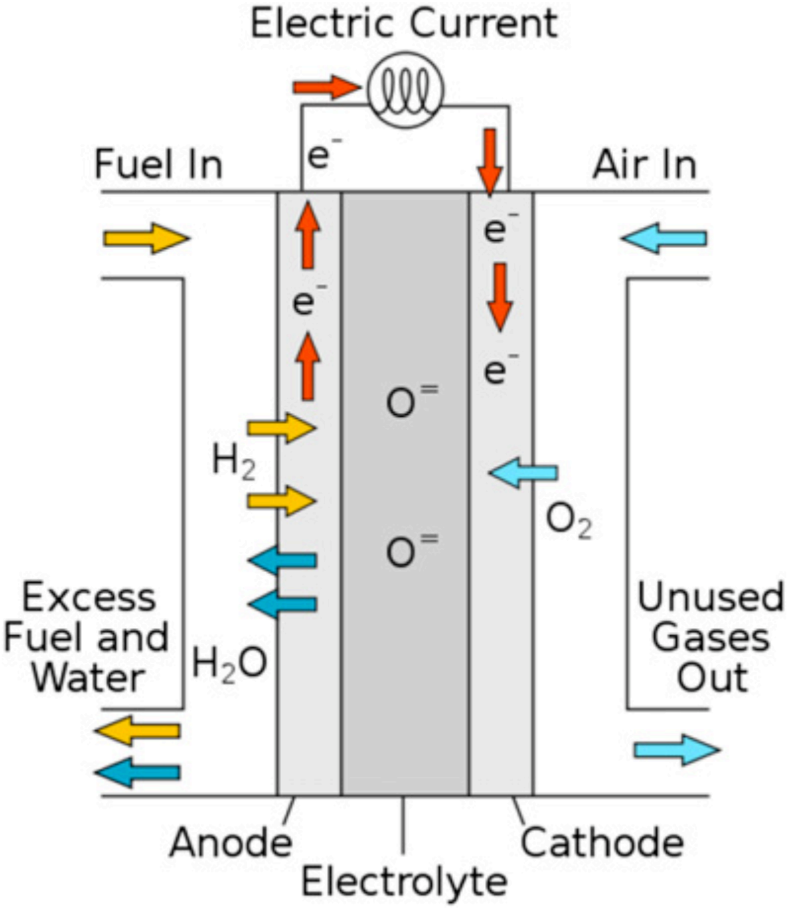


Figure 4. Solid Oxide Fuel Cell (SOFC) schematic [4]

In conclusion, due to the higher temperature, more expensive materials are needed, able to sustain the temperature level and the thermic cycles. For the same reason, both electrodes should have a thermal expansion coefficient similar to the one of the electrolyte to avoid the creation of mechanical stresses within the cell, as well as bipolar plates.

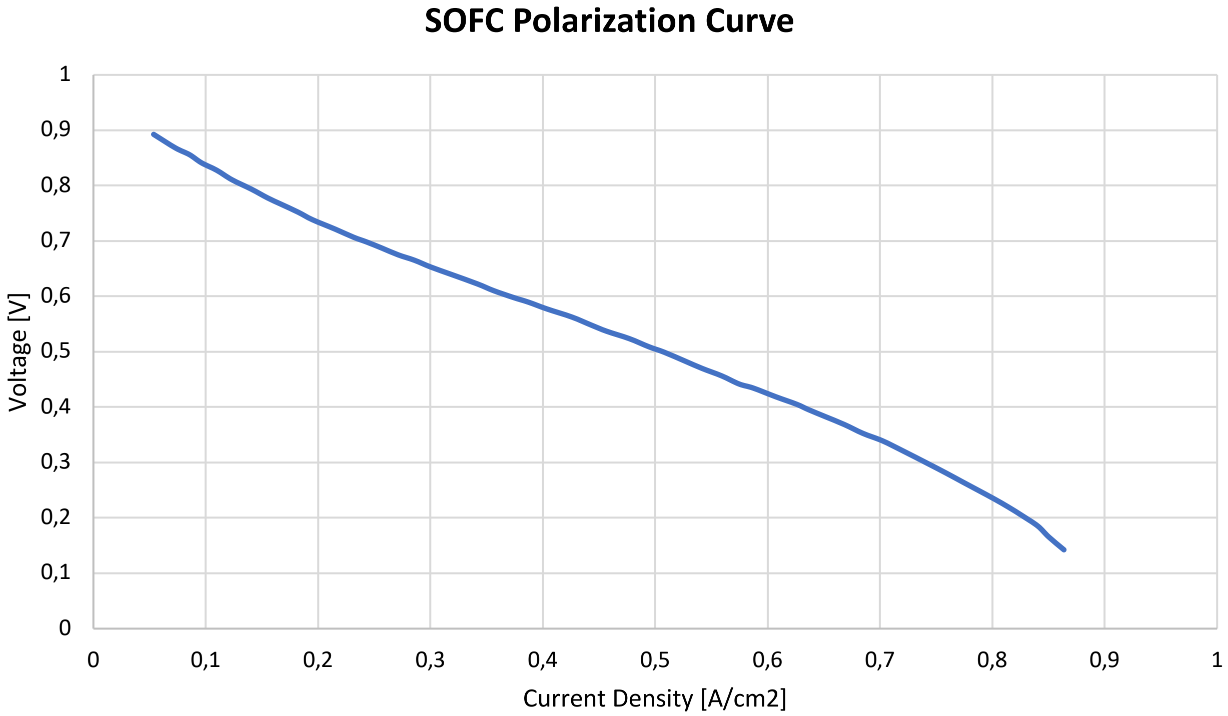


Figure 5. Polarization curve for a SOFC [5]

The presence of a different kind of electrolyte (solid oxide) has a direct influence on the partial reaction taking place at the anode and at the cathode, as shown below:

Anode (HOR)	$H_2 + O^{2-} \longrightarrow H_2O + 2e^-$
Cathode (ORR)	$\frac{1}{2}O_2 + 2e^- \longrightarrow O^{2-}$
Complete reaction	$H_2 + O_2 \longrightarrow H_2O$

In this case, the Oxygen Reduction Reaction (ORR) takes place between the oxygen entering the cell and the electrons, coming from the anode through an external circuit. This reaction creates anions that move through the electrolyte from the cathode to the anode side, where they react with the fuel (hydrogen) to create water and free electrons (Hydrogen Oxidation Reaction, HOR). This way, a current of electrons flow across the load, too.

1.2.3. Fuel Cells: State-of-the-Art

As mentioned above, at the end of the last century an increasing interest in hydrogen and fuel cells has led to major discoveries in research and development in this field.
With the aim to improve performance and lifetime of the technology, an optimization of different components' materials (such as electrodes, electrolyte, bipolar plates, etc.) has been performed.

Nowadays, the state of the art – concerning materials – is:

1.2.3.1. PEM

- *Gas Diffusion Layer*: usually made of porous carbon, their thickness can vary between ~100-300 μm [1]. The cathode side is generally coated with Teflon (which is a hydrophobic material) to improve removal of water from the point of reaction, thus avoiding flooding the electrode
- *Catalyst Layer:* for a hydrogen/air fuel cell, platinum is utilized both as anode and as cathode catalyst. Platinum, being a noble metal, is very expensive and, therefore, the catalyst of the fuel cell represents a major fraction of the overall costs. The usual thickness is ~ 10-15 μm [1]
- *Electrolyte:* is currently based on a perfluorosulphonic acid (PFSA) polymers, one of the most used is Nafion (produced by DuPont); this kind of membrane works properly only if it is fully hydrated. A common membrane's thickness is ~ 25 μm [6]
- *Bipolar Plates:* in order to simultaneously perform all its functions, BPs for stationary applications are typically made out of solid graphite, which is highly conductive, chemical inert, resistant to corrosion, totally dense (usual thickness < 3 mm) [7].

1.2.3.2. SOFCs

- *Anode Electrode*: the usual material for the anode side is *CERMET,* an alloy of a ceramic material (Yttria-Stabilized-Zirconia, YSZ) and of a catalyst (Nickel) [8], [9]
- *Cathode Electrode:* the cathode is constructed from a Mixed Ionic Electronic Conductor (MIEC) material, called *Perovskite*. Nowadays, the most used one is Lanthanum-Strontium-Manganite (LSM) [8], [9]
- *Electrolyte:* the electrolyte material is composed by mixed oxides of metals and rare earth materials (ceramic materials). At this day, the best tradeoff between cost and performance is the Yttria-Stabilized-Zirconia (YSZ) [8], [9]
- *Bipolar Plates:* are usually made out of *CROFER 22 APU,* a high-temperature ferritic stainless steel with very good properties, such as excellent corrosion resistance at high temperature, low coefficient of thermal expansion, good electrical conductivity of the oxide layer (chromium-manganese oxide, $MnCoO_4$), which forms on the surface of the plates for temperature up to 900 °C [9], [10].

1.3. Electrolyzer Basics

At the beginning of the 19th century, shortly after the invention of Alessandro Volta's pile, Anthony Carlisle and William Nicholson first used this device to split water into hydrogen and oxygen. After a little more than a century, during the 1920s and 1930s, different types of alkaline electrolyzer were developed for commercial production.

Moreover, in the 1970s, after the Nafion membrane was discovered by DuPont, General Electric started developing PEM electrolyzer, which present several advantages over the alkaline ones. However, due to their high production costs and limited lifetime, their use was limited until the past two decades when the growing interest for low carbon and Renewable Energy Sources (RES) brought to the commercialization of this technology, able to store excess of energy as hydrogen [11].

The principle of operation of an electrolyzer is opposite with respect to the one of a fuel cell, hence in this case the cell receives electricity (e.g. coming from the grid or a local renewable energy source) and water to produce – through water electrolysis – hydrogen and oxygen; the names given to anode and cathode, too, are reverse, meaning that the anode in an electrolyzer corresponds to the cathode in a fuel cell and vice versa.

1.3.1. Proton Exchange Membrane (PEM) Electrolyzer

PEM electrolyzer shares the same structure as PEM fuel cell, but – as mentioned above – the cell is working in reverse operation. Therefore, the open electrochemical cell is composed by two electrodes (anode and cathode) separated by an electrolyte material, whose purpose is to allow ionic transit from the anode to the cathode.

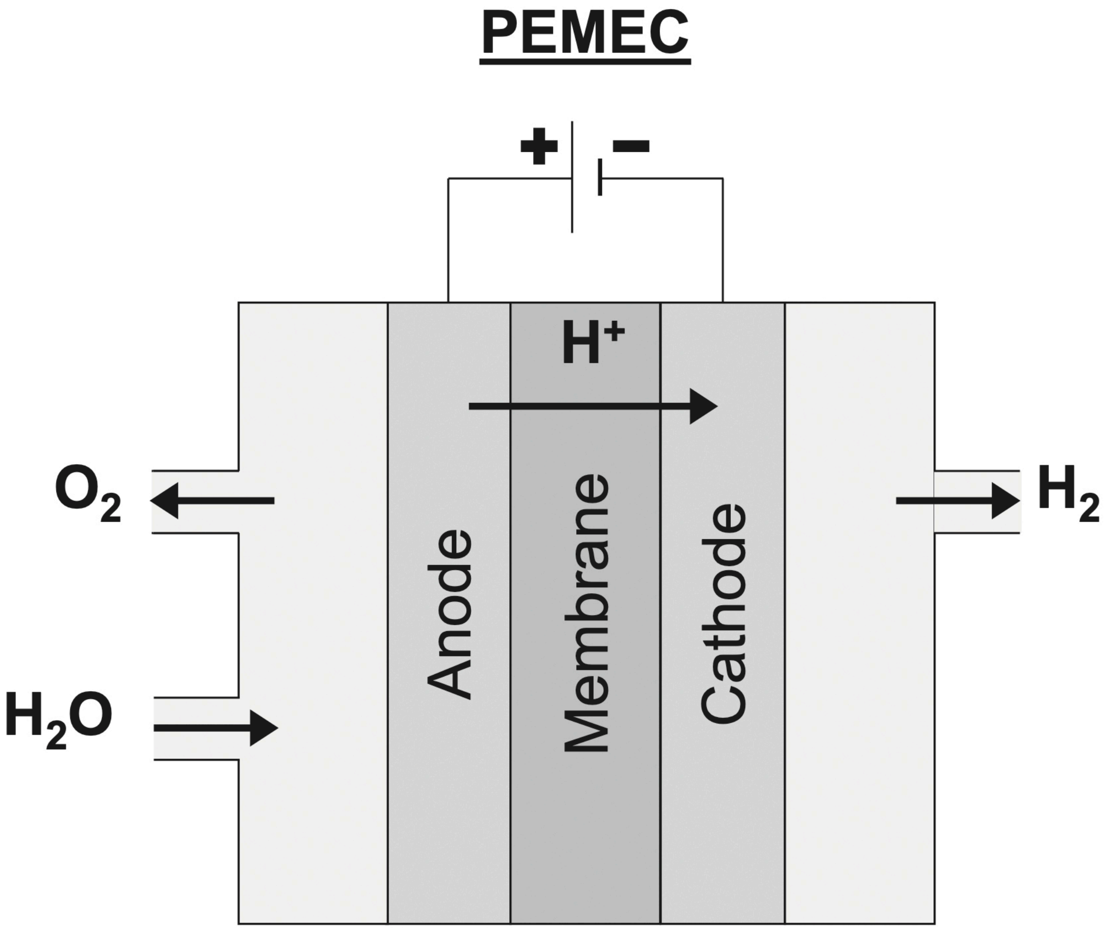


Figure 6. Proton Exchange Membrane Electrolyzer Cell (PEM-EC) schematic [12]

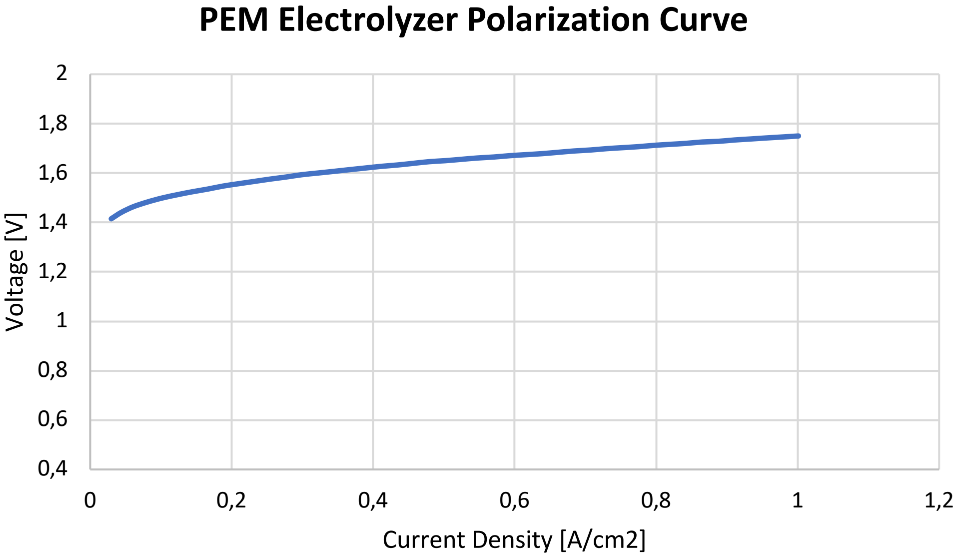


Figure 7. Polarization curve for a PEM electrolyzer [11]

Specifically, the electrochemical reactions taking place in this kind of electrolyzer are:

Anode (OER)	$H_2O \longrightarrow \frac{1}{2}O_2 + 2H^+ + 2e^-$
Cathode (HER)	$2H^+ + 2e^- \longrightarrow H_2$
Complete reaction	$H_2O \longrightarrow H_2 + \frac{1}{2}O_2$

Water is fed to the cell (anode), powered by electricity, where the *Oxygen Evolution Reaction* takes place and delivers oxygen and hydrogen cations. The cations move through the electrolyte towards the cathode, while electrons move through an external circuit to recombine with the cations at the positive electrode to create hydrogen (*Hydrogen Evolution Reaction).*

However, these reactions have different a kinetic from the one of a fuel cell and need a different type of catalyst to optimize the reverse operation over the direct operation. The problem is particularly marked for the catalyst used at the cathode, because the reducing environment and the higher voltage are extremely harmful for platinum.

