Review

A review on alternative processes for green hydrogen production focused on generating hydrogen from biomass.

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A Review on Alternative Processes for Green Hydrogen Production Focused on Generating Hydrogen from Biomass

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Abstract Hydrogen has a leading role in accomplishing a net zero green-house gas emission future. Today's challenge is producing green hydrogen to cover the fuel demand of transportation and industry, to gain independence from fossil fuels. This review's goal is to demonstrate critically the existing methods of biomass treatment and assess their ability to scale up. Biomass is an excellent hydrogen carrier and biomass-derived processes are the main target for hydrogen production as it is an innovative pathway to green hydrogen production. Comparing the existing processes, thermochemical treatment is found to be far more evolved than biological or electrochemical treatment especially with regard to scaling prospects.

Keywords: green hydrogen production; biomass; thermochemical treatment; biological treatment; electrochemical treatment

1. Introduction

1.1. Why Hydrogen?

The rising of the population has increased the energy demand and as per International Energy Agency (IEA), by 2030, the energy demand may be increased by 50% globally [1]. The world's most exploited energy sources are fossil fuels and their derivatives [2–4]. Excessive use of these fuels increases green-house gas (GHG) such as CO_2 which in turn have a noteworthy effect on global warming and climate change [5,8]. The climate goals of the Paris agreement (COP21) aim to strategies for the mitigation of GHG emissions to prevent the global average temperature from rising by more than 2°C above the levels of temperature in pre-industrial era [8].

To tackle the issue, the replacement of conventional energy sources with environmentally friendly energy sources is crucial and vital [2]. The long-term replacement of fossil fuels can be accomplished through the enhanced penetration of renewable energy sources in the energy mix. In this review, particular focus is given to hydrogen (H_2) as an energy carrier produced from renewable

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energy sources and provide solutions for zero or near zero emissions levels in transport, industry, buildings, the energy sector, etc.

Hydrogen is considered to be a green fuel as the product of H_2 combustion is water vapor. Thus, it has zero CO_2 emissions when used to produce energy (e.g.via fuel cells or ICEs) [1,5–7]. It has 2.4-, 2.8- and 4-times higher heating value (LHV, mass basis) than methane, gasoline and coal, respectively and 100 times higher energy density than that of a conventional Lithium-ion battery. Hydrogen, compared to other known fuels, has the highest energy content per unit weight. It also has many aspects and characteristics like storage capability, compared to electricity storage capability, that make it attractive and a probable candidate to play a significant role as a fuel for the future [5–9]. Its transportation can be done by e.g., conventional means for domestic/industrial consumption. H_2 gas safety aspects for transportation and handling can be comparable to those of domestic natural gas [11]. Concerning H_2 production, it is predicted that 50-82 Mt of H_2 are produced with a rate of growth of 5-10% per year estimated by the year 2050 [1,10].

Considering all these characteristics, hydrogen can be applied as a renewable energy carrier and is particularly interesting in applications such as heavy duty, transportation, and other industrial cases, that are hard to electrify [9].

Table 1. Energy contents of different fuels [20,21].

Fuel	Energy Content (MJ/Kg)
Hydrogen	120
Liquefied natural gas	54.4
Propane	49.6
Aviation gasoline	46.8
Automotive gasoline	46.4
Ethanol	45.6
Methanol	29.6
Coke	19.27
Wood (dry)	16.2
Bagasse	9.6

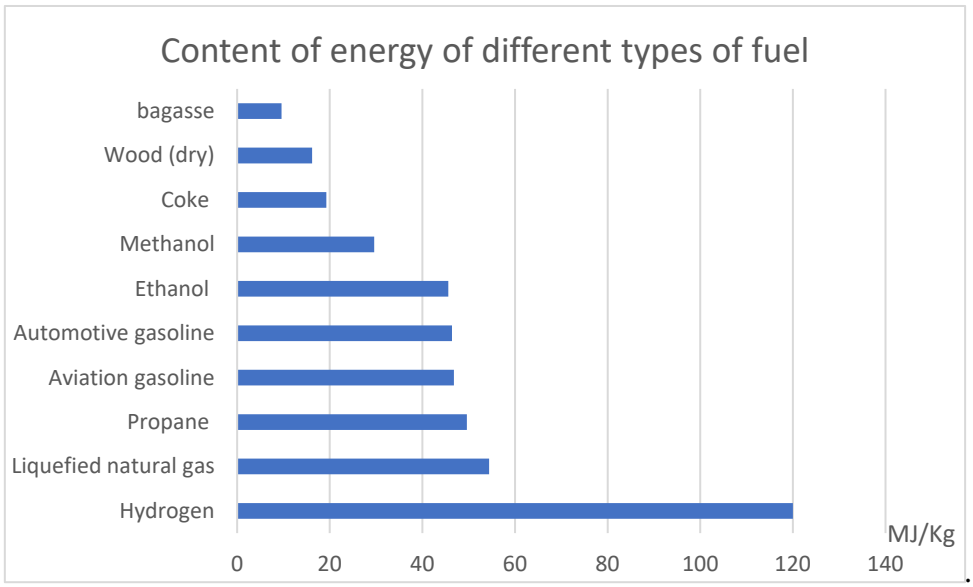

Figure 1. Energy contents of different fuels [20,21].

1.2. Hydrogen Production Methods

Up to date, hydrogen production derives 48% from natural gas, 30% from heavy oils and naphtha, and 18% from coal, consisting a total of 96% from non-renewable energy [1,12–15].

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The most common approaches to hydrogen production based on fossil fuels are:

- Steam reforming of natural gas (SR): The method involves a catalytic conversion of hydrocarbon and steam to hydrogen and carbon oxides. It consists of the main steps of reforming or synthesis gas (syngas) generation, water-gas shift (WGS) and methanation or gas purification [11,12]. This method is the most common for producing hydrogen and has a TRL of 9 [111].
- Partial oxidation process (POX): The method involves the conversion of steam, oxygen and hydrocarbons to hydrogen and carbon oxides. The catalytic process occurs at 950 °C with feedstock from methane to naphtha. The non-catalytic process occurs at 1150–1315 °C with feedstock that includes methane, heavy oil and coal. After sulfur removal, pure O_2 is used to partially oxidize hydrocarbon feedstock. The syngas produced is further treated in the same way as the product gas of the SR process [11,12].

