Chapter 2
Waste Plastics to Hydrogen (H_2) Through Thermochemical Conversion Processes

M. G. Rasul, M. A. Sattar, M. I. Jahirul, and M. M. Hasan

Abstract Plastic products are essential parts of modern life as they are used in household items, packaging, electronics, building, automotive and many more. Amongst about 350 Mt of waste plastics produced worldwide, only about 20% are recycled and 80% are landfilled. The landfilled waste plastic has a serious negative impact on the environment which causes land diversity, havoc in marine, etc. Pyrolysis of waste plastics into usable energy products such as syngas, char and liquid oil can alleviate the burden of plastic waste management. While pyrolyzed liquid oil can be converted to plastic diesel through distillation and hydrotreatment processes, and char can be used for agricultural purposes, the pyrolysis syngas can be processed further through different reforming processes to produce H_2. H_2 is expected to dominate in fuel sector by substituting fossil fuels. In this paper, the experimental findings of waste plastics pyrolysis into oil, char and syngas are reported which shows that waste plastic can be converted to liquid oil by about 80%, remaining are approximately 10% char and 10% syngas. The possibility of H_2 production from pyrolysis syngas through different reforming processes such as steam, partial oxidation, autothermal, plasma, aqueous phase, etc. are reviewed and critically analysed in this paper. The literature indicated that 530 Mt of H_2 is needed to achieve net zero by 2050 worldwide. It is envisaged that converting all waste plastic into H_2 will meet the demand of H_2 and support net zero goal by 2050. Amongst different reforming processes, steam reforming is better than others to produce H_2 from syngas. Therefore, the waste plastics can be a significant potential source of H_2 and will benefit the society and the environment from negative impact and support achieving net zero by 2050.

Keywords Waste plastic · Pyrolysis syngas · Reforming processes · H_2 production · Net zero by 2050

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2.1 Introduction

Plastic is used in every part of daily life which includes food packaging, electrical devices, household utensils and many more because of their favorable properties. The production of plastic items is increasing day by day and the safe disposal of waste plastic has become an urgent need to all countries in the world. Global plastic production exceeds 350 million tons in 2018 (Kusenberg et al. 2022). The main types of waste plastic that are found in the municipal waste are high-density polythene (HDPE), polyvinyl chloride (PVC), low-density polythene (LDPE), polythene terephthalate (PET), polypropylene (PP), and polystyrene (PS) (Kusenberg et al. 2022). Most of the plastics are non-biodegradable and take longer time to degrade which are causing many problems in land and seas. Incineration of plastic produces noxious and toxic fumes which poses health hazard. Therefore, incineration is not a good solution for disposing the plastics. Pyrolysis can an alternative and comparatively safe process of utilising waste plastic. Pyrolysis is a process of decomposing solid wastes into oil, char, and syngas in the absence of oxygen. The oil can be used as a fuel for internal combustion engine after refinement to plastic diesel. The char can be used for soil abatement and other purposes. The Syngas are mostly neglected and released into the atmosphere.

Combustion of hydrocarbons produces CO_2 and other harmful GHG gases which has detrimental effects on the environment. Researchers are searching for environmentally friendly alternative fuel, for example H_2 as a clean energy which can be a suitable substitute for fossil fuel (Hazrat et al. 2022; Sarker et al. 2023). H_2 has many favorable properties such as the highest calorific value and does not produce any harmful gases during the combustion. The demand of H_2 in 2020 was around 88 Mt and expected to increase to 530 Mt in 2050 to achieve net zero goal (IEA 2019; PwC 2017).

The syngas, normally discarded into the atmosphere, can be converted into H_2 using different reforming processes. In this paper, the pyrolysis of waste plastic is discussed first, then different types of reforming processes are briefly described to produce H_2.

2.2 Pyrolysis of Waste Plastics

As introduced earlier, pyrolysis is a thermochemical process of decomposing the waste plastic into oil, biochar, and syngas in the absence of oxygen. There are different types of pyrolysis reactors used for pyrolysis as shown in Fig. 2.1 (Lewandowski et al. 2019; Papari and Hawboldt 2015). Authors used 20L vertical fixed bed reactor for their own research, the pictorial view of which is shown in Fig. 2.2.

