

Progress in carbon emission reduction technology in fossil fuel-based hydrogen production

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(Received January 8, 2021, Revised July 21, 2021, Accepted August 7, 2021)

Abstract. Today, almost all hydrogen production is based on fossil fuels. Hydrogen production plants contribute to harmful emissions in the atmosphere, which is one of the causes of global warming. In order to obtain hydrogen as an entirely green energy source, there is an urgent requirement to significantly reduce or even completely eliminate carbon emissions from fossil fuel-based hydrogen production processes. In this context, new efforts should be increased to develop hydrogen production technologies that produce lower levels of harmful emissions. The development of carbon capture technology by the chemical cycle offers great potential to reduce harmful emissions generated during hydrogen production from fossil fuels. In this study, hydrogen production methods from fossil sources have been reviewed and the recent studies of chemical looping technology for hydrogen production were presented.

Keywords: carbon dioxide; climate change; fossil fuel; hydrogen; renewable energy

1. Introduction

With the world population projected to reach 9 billion by 2050, global energy use is expected to increase by fifty percent (EIA 2019). This increase in energy demand will gradually increase the dependence on fossil resources, which currently constitute 80% of the world's primary energy production unless an alternative energy source becomes widespread. Thus, many researchers are studying alternative energy sources such as nuclear energy and especially renewable energy sources such as solar, wind, geothermal, wave, etc. (Mostafaeipour and Jooyandeh, 2017). Fossil fuels are limited sources and sustainability issues will become a major problem for the human race in the near future (Abdalla *et al.* 2018). Besides, the high amount of CO₂ and other greenhouse gases resulting from the combustion of non-renewable fossil fuels threaten the future of the world by causing global warming (Singh *et al.* 2019). It has been determined that 90% of CO₂ emissions all over the world are due to fossil fuels (Stenberg *et al.* 2018). As a result of the current situation, the search for alternative solutions to fossil fuels or to reduce toxic emissions caused by fossil fuels has become critical.

Recently, the stakeholders in the energy sector interest in a trend of transition to cleaner and reliable energy sources for a sustainable energy future. Hydrogen which is one of the most studied

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Table 1 Properties of various energy sources (Salvi and Subramanian 2015)

Energy source	Chemical formula	Density (kg/m ³)	LHV (Mj/kg)	LHV (Mj/m ³)	Flammability limits (vol% in air)	Stoichiometric composition in the air (vol%)
Hydrogen	H ₂	0.0898	120	9.9	4-75	29.53
Gasoline	C _{7.6} H _{16.2}	730	44.8	32.704	1.4-7.6	1.76
Methane	CH ₄	0.643	50	32.6	4.3-15	9.48

alternative energy sources can be seen as the primary candidate for that transition. Henry Cavendish was the first to discover hydrogen gas in England in 1766 (George and Agarwal, 2010). The name of hydrogen comes from the words of 'hydro' and 'geinomai' meaning the bring forth water. Hydrogen is the most plentiful element on the planet with its high heating value (Mostafaeipour *et al.* 2017). Moreover, the product of hydrogen combustion is only water vapor. Hydrogen does not exist as a single atom in nature, it is found in the form of compounds with the molecules of water (H₂O), hydrocarbons (C_xH_y), biomass (C_xH_yO_z) among other sources (Salvi and Subramanian 2015, Sinigaglia *et al.* 2017). The properties of various energy sources are given in Table 1.

Hydrogen can be used as an energy source for a system. However, several difficulties occur for direct use in internal combustion engines instead of petroleum fuels as the density of hydrogen is only 0.08988 g/L at standard temperature and pressure (Gong and Verstraete 2017). Therefore, in recent years, hydrogen is utilized mostly as an energy carrier rather than an energy source in mobile applications. Electricity generation by using hydrogen as an energy carrier in fuel cells has been recorded as an important improvement in terms of current energy conversion (Krawczyk *et al.* 2014, da Silva Veras *et al.* 2017). Currently, fuel cell solutions are available that can provide power from several watts to 100 kW. Furthermore, Today, 93% of all hydrogen produced in the world is consumed by the chemical industry such as the production of ammonia, methanol, resins, nitrogen fertilizers, and polymers (Protasova and Snijkers 2016). Considering that the annual hydrogen production is approximately 100 million tons and this figure increases by approximately 6 percent each year, it is predicted that the use of hydrogen will increase in near future (Kalamaras and Efstathiou 2013).

In parallel with the increase in demand for hydrogen, the importance of how hydrogen is produced also increases. As can be seen in Fig.1, two main sources are used for hydrogen production, fossil fuels and renewable sources. For hydrogen to be considered a completely clean energy source, there must also be no carbon release during hydrogen production. However, conventional methods of hydrogen production are fossil fuel-based sources that cause massive emissions of greenhouse gases (Deokattey *et al.* 2013). Recently, new and renewable ways to produce hydrogen have received widespread attention from the research community (De Vos *et al.* 2019). However, currently, around 95% of hydrogen is produced from non-renewable sources (Bion *et al.* 2012). In a long period, the rate of 5% of renewable sources can be increased but in the near future, fossil fuels will carry on to be the main source of hydrogen as green hydrogen technologies have not yet reached reasonable levels of efficiency and cost (Salvi *et al.* 2013).

