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Thermal optimization of an hydrogen storage system based on metal hydrides

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Abstract

The production of power using renewable energies carries an intrinsic problem, it is strictly bonded to the availability of wind and sun, the presence of the sources can be usually mismatched with respect to the energy demand, this feature makes it necessary to find valid and efficient alternatives to store the energy produced in order to have energy available in every moment.

Hydrogen, with a large versatility of usage in different fields, can be used as a very interesting alternative for energy storage. There are mainly three ways to store it depending on the phase: gaseous, liquid, solid. In our system the storage is made with a solid phase, performed with metal hydrides technology. This alternative shows very good characteristics in terms of reversibility of the process, volumetric density and storage pressure values. The main disadvantage of this technology is its poor heat transfer characteristics that are the causes of a low hydrogen useful capacity and low velocity of the processes.

The aim of this work is to perform a study of an experimental system, based on hydrogen production, storage, and conversion into energy, with a particular attention on thermal management aspects of the components, with the objective of enhancing its thermal behavior in order to better understand it and increase its efficiency and duration of operation.

We performed an experimental procedure, studying the behavior of the metal hydrides during the adsorption and desorption phase, analyzing different heat transfer configurations and noticing a relation between the heat conductivity of the fluid in contact to the tanks and the duration of the processes. We tried to put in contact the MH canisters with water, obtaining a more stable temperature of the tanks and a longer duration of the desorption process and a higher hydrogen capacity if compared to the normal case. We did the same with coconut oil instead of water, but in this case, we did not obtain results as positive as with water.

We also tried to thermally couple the fuel cell and the MH canisters to exploit the heat released by the fuel cell to obtain a higher and more stable temperature of the water that surrounds the tanks, and to increase the efficiency of the fuel cell. We obtained a slight improvement but there is still possibility to reach better results.

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1 Introduction

1.1 Hydrogen production

The global interest on hydrogen is growing constantly every year, taking into account the projections made by the European union, Europe will have a production of 20 to 35 million tons of renewable-based hydrogen in 2040, accounting for around the 10% of the total energy demand[1], hydrogen is an energy dense, light, and storable gas, and its utilization does not produce direct CO₂ emissions. Indeed, the combustion of hydrogen, as opposed to fossil fuels, releases only water vapor. Its role will become fundamental in several sectors because it can replace fossil fuels in hard-to-abate sectors as heavy industry, power generation and transports.[2] It is suitable to produce chemical products, fuels and fertilizers. In the maritime and aviation transportation will play a fundamental role, replacing gradually a part of the fossil fuels.

Hydrogen is the most abundant element in the universe but is difficult to find it at the molecular state on earth, so it must be produced starting from other compounds that contain it such as water, ammonia, methane, organic compounds and many others.[3] For this reason is considered as an energy carrier and not as a source of energy. To have a positive effect on the process of decarbonization it must be produced starting from renewable sources, in this case is called “green” hydrogen.

Most of the production of hydrogen is performed through steam methane reforming, it is a process in which hydrocarbons react with steam with consequent hydrogen production. This process is characterized by a very low cost of hydrogen, but it has also undesirable emissions, so it is not completely aligned with the environmental requirements. [4]

Another largely used possibility to produce hydrogen is the electrolysis, that does not imply pollution emissions and the only byproducts are oxygen and heat. However, a large limiting factor for this technology is the up scaling, indeed it is possible to use it only for relatively low scale hydrogen production otherwise the cost of production largely increases.

There are also other ways to produce it as we can see in the figure 1 in which are listed some of the possibilities.

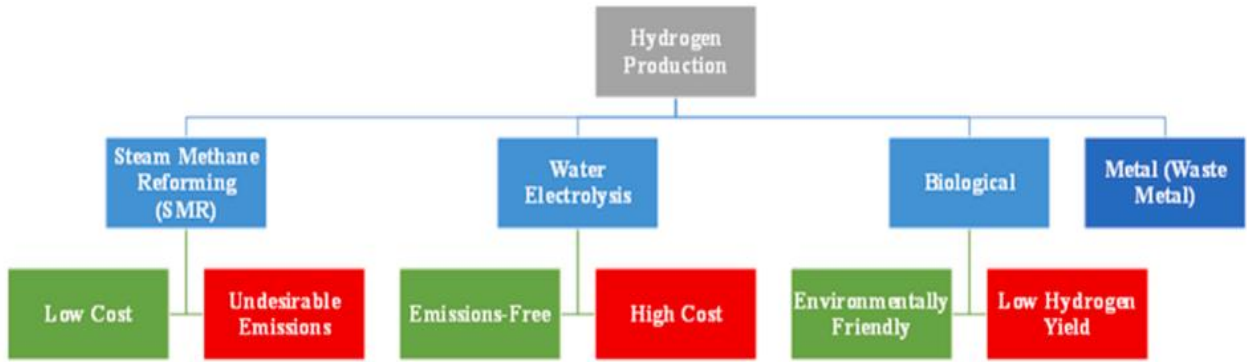


Figure 1 : Hydrogen production

1.1.1 Electrolysis

The electrolysis reaction of water is an electrochemical reaction in which with the use of an electrical current is possible to break the bonds inside the water molecules, by doing that we obtain hydrogen and oxygen. The reaction is:

Anode:



Cathode:



With the general reaction:



It is a key element for the future of hydrogen because it represents a possibility to produce hydrogen in a sustainable and long-term way, since only water is needed for the reaction and the electricity to drive the process can be provided by renewable sources. An important feature is that it can produce hydrogen with high efficiency and without the production of greenhouse gases and pollutants. At the moment over the 95% of the hydrogen used in the hydrogen economy is produced starting from fossil fuels, so we are still very far from a responsible production. Moreover, hydrogen has still a high cost of production using this type of technology, due to the need of noble metal-based electrodes and coatings that make this technology not suitable for large-scale production. [5] An electrochemical cell is a device able to drive a complete redox reaction but forcing the separation of the oxidation step and the reduction step. The device is composed by four different parts: an anode, a cathode, an electrolyte layer, and an external circuit. Inside this type of technology, we obtain two useful effects: A generation of a coherent flow of electrons across the external circuit that defines a current flow, the second effect is related to the charge separation that it happens at the anode and at the cathode when a redox and oxidation reactions take

place. The charge separation generates a voltage gradient, the difference between the anodic and cathodic voltage gradients generates a voltage gradient across the cell. Since we can generate current and voltage, we can generate power. If the redox reaction is spontaneous (ΔG of reaction lower than 0) we have power production, this is the case of the fuel cell that produces power. If the redox reaction is not spontaneous (ΔG of reaction higher than 0) we have consumption of electricity, this is the case of the electrolyser, that consumes power to break water molecules. The utilization of hydrogen as a fuel is very promising due to high density of energy and the absence of CO₂ as product during combustion. Combustion is not the only way to convert hydrogen into energy, it can be done also using a fuel cell. A fuel cell is a device in which is possible to convert the chemical energy of a fuel into electricity, with high efficiency and low emissions of greenhouse gases.[6] Due to the fact that the energy production is not related to a heat engine process, there is not the limitation of efficiency of the Carnot cycle. It is mainly composed by four main parts: anode, cathode, electrolyte, and the external circuit.

1.1.2 PEM electrolysis

Proton exchange membrane electrolysers are devices used to produce hydrogen through electrolysis, splitting electrochemically the water molecule into hydrogen and oxygen. In Figure 2 is reported the structure and the reaction taking place inside the cell. It is possible to notice a negatively charged section called cathode, and a positively charged section called anode, they are divided by the membrane that allows the passage of the protons. At the anode we have the split of water into oxygen electrons and protons, the protons pass through the membrane and reach the cathode where they enter in contact with the electrons, obtaining in this way the hydrogen. [7] Normally this type of cells operate at a temperature between 50 and 90 °C, and pressures between 15 and 30 bars.[8]

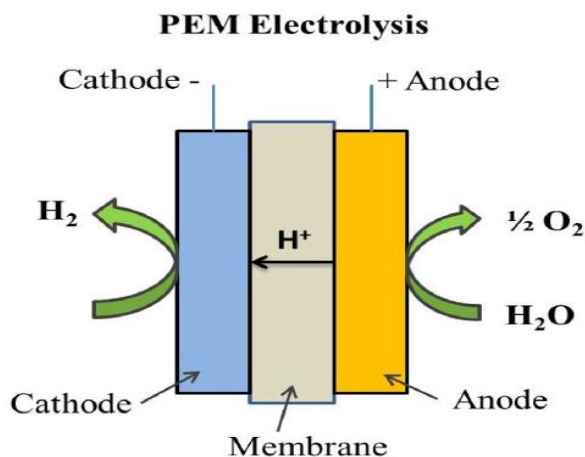


Figure 2: PEM electrolyser

1.1.3 ALK Electrolysis

This is another type of electrolyser cell, based on a different electrochemical reaction. At the cathode two molecules of alkaline solution (KOH/NaOH) are reduced to one molecule of hydrogen and two hydroxyl ions (OH⁻). These ions are able to pass through the porous diaphragm under the motion imposed by the external electrical circuit, reaching the anode where it discharges in half molecules of oxygen, 2 electrons and one molecule of water. The structure of the cell and the reactions described are reported in Figure 3. This typology of electrolyzers operates at a temperature between 30-80 °C, in presence of an aqueous solution (KOH/NaOH) as the electrolyte.[7]

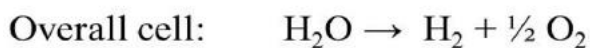
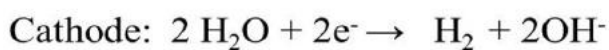
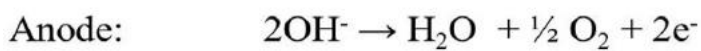
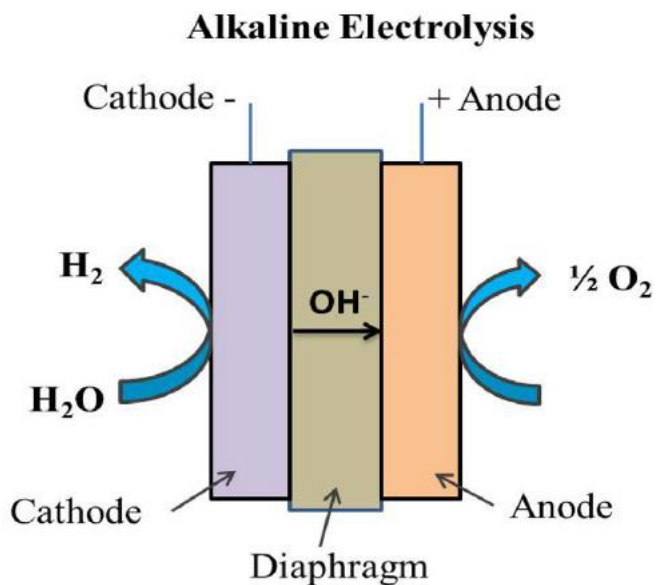


Figure 3: ALK electrolyser

1.1.4 SOEC Electrolysis

This type of technology operates at much higher temperatures, from 500 to 850 °C, with the utilization of water in form of steam. Conventionally as a membrane is used O²⁻ conductors, that are made of Ni/yttria stabilized zirconia. The higher operative temperature causes some issues related to stability of the system and degradation. The reaction that takes place and the structure of the cell are reported in Figure 4.

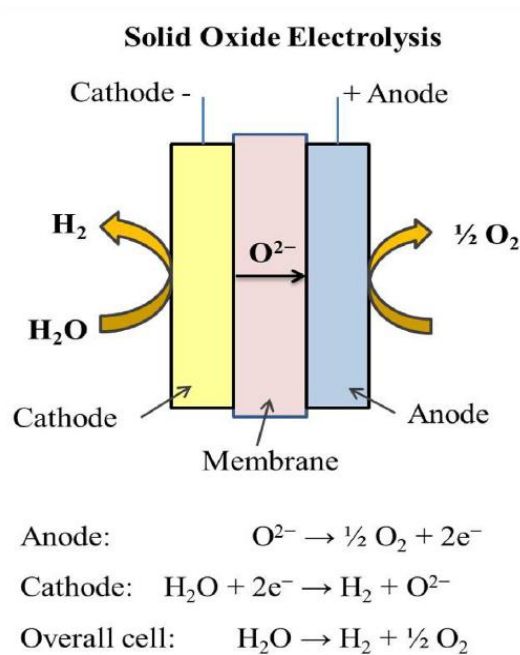


Figure 4: SOEC electrolyser

1.2 Hydrogen storage

Hydrogen is a very promising energy vector in the field of energy because of its high versatility and characteristics. It has a very high energy density by mass, but a low volumetric energy density, for this reason it is important to find alternatives to increase this quantity.

