Chapter 1.14. Hydrogen production research in Mexico: A Review

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ABSTRACT

The insertion of the hydrogen technologies into Mexico's energy portfolio is a complex matter. Historically, the government has monopolized the energy sector. However, very recently (August 2014) an energy reform bill approved by the Mexican Congress and ratified as a law by the President allowed foreign investment in the energy sector. In this context, there is a great opportunity for alternative energy and specifically for hydrogen technologies to flourish within Mexico. Specifically, the opportunity for the hydrogen production research activities to contribute to the advance and possible application of the related hydrogen technologies is today of paramount importance. The present paper is aimed to present a review of research activities in the field of hydrogen production in Mexico. Main research activities are reflected in Journal publications and conference proceedings within the last seven years. These resulted in the following topics and contributions: Hydrogen production (HP) from biological processes and wastes 40.4%, followed by HP through conventional and non-conventional fuels (CO_2 capture & Catalysis) 22.4%, HP by photocatalysis & photo-electrocatalysis 14.1%, HP systems and controls 12.2%, theoretical and thermodynamic studies for HP 7.7%, and HP by electrolysis 3.2%. A wide variety of potential applications can be followed by these contributions, while the spread of this research can be a key for future national or international collaborations that may strengthen this important area within the energy sector to take advantage of the upcoming opportunities in the country.

Keywords: Hydrogen production research, Mexico

1. Introduction

Energy is the engine of economic and social development of the world, therefore continuous supply of all forms of energy is of crucial importance to every nation and Mexico as a country is no exception. Hydrogen has become one of the most promising alternative energy carriers in the country, this in view to decentralize the energy production based on oil. The versatility of its new applications, its high calorific value and the fact that it can be used as a clean fuel are some examples of the high potential in new applications and developments. That is the reason why in several European nations as well as in Japan

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hydrogen use in transportation is currently taking place. However, the most abundant element on our planet is not found in its elemental form, but in molecular form (as in oil or water) and consequently in order to be utilized this must be extracted.

At present, there are different methods for producing hydrogen, and these are characterized by their primary source. Among the primary sources are fossil fuels like natural gas and coal as well as renewable sources such as biomass, solar, wind, hydro and nuclear. Additionally, in production technologies there is also many alternatives, such as chemical, biological, electrolytic, photolytic and thermo-chemical processes. The choice of the primary energy source and the technology to produce hydrogen are strongly linked to parameters such as fuel costs as well as environmental and social impacts. Figure 1 show the most employed current technologies to produce hydrogen [1].

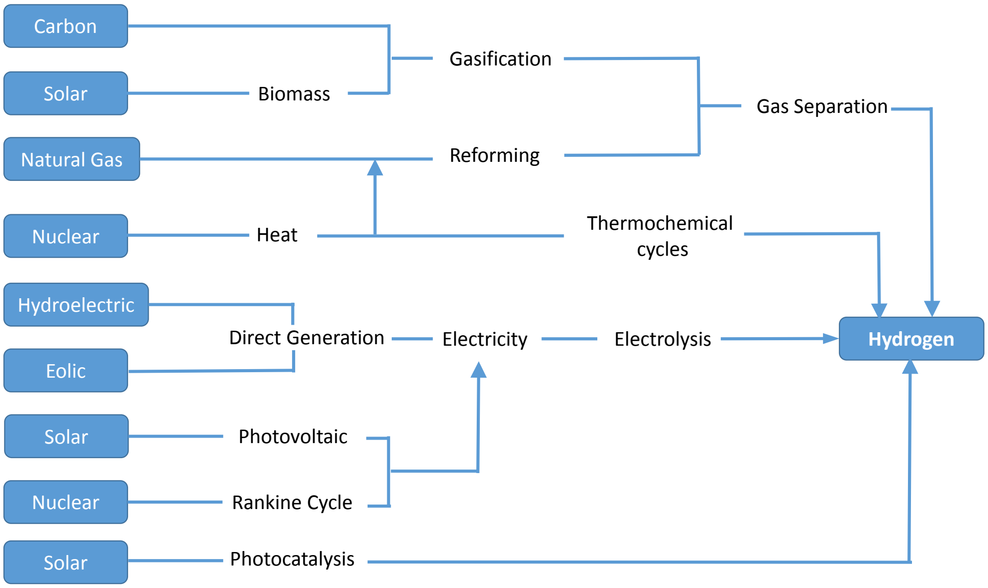

Fig 1. Most employed current technologies to produce hydrogen

In general, electrical and thermal energy can be produced from fossil fuels, nuclear, or recovered energy and renewable energies such as solar, wind, hydro, thermal and biomass. Photonic energy originates from solar irradiation, while biochemical energy is recovered from organic matter. Carbon and biomass (with solar energy as a source) can be gasified to produce syngas (a mixture of H_2 and CO) followed by gas processing and separation to obtain pure hydrogen. Furthermore, natural gas reforming is a mature technology used in many refineries and chemical industries in Mexico for large-scale H_2 production. In some countries (Germany and Japan) small-scale reformers are currently used in demonstration H_2 refueling stations (decentralized production). Reforming options include catalytic steam methane reforming (SMR), partial oxidation (PO) and other variants under development (i.e. CO_2 capture). In SMR, methane reacts with steam at 700°C-950°C to produce syngas, CO is then converted into CO_2, producing additional H_2 by the water-gas shift reaction (WGS). In the POX process, methane reacts initially with pure O_2 to generate syngas [2]. Further purification or gas separation is needed from these processes to obtain pure hydrogen for different needs such as fuel cells in transportation applications.