Considering the issue of hydrogen production with a realistic approach, it is of great importance to carry out innovative studies to reduce carbon emissions in fossil-based hydrogen production technology for a cleaner environment. In this context, the development of carbon

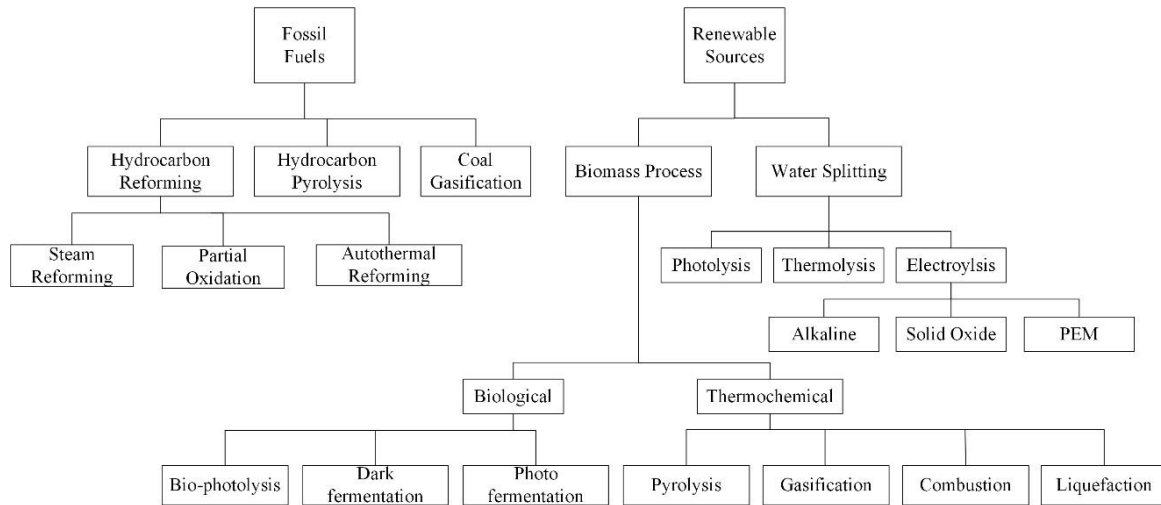


Fig. 1 Hydrogen production methods (adapted from (Shiva Kumar and Himabindu 2019))

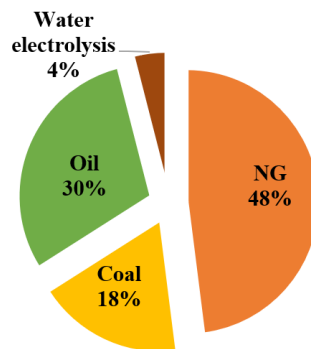


Fig. 2 Hydrogen production distribution from various feedstock (Muradov 2017)

capture technology by the chemical cycle offers great potential to reduce harmful emissions generated during hydrogen production from fossil fuels. In this study, hydrogen production methods from fossil sources with chemical looping technology have been reviewed. The processes of hydrogen production from renewable sources in detail can be found elsewhere (Acar and Dincer 2019, Dincer and Acar 2014, El-Emam and Özcan 2019, Hosseini and Wahid 2016). At present, there are several studies on hydrogen production from fossil fuel sources (Abdalla *et al.* 2018, Dawood *et al.* 2020, Dincer and Acar 2017, Dufour *et al.* 2011, Kang *et al.* 2017, Nikolaidis and Poullikkas 2017, Velazquez Abad and Dodds 2017) however, a comprehensive review of chemical looping technology is limited in the literature. The evaluation of this paper is focused on the reduction of GHG emissions with chemical processes in hydrogen production.

2. Hydrogen production methods from fossil feedstock

The primary hydrogen production feedstock is natural gas (NG) worldwide with 48% as shown in Fig. 2. Oil and coal follow natural gas with a rate of 18% and 30%, respectively (Hosseini and Wahid 2016).

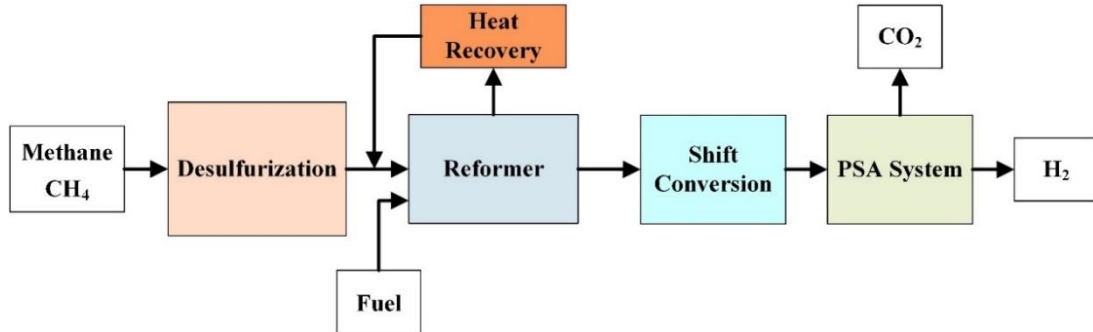


Fig. 3 Steam methane reforming process scheme

When hydrogen production from renewable sources is viewed from a commercial perspective, it can be said that they are not yet at a sufficient level in terms of benefits and costs. (Yilanci *et al.* 2009). For instance, studies about hydrogen production from electrolysis indicated that water electrolysis is more expensive than conventional methods as electricity generation is up to ten times more expensive than fossil fuels. Besides, the energy and exergy efficiencies are less than 5% (Dincer and Acar 2017). For this reason, it can be said that hydrogen production from fossil fuel feedstock will proceed as a primary option for a long time.