Depending on the different typologies of storage, we can have a technology that is more suitable for mobile or on-site applications, some very important factors to determine which one is the best are weight, volume occupied and safety of the plant.

Hydrogen can be stored mainly in three different ways depending on its phase: solid, liquid, or gaseous.

1.2.1 Gaseous Hydrogen

This is the oldest alternative between the above mentioned and it is the most used and mature. The basic idea is to compress the hydrogen and use cylindrical vessels at high pressure to store it. Compressors and vessels of this type are largely used also in other fields of energy, for this reason they are at advanced state of the art. The usual range of temperature is between 350 and 700 bars. They can be used in mobile applications due to a relatively small volume occupied, relatively low weight and relatively high energy density. One of the drawbacks of this technology is the safety due to the flammability and high pressure of hydrogen that can lead to explosions in case of accidents. For these reasons the vessels must guarantee high safety characteristics. Normally they are made of aluminum, high strength steels or carbon reinforced composites; they also have to be diffusion-resistant to avoid hydrogen embrittlement.

1.2.2 Liquid hydrogen

Another possibility to store hydrogen is to liquefy it, under 240 °C that is the critical melting point of hydrogen it changes phase from gaseous to liquid. [9] The density of hydrogen increases with respect to the condition of compressed gas but the need to maintain a so low temperature requires a large amount of energy, it can be up to 40% of the entire energy content, with respect to 10% of compressed hydrogen.[10]

1.2.3 Solid hydrogen

Another alternative is the solid state, it is the result of the reaction between hydrogen and another element, the phenomenon of bonding is called adsorption. It's the process for which we have an adhesion of molecules or atoms of a liquid or a gas to a certain surface. The reaction happens between the adsorbent that is the surface where the process happens, and the adsorbate, that is the substance that reacts with the surface. We have the process of adsorption that is an exothermic reaction with release of heat, and the opposite reaction, called desorption, that is an endothermic reaction with requirement of heat.[11] Adsorption can be of two typologies: chemisorption or physisorption, depending on the kind of interaction that intervene between the atoms and the surface.

Physisorption is the adsorption that happens with physical phenomena, the most relevant interaction force in this case is the Van der Waals force. In this case we do not have any changes in the chemical bonding structure.[12]

Chemisorption is the adsorption in which happens a chemical reaction between the adsorbate and the surface, in this case we have a chemical variation of the chemical bonding structure.[13]

In our case we use a metal hydrides-based technology in which with a chemisorption reaction, chemical bonds are created between a metal powder (mix of different metals), that is the adsorbent, and hydrogen that is the adsorbate.

1.3 Fuel Cell

There are different typologies of fuel cell, depending on the type of electrolyte that is inside the electrochemical cell. The most mature typologies are: Alkaline fuel cell, proton exchange membrane fuel cell and solid oxide fuel cell.

1.3.1 PEM Fuel Cell

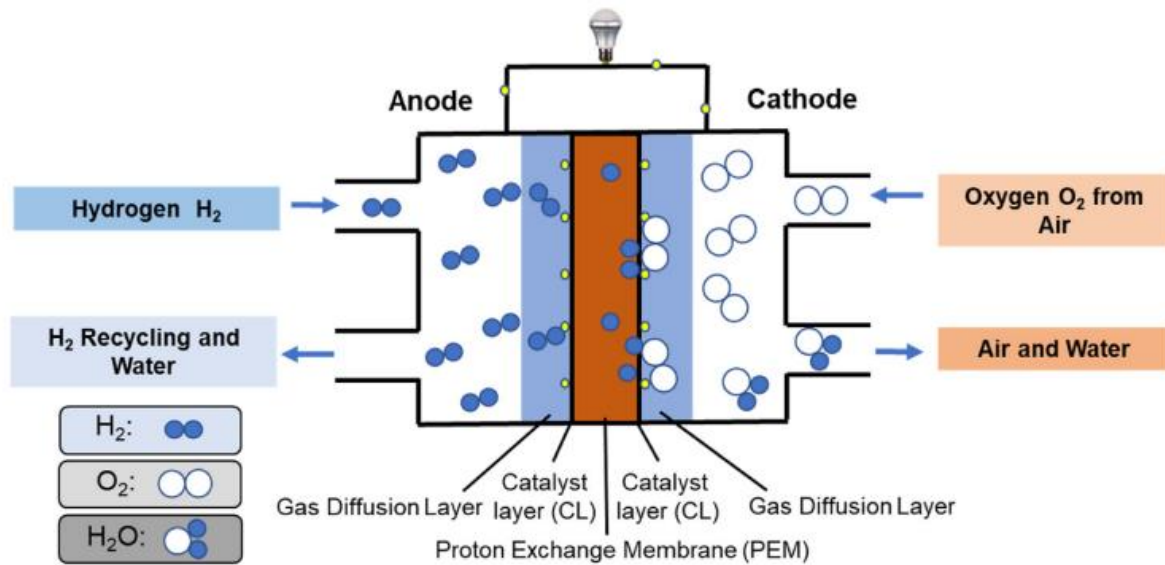


Figure 5: PEM fuel cell electrochemical cell structure [14]

We have 4 main areas: the proton exchange membrane that has the role to divide anode and cathode allowing the passage only of the ions, the catalyst layers that are used to enhance the speed of the reactions, the gas diffusion layers that allow the transport of the gases and electrons to the reaction sites and also to remove the water produced and heat, and the bipolar plates with airflow channels are used to guarantee a more uniform distribution of reaction gases.[14]

In the proton exchange membrane fuel cell these are the reactions that happen at the anode and at the cathode:

Anode:



Cathode:



Total formula:



So, we have the entrance of the hydrogen at the anode, there it loses electrons, and it originates hydrogen ions, the electrons are driven by the external circuit to the cathode, and the protons pass through the proton exchange membrane driven by the electric field that there is between cathode and anode. Ions and electrons arrive to the cathode where they meet oxygen, and they react together generating water and heat.

This production of heat may cause the increase of temperature, it must be controlled to avoid damages of the fuel cell and problems during operation. A good control of temperature and water must be done to avoid flooding, damage of the proton exchange membrane, dehydration that are phenomena that can reduce the performances and the lifetime of the fuel cell.

1.3.2 ALKALINE

It uses an alkaline electrolyte solution (most used is KOH). So, in this technology the ion used is OH^- instead of H^+ , the reaction is:

Anode:



Cathode:



This is the third type of fuel cell that we present, also in this case we are in the field of low temperature fuel cells, with an operational temperature around 70°C . It shows a higher reduction and oxidation kinetics of the oxidants and fuels at the electrodes with respect to other fuel cells. [15]

1.3.3 SOFC

This type of technology is characterized by a high operational temperature range, between 600 and 1000 $^\circ\text{C}$, this feature makes it necessary to use materials able to sustain these conditions. For this reason, this type of technology has a higher cost, but it also shows a higher efficiency. The operation at this high temperature makes it possible to exploit the heat produced during operation for combined heat and power applications (CHP). A usual structure of a SOFC is a sandwich structure, with two porous electrodes for electrochemical reactions, divided by a dense electrolyte that allows the passage of the O_2^- ions. [16] With respect to other fuel cells that work only with hydrogen, SOFC are very versatile in terms of fuel, they can use H_2 , CO, methane, biogas, ethanol, NH_3 , etc.[16]

1.4 Literature Review

Many studies have been conducted on the MH hydrogen storages focusing on the heat transfer characteristics of this technology. As it is possible to see in the work of Mahvash Afzal et al. [17] they studied different configurations of heat transfer to try to overcome the poor thermal conductivity of the metal hydrides, that is considered a main limiting factor for the performances of this technology. They proved that a lower temperature of the heat transfer fluid put in contact with the metal hydrides tank, is a beneficial characteristic to increase the amount of hydrogen adsorbed.

In the study of Yang Ye et al. [18], they analysed the effect of the utilisation of a phase change material as heat transfer fluid put in contact with the metal hydride storage. They proved that a

sufficient amount of PCM can absorb all the heat generated during the adsorption reaction and promote the reaction, and that a PCM with a higher thermal conductivity is beneficial for the duration and the efficacy of the adsorption reaction.

Another study that is very useful to understand the phenomena involved in this type of systems is the one of Disli et al. [19], they studied a MH system performing the experiments with respect to the external convective heat transfer coefficient and they used a phase change material to optimize the heat exchange and varying its thickness to perform a sensitivity analysis. Indeed, phase change materials can be used as passive heat exchangers, they receive the heat from the MH canister during the adsorption process (exothermic reaction) and they release the heat during desorption (endothermic reaction). They noticed that the heat transfer coefficient is much more influent on the performances of the system if compared to the effect of the increase of the thickness of the PCM, this last parameter is influent just for very low levels of latent heat. They also analysed the heat transfer temperature of the heat transfer fluid put in contact with the metal hydride storage, proving that a lower temperature of the heat transfer fluid is beneficial for the amount of hydrogen stored.

A further investigation on the heat transfer techniques was made by Zhu et al. [20], they studied the MH behaviour in presence of different external and internal heat transfer techniques. They proved that the MH performances can be greatly improved by implementing a better external heat transfer exchange. They changed the external environment from natural convection (as in our base case) to a water-cooling system, obtaining an improvement of the absorption time of 72.8 % and the effective hydrogen release of 97.8%. They also proved that the thermal conductivity and the heat transfer surface play a fundamental role in the improvement of the performances, also finding that after a certain value there is not a noticeable effect in increasing more the heat transfer coefficient beyond certain values. They also modified the internal structure of the storage system obtaining very interesting results, but unfortunately, we did not have the possibility to apply this typology of improvement to the system.

A possibility that is under development in the thermal optimization of this type of technologies is to thermally couple the fuel cell with the metal hydride reactor. The fuel cell is connected through an heat exchanger to the canister in order to exploit the heat generated during the functioning of the fuel cell to provide heat to the MH canisters during the release of hydrogen, that is an endothermic reaction. This is an option that allows to obtain two beneficial effects: one is that taking heat from the fuel cell we decrease its temperature, and so we are able to increase its efficiency, because less power is requested to the fans to decrease the temperature; the other beneficial effect is to provide heat to the MH canister during the endothermic reaction, giving to the process more stability and allowing to reach a higher hydrogen release rate for a longer time. Normally better results are obtained if the heat is provided directly to the MH bed and not to the external surface. In the work of Liang Tong et al.

[21], they performed a study on the coupling between a PEM fuel cell and a MH storage and obtained a system that is able to work without any additional auxiliary heating equipment during discharging process.

1.5 Aim of this study

In our system we used a PEM electrolyser, a metal hydride hydrogen storage and a PEM fuel cell to perform our experiments.

The aim of this study is to characterize our hydrogen system from a heat exchange point of view, analysing the behaviour of the components that show more noticeable effects during operation, such as the metal hydrides canisters, that experience a heat release during the adsorption phase, and a heat request during the desorption phase that implies a reduction of the temperature that originates a deterioration of the performances in terms of time of the process and capacity of hydrogen release; and the fuel cell that during its functioning experiences an increase of temperature that causes a decrease of the efficiency, due to the higher amount of energy required for the auxiliary functioning to reduce the operative temperature. When the temperature exceeds a certain value, there is also an immediate shut down of the component to avoid internal damages. This phenomenon happens when the power request it is very high, so the fuel cell it is not able to operate at high powers for long periods without experiencing this phenomenon. Unfortunately, the model that we used for this study was kept off for a long period, and at the moment due to issues related to the long inactivity, it does not have returned to normal operation, so we were not able to work at power higher than 200 W. Working in this condition, the temperature does not increase at the point to originate the emergency shut down and so we were not able to work on it to solve the problem as we wanted to do.

We proposed three configuration improvements to try to obtain a more stable the temperature of the MH canisters putting them in contact first with water, and then with coconut oil.