In this reactor, three electrical heaters are used to heat the reactor and k-type thermocouples are used to measure the pyrolysis operational temperature. Before any experiment, nitrogen gas is purged through the system to make it inert (free of

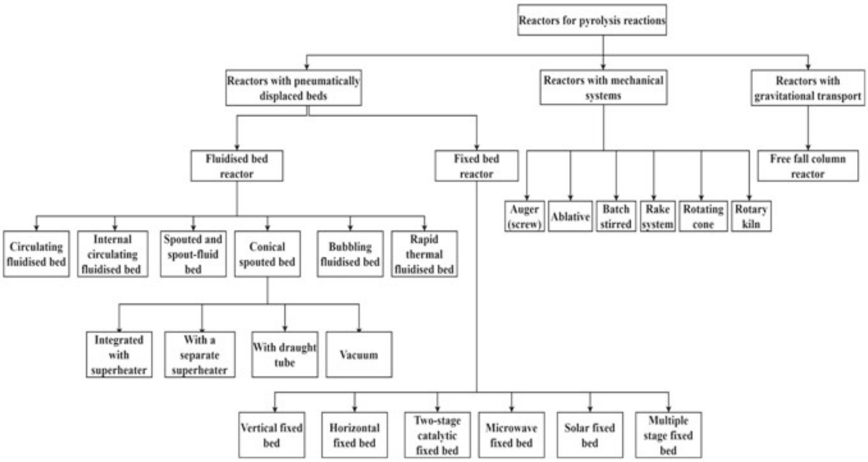

Fig. 2.1 Different types of reactor for pyrolysis (Lewandowski et al. 2019; Papari and Hawboldt 2015)

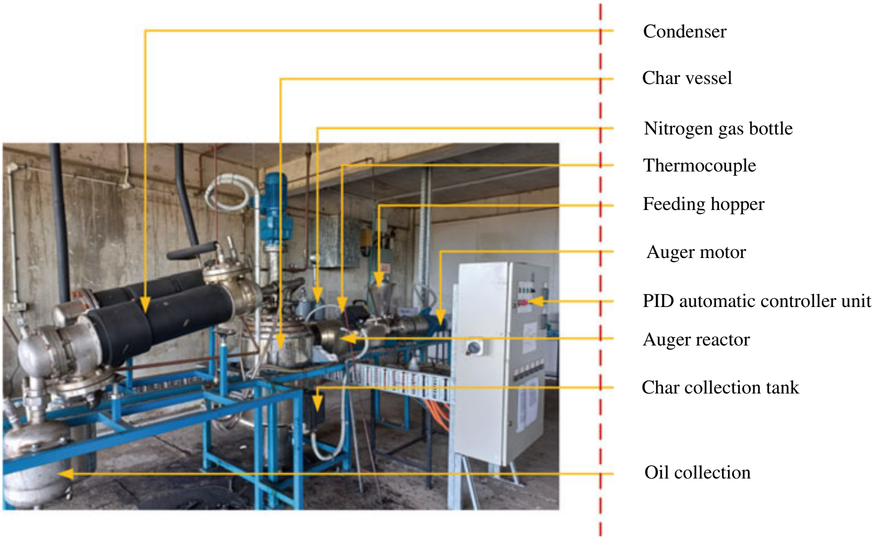

Fig. 2.2 Pyrolysis set up at Fuel and Energy Research Laboratory, CQU Rockhampton campus

oxygen). A PID controller is used to operate the reactor at atmospheric pressure. A chiller unit is used to condense the pyrolysis vapors after decomposition of feed materials into crude oil. Polyethylene glycol solution circulated chiller unit temperature can be set − 5 to 20 °C. The mixed waste plastic was fed into the reactor through a feeding hopper and kept sealed after nitrogen was purged. Then, the data logging system and electrical heater were turned on to heat the reactor to the desired temperature and maintained at the desired time. During this period, the feedstock was converted into vapor and char. The vapor passed through a water-cooled condenser to produce crude oil which was collected from the oil collection tank. The system was turned off until the reactor temperature reduced to room temperature, then char was taken out from the bottom opening of the reactor. The crude oil can be refined through distillation process to produce oil equivalent to standard diesel to use in diesel engine. The ultimate and proximate analysis of different types of waste plastics can be found in Zhou et al. (2014). The yield from the pyrolysis of waste plastic is presented in Table 2.1. The results show that the oil yield can be found $\geq$ 80% including findings of the authors, except yield of PET, PE, and few mixtures (Rasul et al. 2022).