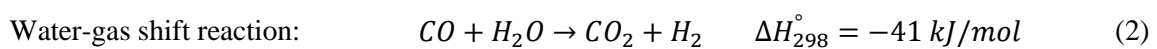
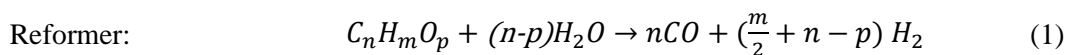
In the next stages of the paper, various hydrogen production methods from fossil fuel-based are discussed with chemical aspects and then the overview of carbon-capturing technologies was presented.

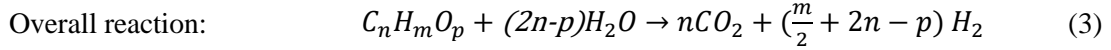
2.1 Steam reforming

Steam reforming (SR) is the most common method over a period of several decades which accounts for nearly half of the hydrogen production all over the world (Kaiwen *et al.* 2018). Currently, natural gas is the primary source of steam reforming of hydrogen. Other raw materials for hydrogen production by steam reform are methanol, butane, diesel, propane, and naphtha (García 2015). The use of nickel as a catalytic component is observed throughout the steam reforming industry, besides, platinum group metals are also used in some applications.

The flow diagram of steam methane reforming (SMR) is illustrated in Fig. 3. In this process, methane is transferred to the reformer to start a reaction with high-temperature steam (800°C - 900°C) in an endothermic reaction (165 kJ mol⁻¹) to produce carbon monoxide and hydrogen. As a result of this reaction, 4 mol H₂ and 1 mol CO₂ are released per one mol methane. After the reforming process, in order to increase hydrogen production, the gas cooled down to 350 °C by a water gas shift conversion unit using catalysts to maximize the efficiency of hydrogen. Nickel-alumina-based catalysts are widely used at the pressure of 3-25 bar during the process (Settar *et al.* 2015).

The general reactions of the steam reforming process are given in Eqs. (1)-(3) (Dufour *et al.* 2011):



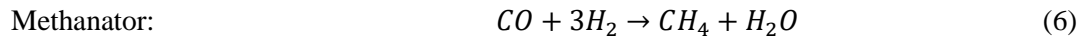
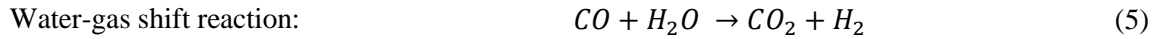
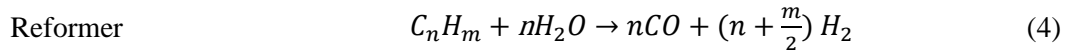


While steam reform is an industrially well-established method, it can be noted that relevant toxic emission production is high (LeValley *et al.* 2015, Román Galdámez *et al.* 2005). About 0.4 million standard cubic meters of carbon dioxide emissions are released into nature in an SR plant that produces 1 million m³ of hydrogen daily. The price of methane raw materials is about 58% of the total cost in large plants, while that number decreases by up to 40% in small plants (Yamaguchi *et al.* 2012).

2.2 Partial oxidations

Partial oxidation (PO_x) which can be expressed as the unfinished combustion of a fuel-rich mixture is another method for hydrogen production (Staffell *et al.* 2019). The reaction is exothermic and indirect heat exchange is not required. For this reason, this system is more compact than SR plants (Ziyad Salameh 2014). As shown in Fig. 4, the system has a partial oxidation reactor, CO shift, and Methanator equipment. The non-catalytic operation of PO_x is more sulfur tolerant than steam reforming and the mixture of natural gas and oxygen is preheated and then combusted with a burner (Zhong *et al.* 2017). This method allows a wider range of fuels to be used, and moves faster without the requirement for external heat, however, the hydrogen production efficiency is lower than steam reforming. Besides, the generated gas requires additional cleaning.

The reactions of the PO_x are given in Eqs. (4)-(6) (Nikolaidis and Poullikkas 2017):



2.3 Autothermal reforming method

The autothermal reforming (ATR) method is a combination of the best properties of steam reformation and partial oxidation systems in a single unit (Demirbaş 2002). Although ATR is not as popular as SR and PO_x, it is more suitable and low cost for relatively small-scale hydrogen production. This method produces more hydrogen than PO_x while providing faster start-up and response time than the SR method (Park *et al.* 2008). Unlike the SR process, ATR does not require an external heat source. As shown in Fig. 5, in this process, steam and air are sent into the reformer