We also proposed a configuration to exploit the heat released by the fuel cell during operation both to decrease its temperature in order to increase the efficiency and to heat up the water in contact with the canisters to maintain the temperature more stable.

2 Experimental setup characterization

In our experimental bench we have the three main components that are: the electrolyser, for the hydrogen production, the Metal Hydride storage for the hydrogen storage and the fuel cell for the conversion of hydrogen into electricity. The electrolyser receives electricity from the grid (or from the PV panels to obtain green hydrogen), and water deionized from a reservoir tank. It produces oxygen, that is released in the room, and hydrogen that goes inside the metal hydride storage. The electrolyser is governed by the operator through a simple on/off configuration, related to the need to produce hydrogen and to stop the production when we reach the desired value. The MH storage is connected to the fuel cell, and through a pressure regulation (a manual valve) it is possible to regulate the pressure at which we want to provide the hydrogen to the fuel cell, respecting the input value of pressure requested by it. Even the fuel cell works in an on/off configuration, depending on the necessity to produce energy or not. It receives in input also oxygen captured inside the surrounding air. As input, it receives also electricity from an electrical generator to guarantee the functioning of the auxiliaries. As output we have water, that is released into a tank, and electricity that is used to supply the energy demand of the programmable load (that normally is set at 200W).

Connected to the fuel cell we have two sensors: the Cassy module, to monitor the internal temperature of the fuel cell, the voltage and the current that can provide, and the flowmeter that gives us the value of the hydrogen flow that the fuel cell is receiving from the storage, to operate at a certain power. To monitor the temperature of the different components (canisters and fuel cell) we applied several thermocouples on the surfaces.

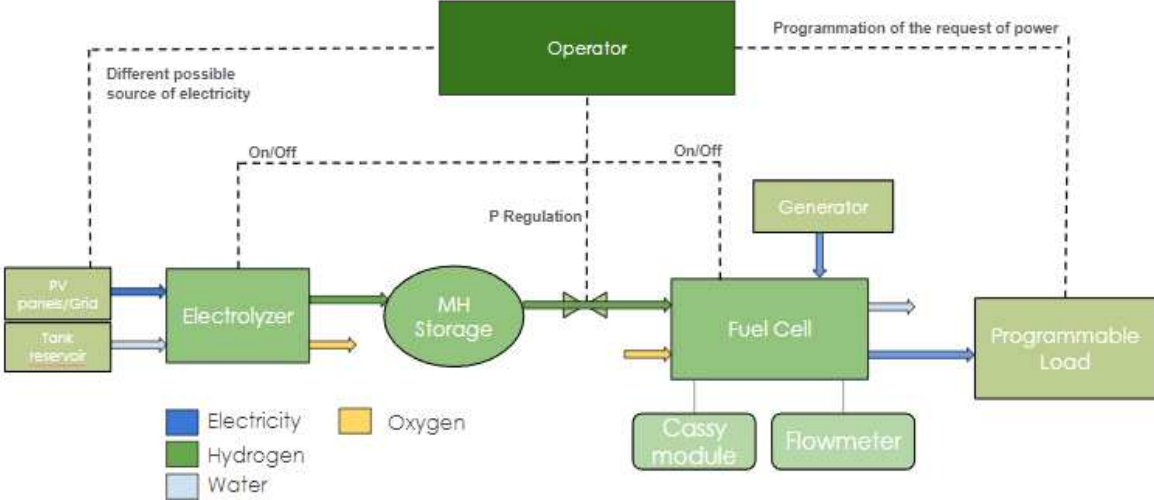


Figure 6: Scheme of the plant

2.1 List of Equipments

2.1.1 Electrolyser HyPEM XP Rack 1000

The electrolyser is the device that ensures the production of hydrogen through the electrolysis of water. In this study we have used an electrolyser of H2 planet company model Hy PEM XP Rack 1000. In Figure 8 is possible to see the parameters of the electrolyser taken from the data sheet provided by the manufacturer H2 planet. We have used the model Hy-PEM XP Rack 1000.[22]



Figure 7: Electrolyser

Electrolyser Model	Hy-PEM XP Rack 1000
Eletrolytic Cell	PEM technology
H2 Purity	>99.99999% (refers to O2), dew point <-70 °C
Output Pressure	16 bar (232 psi)
H2 flow rate cc/min (max)	1000
Dimensions	Standard 19" Rack 4U - depth 40cm
Net weight (empty water tank)	22 kg
Flow Rate at max output*	6.5 L/min
FC Efficiency	41%

Figure 8: Electrolyser technical specifications [22]

2.1.2 Metal Hydrides H2 Storage

To store the hydrogen, we have used a metal hydride system with 3 canisters of the model MyH2 600 each with a storage capability of 600 liters, with metal powder inside where the phenomena of absorption and desorption occur.

The metal powder is constituted by around 40-60% of Manganese, 20-40% of Titanium, 10-30% of Vanadium, 2.5-7 % of zirconium and from 1.5 to 3% of Iron.[23]



Figure 9: Image of the canisters

2.1.3 H-500 PEM Fuel Cell

As already mentioned in the general description of components, we used a PEM fuel cell from Horizon Fuel Cell Technologies model H-500 Fuel Cell Stack. It is the device responsible for the conversion from hydrogen to electricity.

In the table x is possible to see the parameters of the Fuel cell, taken from the data sheet provided by the manufacturer Horizon Fuel cell technologies. The model that we used is a H-500 Fuel Cell stack.[24]

Type of Fuel Cell	PEM
Number of Cells	24
Rated Power	500W
Performance	14.4V @35A
H2 Supply Valve Voltage	12V
Purging Valve Voltage	12V
Blower Voltage	12V
Reactants	Hydrogen and Air
External Temperature	5 to 30°C
Max Stack Temperature	65°C
H2 Pressure	0.45-0,55bar
Hydrogen Purity	$\geq 99.995\%$ dry H2
Humidification	self-humidified
Cooling	Air (integrated cooling fan)
Weight (with fan & casing)	2520 grams (+- 50grams)
Controller	400 grams (+-30 grams)
Dimension	13cm x 26.8cm x 12.25cm
Flow Rate at max output*	6.5 L/min
Start up time	≤ 30 s at ambient temperature
Efficiency of stack	40% @ 14.4V
Low Voltage shut down	12V
Over current shut down	42A
Over temperature shut down	65°C
External power supply	13V (+-1V), 5A



Figure 10: fuel cell [24]

2.1.4 Bronkhorst Flow meter

This device controls and measures the flow rate of hydrogen from the canisters to the fuel cell. It allows to measure the values of efficiency and hydrogen consumption collecting the data through LabVIEW VI software.[25]

2.1.5 Electronic Load

It is a Programmable Electronic DC load, model EA-EL 9000 T 400 W - 600 W. It has the possibility to simulate the power consumption of some typical loads, such as an electrical car, a building etc. We mainly used this device in remote control mode integrating it in the software LabVIEW VI, but it can also be used in manual mode, changing the power consumption through the knobs.[26]

2.1.6 Cassy Module

Is an interface that is used for the acquisition of measurement data such as temperature, current and voltage of the fuel cell through the software LabVIEW VI. [27]

2.1.7 Graphtec GL840

Is the device used to perform the temperature evaluations of the different components of the system during operation. It uses thermocouples to perform the measurements, it can perform up to 30 T measurements at the same time.

For the temperature measurements, as already mentioned we used a Graphtec sensor with thermocouples. Here are reported the images of the different positions for canister temperature, ambient air and external temperature of the fuel cell.[28]

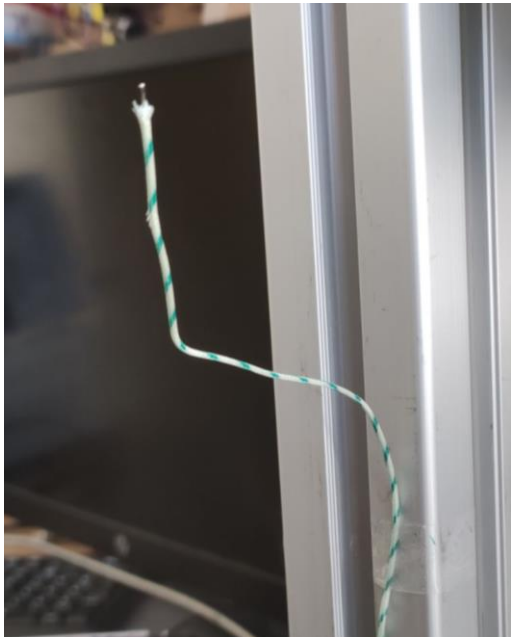


Figure 11: Thermocouple ambient air



Figure 12: Thermocouple on the storage surface

2.1.8 Coconut oil

During the improvement part we used circa 8 liters of coconut oil to try to optimize the heat management of the canisters. The coconut oil shows a phase change temperature around 24 °C, that is exactly in the range between the lower value of the operative temperature of the system, and the higher value. We wanted to exploit its phase change state to store more thermal energy, in latent form, to maintain the temperature of the system more stable.

In the Table 1 are reported the physical properties of coconut oil.[29]

Properties (units)	Coconut oil	
	Solid	Liquid
ρ (kg/m ³)	920	918
μ (Pa s)	-	0.0268
c_p (K/(kg K))	3.750	1.670
k (W/(m K))	0.166	0.166
β (1/K)	0.7×10^{-3}	
h_f (J/kg)	103.000	
T_m (K)	297	

Table 1 : Physical properties of coconut oil

2.1.9 Insulation materials

During the improvement part we insulated the canisters to decrease the heat losses to the environment. We used polystyrene with a heat conductivity of circa 0.01 W/m²/K for the lateral and bottom surface and to cover the system a square piece with heat conductivity of 0.0146 W/m²/K.[30]



Figure 13: Image of the system after insulation

2.1.10 Heat exchangers

During the improvement part we used two heat exchangers of 5,7cm x 5,7cm, to use the heat generated by the fuel cell during operation to heat up the water of the canisters.[31]

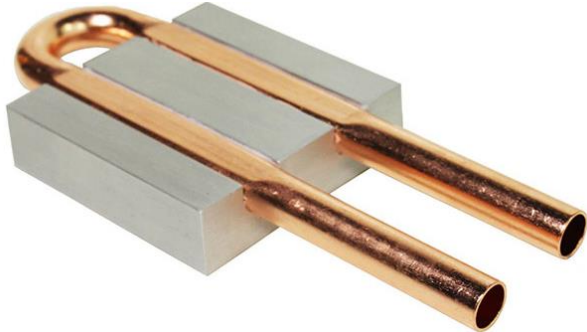


Figure 14: Heat exchanger

2.2 Data extraction and synchronization

2.2.1 LabVIEW

We use the software LabVIEW to extract, synchronize and display the data from the equipment during operation. We can extract: the hydrogen flowrate in L/s provided to the fuel cell, current, voltage and power provided to the load, the efficiency of the fuel cell and the internal temperature of the fuel cell. In input we provide to the software an excel file to precise the format of the data that we want to receive in output and we connect the different measurement instruments that we need to use.

In Figure 15 is reported the user interface of LabVIEW, that allows to start the data acquisition process, through the connection of the sensors of our system. And displays the hydrogen flowrate provided by the MH canisters and the power provided by the fuel cell to the load.

