Hydrogen Production using Solar Membrane Reactor Technology

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Abstract: Fossil fuels as energy resources are decreasing rapidly which will cause a fuel supply crisis in the future. Based on the predictions, global energy consumption has been increased significantly and will continue to rise about two times higher than now till 2050. On the other hand, the extent of the pollutions released from each of the fossil fuels made too many problems for human's lives. Thus, the need for new energy sources has increased recently. The need of affordable and clean alternatives for fossil fuels convinced researchers to test other technologies for energy creation. For example, electrical energy produced by fuel cells is one of the perfect solutions to decrease environmental pollution due to its high efficiency, non-environmental pollution and consumption of hydrogen as a clean fuel. Generally hydrogen can be obtained from various sources such as fossil fuels (natural gas reforming of coal and natural gas), renewable resources, water electrolysis and etc. Of course, each of these processes has its advantages and disadvantages in different conditions. The solar reactor membrane technology in recent years has been proposed as an effective way to produce hydrogen from renewable sources. The purpose of this study is on hydrogen production processes using solar membrane reactor technology. Therefore, various hydrogen production methods have been proposed and the advantages and disadvantages of each method have been analyzed. The combination of membrane technology with solar technology and the developments in solar membrane systems is a good alternative to fossil fuels as a green process for hydrogen production, which is cost-effective in terms of thermal efficiency and fuel consumption rate.

Keywords: Hydrogen production, Membrane reactor, Solar membrane reactor, CSP-Plant, Solar membrane reforming.

1. INTRODUCTION

Due to population growth and energy demand, hydrogen production has become one of the main topics of interest for researchers. Hydrogen can be used in many applications such as desulfurization of natural gas, synthesis of ammonia, methanol synthesis and fuel in fuel cells. 40 million tons of greenhouse gases have been released into the atmosphere because of fossil fuels annually. To minimize the pollution rate, fossil fuel's consumption must be controlled. For this reason, scientists have done extensive studies for renewable energy resources utilization. Solar energy is one of the most appropriate renewable energy sources for exploitation and this study will discuss the methods of using this energy. Hydrogen production by conventional methods caused lots of pollutions and contaminations including ozone layer destruction, acid rain and excessive production of greenhouse gases. Consequently, this will increase the temperature of the Earth. So production of hydrogen using green methods has become more important [1]. Current method of hydrogen production is natural gas reforming that create a large volume of greenhouse gases. About 50% of global demand for hydrogen is supplied by this method, 30% by refining of crude oil in refineries, 18% by coal gasification, 3.9% by water electrolysis and 0.1% is also provided by other methods [2]. Choosing the appropriate method for hydrogen production is based on the limits of these methods.

Some of these limitations are as follow:

1. Feed's local restrictions
2. Technology restrictions
3. Hydrogen market and its price restrictions
4. The relevant political issues

Therefore, in the following the details of hydrogen production methods will be discussed.

2. BACKGROUND OF VARIOUS HYDROGEN PRODUCTION METHODS

As mentioned, hydrogen can be extracted from variety sources such as fossil fuels, biomass and etc. The energy required to extract hydrogen can be classified into four categories; Thermal, electrical, solar and biochemical energies. These energies can be supplied by green sources such as nuclear energy, renewable energies and recovered energy from industries [1].

Hydrogen can be used as a clean fuel for power generation in fuel cells. It has been predicted that this fuel will become the main fuel used to generate energy

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due to its high thermal value. Table **1** shows comparison of hydrogen heat value against other conventional fuels. Generally, hydrogen production methods can be divided to two categories; hydrogen produced by non-renewable resources (conventional) and hydrogen produced by renewable resources (green methods) which will be investigated below.

Table 1: Heat Value Comparison of Different Fuels [3]

Fuel	Heat Value $\left(\frac{Mj}{Kg}\right)$
Hydrogen	120
Liquid gas	54
Propane	50
Gasoline	46

2.1. Hydrogen Production by Non-Renewable Energy Sources

Nowadays about 95% of required hydrogen is supplied by non-renewable energy sources. In the following, investigation of different methods of hydrogen production is discussed.

2.1.1. Hydrogen Production by Natural Gas Reforming Process

Natural gas reforming is the most common method for hydrogen production. Typically hydrogen is produced by steam reforming of hydrocarbons such as methane, oil, naphtha, methanol and ethanol. But for steam reforming in most industries natural gas is used as feed [4]. As shown in Figure **1**, natural gas and steam enter as feed into the first fixed bed reactor as known reformer. In reformer nickel and alumina are used as catalyst usually. As it's obvious in reaction 1, H_2/CO ratio is 3.

$$CH_4 + H_2O \rightarrow CO + 3H_2 \qquad \text{Reaction (1)}$$

Then a compressed stream of hydrogen and carbon monoxide which is called synthesis gas is produced. The object is hydrogen separation from synthesis gas mixture. Since hydrogen is non-polar and small molecule, it is difficult to separate, so we prefer to separate other components from hydrogen. Since absorption of carbon dioxide is more convenient than carbon monoxide, carbon monoxide is converted to carbon dioxide during water-gas shift reaction according to reaction 2. In next step, most of carbon dioxide can be separated from gas mixture in amine unit.

$$CO + H_2O \rightarrow CO_2 + H_2 \qquad \text{Reaction (2)}$$

During the mentioned steps, hydrogen with 95% purity will be created approximately.