Due to the similar structure, PEM electrolyzer also share the same problematics with fuel cells, like the *cross-over effect* that affects the voltage of the cells.

1.3.2. Solid Oxide Electrolyzer Cells (SOEC)

The structure of Solid Oxide Electrolyzer Cells, too, is similar to the one of a SOFC with a few precautions regarding the materials used at the electrodes. More information regarding these issues will be discussed in the "Electrolyzer - State of the Art" section below.

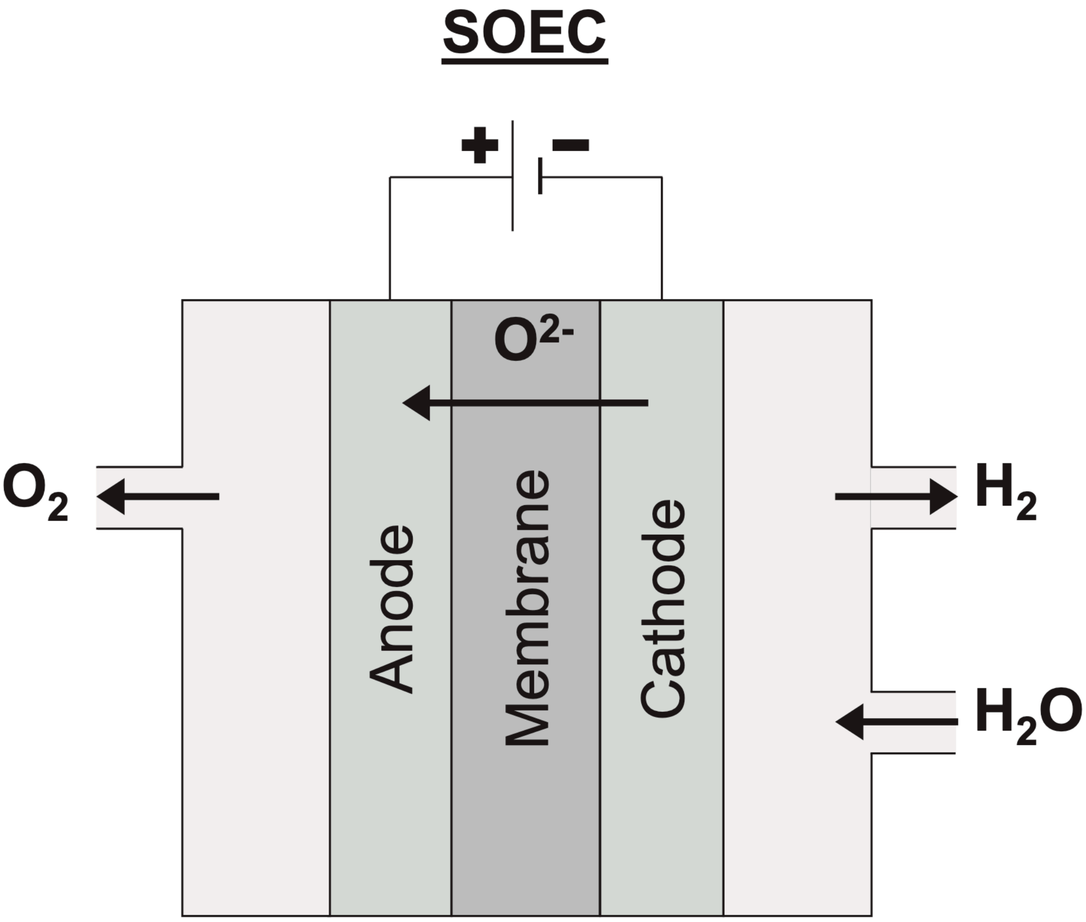


Figure 8. Solid Oxide Electrolyzer Cell (SOEC) schematic [12]

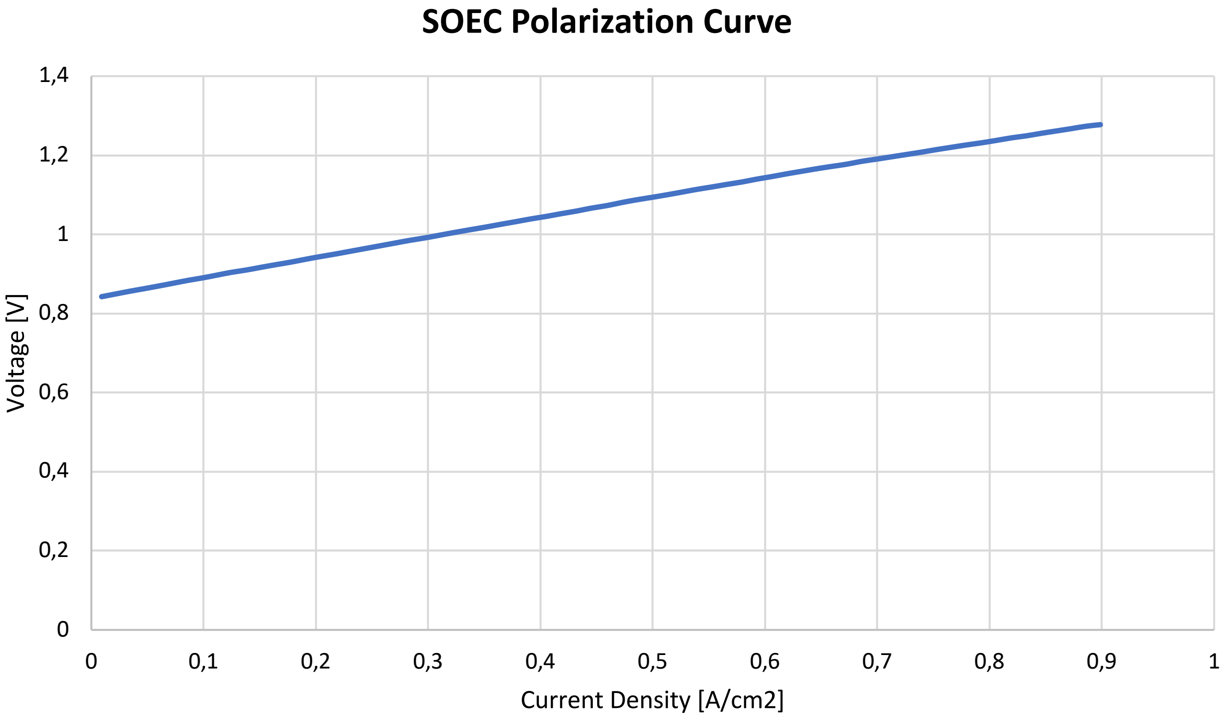


Figure 9. Polarization curve for a SOEC [13]

The following equations describe the operation of a SOEC:

Anode (OER)	$O^{2-} \longrightarrow \frac{1}{2}O_2 + 2e^-$
Cathode (HER)	$H_2O + 2e^- \longrightarrow H_2 + O^{2-}$
Complete reaction	$H_2O \longrightarrow H_2 + \frac{1}{2}O_2$

In this case, due to the high temperature, steam coming into the cell undergoes a reduction reaction at the cathode, where water molecules are dissociated into hydrogen (useful product coming out from the cell) and oxygen anion, under the application of the required electrical potential. The ions move through the electrolyte and are oxidized at the negative electrode to obtain oxygen (biproduct of the cell).

1.3.3. Electrolyzer: State-of-the-Art

Keeping in mind the tradeoff between cost and performance, in this section we analyze the state-of-the-art materials that ensure the optimal operation of electrolyzer.

1.3.3.1. PEM

- *Gas Diffusion Layer:* for the cathode side the simplest solution is to use carbon-based porous materials, because metallic materials suffer from embrittlement (i.e. metals become fragile) when in contact with hydrogen. The highly corrosive environment of the anode, due to the simultaneous presence of water, oxygen and high potentials (>1.5 V), is not compatible with carbon-based porous material; therefore, a good solution is to utilize sintered porous titanium, that has a good electrical conductivity and an excellent corrosion resistance compared to others non-precious metals. A usual value is ~280 μm [11], [14]
- *Catalyst Layer:* anode and cathode need different catalysts, because of the highly corrosive environment at the negative electrode. Hence, for the cathode the typical platinum catalyst can be used, while for the anode a widely used option is iridium or its oxides. Usual Platinum Group Metal (PGM) loadings are 1 mg/cm^2 for the cathode and 2 mg/cm^2 for the anode [11]
- *Electrolyte:* the most widely used is Nafion (by DuPont), which has the same constraints regarding the temperature of operation as the fuel cells; the membrane is, on the other hand, thicker. Usual values vary between 100-400 μm [11]
- *Bipolar Plates:* a widely used option is stamped stainless steel (e.g. 316L sheets) with a thickness ~5 mm. A gold coating (with a thickness of ~ 100 nm) can be used to improve their electrical conductivity and resistance to oxidation [11], [14].

1.3.3.2. SOECs

- *Electrolyte*: the most widely used material is Yttria-Stabilized-Zirconia (YSZ), which has good ionic conductivity (only at high temperature) and good mechanical strength [15]
- *Anode Electrode*: nowadays, is typically made out of Lanthanum-Strontium-Manganite (LSM) or LSM-YSZ composite. These materials present a thermal expansion coefficient close to the one of the YSZ electrolyte to avoid the formation of internal stresses [15], [16]

- *Cathode Electrode*: the material is the same as the one utilized for the SOFC's anode, *CERMET* – an alloy of a ceramic material (YSZ) and of a catalyst (Nickel) – which has as well a thermal expansion coefficient close to the one of the electrolyte
- *Bipolar Plates*: are usually made out of *CROFER 22 APU,* similarly to SOFC [10], [16].

1.4. Aim of the Project

This Thesis – part of an ongoing Lawrence Berkeley National Laboratory (LBNL) Project, under the supervision of the Department of Energy's (DOE) Fuel Cell Technology Office (FCTO) – focuses on the analysis of Reversible Fuel Cells, aiming to assess the capabilities of this technology as a tool to improve the reliability and resiliency of electric grids. The aforementioned project falls under the wider DOE's Beyond Batteries effort – part of the U.S. Department of Energy's Grid Modernization Initiative (GMI) – concentrating on advances in controllable loads, hybrid systems, and new approaches to energy storage to increase the reliability and resiliency of energy systems [17].

Reversible Fuel Cells (RFC) are an energy storage system that is able both to produce and store energy. It consists of a combination of two different technologies, Fuel Cell and Electrolyzer, and hence it is able to produce hydrogen (electrolyzer mode) to be later used to produce energy (fuel cell mode).

Especially appealing is the concept of Unitized Reversible Fuel Cells, i.e. a single stack can operate in either modes (fuel cell or electrolyzer); hence this configuration has some obvious cost benefits – unlike Discrete systems – since it deals with a single stack instead of two and with a simpler Balance of Plant (BOP). However, a few difficulties have to be solved to make competitive a unitized system; difficulties related, for instance, with the kinetic of reaction and the development of an effective bifunctional electrode, able to sustain the different environments of the two modes of operation.

The various kinds of RFCs can be differentiated – based on the temperature of operation – in two groups: Low Temperature ones and High Temperature ones. Among those in the first group the

Project has focused its attention on Reversible Proton Exchange Membrane Fuel Cells (R-PEMFC), while among those in the second group on Reversible Solid Oxide Fuel Cells (R-SOFC), both considered as unitized systems.

As RFC technologies are at an early stage and need further research and development to improve performance and durability, firstly the Project sets itself the objective of developing a set of Technical Targets in order to provide insights on the most critical R&D needs, both at cell/stack and at system level. Secondly the Project proposes to evaluate the overall costs of a RFC system, by estimating the costs of both the stack and the Balance of Plant, and, successively, by making it parametric. Since the Project is still ongoing, the cost analysis is going to be limited to the R-PEM technology alone, while the cost analysis on R-SOFC is still under development. All the above is performed to asses if URFC technology is competitive compared to other energy storage technologies.

2. REVERSIBLE FUEL CELLS

Unitized Reversible Fuel Cells (RFCs) are an innovative and promising energy storage technology able to produce large amounts of energy at potentially lower cost than using discrete FCs and electrolyzer units, since they can accomplish two different tasks: power production (fuel cell mode) and energy storage (electrolyzer mode). In the global trend of including more and more intermittent renewable energy sources into the national energetic mixes, this characteristic of dual operation combined with a potential cost reduction fits extremely well, because it allows to avoid curtailment of energy produced by renewables by transforming into hydrogen the excess electricity.

A RFCs system can be organized in two different configurations: as a Discrete System - composed by two separated stacks, one for fuel cell and one for electrolyzer operation – or as a Unitized System, if both operations are carried out in a single stack.
The second configuration has some important potential economic benefits, since it needs only one stack to perform both functions and there is also potential simplification regarding the Balance of Plant (BOP). However, a unitized system has lower performance because it has to be optimized for two different operations, each with different needs.
It is important to point out that the majority of RFCs demonstration systems to date are discrete systems, due to the challenging technical issues that need to be solved in unitized systems to achieve an effective dual modes operation. To make an unitized system competitive over a discrete one, one of main objectives is to demonstrate its superiority in performance, durability and costs [18].

The first effort to develop a URFC (PEM) was done at the end of the 1960s. However poor performance was recorded due to problems regarding the catalyst and the membrane. During the 1970s General Electric Company made some promising improvements, but we have to wait the early 1990s for the development of the first portable URFC by the Lawrence Livermore National

Laboratory (LLNL), with a high specific energy density (450 Wh/kg). Only at the beginning of the 2000s, the Proton Energy Systems company (now called Proton Onsite) marketed the first URFC commercial system, called Unigen (consuming 15 kW_e in electrolyzer mode and producing 5 kW_e in fuel cell mode). Over the last years – helped also by the concept of a *Hydrogen Economy* – more and more research projects are being funded worldwide [18], [19].

Since the traditional fossil fuels are an unsustainable energy source, as they have limited reserves and they are worsening the already severe climate change problem, in this scenario the RFCs technology has the potential to become a promising solution to face the problem of intermittency of Renewable Energy Sources (RES). An RFC can produce hydrogen when there is an excess of renewable electricity, store it and then provide electricity back to the grid when it is needed by converting hydrogen to electricity using a fuel cell. Therefore, it has the great advantage of using an abundant chemical compound (water) as a fuel carrier and, moreover, the only emission from the system is water (or steam, depending on the temperature of operation), that can be stored and used in the following electrolysis phase or vented to the atmosphere, creating this way an almost perfect loop [19], [20]

Unlike other energy storage technologies – such as batteries – the electrodes of an RFC can be sized just on the basis of rated power required and not of the storage duration. In other words, it is possible to increase the storage duration just by increasing the size of the hydrogen storage itself, instead of increasing the area of the electrodes, which is much more economically feasible. RFCs have a very low self-discharge rate, too. Keeping this in mind, overproducing hydrogen to be sold for other end-uses (e.g. hydrogen based industrial processes, fuel cell electric vehicle fueling stations) can be an interesting possibility to exploit, same as seasonal energy storage in immense underground salt caverns.

Another notable trait that RFCs have compared to other energy storage technologies – like pumped storage hydropower, Compressed Air Energy Storage (CAES) and batteries – is its flexibility and high specific energy (400-1000 Wh/kg) [21].

Nevertheless, there is still a strong need for funds in Research and Development (R&D) to optimize the performance of unitized systems, allowing them to reach performance close to the ones of discrete systems with lower investment costs.

Since our study is focused on Unitized Reversible Fuel Cells, from now on when we mention RFCs, we are referring to a Unitized system.

