

PLASTIC WASTE AS AN ALTERNATIVE SOURCE OF FUEL THROUGH THERMOCHEMICAL CONVERSION PROCESS- A REVIEW

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ABSTRACT.

With the progression of science and technology, the fabrication of plastic products has heightened day by day and the extensive usage of these products has brought convenience to the whole world. Though these products have proved to be a favorable addition to our daily life, they also generated acute environmental concern globally since most waste plastics are landfilled due to poor maintenance and disposal even though they are completely recyclable and reusable. These plastic wastes can become sustainable feedstock and promising alternatives to non-renewable fossil fuels and can become third-generation biofuels using thermal conversion technologies like hydrothermal liquefaction (HTL) and Pyrolysis. This paper explored the technology along with the detected plastic type that would be best for the production of maximum bio-oil yield from plastic waste. The ultimate composition components value range of crude oil obtained from the elemental analysis data comprised C (63.6-90.5%), H (6.9-9%), O (0.9-17%), and HHV (29.44-42.61MJ/kg). This review article also explored the reaction parameters which include the type of reactor, thermal conversion technology used, temperature, pressure, the addition of solvent, and retention time are most important. In a pressurized batch reactor at a temperature of about 425°C, Polystyrene produces the highest yield percentage of about 97% through pyrolysis. However, due to the high abundance and easy availability, and favorable thermal properties of plastic this review article suggests, one of the best courses of action to produce biofuel can be the proper utilization of unmanaged plastic waste as raw feedstocks through a suitable conversion process which will prove a double-edged solution to minimize plastic waste along with getting valuable crude products.

Keywords: *Plastic Waste, Biofuel, Hydrothermal Liquefaction (HTL), Pyrolysis, Higher Heating Value (HHV)*

INTRODUCTION

Currently, plastic is found to be one of the vital pollutants in the environment which is produced on a vast scale and in the most unplanned way due to the reason of manufacturing bags, containers, and various construction materials (Ma, C et al., 2016). Worldwide plastic consumption has reached about 322 million tons in 2015, representing a 4% increment over 2014 (Plastics- The Facts 2016). According to Seshasayee et al. (2020), approximately 368 million tons of plastics were generated worldwide in 2019 and out of which roughly 76 wt % of plastic waste was landfilled. At best, 9 wt % was recycled, and 15 wt % was used for energy recovery via combustion. In developed countries particularly in the US, the quantity of plastic waste has reached 39.3 million tons in 2014, indicating a 13% increment over 2011 (M. E Diaz Barriga, 2011; A. T Bourtsalas, 2014). In the UK, plastic waste has upsurged by over 60% from 2005 to 2015. An approximated 4–12 million tons of plastic waste end up in the oceans annually (Brooks et al., 2018). Recent studies evaluate that a minimum of 5.25 trillion plastic particles are afloat in the ocean, weighing almost 300 000 tonnes (Eriksen et al.,2020). The outcome is the

production of several deleterious substances such as hydrochloric acid, sulfur dioxide, and heavy metals as well as potentially carcinogenic emanations. The lightning rate of plastic consumption throughout the world has led to the generation of incrementing proportions of waste with complicated disposal (Panda et al., 2010).

Untreated and unmanaged plastic waste can prove to be deleterious, hazardous, and destructive for the environment and the whole ecosystem. The different kinds of used plastics include low-density polyethylene (LDPE), high-density polyethylene (HDPE), polypropylene (PP), polyvinyl chloride (PVC), polystyrene (PS), and polyethylene terephthalate (PET), which account for 74.2 wt% of the European plastic demand in 2012 (Yu J et al., 2016). These used plastics produce a vast proportion of municipal solid waste (MSW). The direct combustion method could be a solution to this widespread problem. But for the welfare of the environment and the whole ecosystem, plastic waste disposal through this method is not appreciated due to the development of toxic pollutants such as SO₂, NO_x, heavy metals, and particulates in both flue gas and solid residues. Thus, more efficient handling, control and disposal strategies for plastic waste are mandatory. Landfilling and incineration have become more extravagant and environmentally risky at the present time. Also, the recycling of waste plastics is alluring more and more awareness as a substitute to ingest extensive proportions of post-consumer plastics (Wu et al., 2010; Shah et al., 2018).

