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Iranian hydrogen production insight: research trends and outlook

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Abstract

Hydrogen is the best potential candidate to deliver economically reasonable, socially beneficial, and energetically efficient answers to problems associated with the ever-increasing world energy demand and climate change. In this study, different renewable and non-renewable hydrogen production sources and systems are presented comparatively and then partially discussed. Thermal, photonic, biochemical, and electrical are the selected energy sources for hydrogen production. Moreover, the trend of publishing journal articles and registering patents in the field of hydrogen production in Iran and the rest of the world has been presented and compared. The patent registration trend has been markedly upward in recent years. Iranian articles in the field of hydrogen production are categorized based on hydrogen production technology. Due to the vast and affordable sources of fossil fuels, hydrogen production from reforming technology has attracted the most attention. Biomass gasification is another common method of hydrogen production using renewable energy sources.

1. Introduction

One of the major challenges of the 21st century is the growing global demand for energy as a result of population growth and rising living standards. In this regard, in 2011, 15 TWy of energy was consumed by seven billion people on Earth and is projected to reach 30 TWy for nine billion people by 2050 [1]. Figure 1 shows the annual growth of global energy demand and the share of major energy sources in global consumption growth in 2018. Nearly 32 percent of global energy supply growth in 2018 comes from renewable sources, with the rest coming from fossil fuel sources.

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However, it is not expected that the increasing energy needs will be met by fossil resources due to the limited fossil resources and the lack of homogeneous distribution of these resources in the geography of the earth. In line with economic concerns, the consequences of indiscriminate use of fossil fuels, greenhouse gas emissions (generally CO_2), and their role in global warming have raised environmental concerns. Therefore, a shift to non-fossil fuels, which can significantly prevent CO_2 emissions and global warming, is inevitable.

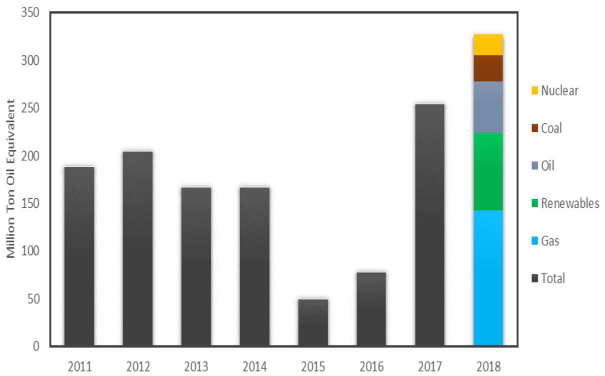

Fig.1. Annual change in global primary energy demand, 2011-2018 (March 2019) [2].

Although technologies like SOFC stacks made of cost-effective materials, with fewer undesirable emissions and using direct natural gas, are fabricated and developed for the near future, it is not enough for the long-term [3]. Hydrogen will be an ideal energy carrier with emissions (greenhouse gases) close to zero or completely zero and with sources that can be continuously renewed. The advantages of hydrogen are 1- High energy conversion efficiency, 2- Ability to produce water without emitting greenhouse gases, 3- Abundant in the world, 4- Ability to store in different ways (including gas, liquid, or in metal hydrides), 5- Ability to move prolonged distances, 6- Easy to convert to other types of energy, and 7- Higher heating value (HHV) and lower heating value (LHV) greater than other fuels. On the other hand, most hydrogen production methods are immature, with high production costs or low production efficiencies. Green hydrogen production technologies with an acceptable cost and efficiency are not currently available. For example, studies on the efficiency and cost of large-scale and small-scale photovoltaic electrolysis of hydrogen production show that photovoltaic electrolysis is expensive with current technologies and that this method has a low conversion efficiency (with energy efficiency and exergy less than 5%); hence, high energy conversion efficiency is not possible at this time [5].

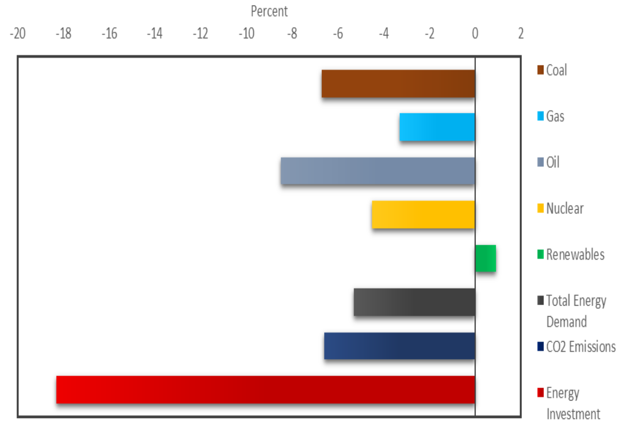

Fig. 2. Key estimated energy demand, CO2 emissions, and investment indicators, 2020 relative to 2019 (October 2020) [4].

