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Different forms of hydrogen production: a review and future perspectives

Diferentes formas de produção de hidrogênio: uma revisão e perspectivas futuras

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Abstract: There was a significant increase in the concern with climate issues, among them highlighted as the derivation of greenhouse gases from the burning fossil fuels, leading several research centers and researchers to seek new sources of less polluting energy, independent of the burn-based matrix of fuels. In this context, the present paper has as main goal a literature review, perspective and comparisons regarding the use of hydrogen as a clean energy source, presenting three main ways of obtaining it: a) through electrolysis using renewable sources; b) biohydrogen production, based on the photosynthesis of plants and algae; c) production through biodigesters.
Keywords: Green hydrogen, biohydrogen, biofuel, renewable sources, clean energy.

***Resumo**: Houve um significativo aumento na preocupação com questões climáticas entre elas destacadamente com as emissões dos gases do efeito estufa provenientes da queima de combustíveis fósseis, levando vários centros de pesquisa e pesquisadores a procurarem novas fontes de energias menos poluentes e principalmente independentes da matriz baseada em queima de combustíveis. Nesse contexto o presente artigo tem como principal finalidade apresentar uma revisão de literatura, perspectiva e comparações quanto à utilização do hidrogênio como fonte de energia limpa apresentando três formas principais de obtenção do mesmo: a) através de eletrólise utilizando fontes renováveis; b) produção de biohidrogênio, com base na fotossíntese de plantas e algas; c) produção através de biodigestores.*
Palavras-chave: Hidrogênio verde, biohidrogênio, biocombustível, fontes renováveis, energia limpa.

1. Introduction

Much has been said about climate change caused by global warming and greenhouse gases. These problems are totally linked to the world's energy consumption. With the growing concern about all these environmental problems, ideas emerged for the use of biofuels that have low or no carbon emissions.

It is known that polluting gas emissions are caused by several factors, including the fossil fuels burning. These fuels, in addition to being non-renewable, have many negative impacts on the environment and for the health of all living beings. In addition to the polluting emissions generated by their burning, which causes great air pollution, fuel leaks and spills can also occur, also causing water pollution.

When talking about the use of fossil fuels, one of the most polluting sectors is transport. In 2016, transport was responsible for 15,9% of greenhouse gas emissions, second Only to the electricity and heating sector, which accounted for 30,4%. Road transport represents about 11,9% of total emissions in this sector (IEA, 2018). Automotive vehicles of light or heavy category are responsible for a large part of the emissions generated, which are produced from the combustion process and incomplete fuel burning. These emissions are composed of toxic substances, such as nitrogen oxide (NOx), carbon oxides (CO, CO_2), sulfur oxides (SOx), hydrocarbons (HC), among others (Teixeira et al., 2008). It is for this reason that the technologies for using hydrogen as an energy source and as a fuel have a very important role to play in this sector. The hydrogen combustion does not generate greenhouse gases and is free from harmful emissions emitted by conventional gasoline and diesel vehicles. Its combustion forms water and a small amount of NOx. Vehicles powered by hydrogen promote better air quality and are already used in many countries, such as Japan, South Korea, China and some countries in Europe.

The world urgently needs to implement concrete actions and make a profound change in energy supply and consumption, using renewable sources, with low or zero carbon content. The transition to a clean, low-carbon energy system can be significantly made possible by hydrogen and fuel cell technologies.

Hydrogen is considered the fuel of the future. In addition to being renewable and non-polluting, it has the highest amount of energy per unit of mass compared to any other fuel known today: around 52,000 BTU per pound (Vargas et al., 2006). It is a versatile, clean and safe energy carrier that can be used as a raw material in industry or as a fuel. It can be transported in the same ways as fossil fuels, through trucks, in liquid or gaseous state, transport in ships and pipelines. It can be burned or used in fuel cells to generate heat and electricity.

Fuel cell technology using hydrogen has a low environmental impact, as it does not emit polluting gases or particulates, there is no vibration or noise. This technology has the basic principle of a battery, which transforms chemical energy into electrical energy. According to Reitz (2007), a fuel cell can reach 80% efficiency, whereas traditional combustion engines reach 40%.

