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Dry reforming of waste polymers for production of hydrogen

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بِسْمِ اللَّهِ الرَّحْمَنِ الرَّحِيمِ

((وَقُلْ رَبِّ زِدْنِي عِلْمًا))

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الاهداء

اللهي لا يطيب الليل الى بشرك ولا يطيب النهار الى بطاعتك ..ولا تطيب
اللحظات الى بذكرك ..ولا تطيب الآخرة إلى بعفوك.. ولا تطيب الجنة الى
برؤيتك الله جل جلاله. إلى من بلغ الرسالة وادى الامانة ..ونصح الامة
..الى نبي الرحمة ونور العالمين . نبينا محمد(صلى الله عليه واله وسلم).

إلى من كلفه الله بالهيبة والوقار ..إلى من علمني العطاء بدون انتظار
..إلى من احمل اسمه بكل افتخار .. وستبقى كلماته نجوما اهتدي بها اليوم
وفي الغد والى الابد ...الى من يرتعش قلبي بذكره ...إلى اجمل ذكرى أبي
الغالي حفظة الله. إلى ملاكي في الحياة ..الى معنى الحب والى معنى
الحنان والتفاني ..الى بسمه الحياة وسر الوجود .إلى من كان دعائها سر
نجاحي وحنانها بلسم جراحي إلى أُمي الحبيبة .

الشكر والعرفان

الحمد لله قبل كل أحدوالحمد لله بعد كل أحدوالحمد لله مع كل أحد
....والحمد لله الباقي بعد فناء كل أحد...وبعد ...

وانا اضع اللمسات الاخيرة في بحثي يطيب لي ان اتقدم بالشكر الجزيل
والامتنان العظيم للأستاذ الدكتور محمد الذي تفضل مشكوراً بقبول
الإشراف على هذا البحث فقد كان لسداد رأيه ونفاذ بصيرته ولمتابعته
المستمرة طوال مدة اعداد البحث الاثر الاكبر في تذليل الصعوبات.

ومن الوفاء ان اتوجه بالشكر والامتنان الى جميع الأساتذة في كلية
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وفي الختام أوجه شكري وامتناني الى كل من ساعدني في اتمام هذا
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Abstract

Dry reforming (DR) processes consist of a reaction between an adequate feedstock and carbon dioxide to produce syngas. In the case of a renewable feedstock (biogas, bioalcohols, wood tar,...), the DR processes become very interesting since they consume greenhouse gases (CO_2 , CH_4 ,...) and produce hydrogen and syngas mixtures that can be considered as renewable alternatives to fossil fuels. The DR processes involve endothermic reactions accompanied by side reactions that decrease the overall process efficiency. The use of a catalytic material is expected to reduce the energy required for the process and to favor the selectivity towards syngas production. Thus, in the last decades, many studies considered the synthesis of catalytic materials that are active, selective and stable in DR reactions. The production of gases through pyrolysis process is affected by some factors such as atmosphere, temperature, feedstock and catalyst type. Nowadays, most of researches are focusing on carbon capture processes due to the effects of carbon dioxide on climate change.

Chapter one

Introduction

1. Introduction

Plastic is a crucial material in many sectors, including construction, packaging, transportation, electronics, textiles, and others[1,2]. The last half century has witnessed the rapidly increasing demand and production of plastics[3], resulting in considerable plastic waste due to the low plastic recycling rate. From 1950 to 2015, only 9% of the cumulative plastic waste generation (6300 million metric tons (Mt)) was recycled, compared to over 60% discarded (accumulating in landfills or in the natural environment). The landfilled or disposed plastic wastes and their fragments, i.e., microplastics and nanoplastics, have caused increasing environmental concerns. Increasing plastic recycling is one essential strategy to reduce plastic waste disposal. There are two common types of plastic recycling, mechanical (e.g., magnetic density separation) and chemical recycling (e.g., gasification). Recently, another type of recycling method, solvent-based recycling (or referred as physical recycling), is also attracting attention. The challenges of plastic mechanical recycling include thermal-mechanical degradation (e.g., caused by heating and mechanical shearing of polymer), plastic degradation (e.g., caused by photo-oxidation process during lifetime), incompatibility among different polymers when recycling blended plastics⁹, and contaminations (e.g., coating, ink, additives, metal residues or cross-contaminations among different plastic streams)[4,5].