2.1. PEM - Reversible Fuel Cells

An RFC is not obtained just by combining the state-of-the-art materials of a fuel cell and of an electrolyzer. There are indeed several issues related to the different material requirements needed by fuel cell (FC) and water electrolysis (WE) modes. From this point of view there are two possible configurations of a PEM-RFC:

- Configuration based on the concept of "*Hydrogen and Oxygen Electrodes*", whereby hydrogen and oxygen get in contact always with the same electrode in both modes of operation. This configuration allows for a simpler management of gases, but shows severe matters of material stability, due to the high potential applied at the anode (cathode in FC operation) during WE mode (the potential can indeed vary by more than 1 V, going from FC to WE mode). Hence, the cell is affected by several degradation factors, the most critical of which is a serious corrosion of the carbon-based materials in GDL and BPs, corrosion that leads to lower performance and durability. Among other factors, it is important to highlight the problems related to the electrocatalyst (i.e. aggregation and migration of the catalyst particles, whose properties suffer a loss) [19], [21]–[23]
- Configuration based on the concept of "*Reduction and Oxidation Electrodes*", whereby each electrode is specialized for a specific redox process, meaning that each electrode is going to perform always the same reaction (reduction or oxidation). This configuration allows for a simplification of the material and structure of the electrodes, since it is possible to optimize them for a single redox function, leading to higher performance and to a more stable operation under cycling conditions. The chambers of the electrodes, however, need to be purged before switching operation mode. Even if this configuration seems more promising to reach a higher efficiency, it is a newer concept that needs further research to reach maturity [19], [21]–[23].

To better understand these two possible configurations, we show below a scheme reporting the operation and the electrochemical reactions taking place at the different electrodes:

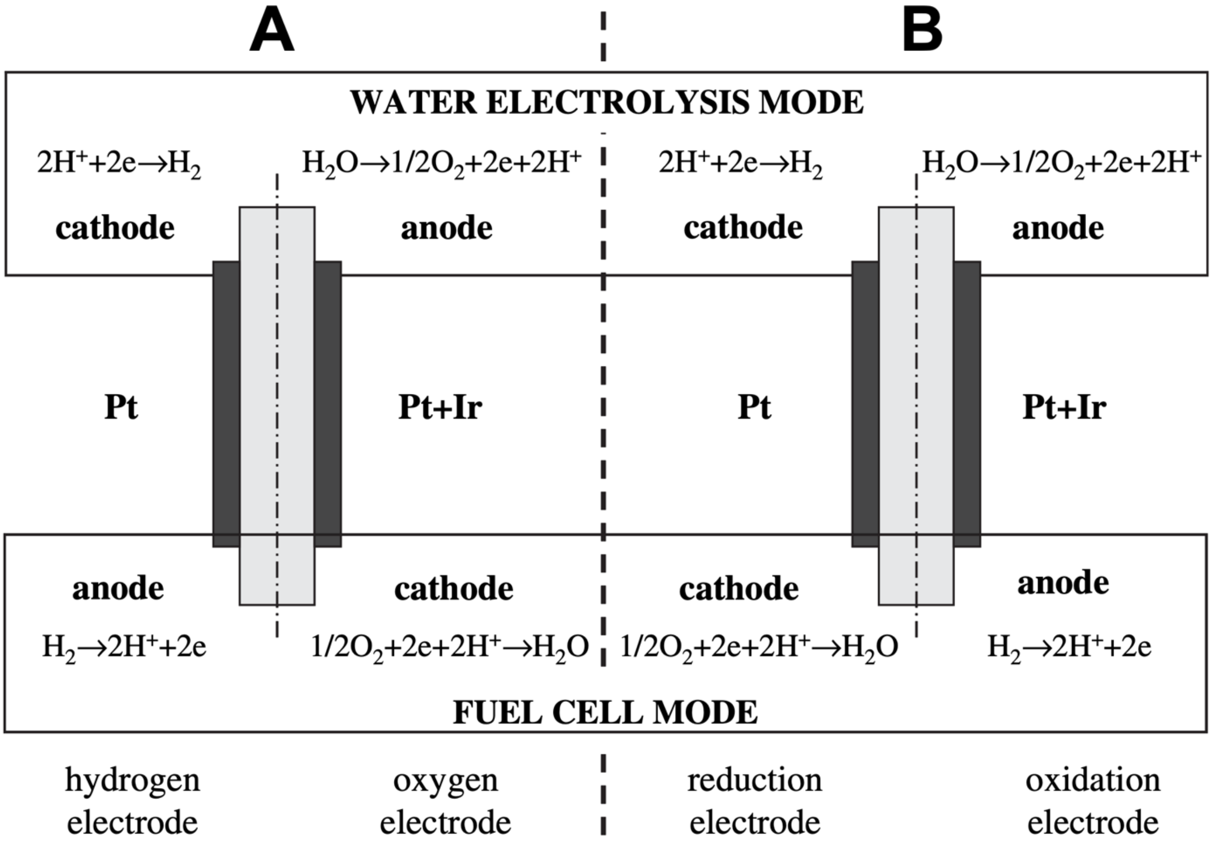


Figure 10. Operation of a Unitized Reversible PEM Fuel Cell in its two possible configurations: (A) hydrogen and oxygen electrodes; (B) reduction and oxidation electrodes [22]

In our study we concentrated on the "*Hydrogen and Oxygen Electrodes*" configuration, because of its greater maturity, hence, the higher number of studies about it. For this reason, we deepen here below the state-of-the-art materials for a R-PEM fuel cell based on this configuration.

2.1.1. State-of-the-Art ("Hydrogen and Oxygen Electrodes" configuration)

Based on several researches and experiments performed, the optimal choice to date regarding materials is described here:

- *Gas Diffusion Layer*: the material needs to face several issues. As the usual GDL material of a PEM fuel cell (porous carbon) is not suitable for a unitized cell, because it is usually hydrophobic – to avoid flooding of the electrode and to help with the water management of the cell – while in electrolyzer mode the material needs to be hydrophilic (since it needs to enhance the water molecules transport to the point of reaction), therefore an appropriate balance between hydrophobic and hydrophilic properties needs to be performed. As stated previously, another issue is the severe corrosion of carbon-based materials in the oxygen electrode, due to the high potential applied. A good material capable of fulfilling these requests is woven metal cloth, usually made out of titanium, because it is able to supply both strength to the electrodes as well as a good electrical connection between the BPs and the electrodes [18], [24], [25]

- *Catalyst Layer*: platinum – the usual electrocatalyst for PEM fuel cell – is still the most efficient Bifunctional Hydrogen Catalyst (BHC) for the hydrogen electrode. On the other hand, regarding the oxygen electrode, it performs extremely well for the Oxygen Oxidation Reaction (ORR), but very poorly for the Oxygen Evolution Reaction (OER). The choice of a suitable Bifunctional Oxygen Catalyst (BOC) is complex; a good compromise is achieved by a mixture of platinum and another noble metal (i.e. iridium and rhodium, which are the preferred catalysts for the OER). The best performance under cycling condition has been obtained mixing platinum with iridium oxide (Pt/IrO_2) [18], [24]–[27]
- *Electrolyte*: as for PEM fuel cells and electrolyzer, the membrane is based on a perfluorosulphonic acid (PFSA) polymer. The most used one is Nafion (by DuPont) [21], [25]
- *Bipolar Plates*: for the same corrosion problem of carbon-based materials, porous carbon is not an option for a RFC. Metal-based BPs, instead, are a good solution; in particular titanium is a good alternative, thanks to its excellent corrosion resistance, mechanical strength and light weight. Its only issue is the formation of a layer of titanium oxide (TiO_2) on its surface, that lowers the electrical conduction of the material; to avoid this problem a coating can be used, usually made out of gold, that is very performant but – obviously – expensive [18], [25], [27].

2.2. SOFC - Reversible Fuel Cells

Among high temperature reversible fuel cells, Reversible Solid Oxide Fuel Cells (which work in a temperature range between 700 °C and 800 °C) stand out for their considerable high

round trip efficiency (RTE) and, for this reason, are a promising technology for large-scale production both of hydrogen and electricity. Moreover, working at high temperature enables the unit to function in WE mode with better thermodynamics of reaction; since a larger fraction of the energy demand, ΔG (Gibbs free energy of reaction), is supplied as heat (ΔH), this determines a considerable reduction of the electrical power required and as a result of the related cost.
An additional advantage, in WE mode, is the possibility to exploit high grade waste heat, from high temperature industrial processes or nuclear power plants, to provide energy for the system.

The operation a R-SOFC is described in the following scheme:

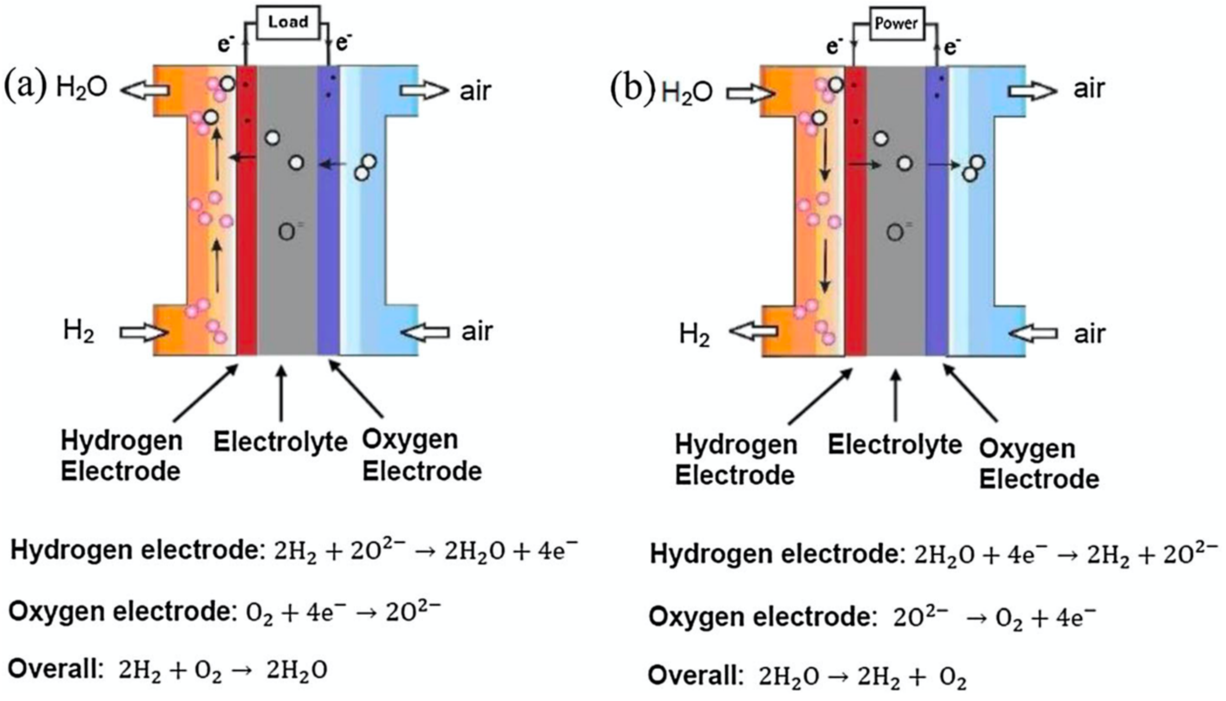


Figure 11. Operation of UR-SOFC: (a) operation as fuel cell; (b) operation as electrolyzer [20]

As for R-PEM, R-SOFC, too, shows issues at the oxygen electrode during WE mode, due to the high potential applied; in particular, the delamination and degradation of the electrode are the outcomes of internal stresses originated at the interface between the electrode and the electrolyte. Nevertheless, over the last years there have been a number of studies demonstrating the beneficial impact of the cathodic polarization (SOFC mode) over the degradation of the electrode, improving – ultimately – its lifetime [20].

2.2.1. State-of-the-Art

Even though the materials utilized for a SOEC are similar to the ones of a SOFC, the atmosphere of the electrodes in the two operating modes imposes some specific material requirements.

Based on several researches and experiments performed, the optimal choice to date regarding materials is described here:

- *Electrolyte*: the most used material for both SOFC and SOEC – Yttria-Stabilized-Zirconia – is suitable for R-SOFC operation too. It is a very good O^{2-} ion conductor and it is stable under both atmospheres of the electrodes [20]
- *Anode Electrode*: CERMET, a mixture of Nickel and YSZ (Ni/YSZ), is the typical material – both for SOFC and SOEC – thanks to its great electrical conductivity, catalytic activity and low price. However, there are two problems affecting the electrode: the deposition of silica impurities (contained in the stack raw materials) at the Three Phase Boundary, that lowers the performance; and Ni loss or Ni agglomeration, if the system is working on clean fuels [20], [28], [29]
- *Cathode Electrode*: a widely used material is Lanthanum-Strontium-Manganite (LSM)-YSZ composite. Compared with LSM on its own, it presents an increased reactive area and as a consequence an increased activity of the electrode [20], [28]
- *Bipolar Plates*: usually made out of *CROFER 22 APU,* similarly to SOFC and to SOEC [30]

3. TECHNICAL TARGETS FOR REVERSIBLE FUEL CELLS

Since the Reversible Fuel Cells technology is a promising opportunity to improve electric grid reliability and resiliency, but it is still at an early stage of development, the Project aims to produce a set of Technical Targets to assist future Research and Development (R&D) towards the most critical needs to upgrade its performance and durability, especially with the focus of making it competitive in the context of energy storage technologies.

Our study – as said before – is focused on Unitized Reversible Fuel Cells technology, since carrying out both modes of operation (fuel cell and electrolyzer) in a single stack has some potential cost benefits compared to Discrete systems. For this reason from now on when we mention RFCs we are referring to a Unitized system.
Two different sets of Technical Targets have been developed: one for Low Temperature (PEM) and one for High Temperature (SOFC) RFCs. In both cases, the targets have been developed both at cell/stack and at system level. Due to the different requirements of low and high temperature RFCs, some specific metrics have been used either for one type or the other.

3.1. Methodology and Metrics Choices

As a first step in approaching the development of these targets, we conducted an in-depth literature review regarding RFCs, fuel cells and electrolyzer, both for performance and cost studies. We have observed that in literature there is much less documentation about cost estimations for unitized RFCs than for discrete ones and, moreover, much less material about performance and durability for R-PEM than for R-SOFC.

Due to this shortage or lack of comprehensive costs information related to RFCs, some assumptions have been made. Specifically, as a starting point, we took as specific cost ($/kW) for this technology the cost of fuel cells, because typically fuel cell costs are higher compared to electrolyzer ones. Therefore, the costs had been largely drawn from existing cost studies on PEM, both fuel cells and electrolyzer, and on SOFC and based on some key assumptions. For instance, because of the lack in literature for SOEC cost studies and due to its similarity in materials and structure, its cost has been assumed to be close to the one of a SOFC.

Additionally – in the case of R-PEM – when some metrics information was lacking, we utilized R-SOFC one, as a first approximation.

The cost of the electrolyzer has been considered into the cost of power (Cp) since the system is composed by a single unit with function of both fuel cell and electrolyzer. This is consistent with the work done by Sandia Laboratory [31] on discrete RFC, where both the costs of fuel cell and electrolyzer were allocated in the cost of power. Similarly, Pumped-Hydro Storage (PHS) system allocates the cost of pumps needed to charge the upper reservoir in the cost of power [32].

Beyond the literature review, to better understand which metrics are to be included in the targets, we started by looking at Table 3.4 of the DOE's QTR (Quadrennial Technology Review, 2015) which reports the costs and performance targets for electric energy storage technologies, as shown below [33].