However, with the depletion of fossil fuel energy and the heightening perception regarding environmental sanctuary, the expansion of new, environmentally friendly, and renewable energy has become the pivoting point of international energy research (Wu, X et al., 2017; Zhang et al., 2014). Among diverse recovery processes of plastics, one of the best options with great potential is the recovery of energy or chemicals through several thermochemical conversion techniques (Panda et al., 2010; Wu et al., 2010; Singh et al., 2016). Bio-oils generated from the thermochemical conversion of several biomass resources can be regarded as a potential substitute for petroleum (Chen et al., 2012; Guo et al., 2021). In this review, we have considered several plastic types as potential biomass resources used in thermochemical conversion.

The abundant constituents in plastic wastes are mainly carbon and hydrogen, and plastics generally have high H/C ratios (Açıklalın, and Karaca, 2012; Shui et al., 2011; Shui et al., 2013; Jongwon Kim, 2016), and correspondingly high energy contents. Therefore, the metamorphosis of waste plastics into biofuel can be a sophisticated solution to both the issue of plastic waste and sustainable energy production. Energy recovery from waste plastics can serve as a prominent alternative. There can be direct energy recovery and also indirect methods. But direct energy recovery can release pollutants such as oxides of sulfur and nitrogen (Verma R et al., 2016). Whereas, indirect methods (e.g., pyrolysis, gasification, hydrothermal liquefaction) are also available. Pyrolysis of plastics can produce energy-dense oil, chemicals, and gases (Hallet et al., 2006; Buekens A et al., 2006). Pyrolysis is a very feasible and sustainable waste-handling technique in the treatment of municipal solid wastes incorporating carbonaceous materials like plastics, and other biomasses (Zhou et al., 2016). The method of pyrolysis necessitates the degradation of complex molecules like polymers (plastics) into short-chain, less composite molecules by the implementation of heat under inert conditions. Most pyrolysis plants employ high temperatures (700°C) to moderate temperatures (500°C) in the presence of the appropriate catalyst (Samanta et al., 2016; Kenny et al., 1995; Kumar et al., 2011; Siddiqui and Redhwi, 2009; Williams and Williams, 2002). As for the quality, the obtained crude oil contains a high calorific value of around 40 MJ/kg (McClain et al., 2014; Desai SB et al., 2015; Pinto F et al., 2002; Blazso M et al., 2010). On the other hand, hydrothermal liquefaction (HTL) is a process that emanated from the valorization of biomass, and it can also be applied to plastic waste to generate an ample proportion of biocrude (Nakagawa T et al., 2015). For wet biomass, hydrothermal liquefaction (HTL) has been stated to be a profitably feasible route, which can generate high biomass conversion yields and can produce bio-crudes with the appreciable proportion of higher heating values (30–39 MJ/kg), lower oxygen contents (10–20 wt%), and a controllable water content (0–5 wt%) (Peterson et al., 2008). HTL evades the requirements for drying the biomass and instead uses pressurized vessels to keep water in the liquid phase and the pressure (10–25 MPa) is generated at moderate temperatures (290–350°C) (Akhtar and Amin, 2011; Andrady et al., 2009; Demirbas A., 2004; Shen et al., 2016). The proximate and ultimate composition of biomass and crude yields is one of the most vital characterization methods of the selected waste biomass. These explorations are mandatory for efficient, systematic, and clean utilization, while the HHV of these materials influences the quantitative energy content of these fuels (Parikh et al., 2006). In these thermal conversion techniques, the most usual parameters inspected in the proximate and ultimate composition of feedstock biomass include moisture content, VM, FC, ash, carbon, hydrogen, nitrogen,

oxygen, and sulfur contents, high heating value (HHV), and also, hemicellulose, cellulose, lignin contents (Vassilev et al., 2016) because these parameters influence the degradation kinetics and they have a considerable relevance during these thermal conversions. The bio-oil generated due to these different thermal conversions has the potential to become substitute fuel due to the adequate amount of HHV.

There were many previous research works regarding plastic waste to the bio-crude issue but very few papers focus on both hydrothermal liquefaction (HTL) and pyrolysis conversions techniques. Focusing on the comparative analysis side by side of both methods, the key aim of this review is to properly utilize this unmanaged robust plastic waste and convert them into a valuable product. Subsequently, this paper is also a foundation to convert this enormous unmanaged plastic waste biomass to the production of next-generation cleaner energy. This process not only safeguards the environment but is also expected to make enough capital from the Waste to Energy industry in the long run.