Some waste plastics are hard to be mechanically recycled due to low bulk density (e.g., films), lightweight (e.g., polystyrene (PS)), low economic value (e.g., PS), and carbon-black pigments that absorb infrared light and confound the sorting machine[6,7]. Hence, relying on the traditional mechanical recycling method alone is insufficient to address the increasing volume and variety of plastic waste. Compared to mechanical

recycling, thermochemical methods, as one type of chemical recycling, have advantages in processing plastic wastes that are difficult to be depolymerized, or mechanically recycled due to economic or technical barriers[8,9]. Thermochemical processes include pyrolysis and gasification, which have potentials to treat waste plastics with high energy, carbon, and hydrogen content, and low moisture content¹³. Thermochemical processes can produce a variety of products, and hydrogen is one product with a mature and growing market. Hydrogen is an important industrial gas widely used in the oil refining and chemical industries, it can also be used as a clean energy source for transportation[10].

Since most plastics are made from fossil fuels, it is necessary to mitigate the fossil-based carbon emissions during the thermochemical conversion of MPW to hydrogen[1,15]. Carbon capture and storage (CCS) is an important technology to mitigate climate change by capturing and geologically storing CO₂. Coupling hydrogen production with CCS offers a means to produce low-carbon hydrogen[11,12]. For the large-scale development and implementation of plastic recycling technologies, it is critical to understand the economic feasibility and environmental performance of plastic waste to hydrogen pathway with/without CCS and policy incentives, as well as to identify the key drivers and future improvement opportunities.

1.1. The impact of plastic waste on the environment:

Plastic production has sharply increased over the last 70 years. In 1950, the world produced just two million tonnes. It now produces over 450 million tonnes. Plastic has added much value to our lives: it's a cheap, versatile, and sterile material used in various applications, including construction, home appliances, medical instruments, and food packaging.

With the development of the world economy, the output of garbage around the world is also increasing rapidly, especially the use of plastic products is becoming more and more widespread, and disposable plastic are most commonly used in people's lives. Disposable plastic products have brought convenience to people's production and life, but due to the difficult degradation of plastic products, "white pollution" has become more and more serious. The large amount of disposable plastic products and the low recycling rate have caused serious pollution to the soil environment and the marine environment. The world is facing an environmental crisis caused by plastic waste. Currently, "Limiting plastic" has become a global consensus, and many countries and regions have launched actions to limit plastic and ban plastic.

The municipal solid waste counted under 10%–12% of the plastic residue is combusted. After the combustion process, the gases are released into the environment, which increases air pollution and causes greenhouse effects. The substance released in the atmosphere are furans, mercury, dioxins, and polychlorinated biphenyls. Immediate measures and implementations are needed to address and manage them properly, which can protect the environment. This is also applied in an aqueous environment for aquatic and aquaculture protection since many of these contaminants are hydrophobic, plastic in the aquatic environment is potentially acting as a sink for contaminants which make them less accessible to wildlife,

particularly if they are buried on the seafloor [13]. The poor disposal and plastic waste mistreatment effects are categorized under three main classes, including the effects of plastic waste on animals, public health, and environmental pollution. Fig. 1 provides an illustration of the fate of plastic materials with harmful impacts on the ecosystem [14].