2.3 Production from Syngas

Syngas produced from pyrolysis of waste plastic is a mixture of H_2, CO and small amount of CO_2 and methane. The general equation for syngas production from pyrolysis can be expressed by:

$$Waste\,platisctics \overset{Pyrolysis}{\Rightarrow} \mathrm{H_2} + \mathrm{CO} + \mathrm{CO_2} + Hydrcarbon\,gases \tag{2.1}$$

Hydrogen can be separated from the syngas. The CO can be transformed into CO_2 by producing H_2 using different reforming process as shown in Fig. 2.3 (Rasul et al. 2022). Reforming is the process of converting low quality hydrocarbon to high quality hydrocarbon which has been used in industry for quite long. Reforming process can be used to produce H_2 by cracking the natural gas, methane, gaseous hydrocarbon, liquid hydrocarbon, ethanol, methanol, coal, and naphtha. Brief description of different types of reforming processes are given below.

2.3.1 Catalytic Steam Reforming

In this process, steam at high temperatures and pressures is used to crack the hydrocarbon to produce CO and H_2 rich syngas. Steam reforming process can be carried out with and without catalyst. After steam reforming process, water–gas-shift (WGS) process is carried out to convert CO into CO_2 and dissociate H_2 from water. This combined process is used to produce H_2 from the hydrocarbon. The highest efficiency of SR can reach up to 80–90%. The reforming reactions (2.2) to (2.4) are given below:

Oxygenated hydrocarbon:

$$\mathrm{C}_x\mathrm{H}_y\mathrm{O}_z + (x - z)\mathrm{H_2O}(+heat) \rightarrow x\mathrm{CO} + (x + y/2 - z)\mathrm{H_2} \tag{2.2}$$

Table 2.1 Yield of pyrolysis of waste plastic

Plastic types	Yield %			References
	Oil	Char	Syngas	
PP	88.86	1.84	9.3	Martínez et al. (2013)
	83.6	0.4	16.0	Williams and Slaney (2007)
PE	92.3	0.1	7.6	Williams (2006)
	85.3	10.9	3.4	Miskolczi et al. (2004)
PS	80.8	6.2	13	Rehan et al. (2016)
	97	0.5	2.5	Onwudili et al. (2009)
PET	38.89	52.13	8.98	Fakhr Hoseini and Dastanian (2013)
	15	53	32	Kunwar et al. (2016)
HDPE	89.1	6.7	4.2	Abbas-Abadi et al. (2014)
	81.00	17.50	1.5	Seo et al. (2003)
LDPE	75.6	8.2	7.5	Uddin et al. (1997)
	84	14.25	1.75	Supriyanto and Richards (2021)
HDPE[a]	58	6.4	4.6	Authors work
PS	85	5.5	9.5	Authors work
PP	90	3.3	6.7	Authors work
HDPE + PP + PS (1:1:1)	81	8.5	10.5	Authors work
HDPE + PP + PS (1:1:2.2)	85	4.5	11.5	Authors work
HDPE + PP + PS + PET(4:4:3:1)	76	10.5	13.5	Authors work
HDPE + PP + PS + PET + PVC (2.1:3.7:3:2.3:2.2)	64	21.5	14.5	Authors work
HDPE + PP + PS + PET (3:4:3:2)	66	18.75	15.25	Authors work

[a] Rest is wax

Non-oxygenated hydrocarbon:

$$C_xH_y + (x - z)H_2O(+heat) \rightarrow xCO + (x + y/2)H_2 \tag{2.3}$$

WGS process.

$$CO + H_2O(-heat) \rightarrow CO_2 + H_2 \tag{2.4}$$

Catalyst is used to enhance the efficiency of the H_2 production. Nahar et al. (2015) found 94% yield efficiency at 650 °C in the presence of 10 wt.% Ni/Ce-Zr catalyst.

