

*Original Research Article*

## Investigation of Standard Natural Gas Synthesis Technologies

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### ABSTRACT

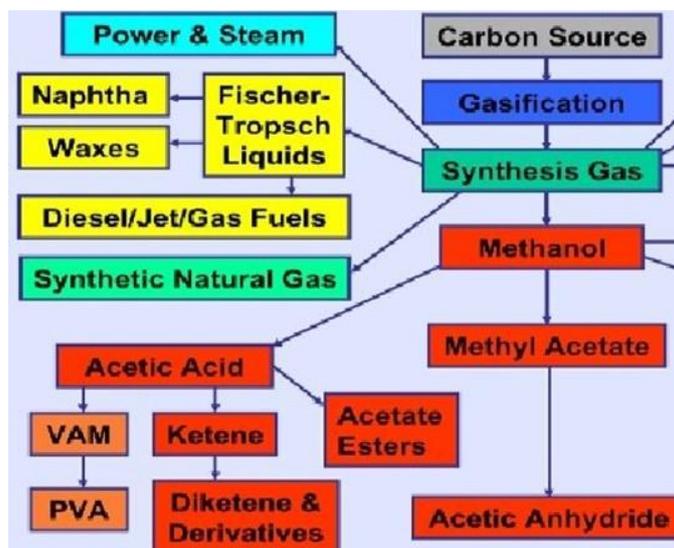
Natural gas is often associated with oil and pushes oil out of the soil into extraction wells. As the oil-gas mixture rises in the wells, gas is released and carries the mixture to the top of the well. In oils which are not saturated with gas and are only under water pressure, the amount of dissolved gas is less and in supersaturated oils, the amount of gas is higher. There are also streams which contain only natural gas and no oil. The term synthetic gas refers to the gas mixtures which contain Co and H<sub>2</sub> in different proportions. H<sub>2</sub> and Co are two important substances in the chemical industry and have many uses. Co is used in the production of paints, plastics, foams, insecticides, herbicides, acids, etc., including the use of hydrogen, ammonia production, hydrogenation, and hydrocracking. Synthesis gas is a very valuable raw material for the production of various chemical substances. Using this gas and various processes, it is possible to produce many different chemical substances which depending on the production method, different ratios of hydrogen to Co are obtained. Likewise, in industrial use, different ratios are required depending on the process in which the gas is used. Synthetic gas feeds can be hydrocarbons, coal, oil, natural gas, and then vegetable and animal products.

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GRAPHICAL ABSTRACT



INTRODUCTION

Most natural gas is composed of methane, and in addition to methane, it contains other gaseous hydrocarbons from C<sub>2</sub> to C<sub>4</sub> in varying amounts, as well as higher hydrocarbons [1]. Natural gas may be dry or wet. The highest growth in the natural gas (Figure 1) demand is expected in the developed countries of the world, where in these areas, the total demand will increase by 3.9% between 2001 and 2025 [2-4].

The level of gas used in developed countries is projected to reach 1.5 or 2 times its level in

2001. Industrialized countries, where the natural gas market is more entrenched, are planning to increase their confidence in natural gas. Over the next 24 years, demand for natural gas in industrial areas is expected to increase by 2.2% per year, which is almost double the rate of increase for crude oil.

Among industrial areas, North America had the largest increase in natural gas use at 19 trillion cubic feet between 2001 and 2005. The United States alone accounts for 66% of the total increase in North American gas consumption [5].