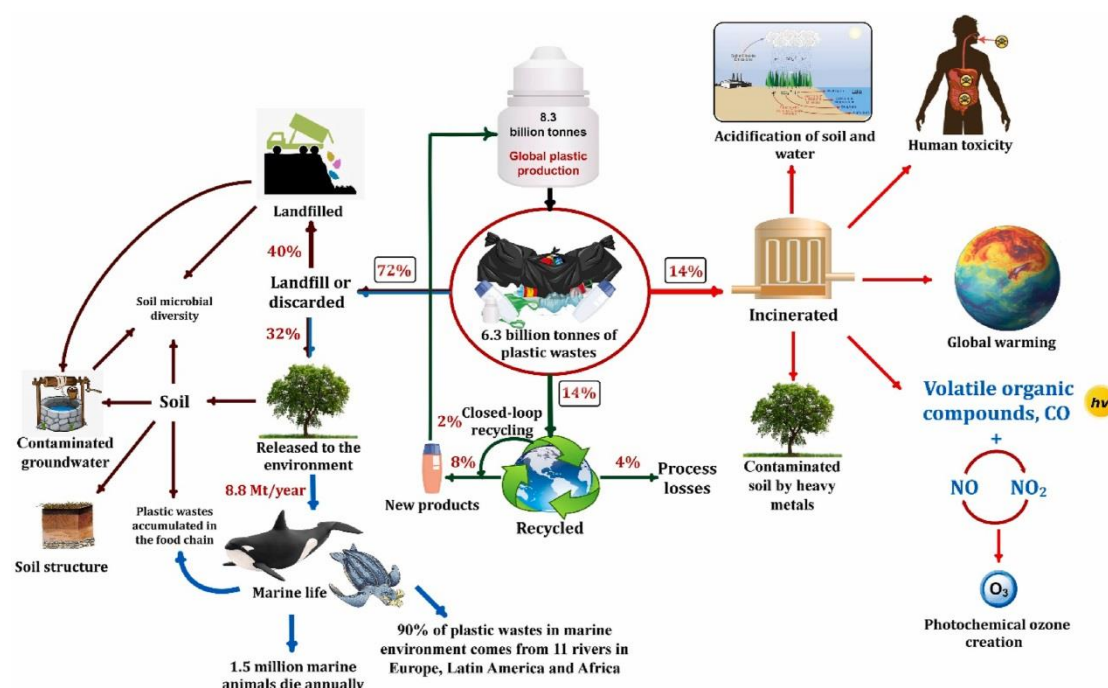


Figure 1. Illustration of the fate of plastic materials with harmful impacts on the eco-system

1.2. The effects of fossil fuel on the environment

When fossil fuels are burned, they emit greenhouse gases like carbon dioxide that trap heat in the earth's atmosphere and contribute to climate change. Some of the climate externalities of fossil fuels include:

- Ocean acidification: At least a quarter of the carbon dioxide emitted from fossil fuels is absorbed by the ocean, changing its chemistry (pH). The increased acidity makes it harder for marine organisms to build shells and coral skeletons. Over the last 150 years, ocean acidity has increased by 30 percent, posing threats to coral reefs, fishing, tourism, and the economy.
- Air pollution: Fossil fuels produce hazardous air pollutants, including sulfur dioxide, nitrogen oxides, particulate matter, carbon monoxide, and mercury, all of which are harmful to the environment and human health (as discussed in the health section below). Air pollution from fossil fuels can cause acid rain, eutrophication (excessive nutrients that can harm aquatic ecosystems by lowering oxygen levels), damage to crops and forests, and harm to wildlife.
- Water pollution: From oil spills to fracking fluids, fossil fuels cause water pollution. Each fracking well uses between 1.5 million to 16 million gallons of water, and the resulting wastewater can be toxic, often containing substances like arsenic, lead, chlorine, and mercury that can contaminate groundwater and drinking water[15].



Figure 2. The environmental impacts of fossil fuels

Chapter two

Literature review

2.1. Processes for hydrogen production

In general, hydrogen gas is produced by several industrial methods. Nearly all of the world's current supply of hydrogen is created from fossil fuels. Most hydrogen is gray hydrogen made through steam methane reforming. In this process, hydrogen is produced from a chemical reaction between steam and methane, the main component of natural gas. Producing one tonne of hydrogen through this process emits 6.6–9.3 tonnes of carbon dioxide[16]. When carbon capture and storage is used to remove a large fraction of these emissions, the product is known as blue hydrogen[17].

Green hydrogen is usually understood to be produced from renewable electricity via electrolysis of water [18][19]. Less frequently, definitions of green hydrogen include hydrogen produced from other low-emission sources such as biomass.[20] Producing green hydrogen is currently more expensive than producing gray hydrogen, and the efficiency of energy conversion is inherently low [21]. Other methods of hydrogen production include biomass gasification, methane pyrolysis, and extraction of underground hydrogen .