In the present study, selected methods of hydrogen production from renewable and non-renewable sources have been studied. H_2 production methods from renewable and non-renewable sources have been introduced and compared. Hydrogen production methods have been selected based on the availability and current status of primary energy sources, technological capabilities, and research interest of institutes in Iran. The main energy sources in this study are electrical, thermal, photonic, and biochemical energies. In general, important methods of hydrogen production are studied, and the trend of publishing journal articles and patents in the field of hydrogen production in Iran and the rest of the world has been presented. Iranian articles in the field of hydrogen production have been studied, and research topics that have received more attention have been introduced.

2. Producing hydrogen from renewable sources

2.1. Electrolysis (electrical decomposition)

Water electrolysis is currently the most important industrial process for producing almost pure hydrogen. Water electrolysis is based on electron movement that is driven by an external bias. The key technologies for hydrogen production by electrolysis are based on alkaline, polymer membrane, and solid oxides membrane electrolyzers [6]. Table 1 shows the general characteristics of alkaline, polymer membrane, and solid oxides electrolyzers. Efficiency and current density as two of the most important parameters are presented in this table. The efficiency of an electrolysis cell is determined by comparing the ideal energies with the actual energy required for the reaction.

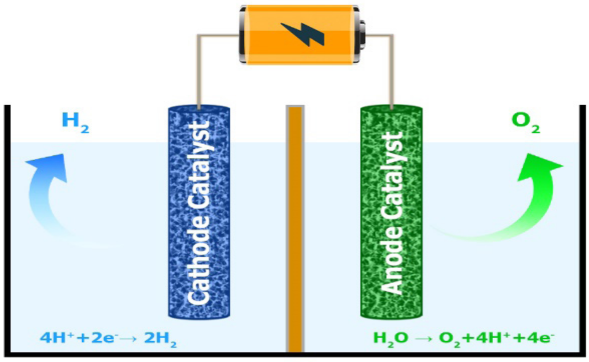

Fig. 3. Schematic illustration of the hybrid water electrolysis system that replaces the OER with other alternative oxidation reactions [7].

Catalysts are used to increase the current density and reaction rate of the electrolysis. Platinum is one of the most widely used heterogeneous catalysts applied to the surface of electrodes [6]. Homogeneous catalysts can also be used in the electrolysis process. Homogeneous catalysts are cheaper than heterogeneous catalysts due to their higher working rates. Because electrolyzers (especially polymer membrane electrolyzers) are very sensitive to water purity, desalination and mineralization processes must be performed before the electrolysis process. However, if the electrolyzer is fed from seawater, along with oxygen, chlorine by-products will be produced. Several methods for preventing side reactions, such as the chlorine production reaction, have been studied. One of these methods is the use of selective ion membranes for the desalination of water. The advantage of solid oxide electrolysis cells is the higher reaction rate due to their high operating temperature [9].

Table 1. Specifications of Alkaline, Polymer membrane (PEM), and Solid oxide electrolyzers (SOE) [10]

Specification	Alkaline	PEM	SOE
Technology maturity	State of the art	Demonstration	R&D
Cell Temperature, °C	80 – 60	80 – 50	-1000 900
Current density, A/cm^2	0.4 – 0.2	2.0 – 0.6	1.0 – 0.3
Cell voltage, V	2.4 – 1.8	2.2 – 1.8	1.3 – 0.95
Power Density, W/cm^2	Up to 1.0	Up to 4.4	-
Voltage efficiency, %	82 – 62	82 – 67	86 - 81
Specific system energy consumption, kWh/Nm^3	7.0 – 4.5	7.5 – 4.5	3.5 – 2.5
Hydrogen production, Nm^3/hr	760>	30>	-
Cold start up time, min	15	15>	60<