Hydrogen is to be used as a sustainable alternative to fossil fuels. Therefore, it is essential to be produced without net emissions of GHG [9]. The above, currently employed, methods are accompanied by considerable GHG emissions, therefore they should be replaced by alternative ones that are primarily based on renewables.

Water electrolysis is one of the most promising and environmentally friendly alternatives for hydrogen production. It is a technology with a technology readiness level (TRL) of 9 and is already producing around 2-3% of the world's hydrogen [16].

- Water Electrolysis: The method uses electrical current in order to separate water into oxygen and hydrogen. This way green hydrogen is produced without any direct emissions of carbon dioxide. The reaction is very endothermic. Thus, renewable energy sources can provide the required energy input [12–16]

In this review, the focus is on the main alternative hydrogen production methods from biomass. They are briefly presented below.

2. Alternative Processes from Biomass

Biomass is a source of energy that derives from plant and animal material. It is renewable and it can consist of energy crops, crop residues, forest wood, forest residue, grass, industrial residues, animal and municipal waste etc. Biomass mainly consists of organic matter in which the energy of sunlight is stored in chemical bonds through photosynthesis. When biomass is utilized to produce energy, CO_2 is released. However, the amount of CO_2 that is emitted is equal to the amount absorbed by the organisms while they are still alive [12,17–19]. Therefore, there is no contribution to the carbon cycle, it is only accelerated [12,86].

2.1. Biological Treatment

2.1.1. Dark Fermentation

Dark fermentation is based on anaerobic bacteria growing in the dark that decompose biomass [9]. Bacteria, such as Enterobacter, Bacillus, and Clostridium, are known to produce hydrogen [11,24]. The bacteria or micro-algae are sustained in dark conditions at 25-80oC. Depending on the strains, they can be sustained even at hyperthermophilic temperatures (>80oC) [29]

Among other carbohydrates, glucose is the most preferred source of carbon for the fermentation process. With glucose used as the main model substrate, acetic and butyric acids are produced and cover over 80% of total end-products. In theory, when 1 mol of glucose undergoes bioconversion, 12 mol of H_2 are being produced [11,12,29].

The reaction is displayed below:

$$C_6H_{12}O + 2H_2O \rightarrow 2CH_3COOH + 4H_2 + 2CO_2 \text{ (acetate fermentation) [25]}$$

$$C_6H_{12}O + 2H_2O \rightarrow CH_3CH_2CH_2COOH + 2H_2 + 2CO_2 \text{ (butyrate fermentation) [26]}$$

The conditions in which the process is occurring, e.g. pH (between 5 and 6) [12,27], hydraulic retention time (HPT) and gas partial pressure, act on the metabolism balance of the bacteria used in fermentation. The H_2 partial pressure is one of the most critical factors because hydrogen synthesis

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pathways are sensitive to H_2 concentration. As H_2 concentration increases, the H_2 synthesis decreases [11,24]. Thus, the H_2 gas must be removed as it is generated [12,28].

2.1.2. Photo Fermentation

Photo fermentation (PF) takes place in nitrogen-deficient conditions and the implementation of purple non-sulfur bacteria [11]. The reaction is catalyzed by nitrogenases enabling the production of hydrogen and carbon dioxide from organic acids (acetic, lactic and butyric) through photosynthetic bacteria [12].

The reaction is displayed below:

$$CH_3COOH + 2H_2O + \text{light energy} \rightarrow 4H_2 + 2CO_2$$

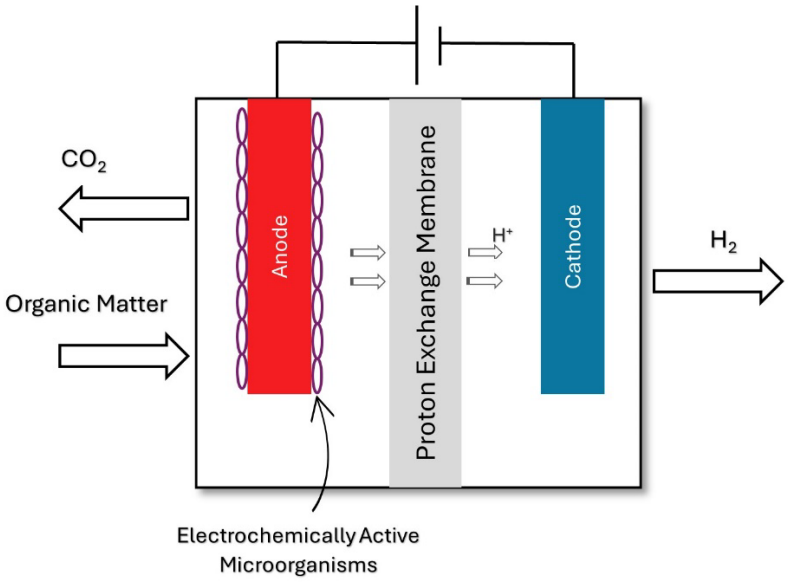
These photosynthetic bacteria are suitable for converting light energy to hydrogen by using organic wastes as substrate [30–32] in batch processes [33] and continuous cultures [34]. The proper ratio of carbon to nitrogen nutrient must be achieved and controlled to increase the nitrogenase activity and decrease the energy demand [11,23]. Moreover, the light intensity is a factor that has a contributing effect on hydrogen production rate and yield but, has an adverse result on the light conversion efficiency [12].

PF has been studied as a waste-prevention process to produce H_2 from industrial and agricultural wastes [29].

2.1.3. Biocatalyzed Electrolysis

This process is another way of oxidizing organic matter for hydrogen production. The different aspect of this method is that the external energy required is in the form of electrical energy [11].

This method takes place in a microbial electrolysis cell (MEC) [84], often mentioned as bio-electrochemically assisted microbial reactor (BEAMR) [78,79], biocatalyzed electrolysis cell (BEC) [80], and electrohydrogenesis [81–83]. Microorganisms that are electrochemically active are able to utilize an electrode as an electron acceptor under anaerobic conditions, as they release the electrons at high energy level. As a result, the electrode turns into a bioanode. The organic matter is electrolyzed and then hydrogen is generated. The required applied voltage for this method of electrolysis is about 10-12mV [11,35]. The anode materials are essential in influencing the bacterial adherence on the anode, the electrode's biocompatibility and the electron transmission [74,75].