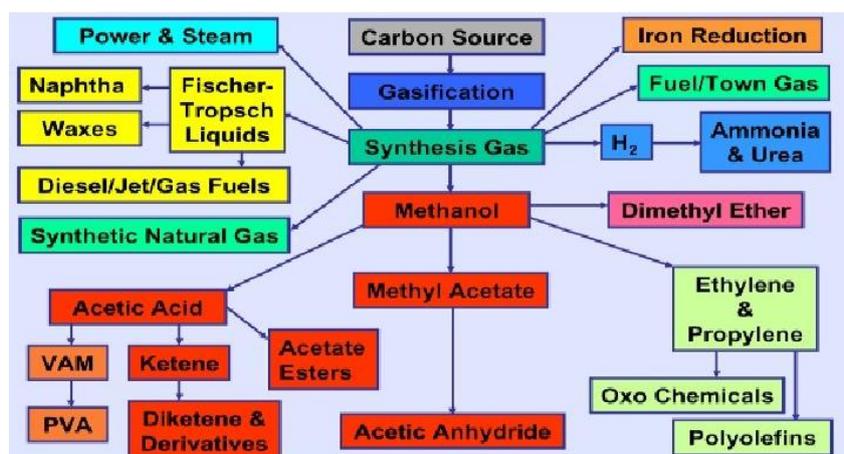


Figure 1. Utilization of Fossil Plant CO<sub>2</sub> [6]

In the United States, the demand for natural gas increases to 1.8% per year, which is mainly for the production of electricity. The new generation capacity expected in the United States is 80% of the total amount of natural gas for the combined cycle or combustion turbine technology [6-8].

It seems that in Western Europe, the use of natural gas will be expanded with more than 2.4% of the average annual growth rate in the forecast period. Free natural gas markets in the European Union have flourished since natural gas laws were adopted in 1998 with the consent of a majority of member states to allow free access to natural gas until 2008. Total natural gas consumption in Western Europe is expected to increase from 14.8 trillion cubic feet in 2001 to 25.9 trillion cubic feet in 2025. In Eastern Europe and the former Soviet Union (EE/FSU), consumption of natural gas is expected to increase by 2.9% annually between 2001 and 2025. The fastest growth is thought to be in Eastern European countries whose economies have been recovering since the collapse of the Soviet Union. The demand for natural gas in Eastern Europe is likely to grow by 4.6% [9].

At FSU, natural gas demand appears to be growing relatively slowly at 2.6 percent per year. The rate of increase in natural gas exchanged across the international borders has increased from 19% of world consumption in 1995 to 25% in 2003. Gas pipelines grew by 39% and liquefied natural gas (LNG) exchanges grew by 55% between 1995 and 2001. Many international pipelines have been designed or are to be built. In fact, large numbers of natural gas reserves are located in areas far from the center of demand, which increases the LNG competitiveness. This has created high expectations for LNG worldwide. In Asia and Europe, as LNG markets are heavily influenced by the oil market so that natural gas prices, the natural gas trade will grow as pricing mechanisms continue to evolve and international trade will be facilitated and pave

the way for creating a global natural gas market [10].

### **Reserves and resources**

Since the mid-1970s, the world's natural gas reserves in general have been on the rise every year. For instance, in January 2003, proven natural gas reserves were estimated at 5,501 trillion cubic feet, according to the Journal of Oil and Gas, which is 50 trillion cubic feet more than in 2002.

Natural gas reserves in industrialized countries increased by 18 trillion cubic feet between 2002 and 2003. EE/FSU reserves decreased by 4 million cubic feet due to an incorrect estimate of Turkmenistan's gas reserves, where gas reserves decreased by 30 trillion cubic feet. Most of the world's natural gas is concentrated in the Middle East and EE/FSU [11].

Natural gas consumption, especially in the last decade, the ratio of reserves to consumption has remained high. Globally, the ratio of reserves to consumption is estimated at 61.9 years. In developing Asian countries, an increase of 11 trillion cubic feet in reserves was seen in 2003. The US Geological Survey (USGS) evaluates the potential for global oil production at regular intervals. (Oil, natural gas, and natural gas condensate) according to the latest estimates of this organization, a huge volume of undiscovered gas reserves still remains, which is approximately 4839 trillion cubic feet worldwide [12].

### **Why do we use natural gas?**

We prefer to use natural gas for four reasons, which are:

- 1- Natural gas is clean gas because it has the least amount of carbon.
- 2- Natural gas is an effective gas, because it does not need to be strengthened and helped by new technologies to build, and it is environmentally acceptable in the power sector.
- 3- It is affordable.

4- It is abundant.