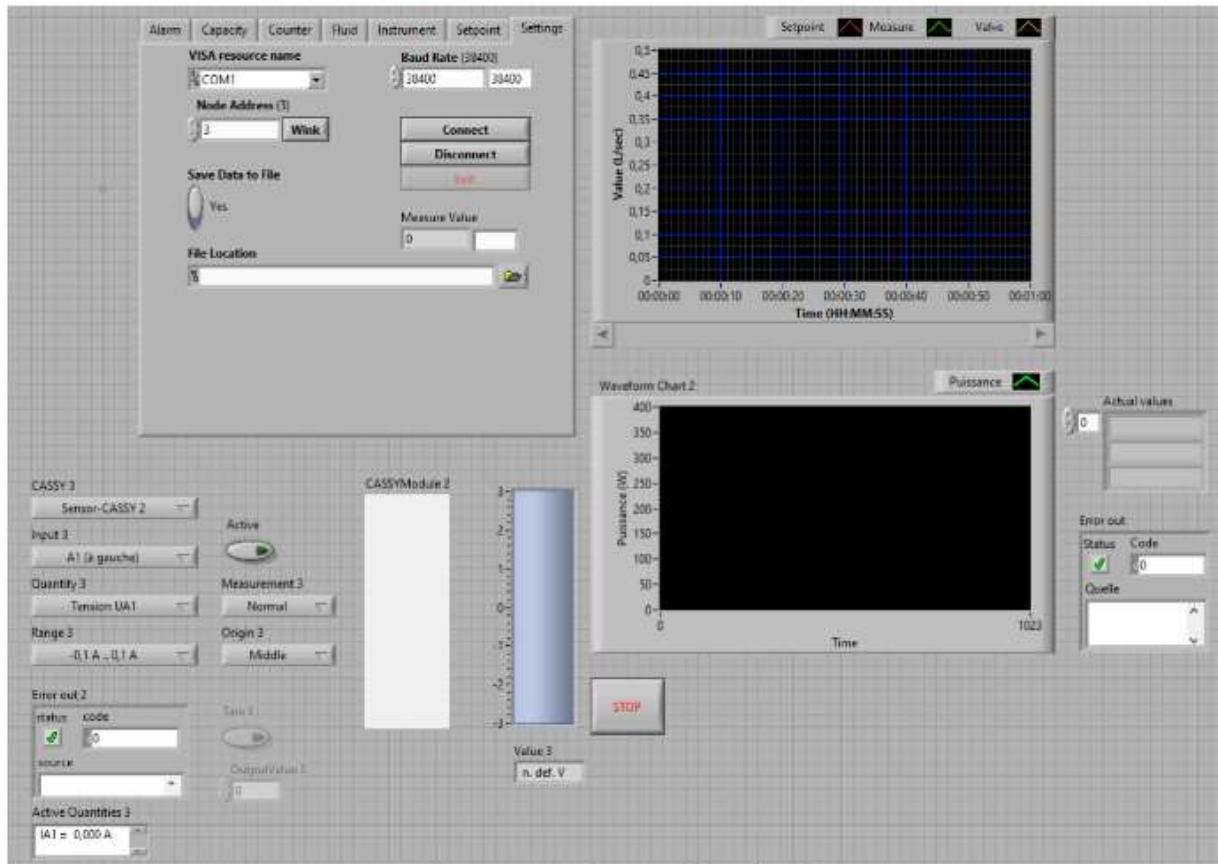


Figure 15: LabVIEW interface

2.3 Experimental plan

The experiments are organized in four main phases:

- 1) Temperature measurements of system in the base case.
We emptied the canisters, we did an entire process of charging (adsorption phase), after that an entire process of discharging (desorption) working with the fuel cell at a power of 200W. Taking the values of temperature that we needed and performing this procedure several times to validate the results. This procedure is repeated in the same way in the following configurations.
- 2) Temperature measurements of the system with the MH canisters in contact with water.
- 3) Temperature measurements of the system with the MH canisters in contact with the coconut oil.
- 4) Temperature measurements of the system in the configuration of the system with MH canisters in contact with water, and water and the fuel cell thermally coupled through the heat exchangers.

All the phases are performed for the adsorption and the desorption process, in this case the fuel cell is always working at a power of 200W.

The variables that we will use to present the results are:

- T_{amb} : That is the ambient temperature of the room.
- T_{water} : It is the temperature of the water inside the box that contains water and the tanks.
- $T_{can 2}$: It is the temperature of the canisters that shows a more noticeable thermal behavior.
- $T_{amb int}$: That is the internal ambient temperature of the insulated box that we used during the coconut oil phase.
- $T_{amb ext}$: That is the external ambient temperature outside of the insulated box during the coconut oil phase.
- $T_{oil can 2}$: It is the temperature of the oil in the proximity of the canister 2.
- $T_{int FC with HE}$: It is the temperature on the internal surface of the fuel cell in the case with the heat exchanger.
- $T_{int FC}$: It is the temperature on the internal surface of the fuel cell in the case without the heat exchanger.
- $T_{ext FC with HE}$: It is the temperature on the external surface of the fuel cell in the case with the heat exchanger.
- $T_{ext FC}$: It is the temperature on the external surface of the fuel cell in the case with the heat exchanger.

3 Thermal behavior of the system

To test the the Metal Hydrides canister I performed an entire process of discharging and charging alternatively, to monitor its evolution of temperature. All the measurements are made utilizing just one canister to have a faster operation, in this way we can perform more tests to better validate the results. The canister that I chose was the one that showed a stronger variation of temperature during operation.

In Figure 16, is reported a scheme in which is put in evidence the structure of the system and the respective thermal behavior of the , we can see that both the electrolyser and the fuel cell during operation experience a release of heat to the environment, while the MH storage can experience both a release of heat or a high request from the environment depending on the process that is happening, adsorption or desorption respectively.

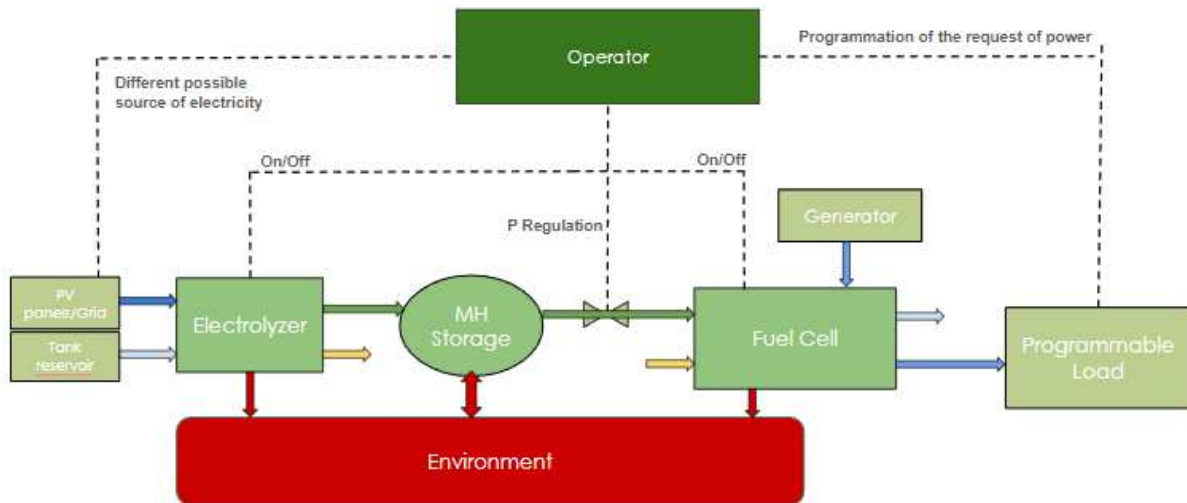


Figure 16: Scheme of the system

3.1 Evaluation of the canister temperature during adsorption phase

The adsorption phenomenon is an exothermic reaction, it is a spontaneous reaction in which we have a release of heat, for this reason during this phenomenon the canisters experience an increase of temperature.

In the **Error! Reference source not found.** we have the case of one canister charging, we see an evolution of temperature from 15 degrees to 33 °C. The temperature of the ambient air remains almost constant. The process finishes in almost 68 minutes.

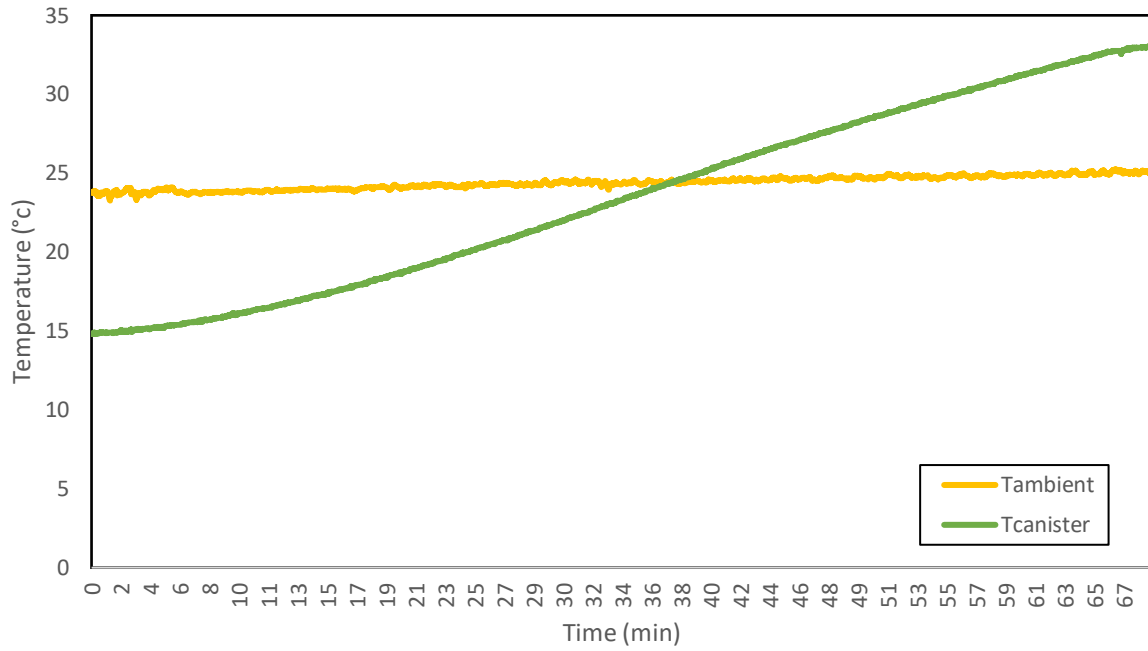


Figure 17: Temperature evolution of the canister during adsorption.

Monitoring the operation of the electrolyser we notice a constant flow rate of 2,125 l/min. The charging processes are all made with canisters at a low level of charge after utilization (p=2 bars).

In Figure 18 we have the same process but with a lower starting temperature, we start from 6,2 °C and we finish at 29 °C, the charging time is 72 minutes, and is higher than the previous case due to a lower operative temperature.

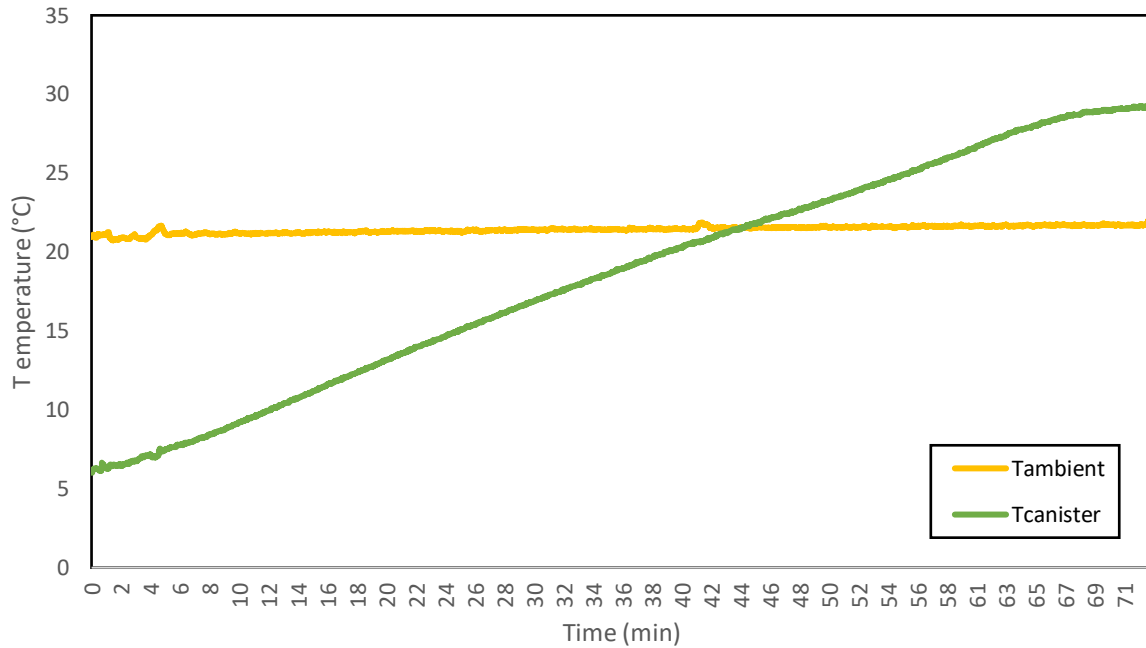


Figure 18: Temperature evolution of the canister during adsorption

3.2 Temperature evolution of the canister during desorption process

3.2.1 Dehydration of the fuel cell

Fuel cells can be affected by many problems during operation, for this reason in the research a lot of effort is made to improve the ability to perform diagnosis to the fuel cell. Some of the problems that PEMFCs can experience are: contamination of the membrane related to gas impurities, starvation of oxygen and hydrogen, leakages, cracking of the membrane, flooding, and membrane dehydration. A great percentage of the faults that can happen are related to hydration level of the membrane.[32]

There are some conditions that enhance this phenomenon: low electric current levels allow a low amount of water formation, and higher mass flow rates of what sprayed out that low amount of produced water. Another thing is that with higher values of stack temperatures we have an increase of the evaporation rate, and we are aware that the fuel cell suffers the increase of temperature during operation.