To achieve higher purities, membranes can be used to bring up the purity to more than 99.5% [5]. Operating condition for this process is a temperature between 700 to 900°C and a pressure between 15 to 30 atmospheres [6]. Restrictions of reforming process are as follow:

* Catalyst deactivation by coke formation

* Heat transfer restrictions

* Environmental pollution and carbon dioxide and nitrogen compounds emissions

According to reports, hydrogen is produced about 100,000 tons per year by this method.

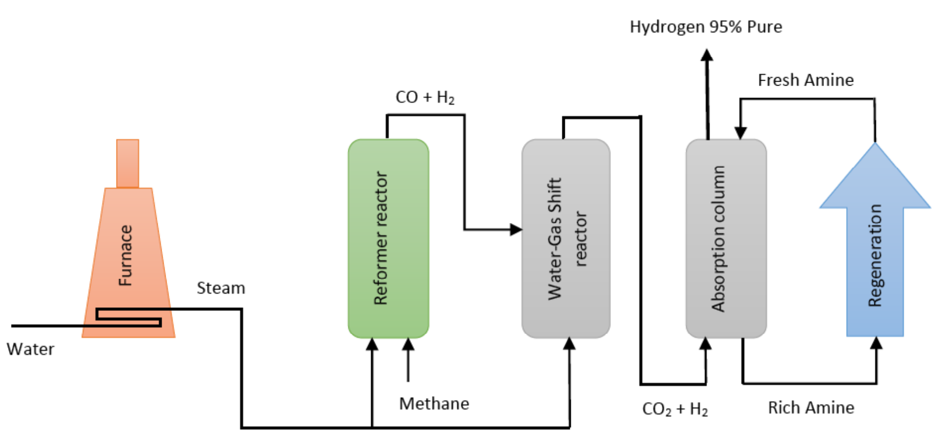

Figure 1: Schematic of hydrogen production by steam reforming process.

2.1.2. Hydrogen Production by Partial Oxidation Process

In partial oxidation process, hydrocarbons are converted to hydrogen through combustion. In order to occur partial oxidation, combustion must be happened in controlled conditions. In normal combustion, steam and carbon dioxide will be produced instead of synthesis gas. Reaction 3 shows Partial oxidation process that the enthalpy of reaction depends on type of hydrocarbon.

$$C_mH_n + \frac{1}{2}mO_2 \rightarrow mCO + \frac{1}{2}nH_2 \quad , \quad \Delta H < 0$$

Reaction (3)

Generally partial oxidation process is exothermic and reaction temperature is about 1100 to 1200 ^{o}C [7]. One of the advantages of partial oxidation process is that the process doesn't require any external heat source. However, this method has limitations such as low H_2/CO. ratio (about 1 to 2), high temperature generated during the process, Coke formation and the amount of entering air control for partial combustion. Since hydrocarbon burning with air causes NO_X emissions, pure oxygen is used instead of air preferably. Figure **2** shows the general schematic of partial oxidation process.

2.1.3. Hydrogen Production by Auto Thermal Reforming Process

Auto thermal process is a combination of partial oxidation and steam reforming process. Since partial oxidation process is exothermic and reforming process is endothermic, combination of these two processes can be optimized in terms of energy consumption. As shown in Figure **3**, auto thermal reactor has three inlet for oxygen, fuel and steam that partial oxidation process takes place when fuel and oxygen in upper

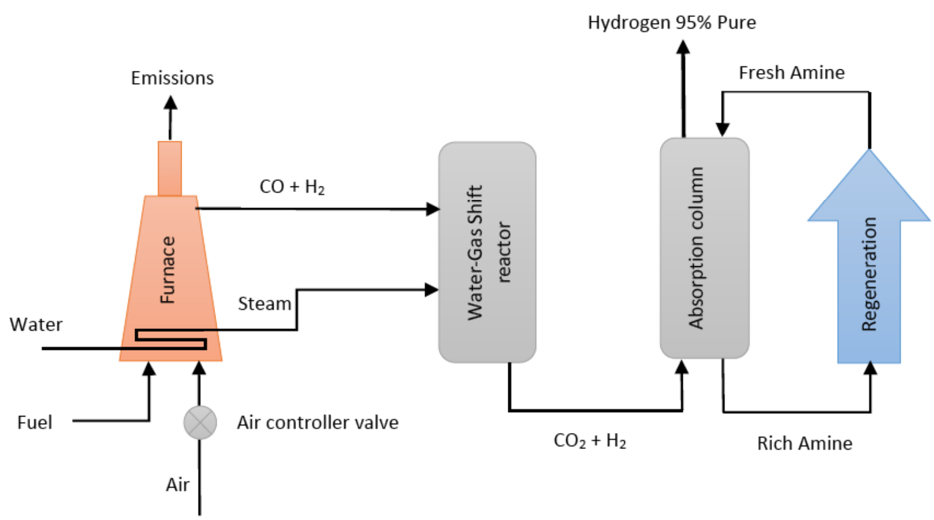

Figure 2: Schematic of partial oxidation process.