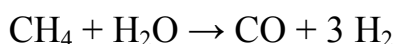
2.1.1. Steam reforming

Hydrogen is industrially produced from steam reforming (SMR), which uses natural gas [22]. The energy content of the produced hydrogen is around 74% of the energy content of the original fuel,[23] as some energy is lost as excess heat during production. In general, steam reforming emits carbon dioxide, a greenhouse gas, and is known as gray hydrogen. If the carbon dioxide is captured and stored, the hydrogen produced is known as blue hydrogen.

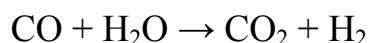
Steam methane reforming (SMR) produces hydrogen from natural gas, mostly methane (CH₄), and water. It is the cheapest source of industrial hydrogen, being the source of nearly 50% of the world's hydrogen.[24] The process consists of heating the gas to 700–1,100 °C (1,300–2,000 °F) in the presence of steam over a nickel catalyst. The resulting endothermic reaction forms carbon monoxide and molecular hydrogen (H₂).

In the water-gas shift reaction, the carbon monoxide reacts with steam to obtain further quantities of H₂. The WGSR also requires a catalyst, typically over iron oxide or other oxides. The byproduct is CO₂ [25]. Depending on the quality of the feedstock (natural gas, naphtha, etc.), one ton of hydrogen produced will also produce 9 to 12 tons of CO₂, a greenhouse gas that may be captured [26].

For this process, high temperature steam (H₂O) reacts with methane (CH₄) in an endothermic reaction to yield syngas [27].



In a second stage, additional hydrogen is generated through the lower-temperature, exothermic, water-gas shift reaction, performed at about 360 °C (680 °F):



Essentially, the oxygen (O) atom is stripped from the additional water (steam) to oxidize CO to CO₂. This oxidation also provides energy to maintain the reaction. Additional heat required to drive the process is generally supplied by burning some portion of the methane.

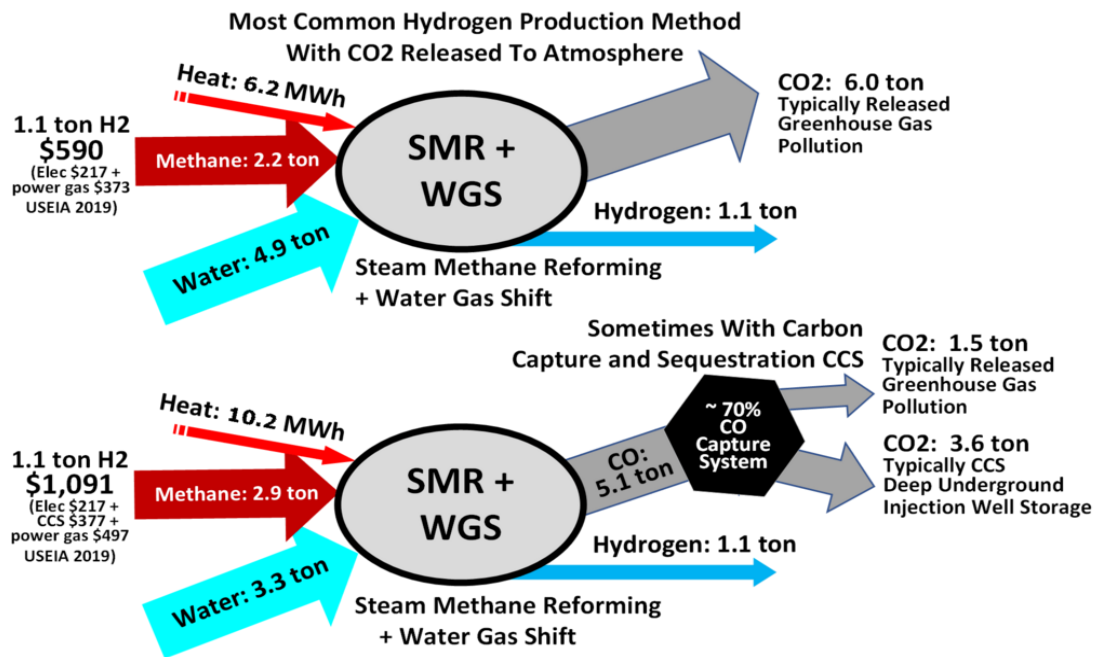


Figure 3. Steam reforming

2.1.2. Electrolysis of water

Hydrogen can be made via high pressure electrolysis, low pressure electrolysis of water, or a range of other emerging electrochemical processes such as high temperature electrolysis or carbon assisted electrolysis.[28]. However, current best processes for water electrolysis have an effective electrical efficiency of 70-80%,[29][30][31] so that producing 1 kg of hydrogen (which has a specific energy of 143 MJ/kg or about 40 kWh/kg) requires 50–55 kWh of electricity.