Scheme 2. Schematic illustration of hydrogen production through biocatalyzed electrolysis.

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Biocatalyzed electrolysis is also capable of converting dissolved organic compounds that might be produced from dark fermentation, addressing the endothermic nature of these reactions. It is also very flexible as a method as it will be designed to produce hydrogen from wastewater [35].

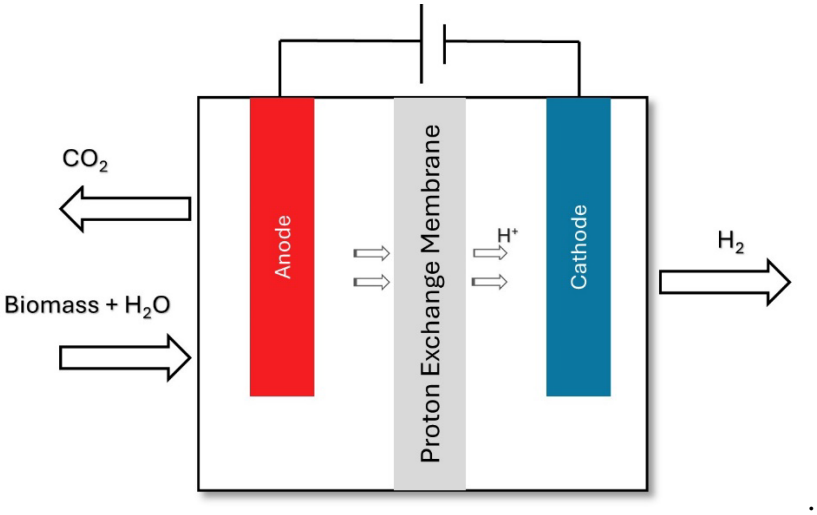
2.2. Electrochemical Treatment

2.2.1. Electrooxidation

Electrosynthesis has seen a rebirth during the last decade due to research for a dual production platform for both molecules and energy carriers. It is referred to as a "Power-to-X" approach, where X refers to fuel or chemical. "X-to-Power" refers to a strategy in which clean energy is produced from an energy carrier, as an inverse approach. A fuel cell, in the broadest sense, is an electrochemical device made up of two electrodes separated by a spacer that turns chemical energy directly into electrical energy (while releasing heat). Thermodynamic and faradaic contributions won't alter during a fuel cell's normal operation if the reaction selectivity doesn't change significantly. Thus, the efficiency is solely influenced by the experimentally measured cell voltage U [55].

The research on fuel cells and electrolysis cells is closely linked with the study of organic selective electrooxidation processes. Their slow electrochemical kinetics have been overcome by a variety of (bio)catalytic interfaces. The key electrocatalytic reaction characteristics that have been established allow the proposal of new materials that could maximize the activity, selectivity, and durability of anode materials [55].

The oxygen reduction reaction (ORR), which is carried out by a cathode, is thermodynamically anticipated to begin at about 1.2 V relative to the reversible hydrogen electrode (RHE). The best activity is attained in the potential range of 1.0-0.7 V versus RHE due to ORR's sluggishness. The primary criterion for choosing anode electrocatalysts for fuel cells (operating voltage optimization) is an electrooxidation at the lowest electrode potential (E), ideally E < 0.5 V vs RHE, with the lowest overpotential [55].



Scheme 3. Schematic illustration of hydrogen production through electrooxidation.

Electro-oxidation of bio-derived feedstocks may provide compounds with significant economic value that can increase income and represent a more cost-competitive option. According to studies, most electrocatalysts for the electro-oxidation of organic compounds have increased activity and stability under alkaline conditions [56–59]. The main drawback of alkaline electro-oxidation is its lower H_2 selectivity. H_2 accounts for no more than 6% of the total product share in the processes. The demand for hydrogen, however, is anticipated to grow significantly over time in comparison to the coproducts [60]. The lack of a sizable enough market for the coproducts may limit the total deployment potential of this technology toward small scale applications [55].

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2.3. Thermochemical Treatment

The most mature process for producing hydrogen from biomass is thermochemical conversion [38]. Aqueous phase reforming, pyrolysis, and gasification are the three main thermochemical methods [28,39]. In these conversion methods CH_4 and CO are produced which can be further processed for more hydrogen production through steam reforming and WGS reaction [12].

2.3.1. Gasification

Gasification is a thermochemical treatment according to which biomass is decomposed at elevated temperatures in an environment with limited oxygen [9,40]. It is highly endothermic, it takes place at temperatures between 500 and 1400 °C and at operating pressures, depending on the plant scale, from atmospheric to 33 bar [12]. The process can be classified as air gasification, oxygen gasification, or steam gasification determined by the oxidizing agent utilized [28].

The gasifier is the most significant element of a gasification plant. It offers enough space to mix biomass and the gasification agent to a certain level, sometimes in the presence of primary catalysts and/or additives [45]. Depending on how the process is set up, gasification can generally be divided into three groups: dual fluidized bed (DFB) steam blown gasification [45], also referred to as indirect gasification, direct blown, steam/oxygen or air, fluidized bed gasification [46], and entrained flow gasification [9,47].

The feedstock normally undergoes pre-treatment before being introduced into the gasification reactor. This frequently entails drying the biomass to increase conversion rates in the gasification reactor, but it can also entail other processes, including milling, to produce a uniform feedstock in terms of size [9,43].

Syngas, a synthesized gas made of hydrogen, methane, carbon monoxide, nitrogen, and carbon dioxide, is produced when it consumes an oxidizing agent [28]. Minor organic and inorganic contaminants have also been discovered in syngas. The inorganic molecules include H_2S, HCl, NH_3, and alkali metals, while the organic compounds include light hydrocarbons (LHC), such as CH_4, and tar, a viscous liquid made up of condensable organic compounds [28,42]. Depending on the desired end-product of the gasification process, a range of product gas upgrading and conditioning sequences may be included in the process design [9].