Hydrogen can be produced using nuclear energy as a source of primary energy, through thermal splitting of the water molecule, electrolysis and thermochemical processes, the three alternatives are free of carbon emissions. The heat obtained from the nuclear reactor is used to achieve the splitting of the water molecule, thus producing hydrogen. Thermochemical cycles were developed already since the 1970s and 1980s when the search for new sources of production of alternative fuels were needed during the petroleum crisis. In thermochemical water splitting, also called thermolysis, heat alone is used to decompose water to hydrogen and oxygen [3]. It is believed that overall efficiencies close to 50% can be achieved using these processes [4]. However, the temperature required for this process is 850 °C; these temperatures are reached only by high temperature reactors [5].

Hydroelectric power has the advantage of being a clean resource that is perpetually renewable and hydrogen production can be carried out by electrolysis. Essentially, this practice is performed to increase the efficiency of hydropower plants through the conversion of water to hydrogen through electrolysis by using the excess energy or wasted energy not yet utilized, and then conversion of hydrogen to electricity via a gas turbine or fuel cells. The electricity produced in the off-peak or no-demand time, or at the time of huge river flows in the spring can be stored in the form of hydrogen, and later, when the peak energy is needed, the hydrogen converted to electricity. One of the advantages of the proposed system is that the resource for production of hydrogen (the water) is available directly at the site. It appears that the conversion of electricity at hydro-power plants to hydrogen, and its utilization via a gas turbine, is technically and economically feasible [6].

Moreover, eolic energy is constantly growing worldwide. In some locations today, wind is cost competitive with conventional, fossil fuel, or nuclear generated electricity. It is the fastest growing renewable energy sector with annual growth of 27%, which means doubling the installed capacity every three years [7]. Direct coupling of an electrolyzer with a wind turbine to produce H_2 implies intermittent operation with a highly variable power output. One particular issue deals with alkaline electrolyzers, when at very low loads the rate at which hydrogen and oxygen are produced (which is proportional to current density) may be lower than the rate at which these gases permeate through the electrolyte, and mix with each other and this may create a hazardous condition inside the electrolyzer [8]. Due to present high costs of electrolyzers and fuel cells, the cost of delivered electricity would be several times higher than the cost of wind-generated electricity [9].

Furthermore, hydrogen production by solar technology can be produced by two methods. Solar energy is converted to electricity in a photovoltaic cell (PV) and hydrogen is generated by electrolysis of water, while the alternative method is carried out in photoelectrochemical cells directly produce hydrogen. Approximately 85% of commercial PV cell technology are based on polycrystalline silicon. Other cells are based on thin plates of amorphous and crystal compounds of group II-IV and I-III-IV of the periodic table [1]. PV based electrolysis process includes photovoltaic (PV) panels, DC bus bar, AC grid, accumulator battery set, electrolyzer and hydrogen storage canisters. PV based electrolysis is one of the most expensive hydrogen production methods; with current technology, the cost of hydrogen from PV electrolysis is about 25 times higher than that of fossil fuel alternatives [10]. However, the cost of this process has been continuously decreasing and this factor is estimated to go down to 6 [11].

Finally, photocatalysis converts photonic energy (solar irradiation) to chemical energy (hydrogen). When a photon encounters the photocatalyst, an electron-hole pair is generated and the obtained electrical charge is employed to dissociate water. In order for a photocatalyst to split water and generate hydrogen, it should have an appropriate band gap and properly located conduction and valance bands for oxidation/reduction reactions. The most studied material so far is TiO_2. In addition to TiO_2, several other semiconductors have been studied, such as, ZnO, Fe_2O_3, $BiVO_4$, and WO_3 [12]. The grand challenge in this technology is to design a catalyst system that can effectively use sunlight and water to generate hydrogen and oxygen [13]. For this technology to be feasible, the development of a photocatalyst must fulfill certain requirements: (a) good (visible) light absorption; (b) high chemical stability in the dark and under illumination; (c) band edge positions that overlap the water reduction and oxidation potentials; (d) efficient charge transport; (e) low overpotentials for reduction/oxidation of water; and (f) low cost [14].

2. Hydrogen production research in Mexico

Hydrogen can be produced from different renewable energy sources, and Mexico is a rich nation in renewable energy resources. Resources, security issues, public acceptance, and appropriate government incentives are the main factors affecting the application of hydrogen technologies as a source of economic growth in Mexico. The inclusion of the hydrogen technologies into Mexico's energy portfolio is a complex matter. Historically, the government has monopolized the energy sector. However, very recently (August 2014) an energy reform bill approved by the Mexican Congress and ratified as a law by the president has allowed foreign investment in the energy sector. In this context, there is a great opportunity for alternative energy and specifically for hydrogen technologies to flourish within Mexico. In 2014, a bill for a general law aimed at the utilization of hydrogen and their related technologies was presented to the Mexican Senate and today this is being analyzed. This bill has the objective to adopt a National Hydrogen Plan with the aim of boosting the energy technological development by hydrogen as a fuel and energy carrier, through scientific and technological research. Therefore, research activities within Mexico are of paramount importance in view of the forecoming utilization of the related technologies. In Mexico, research related to hydrogen technologies is today very active and specifically, one of the major contributions is generated in the field of hydrogen production. In this field major contributions come from the following areas: hydrogen production from biological processes, through conventional and non-conventional fuels (CO_2 capture & Catalysis), by photocatalysis & Photo-electrocatalysis, through systems and controls, theoretical and thermodynamic studies and by Electrolysis. A wide variety of potential applications can be followed by these contributions, while the spread of this research can be a key for Mexico’s future and international collaborations that may strengthen this important area within the energy sector to take advantage of the upcoming opportunities in the country. Therefore, the present paper is aimed to present a review of the hydrogen production research activities in Mexico within the past seven years.