2. Biohydrogen production

The photobiological hydrogen production, or biohydrogen or green hydrogen, is based on the photosynthesis process of plants and algae. Naturally, only bacteria and algae are capable of producing hydrogen. Currently, the most studied are anaerobic bacteria, photosynthetic bacteria, cyanobacteria (blue algae) and green algae (Miyake, 1999). Several types of green algae and cyanobacteria are capable of producing hydrogen through catalyst enzymes they produce. They have capacity to fix nitrogen in the atmosphere and CO_2 through photosynthesis. In cyanobacteria, there are three types of enzymes: nitrogenase, assimilation hydrogenase and bidirectional hydrogenase (Tamagnini et al., 2002; Sacramento et al., 2006). In a high oxygen environment, the nitrogenase can have its activity inhibited.

When we need to develop new processes for obtaining products that have had problems for some time, nothing better than observing nature. Proof of this is that it has been producing energy from water and the sun for a long time. In this sense, several researchers gave up on inventing machines and processes based on obsolete models and that have been shown to be harmful to the environment, looking for new ways to produce energy in a simpler and more efficient way, only imitating nature. Biohydrogen is an expanding technological area mainly due to the fact that it is about clean production from renewable sources (Cheong, 2006). When production is carried out in the laboratory, under controlled conditions, it is possible to achieve the same efficiency as photovoltaic systems. The difficulty is to produce with such efficiency in open places, outdoors, because in high light intensities, directly under sunlight, there is an excessive absorption and a waste of energy (Benemann, 1997). The production of hydrogen through this biotechnological means promises to be a very interesting and viable alternative in the acquisition of new energies, as it presents minimal production costs and required characteristics such as those of renewable energy (Ming, 2002). When we compare this process with thermochemical and electrochemical processes, it has the characteristic of being friendly to the environment in the sense of not causing irreversible or even temporary damage, which is why it is considered clean energy. (Veziroglu, 2001).

Research into the production of hydrogen from microbacteria and micro or macro algae began in the early 1980s (Demirbas, 2008; Zhi, 2008; Li, 2007). Green algae can produce hydrogen after a period of anaerobic conditions and in the absence of light, through the hydrogenase enzyme that is activated and synthesized. When these algae return to light, there is an increase in the rate of hydrogen (Das, 2001). We can classify obtaining hydrogen in this way into two large groups. One of them is obtained mainly from light and the other can be present in the absence of light (Kotay, 2008).

The first large group is characterized by the production of photobiological hydrogen by photosynthesis of microorganisms and has shown to be very promising due to the generation of clean energy, thus Biophotolysis can be described as the action of light on a biological system resulting in the release of water, to produce hydrogen (Das et al., 2008). The process resembles reverse photosynthesis. Present in green plants, reducing carbon dioxide, photosynthesis made by microalgae, due to the presence of enzymes such as hydrogenase and nitrogenase, producing hydrogen under appropriate conditions (Sacramento, 2007).

There are basically two types of biophotolysis available: direct and indirect (Sacramento, 2007). Direct biophotolysis is a biological process where photosynthetic microalgae systems use solar energy, converting it into chemical energy, decomposing water and hydrogen (Das et al., 2008). This technology is promising and particularly interesting since solar energy is used to convert into oxygen and hydrogen, a component that, despite water crises, is still abundant: water. In indirect biophotolysis, cyanobacteria are by far the most studied and widely used microorganisms in laboratory studies. (Sacramento et al., 2006). The technique is based on the knowledge that nitrogenase is very sensitive to oxygen, thus developing strategic defense mechanisms for protection from atmospheric and intracellular oxygen in the photosynthesis process. Thus, with the objective of efficient hydrogen production, researchers try to produce and select species deficient in H_2 assimilation and select those whose bidirectional hydrogenase responds better to the presence of oxygen (Tamagnini et al., 2003).

Photobiological hydrogen production still faces some adversities, such as low solar energy conversion efficiency, low light intensity (where production saturation occurs) or high light intensity (where cell growth and H_2 production photoinhibition occurs). The production process using algae is at an early stage of development. There are many tests performed, but none on a large scale, or energy-efficiently and financially. Currently, there are several countries investing in studies applied to the exploration of photobiological hydrogen producing microorganisms. As the production of hydrogen through algae is dependent on many factors, further studies on this subject are needed (Tamagnini et al., 2003). There is a significant obstacle to photosynthetic activity. In this process, there are two incompatible steps: in the first, water is split to produce oxygen. In the second, there is the production of hydrogen through hydrogenase. As oxygen inhibits hydrogenase activity, feedback inhibition occurs (Benemann, 1996). Every process of obtaining energy has points to be considered, but according to Kandan and Kargi (2006), in this case, the inhibition of the hydrogenase enzyme in the presence of oxygen is one of the most concerning. Some authors cite the fact that no residues are used, that is, there is no factor of reuse of residues in the production of hydrogen, as another aspect that should perhaps be considered. (Kapdan and Kargi, 2006).