The general air gasification reaction of biomass is displayed below [29,41]:

$$\text{Biomass} + \text{Air} \rightarrow N_2 + CO + H_2 + CO_2 + CH_4 + H_2O + \text{LHC} + \text{Tar} + \text{Char}$$

Regarding feedstock, air gasification requires a dry raw material [44]. The main challenge in air gasification is the nitrogen removal from the products, as it is a non-combustible gas that results in the reduction of the heat value of the fuel.

The product of oxygen gasification is a gas of greater purity, containing a higher level of H_2, less tar and char and no nitrogen.

As a middle ground, between air and oxygen gasification, steam gasification is employed. This process can take place with wet biomass (moisture between 5 and 35 wt%) [44].

The general reaction of biomass' steam gasification is presented below [29]:

$$\text{Biomass} + \text{Steam} \rightarrow H_2 + CO + CO_2 + CH_4 + \text{HC} + \text{Tar} + \text{Char}$$

Gasification also produces CO and CH_4 which can be further treated to make more hydrogen if they undergo steam reforming and the WGS reaction [12].

2.3.2. Pyrolysis

Pyrolysis is another thermochemical method which doesn't require an oxidizing agent and can be done at lower temperatures of 400-800oC [9] at 0.1-0.5 MPa [12]. The method is categorized according to rising reaction temperature into three main classifications: slow pyrolysis, fast pyrolysis, and flash pyrolysis [48]. This reaction is also highly endothermic [29].

The three main components of a biomass pyrolysis feedstock are cellulose, hemicellulose, and lignin. These three constituents make up to 90% of lignocellulosic biomass, with ash and extracts

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making up the remaining 10% or so [53,54]. The type of feedstock, the type of catalyst utilized, the temperature, and the time of residence affect the yield of hydrogen production from biomass pyrolysis [12,50,51].

Biomass pyrolysis typically occurs in four steps [52]: 1) pre-heating and drying, 2) pre-pyrolysis, 3) solid decomposition, and 4) the residual char decomposition process, which is a complex chemical process that includes numerous simultaneous reactions [53].

The general pyrolysis reaction is presented below [29]:

$$\text{Biomass} + \text{Heat} \rightarrow H_2 + CO + CO_2 + CH_4 + H_2O + \text{Bio-Oil} + \text{charcoal}$$

Methane and other hydrocarbon gases produced, similar to gasification, can be steam reformed, and the WGS process is utilized to produce even more hydrogen. After pyrolysis, condensed oxygenated molecules (aldehydes, ketones, alcohols, phenolic compounds, and carboxylic acids), water, and ash can create "bio-oil," a complex liquid fraction that resembles tar. Water-insoluble fraction and water-soluble fraction are the two categories into which bio-oils can be separated. For usage in adhesive applications, the insoluble fraction can be broken down into platform molecules like BTX or olefins [49]. To improve the yield of H_2 produced from the soluble fraction, a steam gasification device can be incorporated into the procedure [29].

2.3.3. Biogas Reforming

Biogas may be produced from biomass and agricultural residue, such as rice straw and wheat straw, via anaerobic digestion. It may also be obtained from landfills, which is referred to as landfill gas. The composition of biogas differs according to the feedstock and anaerobic digesters utilized [87]. Biogas as an energy resource is practical and sustainable due to the abundant supply of low-cost feedstocks [87]. In most studies the biogas composition is simulated 60% CH_4 and 40% CO_2. Biogas in Europe is mostly produced by anaerobic digestion of agricultural or industrial wastes as well as biowaste, municipal organic waste and sewage sludge [113,114].

Table 2. Chemical composition of biogas [88–91].

Composite	Percentage
CH_4	55-70 (vol%)
CO_2	30-45 (vol%)
H_2S	500-4000 (ppm)
NH_3	100-800 (ppm)
H_2	<1 (vol%)
N_2	<1 (vol%)
O_2	<1 (vol%)
H_2O	<1 (vol%)

Table 3. Chemical composition of biogas from landfill and anaerobic digestion**.**

Composite	Landfill	Anaerobic digester
CH_4	44.2 (mol%)	58.1 (mol%)
CO_2	34.0 (mol%)	33.9 (mol%)
LHV	12.7 (MJ/kg)	17.8 (MJ/kg)

Steam reforming (SR) and autothermal reforming (ATR) are methods that have been established as prototypes for biogas to hydrogen generation due to their significant similarity to natural gas [92]. Biogas steam reforming (BSR) produces H_2 at temperatures ranging from 600 to 1000 °C through endothermic and reversible reactions, typically coupled with catalytic methods [92]. The ATR process combines two reforming techniques: SR and catalytic partial oxidation (CPOX) [101,102]. Both reforming processes may be carried out at low pressure (usually at atmospheric pressure) in tubular fixed bed or fluidized reactors [92–96]. The mixture of gases produced by the conversion process has

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a high concentration of hydrogen, thus CO_2 and other contaminants must be separated from the gas output [92].

The reforming reactions are displayed below [101]:

Steam Reforming (SR) : $CH_4 + H_2O \rightarrow CO + 3H_2$, $\Delta H = 206.2$ kJ/mol

Catalytic Partial Oxidation (Pox): $CH_4 + ½\ O_2 \rightarrow CO + 2H_2$, $\Delta H = -36$ kJ/mol

Autothermal Reforming (ATR): $CH_4 + x½O_2 + yCO_2 + (1 - x - y)H_2O \rightarrow (y + 1)CO + (3 - x - y)H_2$

Biogas reforming takes place in several steps in order to obtain purity. The process includes the following units in order: a high temperature reformer, a high temperature shift reactor (HT), a low temperature shift reactor (LT) and a separation unit [87]. At the moment, several gas separation technologies are accessible for separating hydrogen from the synthesis gas via reforming or gasification methods. The membrane gas separation system is simple to use, requires less energy, has a compact footprint, can operate continuously, is easy to scale up, etc. [99,100]. Steam methane reforming demands a feedstock stream with high purity. Thus, we make the assumption that biogas is upgraded with 99.5% purity [113].

3. Prospects of Hydrogen Production from Biomass in Scale

To assess the previously mentioned processes, it is a necessity to examine the type of feedstock and pre-treatment that may be needed, the production materials/energy requirement and the purity of the gas output.