2.1 Biological hydrogen production

Research activities in Mexico focused on biological hydrogen production fall within six main fields: bioreactors their operation and specific applications, microalgae as source for photosynthetic hydrogen, bacteria and microorganisms, fermentation of organic matter and microbial electrolysis cells (MEC).

2.1.1 Bioreactors

In the field of bioreactors, Torres Zúñiga et al. [15-16] developed a strategy that make use of a Luenberger observer to estimate the glucose and the biomass concentrations inside a fermentative reactor to produce hydrogen. This was established in order to accurately determine unknown inputs, which is a key issue in reactor control of biotechnological systems. Ramos et al. [16a] studied several variables that influence the hydrogen production from the organic fraction of a cafeteria food waste at mesophilic conditions in batch reactors. Main variables studied were the initial pH and the total solids concentration (TS). Their results indicate that the highest specific degradation rate was obtained with the lowest TS concentration and an initial neutral pH. While, it was found that the influence of the TS concentration on hydrogen production was more significant than that of the initial pH for this type of residues. Alzate-Gaviria et al. [17] compared two coupled laboratory scale anaerobic digestion reactors for hydrogen production from an organic fraction of municipal solid waste (OFMSW) and synthetic wastewater. These reactors were a packed bed reactor (PBR) containing OFMSW and an upflow anaerobic sludge bed reactor. The reactors were inoculated with a mixture of non-anaerobic inocula and a hydrogen yield of 37% was reached in mesophilic range. In other work, Espinoza-Escalante et al. [18] performed an optimization analysis of the pretreatments of Tequila's stillages (waste) for volatile fatty acids (VFA's) and hydrogen production. It was found that hydrolyzation of the organic matter present in Tequila's stillages is the adequate treatment to enhance the VFAS's and hydrogen production. While, Buitron et al. [19] studied the H_2 and methane production in a two-stage sequencing batch (SBR) and up-flow anaerobic sludge blanket (UASB) reactors using tequila vinasses achieving an overall organic matter removal of 73–75% and the optimization of such reactors were analyzed in a further study [20]. Furthermore, Carrillo Reyes et al. [21] proposed strategies to cope with methanogens in hydrogen producing UASB reactors by analyzing the community dynamics. In another study, Espinoza Escalante et al [22] analyzed the effect of pH, temperature and hydraulic retention time on the production of hydrogen and methane in the anaerobic digestion of vinasses from the fermentation of Agave tequilana Weber to tequila.

Moreover, other kind of bioreactors studied were stirred tank reactors (CSTR). Cheese whey (CW), as an industrial waste also constituted the object of several studies towards the biological production of hydrogen. That is the case of the study dealing with the improvement of the hydrogen production rate in a continuous CSTR [23]. In that study it was found that continuous fermentative Bio-H_2 production from CW can be significantly enhanced by an appropriate selection of parameters such as hydraulic retention time and organic loading rate. While, start-up and operation analyses were performed on a CSTR for biohydrogen production from restaurant organic solid waste [24]. Furthermore, a biotrickling filter was employed as a bioreactor using oat straw acid hydrolysate with some H_2 production being reported [25]. However, implementation of strategies for biomass control to avoid reactor clogging was needed.

The comparison of in continuous and discontinuous reactors was studied for the production of H_2 in bacterial communities [26]. It was found that the feeding-regime conditioned the diversity and bacterial population abundance, while high H_2 production was obtained when low diversity was observed. Using a fluidized bed reactor, Muñoz-Paez et al. [27] studied two levels of operational temperatures (ambient and 35 °C) and two organic volumetric loading rates. They found that the H_2 production in the reactor operated at ambient temperature was superior to that of the mesophilic units and showed encouraging results for H_2 production in submerged fermentation of moderate concentration of sucrose. Whereas, biofilm reactors were used to study the biohydrogen production from diary processing wastewater [28] with dried stems of *Opuntia imbricata* were used as substrate along with a pretreated mixed culture for biofilm formation. Hydrogen yield was significantly affected by initial COD concentration, temperature and dairy wastewater pH. Also, the use of anaerobic sludge blanket (UASB) reactor was used for the study of effect of hydraulic retention time (HRT) on the H_2 production employing paper industry wastes [29]. Finally, in a study aimed to evaluate the application of hybrid membranes for the enrichment of hydrogen gas produced in biodigestors Quechulpa-Pérez et al. [30] showed a feasible separation of these gases in a membrane reactor set.

2.1.2 Microalgae

Microalgae are photosynthetic microorganisms that convert solar energy into chemical energy, which can be redirected physiologically to produce hydrogen (H_2). Photosynthetic H2 production by microalgae has many advantages, such as CO_2 sequestration; bioremediation of wastewater when it is used as culture medium to grow algal biomass; microalgae growth is not seasonal; cultures can be installed in non-arable lands; and depending on the microalgae species, valuable byproducts can be obtained. One of the most significant challenges in hydrogen (H_2) production by microalgae is the incompatibility between oxygenic photosynthesis and anaerobic hydrogen production due to the high sensitivity of hydrogenase to oxygen (O_2). Marquez Reyes et al. [31] studied the reduction of photosynthetic O_2 in a culture medium to enhance the production of hydrogen. A remarkable finding of this work is that when the photosynthetic O_2 in the culture medium is depleted (anaerobic conditions), it is possible to produce hydrogen. Moreover, Martin del Campo et al. [32] studied the hydrogen production by *Chlamydomonas reinhardtii* under light-driven and sulfur-deprived conditions, using biomass grown in outdoor photobioreactors at the Yucatan Peninsula, while Marquez-Reyes et al [33] explored the hydrogen production by native species of microalgae isolated from México. Finally, Martin del Campo and Patiño [34] designed a prototype for the bioproduction of H_2 by *Chlamydomonas reinhardtii*.