The cultivation of algae for production of biofuels, which includes hydrogen, is one of the most promising alternatives compared to other resources. According to Satyanarayana (2011), they have high oil production capacity, high growth rate and require sunlight and CO_2, which makes them more photosynthetically efficient than oilseeds. Also, growing algae has some advantages over growing other crops. The fact is that they do not need large lands that could be used for food production and that they can be cultivated without the use of drinking water. Another advantage is that they have a great variability of habitats (fresh and salt water, frozen lakes, thermal springs) and the ability to survive in different environmental conditions. More strains of cyanobacteria and microalgae should be studied to improve the applicability of hydrogen production. According to Tamagnini et al. (2002), genetic engineering has become an important ally in determining molecular biology techniques when it comes to cyanobacteria.

Microalgae cultivation systems can be classified into two main types: open and closed. Open systems are those in which microalgae are cultivated in contact with ambient air and have little sophistication in terms of control of temperature, light, pH, etc. These systems have lower costs, but their productivity is low, which causes very limited efficiency. Closed systems, which are photobioreactors (PBR), do not have contact with the atmospheric air. They already designed to obtain greater volumetric algae productivity, and consequently have higher operating costs. In closed systems, it’s possible to control several conditions, such as the amount of light, temperature, gas dissolution, among others.

The hydrogen production from biomass composed of algae has the potential to be economically viable and sustainable, as it uses water as a renewable source and also uses CO_2, thus considering an excellent opportunity to make a positive balance of carbon dioxide as it is considered a today's biggest pollutants. When it comes to carbon sequestration, microalgae have the capacity to absorb CO_2 up to 15 times more than tropical forests (Carvalho, 2011). Microalgae are able to use the CO_2 from waste burning to generate energy, which strongly contributes to the reduction of greenhouse gas emissions. At UFPR's Self-sustainable Energy Research and Development Center, located in the city of Curitiba, Brazil, a system was built that handles solid waste treatment, electricity generation, emissions control, biomass production, biogas generation and biofuels, and that uses microalgae (Munõz, 2018). In short, gases from the burning of solid waste are injected into the photobioreactors, which are used for the microalgae cultivation. These microalgae generate biomass, where oil can be extracted for production of biodiesel, and the solid residues not used in oil extraction are used to generate biogas, where hydrogen can be produced. With this system, the environmental relevance of using algae for energy production and carbon sequestration at the same time is remarkable. The Figure 1 exemplifies the use of algae in the CO_2 capture process, where the algae would later be used to produce H_2.