The energy efficiency of standard water electrolyzers is strongly limited by the kinetic properties of the anodic oxygen evolution reaction (OER). Recently, a hybrid water electrolysis approach of replacing the OER with more feasible oxidation reactions and coupling with the cathodic hydrogen evolution reaction (HER) has been developed. Figure 3 illustrates the operation of the hybrid water electrolysis cell for electrochemical hydrogen production. Such an innovative strategy improves the efficiency of electrochemical water electrolysis and also eliminates the formation of an explosive H_2/O_2 mixture by producing useful byproducts or inert gas at the anode [7]. Mehrpooya et al. proposed a newly developed system for producing a continuous flow of hydrogen from solar energy. The system includes four main subsystems. These subsystems are linear Fresnel solar collectors, a solid oxide electrolyzer cell, a Rankine power generation cycle, and a thermochemical energy storage unit. This system can produce 50.4 kg of hydrogen in an hour. Results show that the Rankine cycle has the capability of providing 42.78% of the electricity required for the electrolyzer [11].

2.2. Water thermolysis

Water thermolysis is a single-stage water splitting process, which completes through the following reaction:

$$H_2O \xrightarrow{heat} H_2 + \frac{1}{2}O_3 \quad (1)$$

To perform thermal water splitting at acceptable rates, the reaction requires a heat source that can supply temperatures up to 2,500 K. For example, at 3000 K and 1atm pressure, the degree of separation will be 64%. One of the challenges of this method is to separate oxygen and hydrogen gases after splitting. Existing semi-permeable membrane technologies can be used only up to 2500 K, so the gas mixture must be cooled before the membrane separation process[12]. The solar water thermolysis test has been studied at a temperature of 2500 K and detention time of one millisecond, and the system reached 90% equilibrium. The results also show that if the gaseous product cools rapidly (in a few milliseconds) to a temperature between 1500 and 2000 K, it is possible to prevent the recombination of oxygen and hydrogen by using palladium coated membranes; thus, allowing water thermolysis to be used to produce hydrogen [13].

2.3. Thermochemical water splitting

Thermochemical water splitting has the great advantage of not requiring a catalytic component for the hydrogen production reaction. In this method, all chemicals used in the thermochemical cycle are recyclable except for water, which is the main material for hydrogen production Other advantages of thermochemical water splitting are 1- No need for oxygen-hydrogen separation membranes, 2- Suitable operating temperature in the range of 600 to 1200 K, and 3- Little or no required electrical energy [1]. Although different thermochemical cycles have been studied since the 1970s, only some of these methods have been combined with a green energy source for H_2 production.

The S-I cycle seems technically viable, but it must be proven that this method can be commercialized. The first reaction in the S-I cycle takes place with heat and is written as follows:

$$H_2SO_4 \xrightarrow{heat\,(300-500°C)} H_2O + SO_3 \quad (2)$$

Gas products (water and SO_3) are separately heated to 800-900 ° C to decompose SO3 according to the following reaction.

$$SO_3 \xrightarrow{heat\,(800-900°C)} \frac{1}{2}O_2 + So_2 \quad (3)$$

After oxygen separation, SO2, in an exothermic reaction with iodine and water at low temperature, undergo the following reaction.

$$SO_2 + I_2 + 2H_2O \rightarrow 2HI + H_2SO_4 \quad (4)$$

Finally, HI decomposes in the temperature range of 425 to 450 ° C according to the following reaction.

$$2HI \xrightarrow{heat\,(425-450°C)} H_2 + I_2 \quad (5)$$

Figure 4 illustrates the S-I cycle for thermochemical water splitting. Since no adverse reactions occur during the S-I cycle, it is possible to recycle and reuse the chemicals used in the above reactions. Because the S-I cycle reaction temperature is relatively high, nuclear energy, solar energy concentration, and biomass combustion heat can be considered as the few sustainable sources of thermal energy required for the S-I cycle reaction. In the hybrid version of the S-I cycle, the hydrogen production reaction will be electrochemically supported [14].

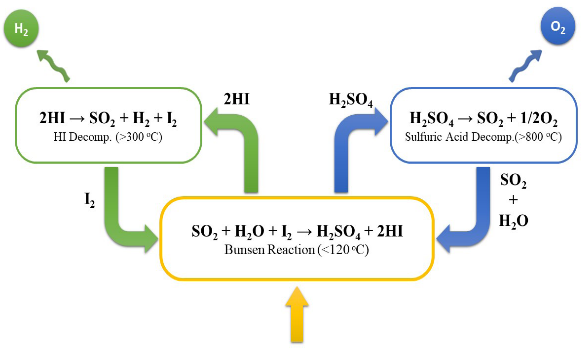

Fig.4. Sulfur-Iodine Thermochemical Cycle [15].