MATERIALS AND METHODOLOGY

Selected Plastic Waste Resources Overview

The selection of plastic waste for the generation of biofuel mostly depends upon the type of plastic, availability, recycling percentage, decomposition longevity, and toxicity level. For example, PETE is the most commonly recycled plastic worldwide and it takes lesser time (5-10 years) to decompose. From several experimented results, it can be demonstrated that the crude oil production rate from PETE is not up to the mark. The recycling percentage of PVC is <1 and it rarely decomposes. But due to the production of toxic gases in presence of heat and low energy conversion rate, it is not so eligible either. Polystyrene (PS) is not so eligible in recycling programs due to its lightness that is easily blown away and generates litter during collection. PS can also be hazardous in presence of heat and can produce toxic gases (Sharuddin et al.,2016). But it is the most eligible for the WtE resource program due to the production of elevated yield percentage. Also, PP and PE can be appreciable addition as the ideal feedstocks due to their abundance and adequate biofuel fabrication rate. Also, the co-processing of plastic with another feedstock (microalgae, wood, and agricultural forestry residues) can provide adequate hydrogen with a depletion in the oxygen percentage, which can lead to an increment in the liquid yield and improvement in the oil quality (Bhattacharya et al.,2009).

Feedstock Sampling and Collection

In this review, collection, inspection, and evaluation of several high-density plastic biomass like High-Density Polyethylene (HDPE), Low-Density Polyethylene (LDPE), Polypropylene (PP), Polystyrene (PS), Polyvinyl Chloride (PVC), Poly (butylene terephthalate) (PBT), Polycarbonate (PC), Poly (ethylene terephthalate) (PET), Poly (lactic acid) (PLA), Poly (methyl methacrylate) (PMMA), Poly(oxymethylene) (POM), Poly (phenylene oxide) (PPO), Poly (vinyl alcohol) (PVA), Styrene butadiene (SB) and other feedstocks like microalgae, wood and forestry residues (additional feedstocks data is essential for demonstrating yield variation due to co-processing) data were obtained from the previously published journals and literature. According to the previously published journals, waste plastic samples were either gathered from the location close to the experimentation site, or the rarer and unfamiliar biomass categories were conveyed from their origin to the laboratory via transfer vehicles. For example, waste plastic samples were either collected from household plastic waste materials or post-consumer municipal waste mixed plastic (Das et al.,2018; Paul T. Williams et al.,2007). Pellet-type fractions of waste plastics like polypropylene (PP) and polyethylene (PE) were obtained from a plastic recycling center. Virgin plastic pellets like LDPE, HDPE, and PP samples were obtained from petrochemicals and plastic recycling industries (Das et al.,2018). The plastic materials were sieved to obtain materials with a certain diameter appropriate for proper reaction conditions. These virgin plastic biomasses or their mixture (VMIX) and the mixture of plastic and additional feedstock samples were collected from reliable resources for proper reaction and appreciable augmentation in the bio-oil yield percentage.

Available Thermal Conversion Techniques for Bio-oil Production

Plastic wastes can be turned into valuable energy resources since they are extracted from the petrochemical source which has significant calorific value and it can be a promising alternative to the waste to energy conversion. This conversion can be made possible through several thermal treatment technologies such as gasification, pyrolysis, plasma process, hydrothermal liquefaction (HTL), Supercritical Liquefaction, and incineration (Moustakas et al.,2010). But in this review article, the most effective and worldwide used conversion technologies which include pyrolysis, and hydrothermal

liquefaction (HTL) have been briefly demonstrated, and crude oil yield data has been analyzed and evaluated using these techniques.

Pyrolysis

Pyrolysis is a chemical recycling process that disintegrates organic macromolecules into smaller molecules at elevated temperatures in the absence of oxygen via free radical degradation pathways (Achilias DS et al.,2008). Pyrolysis can occur in a reactor tube and vapor is promptly expanded into a condenser train where it is condensed to the bio-oil (Mohan D et al.,2007). Pyrolysis reactors can be of different types like fixed bed, horizontal steel, semi-batch, batch, fluidized bed, vacuum batch, pressurized batch reactor, etc. In pyrolytic reaction conditions, individual single plastic types in a nitrogen atmosphere can be reacted under higher pressure and at a temperature of about 500 °C in the reactor (Paul T. Williams et al.,2007). Both the pyrolysis and co-pyrolysis experiments can be executed by using an updraft stainless steel fixed bed reactor under atmospheric pressure (Sharuddin et al.,2017). The pyrolysis plant consisted mainly of a feeding system, a fluidized bed reactor, a char removal system, a quench system, and a gas-circulating system. The solid residues can be accumulated in the fixed bed reactor as biochar. And then, the weight of bio-oil and bio-char can be directly measured (Bai et al.,2019).