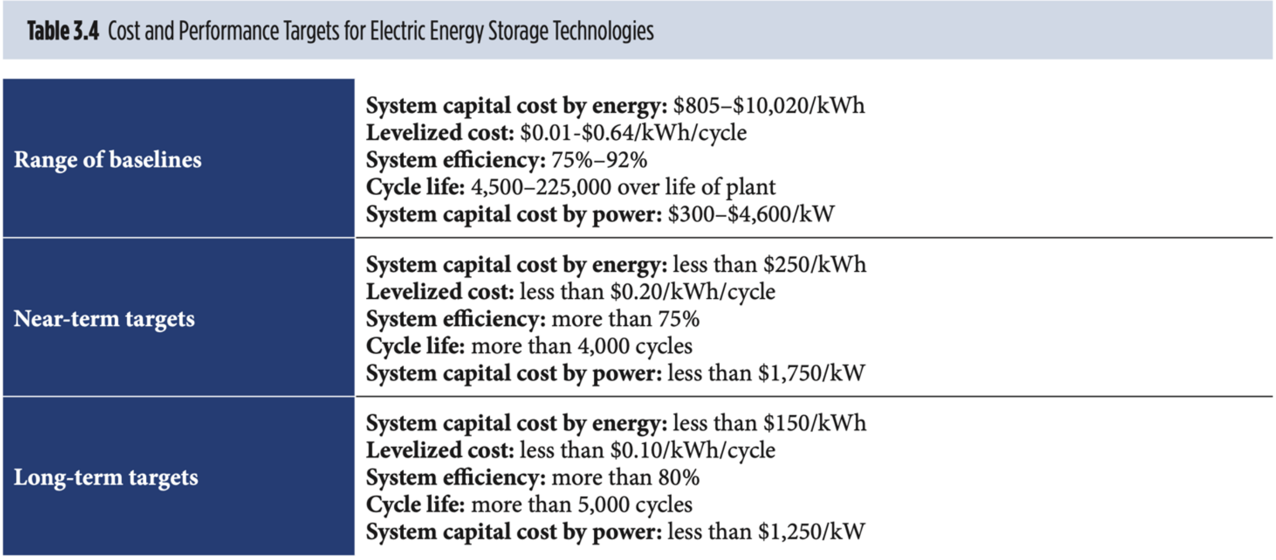


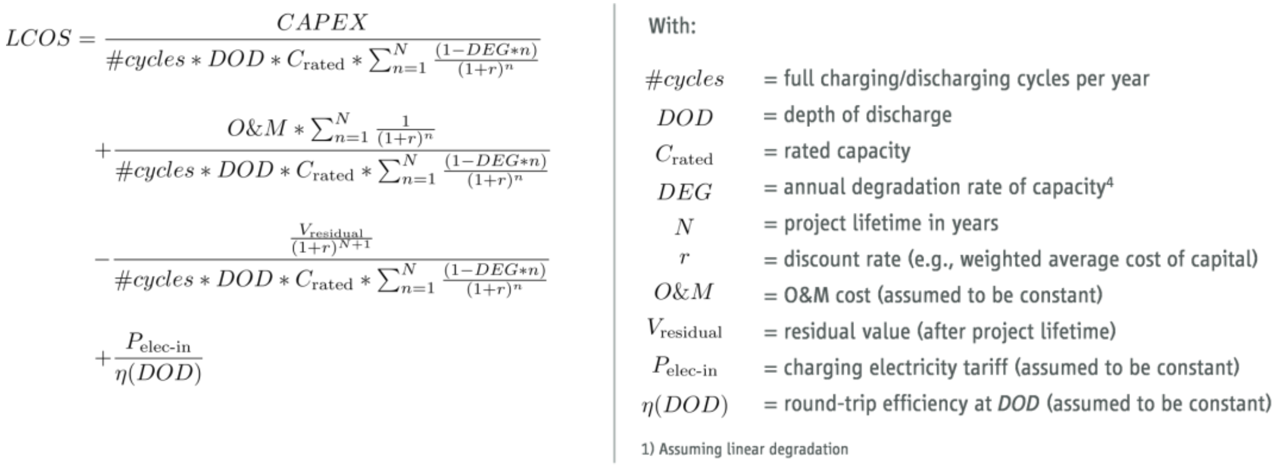
Table 3.4 Cost and Performance Targets for Electric Energy Storage Technologies

Range of baselines	**System capital cost by energy:** $805–$10,020/kWh **Levelized cost:** $0.01-$0.64/kWh/cycle **System efficiency:** 75%–92% **Cycle life:** 4,500–225,000 over life of plant **System capital cost by power:** $300–$4,600/kW
Near-term targets	**System capital cost by energy:** less than $250/kWh **Levelized cost:** less than $0.20/kWh/cycle **System efficiency:** more than 75% **Cycle life:** more than 4,000 cycles **System capital cost by power:** less than $1,750/kW
Long-term targets	**System capital cost by energy:** less than $150/kWh **Levelized cost:** less than $0.10/kWh/cycle **System efficiency:** more than 80% **Cycle life:** more than 5,000 cycles **System capital cost by power:** less than $1,250/kW

Figure 12. QTR targets for electric energy storage technologies

Among the metrics reported in the QTR, a crucial one is the Levelized Cost of Storage (LCOS), because it allows to compare different kind of energy storage technologies – similar to the Levelized Cost of Energy (LCOE) which is useful for power producing technologies. The LCOS is "the constant, thus levelized price per kWh at which the net present value of the Energy Storage System (ESS) project is zero" [34].

However, in our case, with respect to the formulation used in the QTR, we have adopted a different one, that is:



$$
\begin{aligned}
LCOS = & \frac{CAPEX}{\#cycles * DOD * C_{\text{rated}} * \sum_{n=1}^{N} \frac{(1-DEG*n)}{(1+r)^n}} \\
& + \frac{O\&M * \sum_{n=1}^{N} \frac{1}{(1+r)^n}}{\#cycles * DOD * C_{\text{rated}} * \sum_{n=1}^{N} \frac{(1-DEG*n)}{(1+r)^n}} \\
& - \frac{\frac{V_{\text{residual}}}{(1+r)^{N+1}}}{\#cycles * DOD * C_{\text{rated}} * \sum_{n=1}^{N} \frac{(1-DEG*n)}{(1+r)^n}} \\
& + \frac{P_{\text{elec-in}}}{\eta(DOD)}
\end{aligned}
$$

With:

$\#cycles$ = full charging/discharging cycles per year
DOD = depth of discharge
C_{rated} = rated capacity
DEG = annual degradation rate of capacity[4]
N = project lifetime in years
r = discount rate (e.g., weighted average cost of capital)
$O\&M$ = O&M cost (assumed to be constant)
V_{residual} = residual value (after project lifetime)
$P_{\text{elec-in}}$ = charging electricity tariff (assumed to be constant)
$\eta(DOD)$ = round-trip efficiency at *DOD* (assumed to be constant)

1) Assuming linear degradation

Figure 13. Levelized Cost of Storage (LCOS) formulation by Apricum [34]

where the first term represents the actual cost of the system (we considered the CAPEX as the summation of the cost of the stacks, the cost of balance of plant and the cost of the hydrogen storage, all values expressed in $), the second term stands for the operational and maintenance costs, the third one for the residual value of the system at its end-of-life and, lastly, the fourth one for the charging costs.

We have utilized this formulation of the LCOS with some assumptions:

- the *capital cost by power* has been estimated for 250 kW system in fuel cell mode
- the *duty cycle* has been assumed – as a starting point – to be daily, in particular 10 h charge, 8 h discharge and a total of 350 cycle/year
- the *depth of discharge* (DOD) has been kept constant to 100%
- the *annual degradation rate of capacity* (DEG) has been assumed to be 0%
- the *system round trip efficiency, capital cost* and *lifetime* used are those given in the system targets table below
- the *discount rate* (r) has been assumed to be 8%

- the *O&M* cost assumed to be constant and equal to 3.5% of capital cost [35]
- the third term of the formulation has been neglected, namely the residual value of the system at its end-of-life
- the *cost of the electricity* to charge the system ($P_{elec-in}$) has been assumed to be constant and equal to $ 0.04/kWh as a first approximation for future low-cost renewable energy costs
- the *installation costs* plus *soft costs* (such as permitting and project management fees) has been assumed to be 33% of the capital cost [6]
- a 10% degradation over the system lifetime has been utilized and accounted for via oversizing the stack and increasing system costs accordingly so that at the end of life the RTE specification is met; thus, the corresponding new power level is 278 kW
- the hydrogen storage cost has been accounted only for the amount of hydrogen needed for fuel cell operation, since the LCOS metric does not consider the excess of hydrogen produced as a revenue, not contemplating the possibility to sell it for other end-uses.

Despite an increasing number of studies on this topic, a common definition of LCOS is still missing: for instance, some studies neglect replacement and end-of-life cost, as well as capacity degradation and self-discharge [36].

Based on the information gathered, we decided to specify the metrics in cell/stack and system level. The common metrics between low and high temperature RFCs *at cell/stack level* are the following:

- *Stack Capital Cost ($/kW)*: is the cost of the stack per unit of power
- *Degradation Rate (%/1000h of operation hours)*: performance loss based on loss in RTE (Round Trip Efficiency) at fixed current density

whereas *at system level* all the metrics are in common, as follow:

- *System RTE (%)*: for a RFCs working with air; in the case of high temperature RFCs, a thermal energy input is included, too
- *Levelized Cost of Storage ($/kWh)*: based on the aforementioned assumptions
- *System Capital Cost by Power ($/kW)*: is the cost of the system per unit of power (it does not include the cost of hydrogen storage)

- *Lifetime/Durability (h)*: in technical targets tables the lifetime is also reported in number of cycles, based on the assumption of a daily duty cycle.

For high temperature RFCs the specific metric used at *stack level* is:

- *Area Specific Resistance (Ω cm^2)*: used instead of RTE to describe the cell's efficiency in order to avoid specifying the current density, since $RTE \sim \left(1 - \frac{i \cdot ASR}{V_{oc}}\right)^2$

whereas, for low temperature RFCs the specific metrics used at *stack level* are:

- *Electrical RTE (%)*: at 1A/cm^2 for electrolyzer operation and at 0.5 A/cm^2 for fuel cell operation
- *Total Cell PGM loading (mg/cm^2)*: is the quantity of catalyst needed per cm^2 of cell area.

For all these metrics two projections have been made, one for a near-term case, namely *2025 Targets,* and one for a long-term case, namely *Ultimate Targets.* Moreover – where available –a *current status* of the metrics has been included, too. It is important to notice that an effort was made so that all the targets for both near and long term were self-consistent; this is not verified for the current status since the values may have been taken from different systems.

As a methodological choice, various versions of these targets have been developed and sent out to a number of experts, within LBNL, NREL and worldwide, both in the research and in the industry fields, to be vetted. Thanks to the precious feedback received from experts, we have then accordingly confirmed, updated or modified these targets and the assumptions made.

Here below the technical targets developed have been organized in four tables, the first two for Low Temperature RFCs, at cell/stack and system level respectively, the following two for High Temperature RFCs, at cell/stack and system level respectively.

Low Temperature Reversible Fuel Cells - CELL / STACK					
Characteristics	**Units**	**a | 2019 Status**	**b | 2025 Targets**	**c | Ultimate Targets**	**Notes**
Roundtrip Electrical Efficiency (at 1 A/cm^2)	%	42	45	55	***a*** | S.-Y. Huang, et. al. J. Power Sources 198 (2012) 23
Stack Capital Cost	$/kW	980	540	310	Costs based on modeled 250 kW PEM stack cost @100, 1000 and 10,000 units per year (Avg. values in $/kW from cost studies by LBNL, Battelle, and SA Inc. – references provided in the Bibliography) with PGM loadings as given below for the Total cell PGM loading metric and 15% increase in costs to account for oxygen electrode corrosion mitigation
Durability / Degradation Rate	%/1000 hr	-	0.25	0.125	***a*** | There does not appear to be data in the literature supporting operation beyond several days ***b***, ***c*** | Based on end of life being 10% loss in performance over lifetime (e.g., 40,000 hr for 2025 target). Performance loss can be based on loss in RTE at fixed current densities. ***b***, ***c*** | With daily cycling between fuel cell and electrolyzer modes with a minimum of 40% time in electrolysis mode and 20% in fuel cell mode. Standby and/or transient operation could be included in the daily cycling
Total Cell PGM loading	mg/cm^2	1.5	1.0	0.5	***a*** | S.-Y. Huang, et. al. J. Power Sources 198 (2012) 23; Review article by Y. Wang, Renewable and Sustainable Energy Reviews 65 (2016) 961 provides a range of 0.5-8 mg/cm2 for RPEMFC

Table 1. Technical Targets at cell/stack level for Low Temperature Fuel Cells for electrical energy storage applications

Low Temperature Reversible Fuel Cells - SYSTEM

Characteristics	Units	*a* | 2019 Status	b | 2025 Targets	c | Ultimate Targets	Notes
Roundtrip System Efficiency (working with air)	%	-	40	50	***b***, ***c*** | IEA Technology Roadmap Hydrogen and Fuel Cells (2015), Table 15
Levelized Cost of Storage	$/kWh	-	0.40	0.25	***b***, ***c*** | These LCOS numbers are consistent with the other PEM system target numbers here for cost and performance. These are higher than the DOE's Quadrennial Technology Review (QTR) Electrical Energy Storage Targets which have near-term and long-term targets of $0.20/kWh and $0.10/kWh, respectively (cf. Figure 5)
System Capital Cost by Power	$/kW	-	1550	1210	Cost based on modeled 250 kW PEM system cost @100, 1000 and 10,000 units per year (Avg. values in $/kW from cost studies by LBNL, Battelle, and SA Inc.) with PGM loading as given below for the Total cell PGM loading metric, with 15% increase in costs to account for oxygen electrode corrosion mitigation and with no fuel processing.
Lifetime / Durability	hr [Cycles]	-	40,000 [1667]	80,000 [3333]	***b***, ***c*** | With daily cycling between fuel cell and electrolyzer operating modes. ***b***, ***c*** | Lifetime targets are consistent with FCTO MYRDD Plan stationary fuel cell durability targets (https://www.energy.gov/eere/fuelcells/doe-technical-targets-fuel-cell-systems-stationary-combined-heat-and-power; Table 2)

Table 2. Technical Targets at system level for Low Temperature Fuel Cells for electrical energy storage applications

High Temperature Reversible Fuel Cells - CELL / STACK

Characteristics	Units	*a* | 2019 Status	*b* | 2025 Targets	*c* | Ultimate Targets	Notes
Area Specific Resistance (ASR)	Ω cm^2	0.125	0.125	0.10	A linear response of i-V polarization curve over current density range (to include both FC and electrolyzer performance) is needed. As an example, for ASR of 0.10 Ω-cm2 the RTE is ~80% @1 A/cm^2 (with V$_{oc}$ ~1.0 V). ***a*** | Z. Gao, V. Zenou, D. Kennouche, L.D. Marks and S. Barnett, J. Mater. Chem. A 3 (2015) 9955-9964 (Conditions: 800 °C, 50 vol% H2-50 vol% H2O/air)
Stack Capital Cost	$/kW	500	330	300	***a***, ***b***, ***c*** | Costs based on modeled 250 kW SOFC stack cost @100, 1000 and 10,000 units per year (Avg. values in $/kW from cost studies by LBNL, Battelle, and SA Inc. – references provided in Bibliography)
Durability / Degradation Rate	%/1000 hr	< 1.5	0.25	0.125	***a*** | Single cell with daily cycles over 14,500 hr (Conditions: ≥0.25 A/cm^2 SOFC; ≥0.50 A/cm^2 SOEC); Versa Power Final Technical Report (https://www.osti.gov/servlets/purl/1058912) ***b***, ***c*** | Based on end of life being 10% loss in performance over lifetime (e.g., 40,000 hr for 2025 target). Performance loss can be based on ASR degradation rate or loss in RTE at fixed current densities. ***b***,***c*** | With daily cycling between fuel cell and electrolyzer modes with a minimum of 40% time in electrolysis mode and 20% in fuel cell mode. Standby and/or transient operation could be included in the daily cycling.