We experienced a decrease of more of 50% of the performances of the fuel cell, we tried to rehydrate the fuel cell because we hypothesized that this was the problem. But even if we tried to do that the performances did not change, and we were obliged to work with a maximum power of 200 W and with an efficiency usually around 15%.

3.2.2 Desorption process

In **Error! Reference source not found.** is reported the evolution of temperature of one canister in ambient air, during fuel cell operation at 200 W, we will do all the desorption processes operating with this value of power in the following paragraphs.

The canister starts from a temperature of 27.8 °C and finishes at 10.6 °C, the duration is of around 32 minutes. The test is stopped when we arrive in a condition in which the fuel cell is no longer able to work at 200 W because the canister is not able to provide enough hydrogen. The ambient temperature in this case remained almost unchanged.

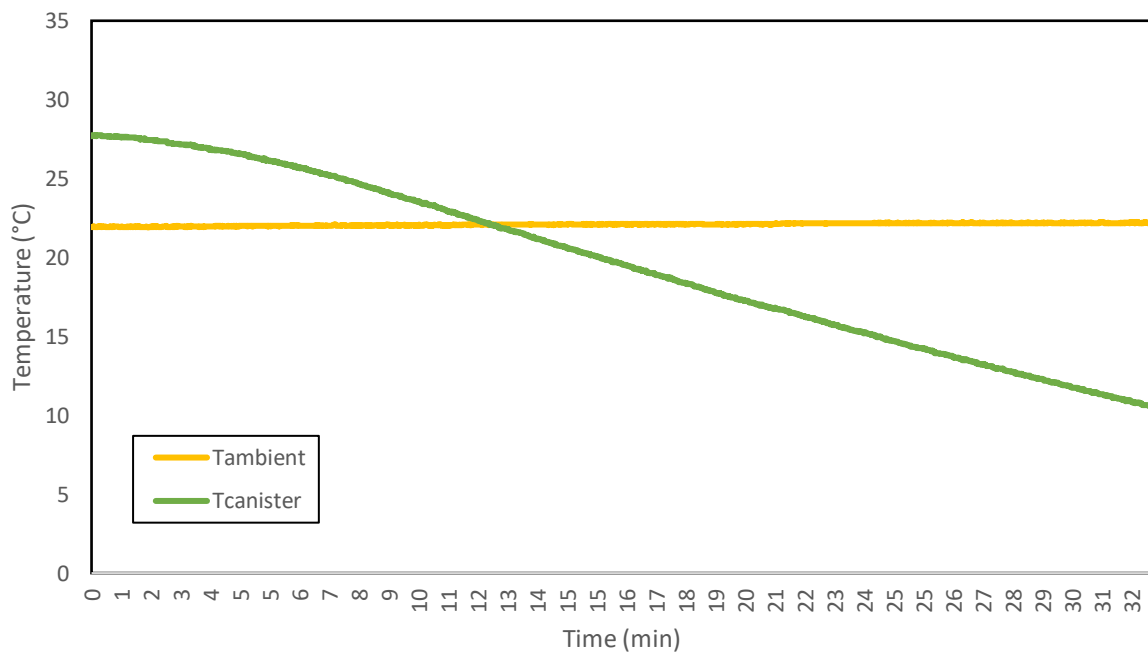


Figure 19: Temperature evolution of the canister during desorption

4 Configuration Improvements

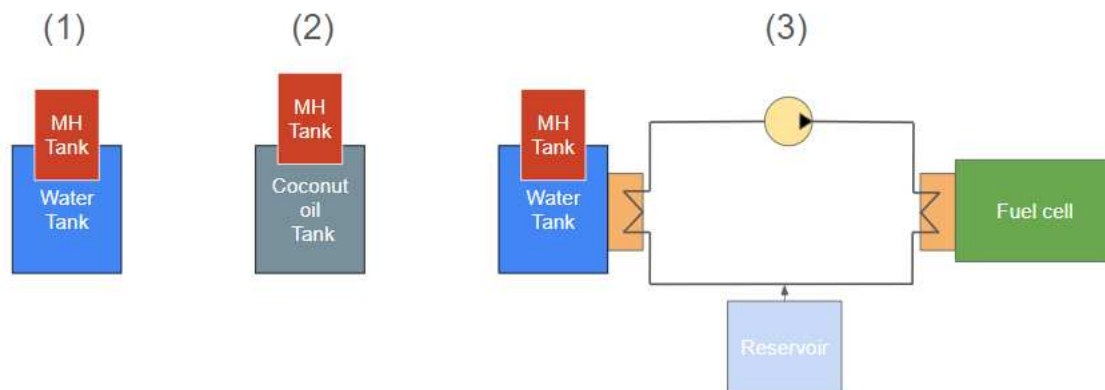


Figure 20: Different configurations proposed

- 1) The first configuration improvement that we propose is to put in contact the MH canisters with water.
- 2) The second improvement that we propose is to put in contact the MH canisters with coconut oil PCM.
- 3) The third improvement that we tried to implement is to thermally couple the fuel cell with the MH canister water tank.

4.1 Water contact with the metal hydrides canister

A first approach that we tried to improve the system performances, is to put the canisters inside a box filled with water. Being in thermal contact with the water we exploit the high thermal capacity of water to maintain the temperature of the canisters much more stable. We added 11 liters of water to the canister box. As it is possible to see, the canisters are not completely submerged, we have a height of water around 15 cm on a total height of the canister (red part) of 23 cm. We still have a portion of the canisters in contact with air.

The following picture shows the canisters inside water.



Figure 21: Canisters inside water

4.1.1 Adsorption process in presence of water

In Figure 22 it's possible to see the evolution of temperature of the canister during the adsorption process, and the relative evolution of water's temperature. We see that the temperature evolution of the canister is much less steep than the case without water and the maximum temperature reached is around 24.5 °C instead of reaching values over 30 °C. The time needed for the process, using the same hydrogen flowrate from the electrolyser of the previous case is around 94 minutes, we notice that is much longer than the previous case (circa 70 minutes).

In this case the addition of water can be seen as disadvantageous because we need the process of adsorption to be as fast as possible.

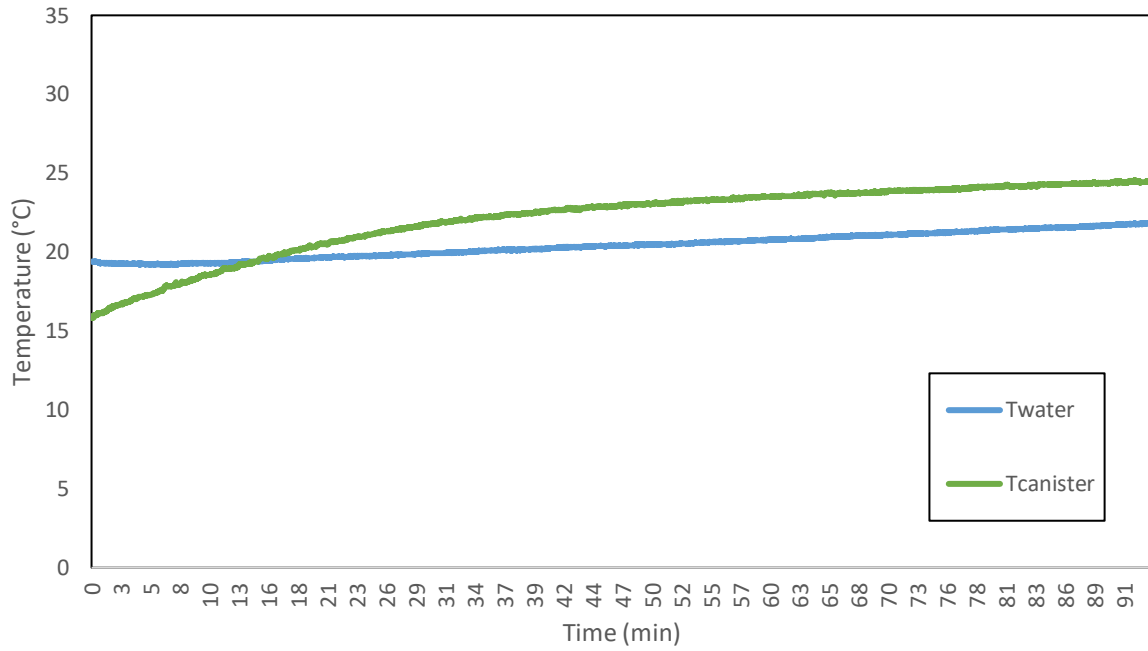


Figure 22: Temperature evolution of the canister inside water during adsorption

4.1.2 Desorption process in presence of water

In Figure 23 I reported the evolution of temperature of one canister during discharging at 200 W of operation. In this case we are able to obtain a much longer operation with respect to the base case (just with air), with an increase of time of 62.5%. It is possible to notice how the temperature is much more stable and the variation of it during the process is just of 6.6 °C instead of 17.2 °C of the previous case.

The addition of water in this case allows a very huge advantage for the system.

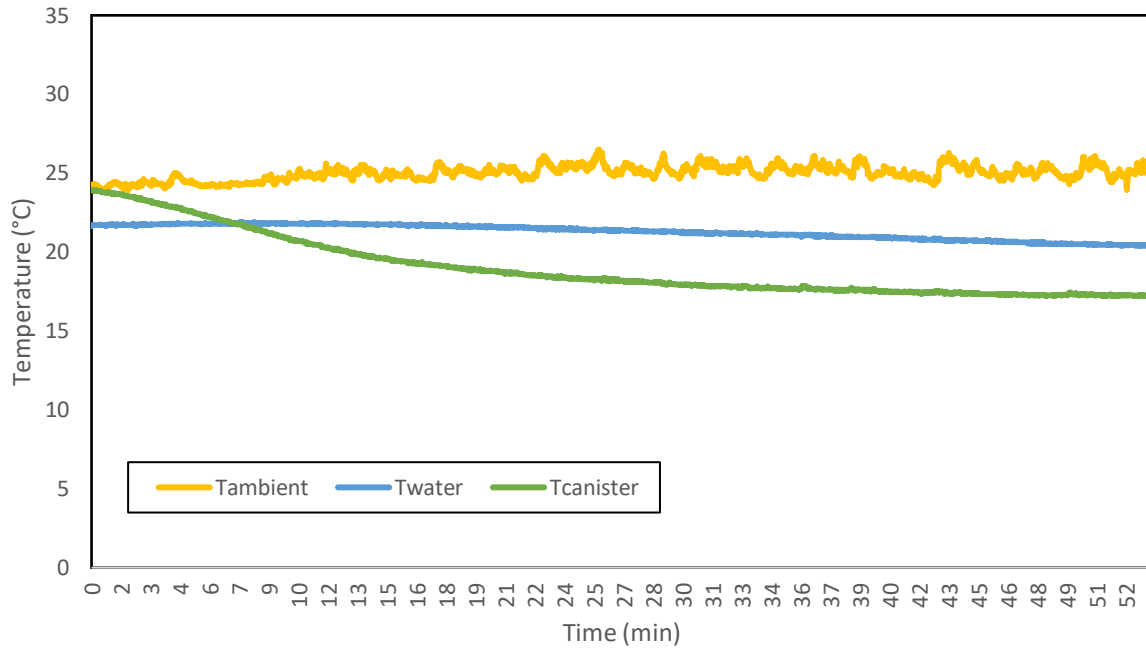


Figure 23: Temperature evolution of the canister inside water during desorption

4.2 Coconut oil contact with metal hydride canister

4.2.1 Phase Change Materials

Phase change materials are substances that when they reach the phase change state can pass from one state of matter to another by absorbing or releasing heat. The most common transition phases used are the transition from liquid to solid and vice-versa. The energy released or absorbed to perform the phase change, that is called latent heat, is generally much higher than the sensible heat of the substance. The general behavior is that when you provide energy to them, you increase their temperature until the phase change temperature is reached, at this point you continue to provide heat and their temperature do not change until the change of phase, this also happens for the heat release for the opposite phase change.