### Synthesis gas production methods

#### Importance of synthesized gas

Most methods of using natural gas to produce valuable products require the production of synthetic gas as an intermediary [13]. Unfortunately, despite the fact that a long time has passed since the production of synthetic gas in the world, all synthetic gas production units in the country by companies' external are installed and commissioned. In recent years, the extensive efforts have been made to effectively use natural gas and convert it into valuable and easy-to-transport products such as methanol, liquid hydrocarbon fuels, and petrochemical raw materials such as ethylene and other light olefins. This between the production and use of synthetic gas has a special place in industry.

#### Major uses of synthesized gas

**1- Preparation of methanol:** Since methanol is used in large quantities in the synthesis of acetic acid, it is of great importance in industry. Methanol is prepared from the synthesis gas according to the following reaction:

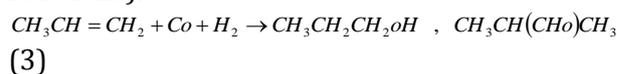


**2- Preparation of ethylene glycol:** In this method, ethylene glycol is obtained due to the

density of synthesized gas at high pressure and in the presence of a catalyst.

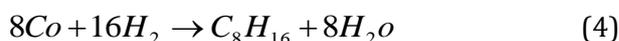


**3- Hydroformylation reactions:** In this type of reactions, aldehydes are produced from olefins using synthetic gas. This reaction is called (oxo reactions).

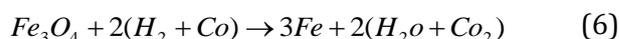
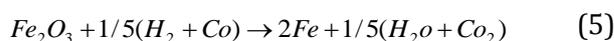


#### 4- Fischer-Tropsch reaction

In this process, the synthesized gas is converted to gasoline molecules in the C<sub>8</sub> range. In essence, this reaction is the first ammonization of carbon monoxide by hydrogen to form organic products.



**5- Reduction of iron ore:** To regenerate iron ore obtained from mines, the synthetic gas is used. In this process, iron or its powder is obtained by direct reduction of iron ores.



**6- Other uses:** Among the uses of synthetic gas, we can mention the preparation of heavy alcohols, dimethyl ether, esters, ketones, hydrocarbons, etc. among which is the preparation of ammonia [14].

**Table 1.** Some important ratios for various applications of synthetic gas [15]

Mixtures	Main uses
H <sub>2</sub>	Hydrocracking and hydro-treating for treatment
3H <sub>2</sub> : 1 N <sub>2</sub>	Ammonia plant feed
2H <sub>2</sub> : 1 Co	Alkanah (Fisher Reaction - Truth)
2H <sub>2</sub> : 1Co	Methanol plant feed
1 H <sub>2</sub> : 1 Co	Alcohols and alcohols (exo reactions)
Co	Acids (formic and acetic)

### Synthesis gas production methods

#### Coal Gasification

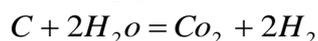
This method is initial in synthesis gas production. It has been one of the oldest methods in producing industrially rich mixtures

of carbon monoxide and hydrogen in which a bed of hot coke was exposed to air and water vapor, respectively [16]. During the blowing period, some coke was heated and burned in the bed until it reached a temperature of

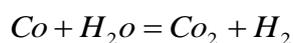
approximately 1000 °C. After blowing air, blowing steam causes the following general reaction:



The use of steam at this stage leads to the re-use of the "water gas" method with reference to the mixture of carbon monoxide and hydrogen formed reaction unlike the highly exothermic reactions of carbon with oxygen which occur during the blowing of air. It is a heat sink ( $\Delta H^0_{298} = +31kcal$  for granite carbon). Increasing the bed temperature during the exothermic combustion phase provides the heat required for the subsequent hot-water reaction of the water vapor with the coke. At 1000° C, carbon dioxide is formed in part according to the following general reaction:

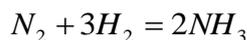


If equilibrium is assumed for reactions, the thermodynamic calculations of the gas mixture composition indicate that carbon dioxide and water are only in small amounts at 1000° C according to the observations made in water gas generators, as illustrated in reactions 2-8, it is endothermic, but the heat of reaction is lower ( $\Delta H^0_{298} = 21kcal$  for granite carbon). We note that the reaction equation is a simple combination of the reaction equation which is called the "water-gas displacement" and results from the combination of Co with H<sub>2</sub>O.