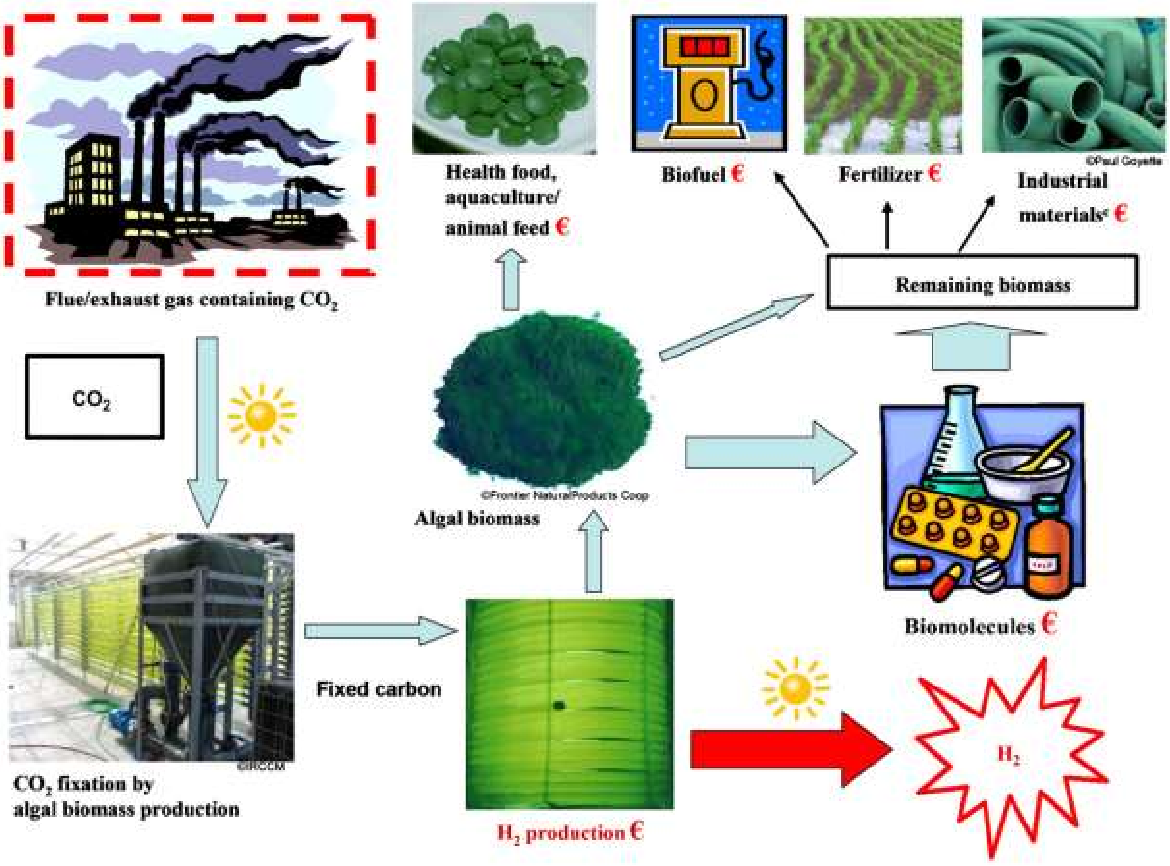

Figure 1. Different uses of hydrogen as an anergy source. Source: Skjanes (2007).

Determining the ideal conditions for the hydrogen production is still an open question, but one that has been studied in the laboratory. Mixed cultures in batch tests show that environments where the pH is very low and where there is a large availability of organic matter can cause a reduction in hydrogen production.

3. Hydrogen production by water electrolysis

3.1 Electrolysis

The electrolysis of water is one of the simplest ways used to produce hydrogen. It has the ability to produce hydrogen using only renewable energy. It produces extremely pure hydrogen (>99.9%), ideal for some processes with high added value, such as the manufacture of electronic components. Water electrolysis also shows good results on a small scale and the process is even more sustainable if the electricity used is derived from renewable energy sources, such as wind, solar, hydro, etc. When abundant energy is available, the extra energy can be stored in the form of hydrogen used in the electrolysis of water. A good use for the hydrogen that has been stored is to be used in fuel cells (FCs) to generate electricity. Therefore, electricity that was generated by renewable energy is directly incorporated into the grid or used to produce hydrogen (Santos et al., 2013).

The water electrolysis process consists in the separation of molecules in the hydrogen and oxygen gases through the passage of an electric current. Current flows between two separate electrodes immersed in an electrolyte to increase ionic conductivity. For this process to occur, it is necessary to use a diaphragm or separator in order to avoid mixing of gases generated in the electrodes (Hamann et al., 2007; Ursúa et al., 2012).

According to Ursúa et al. (2021), the electrodes must present resistance to corrosion, have good electrical conductivity and good catalytic properties, during the process, the electrolyte must not change or react with the electrodes. The diaphragm will serve to avoid short circuits between the electrodes, and must have high ionic conductivity and physical and chemical stability.

3.2 Fundamentals of water electrolysis

Generally, the general electrolysis reaction can be divided into two half-cell reactions: hydrogen evolution reaction (HER) and oxygen evolution reaction (OER). In HER the reaction is reduced at the cathode to produce H2, and OER is the reaction where water is oxygenated at the anode to produce O2. One of the obstacles preventing water splitting from being practical is the slow reaction kinetics of OER and HER due to superpotentials. Researches carried out indicate the use of catalysts as highly effective to minimize superpotentials, for OER and HER (Wang et al., 2021).

The simplest water electrolysis is composed of an anode and a cathode connected through an external power supply and immersed in a conductive electrolyte. Direct current (DC) is applied to the unit, electrons flow from the negative terminal of the DC power source to the cathode, where they are consumed by hydrogen ions (protons) to form hydrogen atoms. In the water electrolysis, hydrogen ions end up moving towards the cathode, the hydroxide ions move towards the anode, while the diaphragm is used to separate the two compartments. The gas receivers are used to collect the hydrogen and oxygen gases, which are formed at the cathode and anode (Santos et al., 2013).