This feature makes them very useful heat storages, because exploiting their phase change they can store or release high quantity of heat without a consequent large increase of temperature.[33]

They can be divided in different typologies with respect to their characteristics:

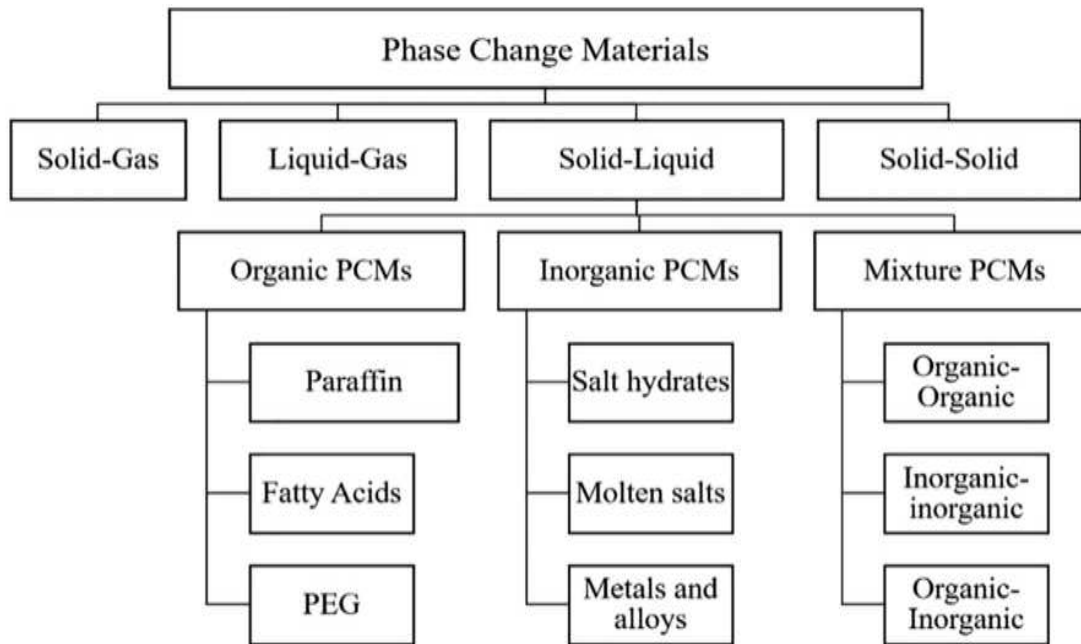


Figure 24: Phase Change Materials categories [34]

During operation, the metal hydrides tanks release heat during adsorption, and absorb heat during desorption. The heat released and absorbed are exchanged through the surrounding ambient air in the general configuration. During desorption the temperature of the canisters decrease, during absorption it increases, and the two phenomena lose kinetics and velocity due to the decrease of temperature. We want to implement a phase change material in the system to make the canisters' temperature more stable and ensure better performances. The way in which we do that is to put in contact the canisters with the PCM, in this way it acts like a latent energy storage for the canisters, absorbing heat from the canisters during adsorption, and releasing to them during desorption.

To do that we have considered to use coconut oil, it is bio PCM made of unsaturated fatty acids[35]. It is a very cheap material, easy to obtain, non-corrosive, chemically stable. The most important feature is its melting temperature, for our purpose we needed a PCM with a melting temperature near to ambient air, the coconut oil has a value around 25 °C. The idea is that we can provide heat to the oil during adsorption and go over 25 °C, and during desorption ask for heat but having the phase change to overcome, in this way we want to keep the temperature of the system as high as possible.

4.2.2 Insulation box

During the experiments made with coconut oil we put the system into an insulated box to decrease the losses to the environment, in order to better maintain the heat that we are providing to the oil. We used polystyrene with a heat conductivity of circa 0.01 W/m²/K for the lateral and bottom surface and to cover the system a square piece with heat conductivity of 0.0146 W/m²/K.



Figure 25: Insulated canisters into coconut oil

4.2.3 Adsorption Process in presence of Coconut oil

For this part of the experiment, we used 7.6 kg of coconut oil that was the amount required to cover the canisters as much as possible. In Figure 26 is possible to see the configuration. In this case the temperature of the oil is around 18 degrees, and it is completely solid. During the adsorption process we have a release of heat, and we obtain the configuration of Figure 27, in which is possible to notice the melted oil around the canister. (in this specific case was working also the other canister for a test so there is coconut oil melted around the other canister)



Figure 26: Canisters into coconut oil in solid state



Figure 27: Canisters into coconut oil after desorption

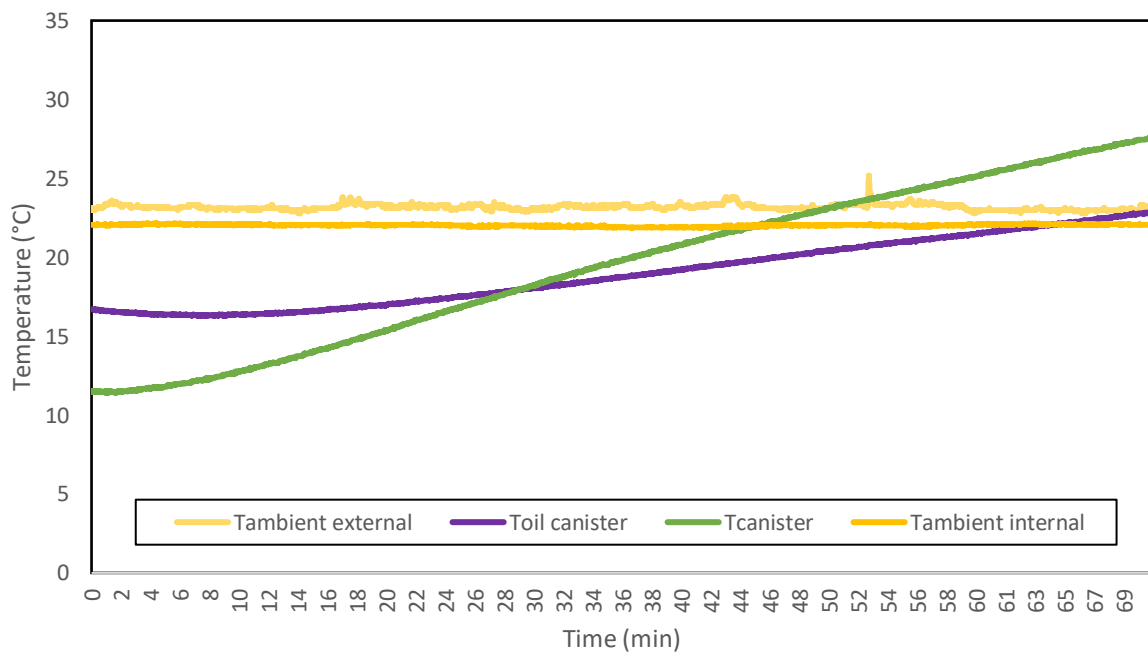


Figure 28: Temperature evolutions during adsorption process with coconut oil

In Figure 28 is possible to see the evolution of temperature of one canister during adsorption, it is evident that even if we use the coconut oil, the adsorption time remains almost unchanged from the case without fluids. (Here 71 minutes as in the base case) The starting temperature of the canister is around 11.5 °C and it ends up at 27.5 °C. With respect to the case with water ($\Delta T=18$ °C), here the ΔT is still high, 16 °C, for this reason we have a very similar trend.

4.2.4 Desorption Process in presence of Coconut oil

In this section we performed the discharging process with the coconut oil, always providing a power of 200 W. The duration time of the process is around 38 minutes that is an increase of 18.8 % with respect to the base case (with air). The canister starts at a temperature of 26.2 °C and finishes at 12.8 °C with a ΔT of 13.4 °C. (Base case 17.2 °C, 32 minutes)

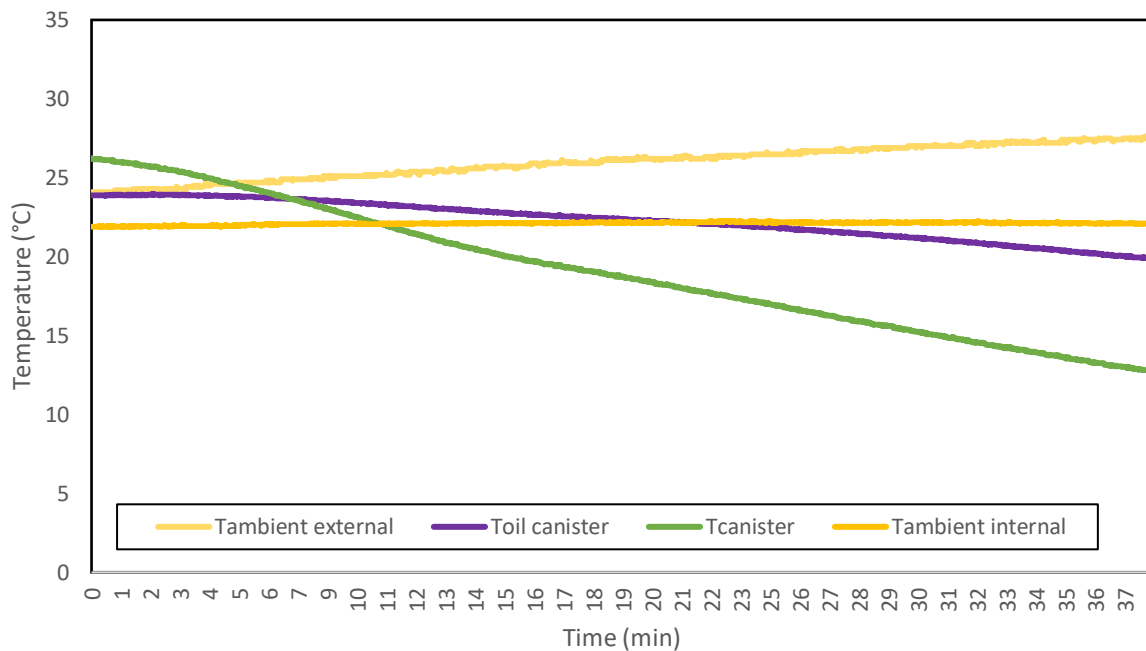


Figure 29: Temperature evolutions during desorption process with coconut oil

4.3 Fuel cell and Metal hydrides canister coupling

In this last part, we tried to implement a final configuration to decrease the external temperature of the fuel cell during operation and exploit the heat generated to heat up the water in which the canister is submerged. We did that constructing a simple water circulation circuit. We used a pump for the water circulation, 3 plastic tubes with an external diameter of 1.2 cm, two heat exchangers of 5,7cm x 5,7cm of dimension, we also used a reservoir to inject the water into the circuit.

In **Error! Reference source not found.** is possible to see a simple scheme of the system.