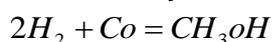


This reaction is exothermic ( $\Delta H^0_{298} = -10kcal$ )

Due to the high calorific values of these reactions, mixtures of carbon monoxide-rich water gas and hydrogen were initially considered as fuels. Then, instead of sources of H<sub>2</sub> or Co or mixtures of the two, they became important for the synthesis of various chemicals. An important initial instance was the recycling of hydrogen by water gas for usage in the catalytic synthesis of ammonia from the elements nitrogen and hydrogen.

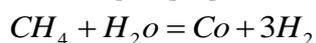


In this particular case, the reaction nitrogen can be obtained from the same operation used for the water-gas mixture, because the gas obtained from the reactor during the blowing of the air contained the highest amount of carbon and carbon monoxide, in which oxygen was completely consumed. The products combine with the coke during intermittent blowing of air and water vapor, although they contain a large amount of a mixture of N<sub>2</sub>, Co, and H<sub>2</sub> with an excess of CO<sub>2</sub>. When the Co concentration in the mixture decreases to a very low level, through the water-gas displacement reaction can be exploited. The reaction equilibrium shifts to higher concentrations of Co and H<sub>2</sub> by adding excess water vapor to the mixture. Simultaneously, the mixture is cooled to a temperature of about 500 °C. Satisfactory reaction rate is obtained by the mixture contact with the iron oxide catalyst. The Co concentration decreases to about 1% and Co as a strong toxin for the iron metal catalyst used in ammonia synthesis by contact of the gas mixture with the ammonia solution of a copper salt is separated. Before Co is separated in this method, CO<sub>2</sub> is separated by contact with high pressure water in an adsorption tower. One of the further advances in the separation of small amounts of Co is the reaction of hydrogen with methane on a nickel catalyst.



### Steam reforming

In the 1920 s and 1930 s, the use of natural gas (methane) in the production of synthesized gas received more attention than the molten coke. An overall reaction occurs between methane and water vapor [17].



That is extremely hot ( $\Delta H^0_{298} = +49kcal$ ). A conventional included catalyst is used as the active component. The process usually operates

with additional steam at temperatures above 800 °C. (Most H<sub>2</sub>O to CH<sub>4</sub> ratios are from 2:1 to 4:1).

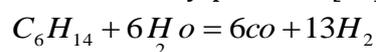
The produced gas composition is generally close to the required equilibrium. At a reactor pressure and the molar ratio of water to methane, at the gas inlet flow, the Co amount increases with increasing temperature, to the extent that the amounts of carbon dioxide and methane decrease. If the reactor pressure increases, the amounts of methane and water in general increase.

In the 1950s and 1960s, there was an interesting development in the use of naphtha as a light feed, especially the highly paraffinic naphtha of the Middle East, which is an undesirable feed for the production of aromatic hydrocarbons by catalytic reforming.

The catalytic reforming process does not use steam as a reagent or diluent. It is used to produce high-octane aromatic hydrocarbons for gasoline with the help of so-called "two-factor" metal catalysts. Light crude naphtha vapor reforming is primarily specific to areas of the world where, although vapor reforming is primarily used to produce mixtures of hydrogen and carbon monoxide, a process at a lower temperature than naphtha crude is used to produce methane used in residential areas. The high activity and resistance of nickel catalysts for usage in the city gas production in the laboratories of Exxon Engineering Research Company in the 1960s greatly advanced. The use of these catalysts to produce hydrogen from naphtha was considered [18].

In catalytic steam reforming, high pressures (30 to 40 atm) are used to produce city gas, and operating temperatures (300 to 400) are much lower than those used to produce synthetic gas. The molar ratios of vapor to hydrocarbons are typically in the range of 8:1 to 12:1. High ratios are important to limit the formation of inactive carbon residues on the catalyst. If we consider the normal hexane from the hydrocarbons

contained in light crude naphtha, we can consider the overall vapor reforming process for methane production consisting of two major components. The first part is the decomposition of reaction hydrocarbons and the formation of Co and H<sub>2</sub> as by-products [19].