Santos et al. (2013) explains that when water electrolysis occurs in an acidic or neutral aqueous electrolyte, the processes that occur on the electrode surface are Boundary layers on an electrode surface. The described electrolyte reactions are heterogeneous reactions, occurring between the electrode phase (solid metal or carbonaceous material) and the electrolyte phase (aqueous salt solution).

The "interphase" region experiences differences in electrolyte velocity, concentrations of electroactive species and electrical potential with electrode distance. Each of these gradients gives rise to a different boundary layer near the electrode surface. According to Eq. (1) and Eq. (2): (Santos et al., 2013).

$$\text{Cathode } 2\,H^+(aq) + 2\,e^- \rightarrow H_2(g)\ (E0 = 0.00\text{ V } vs.\text{ SHE}), \tag{1}$$

$$\text{Anode } H_2O(l) \rightarrow \tfrac{1}{2}\,O_2(g) + 2\,H^+(aq) + 2\,e^-\ (E0 = 1.23\text{ V } vs.\text{ SHE}). \tag{2}$$

The sum of these two equations leads to the general reaction of the electrolysis of water, according to Eq. (3):

$$\text{Overall } H_2O \rightarrow H_2 + \tfrac{1}{2}\,O_2\ (E0 = -1.23\text{ V } vs.\text{ SHE}). \tag{3}$$

According to Santos et al. (2013) in alkaline water electrolysis where the strong base is used as the electrolyte, the hydroxide anions are transferred through the electrolyte to the anode surface, where they lose electrons, the electrons return to the positive terminal of the source of DC power. Nickel (Ni) is a good choice due to its low cost and easy availability. To increase conductivity, the electrolyte used in the cell must have high mobility ions, potassium hydroxide (KOH) is generally used in alkaline water electrolysis to avoid corrosion problems that are caused by acidic electrolytes. KOH is used more than sodium hydroxide (NaOH) because previous electrolyte solutions have higher conductivity. However, when the process is run in alkaline electrolyte

$$\text{Cathode } 2\,H_2O + 2\,e^- \rightarrow H_2 + 2\,OH^-\ (E0 = -0.83\text{ V } vs.\text{ SHE}), \tag{4}$$

$$\text{Anode } 2\,OH^- \rightarrow \tfrac{1}{2}\,O_2 + H_2O + 2\,e^-\ (E0 = 0.40\text{ V } vs.\text{ SHE}). \tag{5}$$

The sum of Eq. (4) and Eq. (5) will lead to the same general reaction as Eq. (3) with the same value (-1.23 V) for the theoretical cell voltage. For these reactions to occur, barriers must be overcome, which depend on the components of the electrolysis cell: electrode phase, electrolyte phase, boundary layers on the electrode surface and separator and electrical resistances of the circuit (Santos et al., 2013).

3.3 Relationship between hydrogen production and solar energy

Hydrogen has some weaknesses, among them low energy content by volume, high infrastructure cost, very high cost, and high rates of gas emissions. It was said that hydrogen would be the fuel of the future. Solar hydrogen is an excellent and clean fuel, it can replace many fuels such as gaseous and fossil fuels. Thus, it helps the environment with carbon gas reduction, helping to prevent global warming (Romm, 2004).

Solar energy is considered a renewable energy source. It is present all over the world and can be used to generate the electricity it needs for the electrolysis process. The amount of solar energy that is received by the earth is approximately 10,000 times more than the annual energy expenditure. With this, all human needs would be met, if we could efficiently harvest solar energy (Markandya, 2007).

Fuel cell prices have been decreasing with the production of hydrogen and the extra electricity generated with this process is used as a battery to produce more electricity when solar energy is not used. Thus being very cost efficient. However, due to the amount of existing solar energy, a schedule for storing all this reserve of electrical energy is necessary (Gotz, 2016).

Hydrogen generated through solar energy is an excellent system being used together with the electrolysis process. Solar hydrogen can be used in several ways, such as: domestic and industrial use, as a fuel for transport and energy generation in different ways (Gotz, 2016).

The best and most accessible technique to be able to use the solar energy found is the photovoltaic system, which through irradiation can convert solar energy into electricity quickly and directly. Studies show that solar energy must be prepared in a large amount to be stored through methods created to reduce the intermittence of solar energy, after this process can provide electricity continuously (Hosseini, 2019).