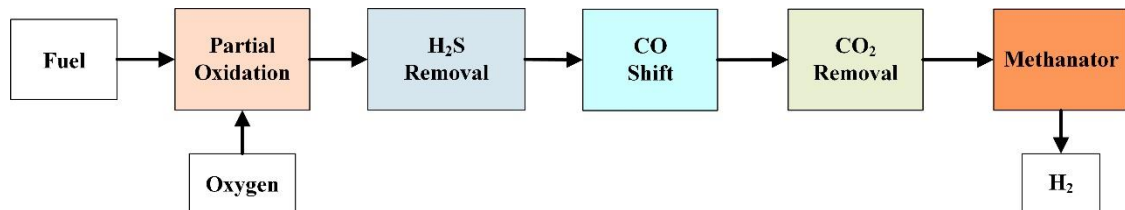


Fig. 4 Partial oxidation process scheme

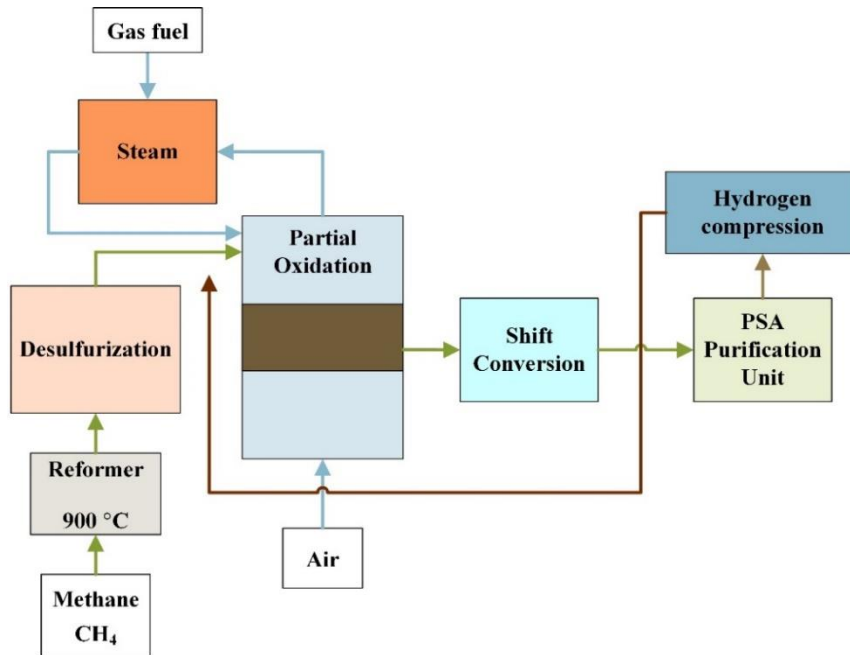
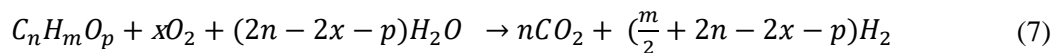


Fig. 5 Autothermal reforming process (adapted from (Kayfeci *et al.* 2019))

meanwhile oxidation and reforming reactions occur at the same time (Chen *et al.* 2008). The output temperature of the reactor is in the range of 800°C to 1200°C, and the pressure of the mixture for the ATR reactor is up to 100 bar (Abdalla *et al.* 2018).

The general reaction of the ATR is given in Eq. (7) and the enthalpy change is calculated with Eq. (8) (Dufour *et al.* 2011).



$$\Delta H_{298}^\circ = 60472 - 136634x \left(\frac{cal}{mol}\right) \quad (8)$$

where x depends on the numbers of hydrocarbon.

2.4 Coal gasification

The gasification process utilizes oxygen and heat in a controlled manner to convert a carbonaceous structure into hydrogen. Indeed, as a result of this process, a mixture of hydrogen and carbon monoxide called syngas (synthesis gas) emerges (Dufour *et al.* 2011). Gasification is a very effective method of obtaining energy from hydrocarbons. Coal gasification is the oldest hydrogen production method (Sharma and Ghoshal 2015). In this method, the coal is converted into the gas state, then the coal gas is mixed with oxygen and steam in the presence of a catalyst. Fig. 6 illustrates the coal gasification process.

Reactions of the gasification process are given in Eqs. (9)-(14) (Dufour *et al.* 2011),



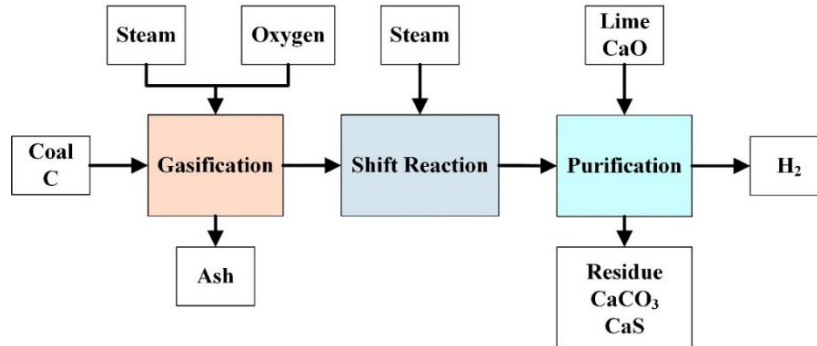


Fig. 6 The coal gasification process (adapted from (Kayfeci *et al.* 2019))



In this production method, there are still some issues that need to be resolved. In coal gasification, hydrogen is produced with several contaminants, thus hydrogen must also be refined before it can be used, in particular in some applications that require high purity (Yamaguchi *et al.* 2012). Although natural gas has generally higher costs compared to coal raw materials, the unit cost of produced hydrogen from coal gasification is 70% and 50% higher than SMR and POX, respectively. The main reason for the higher cost of coal gasification is caused by the initial investment cost.