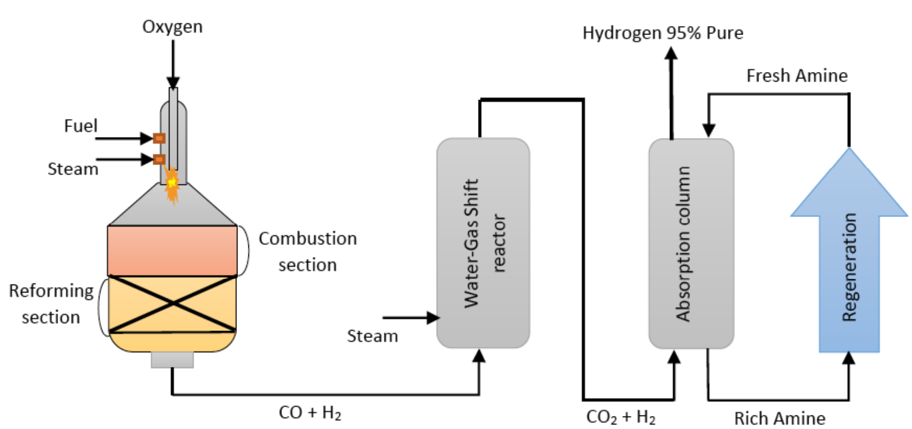

Figure 3: Schematic of auto thermal process for hydrogen production.

part of reactor are entered, then excess fuel and steam are arrived in catalytic section and reforming process occurs. The needed energy for reforming process is funded by the energy produced in combustion section of auto thermal reactor.

To compare three conventional reforming methods (steam reforming, partial oxidation and auto thermal reforming) for hydrogen production, advantages and disadvantages of these methods are shown in Table **2** [5].

2.1.4. Hydrogen Production by Coal Gasification

Among the available hydrogen production methods, coal gasification seems beneficial. During this process, based on reaction 4, coal is partially oxidized at high-temperature high-pressure reactor. The main product of this process is synthesis gas consisting steam and carbon dioxide.

$$2C(s)+4H_2O \xrightarrow{Heat} CO+3H_2+CO_2+H_2O \qquad \text{Reaction (4)}$$

In order to increase hydrogen production, water-gas shift occurs after coal gasification according to reaction 2. Because of the abundance of coal, cost of raw materials is less while total cost of this method is more than conventional hydrogen production methods [8].

2.2. Hydrogen Production by Renewable Energy Sources

As mentioned before, only 5% of hydrogen is produced by renewable energy sources today. Various methods of hydrogen production from renewable energy sources will be discussed.

2.2.1. Hydrogen Production by Water Electrolysis

In this method, hydrogen and oxygen are produced by passing electric current through electrodes in water, water molecules are decomposed. At first based on reaction 5, water molecules decomposed to hydrogen ions, electrons and oxygen. Then based on reaction 6, hydrogen ions combine with electrons and hydrogen gas is produced.

$$2H_2O(l) \rightarrow 4H^+(aq)+4e^- +O_2(g) \qquad \text{Reaction (5)}$$

$$4H^+ +4e^- \rightarrow 2H_2(g) \qquad \text{Reaction (6)}$$

High purity hydrogen obtained by this method and it can be used in pharmaceutical and medical applications. High cost and considerable energy required in water electrolysis are the main disadvantages of this method in comparison with steam reforming and coal gasification [9].

2.2.2. Hydrogen Production by Thermochemical Cycle

During thermochemical cycle, hydrogen can be produced in a nuclear reactor. In this method hydrogen is generated in sequential reactions in a thermochemical cycle. Sulfuric acid decomposed to H_2O and SO_3 at 300 to 500 ° C according to reaction 7 and by raising temperature to 800 ^oC, SO_3. decomposes according to the reaction 8 and oxygen is achieved. The produced SO_2 in reaction 8 enters to reactor with I_2 and H_2O during an exothermic reaction (reaction 9) at low temperature. Produced H_2SO_4 can be used in reaction 7 as feed. Finally based on reaction10, HI decomposes and hydrogen is produced at a temperature of 450 ° C. Produced I_2 can be used in reaction 9.

$$H_2SO_4(aq) \xrightarrow{300-500^oC} H_2O(g)+SO_3(g) \qquad \text{Reaction (7)}$$

$$SO_3(g) \xrightarrow{800-900^oC} SO_2(g)+\frac{1}{2}O_2(g) \qquad \text{Reaction (8)}$$

Table 2: Advantages and Disadvantages of Hydrogen Production Methods [5]