2.1.3 Microbial hydrogen production

Mexico is a rich country in terms of natural resources. From the exploitation of these resources, an important amount of biological waste is generated, while most of this is not adequately handled towards its utilization in the energy sector. This strategy can be of relevant importance in this critical sector in Mexico’s economy. This is the reason why an important research effort is being taking place in the country in order to take advantage of such industrial wastes employing the microbial route for H_2 production.

One important agricultural waste in Mexico is the wheat straw that has been subjected of several important investigations. Diverse microbial consortia were designed in order to efficiently degrade recalcitrant lignocellulosic substrates from wheat straw for hydrogen production [35]. Perez Rangel et al. [36] studied the natural anaerobic consortia performance for hydrogen production from wheat straw under isolation conditions, while Torres-Aguirre et al. [37] studied the effect of the C/N and C/P ratios over the fermentation of wheat straw. In another study, hydration treatments were performed to increase the biodegradability of native wheat straw for hydrogen production by a microbial consortium [38], while the combined effect of particle size and hydration treatment on the wheat straw biodegradability was studied by Lara Vazquez et al. [39].

Different kinds of bacteria have been employed for the production of H_2, which is the case of diazotrophic Burkholderia species [40]. In another study, Arreola Vargas et al. [41] explored the hydrogen production from acid and enzymatic oat straw hydrolysates in an anaerobic sequencing batch reactor by evaluation of the performance and microbial population analysis. Other bacteria extensively used by Mexican researchers in the H_2 production is *Escherichia coli* using cheese whey as an industrial waste as substrate. In particular, nitrogen sources impact on the hydrogen production was evaluated [42], while maximum yield obtained was comparable to the yield reached in other hydrogen production processes with *Clostridium sp*. or mixed cultures [43], furthermore, the influence of pH control on hydrogen production was studied by Rosales Colunga et al. [44-45].

In other studies, the buffer composition effect over the hydrogen production and the microbial community composition in non-axenic cultures, it was concluded that the formulation of culture media had a strong effect on hydrogen production, kinetics and also on the microbial diversity [46]. The cyanobacteria *Spirulina maxima 2342* was autotrophically obtained in a bioreactor under illumination and air bubbling and analyzed for its photobiological hydrogen production [47], while in a further study the quantification of hydrogen production was performed at different light intensities and through a PEMFC [48].

Moreover, Valdez-Vazquez et al. [49] researched how nutrients related to spore germination improved H_2 production from heat-shock-treated consortia. Cepeda-Rodríguez et al. [50]employed a natural substratum (*Opuntia imbricata*) to immobilize a microbial mixed culture for H_2 production under anaerobic conditions on biofilms. While Herrera Ramirez et al. [51] studied the use of biofilms to produce H_2 by exploring on the influence of pH in the fermentative hydrogen production in batch reactors, by simultaneous saccharification and fermentation (SSF) from paper industry wastes using anaerobic biofilms. Also from industrial paper waste, the study of Morales-García et al. [52] who evaluated different natural microbial consortia for H_2 production from crystalline cellulose. While, Navarro Diaz et al. [53] evaluated the differences in ecological structure between H_2 production under anaerobic digestion consortia.

2.1.4 Fermentation

Plenty biomass from various industries wastes can be used as a source for biohydrogen production where the combination of waste treatment and energy production can be a valuable asset in Mexico's energy portfolio. This section summarizes research activities related to the fermentative biohydrogen production from biomass. Potential biomass that could be the source for biohydrogen generation are glucose wastes, cellulosic materials, dairy wastes, among others, while a special emphasis was given to the microorganisms and factors affecting the biohydrogen production.

Davila-Vazquez et al. [54] used lactose, cheese whey and glucose in batch experiments for the fermentative hydrogen production. Rosales Colunga et al. [55] studied the fermentation of lactose and its constituent sugars by *Escherichia coli* and its impact on the hydrogen production. In addition, this research group explored the estimation of hydrogen production in genetically modified *E. coli* fermentations using an artificial neural network [56]. In another research, anaerobic mixed cultures were evaluated towards the H_2 production among them anaerobic microbial communities and their H_2 production inhibition mechanisms [57].

Whereas, the solid substrate fermentation of agroindustrial wastes and the influence of total solids content and initial pH on batch operation were evaluated by Robledo-Narváez et al. [58]. Furthermore, Moreno-Dávila et al. [59] studied the fermentative hydrogen production by anaerobic biofilms from a pretreated mixed microflora in a continuous fixed bed-reactor, in this research microbial cultured community was mainly formed by lactic acid bacillus from *Phylum firmicutes*. Whereas, Garcia-Lopez et al. [60] used vegetable fermentation products as substrate for hydrogen production by *Rhodopseudomonas palustris*. Alamilla et al. [61] studied the richness, composition and predominance in undefined mixed cultures for fermentative hydrogen production and Montiel-Corona et al [62] explored the effect of flushing method, bicarbonate addition, and outdoor–indoor conditions within the H_2 production by an enriched photoheterotrophic culture using dark fermentation effluent as substrate. Cisneros-Pérez et al [63] employed an inoculum pretreatment to promote differences in hydrogen production performance in EGSB reactors under dark fermentation conditions; they found that inoculum pretreatments allow the selection of bacteria that have better performance in hydrogen production. Finally, Hernández-Rojas et al. [64] reported the effect of the mixed inocula and pH during the dark fermentation process for the production of H_2 using molasses as substrate.

2.1.5 Microbial electrolysis Cells (MEC)

The microbial electrolysis is a biological process that allows the production of hydrogen (H_2) as a result of the cathodic reaction of protons coming from the oxidation of the organic matter contained in waste water. For the application of this technology it is necessary to determine suitable operating conditions that allow to scale-up a microbial electrolysis cell to produce hydrogen efficiently at low cost.