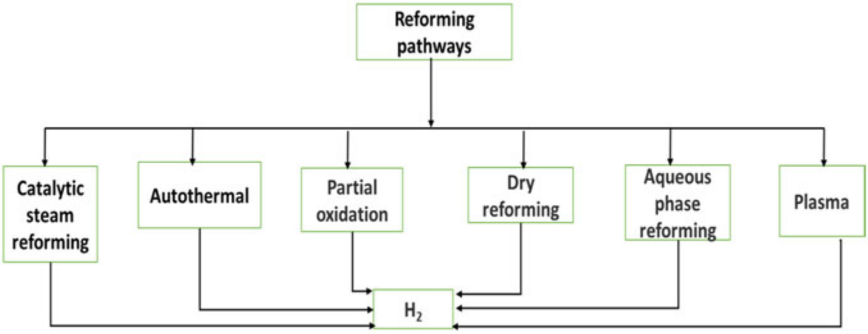

Fig. 2.3 Different types of reforming process to produce hydrogen

Typical operating conditions for cracking natural gas is 3–25 bar and 700–1000 °C in the presence of a catalyst. Steam reform process requires high temperature if catalyst is not used for example CH_4 cracks into various radicals (e.g., C_2H_4, C_2H_2, and C) at 1000 °C, and at over 1500 °C to produce H_2 gas (Rostrup-Nielsen et al. 2011). The water–gas shift process is performed after the steam reforming process to decompose water with CO to increase H_2 production. Haber–Bosch process is being used for quite long to produce H_2. The combination of SR and WGS process has drawn the attention to produce H_2 in recent time. Globally about 48 and 30% of the total H_2 is produced using the SR and WGS methods (Ugurlu and Oztuna 2020).

2.3.2 *Autothermal Reforming*

Autothermal steam reforming (ATR) is a combination of SR and partial oxidation (POX) where fuel, steam, and water input into the reactor to produce H_2. The heat is produced through POX therefore no additional heat is required. This process requires oxygen, so oxygen separation plant is needed to supply and carry out this process. WGS process is carried out to enhance the H_2 concentration in the mixture. ATR process can produce H_2 at a cost of \$1.69–\$2.55 per kg and adding the carbon capture, utilization, and storage (CCUS) techniques can make it green process (Ahmed and Krumpelt 2001). The CO_2 emissions from this process is less than other processes. Catalysts can enhance the production of H_2 as for example the addition of Pd-Zn/γ-Al_2O_3 at 400 °C can produce about 45% (v/v) of H_2 efficiently (Oni et al. 2022). The typical reaction process of the ATR or OSR process is shown in Eq. 2.5.

$$Fuel\left(\mathrm{C}_x\mathrm{H}_y\mathrm{O}_z\right) + air + \text{steam} \rightarrow \mathrm{CO}_2 + \mathrm{H}_2 + \mathrm{N}_2(-\Delta H) \tag{2.5}$$

Cortazar et al. (2022) reported the production of H_2 from several waste plastics (HDPE, PP, PS, PET), mixed plastics, biomass, and HDPE and found that the highest H_2 production was from PP (64.1%), and HDPE (64%).

2.3.3 Partial Oxidation Reforming (POX)

POX process generates heat during the reaction. This process does not require external heat like SR process. POX process produces less H_2 than that of SR process. POX reforming process is carried out at sub-stoichiometric quantity of oxygen as can be seen in Eqs. 2.6 and 2.7 (Rasul et al. 2022). POX is exothermic whereas SR is endothermic process.

$$CH_4 + \ 1/2O_2 \rightarrow CO + 2H_2(+\Delta H) \tag{2.6}$$

$$\text{WGS reaction}: \ CO + H_2O \rightarrow CO_2 + H_2\,(+\,\text{small amount of heat}) \tag{2.7}$$

Hydrogen is separated using the pressure swing absorption process. Three-fourth of the total global H_2 is produced usually using this technology. The generic POX reaction of hydrocarbon fuels is presented in Eq. 2.8.

$$Hydrocarbon\ fuel: C_nH_m + (n/2)O_2 \rightarrow nCO + \left(\frac{m}{2}\right)H_2 + heat \tag{2.8}$$