Method	Benefits	Challenges
Steam reforming	I. Extensive performance in industry II. No need for oxygen III. Operating temperature is relatively low IV. High ratio of H_2/CO.	I. High production of emissions II. Costs for large units III. Sensitive to natural gas qualities
Partial oxidation	No need for catalyst	I. Low ratio of H_2/CO. II. High operating temperature III. coke formation
Auto thermal	Average Operating temperature	I. Trade restrictions II. Oxygen needs

$$SO_2(g)+I_2(g)+2H_2O(l)\rightarrow 2HI(g)+H_2SO_4(aq)$$
Reaction (9)

$$2HI(g)\xrightarrow{425-450^{o}C} H_2(g)+I_2(g)$$
Reaction (10)

It should be mentioned that this method is one of the interesting topics for scientists and researchers because of two reasons [9]:

- There is no need for a catalyst.
- Temperature required for reactions is in an acceptable range. (300 to 900°C)

2.2.3. Hydrogen Production by Biomass Gasification

Biomass is a primary source for hydrogen production in gasification method with steam. In case of high water content in biomass, it should be completely dried before gasification. Gasification is conversion of carbon-contained materials to synthesis gas at high temperatures. Biomass are plant based materials like bagasse, wood sawdust and etc. So we can produce hydrogen from materials that are generally waste [10]. Biomass gasification main reaction is as follows:

$$\alpha C_l H_m O_n + \gamma H_2O \xrightarrow{Heat} \alpha H_2 + cCO_2 + dCH_4 + eC$$
Reaction (11)

Acharya *et al*. studied the potential of hydrogen production from steam gasification of saw dust in the presence of CaO and the effect of different operating conditions applied. Results indicated that a drop of 93.33% in CO_2 concentration was achieved when the ratio of $CaO/biomass$ was 2 compared to the gasification without CaO. The concentration of hydrogen was obtained up to 54.43% at 670°C [11].

Due to population growth and the increase global faced in waste generation, it is expected that more attention will be paid to this method to produce hydrogen in future. A major problem in hydrogen production by biomass gasification is the limited sources as feedstock. Since this method is economically feasible on large scale, researches are trying to solve this problem.

2.2.4. Hydrogen Production by Water Thermal Decomposition

Water thermal decomposition is simple one-step process. According to reaction12, water decomposes to its components at very high temperatures.

$$H_2O \xrightarrow{Heat} H_2 + \frac{1}{2}O_2$$
Reaction (12)

Reaction 12 requires heat source with temperature of 2,500 to 3,000 Kelvin. The products of this method are hydrogen and oxygen which can be separated in two ways; separation by semi-permeable membrane or separation by cooling that is the main problem in this step [12, 13]. Water thermal decomposition is not economical and requires more optimization due to its high energy consumption.

Heretofore we discussed various methods for hydrogen production by non-renewable and renewable energy sources. Since one of the environmental problems is the excessive consumption of fossil fuels, hydrogen production by environmental friendly methods can be employed for this purpose. Hydrogen burning fuel cells are appropriate alternative for fossil fuels. According to reaction 13, due to exothermic reaction, this process is associated with energy generation.

$$2H_2 + O_2 \rightarrow 2H_2O + Energy$$
Reaction (13)

As shown in figure **4**, energy generated by fuel cells can be saved in the form of electrical energy and used for different goals.

Use of hydrogen as a fuel is still not developed enough and needs more study to provide more efficient and practical methods. One of the new methods for hydrogen production is solar membrane reactors.

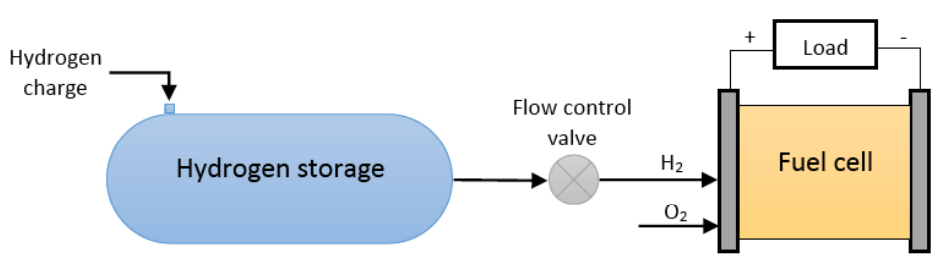

Figure 4: Use of hydrogen as fuel in fuel cells.

Introduce of this technology and how to produce hydrogen using this method will be discussed in the following.

3. MEMBRANE REACTOR TECHNOLOGY

Nowadays membrane has an important role in industry and has become the main mechanism of distillation and absorption in developed countries. One of the applications of membranes is in processes that require ultra-pure products or in processes that end-products are difficult to separate. Although the initial cost is high, but long-term use of this technology is economically justified.

Various methods of hydrogen production were discussed in previous section. As mentioned, hydrogen can be used as fuel in fuel cells to generate energy. Since hydrogen used in fuel cells must be pure, the presence of few impurities can causes great problem in system. Therefore, it is very important to use membranes for hydrogen purification and separation.