There are two forms of feedstock that can be utilized to provide the required amount of biomass to create hydrogen: lignocellulosic residue and dedicated crops. Because they are produced towards the end of the harvest season for crops like cereal wheat or during the transformation process, lignocellulosic waste is easily accessible and inexpensive. Dedicated energy from crops like sorghum, however, demands land utilization and growth time, which might be difficult. Clustering biomass kinds based on their chemical components (carbohydrates, lignin, and other components) can make the necessary pre-treatment easier [10,73].

Comparing the biological processes of biomass, dark fermentation seems to be the most competitive when considering scale-up possibilities. It manifests an efficiency of 60-80% [12], yield between 0.004 - 0.044 kgH_2/kg biomass [10] and has a TRL of 5 [9,10,66]. It is a simple method that can produce H_2 without light, contributing to waste recycling. It is also CO_2-neutral and has no O_2 limitation [12]. The main disadvantages of this process are fatty acids removal, low H_2 rates and yields, low conversion efficiency, and the requirement of large reactor volume [12]. In addition, dark fermentation is limited by poor catalyst durability and product contaminants [62]. The production cost is around 0.332 – 2.63 €/kgH_2 [10,66] but the numbers are indicative because there are few full plant cost estimations [61]. There are no commercial scaled plants yet. Also, feedstock pretreatment is significantly expensive in order to be enzymatically fermentable. Thus, the industrial scale of the process development is limited [10].

Photo fermentation is also a CO_2-neutral process that contributes to waste recycling and can use different organic wastes and wastewaters [12] but, the light conversion efficiency is only 1–5% [11], the production cost is 0.362 – 3.66 €/kgH_2 [10,66,69] and the possibly achieved yield is 0.004 - 0.049 kgH_2/kg biomass [9]. The method also requires sunlight, large reactor volume and is sensitive to O_2 and has a TRL of 4 [9,10,12,66]. Problems that might occur with industrial effluents being used for H_2 production are the color of the wastewaters, which will prevent light penetration, and heavy metals or other toxic compounds that might be carried in the wastewaters [12,22]. Lastly, there are difficulties in controlling the various bacteria [10]. Compared to dark fermentation, it is thought to be less financially competitive because it achieves approximately the same yield while requiring higher production cost [63].

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The last biological process, biocatalyzed electrolysis, has an electrical requirement that remains far below that of commercial water electrolysis. Even when microbial metabolic and other energy losses increase the energy demand, it is expected to remain below 1 kWh/m^3 H_2. Biological anodes can easily be operated under non-sterile conditions because electrochemically active consortia can be naturally selected from a wide range of inocula. This process can theoretically produce hydrogen as a pure gas in the cathode chamber instead of a mixed gas output [35]. The most significant barrier to these techniques in terms of viability is the low hydrogen production rate [72]. In addition, enzymatic biocathodes are relatively unstable and they are not self-regenerating [35]. There has not been thorough recent research on this method, especially from a techno-economic standpoint. Another challenge that this method faces is that the lignocellulosic biomass, a plentiful natural resource, cannot be directly processed by the microorganisms in the microbial electrolysis cell. In order to transform it into monosaccharides or low-molecular-weight molecules, it must first undergo fermentation [77]. Therefore, biocatalyzed electrolysis emerges as a supplementary technology to fermentative processes. Their combination allows the recovery of the energy content in the substrate up to 90% [36,67].

Biomass electrolysis offers higher hydrogen production efficiency and lower ΔE_{eq} than water electrolysis because the oxidation of the biomass-derived material has lower thermodynamic requirements. The production cost when applying the current density range of 0.2-1.0 A/cm^2 in biomass-based organic molecules is approximately 8 - 10 €/kgH_2, but it can be reduced significantly considering the value-added chemical(s) that are co-produced [55]. This process's disadvantage is its slow kinetics due to the numerous electron transfer mechanisms [56]. Electro-oxidation has a TRL of 2-4 [10] and there has not been extended recent research on this method.

For thermochemical processes, the industrial design has already been defined [10]. The approach was developed using comparable techniques used with biofuels like biomethane and adapted from steam methane reforming (SMR) [38]. Gasification has an efficiency up to 50 %, a TRL of 7, production cost between 1.14 – 3.29 €/kgH_2 and produces the highest yields [10,66–68]. Steam gasification has a 40% H_2 percentage in the gas, higher H_2/CO ratio (1.6), reduced impurities compared to air gasification and produces 0.040 kg H_2/kg biomass without catalyst and 0.070 kgH_2/kg biomass with catalyst [29,76]. This process is thought to be the most suitable process for hydrogen production. Concerning the feedstock, steam gasification is feasible for wet biomass (moisture from 5 to 35 wt%), while air gasification requires a dry raw material [37]. However, because of the production of tar and char, the processes are more vulnerable to catalyst deactivation and the gas products need to be separated and purified [10,76]. It was also seen that the gasification process was economically non-viable due to ash-related issues such as corrosion, erosion, agglomeration, and sintering [2,40].

Pyrolysis has shown efficiency up to 65% using HDPE as feedstock, yield 0.100 kg H_2/kg biomass & HDPE and 0.373 kg H_2/kg HDPE [9] and TRL of 7 [10,66]. The production cost of this process is around 1.14 – 2.41 €/kg H_2 [10,66,69]. Studies show that at the same temperature, fast pyrolysis of biomass releases more volatiles than slow pyrolysis [53]. It has been observed that the presence of a catalyst raised H_2 gas yield while reducing CO, C2–C4, and CH_4 yield [2]. On the contrary, because of the production of tar and char, the processes are more vulnerable to catalyst deactivation [10]. There is less data for biomass-to-hydrogen yields and production costs in the scientific literature on techno-economic analyses of pyrolysis than of gasification. The gasification and pyrolysis of biomass use similar procedures to those used to treat fossil fuels. They are projected to develop and reach a TRL of up to 9 in the following two decades [10]. However, to produce negative emissions, thermochemical processes that release CO_2 must be combined with carbon capture systems (CCS) [10].