Table 3. Technical Targets at cell/stack level for High Temperature Fuel Cells for electrical energy storage applications

High Temperature Reversible Fuel Cells - SYSTEM					
Characteristics	**Units**	***a* | 2019 Status**	***b* | 2025 Targets**	***c* | Ultimate Targets**	**Notes**
Roundtrip System Efficiency (includes thermal energy input and working with air)	%	43	55	65	***a*** | FZJ Institute in Germany (https://fuelcellsworks.com/news/fuel-cell-system-breaks-efficiency-record/accessed; June 10, 2019) ***b*, *c*** | Modeled efficiency from following references: - A. Perna, et al. Designing and analyzing an electric energy storage system based on reversible solid oxide cells. Energy Conversion and Management, 159 (2018) 381-395 - S. Santhanam, et al. Theoretical and experimental study of Reversible Solid Oxide Cell (r-SOC) systems for energy storage, Energy 141 (2017) 202-214
Levelized Cost of Storage	$/kWh	-	0.25	0.15	***b*, *c*** | These LCOS numbers which are consistent with the other solid-oxide system target numbers here for cost and performance. These are relatively consistent with DOE's Quadrennial Technology Review (QTR) Electrical Energy Storage Targets which have near-term and long-term targets of $0.20/kWh and $0.10/kWh, respectively (cf. Figure 5 above)
System Capital Cost by Power	$/kW	1300	1000	900	Cost based on modeled 250 kW SOFC system cost @100, 1000 and 10,000 units per year (Avg. values in $/kW from LBNL, Battelle, and SA Inc. cost analysis) with fuel processing system removed
Lifetime / Durability	hr [Cycles]	10,000 [-]	40,000 [1667]	80,000 [3333]	***a*** | https://www.green-industrial-hydrogen.com/news-detail/grinhy-project-ends-with-submission-of-final-report ***b*, *c*** | With daily cycling between modes ***b*, *c*** | Lifetime targets are consistent with FCTO MYRDD Plan stationary fuel cell durability targets (https://www.energy.gov/eere/fuelcells/doe-technical-targets-fuel-cell-systems-stationary-combined-heat-and-power; Table 2)

Table 4. Technical Targets at system level for High Temperature Fuel Cells for electrical energy storage applications

In the Technical Targets tables, focusing our attention on the LCOS metric – useful to compare different storage technologies – it is possible to observe that R-SOFCs ($0.25/kWh for *2025 Targets* and $0.15/kWh for *Ultimate Targets*) appear more promising than R-PEM fuel cells ($0.40/kWh for *2025 Targets* and $0.25/kWh for *Ultimate Targets*). This values difference could be due to the major interest that R-SOFC has seen compared to R-PEM, confirmed by the more material present in literature, too, for this technology. As a reference, about electric energy storage technologies, the QTR's LCOS values are $0.20/kWh for near term and $0.10/kWh for long term targets.

3.2. R&D Needs at Stack and System Level

For all these RFCs technologies, a limited number of system tests have been carried out, since they can be considered a "new" energy storage technology, while – on the other hand – discrete RFCs systems have attracted more interest and thus more demonstration systems have been deployed. Therefore, a unitized system, since it will likely never match performance and durability of a discrete system (because a single stack has to be optimized to operate in two different modes with opposite needs), has to be superior in other ways (for instance, capital cost).

Because of the differences between high and low temperature RFCs, it is more profitable to carry out this analysis separately.

3.2.1. R-PEM

For PEM technology the main problem to overcome is to optimize the structure and the materials utilized to create an effective and durable bifunctional oxygen electrode. This electrode presents some issues related to the intrinsic properties of the materials utilized, to the corrosion problem in electrolyzer mode and to the slow kinetic of the reactions taking place here.

The first issue is due to the different water management needed, specifically in fuel cell mode the cathode material (anode in electrolyzer mode) – where the ORR takes place – has to have hydrophobic properties to remove the produced water from the point of reaction, avoiding in this way the flooding of the electrode itself, while during electrolyzer operation the supplied water needs to easily reach the reaction point – where the OER takes place – thus the electrode material needs to have, on the contrary, hydrophilic properties.

The second issue is a corrosion of the carbon-based materials composing both the oxygen electrode as well as the bipolar plates, due to the high voltage applied to the cell during electrolyzer mode; for this reason the typical porous carbon materials used for PEM fuel cell are not suitable to be utilized in a dual operation device. Nowadays, the state-of-the-art material able to accomplish a proper water management and to sustain the corrosive environment is a woven metal cloth, usually made out of titanium; however the durability of this material under cycling condition still needs to be research, as no information on durability beyond several days is reported in the literature.

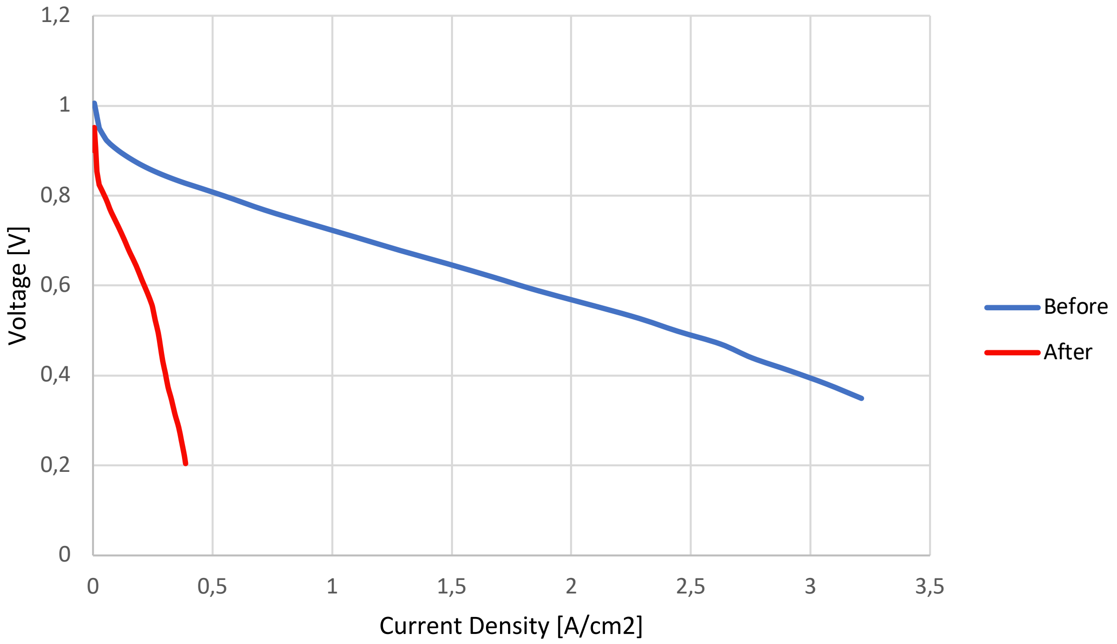


Figure 14. I–V curves of unit cell with carbon-based bipolar plate in fuel cell mode at 75 ◦ C (before and after operation of the unit cell in electrolyzer mode at 2.0 V for 1 h, 4 mg cm^{-2} of Pt black catalyst) [37]

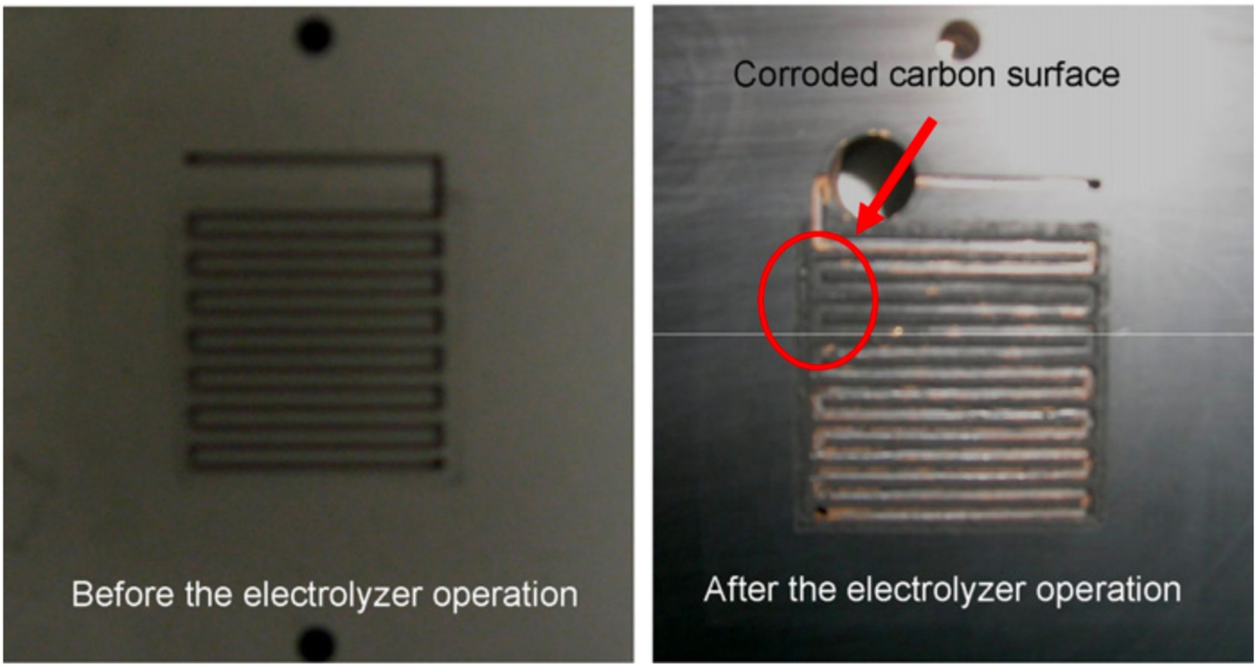


Figure 15. Corrosion on the surface of carbon-based bipolar plate before and after operation of water electrolysis reaction at 2.0 V for 1 h [37]

The third issue is related to the kinetic of the reactions taking place at the positive electrode, because platinum – the preferred catalyst for the ORR (operation as fuel cell) – is not fitted to catalyze the OER; a good kinetic in both reaction has been achieved using a mixture of platinum and iridium (or iridium oxide). However, since both these catalyst materials are noble metals and therefore very expensive, further research is needed to reduce the total PGM loading in order to lower the overall cost of the stack. A further improvement would be achieved with the introduction of a proper catalyst support layer – structure in which the catalyst is dispersed – able to stand the characteristic voltage difference in the two modes of operation.

Another metric – on which further improvement is needed – is the round-trip-efficiency; in fact, compared to PEM fuel cell and electrolyzer range of efficiency of 40-50%, the RFC efficiency varies in the range of 30-40%, but at lower current density (for instance, at 1A/cm^2 for electrolyzer operation and at 0.5 A/cm^2 for fuel cell operation) and using usually PGM loading one order of magnitude higher. A further factor affecting the RTE is the fuel cell mode efficiency, meaning that the increased thickness of the membrane due to the electrolyzer requirements (>100 µm) lowers the performance in fuel cell mode compared to electrolyzer one.

Over last years, reversible PEM fuel cells have witnessed an increased interest due to their peculiar characteristics of fast dynamic response – thanks to the low temperature range of operation – that enables, for instance, great start-up performance.

3.2.2. R-SOFC

Reversible SOFCs present overall less problems, starting from the fact that typically used materials for fuel cells (e.g. Yttria-Stabilized-Zirconia electrolyte) are suitable to be adopted for reversible system with just minor modifications [29]. On the other hand, the high temperature of operation (i.e. 700-800 °C) affects the material durability, making it one of the greater challenges to overcome for this technology.

The elevated temperature range has, however, a positive impact on the kinetic of reaction and solve one of the major problems of low temperature RFCs, allowing nickel, that already guarantees a fast kinetic, to be used as a catalyst, thus avoiding the need for an expensive material.

Another issue that high temperature allows to solve – with respect to the reversible PEM – is water management, since water being in steam phase excludes the electrode flooding.

Regarding the main problem of durability for R-SOFC – especially caused by electrolyzer mode – recent studies state that under cycling condition fuel cell mode is beneficial to the degradation of the electrode, thus increasing its durability [20], [38].
Another option that has been investigated is to utilize a *proton conductor electrolyte*, that would lower the operation temperature to 500-700 °C, allowing the use of less resistant materials (e.g. bipolar plates) [39].

4. COST ANALYSIS

To better assess the opportunity and feasibility of Unitized Reversible Fuel Cells, compared to Discrete systems and to other energy storage technologies, we developed a Cost Study to estimate the actual costs of the system and its possible cost reduction trend due to the increase in manufacturing volume. Since the Project is still ongoing, the cost analysis is going to be limited to the R-PEM technology alone, while the cost analysis on R-SOFC is still under development.

The cost estimation for the stack and for the balance of plant components – due to the Project timing – has been largely drawn from existing cost studies on PEM, both fuel cells and electrolyzer, and based on some key assumptions. The main cost studies that we have taken as references are: *A Total Cost of Ownership Model for Low Temperature PEM Fuel Cells in Combined Heat and Power and Backup Power* by the Lawrence Berkeley National Laboratories [6]; *Manufacturing Cost Analysis for Proton Exchange Membrane Water Electrolyzers* by the National Renewable Energy Laboratory [14]. It is important to highlight that these references report also manufacturing cost (that do not include any profit margins).

4.1. Framework

A sequence of steps has been followed to build the framework for our cost study. We started by developing a series of functional specification for our system with a few key simplifying assumptions. Hence, we worked out a basic system design with main balance of plant components and with evaluation of stack parameters. From here we moved to a simplified cost estimation of the stack and balance of plant components, based mainly – as said above – on existing cost studies.

Afterwards we focused our attention on identifying key indicators on which make the cost study parametric. The logic behind these choices was to determine the parameters that have the more significant impacts on the stack and the BOP costs. We identified, from our analysis, these metrics: *System Size (FC mode)*; *Duty Cycle*; *Current Density* (in both modes of operation); *Platinum Group Metal* (PGM) *Loading*. All the above has been done with the objective of enabling an easier comparison with other energy storage technologies, utilizing parameters such the levelized cost of storage (LCOS).

4.1.1. Functional Specifications

The starting point in the development of functional specifications (such as power, current density, voltage, efficiency, active area of the cell, etc.) for our system was to impose the *power in fuel cell mode* equal to 250 kW, keeping the power in electrolyzer mode as a variable. Moreover, we increased this power considering a 10% degradation over its lifetime to assure that the power output from the system would be met still at its end-of-life: as a result of this, the corresponding power became 278 kW.

The design choice that we made afterwards was to assume as *area of cell* for a RFC the one of a fuel cell – typically smaller than that of an electrolyzer – basing our assumption on the fact that fuel cells are more critical components compared to the more flexible electrolyzer, prioritizing in this way the less performant element. About the values of the area of the cell, we took the following [6]:

Area of the Reversible Fuel Cell			
Characteristics	**Units**	**Fuel Cell mode**	**Electrolyzer mode**
Total Plate Area	cm^2	363	363
Catalyst Coated Membrane (CCM)	cm^2	306	306
Single Cell Active Area	cm^2	285	285
Gross Cell Inactive Area	%	21	21

Table 5. Assumptions for the Area of the RFCs

However, before that, some design choices and assumptions had to be made. We explored two design choice alternatives for the cell of a unitized system able to work in both modes:

- one option was to make the single stack work at the same current density (A/cm^2) in both operation modes, even though usually the current density of an electrolyzer is higher with respect to the one of a fuel cell. Therefore, with this option the operation point of the device (namely the nominal voltage and nominal current density) would be optimized for just one mode and – since the performance of a fuel cell is usually more critical than the one of an electrolyzer in terms of range of operation and efficiency – the mode to optimize would be the fuel cell one
- the other option was to utilize two different current densities for the two operating modes; this way both fuel cell and electrolyzer modes would work at a more suitable current density, increasing the respective efficiencies and, overall, the round-trip efficiency.