About 80% of hydrogen produced by various methods such as reforming, partial oxidation and auto thermal. Since the reactions in these three methods are equilibrated, temperature must be raised extremely to increase conversion of hydrogen that it is consuming lots of energy. Produced hydrogen adsorb by membrane and separation occurs quickly using hydrogen permeable membranes, thus according to Loshatolie principle, thermodynamic equilibrium has never developed and more hydrogen can be produced at lower temperature, and this is the most important advantage of membranes in reactors [14]. Membrane separates the Flow of a component from gas mixture. As shown in Figure **5**, components that are selective in membrane separated in permeate flow. In other words membranes for hydrogen separation are membranes that have high selectivity for hydrogen [15].

Catalytic membrane reactors are consist of a section with membrane which separates desired product from the other components. The separated flow in shell or tube of membrane reactor (depending on type of reactor) driven out as product by vacuum. The catalyst used in membrane reactors, can be either packed or fluidized in reactors.

3.1. Packed Bed Membrane Reactors

The most common type of membrane reactors in researcher's studies is packed bed membrane reactor. Main use of this reactor is in reforming process [14]. Packed bed membrane reactors work structurally similar to shell and tube system and divided in two categories.

3.1.1. Tube Packed Membrane Reactors

In this type of membrane reactors, catalysts are placed inside the tube and desired product separated by membrane in shell part. Tube packed membrane reactors are used if separated flow has high flux. Since shell of reactor has more space than tube, it is more appropriate for separated component that has high flux.

3.1.2. Shell Packed Membrane Reactors

In this type of membrane reactors, catalysts are placed inside the shell and desired product separated by membrane in tube part. Shell packed membrane reactors are used if separated flow has low flux. Since reaction occurs in shell which has more space than

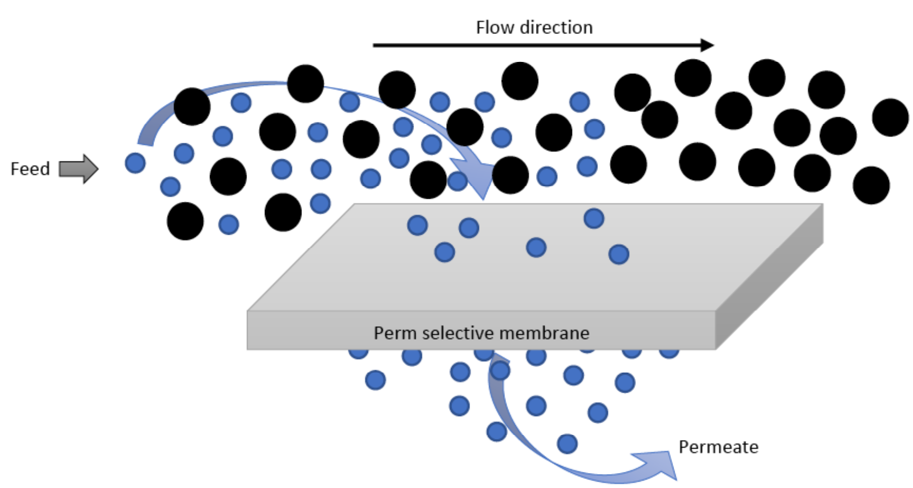

Figure 5: Separation of a component by membrane.

tube, these membrane reactors are more appropriate to separate component that has low flux. Since separated hydrogen flux in reforming method is not very high, shell packed membrane reactor is used for this method [14]. The schematic of packed bed reactors is shown in Figure **6**.

3.2. Fluidized Bed Membrane Reactors

If feed is not gaseous, use of packed bed membrane reactors are not recommended. In other words, if feed is in liquid phase, the conditions for using packed bed membrane reactors are not provided and fluidized bed membrane reactors should be used [14]. For example, in natural gas reforming process, packed bed membrane reactors can be used, but in auto thermal process that the feed is liquid, it is recommended to use fluidized bed membrane reactors [16]. Schematic of fluidized bed membrane reactor is shown In Figure **7**.

4. CONCENTRATED SOLAR POWER PLANT

Due to limited resources for fossil fuels, scientists should develop new methods with low fossil fuel consumption for energy generation. Benefits of these new methods must be more than previous methods or at least be competitive with them. These methods should use renewable energy sources as much as possible to provide required energy. Concentrated solar power plant (CSP-Plant) is a technology that saves solar energy which is clean and endless energy to use in various applications. Receiving and storing solar energy by CSP-Plant has become one of the main methods to use solar energy recently [17]. CSP-Plant is a system with mirrors and lenses which received solar radiation from large area and concentrated them in small area. As shown in Figure **8** concentrated radiations are centralized by parabolic mirrors in their focal length to determine suitable location for maximum energy. Solar energy should be received and stored by an energy receiver.