Comparing biogas from landfill and anaerobic digestion of biomass the performing efficiency is slightly higher when it comes to biogas from anaerobic digestion. When undergoing SR and ATR at 20 bar it is shown to achieve a maximum of 51.7% and 27.8%, respectively [92]. As the temperature of the reforming process increases, so does the yield of H_2. It reaches a peak before slightly decreasing [103]. Although this approach produces significant hydrogen yields, achieving the required high purity hydrogen, it involves complicated energy integration, costly heat exchangers (high

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temperature), and numerous process units. The amount of distinct process stages affects system efficiency, making scaling down uneconomical [87]. The approximate production cost is between 4.21-4.29 €/ kg H_2 for SR and 6.41-6.60 €/ kg H_2 for ATR, when both processes are performed at 20 bar using biogas from anaerobic digestor and landfill respectively. As far as TRL, SMR reach a TRL of 9, and ATR reach a TRL of 8 when processing natural gas [105]. The process' model generates 0.29 kg H_2 per kilogram of bio-methane [113,116].

Antonini et al [113], have conducted research that compares natural gas to biomethane and investigates CCS and storage systems. Results show that natural gas and biowaste-based biomethane have minor differences in performance when undergoing SR and ATR, despite their different composition. However, addressing the life cycle impacts on climate change, biomethane is clearly more sustainable as the CO_2 that is released from biomethane does not contribute to the carbon cycle in contrast with the contribution of natural gas to GHG.

The following table, Table 4, displays a summary of the most important features of each process from various literature references which were mentioned in the previous paragraphs. The table highlights each methods efficiency (%), yield (kg H_2/kg biomass), production cost (€/ kg H_2) and TRL. In order to compare them, the necessary unit conversions were made. In addition, data from water electrolysis are shown in Table 3 to compare the previously mentioned methods of green hydrogen production to water electrolysis, another method of green hydrogen production. The most advanced methods of water electrolysis are alkaline electrolysis (AEL), proton-exchange membrane electrolysis (PEMEL), solid oxide electrolysis (SOEL) [106].

Table 4. Important features of each method.

Process	Efficiency %	Yield (kg H_2/kg biomass)	Production Cost (€/ kgH_2)	TRL
Dark fermentation	60-80 [12]	0.004 - 0.044 [10]	0.332 [9] 2.42 – 2.63 [10,66]	5 [9,10,66]
Photo fermentation	Light conversion efficiency 1–5 [11]	0.004 - 0.049 [9]	0.362 [9] 2.50 – 3.66 [10,66,69]	4 [9,10,66]
Biocatalyzed electrolysis		0.095 (kg H_2/ kg glucose at 0.6 V) [84,85]	The cathode: 44.50, ~80.55 (based on laboratory materials, not recent) [35].	
Electrooxidation			8 - 10 [55]	2–4 [10]
Gasification	50	(SG) Without catalyst: 0.040 With catalyst: 0.070 [76]	1.14 – 3.29 [10,66–68]	4-7 [10,66–68,112]
Pyrolysis	65 using HDPE [9]	0.100 (kg H_2/kg biomass & HDPE) 0.373 (kg H_2/kg HDPE) [9]	1.14 – 2.42 [10,66,69]	7 [10,66]
Biogas Reforming	46.2-51.7 (SR) 24.5-27.8 (ATR) [92]	0.29 (SR, kg H_2/kg bio-methane) [113,116]	4.21-4.29 (SR) 6.41-6.6 (ATR) [92]	9 (SMR) 8 (ATR), for natural gas [105]
Water Electrolysis	51-60 (AEL) 46-60 (PEMEL) 76-81 (SOEL) [106]		3.38-5.45 [108] 5.87 (PEM) (including capital cost and maintenance)	9 (AEL), 6-8 (PEM) [110]

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[107]

The Figures (Figure 2-4) that are following are based in Table 2. The two colors on the bars represent the range of the values that are mentioned on Table 2, with orange depicting the minimum value and yellow the maximum value.

In Figure 2, the comparison of the method's yield is presented. The chart clearly shows the advantages of thermochemical methods in aspects of yield. This conclusion is justified because of the various and extended research and implementations of these technologies in fields other than hydrogen production. Biocatalyzed electrolysis has a lead among the biological methods, even though their performance does not differ much. In general, two obstacles are preventing biological processes from evolving and expanding on a global scale. Even while dark fermentation has made biological methods more competitive, the H_2 yield and production pace are significantly lower compared to thermochemical methods [10,71]. Lastly, pilot-scale processes are limited by the need for pre-treatment throughout the synthesis of complex biomass [10].

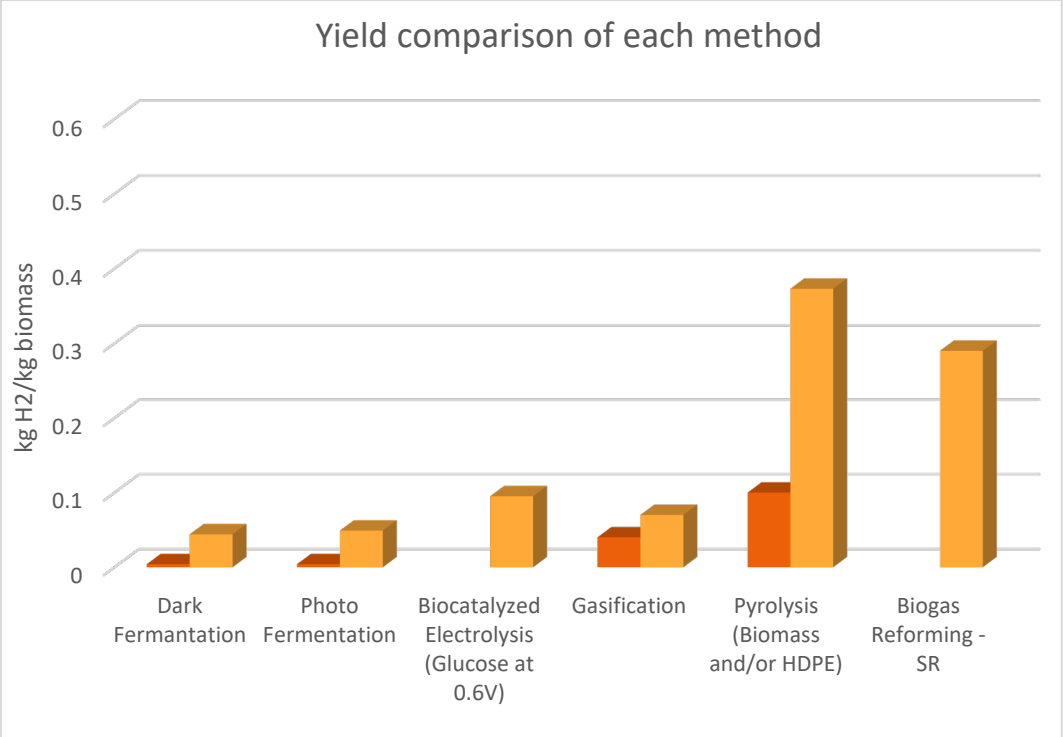

Figure 2. Comparison of each method's yield.