To sum up fossil fuel reforming technologies, steam reforming is the most developed industrial process and it offers the best H₂/CO ratio among the fossil fuel-based methods. The advantage of the POX method is that the catalysts are not required but the operation can occur at high operating temperatures which decreases the overall efficiency. ATR has limited applications in the industry due to the cost of installation. Furthermore, SMR produces approximately 10 kg of CO₂ per kg of hydrogen, while coal gasification produces 16.5 kg of CO₂ per kg of hydrogen (Voldsund *et al.* 2016). A comparison of various hydrogen production methods is given in Table 2.

3. Chemical looping technology for hydrogen production

3.1 Overview of chemical looping technology

A large number of researchers are conducting studies on the use of hydrogen as a green energy

Table 2 The properties of various hydrogen production methods (Unit cost of gasoline is referred to as 0.93 U.S. dollar) (Hosseini and Wahid 2016)

Method	Chemical reaction	Efficiency (%)	Cost (\$/kg)
SMR/SMR with carbon-capturing	$CH_4 + H_2O \rightarrow CO_2 + 4H_2$	70-85	1.03/1.22
Partial oxidation	$CH_4 + 1/2O_2 + H_2O \rightarrow CO_2 + 3H_2$	60-75	1.48
Autothermal reforming	$CH_n + 1/2O_2 + xH_2O \rightarrow CO_2 + yH_2$	60-75	1.48
Coal gasification/ Coal gasification with carbon-capturing	$CH_n + 1/2O_2 + xH_2O \rightarrow CO_2 + yH_2$	45-65	0.93/1.03
Wind Electrolysis	$H_2O + Electricity \rightarrow H_2 + 1/2O_2$	51-75	6.64

Table 3 CO₂ emissions from various hydrogen production processes (adapted from (Muradov 2017))

Process	Feedstock	CO ₂ /H ₂ ratio (per 1 kg hydrogen)	
		Stoichiometric	Process (in real)
Steam reforming	CH ₄	5.5	8.7-10.4
Steam reforming	Naphtha	6.9	10.5
Partial oxidation	CH ₄	7.3	9-10
ATR	Coal	15.7	19-24
Water Electrolysis	H ₂ O	0	24-28

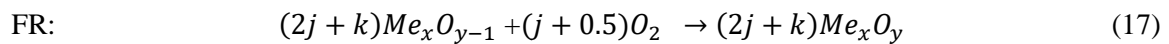
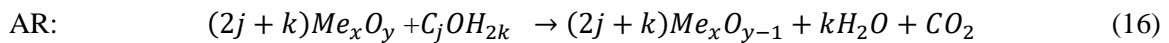
source and to reduce harmful carbonaceous gases emitted during the hydrogen production process. Because, hydrogen is considered an important energy carrier in reducing harmful gas emissions if it does not contribute to CO₂ emissions during the production, compression, or liquefaction, and transport stages. But since this desired situation has not yet been realized, 0.5 Gtons/year CO₂ from the usage of hydrogen is released into nature per year. Although there are efforts to increase hydrogen production from renewable and clean energy sources in the future, it is assumed that fossil resources, the main raw material of hydrogen production in the short and medium-term, will maintain their dominance in this area for economic reasons. Therefore, further efforts are critical to decrease carbon emissions for current hydrogen production technology.

Currently, initiatives and efforts to develop and reduce all systems that lead to CO₂ emissions have become increasingly important due to greenhouse gas concerns. Some transformations and innovations that will be carried out in hydrogen production plants, which significantly lead to CO₂ emissions, are one of these efforts. In these plants, fossil fuel sources are first converted to syngas by existing commercial hydrogen production technologies. CO₂ and hydrogen products are obtained from carbon monoxide and water reactions by the water gas shifting phase. It is important to decompose carbon dioxide both from an environmental point of view and because of the very high purity hydrogen requirements of hydrogen systems (such as fuel cells).

The hydrogen production industry causes 830 million tonnes of CO₂ emissions per year which is equal to the total CO₂ emissions of the United Kingdom and Indonesia. This is a significant indicator of the necessity of cleaner hydrogen production (IEA 2019). The values of CO₂ emissions from various hydrogen production processes are given in Table 3. Although SMR is the least carbon-emitting technology among the hydrogen production from fossil sources, 10 kg of CO₂ is emitted to nature for 1 kg hydrogen production with that method (Muradov 2017). Those emissions are emitted due not only to the SMR reaction itself but also to the energy consumption by facilities during the operation (Borschette 2019).

Carbon capture with chemical looping technology offers great potential with a low energy penalty to decrease toxic emissions during hydrogen production from both renewable and fossil fuel feedstocks (Boot-Handford *et al.* 2014). The main view of chemical looping technology was presented in 1983 by Richter and Knoche (Rydén 2006). This technology is different from the conventional combustion process in which an oxygen carrier referred to as MeO (metal oxide) in Eq. (15) is used to transfer oxygen from the combustion air to the fuel.