In addition to conventional thermochemical water splitting cycles, it is expected that the NaOH cycle, with a high energy efficiency of more than 80%, will be developed. Moreover, Cobalt chlorine and Boron-based Magnesium chloride cycles operating in the 700–750 °C temperature range are some of the most popular future technologies due to their low operating temperatures [15].

2.4. Thermochemical conversion of biomass, biofuel gasification, and reforming

To use biomass to extract hydrogen, the moisture content must be kept below a certain level by drying or supercritical gasification of the vapor. Some types of biomass include sawdust and cane husks. The general reaction of biomass conversion is as follows:

$$\alpha C_1H_mO_n + \beta H_2O \xrightarrow{heat} aH_2 + bCO + cCO_2 + dCH_4 + eC + fTar \quad (6)$$

In the above reaction, C1HmOn is the general sign of biomass. Tar is an unsuitable by-product that harms the process. Several different catalysts have been used to control, reduce, and prevent the formation of fibers in this reaction. To produce hydrogen, solid biomass enters the following gas production reaction:

$$C_\alpha H_\beta + \alpha H_2O \xrightarrow{high\ temperature\ heat} (\frac{\beta}{2} + \alpha) + xCO \quad (7)$$

Depending on the amount of heat produced or supplied, this process is called autothermal or thermal. In the process of self-heating gas production, the required heat is supplied based on partial oxidation in the gasifier. Hydrogen production from liquid biofuels, such as ethanol and methanol, can also be based on thermochemical reactions [16].

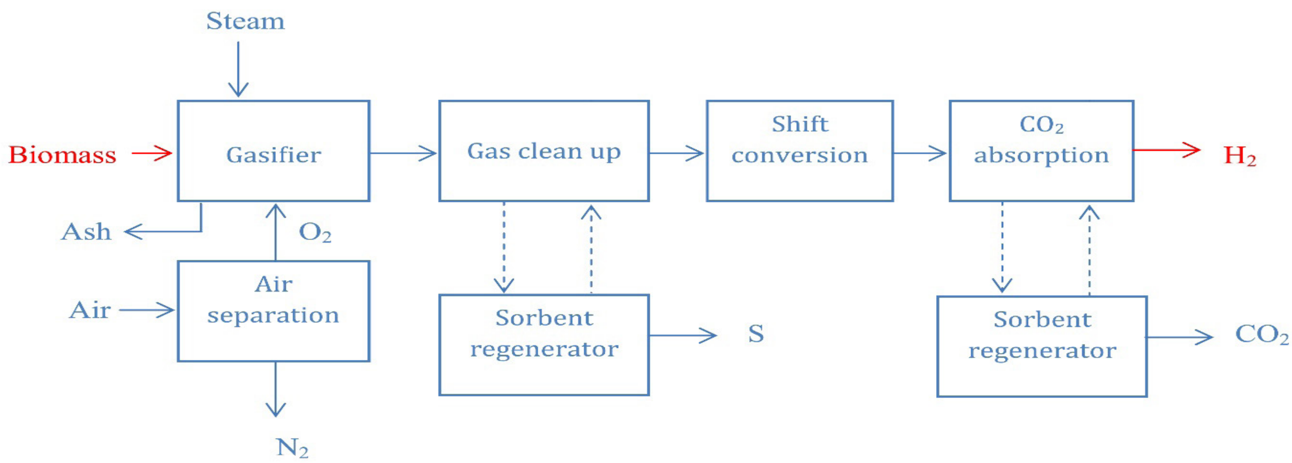

Fig. 5. Schematic illustration of Biomass gasification process [16].

Samimi et al. studied and compared the energy and exergy efficiencies of the gasification process of horse manure, pinewood, and sawdust using conventional gasification agents. Comparisons showed that pinewood steam gasification had much higher exergy efficiency than other gasification agents and biomass sources. It has also been observed that the higher molar ratios of vapor to biomass, higher temperature, and lower humidity produce higher energy and exergy efficiencies for the product gas and lower exergy efficiencies for unreacted carbon and irreversibilities [17].

2.5. Photovoltaic electrolysis

Photovoltaic-based electrolysis is one of the most expensive methods of hydrogen production. With today's technology, the cost of hydrogen produced by photovoltaic electrolysis is about 25 times that of fossil fuel production methods. However, the cost of this process is steadily declining and is expected to drop to 6 times that of fossil fuel in the coming years [18].
Fereidooni et al. have studied the annual performance of the 20 kW power plant installed in Yazd. The results show that in one year, the solar panels have re-

ceived 299,376 MWh of solar radiation on the surface of the panels, and the system has converted 12.32% (equivalent to 36.81 MWh) of this total energy into AC power. The results show that the actual output of the Yazd photovoltaic power plant has the potential to produce 373 tons of hydrogen per year, which can be considered as an average of 48.6 tons per month for real power and 48.3 tons for simulated power [19].