We chose the second *configuration* because it allows better performance in both modes and – since we are interested in using this technology to store potentially large amounts of energy in the form of hydrogen to support electric grids – in this perspective it was a better choice. Various experts' inputs had confirmed this choice: for instance, Giner suggested that EC mode should work at a current density approximately three times the one of FC mode.

At this point – once the power in FC mode, the area of the cell and the operation configuration (i.e. the cell works at different current density in the two modes) were defined – we set the *point of operation* (i.e. the nominal voltage and nominal current density) in both modes, based on typical polarization curves of fuel cells and electrolyzer respectively. With all this information, then, we were able to calculate the *power density* and the *power of a single cell* for both modes, as follows:

Electrical Parameters			
Characteristics	**Units**	**Fuel Cell mode**	**Electrolyzer mode**
Cell Current	A	71	214
Current Density	A/cm^2	0.25	0.75
Reference Voltage	V (cell voltage)	0.67	1.69
Power Density	W/cm^2	0.17	1.27
Single Cell Power	W	48	361

Table 6. Assumptions for the Electrical Parameters of the RFCs

Using the electrical parameters above and, in particular, by dividing the *power of the fuel cell mode* by its *single cell power,* we were able to obtain the overall number of cells needed to reach the required power level (*cells per system)* and the value of the *power of the electrolyzer mode,* calculated as the product between the number of cells per system and the single cell power for the EC mode. Lastly, we decided to impose the number of *stacks per system* to get the number of cells composing a stack (*cells per stack*). Below are reported the functional specifications as a whole:

Functional Specifications - STACK			
Characteristics	**Units**	**Fuel Cell mode**	**Electrolyzer mode**
Stack Power	kW	278	2110
Total Plate Area	cm^2	363	363
Catalyst Coated Membrane (CCM)	cm^2	306	306
Single Cell Active Area	cm^2	285	285
Gross Cell Inactive Area	%	21	21
Cell Current	A	71	214
Current Density	A/cm^2	0.25	0.75
Reference Voltage	V (cell voltage)	0.67	1.69
Power Density	W/cm^2	0.17	1.27
Single Cell Power	W	48	361
Cells per Stack	cells	234	234
Cells per System	cells	5841	5841
Stacks per System	stacks	25	25
Efficiency	%	54	73
Round Trip Efficiency (RTE)	%	39	

Table 7. Stack Functional Specifications

4.1.2. System Design and Balance of Plant (BOP) Components

As we are dealing with a new technology and there are few demonstration systems, in order to understand which main elements of the BOP are needed in a unitized reversible fuel cell system, we have developed a simplified system design. A useful reference in this sense has been the pilot-scale URFC system that has been installed in the National Institute of Advanced Industrial Science and Technology (AIST), in Japan, composed by a PEM stack and by some major BOP elements [40]. Moreover, we looked separately to PEM-fuel cells and PEM-electrolyzer systems designs to look for possible simplifications of the balance of plant with respect to discrete systems.

In our system design and, successively, in our cost study we have not included the *fuel processing subsystem* – which typically represents one of the major fractions of the overall investment cost for PEM-fuel cell system – because we assumed that in our case the fuel (hydrogen) needed in fuel cell mode had been previously produced and stored by electrolyzer mode. Keeping in mind one of the main objectives of this cost study, that is an easy comparison with other energy storage technologies, we considered as a cost only the portion of the hydrogen storage needed for fuel cell operation. We did not account for the excess of hydrogen produced, as the LCOS metric formulation we are using (analyzed in the Technical Targets chapter) does not contemplate the possibility of utilizing the hydrogen for other end-uses than the production of energy.

A few more assumptions on the design were made:

- the system works at ambient pressure (both modes of operation)
- the hydrogen produced is stored at 100 bar (electrolyzer mode)
- the system is fed with air and pure hydrogen in fuel cell mode, while city water is fed for the operation in electrolyzer mode
- the oxygen produced from water electrolysis is vented to the atmosphere (electrolyzer mode)
- exhaust cathodic air is used to pre-humidify inlet air (fuel cell mode)
- partial recirculation of anodic exhausts to exploit the residual hydrogen (fuel cell mode)

We grouped together the balance of plant components on the basis of the function they absolve in the system. Below are reported the subsystems and their functions:

- *Fuel subsystem*: takes care of reducing the fuel pressure from the storage pressure value to the one required by the device, of preheating the inlet fuel and of recirculating anode exhausts (in fuel cell mode)
- *Water subsystem*: takes care of preheating the inlet water to the required temperature and of pumping it to the cell (in electrolyzer mode)
- *Hydrogen Processing subsystem*: takes care of bearing the produced hydrogen to the storage conditions, hence it is composed by a hydrogen/water separator, a dryer and a hydrogen compressor (in electrolyzer mode)
- *Air subsystem*: takes care of bearing the inlet air to the required conditions for the fuel cell operation, hence it is composed by air filters, a humidifier and air blowers
- *Coolant subsystem*: takes care of the thermal management of the device (fuel cell mode); it is not represented in the system schematic

- *Power System subsystem*: takes care of transforming AC current to DC to feed the electrolyzer and of transforming the electricity produced in fuel cell mode from DC to AC (hence, it works in both fuel cell and electrolyzer modes)
- *Controls/Meters subsystem*: monitors the system parameters (e.g. pressure, temperature, flow rates, etc.) in both modes of operation
- *Miscellaneous Components subsystem*: it includes the tubing and the wiring needed as well as hazard prevention and labor cost.

The system design we developed is schematically represented below, highlighting the main components of the BOP:

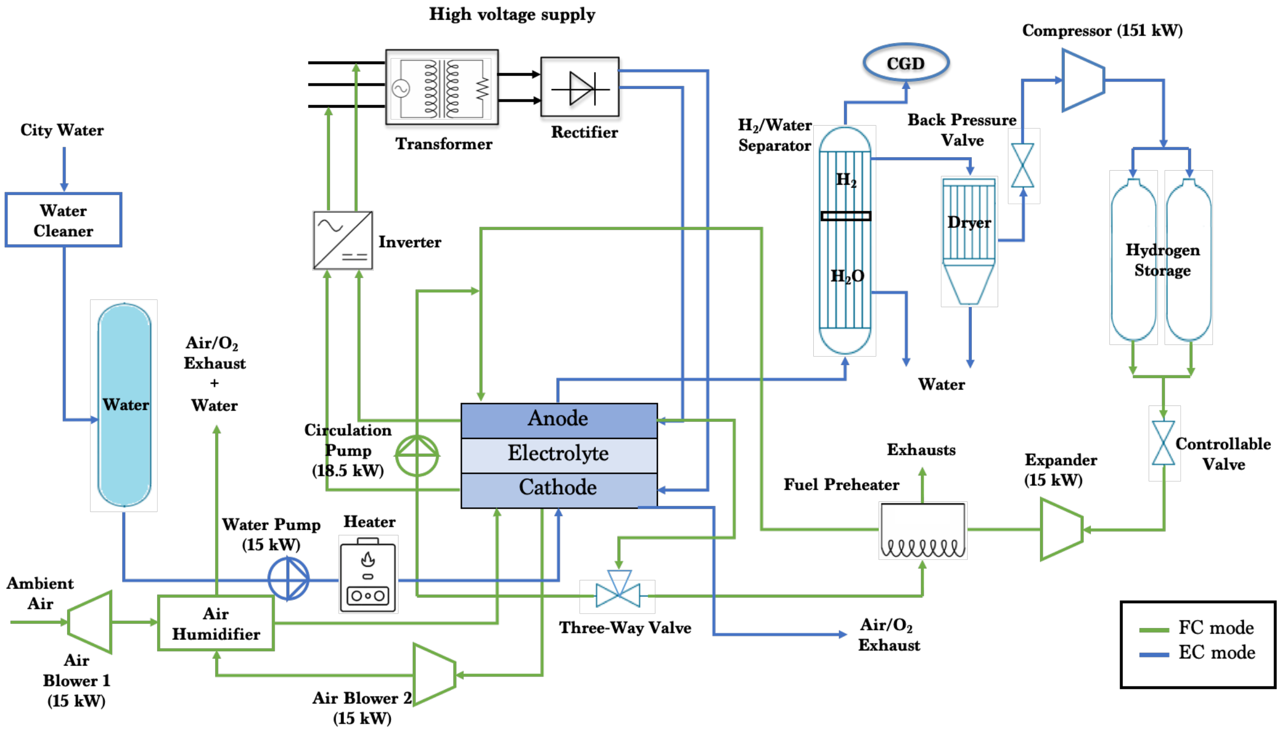


Figure 16. System Schematic developed for the cost analysis; CGD is the Combustible Gas Detector

The operation in fuel cell mode is articulated in the following way: first of all, the two reactants flow rates, namely ambient air and hydrogen, are taken to the conditions required by the operation. The air flow rate is taken into *the air humidifier* by an *air blower*, where it gets in contact with the cathodic exhausts (i.e. air and water) and it is humidified, since the PEM membrane has a good ionic conductivity only if it is hydrated; the exhausts (cathodic) are then vented to the

environment. Now the air – preheated and humidified – is ready to enter the cathode, where the ORR takes place.
Simultaneously, the hydrogen coming from the storage flows across the *expander*, where its pressure is reduced from 100 bar to ambient pressure; then, it is preheated to the required temperature within the *fuel preheater* by exploiting the anodic exhausts; at this point it is ready to enter the anode, where the HOR takes place. A fraction of the exhausts from the anode is mixed – through a *circulation pump* – with the inlet fresh fuel. The produced current goes through the *inverter*, where it is transformed from DC to AC, then it is sent to the grid.

In electrolyzer mode, on the other hand, the electricity coming from the grid goes first through the *transformer* (that reduces its voltage from high to low), then it goes through the *rectifier* (that converts the current from AC to DC) and finally it feeds the system. At the same time water from the mains is pumped (by the *water pump*) into the anode (cathode in FC mode), where the OER takes place, after being preheated by the *heater*. At the cathode (anode FC mode) the hydrogen – produced by the HER – goes into the *hydrogen-water separator*; from here water is removed, while the hydrogen goes through the *dryer*, which has the function of removing the residual water from the flow rate. The now pure hydrogen is then compressed to 100 bar (in the *compressor*) and is sent to the *hydrogen storage*.

Once we had defined the system schematic, we moved to the calculations of the flow rates composing the RFC operation. These calculations are meant to be "medium fidelity" – as for the BOP – but representative of the actual operation of our system and useful, anyway, to evaluate the costs of some components of the balance of plant. Further on they were used to develop the parametrization of the cost study. Below are reported the most significant flow rates for both modes of operation of the device.

Operation Specifications of the System			
Characteristics	**Units**	**Fuel Cell mode**	**Electrolyzer mode**
Hydrogen Volume Flow Rate	Nm^3/h	209	365
Water consumption	l/Nm^3, H_2	-	1.15
Power consumption	kWh/Nm^3, H_2	-	6.31

Table 8. Most significant flow rates values of the system

To cross check these values obtained from our calculations, we looked for specifications of commercially available systems for PEM-fuel cells and PEM-electrolyzer separately. We took two Hydrogenics systems, one for MW-scale PEM fuel cells (1 MW) and one for MW-scale PEM electrolyzer (1.5 MW): due to the different power levels, in both cases we had to match them with our 250 kW (fuel cell mode) system, by proportionally scaling the hydrogen volume flow rate, the water consumption and the power consumption [41], [42].

Commercial Systems Specifications			
Characteristics	**Units**	**PEM - Fuel Cell 250 kW [1 MW]**	**PEM - Electrolyzer 2.11 MW [1.5 MW]**
Hydrogen Volume Flow Rate	Nm^3/h	188 [750]	300 [422]
Water consumption	l/Nm^3, H_2	-	< 1.4 [2]
Power consumption	kWh/Nm^3, H_2	-	5-5.4 [7-8]

Table 9. System specifications for MW-scale commercial systems by Hydrogenics [41], [42]. *Real case values are reported in square brackets, while the scaled values to our system size are presented out of the parenthesis*

4.2. Cost Study

The framework just analyzed represents the base for the development of our cost study. This has been carried out with the objective of providing researchers and investors with an additional tool to assist them in future R&D and deployment of this promising technology (Unitized Reversible Fuel Cell).

To make the point of the situation, we set the framework for the cost analysis, meaning that: we defined the functional specifications, developed the system design and identified the main components of the balance of plant by reviewing and analyzing different schematics for both fuel cells and electrolyzer. Now we are going to analyze individually the costs for the stack and those for the balance of plant.

4.2.1. Stack Cost

To evaluate the cost of the stack, or better of its single components, we utilized the reference *A Total Cost of Ownership Model for Low Temperature PEM Fuel Cells in Combined Heat and Power and Backup Power Applications*, by Wei et al. [6]. In order to be conservative, we decided to approximate the cost of the stack with the one of fuel cells, as it is typically higher with respect to the electrolyzer one.

Moreover, the chosen reference reports in the appendix a series of detailed tables containing the specific cost (e.g. $\$/m^2$, \$/part, etc.) for all the components that form the stack, at different power levels of the system (for our study we utilized the 250 kW case).

Starting from this data, which has given us a lot of flexibility allowing us to better adjust the costs to our case, we evaluated the overall cost of the stack.

Regarding the materials selected, we made the assumption of using the same ones as for fuel cells, but with some forethoughts. Specifically, we updated the PGM loading to the new catalyst composition for a unitized reversible fuel cell (i.e. a mixture of platinum and iridium instead of pure platinum) and the thickness of the membrane, since a fuel cell membrane is too thin to work properly in electrolyzer mode; as a result, the membrane cost was updated, too. We assumed a total PGM loading of 1.5 mg/cm^2, in particular the anode (FC mode) – whose HOR and HER are well catalyzed by platinum alone – has a loading of 0.5 mg/cm^2, while the cathode (FC mode) – whose ORR is well catalyzed by platinum, but OER needs another catalyst (namely iridium or its oxide) – has a loading of 0.1 mg/cm^2 for platinum and a loading of 0.9 mg/cm^2 for iridium [18], [24]–[27]. To account for the variability of these noble metals' prices, we decided to take an average value over the last three years; the two graphs below show the trend of their prices:

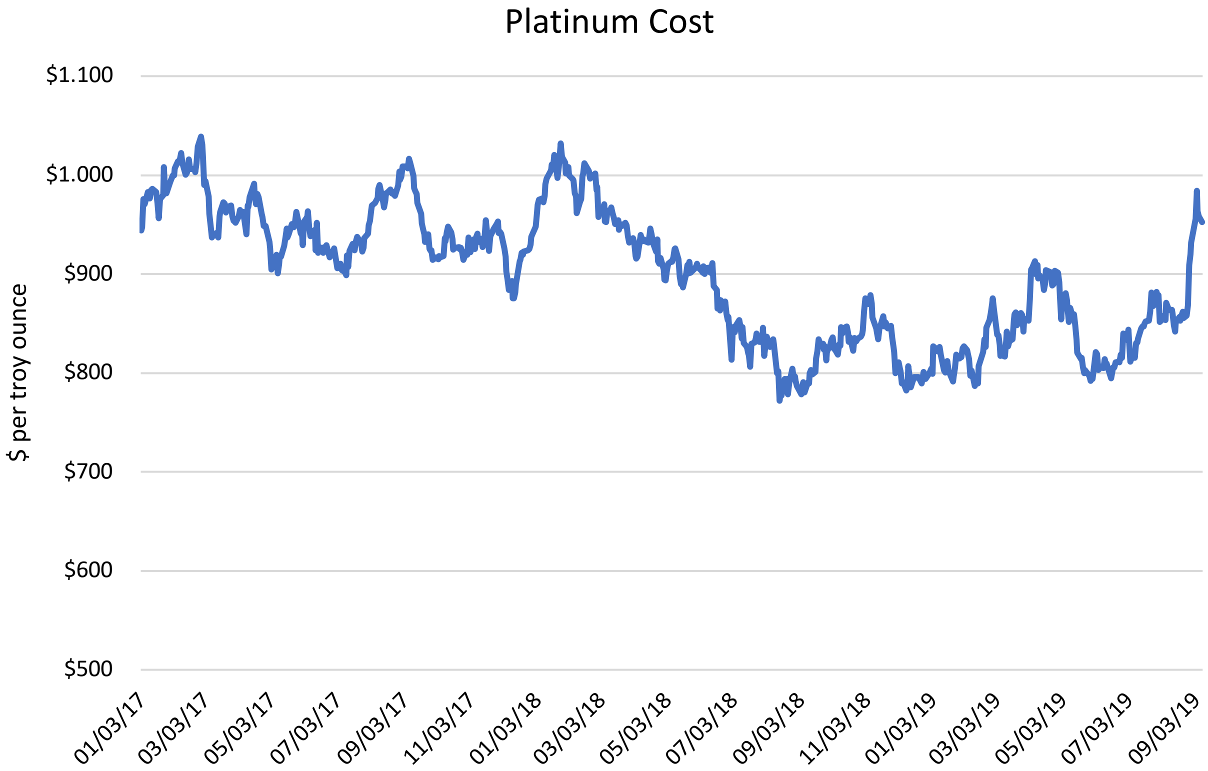


Figure 17. Platinum price per troy ounce trend (2017-2019)