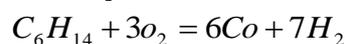


This reaction is very hot.

### Spontaneous thermal reforming (ATR)

Steam reforming was an important process at first, but progress has been made in catalysts and in engineering forms of reformers. One of these advances was the spontaneous heat reformer introduced by Haldor Topsøe after the 1950s. This method is a combination of partial oxidation and steam reforming in which oxygen and water vapor enter a reforming reactor. Partial oxidation is performed at the inlet of the reactor and heat is prepared for the steam reforming reaction in the second part of the tank, which is filled with catalyst. As a result, there is no need to heat the reactor less or more than the amount provided by preheating the reactions. This advancement in steam reforming technology improves the overall efficiency of the reactor and increases process flexibility. The ratio of O<sub>2</sub> gas molecules to carbon atoms in hydrocarbon reactions ( $\frac{O_2}{C}$ ) is usually about

0.5 to 0.6, which is much less than what is needed for complete combustion. The partial oxidation part of the process uses the normal hexane present in the feed according to reaction.

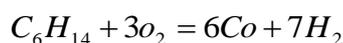


It is assumed that the conversion factor of Co and H<sub>2</sub> is 100%. The reaction (2-14) is highly exothermic ( $\Delta H^0_{298} = -118kcal$ ). Interestingly, the main idea in using an exothermic reaction is to provide heat for an endothermic reaction. The ATR provides a completely independent one-step process for converting raw feed into synthetic gas [20].

### Minor Oxidation (POX)

Another type of synthetic gas production process was developed in 1940 by the Texas and Shell oil companies. This method is especially significant for carbonating low-quality feed, such as oil residues and even coal or coke. The reaction takes place in the presence of oxygen in a refractory tank. There is usually no excess steam and the reactor is not filled with catalyst. Oxygen is used more than the air, because it can bring reactors to the required high temperatures (around 1400 °C) by avoiding the problems which occur with carbon formation. The ratio of O<sub>2</sub> gas molecules to carbon atoms in hydrocarbon reactions ( $\frac{O_2}{Co}$ ) is usually about

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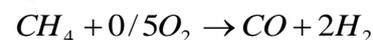
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which occur with carbon formation. The  $\frac{O_2}{c}$  ratio of about 0.7 is required for this purpose. In other words, in spontaneous heat reformatting, the design of process burners is a sensitive and important category. Reactors, commonly called gasifiers or Pox heating units, operate at pressures above 70 atmospheres in the total temperature range of 1200 to 1600 °C are located. The ratios  $\frac{H_2}{Co}$  in the products are generally less than about 2 and this ratio may be adjusted through an additional process depending on the final application of the synthesis gas. The technology of this work is highly advanced and widely used.

### Catalytic partial oxidation (cpo)

In recent years, the partial catalytic oxidation of hydrocarbons has benefited from alloy metals coated with porous ceramic monoliths, which has been the subject of much research. Professor Lenny Schmidt of the University of Minnesota has done this research. Other researchers are currently working on this. In this method, reaction times are very short (milliseconds) and widely used in the Ostwald process for the oxidation of ammonia or platinum or gases (gauzes) with platinum alloys in the commercial production of nitric acid.

The catalytic form of the monolith is similar to the structural coating used in catalytic converters to control contamination in car exhaust systems. In methane oxidation, the optimum reaction is as follows:



The efficiency of H<sub>2</sub> and Co is in a 2:1 molar ratio. Such a ratio is ideal for the reaction flow in methanol synthesis or Fisher-Trich synthesis. The reactions were subsequently preheated to the desired inlet temperature

( $\Delta H^0_{298} = -8/5kcal$ ). There is no longer a need for additional heat to enter through the

reactor walls or through a provision for a separate inlet section of the reactor where the exothermic combustion reactions provide heat for the downstream heat capture process.