3. Producing hydrogen from non-renewable sources

3.1. Fossil fuel reforming

Steam reforming, partial oxidation reforming, and self-heating reforming (autothermal reforming) are the three main technologies for reforming fossil fuels to produce hydrogen. In addition to hydrogen gas, carbon monoxide and carbon dioxide gases will be present at the end of the reforming process. Steam reforming generally requires an external heat source, but oxygen will not be used to perform these processes. This method has a lower operating temperature and a higher ratio of hydrogen to carbon monoxide compared to partial oxidation and self-heating reforming. In the partial oxidation method, hydrocarbons are partially oxidized to produce hydrogen with oxygen. The heat source for this process is provided by a partial oxidation reaction (combustion). There is no need for catalysts in partial oxidation, and this method tolerates the presence of higher amounts of sulfur compared to steam reforming and self-heating. The pressure required for the self-thermal method is lower than the partial oxidation method, and as mentioned, the partial oxidation and self-thermal methods do not require an external heat source. However, because both of these methods require pure oxygen, the complexity and cost will increase due to the need for an oxygen separation unit. Compared to other fossil fuel reforming technologies, water vapor reforming (specifically methane vapor reforming) is the cheapest and most common method of hydrogen production [20].

Song et al. studied the produced heat of kerosene, petrol, and diesel reformed by different reforming methods through thermodynamic analysis. They concluded that steam reforming has the highest degree of hydrogen enrichment among the three reforming methods, regardless of fuel type. The ideal efficiency of the reforming system can be regarded as a function of the oxygen-to-carbon ratio. Petrol, kerosene, and diesel have similar ideal reforming efficiency under different O/C ratios. The ideal thermal efficiency of the autothermal reforming is the largest among the three reforming methods. The ideal efficiency of the petrol autothermal reforming system is 91.22%, that of the steam reforming system is 89.57%, and the ideal efficiency of the partial oxidation reforming system is 42.97%. The heat of vaporization of consumed water and the heat effect for steam reforming and partial oxidation caused the loss of ideal efficiency of the system [21].

Varmazyari et al. have studied the effect of using Cu-(BDC) MOF support on the selectivity and activity of a copper catalyst in methanol steam reforming. This study has shown that increasing the specific surface area of X-Cu(BDC) (X=La, Y, Pr, Gd, Ce, Sm, and Zn) decreased the carbon monoxide selectivity. On the other hand, the cerium catalyst had the best distribution in MOF pores; and therefore, the nanometer dimension of the catalyst has been preserved [22].

3.2. Plasma arc decomposition

Plasma is an ionized state of matter that carries electrons and atomic components in the excited state. Plasma has the potential to be used as a substrate for high voltage electrical current due to the presence of electrically charged particles. Natural gas decomposes into hydrogen and soot (carbon black) as a result of the thermal activity of plasma (mainly methane). Soot in the solid phase at the bottom of the reactor and hydrogen gas in the gas phase can be collected from the system. In this method, the decomposition reaction of methane to hydrogen and carbon is as follows.

$$CH_4 \rightarrow C_{(s)} + 2H_2 \quad \Delta H = 74.6\ Mj/kmol \tag{8}$$

Fulcheri et al. have studied the above reaction. To do this, they used a thermal plasma reactor with three electrodes connected to 3-phase electricity. Plasma gas is generated around the two electrodes, and then methane gas is introduced to the reactor from above [23].

The results showed the production of 100% pure hydrogen with zero emissions of carbon dioxide. Plasma arc decomposition can be classified as high-temperature pyrolysis. Gaudernack et al. describe the reaction of hydrogen production by plasma with the potential to reduce the cost of hydrogen production by 5% compared to the reforming of methane vapor with the cost of trapping carbon dioxide. The need to use large amounts of electrical energy to produce plasma and pollutants from the natural gas supply, including energy consumption and gas emissions in the stages of extraction, processing, and transmission of natural gas, are the weaknesses of this method [24]. Ghanbari et al. studied the effect of carrier gas flow, applied voltage, and Ni-K_2O/Al_2O_3 catalyst loading on methane conversion to hydrogen via a nano-second pulsed Dielectric Barrier Discharge (DBD) plasma reactor. They concluded that by increasing the applied voltage, the methane conversion and hydrogen production increased while the energy efficiency decreased. In contrast with the optimum condition without the catalytic agent, the presence of a 6 g Ni-K_2O/Al_2O_3 catalyst increased the methane conversion, hydrogen production, hydrogen selectivity, and energy efficiency by 15.7, 22.5, 7.1, and 40%, respectively [25].