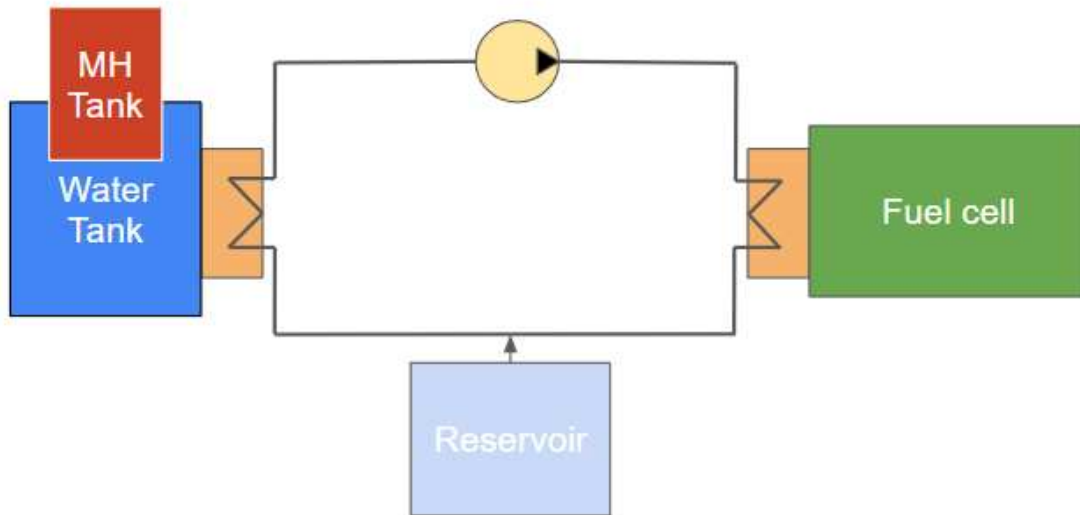


Figure 30: Water circulation system scheme

4.3.1 Adsorption process with water and the HE

In Figure 31 we see the same adsorption process but with water with a starting temperature of 22.6 °C instead of 19.3 °C (as in the case without HE), in this case the process is faster, 85 minutes instead of 94, but still slower than the case without water (70 minutes). The canister experiences a ΔT of just 6,7 °C.

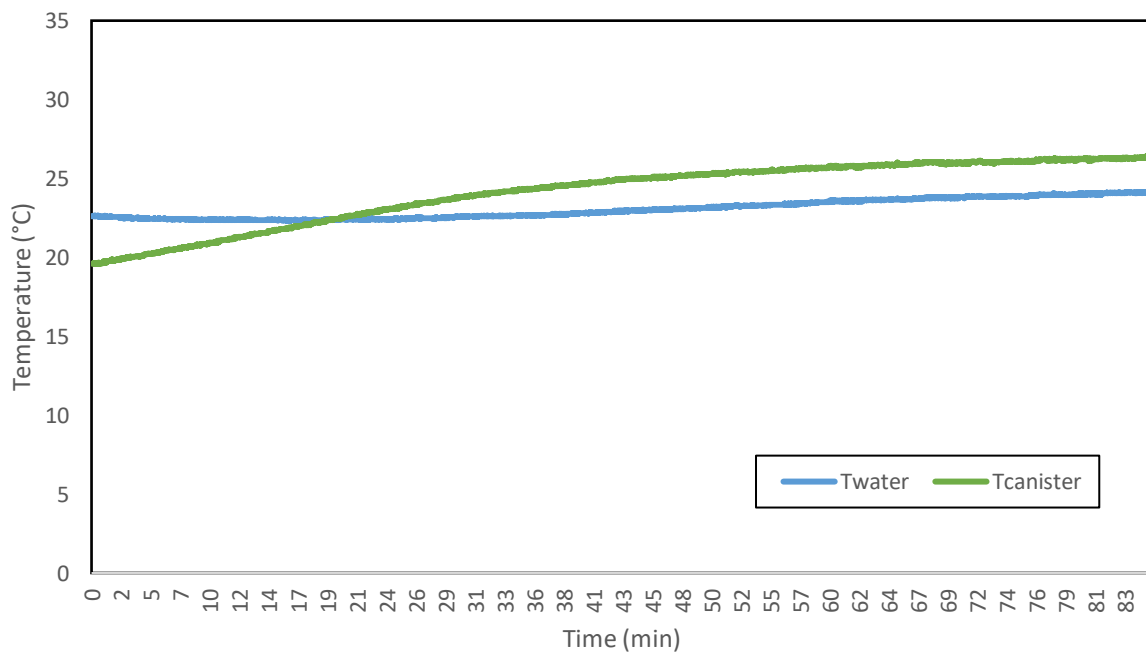


Figure 31: Temperature evolutions during adsorption process with water and HE

4.3.2 Desorption process with water and the HE

In Figure 32 we see the evolution of temperature of 1 canister during discharging. We see how the temperature of the water remains very stable, and the canister experiences a ΔT of just 6.8 °C during the entire process.

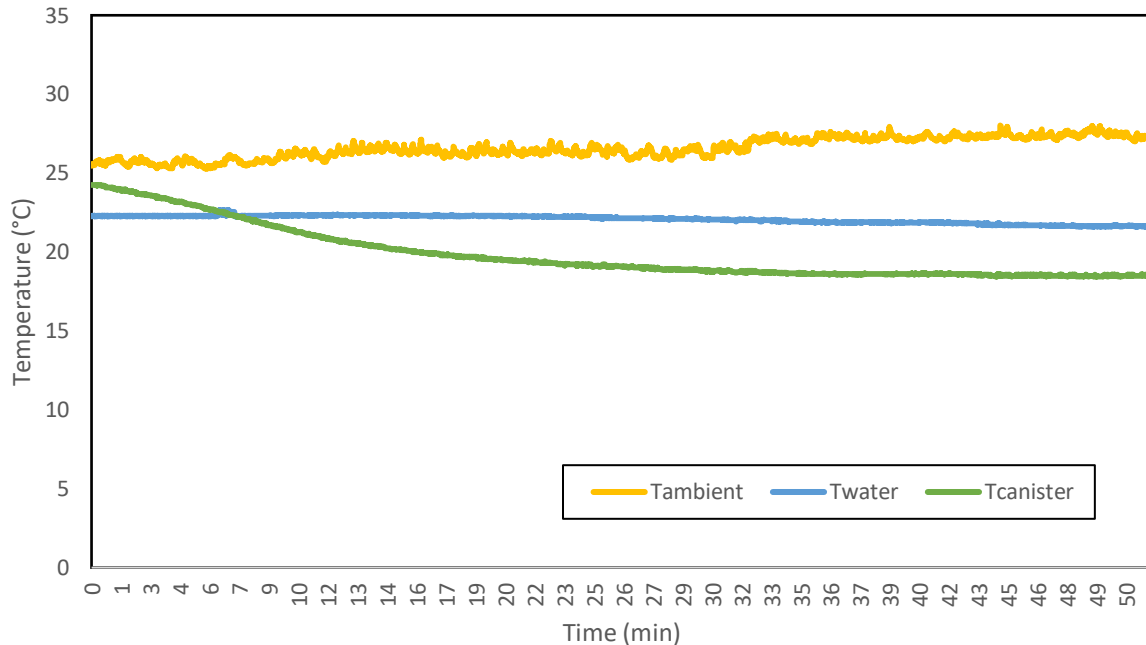


Figure 32: Temperature evolutions during desorption process with water and HE

4.3.3 Efficiency, Internal and External temperature of the fuel cell

The objective of this configuration was both to maintain a more stable temperature of the water, and to decrease the internal temperature of the fuel cell to decrease the amount of energy required for its cooling from the fans, and in this way to increase the efficiency of the fuel cell.

In Figure 33 is reported the evolution of temperature of the external face of the fuel cell in the case without the heat exchanger system (Text FC), and the evolution of the external temperature of the fuel cell in the case with the heat exchanger (Text FC with HE). It is possible to notice how at steady state operation we pass from a temperature of 42.6 °C to a temperature of 37.2 °C.

This difference unfortunately does not allow us to also decrease the internal temperature of the fuel cell (Figure 34). We see in this figure the internal temperature evolution of the fuel cell, in the case with the heat exchanger (Tint FC with HE) and the case without (Tint FC).

Because of this we do not have any appreciable effect on the efficiency.

To solve this problem, it's possible that we would have implemented a higher surface of heat exchangers to further decrease the external temperature of the fuel cell possibly on every face.

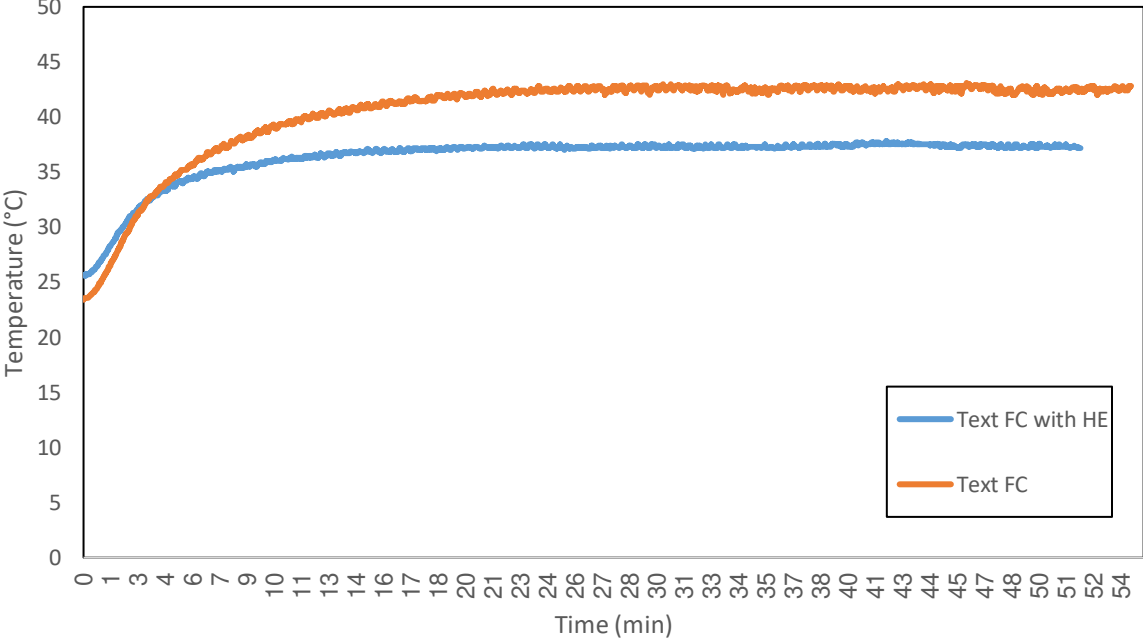


Figure 33: Temperature evolution of the fuel cell during operation

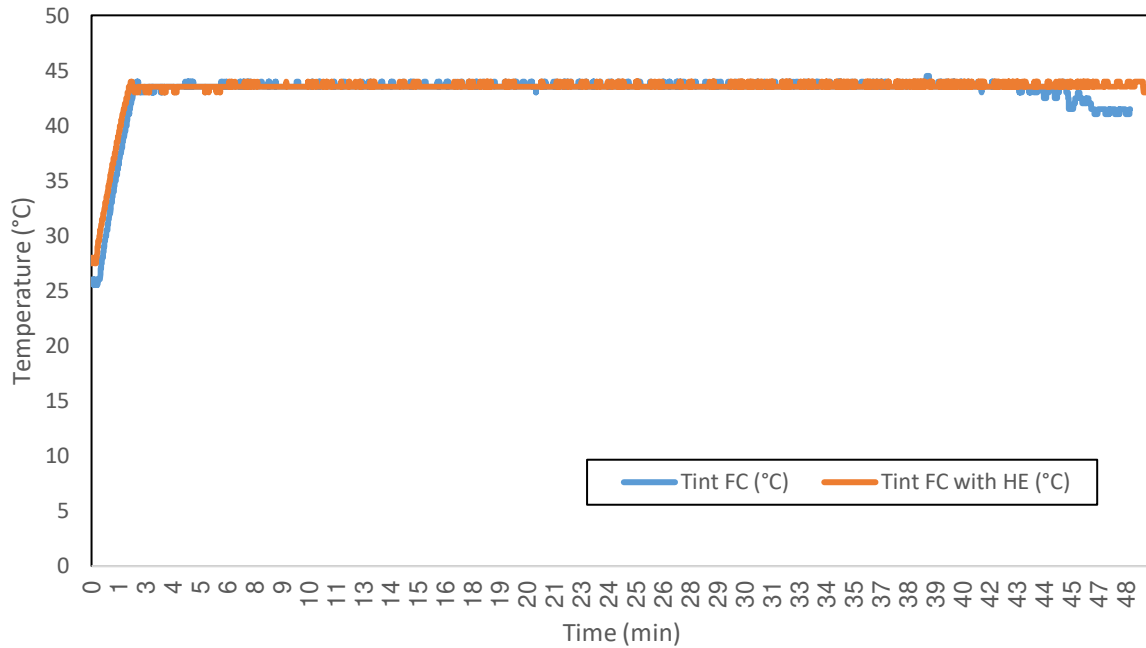


Figure 34: Internal Temperature evolution of the fuel cell during operation

4.4 Results comparison

4.4.1 Adsorption process considerations

In **Error! Reference source not found.** are listed the behaviors in all the different configurations. I considered the initial and final temperature of the canister during the process, the ΔT to compare the temperature variation, and the duration of the process. It is possible to notice that in the case of adsorption, the best results are the one without any improvement, the base case gives the fastest adsorption process. The case with water shows the worst behavior in this case because to complete the charge process a much higher time is required (+38%). The other 2 cases are not as negative on the process duration, but they imply anyway a slowdown of the process.