The process can be carried out without catalyst therefore there is no drawback of degradation of catalyst effectiveness. The main challenge of POX is the requirement of high temperature and lower H_2/CO ratio (Rasul et al. 2022). WGS is conducted after POX to convert CO to CO_2. POX reaction for methanol and ethanol can be expresses as in the Eqs. 2.9 and 2.10, respectively (Rasul et al. 2022).

$$POX of methanol: CH_3OH + \frac{1}{2}O_2 \rightarrow 2H_2 + CO_2 + \left(\Delta H \approx 192.2\frac{kJ}{mol}\right) \tag{2.9}$$

$$POX of ethanol: C_2H_5OH + \frac{3}{2}O_2 \rightarrow 3H_2 + 2CO_2 + \left(\Delta H \approx 620.3\frac{kJ}{mol}\right) \tag{2.10}$$

Catalyst can lower operating temperature. Agrell et al. (2001) used Cu (%40) Zn (%60) to conduct POX reaction of methanol at 185–215 °C and found that the H_2 production increases.

2.3.4 Dry Reforming (DR)

In drying reforming process, CO_2 and CH_4 react to produce CO and H_2 at 700–900 °C. The main challenge of this process is the deactivation of the catalyst and low H_2/CO ratio (Uddin et al. 1997). In the dry reforming process CO_2 is used to crack the hydrocarbon to produce the syngas. The dry reforming process can be expressed in Eq. 2.11 below.

$$2C_xH_y + CO_2(+heat) \rightarrow 2(x + 1/2)CO + yH_2 \tag{2.11}$$

The Boudouard and reverse water–gas shift (RWGS) reaction can be written as:

$$Boudouard\,reaction : 2CO \leftrightarrow CO_2 + C_{(s)};\ (\Delta H = -172\,kJ/mol) \tag{2.12}$$

$$RWGS\,reaction : CO_2 + H_2 \leftrightarrow H_2O + CO;\ (\Delta H = 41\,kJ/mol) \tag{2.13}$$

The efficiency of this process depends on the performance of the catalyst therefore a highly active catalyst is desired. Ru, Pt and Pd show higher catalytic activity for DR process (Medeiros et al. 2022). Ballarini et al. (2019) reported that K-L Zeolite, K-Al_2O_3, K-Mg/Al oxides, and MgO along with Pt-based catalyst has high stability to produce H_2. The maximum yield was obtained when MgO/Pt was used. Xie et al. (2018) found that PtCo/CeO_2 has high stability and effectiveness to produce H_2. Hajizadeh et al. (2022) found that 48.07 kg/h biogas produces 8.11 kgH_2/h in presence of Co-Ni-Al_2O_3 catalyst.

2.3.5 Aqueous Phase Reforming

In the aqueous phase reforming (APR), oxygenated/non-oxygenated hydrocarbon is cracked in aqueous solution at lower temperature to produce H_2. This process operates at 200–250 °C and at 60 bar in the presence of catalyst. Platinum (Pt), tin (Sn), cobalt (Co) or nickel (Ni)-based metallic, and alumina can be used as a catalyst support (Shabaker and Dumesic 2004). The reaction can be given by Eq. 2.14.

$$C_xH_y + 2xH_2O \rightarrow +\left(2x + \frac{y}{2}\right)H_2 + xCO_2 \tag{2.14}$$

This process consumes less energy than other processes and is termed as greener and therefore this process is an economical process to produce H_2 from organic compounds. There is complexity of producing H_2 directly from the biomass which can be overcome by converting the biomass into liquid as an intermediate material and then producing H_2 through reforming. The stoichiometric APR reaction of sugar–alcohol sorbitol ($C_6O_6H_{14}$) in the presence of Pt catalyst can be shown as in Eq. 2.15 (Shabaker and Dumesic 2004):

$$C_6O_6H_{14}(l) + 6H_2O(l) \leftrightarrow 13H_2(g) + 6CO_2(g) \tag{2.15}$$

H_2 and CO_2 react in the presence of catalyst therefore it is essential to capture CO_2 otherwise the overall efficiency of the process will decrease. In the APR process, bimetallic catalysts like PtNi, PdFe, PtFe show better performance than that of monometallic catalysts.