Figure 3 was created by comparing the cost of each method. This chart also indicates that the thermochemical methods are quite advantageous but not far more inviting than the fermentation processes in terms of production cost. Electrooxidation is more expensive than the previously mentioned methods but not as unviable as biocatalyzed electrolysis, which is far more expensive than any other method even if the data we have are from laboratory materials. Biogas reforming methods are costly compared to the other thermochemical and biological methods but have similar cost to water electrolysis. In general, we can conclude that thermochemical and biological methods are the most advantageous financially.

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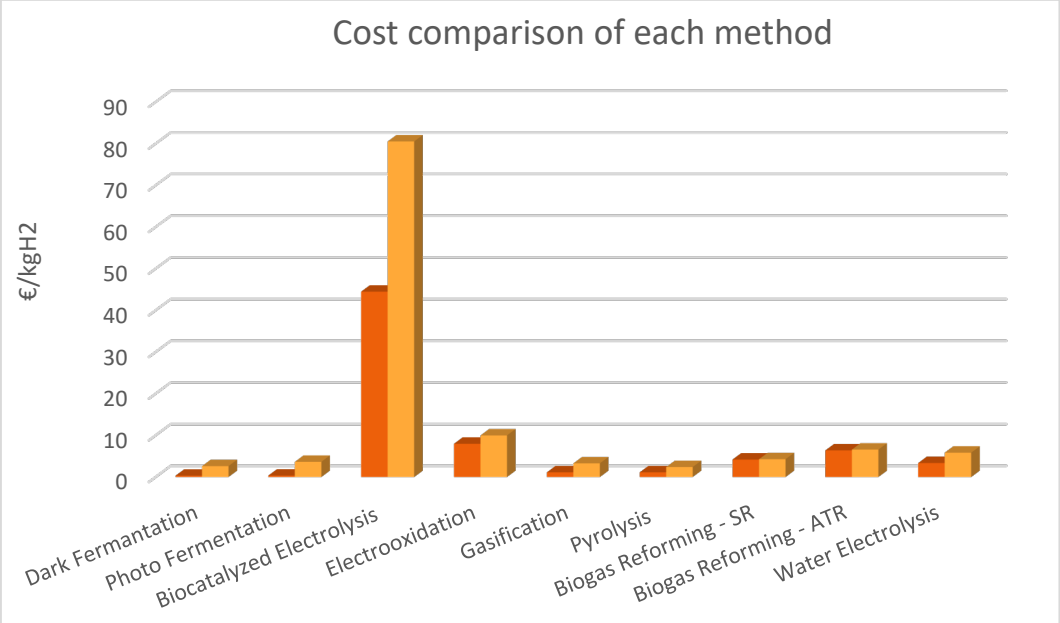

Figure 3. Comparison of each method's cost.

In Figure 4, the TRL of each method is depicted. The purpose of this figure is to compare the TRL of hydrogen production methods based on biomass to other established methods such as SR and ATR of natural gas and water electrolysis via PEM and AE. It is clearly shown through Figure 4 that the methods that are competitive against the dominant methods of SR and water electrolysis are gasification and pyrolysis. Biological and electrochemical methods do not present TRL above 5, which means that they are still under development while thermochemical methods are in a state of pre-commercial demonstration.

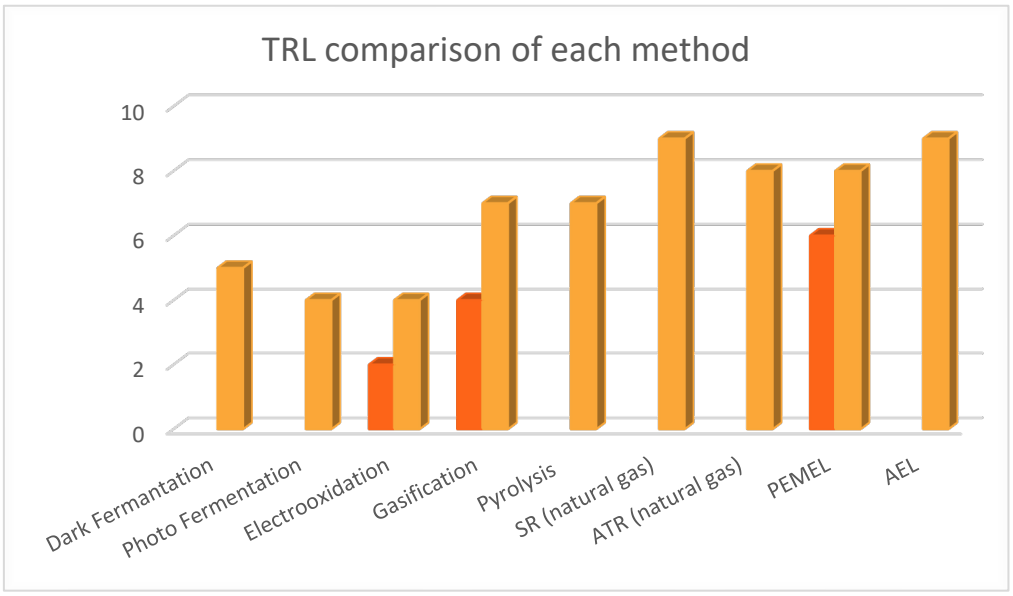

Figure 4. Comparison of each method's TRL.