The direct cpo reaction is approximately twice as fast as the similar catalytic vapor reforming reaction. Complete conversion of methane (more than 99%) to H<sub>2</sub> and co is performed in molar ratios of 2 to 1, at which temperatures reach above 850 °C [21].

### **Two-stage reforming**

The new reformer reactor method, specifically through the Exxons AGC-21 process for a subset of ATR, is commonly referred to as the secondary reforming [22-24]. As a result, variations in feed composition, especially lower concentrations of combustibles in secondary reformer feeds, ATR reactors and secondary reformers, will have different soot and thermal properties which require different reactor and burner designs. However, the difference between ATR and secondary reforming is not of constant concern to consumers and suppliers of this technology, and that is why secondary reformers are often referred to as ATRs [25-27]. Most commercial experiences with spontaneous heat reforming have actually secondary reform problems, the most notable of which are the oxygen blower units for methane production and the air blower units for ammonia production [28-30].

### **Combined reforming methods**

Combining methods such as steam reforming, carbon dioxide reforming, partial oxidation and ATR, various processes for the production of synthetic gas have been proposed so far. The purpose of combining these processes is to obtain different ratios for H<sub>2</sub>/CO [31-33]. The combination of different methods can lead to the combination of the desired percentage for the synthesized gas. Also, combined methods lead to the development of reactors and processes which have higher energy efficiency and are

more balanced in terms of dimensions and lower manufacturing costs. It normally acts as an adiabatic [34-36]. The pre-refurbishment of the overall unit user development contributes to a wider range of hydrocarbons. The pre-reformer also conducts some of the primary reformer tasks in the conversion process. In this way, it is possible to use cheaper materials in construction, and also a lower level of heat transfer, which will mean lower costs. In this process, reaching the outlet synthesis gas temperature up to 1050 °C is common and methane conversion rate of more than 99.6% will be possible. Reduction of energy consumption in this process up to 3% and reduction of operating costs up to 6% are the advantages of this method. The initial investment is between 72 and 76% of a single SMR unit. The main reason for the decrease in initial investment is due to the savings in reformer equipment. Companies such as Lurgi, Haldor-Topsoe and Kellogg are among the main contenders for these combined processes [37].

It is clear that only 50% of the heat generated by the fuel in the SMR unit is used for reforming reactions and the other 50% should be absorbed by a complex steam generating unit using a heat exchanger reformer (heat exchange reformer). It is possible that heat transfer to the catalytic bed reaches 80% [38]. The combined reformers described below (KRES, CHR, and CAR) use this principle to increase efficiency and reduce environmental issues (emissions). Kellogg Reforming Exchanger System (KRES) is the first type of these reformers. This method was registered in 1991 by M.W. Kellogg in the United States. In this technique, the feed flow is divided between the tubes containing SMR reformer catalyst and ATR secondary reformer [39].

The distribution rate is 75% for the secondary ATR reformer and 25% for the SMR primary reformer. The mixture of natural gas and water vapor enters the SMR from above and passes through the catalyst bed and exits from below. In

this possibility, the SMR synthesis gas is mixed with the ATR output and flows upwards as the heating medium of the SMR unit in the shell. The advantages of this method over individual SMR are reduction of initial investment, improvement of return, thermal, reduction of NO<sub>x</sub> and CO<sub>2</sub>, reduction of maintenance costs, and reduction of operating space. It should be noted that this method has not yet been industrialized. Gas heated reformer (GHR) is the process by which the heat required for heat treatment reactions is provided by the cooling of the converted gas leaving the secondary reformer. The technology was introduced by ICI under the brand name Syntex in the 1960s which was used in 1988 in two ammonia units in Severnside, England. Unlike the KRES method, the feed in our GHR technique is not split between the reformers, but is first passed through the SMR reformer and 25% of the reforming is completed. The converted partial gas, then, passes through the secondary ATR reformer. The ATR output is used to heat the primary reformer feed. An auxiliary torch is used in the initial start-up. The volume of a GHR is about 15 times smaller than that of an SMR with the same capacity. The initial investment of a GHR is 40% less than that of a similar SMR, but the operating costs are relatively the same [40]. A GHR unit consumes 33% less oxygen than a single ATR unit. Decreasing the S/C ratio will lead to two problems, including an increase in the possibility of coke formation in the preheating units, a pre-reformer, and an increase in metal dust in the heat recovery section. Coke formation depends on the composition of natural gas, temperature, pressure, and S/C. It can be prevented by selecting suitable operating conditions. In GHR units, nickel alloys with a high percentage of chromium can be used to prevent metal dusting. In general, increasing the S/C ratio reduces the risk of corrosion of metals by metal dusting. Combined auto-thermal