Chemical looping technology used in the production of hydrogen is divided into two categories: chemical looping reforming (CLR) and chemical looping hydrogen production (CLH). The basic elements of the chemical looping system are a fluidized bed and a solid oxygen carrier with two reactor vessels (Stenberg *et al.* 2018). In this process, fuel is supplied together with steam or air in the fuel reactor (FR), which leads to steam methane reforming via the oxygen transporter, or catalytic PO_x , air or steam is adopted in the air reactor (AR) for oxygen transporter regeneration or H_2 production (Bayham *et al.* 2016). The chemical reactions of AR and FR are given in Eqs. (15)-(16) (Rydén and Lyngfelt 2006). The schematic of the chemical looping process and hydrogen production from fossil feedstocks with carbon capture technology are shown in Figs 7 and 8, respectively.



In gasification and ATR systems, it is oxidized in an internally heated reactor, while in SMR it occurs thanks to an externally heated furnace. For this reason, a separate fuel flow is required for heating in the SMR, and therefore CO_2 emission occurs in the first stage. A separate unit in the flue gas is required for this stage in SMR systems, otherwise only the CO_2 retention rate after synthesis gas will remain at a maximum of 60 % (Voldsund *et al.* 2016).

3.2 Recent progress on hydrogen production with chemical looping process

Two of the methods used to produce hydrogen that emits minimal CO_2 emissions from fossil fuels are: the first one is to use carbon capture technologies in production facilities, and the second one is to integrate carbon-free energy sources into production stages. In the case of applying these methods, up to 90% of the emissions from fossil fuel-derived hydrogen production can be decomposed and stored. In order to achieve this, it is necessary to place CO_2 -absorbing material at the flue gas outlet of hydrogen production plants or an extra pressure oscillating absorption system. The CO_2 produced in these processes is compressed or liquefied in cleaning units and then injected into a suitable pool where it can be stored permanently for use. As carbon-capturing technology needs additional energy to clean, compress, and transport CO_2 , this will reduce operating efficiency by at least 5% (Velazquez Abad and Dodds 2017). Since CO_2 separation in chemical looping technology is involved in the fuel conversion step, it is considered more efficient and more useful than other solutions.

Chemical looping technology is receiving great attention all over the world and continues to be developed by many different research groups for hydrogen to be a near-zero emission energy carrier. There are various studies in the literature regarding the establishment of chemical looping technology and steam reform in the same system (Pans *et al.* 2013, Rydén and Lyngfelt 2006). These studies have demonstrated the potential of obtaining hydrogen and pure carbon dioxide using chemical cycle technology (Adanez *et al.* 2012, Rydén 2006, 2008).

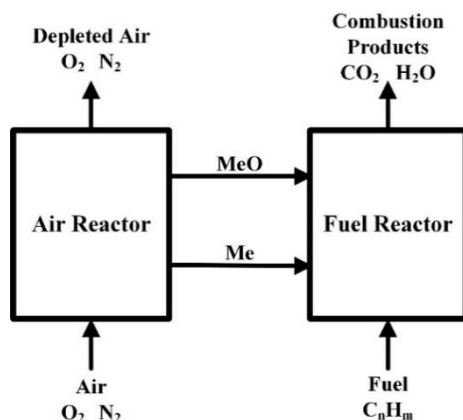


Fig. 7 Chemical looping process scheme (Rydén and Lyngfelt 2006) (Me: Metal, MeO: Metal oxide)

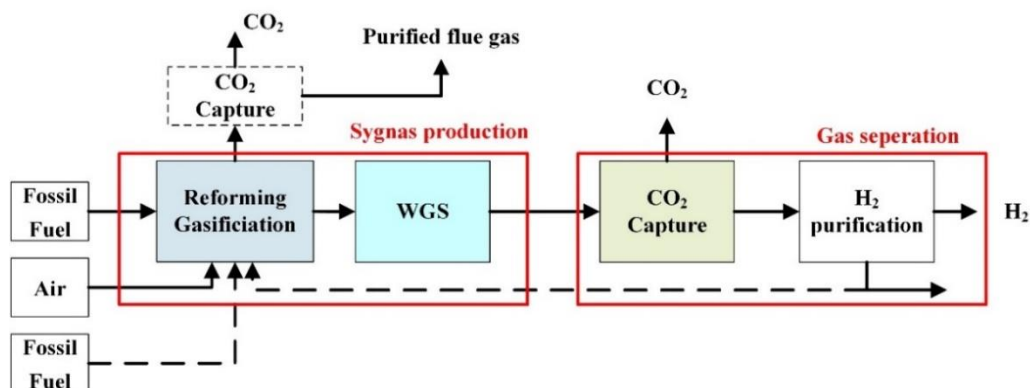


Fig. 8 Hydrogen production from fossil feedstocks with carbon capture technology (Voldsund *et al.* 2016) (Solid black lines show Gasification and ATR, dashed black lines in addition to others show the units required for the SMR system)