Verea et al. [65] employed a new approach to get high hydrogen production rate in a MEC by achieving in a very short time the process for making the anode's bacteria enrichment for the biofilm formation. In this study, the hydrogen production efficiency was optimized through the change of the electrolyte conductivity and the electrode surface area/electrolyte volume ratio. It was found that the hydrogen production rate increased with the increase of the electrolyte conductivity. In another study, Rivera et al. [66] used a two-chamber microbial electrolysis cell (MEC) to study the organic matter consumption and hydrogen production rate. The chemical oxygen demand (COD) was composed of a mixture of volatile fatty acids (VFAs) present in the effluent of a dark fermentation process. Results indicated that the substrate containing glucose was more slowly degraded because glucose was first transformed into VFAs, and then the VFAs were consumed to produce hydrogen. Finally, Gomez Roquea et al. explored the hydrogen generation in a microbial electrolysis cell (MEC) using two configurations; catalyzed by platinum and biocathode.

2.1.5 Hydrogen production from industrial wastes

Industrial food wastes contain high levels of carbohydrate and protein and these are mostly treated anaerobically. However, lactic acid, biocompost and energy from food wastes can be a valuable strategy for treatment of food wastes. Furthermore, the organic constituents especially carbohydrates in food wastes can be employed as potential substrates for anaerobic hydrogen production [67]. Sugar cane bagasse, cellulosic waste biomass, food waste derived from restaurants, food-processing industries and organic municipal wastes are among the raw materials for the production of hydrogen that Mexican researchers have used in their investigations. Lopez Ortiz et al. [68] used low temperature sugar cane bagasse pyrolysis for the production of high purity hydrogen through steam reforming and CO_2 capture. In this study, slow and low temperature bagasse pyrolysis produced the highest yield of liquid products, while a thermodynamic analysis of acetic acid (main product from this pyrolysis) steam reforming and CO_2 absorption reactions predicted a high purity H_2 gas. Sanchez et al. [69] explored the co-production of ethanol, hydrogen and biogas using agro-wastes. They proposed a conceptual plant design and NPV analysis for mid-size agricultural sectors. Escamilla-Alvarado et al. [70] examined the hydrogen production from organic wastes and re-use of fermented solids to produce holocellulases.

Whereas, Poggi-Varaldo et al [71] reported an overview of hydrogen, fermentation, methane and bioelectricity as key contributions to biorefineries of organic wastes. In another research, Moreno-Andrade et al. [72] evaluated several parameters that affect the H_2 production using organic solid waste in a discontinuous process, such as the hydraulic retention time (HRT), where the *Genus Megasphaera* was found to be the dominant genus in the microbial community.

Escamilla-Alvarado et al. [73] used municipal solid waste biohydrogen production through solid substrate fermentation. They found that in the semi-continuous fermentation, the factors that had significant influence on hydrogen production followed the order: total solids > mass retention time > temperature. While, in the batch fermentation, supplemental nitrogen did not show a significant effect.

2.2 Hydrogen production from conventional and non-conventional fuels

Conventional processes (commercially mature technologies) for H_2 production have been used for years to obtain large-scale hydrogen (steam reforming, coal gasification and partial oxidation of hydrocarbons) are characterized by having high operating costs, be energy intensive, low efficiency and large CO_2 discharges. However, these can be considered as a bridge technology, while new photocatalytic sustainable technologies as water electrolysis, bio-hydrogen, etc., are able to produce H_2 in large quantities with acceptable yields and efficiencies to be used in combination with other alternative energy sources (wind, solar, hydro, etc.). Specifically, traditional methods of H_2 production can be modified by the CO_2 capture at high temperature. Among the most important advantages of this strategy are: the use of extensive experience in operating these processes, as well as the reuse of existing infrastructure in Mexico. Additionally, derived from these modifications, it is possible to reduce emissions of carbon dioxide, resulting in processes that are more efficient. Thus, reducing the effects of global warming. Consequently, changes to conventional processes, which result in greater efficiencies in the production of hydrogen, have become the essential bridge to the hydrogen economy.

Escobedo Bretado et al. [74, 75] studied the absorption enhanced water gas shift (AEWGS), which is a reaction that combines the WGS reaction and CO_2 capture by a solid absorbent to produce high purity H_2 from synthesis gas in one single step. They obtained a 97% H_2 product using calcined dolomite suggesting this last to act as a WGS catalyst. The use of metal oxide to serve as oxygen source for the partial oxidation of hydrocarbons was explored by de los Ríos Castillo et al. [76, 77]. They used $CoWO_4$ as oxygen source promoted with 10% Ni and 10% La_2O_3 in redox cycles of CH_4 and H_2O to evaluate their global kinetics and found that Ni and La_2O_3 resulted in a greater reaction rate with respect to $CoWO_4$ alone. In a similar study, Sosa Vázquez et al. [78] studied the stabilizing effect of Al_2O_3 and ZrO_2 in mixed metal oxides of Cu for hydrogen production through redox cycles. Results showed that these metal oxides were excellent stabilizers for CuO.

The combination of the steam reforming of hydrocarbons and the CO_2 capture by solid oxides is an important research field that has been very active in Mexico in recent years. Aceves Olivas et al. [79, 80] presented results of thermodynamic analysis and experimental evaluation of hydrogen production by steam reforming of ethanol (SRE) combined with CO_2 absorption using a mixture of a solid absorbent (CaO, CaO*MgO and Na_2ZrO_3) and a Ni/Al_2O_3 catalyst. They found that a H_2 concentration of 96.6, 94.1, and 92.2% using CaO, CaO*MgO, and Na_2ZrO_3, respectively can be produced under this reactions scheme. Lopez Ortiz et al. [81] evaluated process variables for the production of hydrogen using calcined dolomite combined with methane reforming. Since the solid CO_2 absorbent is a crucial material in this modified process, Escobedo Bretado et al. [82] performed an experimental and modeling kinetic study of the CO_2 absorption by Li_4SiO_4, a synthetic absorbent. Finally, thermochemical cycles using biomass and carbon were employed by Valverde-Ramírez et al. [83] to produce H_2 at high temperatures.