3.3. Coal gasification

In the current context, given global technology and reserves, coal is an economically and technologically viable option for large-scale hydrogen production. Compared to existing methods (such as electrolysis), coal gasification is a much more suitable method for hydrogen extraction. In gasification, coal is partially oxidized by water vapor and oxygen in high pressure and temperature reactor, and its main product will be hydrogen, CO, and a mixture of water vapor with CO2 (synthesis gas). In another reaction, this synthesis gas will help produce hydrogen and increase the amount of hydrogen production. The gaseous product is refined in a process to recycle elemental sulfur and sulfuric acid. Part of the synthetic gas can be burnt in gas turbines to generate electricity. Despite the advantages of coal gasification, compared to other hydrogen production technologies, this method leads to the production of high amounts of CO2 due to the high amount of carbon in coal. At present, the hydrogen production efficiency of coal is slightly higher than that of natural gas steam reforming. However, coal gasification technologies have been studied less than natural gas reforming. In the economic sphere, the lower price of raw materials compared to other methods of producing hydrogen from fossil fuels is an advantage of coal gasification [26].

Underground coal gasification (UCG) converts deep coal resources into syngas to use for producing electricity, fuels, and chemicals. Although underground coal gasification is a well-known method, the process has not been implemented commercially outside of the former U.S.S.R. despite many decades of development. The complexity of UCG results from the fact that strong interactions occur between the chemical, thermal, and mechanical processes during gasification. The UCG process is strongly impacted by the properties of the coal, surrounding strata, and the environment. The factors which affect the performance of UCG, such as oxidant choice, coal properties, coal seam depth and thickness, process scale, gasifier design, and site conditions, have been reviewed. Work over the past several decades has shown that the application of the controlled retracting injection point (CRIP) method has many advantages, such as linked vertical wells, over earlier methods and results in superior performance [27].

4. Research trends and outlook

Figure 6 shows the patents issued in the field of hydrogen production. Hydrogen production methods have been patented since 1879, with 19,168 inventions. The patent registration trend has been significantly upward in recent years. This upward trend shows the importance and investment of research in this field. Hydrogen production has also been an attractive topic for researchers in recent years due to its use in multiple markets in various industries. Unfortunately, the authors of the present study could not find any international patents registered by Iranian researchers. However, it should be noted that 49 patents have been registered in the Iranian Intellectual Property Office since 2011.

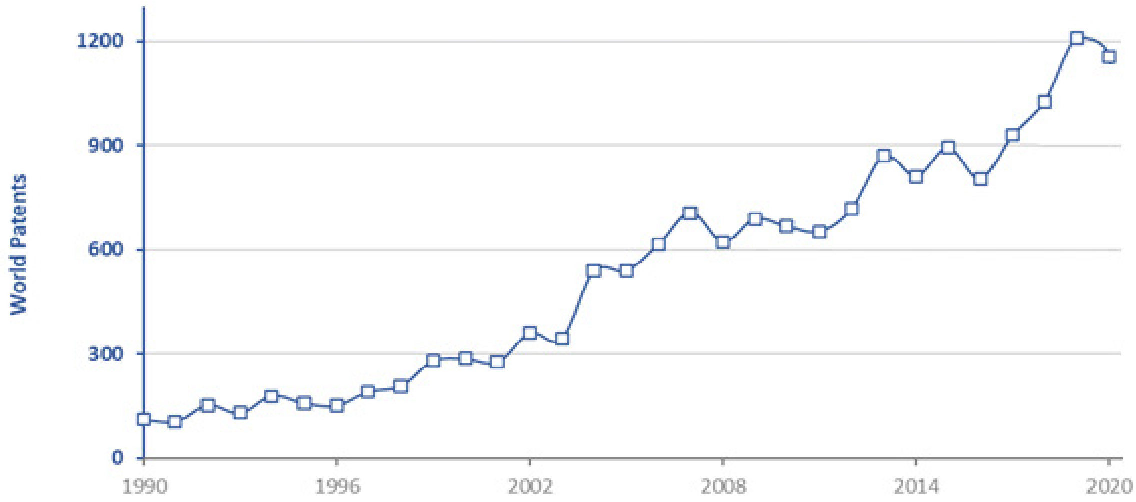

Fig. 6.Comparison of worldwide published patents on hydrogen production.