Usually molten nitrate salts mixture such as $NaNO_3 - KNO_3\left(60-40\right)\%\,wt$ is used and warmed at 550 to 600 ^oC. Tubes containing molten nitrate salts are placed in focal length of parabolic mirrors to receive maximum energy. Molten salts receive solar energy and they are collected in isolated tanks to provide energy for various processes such as producing steam to generate electricity in steam turbine or providing required energy for electrochemical and thermochemical processes. After using in process and

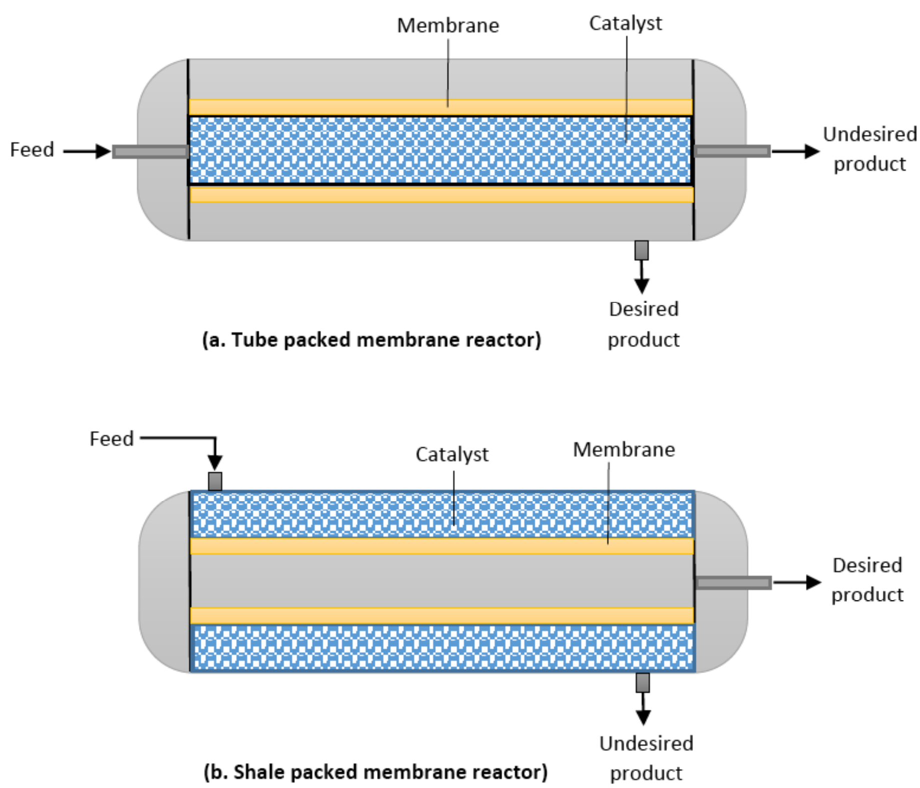

Figure 6: schematic of packed bed reactors.

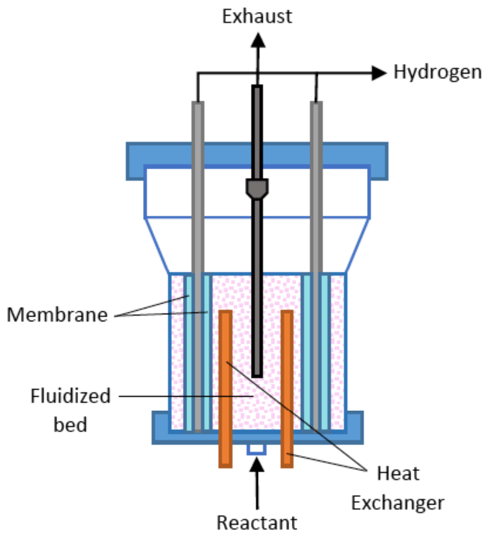

Figure 7: Schematic of fluidized bed membrane reactor.

reducing the energy, flow of molten salt is sent to another tank to get ready to return to the cycle. In these systems there are other methods of energy generation such as fossil fuels combustion that remain standby that when solar energy is not available, process does not stop [17, 18]. The flow of molten nitrate salts retrieves 99% of stored energy and can deliver over 24 hours with constant heat flux. Because of these applications on CSP-Plant, it is also called solar salts [19].

It is expected that power generation capacity by CSP-Plant technology reach to more than 1,500 GW till 2050 [18].

In previous sections membrane technology, solar technology and its details and applications were investigated. Combination of these two technologies results solar membrane technology. This means that advantages of each technology are available in solar membrane technology.

5. HYDROGEN PRODUCTION BY SOLAR MEMBRANE REACTORS

As mentioned in previous section, hydrogen production methods can include a membrane separation stage or can occur in membrane reactors. The presence or absence of membrane separation step depends on purity of required hydrogen. For example, the hydrogen use as a fuel in fuel cells must have high purity, therefore a membrane separation stage must exist in hydrogen production process. Since the main issue is hydrogen separation from final products, the use of membrane reactors that only hydrogen can pass through the membrane, can be effective and can increase the hydrogen purity to 99.9% [19]. Solar membrane reactor is a membrane reactor that it's required energy supplied by solar energy in CSP-Plant. Hydrogen production by water thermochemical decomposition cycle and solar membrane reforming process will be described to know more about solar membrane reactors performance.