In parts of the world, steam methane reforming is between \$1–3/kg on average excluding hydrogen gas pressurization cost. This makes production of hydrogen via electrolysis cost competitive in many regions already, as outlined by Nel Hydrogen[32] and others, including an article by the IEA[33] examining the conditions which could lead to a competitive advantage for electrolysis. Water electrolysis is using

electricity to split water into hydrogen and oxygen. As of 2020, less than 0.1% of hydrogen production comes from water electrolysis.[34] Electrolysis of water is 70–80% efficient (a 20–30% conversion loss)[35][36] while steam reforming of natural gas has a thermal efficiency between 70 and 85% [37]. The electrical efficiency of electrolysis is expected to reach 82–86%[38] before 2030, while also maintaining durability as progress in this area continues apace [39].

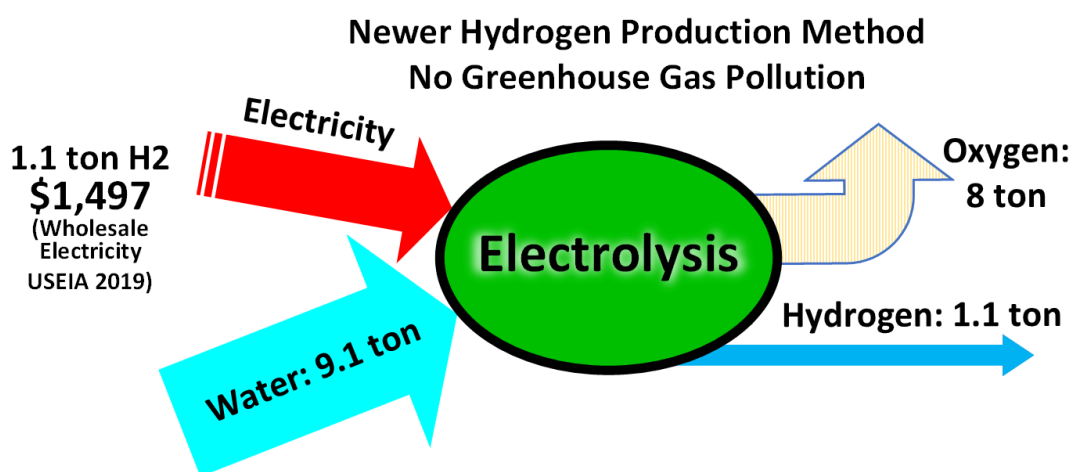


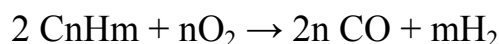
Figure 4. Electrolysis of water

2.1.3. Partial oxidation

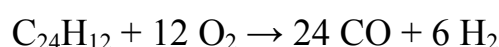
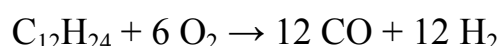
Hydrogen production from natural gas and heavier hydrocarbons is achieved by partial oxidation. A fuel-air or fuel-oxygen mixture is partially combusted, resulting in a hydrogen- and carbon monoxide-rich syngas. More hydrogen and carbon dioxide are then obtained from carbon monoxide (and water) via the water-gas shift reaction [32]. Carbon dioxide can be co-fed to lower the hydrogen to carbon monoxide ratio.

The partial oxidation reaction occurs when a substoichiometric fuel-air mixture or fuel-oxygen is partially combusted in a reformer or partial

oxidation reactor. A distinction is made between thermal partial oxidation (TPOX) and catalytic partial oxidation (CPOX). The chemical reaction takes the general form:



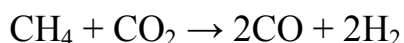
Idealized examples for heating oil and coal, assuming compositions $\text{C}_{12}\text{H}_{24}$ and $\text{C}_{24}\text{H}_{12}$ respectively, are as follows:



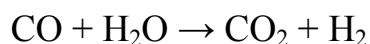
2.1.4. Dry reforming

Is a method of producing synthesis gas (mixtures of hydrogen and carbon monoxide) from the reaction of carbon dioxide with hydrocarbons such as methane with the aid of noble metal catalysts (typically Ni or Ni alloys).[40][41] Synthesis gas is conventionally produced via the steam reforming reaction or coal gasification. In recent years, increased concerns on the contribution of greenhouse gases to global warming have increased interest in the replacement of steam as reactant with carbon dioxide [42].