Studies indicate that in terms of environmental performance, solar-based methods have shown the best and nuclear energy the worst performance in hydrogen production. If all the economic, environmental, social, technical, and reliability advantages of the systems are taken into account, wind-based hydrogen production has the highest performance while geothermal hydrogen production has the lowest. In general, the production of hydrogen by electrical methods had the highest, and biological systems had the lowest performance. According to studies by Acar et al., electrical hydrogen production based on solar energy (such as photoelectrochemical) along with hydrogen storage in nanomaterials can be the most environmentally sound and sustainable choice [28].

The first Iranian article on hydrogen storage was published by Kianvash et al. in 1999. So far, 484 articles have been published focusing on various hydrogen production technologies. Figure 7 shows the published articles in the field of hydrogen storage. This figure shows the articles in Iran and the rest of the world and the number of articles supported by sponsoring organizations. The publication of articles on hydrogen storage has been on the rise in recent years. Research trend analysis has shown that, with the development of technology and the rising price of fossil fuels, renewables will be the most cost-effective source for hydrogen production [29].

Gnanapragasam et al. reviewed research in hydrogen production using coal, biomass, and other solid fuels. Research trends and achievements show that in the next few years, there will be a major shift in research trends toward the conversion of solid fuels into various energy forms. As hydrogen seems to be the highest priority among renewable fuels, extensive research in academic and industrial sectors is expected to make the cost of hydrogen produced from solid fuels more competitive and lower than other sources of hydrogen production [26].

Due to global attention and existing environmental concerns, attention to hydrogen production has increased in Iran.

By comparing the amount of research done in Iran with the rest of the world, the smaller amount of support for research and publishing articles in Iran is significant.

Most of the research funding support in Iran has come from the Nanotechnology Initiative Council. The support of this organization has had a visible effect on the research process in the field of hydrogen production. Hydrogen has a diverseconsumer market, which has led to the consolidation of international financial support for research in this field.

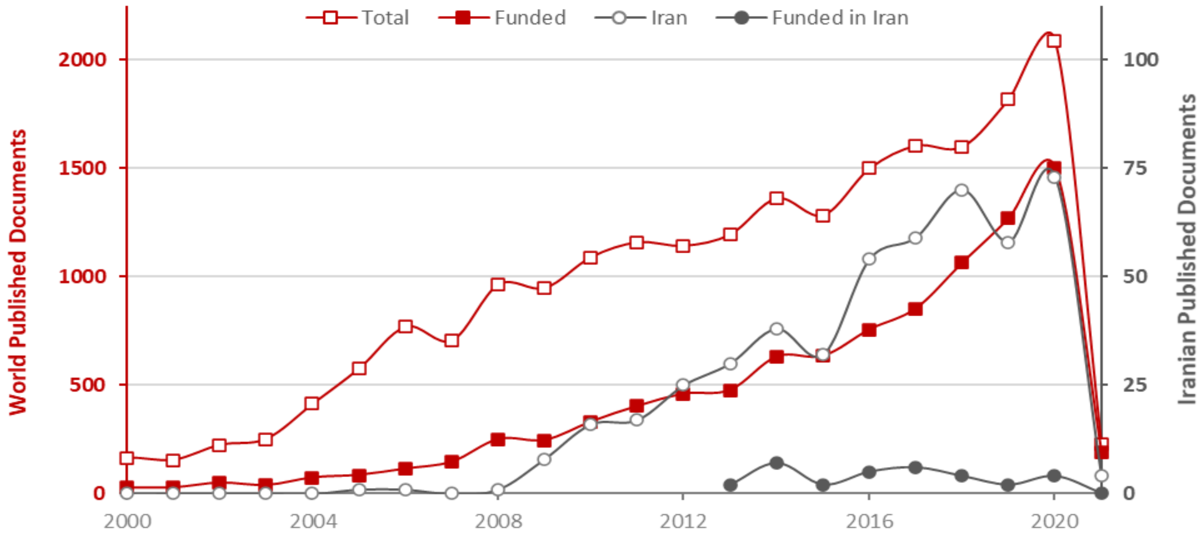

Fig. 7. Comparison of research articles on hydrogen production in Iran and the rest of the world.