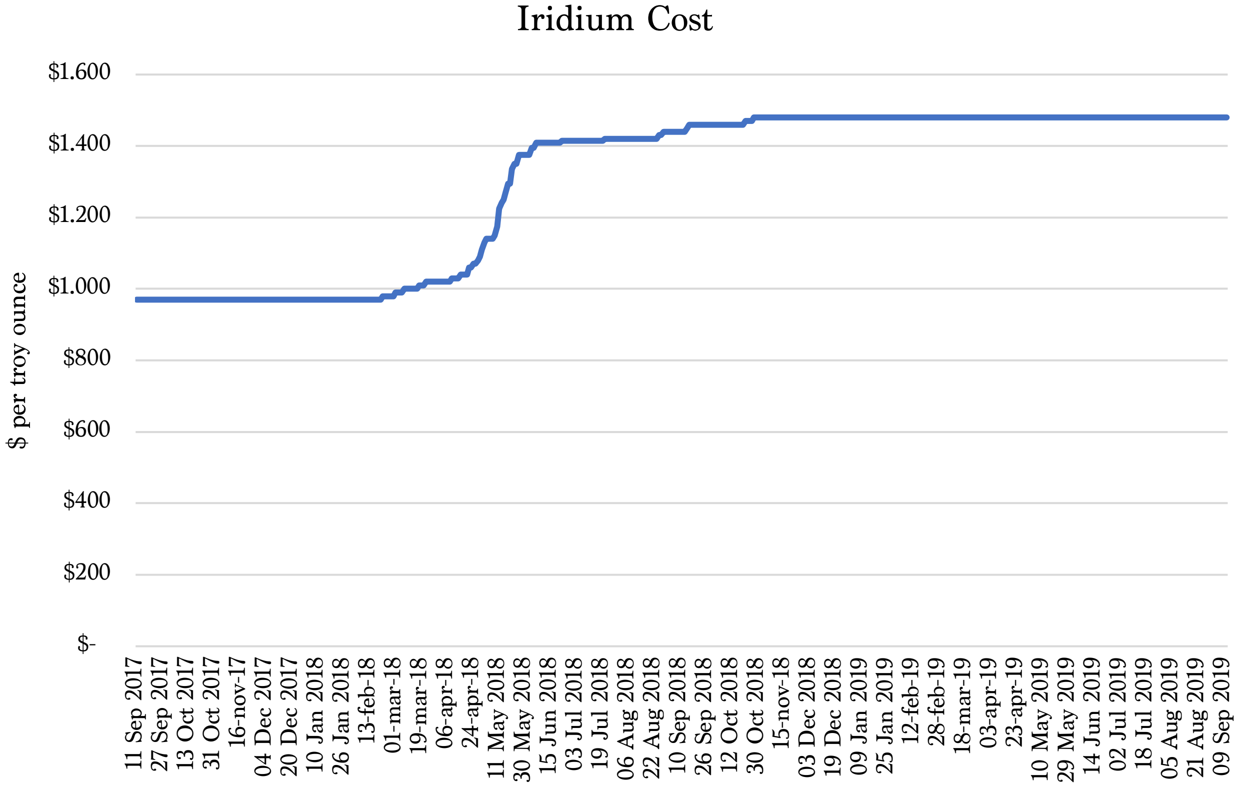


Figure 18. Iridium price per troy ounce trend (2017-2019)

For platinum the price average value is \$898/tr. oz, while for iridium it is \$1303/tr. oz. Regarding the membrane we assumed Nafion 117 – thick membrane typically used for electrolyzer – that is very expensive (\$500/m^2, [14]) with respect to a fuel cell membrane (~\$300/m^2, [6]), therefore here too, we decided to be conservative by assuming the higher cost between the two; moreover, we considered the value of the membrane to be constant (no cost reduction with increasing production volume).

This is how we changed the materials that represent the bigger fraction of the total cost of the stack, in order to get closer to the real cost of a unitized system, keeping in mind that our study has the objective of giving a realistic, though obviously not perfect, cost estimation for a URFC system.

Therefore, because of the lack of cost studies on R-PEM, we assumed to utilize PEM fuel cells materials for the different components, even though they are not suitable to stand the corrosive environment of the oxygen electrode:

- *Catalyst Coated Membrane*: composed by the Nafion membrane and the catalysts (platinum and iridium, descripted above)
- *Gas Diffusion Layer*: made out of carbon fiber paper
- *MEA Frame/Seal*: made out of Polyethylene naphthalate (PEN)
- *Plate*: made out of stainless steel (SS304) with a chromium nitride (CrN) coating.

Applying these considerations, the cost of the stack as a function of the annual production rate is:

Stack Cost per Components					
Production units	**100**	**1000**	**10000**	**50000**	**Units**
Catalyst Coated Membrane (CCM)	770	647	548	488	$/kW
Gas Diffusion Layer (GDL)	77	46	25	12	$/kW
MEA Frame/Seal	49	45	44	44	$/kW
Plate	117	58	50	49	$/kW
Stack Assembly Process	10	2	1	1	$/kW
TOTAL	1022	797	668	594	$/kW

Table 10. Costs per unit of power for the stack components, as a function of the annual production rate

The same costs are shown below reported – more intuitively - in a graphical representation:

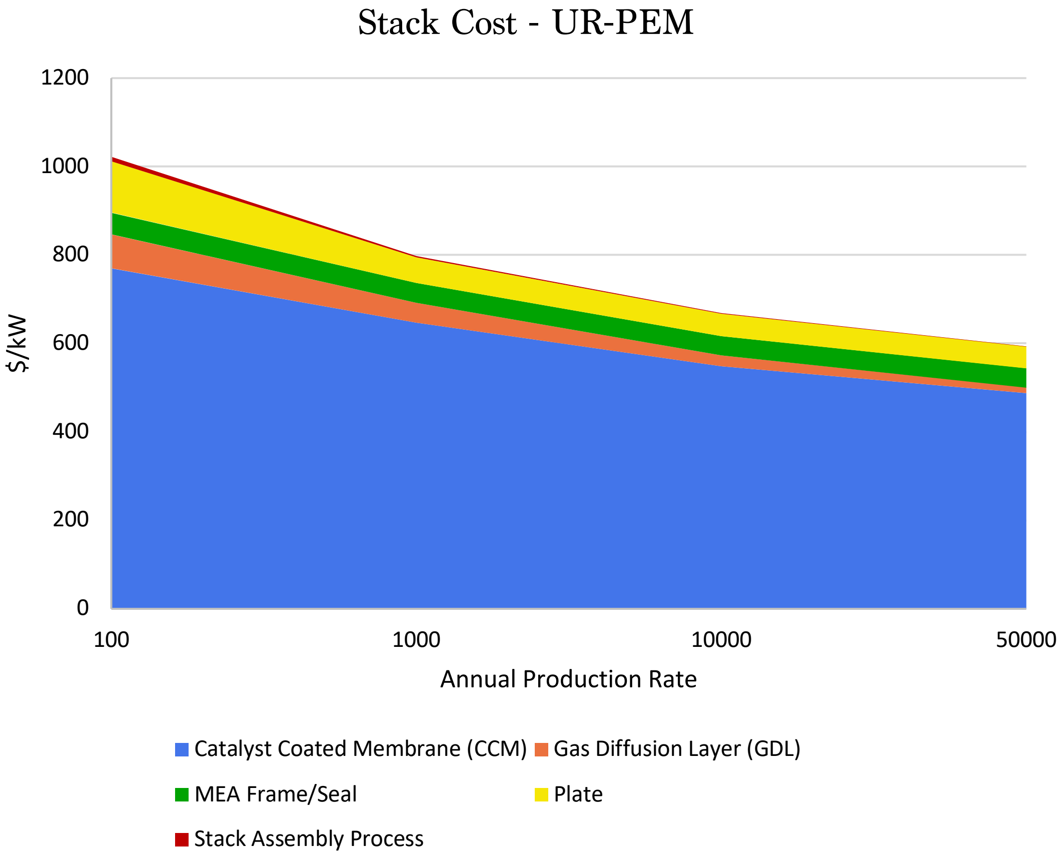


Figure 19. Costs breakdown of the stack of UR-PEM as a function of the annual production rate for a 250 kW (FC mode) system

As shown in the graph, the stack cost goes down with increasing production volume, but it remains dominated by the cost of the catalyst coated membrane; this is due to the high price of both the catalysts and the membrane.

4.2.2. Balance of Plant (BOP) Cost

For BOP components, too, the aim was to show the decrease in costs with an increasing production volume; the costs of components taken from fuel cells systems (e.g. air humidifier, coolant system, etc.) were already given in function of the production volume [6]; for elements like the inverter, that have seen a huge production increase thanks to the diffusion of solar photovoltaic systems (causing a significant cost reduction trend), we updated the costs to take into account this phenomenon by searching for prices of commercially available items.

On the other hand, the cost of components taken from electrolyzer systems (e.g. hydrogen-water separator, dryer, rectifier, etc.) were referred to a single unit [14]; therefore, to account them for different manufacturing volumes, we used the rule of the thumb of 20% decrease in costs for one order of magnitude increase in the production volume. For some components that were not explicitly costed in Mayyas' analysis [14] we searched for commercial available items' costs, applying to them the same rule of the thumb of 20% decrease.

The results obtained combining all these costs are shown in the table below:

BoP Cost per Subsystems					
Production units	**100**	**1000**	**10000**	**50000**	**Units**
Fuel Subsystem	57	45	36	29	$/kW
Water Subsystem	4	4	3	2	$/kW
Hydrogen Processing Subsystem	745	596	477	381	$/kW
Air Subsystem	37	29	24	21	$/kW
Coolant Subsystem	49	42	36	34	$/kW
Power System Subsystem	888	711	569	459	$/kW
Controls/Meters Subsystem	40	33	27	23	$/kW
Misc. Components Subsystem	124	82	52	49	$/kW
TOTAL	1944	1541	1224	1000	$/kW

Table 11. Costs per unit of power for the Balance of Plant subsystems, as a function of the annual production rate; Fuel Subsystem, Air Subsystem and Coolant Subsystem are fore FC mode; Water Subsystem, Hydrogen Processing Subsystem are for EC mode; Power System Subsystem and Controls/Meters Subsystem are for both modes

The following is a representation of the same table as a graph:

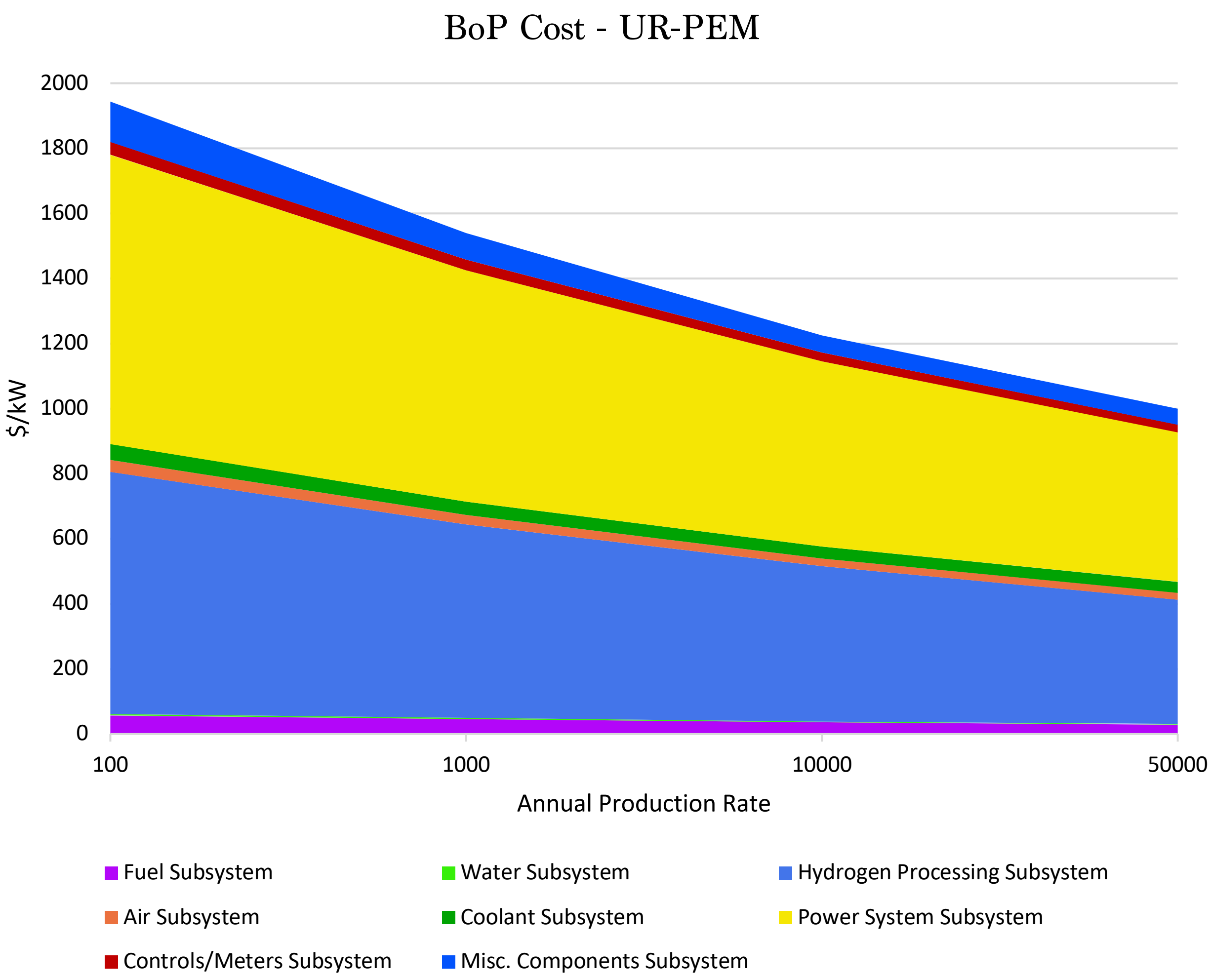


Figure 20. Costs breakdown of the BOP subsystems for UR-PEM as a function of the annual production rate for a 250 kW (FC mode) system

As can be clearly seen, the overall cost of the balance of plant is dominated by two subsystems, namely the subsystems of the Hydrogen Processing and, even more so, of the Power electronics. The more expensive component in the Hydrogen Processing subsystem is linked to the hydrogen compressor, which needs to increase the pressure of the produced hydrogen from the reference pressure of the stack to the storage pressure (i.e. 100 bar). While, among the power electronics, the bigger fraction of the cost is due to the rectifier, which feeds the stack during electrolyzer operation and represents the most expensive component of the entire balance of plant.