5.1. Hydrogen Production by Thermochemical Cycle using CSP-Plant

The required energy for thermochemical cycle is supplied by CSP-Plant. This cycle involves a series of reactions and separation processes. As shown in

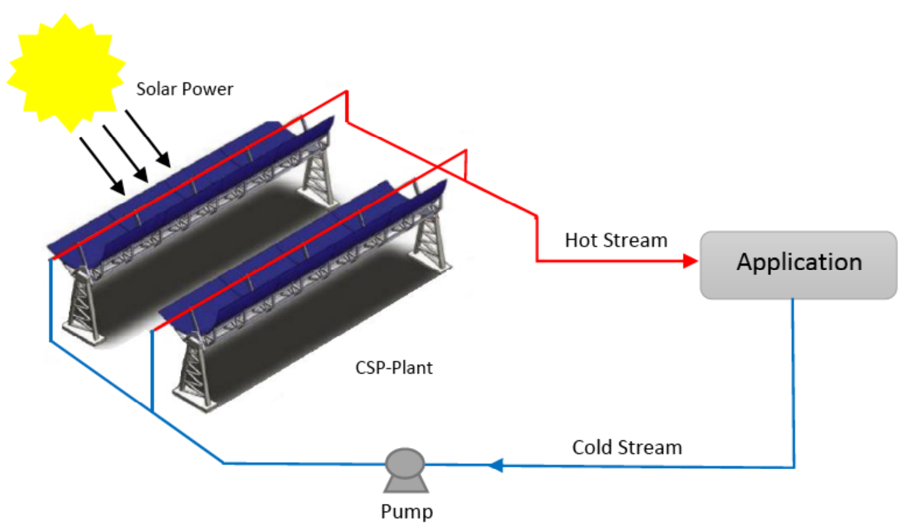

Figure 8: Schematic of concentrated solar power plant.

Figure **9**, H_2SO_4 is decomposed to H_2O and SO_3 where required energy in this step is supplied by CSP-Plant. In the next step H_2O is separated from the mixture in liquid form in a cooling tower and gaseous SO_3 is decomposed to SO_2 and O_2 in solar membrane reactor. Oxygen produced in the process is stored and SO_2 enters to the next reactor with I_2 and H_2O Products are H_2SO_4 and HI In order to complete the cycle, H_2SO_4 returns to the first reactor and produced HI enters to the next reactor. HI decomposition reaction is thermodynamically limited and conversion of the reaction is about 25%, therefore using solar membrane reactor will provide required driving force for reaction and can improved conversion of the reaction. The products of reactor are H_2 and I_2 that I_2 returns to the previous step while hydrogen separated and stored by membrane in reactor [19].

5.2. Hydrogen Production by Solar Membrane Reforming Process

More than 75% of hydrogen, produced by reforming of hydrocarbons. Among the various hydrocarbons as feedstock in reforming process, natural gas is used more than other hydrocarbons. Membrane reforming process is generally endothermic and requires high temperature so conventional reforming process performed by reactors including catalyst tubes inside the furnace, where operating temperatures can reach 1000℃. In such circumstances the conversion of methane is more than 95%.

The use of solar membrane reactors in the process, decrease the operating temperature to about 550°C. Generally in conventional reactors at this temperature, the conversion of methane is less than 25%. But using solar membrane reactor instead of conventional reactors, always provide the necessary driving force for more methane conversion at lower temperature [20, 21]. As shown in Figure **10**, methane and steam enter into the shell of solar membrane reactor. To provide the energy for this process, the flow of molten nitrate salts passes the tube of reactor at 550℃ and re-enter to CSP-Plant after cooling to recover the required energy.