reforming (CAR) system integrates SMR and POX reforming into a pressure reactor. In fact, it's a kind of GHR extension. In 1982, Uhde GmbH introduced this process. In 1987, a leading unit of this type was built in Germany [41].

An industrial prototype unit has been operating at the Strazke refinery in Slovakia since 1991. In this method, a mixture of water vapor and feedstock is first converted into heat generated by the POX process in the SMR reformer. The POX process takes place in the lower part of the reactor and between the uncharged feed and oxygen at a temperature of 1200 to 1300 °C. Similar to GHR, since there is no external flame, no pollutants are generated. The CAR method reduces oxygen consumption by up to 35% and natural gas consumption by up to 15% compared to a unit with similar pox capacity, but operating costs will be 10% higher. Initial investment is also 20% less.

### Conclusion

Since synthetic gas has many uses in industry, so its production requires a process which is firstly able to generate abundantly and secondly is cost-effective. Synthesis gas was initially produced from a mixture of coke, the air, and water vapor, but the most common and cost-effective method used since the 1930s is hydrocarbons' reforming. Among hydrocarbons, only light ones are commonly used, commercially and cost-effectively, among which the natural gas is the most common and propane and butane are next. With the development of special catalysts, the use of naphtha as feed has also been already common. As mentioned above, it is obvious that the process of reforming natural gas to produce synthetic gas is the most cost-effective method. However, some specific reasons such as easy access to raw materials, cheapness of one feed compared to another, the simplicity of technology, etc. may change the choice of process.

**Table 2.** Comparison of synthesis gas production technologies (natural gas feed)

Losses	Profits	Technology
<p>The ratio <math>\frac{H_2}{Co}</math> is often higher than the required amount. When Co is also produced. The highest value of air pressure</p>	<p>Most industrial experience Oxygen is not required Minimum temperature required in the process The best ratio <math>\frac{H_2}{Co}</math> for hydrogen production applications</p>	SMR
<p>Low business experience In some units, it should be used in conjunction with other synthetic gas production technologies</p>	<p>Dense and compact size and capacity Utilizing flexibility which offers additional ways to increase capacity.</p>	Thermal conversion encoding
<p>Increase process complexity Process temperature is higher than SMR. Oxygen is needed at all times.</p>	<p>The size of the SMR unit decreases Less slip and drop of methane, this helps to obtain synthetic gas with higher purity. The volume and capacity of methane synthesis gas should be reached by adjusting the output temperature of the secondary code.</p>	Two-stage encryption
<p>Limited experimental experience Oxygen is always needed.</p>	<p>The natural ratio <math>\frac{H_2}{Co}</math> is often provided. Slip and drop less methane The capacity of methane synthesis gas can be reached to a suitable level by adjusting the outlet temperature of the reformer. The process temperature needs to be lower than Pox.</p>	ATR
<p>Ratios are inherently <math>\frac{H_2}{Co}</math> less harmful where ratios of more than 2 are required. The operating temperature of the process is very high. The heat recovery temperature and soot formation are high, which adds to the complexity of the process. The volume of methane synthesis gas is basically small and does not change easily in the face of downstream process equipment and supplies.</p>	<p>Raw feed desulfurization is not required. The absence of a catalyst allows carbon to form, and therefore, steam-free operation significantly reduces the volume of CO<sub>2</sub> synthesized. Inherently low ratios are useful for applications where ratios of less than 2 are required.</p>	Pox

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