2.2.1 Catalysis

One of the energy challenges that today's world is facing is the development of practical alternatives to fossil fuels. With increasing levels of global energy demand, as well as climate change triggered by CO_2 emissions, there is an imminent need to find chemical alternatives (energy carriers) to fossil fuels with smaller carbon footprints. Hydrogen, a clean and energetically rich molecule, is one of the most promising candidates. Heterogeneous catalysis is of paramount importance in today's hydrogen production processes from fossil as well as in renewables fuels. The catalytic research performed in Mexico here is presented in terms of their components with emphasis on noble metals deposited over special oxide particles; the emphasis is given in the activity catalysts for the hydrogen production reactions.

Pérez-Hernández et al. [84] proposed the use of Ag nanowires as precursors to synthesize novel Ag-CeO_2 nanotubes for H_2 production in methanol reforming. They found that the catalytic activity was improved as the nanotubes concentration was increased in the catalysts and Ag was found to be the phase mainly responsible of the hydrogen production. Guevara et al. [85] studied the Ni/Ce-MCM-41 mesostructured catalysts for simultaneous production of hydrogen and nanocarbon via methane decomposition. Four types of carbon nanofibers/nanotubes were detected and their formations greatly depend on the reaction temperature, time on steam and degree of the interaction between the metallic Ni and support. The respective mechanisms of the formation of nanocarbons were postulated and discussed. Galindo-Hernández et al. [86] studied the structural and textural properties of $Fe_2O_3/\gamma\text{-}Al_2O_3$ catalysts and their importance in the catalytic reforming of CH_4 with H_2S for hydrogen production. They proposed a novel mechanism for the methane reforming with H_2S. Martínez-Salazar et al. [87] performed further studies in this field when they examined the kinetics and modeling over $Mo/La_2O_3\text{–}ZrO_2$ catalyst within the hydrogen production by methane and hydrogen sulfide reaction. Further studies examined the high H_2 production from the reforming of CH_4 by H_2S using Mo-Cr supported on heterogeneous catalysts.

In the field of ethanol steam reforming, González Vargas et al. [88] examined the H_2 production over Rh/Ce-MCM-41 catalysts. They found that the incorporation of Ce in the framework of MCM-41 is favorable to the reduction of Rh_2O_3, with the introduction of Ce profoundly enhancing the ethanol conversion and H_2 yield by approximately 2–3 times. Melchor-Hernández et al. [89] studied modification of the nickel supported on La-modified alumina catalysts prepared by sol–gel in ethanol steam reforming. They found that, as lanthanum species are progressively present in the support its contribution to stabilization of the catalyst is certainly related to the enhancement of gasification of carbon residues on the surface of the catalyst but also to a close interaction with nickel particles. Contreras et al. [90] elaborated a complete review on catalysis by the steam reforming of ethanol.

Moreover, Pérez-Hernandez et al [91-93] evaluated the hydrogen production by oxidative steam reforming of methanol over $Ni/CeO_2\text{–}ZrO_2$ catalysts and concluded that nickel is the phase mainly responsible of hydrogen production although the CeO_2/ZrO_2 support reduced the CO formation. While, Ortiz et al. [94] evaluated the effect of WOx on catalysts Co, Ni/hydrotalcite to obtain hydrogen from bioethanol and Pt/ hydrotalcite [95].

Furthermore, Lopez et al [96] studied the effect of Cu and Ni impregnation on ZrO_2 and their molecular simulation studies and fou5d that the bimetallic Cu–Ni and core–shell Ni/Cu nanoparticles on the catalysts, suggest that the oxidative steam reforming of methanol reaction may be a structure–sensitive. While other studies examined the effect of Cu loading on CeO_2 [97].

Moreover, Salmones et al. [98] examined the deactivation behavior of Ni-based catalysts for simultaneous production of hydrogen and nanocarbon and the influence of the pore geometry and they found that the catalysts with 15 and 25 wt.% of Ni showed much longer lifetime, which can be explained by assuming a new model related to the pore geometry of the catalysts. In another work, Pérez-Hernández et al. [99] evaluated the effect of the bimetallic Ni/Cu loading on the ZrO_2 support for H_2 production in the autothermal steam reforming of methanol.

Furthermore, in the methane decomposition reaction Zapata et al. [100] studied the effect of Ca, Ce or K oxide addition on the activity of Ni/SiO_2 catalysts. Their results suggest that Ce addition prevents the sintering of nickel particles during the reduction process maintaining a random distribution between the silica and cerium oxide improving the distribution and migration of deposited carbon.