The dry reforming reaction may be represented by:



Thus, two greenhouse gases are consumed and useful chemical building blocks, hydrogen and carbon monoxide, are produced. A challenge to the commercialization of this process is that the hydrogen that is produced tends to react with carbon dioxide. For example, the following reaction typically proceeds with lower activation energy than the dry reforming reaction itself:



Another issue with dry reforming is situated in the fact that it operates at conditions that produces water. As a result, this water can lead to unwanted back-reaction to CO_2 via the water-gas shift reaction. To prevent CO_2 from being formed, and consequently losses in CO yield, CO_2 can be adsorbed onto calcium oxide. Consequently, the process forms only CO and H_2O , increasing the utilization efficiency of the feedstocks. This process is better known as super-dry reforming.[43]

CO_2 can be dry reformed in to CO gas at 800-850 °C by reacting with petcoke, biochar, coal, etc. using low cost iron based catalysts. Using cheaper renewable electricity like solar or wind energy, this cheaper method converts petcoke and green house gas CO_2 in to useful fuel like methanol achieving carbon capture and utilization[44]. Some CO gas is converted in to hydrogen via water-gas shift reaction.

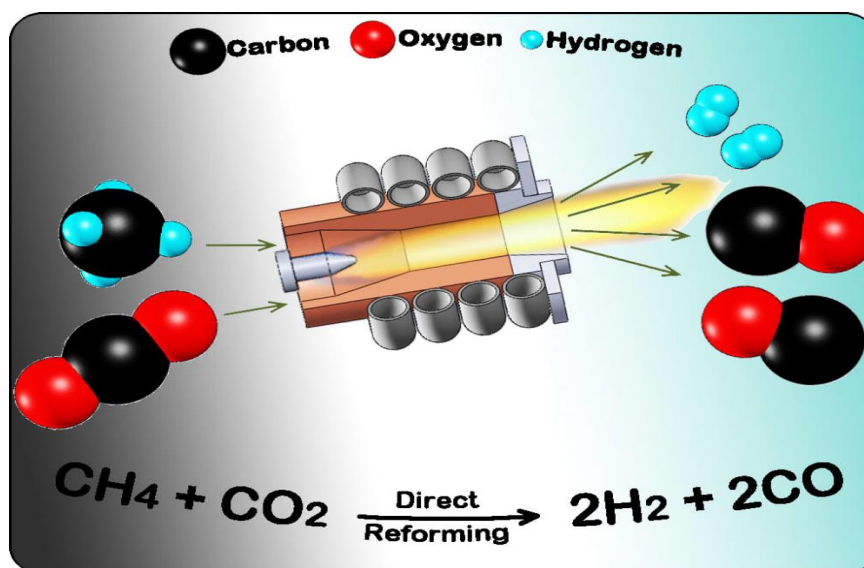


Figure 5. Dry reforming

2.1.4.1. Dry reforming of waste polymers

The vast majority of plastic recycling is through mechanical recycling. However, alternative methods for producing fuels and petrochemical feedstocks from waste plastics are being investigated [45]. One such process is dry reforming, where carbon dioxide is used instead of steam for the catalytic reforming of hydrocarbons for the production of syngas. Dry reforming processes involve cracking of the high molecular weight feedstock hydrocarbons with carbon dioxide to produce synthesis gases (syngas - hydrogen and carbon monoxide). Current research is more focused on the dry reforming of methane, ethanol and glycerol [46]. However, waste plastics represent a feedstock which has a high content of carbon and hydrogen which may be suitable as an alternative source for the process [47]. Therefore, the dry reforming of waste plastics could be a potential treatment process for the production of syngas. The waste plastics may be pyrolysed to produce a suite of hydrocarbon gases which are passed directly to a second stage reactor where dry reforming takes place in the presence of the carbon dioxide and a catalyst [48].