Nowadays, when solar photovoltaic systems, used on the roofs of homes generate more energy than the home spends, the extra amount is used for other purposes, outside the home, these applications outside the owner's network are rewarded again in proportion to the energy donated (Tebibel et al., 2017).

Several experiments were carried out with direct and indirect irradiation prototypes in order to create a solar hydrogen process by solar cracking of methane. When metadata is absorbed in infrared radiation, the entire process must be heated in the same spectral range. In direct irradiation tests, black carbon is placed to absorb solar energy and transfer to methane (Abanades et al., 2015).

When using the photovoltaic system to generate electricity, using together the electrolyser with a low temperature, it is one of the most accessible processes to generate hydrogen with solar energy. The production of hydrogen with the photovoltaic system together with electrolysis started in the 70s. Considering several aspects regarding economic and ecological issues, the most viable production of solar hydrogen would be using a photovoltaic current (Bilgen, 2001).

Hydrogen production based on solar energy requires techniques that require additional deployment to produce hydrogen. Solar hydrogen has a great reach due to the technologies used, in theory the lowest cost for the production of hydrogen using solar energy, should be produced in more equipped solar plants, with more efficiency (Steward et al., 2008).

4. Hydrogen production through biodigesters

4.1 Biomass

Currently 3% of the energy consumption comes from the use of hydrogen technologies and - in addition to being produced from water and fossil fuels - it can also be produced through biomass and biodigesters (Das, 2001).

Nowadays, biomass can be considered as the best option for the replacement of fossil fuels: besides being present in large numbers in nature, it can also be sustainable. The energy from biomass, specifically wood, has been widely used today. In addition, we have municipal solid waste, animal waste and waste from food processing, aquatic plants and algae as other main sources of biomass (Balat and Kirtay, 2010).

The production of hydrogen from renewable biomass has drawn attention due to the advantages it brings, such as the independence of fossil fuels, the consumption of renewable energy sources from the country itself, decreased imports of fuels and increased co2 capture in the atmosphere. The use of biomass for hydrogen production can be made by thermochemical or biological processes (Balat and Kirtay, 2010).

However, the cost of cultivation, harvesting and transporting biomass are still adversities found for hydrogen production, which hinders its economic competitiveness. Hydrogen production processes that use thermochemical and electrochemical processes consume a lot of energy, besides not being environmentally suitable (Balat and Kirtay, 2010).

A timely solution to this would be the use of biomass, such as organic waste, for hydrogen production.

4.2 Biodigesters and biogas

Biogas is generally produced in the anaerobic digestion process of residual biomass, animal, and plant residues in a biodigester, by the action of microorganisms in the absence of free oxygen. During the last stage of digestion, methanogenesis, two groups of bacteria produce methane, one from acetate and the other from hydrogen and carbon dioxide (Weiland, 2010).

Due to the ease of obtaining the raw material, the correct destination of the same (associated with different agricultural activities such as the production of pigs, cattle, and poultry breeding) and the possibility of use as a source of thermal, mechanical, and electrical energy, this biofuel is extremely attractive.

The potential for hydrogen production from biogas is very large, with various techniques and reform processes. To choose the right method, it is necessary to check some factors about the application of the generated hydrogen, such as the composition of biogas, the required purity content of H_2, the production of the volume of H_2 to be used and the availability of investment. All these points influence the decision-making (Alves et al., 2013).

Biogas has been the object of study to obtain products of higher added value through the processes of purification and reform of biogas. It is composed by a mixture of gases, the larger part being methane and carbon dioxide and the rest being formed by hydrogen sulfide, ammonia, hydrogen, nitrogen, oxygen, and water vapor.

One possibility is the production of hydrogen-rich gas from biogas, which can generate benefits such as higher efficiency and lower emission of nitrogen oxides when compared to the direct combustion of biogas (Ashrafi et al., 2008).