2.3 Hydrogen production from photocatalysis & photoelectrolysis

The need to develop new alternatives for sustainable energy has drawn the attention to emergent clean renewable technologies, since they proceed from natural and lasting sources like solar light. The alternative method of photocatalytic water splitting to produce hydrogen is promising since it involves the absorption of light to produce H_2 and O_2 by irradiating oxide semiconductors. The incorporation of metals or metal oxide nanoparticles on the surface of semiconductors as co-catalysts has proved to enhance the photoactivity for the water splitting reaction. These photocatalysts must be efficient, stable, harmless, abundant and inexpensive. In this field in Mexico, many research groups intensively work to overcome these challenges. Oros-Ruiz et al. [101] studied the photocatalytic hydrogen production by Au–M_xO_y (MAg, Cu, Ni) catalysts supported on TiO_2 and their results indicate that the electron charge transfer from TiO_2 to the Au–M_xO_y systems and the effect of surface plasmon resonance of gold nanoparticles produce an enhancement of the photocatalytic activity. Also from this research group, the photocatalytic hydrogen production by water/methanol decomposition using Au/TiO_2 prepared by deposition–precipitation with urea was also evaluated [102]. In other studies Galindo Hernandez et al. [103] explored the the role of Fe^{3+} ions in Fe_xO_y/C catalysts for hydrogen production from the photodehydrogenation of ethanol. While, Escobedo Salas et al. [104] evaluated a Pt modified TiO_2 for hydrogen production via water splitting and they found 7.8% quantum efficiency of photon utilization. In Further studies, Perez-Larios et al. [105] improved the hydrogen production from water splitting using TiO_2–ZnO mixed oxides photocatalysts; they found that these solids were resulted in six times more activity than the reference TiO_2 photocatalyst. Torres-Martinez at al. [106] examined the enhanced photocatalytic water splitting hydrogen production on RuO_2/La:$NaTaO_3$ prepared by sol–gel method. The higher activity of the RuO_2/tantalates was attributed to an important reduction in the electron–hole pair recombination due to the effect produced by the presence of La in the $NaTaO_3$ structure as well as by the role of RuO_2 as electron-trap.

Moreover, Valderrama et al. [107] evaluated the photoelectrochemical activity of CIGS thin films for hydrogen production. It was found that under illumination the quantity of hydrogen produced was two orders of magnitude higher than the hydrogen produced in the dark. While, the characterization of screen-printed Ti/CdS and Ti/CdSe photoelectrodes for photoelectrochemical hydrogen production was performed by Morales et al. [108]. Furthermore, Jaramillo et al. [109] showed the application of fiber optics in the hydrogen production by Photoelectrolysis. They proposed an alternative way to transmit concentrated solar energy which may be applied in the hydrogen production by photoelectrolysis. Whereas, Arriaga et al. [110] prepared and characterized (Zn,Cd)S photoelectrodes for hydrogen production.

Furthermore, Huerta-Flores et al. [111] synthesized $SrZrO_3$ powders as photocatalysts for hydrogen evolution from water splitting. Their results confirm that $SrZrO_3$ is a suitable photocatalyst for clean hydrogen generation from water under UV light irradiation, since crystallinity exhibited the greatest effect on the catalytic activity. In other study, Hernández-Gordillo et al. [112] explored the enhanced blue-light photocatalytic H_2 production using a CdS nanofiber. The high photoactivity was attributed to the quantum confinement effect generated by the small particle size of the nanofibers. Ruiz-Gómez et al [113] evaluated the activity of Sm_2GaTaO_7 photocatalyst. They found that the hydrogen evolution activity was enhanced by using 0.2 wt.% of RuO_2 amount, which exceeded 2.4 times compared with the base photocatalyst, Sm_2GaTaO_7. Pérez-Larios et al. [114] used CoO-TiO_2 and WO_3-TiO_2 mixed oxide as photocatalyst for H_2 generation. They found that under visible irradiation, the catalyst with 5.0% TiO_2-CoO produced the best results. Furthermore, this research group also evaluated zeolites type SBA-16 modified with phosphorous as photocatalyst for hydrogen production [115]. Campos Badillo et al. [116] synthesized GaN by hydrothermal method as promising photo-electrocatalyst for hydrogen production. While, Alvaro-Ruiz et al. [117] evaluated the effect of synthesis parameters over the hybrid zinc sulfide photocatalyst. Mendoza et al. [118] studied the photocatalytic decomposition of H_2O using mixed semiconductor Bi_2S_3/TiO_2. Whereas, Arzola Rubio et al. [119] proposed $W_{1-x}Mo_xO_3\bullet 0.33H_2O$ as solid solutions as a tunable band gap photocatalytic material for hydrogen production.

In a fundamental study, Collins-Martinez et al. [120] studied the influence of the anatase/rutile ratio on the TiO_2 photocatalytic activity and concluded that the thermal treatment used to induce the formation of rutile by calcination would presumably reduce water adsorption capacity and surface area, leading to a decrease in photocatalytic activity. In an effort to employ cheaper materials and the utilization of the solar light spectrum, Ortega López et al. [121] studied the effect of $CoFe_2O_4$ as a photocatalyst for H_2 production from water and visible light. They found that cobalt ferrite by ball milling had a higher photocatalytic activity and this was attributed to the vacancies generated during the milling process at which the sample was exposed.

2.4 Theoretical studies

Theoretical studies constitute an invaluable tool towards the understanding and discussion of experimental results in a wide variety of research areas where the hydrogen production is the final target.

In this context, Ramírez-Morales et al. [122] proposed an on-line heuristic optimization strategy to maximize the hydrogen production rate in a continuous stirred tank reactor, while, Ramirez-Morales et al. [123] studied the life cycle assessment of hydrogen production from a high temperature electrolysis process coupled to a high temperature gas nuclear reactor. Furthermore, Blanco-Cocoma et al. [124] proposed a mathematical model for a continuous hydrogen production system on a stirred fermenter connected to a biocatalyzed electrolysis cell. Guillen-Arguelles et al. [125] evaluated the implementation of hydrogen-based technologies in the feasibility study on the use of hydrogen technologies by hotels in the Mexican Caribbean. While a study of the sustainable-hydrogen, production in the Yucatan peninsula was performed by Patiño et al. [126] and Bautista et al. [127] performed a theoretical analysis of the direct decomposition of methane gas in a laminar stagnation-point flow in a CO_2-free production of hydrogen.