Thermochemical methods have shown an advantage both in yield and TRL aspects. However, the CO_2 emissions intensity and the CCS incorporation need to be taken into account to assess these methods' potential to meet our goal to mitigate GHG emissions. The life cycle assessment conducted by Antonini et al [113] which was previously mentioned, also indicates that the CO_2 emissions intensity with CCS for hydrogen production from bio-methane is a net-negative CO_2 emissions

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process. ATR performs higher CO_2 capture than SMR and the addition of a low-temperature WGS is generally improving the life cycle performance. It is safe to conclude that in general, adding CCS leads to clear benefits considering the implications of climate change. However, results manifest that these methods perform worse with, than without, CCS because the CCS integration is increasing the energy demand and consumption, which leads to other environmental pressures.

Table 5 [116] displays the CO_2 emission that each thermochemical method produces when used for hydrogen production. In addition, Figure 5 is presented based on Table 5.

Table 5. Hydrogen production methods and CO_2 emission intensity [116].

Process	TRL	Emission (kg CO_2/kg H_2)	Source
Fossil methane - SMR	9	10.09 - 17.21	[24]
Fossil methane – SMR & CCS	7-8	2.97 - 9.16	[24]
Coal gasification	9	14.72 – 30.90	[24]
Coal gasification & CCS	6-7	2.11 – 10.35	[24]
Electrolysis (with wind and solar)	9	0.47 - 2.5	[24]
Biomass gasification	5-6	0.31 - 8.63	[24]
Biomass gasification & CCS	3-5	(-)17.50 - (-) 11.66	[24]
Bio-methane -SMR	9	1.20 - 8.60	[113]
Bio-methane -SMR & CCS	7-8	(-)11.60 – (-)8.84	[113]

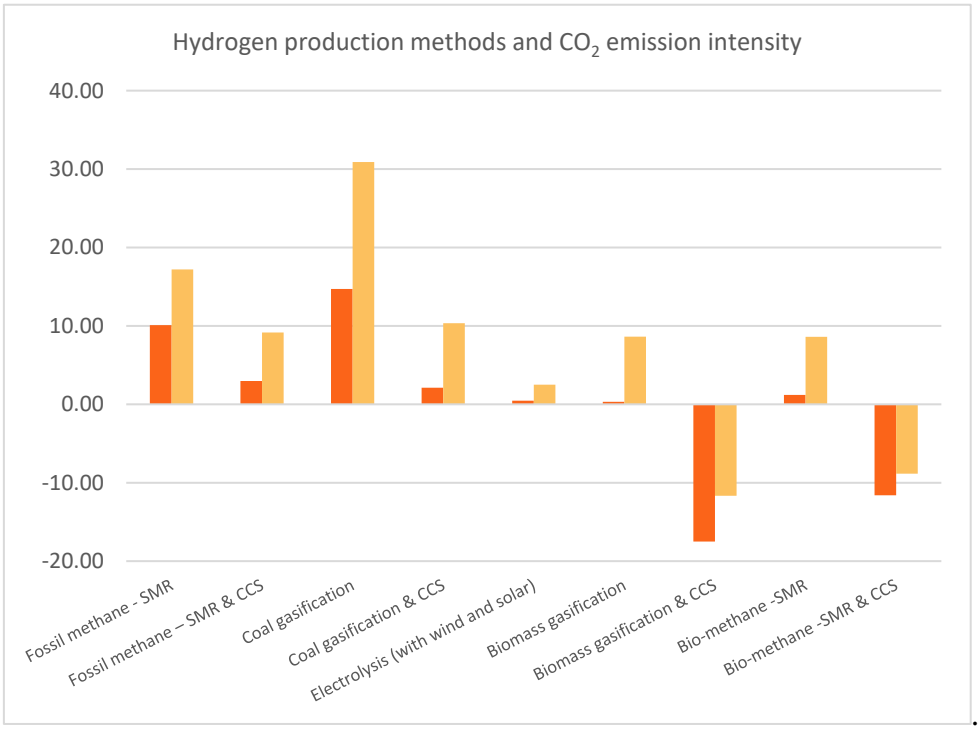

Figure 5. Comparison of CO_2 emission intensity.

From Table 5 [116], we conclude that in every case, the TRL is lower when the process is combined with CCS. Nevertheless, biomass gasification and bio-methane SMR show significant results when CCS is included in the system. Comparing these two methods, as the two most inviting methods of green hydrogen production, with their non-green equivalent it is shown that SMR with CCS from fossil methane and SMR with CCS from bio-methane present the same TRL but a significant difference in CO_2 emissions. The latter demonstrates net-negative CO_2 emissions which is a very important factor considering a net zero economy that requires carbon absorption and negative emissions from the atmosphere by 2030.

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4. Conclusions

This review has comparatively shown that among the biological processes, dark fermentation has the most potential for large scale development. Bio-catalyzed electrolysis, as well as electrochemical methods, is advantageous compared to water electrolysis concerning electrical demand. However, the current status of the method reveals that it is not yet financially viable, and the obstacles are numerous. Also, there has not been extended techno-economic analysis of full scaled plants for any biological processes nor for electrochemical methods. Examining the methods with lower TRL, the combination of dark fermentation and biocatalyzed electrolysis as two consecutive treating methods has more potential than the two methods separately and may be subject to further exploration.

Thermochemical methods are significantly more advanced because they have already been applied in studies concerning hydrogen production from fossil fuels. Steam gasification as well as bio-methane reforming are considered the most suitable processes for a scaled-up plant in the near future. Some of their distinct advantages compared to other methods are the high purity in the gas output, the high ratio of H_2/CO_2, low production cost and high yield. The prospect of a scaled plant is much more realistic for these methods due to the extended research that has already been conducted. However, it is important to consider that to produce negative emissions, thermochemical processes that release CO_2 must be combined with carbon capture systems (CCS) in which case, the TRL comparison shows that the systems are still under development and optimization to be fully efficient but still, demonstrate very promising potential of large-scale development.

The need to switch from fossil fuels to renewable energy sources is urgent and hydrogen is a very promising energy carrier that may give the solution to this problem. Biomass has been studied through various processes since it is a significant hydrogen carrier, and it seems like an encouraging alternative for hydrogen production. In hopes of a fully independent plant there is still room for research, specifically in subjects such as a complete techno-economic analysis, the sustainability of these methods, their life cycle analysis along with their impact on the environment.

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