As studies on hydrogen production with chemical looping are examined in detail, the common result shows that the choice of oxygen carrier materials is a vital issue for optimal efficiency. To select this oxygen carrier material, it is noted that the following properties: chemical stability, simple preparation methods, sufficient reactivity throughout cycles to reduce solid inventory, high resistance, low environmental impact, good properties for fluidization, and complete fuel conversion to H_2 (Khan and Shamim 2014). The fact that the oxygen carrier tends to combine strongly with the fuel and in high reactivity with steam provides significant advantages at the point of hydrogen production (Gu *et al.* 2019). Transition metals can be said to be widely studied oxygen carriers in the chemical cycle process (Adanez *et al.* 2012). In terms of chemical and cost aspects, Fe-based oxygen carriers can be considered the most appropriate for hydrogen production with chemical looping technology compared to Ni, Cu, Co, and Mn-based. Furthermore, inert materials (as support) used in combination with metal oxides can be involved in the process by increasing the surface of oxygen carriers, increasing their resistance to wear, the stability of reactivity, and supporting ionic conductivity. In Table 4, the advantages and the disadvantages of various oxygen carriers used in chemical looping technology are given.

Table 4 States of various oxygen carriers that have been applied in chemical looping technology

Type of oxygen carriers	Advantages	Disadvantages	The commonly used supporting material	References
Ni-based	<ul style="list-style-type: none"> • high stability at high temperatures • more reactive • almost complete transformation 	<ul style="list-style-type: none"> • rapid catalyst deactivation • high cost • more toxic 	MgAl ₂ O ₄ , Al ₂ O ₃ , MgO, La ₂ O ₃ , CeO ₂ , TiO ₂ , SiO ₂ , ZrO ₂	(Adanez <i>et al.</i> 2012, de Diego <i>et al.</i> 2008, Nandy <i>et al.</i> 2016, Rydén and Ramos, 2012, Tang <i>et al.</i> 2015)
Fe-based	<ul style="list-style-type: none"> • the low tendency to carbon formation • strong resistance to agglomeration • no risk of sulfide or sulfate formation • low cost 	<ul style="list-style-type: none"> • the low reactivity for gaseous fuels • the weak redox characteristics 	Al ₂ O ₃ , MgAl ₂ O ₄ , TiO ₂ , SiO ₂ , ZrO ₂ , CeO ₂	(Kang <i>et al.</i> 2010, Nasr and Plucknett, 2014, De Vos <i>et al.</i> 2019)
Cu-based	<ul style="list-style-type: none"> • high oxygen storage capacity • good reactivity at low temperatures • cheaper than Ni and Co • environmentally friendly 	<ul style="list-style-type: none"> • low melting point • agglomeration formation above 750°C • the poor mechanical strength 	TiO ₂ , SiO ₂ , ZrO ₂ , MgAl ₂ O ₄ , Al ₂ O ₃	(Alirezai <i>et al.</i> 2016, De Diego <i>et al.</i> 2004, Rydén <i>et al.</i> 2014)
Co-based	<ul style="list-style-type: none"> • high oxygen storage capacity 	<ul style="list-style-type: none"> • expensive 	Al ₂ O ₃ , TiO ₂ , MgO, YSZ	(Svoboda <i>et al.</i> 2008, Xiong <i>et al.</i> 2020)
Mn-based	<ul style="list-style-type: none"> • toxic-free • low cost • high oxygen storage capacity 	<ul style="list-style-type: none"> • highly irreversible • have some unreactive phases • low mechanical strength 	TiO ₂ , SiO ₂ , ZrO ₂ , MgAl ₂ O ₄ , Al ₂ O ₃	(Adánez <i>et al.</i> 2004, Johansson <i>et al.</i> 2006)

Ma *et al.* (2017) analyzed various working behaviors of Fe-based oxygen carriers to examine the effects of support materials on carbon capture and performance. Al₂O₃, SiO₂, MgAl₂O₄, ZrO₂, and YSZ (yttrium-stabilized zirconia) were used as inert materials in the experiments that were conducted at 900°C in a laboratory-scale batch fluidized bed reactor. The results showed that the properties of oxygen carriers depend mainly on the supporting material and its interaction with iron oxides, from the point of view of hydrogen efficiency, MgAl₂O₄ inert material together with Fe₂O₃ produced the best results, while SiO₂ was the lowest-yielding material. They found that in terms of carbon accumulation, Al₂O₃, MgAl₂O₄, ZrO₂ materials are higher than SiO₂ and YSZ. Hafizi *et al.* (2015) investigated the effect of calcium loading in Fe/Al₂O₃ (10-40 percent by weight) oxygen carrier during the hydrogen production process with chemical looping - steam reforming. That study also aimed to compare the results with different reaction temperatures between 550 and 800°C. The results showed that the 15% Fe/g-Al₂O₃ oxygen carrier synthesized by impregnation had the best activity in this process. The results showed the best catalytic activity of 15% Fe by weight loaded onto alumina by both co-precipitation and impregnation synthesis methods, and also temperature is also a factor that increases the efficiency of oxygen carriers.