2.5 Thermodynamic and energy analyses

Chemical equilibrium in a reaction gas mixture is a dynamic process. A spontaneous approach toward equilibrium is observed when the initial partial pressures of the reactants are high and collisions between reactant molecules cause product molecules to form. Once the partial pressures of the products have increased sufficiently, the reverse reaction (forming “reactants” from “products”) begins to occur. As the equilibrium state is approached, the forward and backward rates of reaction become equal and there is no further net change in reactant and product partial pressures. Furthermore, the total Gibbs free energy function governs the forward and backward directions of chemical reactions. The composition at thermochemical equilibrium can be calculated by minimizing the Gibbs free energy, and so such equilibrium calculations provide a means to estimate the extent to which spontaneous chemical reactions will proceed. Equilibrium calculations provide a best-case effective-catalyst scenario for reaction dynamics without the need for complicated kinetic considerations. Throughout the years thermodynamic and energy analyses have been of paramount importance to assess possible reaction routes and conditions for the proposed hydrogen production processes.

In this context, Romero-Paredes et al. [128] performed an exergy and separately energy analysis of a thermochemical nuclear cycle for hydrogen production. While, Collins-Martinez et al. [129] performed a thermodynamic modeling for the absorption enhanced reforming of light alcohols (methanol and ethanol) for the production of hydrogen. In other study, Gutierrez et al. [130] performed an entropy generation minimization for the thermal decomposition of methane gas in hydrogen using genetic algorithms. Furthermore, Collins-Martínez et al. [131, 132] performed a thermodynamic analysis of the absorption-enhanced autothermal reforming of ethanol.

2.6 Hydrogen production systems

Coupled systems to produce hydrogen and its utilization is a very active research area in view of its implementation in practical energy-related applications. This is the case of the study of Arriaga et al. [133]. They explored the direct coupling of a solar-hydrogen system consisting of a commercial electrolyzer stack and a photovoltaic (PV) solar system of 36 panels (75 W each) of monocrystalline silicon (Siemens) interconnected in a configuration for 2.7 kW power at 48 VDC. In another interesting system, Sanchez-Dirzo et al. [134] proposed a basic system that stores the energy from waves in the form of hydrogen. While, Sebastian et al. [135] modeled a local grid connected integrated solar-hydrogen-fuel cell systems in Mexico. Furthermore, Espinosa-Paredes et al. [136] made a comparative study of the hydrogen generation during a short-term station blackout (STSBO). In addition, Navarro-Solís et al. [137] performed an analysis of the H_2 production by PEM electrolysis, assisted by textile effluent treatment and a solar photovoltaic cell.

Moreover, Aceves et al. [138] proposed safe, long-range, inexpensive and rapidly refuelable hydrogen vehicles with cryogenic pressure vessels. Several other studies have focused on specific applications of hydrogen production and renewable energies [139-140].

Finally, some studies of systems have proposed the utilization of hydrogen as additives to an automobile[142-142] for supplying electrical power to a sustainable housing with a range of 8h/ day and a hydrogen generation system for a PEM fuel cell and performance evaluation using an on-line LabVIEW data acquisition arrangement [143-144], while other research have focused in the use of use of a dielectric-barrier discharge plasma reactor for hydrogen production [145-146], whereas some other systems have proposed the use of recycled waste aluminum for hydrogen generation [147].

2.7 Electrolysis

Ortiz Verdín et al. [148] reported the study of evaluation of rich in nickel alloys for hydrogen generation in an electrolytic alkaline medium. Furthermore, Olvera Vazquez et al. [149] evaluated the electrochemical behavior of mechanical milling synthesized Al_xSn_y in HER (hydrogen evolution reaction). Ortega Chávez et al. [150] performed a mathematical modeling of the the reaction of hydrogen evolution electrodes of Pt/C considering spillovers. In addition, Gutiérrez et al. [151] made an evaluation of electrocatalysts for the production hydrogen from water electrolysis. Finally, Lucatero et al. [152] evaluated the electrocatalytic Properties of NiMo nanoparticles for the hydrogen evolution reaction.

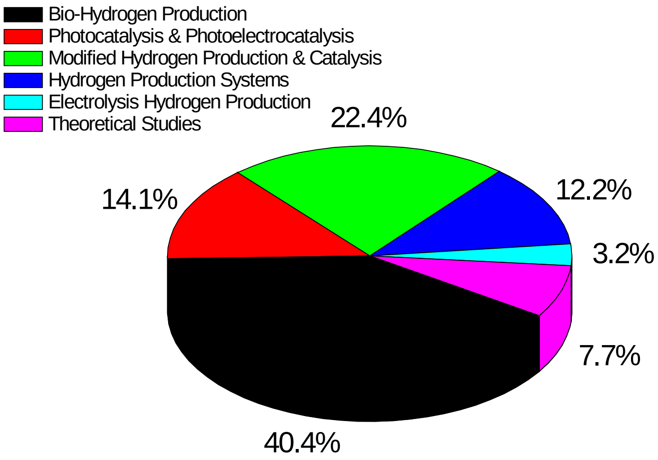

Fig 2. Hydrogen production distribution in terms of research topics in Mexico

3. Conclusion

Figure 2 presents the hydrogen production distribution in terms of research topics in Mexico. From this Figure it can be seen that the distribution of main research activities are quite unique and are reflected in Journal publications and conference proceedings within the last seven years. These resulted in the following topics and contributions: Hydrogen production (HP) from biological processes and wastes 40.4%, followed by HP through conventional and non-conventional fuels (CO_2 capture & Catalysis) 22.4%, HP by photocatalysis & photo-electrocatalysis 14.1%, HP systems and controls 12.2%, theoretical and thermodynamic studies for HP 7.7%, and HP by electrolysis 3.2%. A wide variety of potential applications can be followed by these contributions, while the spread of this research can be a key for future national or international collaborations that may strengthen this important area within the energy sector to take advantage of the upcoming opportunities in the country.

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