A further point in support of the dry reforming process is the current interest in finding viable uses for the large amounts of carbon dioxide which are predicted to be available in the future through carbon dioxide capture and storage processes. The prediction of the expansion of such carbon capture processes are expected to mitigate against climate change.

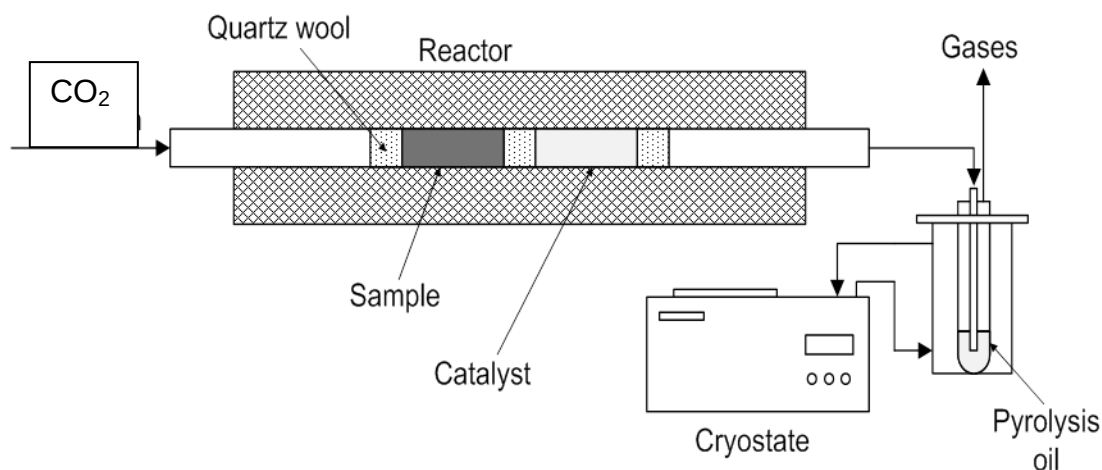


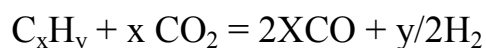
Figure 6. Reactor for dry reforming of waste polymers

2.2. The influence of catalyst type on dry reforming

To improve the dry reforming process, there has been research into the development of catalysts to enhance the production of syngas. Recent reviews on the use of catalysts in the dry reforming process suggest that Ni-based catalysts are the most suitable option due to their low cost and relatively high catalytic activity [49]. Studies have reported that the addition of a metal promoter to Ni-based catalysts can further improve the production of syngas from the dry reforming process by improving the structure and uniformity of the catalyst particles resulting in better metal dispersion. Nickel based catalysts are proved their efficiency in the decomposition reaction of waste plastics. While zeolite catalysts are considered as the best supporter due to their ability to promote the more cracking of waste polymers

2.2. The influence of other parameters on dry reforming

Reaction temperature has significant influence on the hydrogen yields and other gases product as the H₂ and CO yields were increased with temperature. The marked increase in the syngas yields at high temperature was attributed to the CO₂ dry reforming reaction since the reaction require elevated temperature to occur as a result of low potential energy of methane and carbon dioxide



3. Conclusion

There is a large debate on whether to use dry reforming or steam reforming to produce H₂-rich stream from different feedstocks. The CO₂ atmosphere influences the products yields by increasing the amount of hydrogen. The syngas yields from waste polymers were improved by the using of carbon dioxide as a carrier gas, furthermore, the carbon deposition on the catalyst surface was reduced for the same reason. the DR reactions show significant environmental benefits by consuming the CO₂ (greenhouse gas) as an oxidant. In DR reactions, the CO₂ to feedstock molar ratio is the very important parameter to produce H₂ rich stream or syngas mixture with different H₂/CO ratios. The use of catalysts for the DR reactions leads to the decrease of the energy required for the process and the increase of the rate and selectivity towards the desired products. From literature, the results show that the introduction of CO₂ dry reforming of the products of plastics pyrolysis in the absence of a catalyst dramatically increased the total gas production to over 90 wt.% for all of the plastics. Furthermore, The introduction of catalyst significantly improved the production of syngas comprising the H₂ and CO content of the product gases.

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