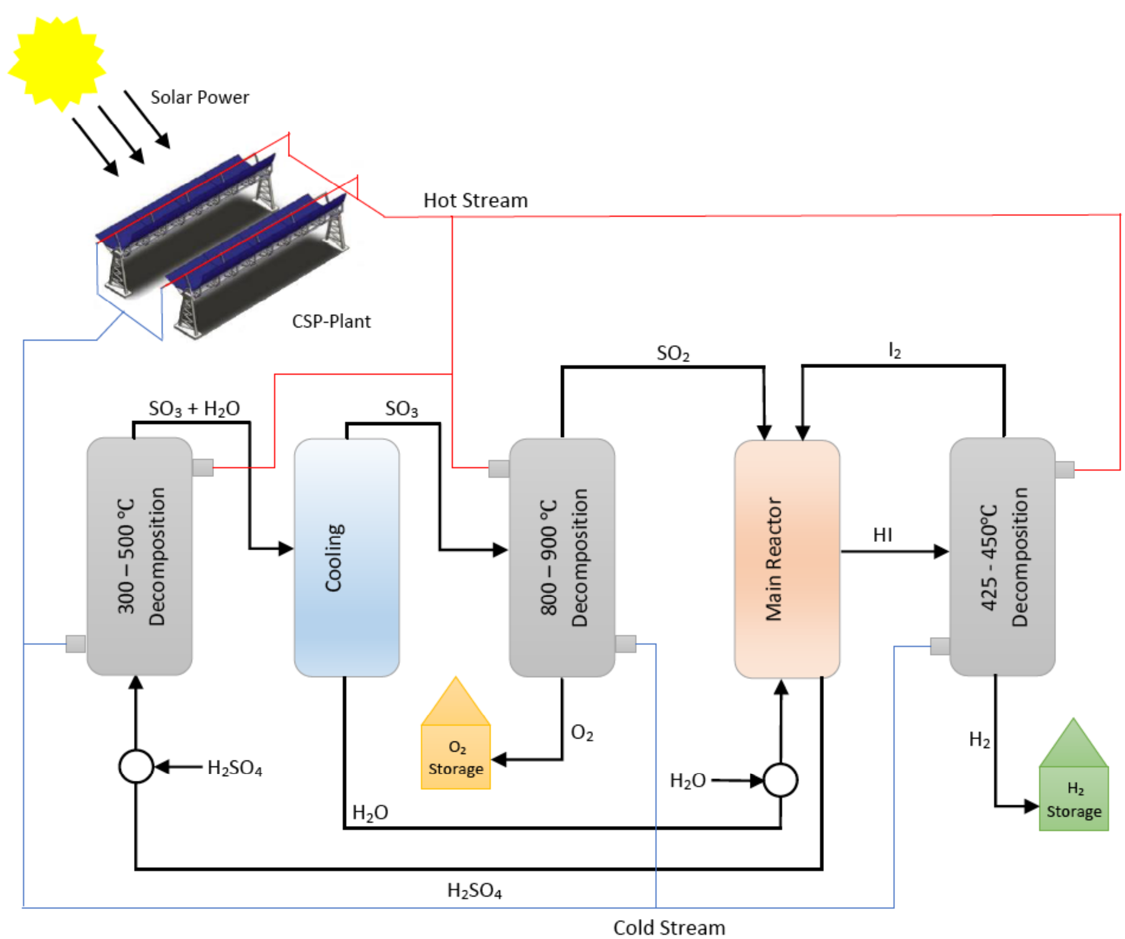

Figure 9: Schematic of hydrogen production by thermochemical cycle using solar membrane reactors.

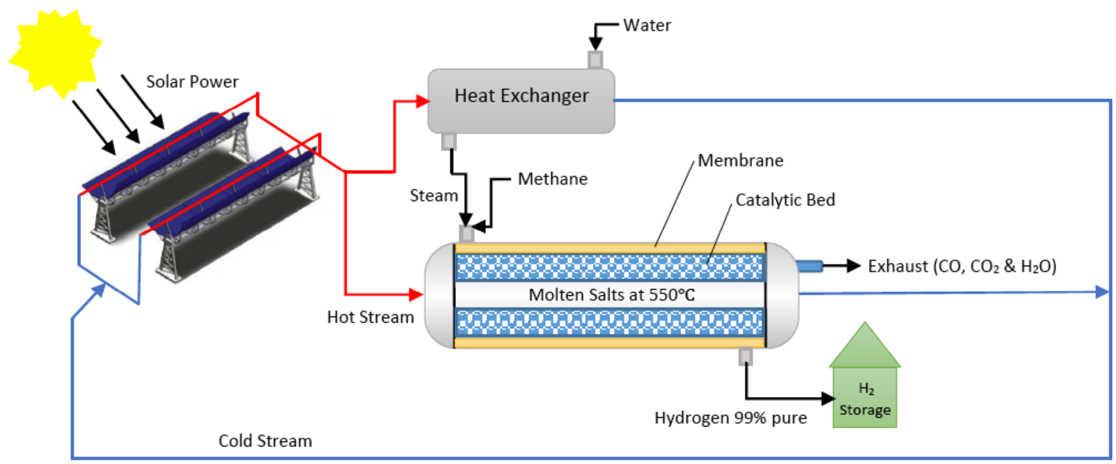

Figure 10: Schematic of hydrogen production by solar membrane reactor.

Reaction 1 is done in shell part of reactor which is filled with granular catalysts and products are CO, CO_2 and excess steam. Hydrogen is separated by membrane and CO, CO_2 and H_2O are exited from the reactor [17]. Cost of hydrogen production by solar membrane reforming is lower than the conventional reforming methods.

CONCLUSION

In this paper, conventional methods for hydrogen production and also combination of membrane technology with solar technology are investigated and advantages and disadvantages of each methods are determined. It is shown that membrane reactor technology can improve conversion of reactions that results more hydrogen production and CSP-Plant as a new source can provide required energy for chemical processes. Hydrogen production methods compared based on thermal efficiency, energy consumption and cost. The concluding remarks of this study can be summarized as following:

- The development of CSP-Plant and its application to provide required energy for various processes can be an appropriate alternative for fossil fuels.

- Using membrane reactors can reduce operating temperature in endothermic processes and necessary driving force for reaction is always provided. Therefore in solar membrane reforming process the required temperature is 550 ^oC instead of 1000 ^oC in conventional reforming.

- Due to the hydrogen separation by membrane in solar membrane reforming process, WGS reactor and amine plant can eliminated that reduces operating unit's volume and reduces costs.

- In last reactor in thermochemical cycle for hydrogen production, presence of membrane reduces the operating temperature to 550 ^oC that can be supplied by CSP-Plant.

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