Iranian articles in the field of hydrogen production are categorized based on production technology. Figure 8 shows the popularity of hydrogen production technologies in the last three years. Steam reforming, biomass gasification, and fossil fuel reforming have been the most frequent technologies in research articles. Due to the vast and affordable sources of fossil fuels, hydrogen production from reforming technology has attracted the most attention. Biomass gasification has also been the most common method of hydrogen production using renewable energy sources. The production of hydrogen by electrochemical method and using PEM and solid oxide electrolyzers has also been considered due to the high purity of the hydrogen produced.

Hydrogen production methods from renewable energy sources that are of interest to Iranian researchers include biomass gasification, biochemical production of hydrogen, production of hydrogen using photocatalysts, and thermochemical cycle. Iranian researchers have focused on the main energy sources, including fossil fuels (mainly focusing on natural methane), biomass, photovoltaic and solar-thermal, wind, and geothermal, according to the resources available in Iran.

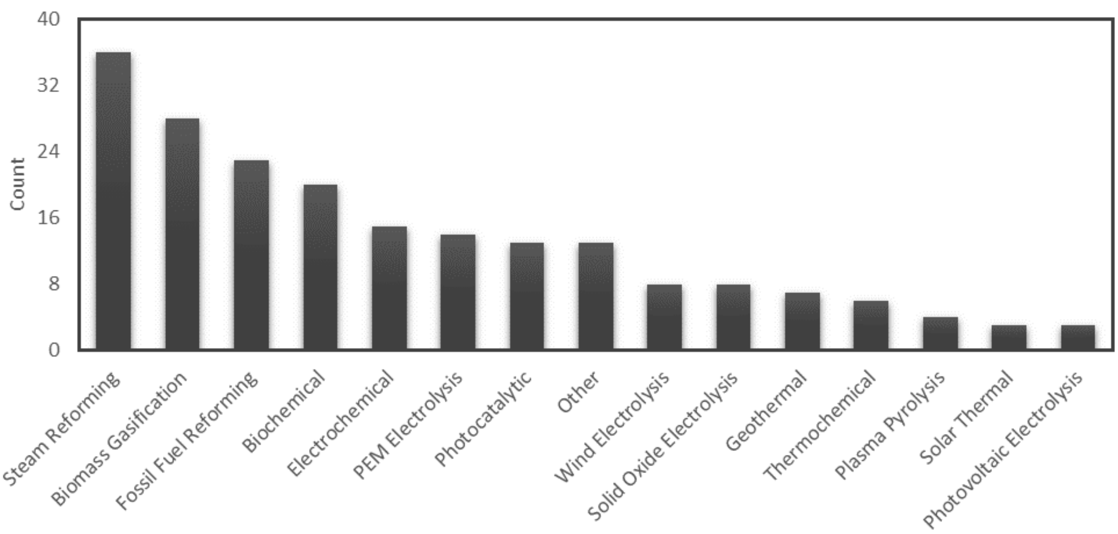

Fig. 8. Attractive research subjects in the field of hydrogen production for Iranian authors 2018 – 2021.

In summary, it seems that due to the limited financial support for Iranian researchers, the small share of Iranian researchers' patent registrations, as well as the breadth of research topics considered by these researchers, there seems to be a lack in policy and planning for research on topics with greater advantages. Accordingly, it seems that policymakers should begin setting medium- and long-term goals in the field of hydrogen production research, and then with the financial support of the research topics of interest, guide researchers to the designated path. Due to the diversity of developed technologies and the diversity of expertise of researchers active in the field of hydrogen production research, there is a promising potential to achieve the latest technologies of hydrogen production in Iran. Since the primary energy sources in Iran are abundant and available, with the guidance of researchers, Iran can become one of the countries supplying hydrogen fuel for the future of the world.

5. Conclusions

Hydrogen production has been an attractive topic for researchers in recent years due to its use in multiple markets in various industries. Due to global attention and existing environmental concerns, attention to hydrogen production has increased in Iran. By comparing the amount of research done in Iran with the rest of the world, the smaller amount of support for research and publishing articles in Iran is significant. Most of the research funding in Iran has come from the Nanotechnology Initiative Council. Steam reforming, biomass gasification, and fossil fuel reforming have been the most frequent technologies in research articles. Since the primary energy sources in Iran are abundant and available, with proper research and guidance, Iran could become one of the countries supplying hydrogen fuel for the future of the world.

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