Chen *et al.* (2011) investigated the effect of the oxygen carrier obtained by mixing two inert materials (Al_2O_3 and TiO_2) with Fe_2O_3 in different proportions on hydrogen production and carbon output using chemical looping technology. They prepared mechanical mixtures with the mass ratio of Fe_2O_3 and Al_2O_3 being 9/1,3/2 and the mass ratio of Fe_2O_3 and TiO_2 being 3/2 and experimentally tested three different carriers in total. It was concluded that its reactivity was better than TiO_2 because Al_2O_3 was more porous and provided a higher surface area at the same mixture ratios. Similar results were observed as carbon output, while it was found that there was a decrease in carbon products with a decrease in temperature. Siriwardane *et al.* (2015) prepared new oxygen carriers by mixing Fe_2O_3 and CuO in different proportions with Al_2O_3 inert material to increase reactivity and create a stable reaction. Copper and iron oxides have been shown to yield higher reaction rates, higher combustion efficiency, and higher oxygen utilization.

In addition to experimental studies on hydrogen production with chemical cycle technology, simulation studies with the help of commercial software have also contributed to the literature. In one of these studies, a thermodynamic model using Fe_2O_3 as oxygen carriers were produced using Aspen Plus software, and the effect of reactor temperatures in H_2 and CO_2 production was investigated by Khan *et al.* (2014). In the scope of the study, the thermodynamic model was used to evaluate the effect of various parameters such as the mass flow rate of the oxygen carrier, air, fuel, and vapor on H_2 production. In the basic case, Fe_2O_3 was considered to be full oxidation, assuming that the fuel is completely converted to CO_2 , the production of CO_2 and H_2O , H_2 fuel was monitored by changing the specified parameters. In an experiment based on this basic situation, they concluded that H_2 and CO_2 production increased with air, fuel, and steam flow rates rising to a certain limit and remained constant for higher flow rates. Stenberg *et al.* (2018) suggested two novel process configurations including fluidized bed heat exchanger and biomass utilizing chemical-looping with SMR and simulated their processes in Aspen Plus. They concluded that using fluidized bed burning as a heat source can achieve a significant reduction in fuel consumption and CO_2 emissions compared to traditional methods. Spallina *et al.* (2017) presented technical and financial assessments of two chemical cycle technology methods that produce hydrogen from natural gas containing carbon separation that they proposed. In the first configuration, the chemical looping combustion was used as an oven for reforming reaction with a double circulation fluidized bed system. In the other, a chemical cycle reforming system was used in pressurized conditions for the production of converted syngas. Wang *et al.* (2016) proposed a new hydrogen production system by integrating chemical cycle combustion technology into the methane cracking method. Their simulation studies with Aspen Plus software showed that the proposed model achieved high exergy efficiency due to the lack of extra weight for chemical cycle combustion in the model. Zhang *et al.* (2013) present a thermodynamic analysis of chemical cycle technology in hydrogen production to demonstrate the efficiency potential in their work. Basically, they have come up with a method to determine the oxygen carriers used in chemical looping combustion. In this method, they used the thermodynamic properties of several previously selected candidate materials for selection. At the end of the study, they found that nickel and iron are more suitable thermodynamically as oxygen carriers, but metals such as calcium and cadmium also increase efficiency. Diglio *et al.* (2016) performed numerical analysis for filled bed reactor inspection in a hydrogen production method that used an autothermal chemical cycle reform. Oxidation, seen theoretically in the study, quantifies challenges such as initial temperature through numerical simulations. It has been found that the correct selection of both the initial temperature and the length of the oxidation and reduction stages are important parameters that affect the performance of hydrogen production processes.

4. Conclusions

In this paper, an overview of hydrogen production technologies with chemical looping technology is presented. Today, it has become increasingly important to improve fuels with low CO₂ emissions due to greenhouse gas concerns that cause global warming. Hydrogen is the most promising energy source and carrier for a sustainable and clean environment. Besides, higher heating values of hydrogen are calculated to be three times more than other petroleum products. Nevertheless, even combustion or usage of hydrogen does not emit any toxic emission, production of hydrogen that is not found free causes a significant amount of carbon emission as most of the hydrogen is produced by fossil fuel feedstock. In order to reduce the harmful effect of hydrogen production on the atmosphere, studies on the use of carbon capture technologies in factories have gained importance. Chemical looping technologies offer the potential to control carbon emissions during hydrogen production. If the carbon capture system is added to an SMR hydrogen production plant, the cost of hydrogen production is estimated to increase by more than 22%. The unit cost per kg of hydrogen produced in a non-CO₂ capture facility is \$ 1.03, while the unit price of hydrogen in a facility using CO₂ capture technology of the same size increases by 18.5% to \$ 1.22, but it prevents the carbon dioxide emitted by the facility from damaging the atmosphere by storing. Taking into account CO₂ emissions from the plant's electricity consumption, there is an emission reduction of up to 83%. Despite recent significant advances in carbon capture and storage technology in hydrogen production, it is currently not implemented on a large commercial scale due to energy losses and an increase in operating costs. Hydrogen production with CO₂ capture can be a key technology to initiate the sustainable hydrogen era by reducing massive emissions from the hydrogen production industry. However, a few efforts have been practiced in the literature for hydrogen production with chemical looping technology. Much more studies are needed to develop and provide to establish carbon-capturing in the plants.

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