2.3.6 Plasma Reforming

Electron at high temperature in the plasma supports the decomposition of organic material. At high temperature, waste plastic and other hydrocarbon decomposes into CO, H_2, and other hydrocarbons. There are different types of plasma reactors exists, such as dielectric barrier discharge (DBD) reactor, pulse plasma reactor, gliding arc plasma reactor, and microwave plasma reactor etc. (Budhraja et al. 2023). Comparative analysis of different types of plasma reactors can be found in Budhraja et al. (2023). Song et al. (2019) investigated the conversion of ammonia into methane and H_2 using DBD plasma reactor and found that catalyst enhances the conversion of methane to H_2. Morgan and ElSabbagh (2017) used a pulse plasma reactor to convert methane into H_2 and found that 92% of methane converted into H_2. Wang et al. (2019) used gliding arc reactor to convert n-heptane and found 50.1% H_2. Wang et al. (2021) used MW plasma reactor and found 94% methane conversion with 74% H_2 yield. The comparison of different reforming processes is given in Table 2.2.

2.4 Challenges and Conclusions

Waste plastic is posing a global threat for environment as only 20% of the total used plastic is disposed safely and the rest is thrown to landfill which takes more than hundred years to decompose. Incineration and other technique are not sustainable and environmentally friendly because it produces toxic gases. Literature review suggests that pyrolysis of waste plastic can produce more than 80% liquid oil which can be refined to diesel fuel for engine. The syngas produced pyrolysis of waste plastic can be used as feedstock to produce H_2 through reforming processes. It is found from the literature that steam reforming process is better than other reforming processes to produce H_2 which has efficiency of more than 90%. The conversion of waste plastic into H_2 through pyrolysis and reforming process can solve the issue of safe disposal of waste plastics. The production of H_2 can also help meet the demand of H_2 and achieve the net zero by 2050. The main challenges of producing hydrogen from waste plastic using reforming process are:

- Cost of producing hydrogen from waste plastic.
- Scale up the production process for mass production.
- Deactivation and reduction of catalyst effectiveness.

Table 2.2 Comparison of different reforming process to produce H_2

Reforming Process	Catalyst	Operating condition	Feedstock	Advantage	Disadvantage	References
Steam	$Cu/ZnO/Al_2O_3$, Cu/Zn/Mg, Pt, Ru, Rh/SiO_2	200–750 °C 0.05–37 bar	Organic compound	Highly efficient (65–75%)	Endothermic	Supriyanto and Richards (2021), Rasul et al. (2022) and Nahar et al. (2015)
Autothermal	Pd-Zn/γ-Al_2O_3	950–1050 °C 30–50 bar	Plastic, biomass	No external heat required	Need Oxygen supply	Rostrup-Nielsen and Hansen (2011), Ugurlu and Oztuna (2020) and Ahmed and Krumpelt (2001)
Partial oxidation	Au-CuO-ZnO, Cu40Zn60	1150 – 1315 °C, 6 bar	Heavy hydrocarbon, Petrol, Diesel	Produce heat, less CO_2	lower H_2/CO ratio, High temperature	Uddin et al. (1997) and Oni et al. (2022)
Dry	K-L Zeolite, K-Al_2O_3, K-Mg/Al oxides, MgO/Pt, PtCo/CeO_2	700 °C and 900 °C	Hydrocarbon, methane	Sustainably promising thermos-catalytical processes	Catalyst deactivation, Equilibrium, endothermic	Uddin et al. (1997), Cortazar et al. (2022), Agrell et al. (2001), Medeiros et al. (2022) and Ballarini et al. (2019)
Aqueous phase	Pt, Sn, Co or nickel (Ni)-based metallic catalysts	200–250 °C, up to 60 bars	Oxygenated/non-oxygenated hydrocarbons, biomass	Less energy consuming	H_2 and CO_2 react	Xie et al. (2018)
Plasma	NiO/Al_2O_3 Cu/ZnO	300–400 °C	Hydrocarbon, Biomass,	Less costly and bulky	Unstable plasma discharge, nonuniform temperature	Hajizadeh et al. (2022), Shabaker and Dumesic (2004), Budhraja et al. (2023), Song et al. (2019) and Morgan and ElSabbagh (2017)

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