We have now all the elements to obtain the overall cost of a Unitized Reversible Fuel Cell system (stack plus BOP), reported in a graphical representation below:

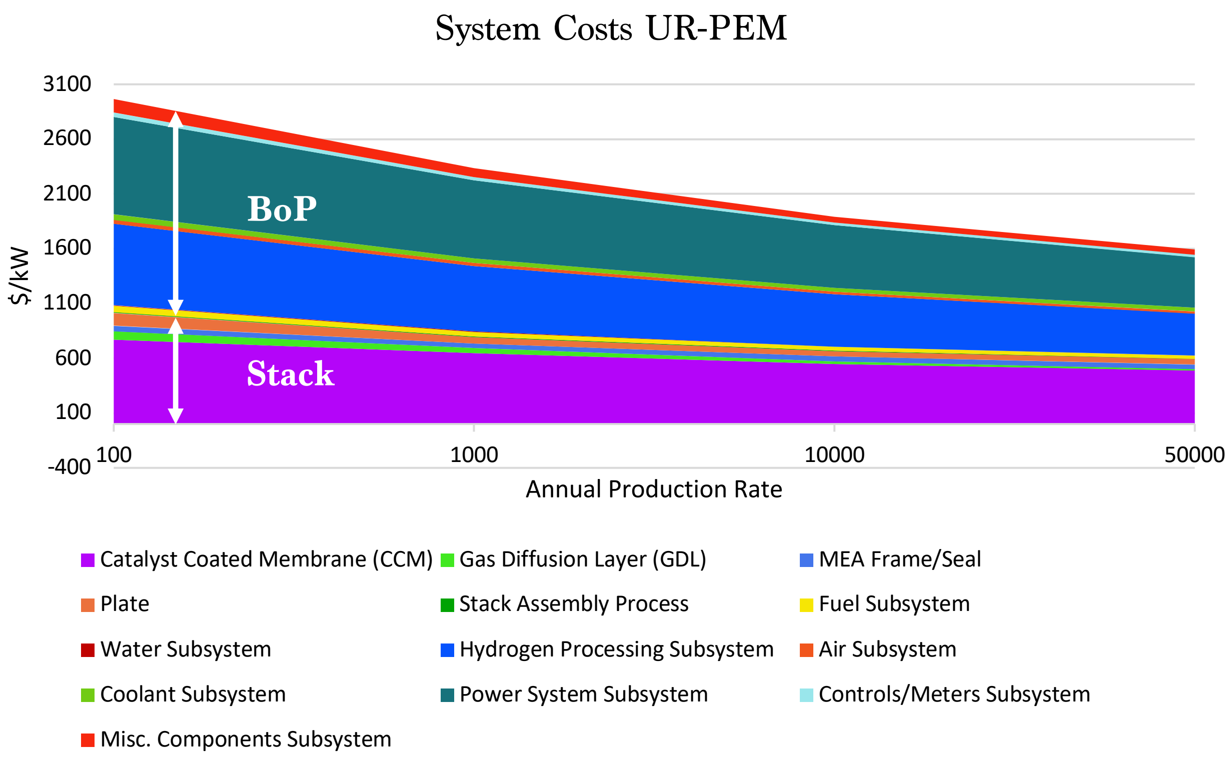


Figure 21. Costs breakdown of the UR-PEM system as a function of the annual production rate for a 250 kW (FC mode) system

This graph allows us to make some observations: at 100 units/year the stack cost is about 34% of the overall costs of the system, while the remaining 66% is to be imputed to the BOP. With the increase in production rate there is a progressive cost reduction, which is not homogeneous between stack and BOP, but mainly related to the cost reduction of the latter so at 10,000 units/year the stack represents 35% of the overall cost, while the BOP represents 65%. The histogram below can help to better understand this trend.

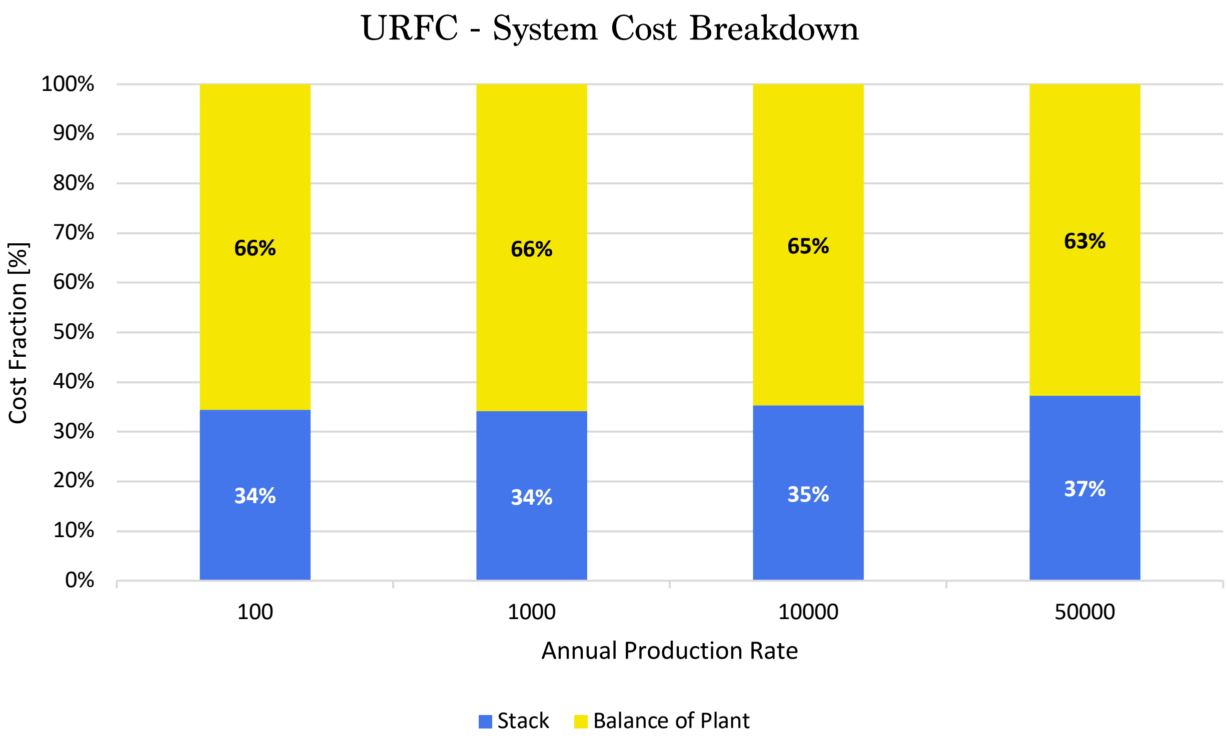


Figure 22. Disaggregation of the system by relative percentage of overall system cost for a 250 kW (FC mode) system

4.2.3. Parametric Cost Study

Since the beginning of the Project we aimed to develop – beyond the Technical Targets – a parametric cost study, which could be a useful tool to assist researchers and investors in future R&D and deployment of Unitized Reversible Fuel Cells, keeping in mind that it is a simplified parametrization focused on the most critical or expensive components of the system in order to give a general idea of the cost variations with increasing production volume.

In order to always be able to compare this technology with other energy storage technologies, we set up a tab in our Cost Study spread sheet dedicated to the calculation of the Levelized Cost of Storage (LCOS), capable of evaluating this metric under varying conditions. Moreover, a parametric cost study could be helpful to assess the current status of this technology with respect to the technical targets we set.

Our workflow has been to first develop a cost study for a 250 kW (fuel cell mode) system and then to identify some key input parameters that have significant impacts on the system design and on its costs. To understand how to make our study parametric, we analyzed all the repercussions that may occur on the system in case the values of key input parameters are modified.

The list of the chosen *key input parameters* and, in brief, their effects on the system is reported below:

- *System Size (FC mode)*: its variation has an effect on the power in EC mode, on the cost of the stack, on the cost of the balance of plant and therefore on the LCOS
- *Duty Cycle*: its variation has an effect on the rated energy, on the produced/consumed hydrogen and on the LCOS
- *Current Density*: the current density variation in both modes has an effect on the respective voltage of the cell and on the efficiency, on the power in EC mode, on the stack and BOP costs and, finally, on the LCOS
- *Platinum Group Metal (PGM) Loading*: its variation has an effect on the stack cost and consequently on the LCOS.

At stack level, the impact of these parameters on the system was considered in the following way: the variation of the *PGM loading* has a direct effect on the stack cost, since we are simply paying for a different quantity of catalyst needed by the Catalyst Coated Membrane (CCM) – as we described above, the CCM represents the major fraction of the overall stack cost.

A variation in *current density* affects the number of cells per system as shown below:

$$Number\ of\ cells = \frac{P_{FC}}{P_{single-cell(FC)}} = \frac{P_{FC}}{i_{FC} \cdot V_{FC} \cdot S}$$

where P_{FC} is the power in fuel cell mode in W, $P_{single\text{-}cell(FC)}$ is the power of a single cell in W/cm^2, i_{FC} is the current density in A/cm^2, V_{FC} is the cell voltage in V and S is the active area of the cell in cm^2. Having parameterized the polarization curve so that at each input value of the current density the spread sheet is able to calculate the respective cell voltage and the efficiency (parametrization only valid in the ohmic region of the curve), the power of the single cell varies, thus also the number of cells per system. It is precisely the number of cells that affects the stack cost.

The *power in FC* mode impacts the number of cell, too, as can be observed in the equation above: a variation of the numerator varies also the number of cells, therefore affecting the overall cost of the stack.

At balance of plant level, both the *power in fuel cell mode* and the *current density* have an impact on BOP components, meaning that they vary the flow rates that each component has to manage (e.g. consumed/produced hydrogen flow rate, water flow rate, etc.). Specifically, the components we decided to analyze and parametrize are:

- *Fuel Preheater*
- *Hydrogen Compressor*
- *Dryer*
- *Hydrogen-Water Separator*

The first two components were parametrized using the CAPCOST Program by Turton [43], which provides the purchased cost of equipment – for the most common components in chemical processes – calculated with a logarithmic function depending on three constant values and a specific parameter for the equipment under consideration, as follow:

$$log_{10}C_p^0 = K_1 + K_2 \cdot log_{10}(A) + K_3[log_{10}(A)]^2$$

where C_p^0 is the purchased cost of the equipment, A is the capacity or size parameter for the equipment and K_1, K_2, K_3 are constant for the specific equipment under consideration.

For the *Fuel Preheater* we set up the spread sheet so that it was able to calculate the needed exchange area with varying flow rates; in detail, the specific values utilized are:

Heat Exchanger					
K_1	**K_2**	**K_3**	**Units**	**Min Size**	**Max Size**
3.9912	0.0668	0.243	m^2	1	100

Table 12. Heat Exchanger - Equipment cost data from CAPCOST Program [43]

For the *Hydrogen Compressor* we set up the spread sheet with the same logic, applying the specific values below:

Compressor					
K_1	**K_2**	**K_3**	**Units**	**Min Size**	**Max Size**
5.0355	-1.8002	0.8253	kW	18	950

Table 13. Compressor - Equipment cost data from CAPCOST Program [43]

The costs of the Fuel Preheater and of the Compressor, obtained from the Turton's equation above, are expressed in \$2001: we later transformed them in \$2018 to make them consistent with the rest of our cost study.

For the last two BOP components (*Dryer* and *Hydrogen-Water Separator*), which were not present in the Turton reference, we decided for simplicity to linearly scale their cost starting from the reference values taken from A. Mayyas' study [14]; by doing so we overestimated their costs, since the trend of the cost increase in function of the component size is not linear (i.e. the cost of a component double the size of another one is less than the double of the original cost).

At balance of plant level, the *power in fuel cell mode* influences the power electronics, too: in our case we focused only on the *Power Inverter*; specifically, since this typology of components is not reported in the Turton CAPCOST Program (which deals only with components in chemical processes), we parameterized the inverter by linearizing its cost between the reference costs of two commercially available inverters (the only data point that were found), with identical characteristics except for the power level. The validity range for this last parameterization is:

Inverter: Range of Validity for its Parameterization		
P_{min}	**P_{max}**	**Units**
250	500	kW

Table 14. Inverter: Range of Validity for its Parameterization

In conclusion, all the key input parameters we chose for our study have an impact on the Levelized Cost of Storage (LCOS, whose equation is given in the Technical Targets Chapter), since this metric, allowing to easily compare a system with other energy storage technologies, has to include all the main quantities of a storage system.

Specifically, the *Power in Fuel Cell mode* has an effect on both the investment and the O&M (operation and maintenance) terms of the LCOS metric; the *Duty Cycle* (that we assumed to be daily as a first hypothesis) determines a variation of the rated capacity, thus, again, on both the investment and the O&M terms; the *Current Density* (of both modes), which affects the number of cells and, consequentially, the cost of the stack, has an effect on the investment, on the O&M terms and, finally, on the charging terms (since the current density determines the efficiencies in both modes of operation and, therefore, the RTE); lastly, *the PGM loading*, too, have an effect on the investment term, changing the quantity of catalyst needed for the cell operations.

5. CONCLUSIONS

Two sets of *Technical Targets* were developed (both at cell/stack and system level), one for Unitized R-PEM and one for Unitized R-SOFC after an in-depth literature review and feedback from experts, in order to assess the status of these new technologies and to identify the most critical R&D needs. In R-PEM technology the major issues are related with the oxygen electrode, as it presents a corrosion problem of carbon-based materials during electrolyzer operation, a slow kinetic of reaction (different catalyst needed for ORR and OER) and difficult water management (tradeoff between hydrophilic and hydrophobic properties). In R-SOFC technology the major problem is the durability related with the delamination of the oxygen electrode, especially under SOEC operation; however recent studies report that switching from FC mode to EC mode is advantageous for the lifetime of the electrode.

In both the *2025 Targets* and in the *Ultimate Targets*, the LCOS values set for R-PEM technology are \$0.40/kWh and \$0.25/kWh respectively, while for R-SOFC technology are \$0.25/kWh and of \$0.15/kWh respectively; it is important to point out that these are preliminary targets based on researches and engineering knowledge.

Furthermore, a *Parametric Cost Study* for R-PEM was developed as a tool to estimate a medium-fidelity cost of a unitized system and its possible cost reduction trend with increasing manufacturing volume; an effort was made to parametrize both the stack and the Balance of Plant (BOP). This tool receives as inputs the system size (in FC mode), the duty cycle, the current density (in both modes of operation), the Platinum Group Metal (PGM) loading (note that this parameter could affects the current density) and it is able to calculate the respective investment cost, assisting researcher and investors for future Research, Development and Deployment (R&DD) of this technology. Moreover, an LCOS calculator was created, capable of evaluating this metric under varying conditions of the parametric cost study (e.g. capital costs, duty cycle, fuel cell and electrolyzer current density), to facilitate the comparison with other energy storage technologies.

To date, our R-PEM parametric cost study estimates a capital cost of $4,557/kW for a 250 kW fuel cell based system, at 100 units/year manufacturing volume, including hydrogen storage, installation costs and margin profits, and a LCOS of $0.37/kWh (assuming a daily duty cycle). Capital cost, efficiencies and lifetime of the system are the most significant factors affecting the LCOS metric for R-PEM. Platinum Group Metal loading and power electronics potentially stand as a considerable cost reduction for R-PEM.

In conclusion, Unitized Reversible Fuel Cells are a flexible and a promising technology, still at an early stage, capable of transforming into hydrogen potentially large amounts of excess electricity from renewable energy sources: this can improve electricity grid's reliability and resiliency. Key topics for further research are in-depth study on PEM durability and related costs, additional simplification or consolidation of the BOP and investigation of a broader range of duty cycles (e.g. for seasonal storage applications).

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