The variation of the delta T seems to have an effect on the duration of the process if we consider the case with water and with coconut oil we have that a decrease of the delta T implies a longer duration of the process. Coconut oil with 16 °C of delta T instead of 18°C of the base case registers a duration increase of 3%, and water with a delta T of only 8.6 °C shows an increase of duration of 38.2 %. This relation is not confirmed by the behavior of the case with water and heat exchanger, in which we obtain an increase of 25% even if the delta T decreases with respect to the other cases (6.7 °C), so we would have expected a further increase of the duration of the process.

	T (°C) (t=0)	T (°C) (t=end)	ΔT (°C)	Time (min)	Duration Increase (%)
Base case	15	33	18	68	-
With Water	15.9	24.5	8.6	94	+38.2
With Water (HE)	19.6	26.3	6.7	85	+25
With Oil	11.5	27.5	16	70	+3

Table 2: Resume of the Temperature evolutions of the adsorption process

4.4.2 Desorption process considerations

In **Error! Reference source not found.** are reported the same quantities of the previous point. In this case the base case is the worst in terms of duration. We obtain excellent results with the water addition to the canister box. It is possible to notice how the variation of temperature of the canister in the cases with water is very small, this characteristic allows to obtain much better results in terms of duration (+62.5%). Is possible to link this value to the longer process of adsorption in this case, that is possible that allows us to store more hydrogen and, in this way, to have a longer process of discharge. The case with water and heat exchanger provides the best results, but also in this case the difference with the case with water it's not that relevant. The coconut oil gives a small improvement to the duration (+18.8%) of the process but compared to water it is not advantageous.

	T can (°C) (t=0)	T can (°C) (t=end)	ΔT (°C)	Time (min)	Duration Increase (%)
Base case	27.8	10.6	17.2	32	-
With Water	23.9	17.3	6.6	52	+62.5
With Water (HE)	24.3	18.5	6.8	53	+65.6
With Oil	26.2	12.8	13.4	38	+18.8

Table 3: Resume of temperature evolutions of the desorption process

4.4.3 Hydrogen Useful capacity

In Figure 35 is reported the hydrogen flowrate provided to the fuel cell during operation in the four different configurations.

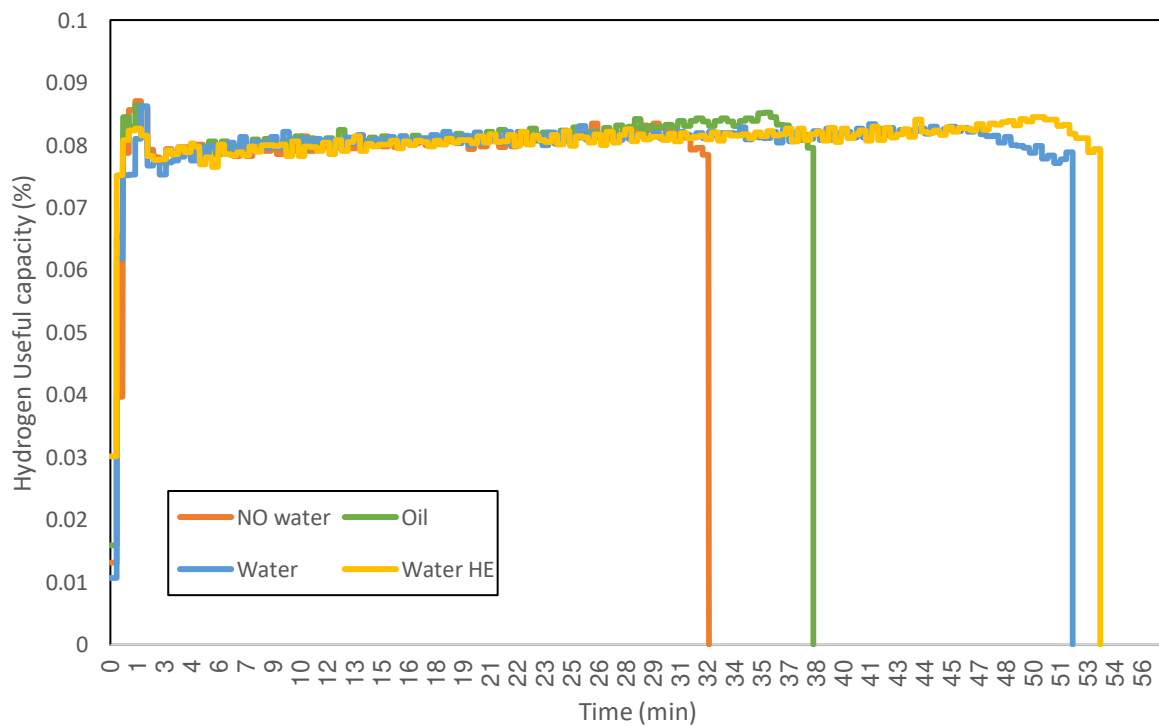


Figure 35: Hydrogen useful capacity of the different configurations

It is possible to notice the huge difference of operational time between the base case (No water curve) and the cases with water (water curve), from around 32 minutes of duration of the first case to around 51 minutes of the case with water. To compare the configurations, I calculated the hydrogen useful capacity in every case, considering 600 liters as the maximum amount of hydrogen that one canister can contain, and the results are reported in Table 4. Performing the integral of every curve we can obtain the amount of hydrogen provided to the fuel cell in every case. From the table is possible to notice how the case with the use of water provides a huge advantage in terms of useful capacity of hydrogen achieved, passing from a value of 26.3% in the base case to a value of 42.8% in the case with water. Also, the case with coconut oil provides an improvement but not as significant as the case with water. The case with the implementation of the heat exchanger allows to obtain the highest value of hydrogen capacity, but if compared to the value with only water we have an improvement of just 1.4%, this small enhancement does not justify the cost to build the water circulation system for the heat exchanger, so the best case remains the one with just water.

	Hydrogen Useful capacity
Base Case	26.3 %
With Water	42.8 %
With Water (HE)	44.2 %
With Coconut Oil	31.5 %

Table 4: Hydrogen useful capacity

4.4.4 Interpretation of the results obtained

We have three situations, with the MH canister in contact with three different materials, air with a thermal conductivity of 0,026 W/m/K[36], water 0,6 W/m/K[37], and coconut oil 0,166 W/m/K [24].

As treated in the article of Yang Ye et al. [38], during adsorption, that is an exothermic reaction, the rise of temperature causes the restriction of the reaction process if the adsorption heat is not discharged effectively. Indeed, during an exothermic reaction, an increase of temperature shifts the equilibrium to the reactants, making the reaction slower. Performing a comparison between the case with water (case 2) and without (case 1), this phenomenon can appear to be not confirmed, considering our results. In case 2 we obtain a much slower adsorption process with respect to case 1 even if in this last case a higher temperature is reached.

The equilibrium pressure of metal hydrides is influenced by the temperature, increasing the temperature increases also the equilibrium pressure[39]. With a higher value of this quantity at the same pressure we are able to store less hydrogen, this can partially explain why the adsorption process in case 1 takes less time, because when we reach the value of 16 bars in the canister, that is the maximum value, in case 1 we have less hydrogen in comparison to case 2 at the same pressure, due to the difference of temperature between the two cases. In this way, having the possibility to store less hydrogen, the process will also be faster, this would explain why, after adsorption in case 1, the desorption lasts less than case 2, because we are able to store less hydrogen in case 1. In any case the temperature difference between the two cases during adsorption is around 10 °C, is not clear how much a difference of this magnitude can influence the storage capacity.

During the process of adsorption, we have a higher temperature increase in case 1 than in case 2, the increase of temperature difference enhances also the heat exchange. But in any case, this increment of temperature difference between the canister and the surrounding fluid does not counterbalance the higher thermal conductivity of water with respect to air, so it's not possible to say that in case 1 we obtain a better heat exchange because of the higher temperature difference reached.

The heat exchange also during the desorption process, in case 2 is more effective due to the higher thermal conductivity of water with respect to the one of air, in this way the endothermic reaction receives more effectively the heat from the water, and this allows the process to last longer. This is a possibility to explain the longer duration of the desorption process in case 2.

The same considerations can be made also for case 3 (with coconut oil), we obtain results that are between case 1 and 2, due to the higher thermal conductivity of coconut oil with respect to air and lower with respect to water, that explains why the phenomena are much less evident in this case.

Before our study we considered to use coconut oil to exploit its phase change temperature to maintain the temperature of the system more stable, but the lower temperature difference that is obtained using this phenomenon, causes a decrease of the heat exchange between the coconut oil and the canister, without any considerable advantage.

Considering also the results obtained in the article of Jianhui et al.[40], we see how the usage of a water cooling system, with respect to a natural convection and air cooling system allows to significantly decrease the absorption time. Unfortunately, this does not help to give an explanation to our results.

5. Conclusions

The aim of this work was to try to enhance the thermal properties of the system in order to obtain better characteristics in terms of durability of the processes, hydrogen useful capacity of the metal hydride canister and to increase the efficiency of the fuel cell.

To achieve that we have performed an analysis of the thermal behavior of the system using an experimental approach. We have used a set of different instruments to obtain the evolution of temperature of our system during operation to try to find some trends or correlations related to the different heat exchange configurations analyzed. The objective was to find a configuration that shows a better and faster exploitation of the hydrogen stored and to further understand the mechanisms of heat exchange that are involved in the processes. This goal was partly achieved but the phenomena involved in the process would need further study to be understood with higher level of detail.

From the data that we have obtained is possible to say that in the case of adsorption none of the improvements that we tried was beneficial for the system to decrease the duration of the process, the best configuration in these terms remains the one without any addition in terms of duration. But in any case, a possible explanation of this is that the adsorption process in the base case takes less time because it achieves a lower level of hydrogen stored, this is due to a poorer heat exchange and a higher temperature reached during the process. This aspect needs further in-depth analysis because unfortunately with our instruments we had just the possibility to know the pressure reached inside the canister, and not the exact amount of hydrogen stored.

For what concerns the desorption process, we obtained an improvement in every configuration that we tried, in terms of duration of the process and useful capacity of hydrogen reached. It is noticeable the improvement of the duration time of the process in the case with water (+62.5% with respect to the base case), using a very simple and cheap configuration we were able to improve significantly the system. The coconut oil configuration does not give a gain comparable to water that, in the context of this study for its cheap cost, its availability and performances, remains the best option in the desorption case (coconut oil in any case gave an increase of the time duration of +18.8%).

The configuration of water in the canister box added to the heat exchange circuit coupled with the fuel cell gave the best results in terms of desorption, both in the duration of the process and useful capacity of hydrogen, but implementing our configuration, the improvement was not very significant with respect to the case with just water, and so the construction of the heat exchanger system is not completely justified. We did not obtain the desired effect on the efficiency of the fuel cell, for a further improvement it would be interesting to implement a bigger heat exchanger circuit to decrease more the overall temperature of the fuel cell during operation. In this case it would be also necessary to be studied the effect related to a higher delivery of heat to the water inside the canister box, indeed a too high

temperature of water will have a negative effect on the heat transfer characteristics of the system. It would be crucial to find a optimal configuration to decrease the temperature of the fuel cell but to not increase too much the temperature of the water inside the box.

It would have been very interesting to analyze the system in different heat transfer conditions, for example using a water circulation system to provide to the storage always water at a low temperature to obtain a more efficient heat exchange, or to analyze the system in presence of many other PCMs with a different heat conductivity to obtain a comparison in a wider range. Or also to vary the amount of PCM and water that was used during the experiments.

To conclude, we obtained some interesting results, but a lot of different configurations can still be implemented starting from this preliminary analysis to better understand the functioning of the phenomena involved in the process.

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