The whole process of biogas to hydrogen and consequently fuel production is represented in the Figure 2 bellow:

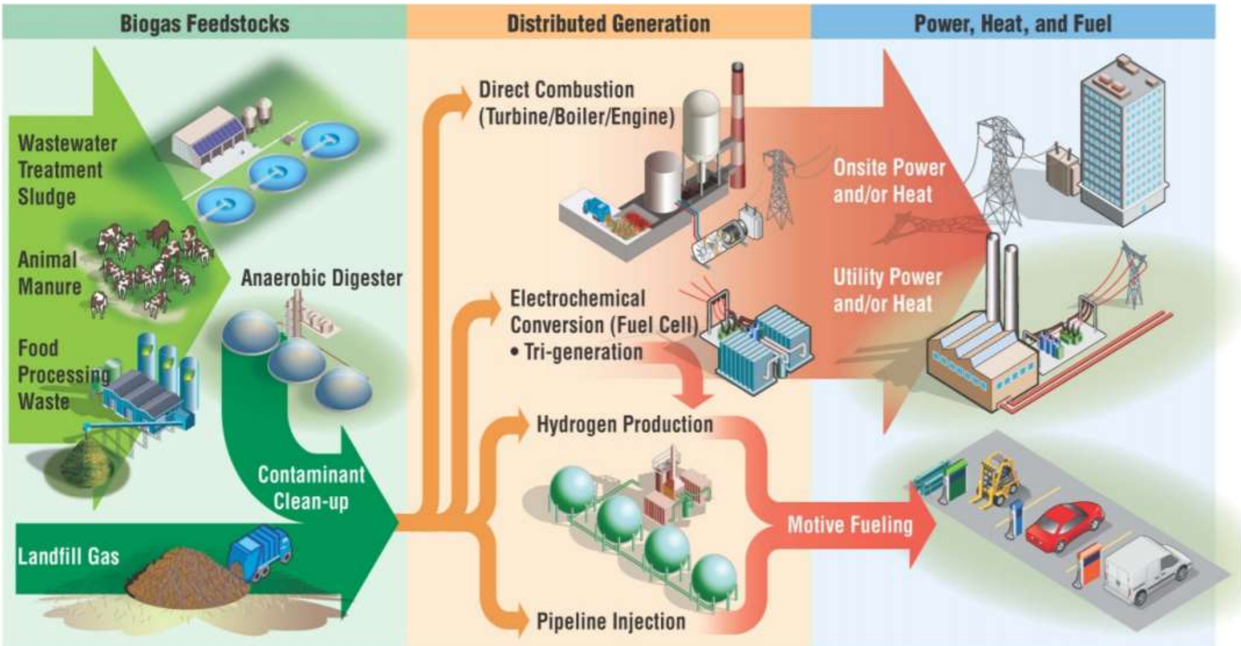

Figure 2. The biogas to energy process. Source: Connelly (2019).

4.3 Purification and reform of biogas

Purification is necessary because biogas in natura is completely saturated with water vapor and, in addition to methane and carbon dioxide, has significant amounts of hydrogen sulfide, among other substances that are considered impurities and must be removed before use, so that they do not cause air pollution or corrosion of equipment (FNR, 2010).

Therefore, when biogas is purified for the removal of water vapor and contaminants such as hydrogen sulfide and also desulfurized to reduce the carbon dioxide content, it ends up possessing many similar characteristics to natural gas and is now referred to as biomethane. As natural gas is the main source of hydrogen extraction, the use of biomethane can enable the reform for hydrogen production. This enables a nobler use of biogas and adds value, since hydrogen is the fuel that has the highest calorific value per unit of mass and has high energy efficiency (FNR, 2010).

Reform is a thermochemical process which fuel is converted in a mixture of gasses rich in hydrogen, the main process used for hydrogen production is the catalytic vapor reform of natural gas. For the production from biogas, the most common processes for obtaining hydrogen gas through methane reform are steam reform, partial oxidative reform, autothermal reform, dry reform, and oxidative dry reform (Alves et al., 2013).

5. Final remarks

In this work was presented three ways to obtain hydrogen as an energy source: from the electrolysis of water, using photosynthesis of plants and algae and finally through the use of biodigesters and making use of waste.

a) Some countries are not geographically well located for the production of solar and wind energy, then the use of hydrogen as an energy source becomes a very interesting option for these places;
b) The hydrogen production using solar energy through the photolysis process is attractive, since it does not require the use of water exclusively, in a period of great scarcity, for example;
c) Hydrogen has flexible and viable forms of storage and transport, so it can be imported from elsewhere;
d) More studies are needed on the microorganisms that can produce hydrogen, as well as more efficient and economically viable processes for the biological production of hydrogen;
e) Another aspect that needs further research is the cost of production and optimization and improvement in the quality of materials used, whether algae, water or organic waste.

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