Hydrothermal Liquefaction (HTL)

Hydrothermal liquefaction (HTL) is a thermal depolymerization process that is used to convert wet biomass and other macromolecules into crude-like oil under high temperatures (200-375 °C) and high pressure (4-25 Mpa) (Akhtar et al.,2011). On the commercial scale, the mechanical shredder should be used for plastic pellets size reduction, followed by a continuous flow HTL reactor. Retsch Cryomill can be used to reduce the size of the plastic particles to the relevant shape required for the reaction. Both the HTL and Co-HTL experiment can either be coordinated in stainless steel batch reactor comprising a certain working volume and in each HTL run, the plastic biomass can be added up directly onto the reactor which remains mechanically sealed and secured then it can be accommodated in the furnace for further heating. After the implementation of the reaction, the reactor can either be cooled down (in the case of the autoclave reactor) to room temperature or can be cooled down for 10 minutes by using tap water. In the HTL Reactor, the temperature and reaction time range differs, depending upon the varying categories of plastic.

Characterizations of Various Plastic types and Co-Processing Feedstocks

Proximate analysis can be defined as a technique to compute the chemical properties of composite plastic compounds based on four particular elements which are moisture content, volatile matter, fixed carbon, and ash content. Volatile matter and ash content are the crucial elements that exert influence upon the liquid oil yield in the pyrolysis and HTL process. High volatile matter heightens the liquid oil production while high ash content lightens the amount of liquid oil, consequently increments the gaseous yield and char formation (Abnisa F et al.,2014). On the other hand, the ultimate analysis establishes the mass fractions of carbon, hydrogen, nitrogen, and heteroatoms of a sample. The elemental composition of plastics can be determined according to ASTM D-5291 by using the Elemental Analyzer and the oxygen content can be estimated by difference (Sharuddin et al.,2017) in the pyrolysis process. Proximate and ultimate analysis data can be acquired according to ASTM D3172 standards in the HTL process.

Table 1 Characterizations and conversion techniques of Plastic and Co-Processing Feedstocks

Component s	PET	HDPE	PVC	LDPE	PP	PC	Dunaliella tertiolecta and PP	
Conversion process	Pyrolysis	Pyrolysis	Pyrolysis	Pyrolysis	Pyrolysis	HTL	Co-HTL	
Proximate Composition (wt., %)								
Volatile matter	86.75	99.77	85.77	99.85	99.64	81.51	64.7	100
Ash content	6.83	0.22	9.11	0.12	0.36	0.39	16.5	0
Fixed carbon	6.2	0	5.1	0	0	18.1	17.2	0
Elemental composition (dry basis, wt., %)								
C	63.94	86.99	37.24	85.6	86.88	77.87	43.31	84.55
H	4.52	12.12	4.99	13.4	12.5	5.98	5.96	13.58

N	0.05	0.33	57.77	0.26	0.3	1.96	20.83	N/A
O	31.49	0.56	0	0.74	0.32	14.19	29.9	N/A
Elemental molar ratio								
H/C	0.85	1.67	1.61	1.88	1.73	1.02	1.65	1.93
O/C	0.37	0.005	0	0.007	0.003	0.14	0.52	N/A
H/C _{eff}	0.11	1.66	1.61	1.87	1.72	0.65	0.62	N/A
HHV (MJ/kg)	22.5	46.6	19.7	47.9	47.1	32.3	17.8	45.5
References	(Sharuddin et al.,2017)	(Sharuddin et al.,2017)	(Sharuddin et al.,2017)	(Sharuddin et al.,2017)	(Sharuddin et al.,2017)	(Jin et al.,2020)	(Wu et al.,2017)	

To anticipate the coke generation during different thermal conversion reaction, the H/C_{eff} ratio can be estimated by using the following equation (Chang et al.,2011).

$$H/C_{eff} = (H - 2O)/C \quad (1)$$

where H, O, and C are mole percentages of hydrogen, oxygen, and carbon which can be attained from elemental analysis.

Product Yield Computation

The following empirical equations can be used to obtain C, O, H, and higher heating values of the acquired crude oil yield of the plastic biomass samples based on elemental analysis (Parikh et al., 2007).

$$C = 0.635FC + 0.460VM - 0.095AC \quad (\%) \quad (2)$$

$$H = 0.059FC + 0.060VM + 0.010AC \quad (\%) \quad (3)$$

$$O = 0.340FC + 0.469VM - 0.023AC \quad (\%) \quad (4)$$

$$HHV_{Biomass} = 0.3536FC + 0.1559VM - 0.0078AC \quad (MJ/kg) \quad (5)$$

$$HHV_{Biocrude} = 0.3383C + 1.422(H - O/8) \quad (6)$$

$$HTL_{yield} \quad (\%) = 100 - (\text{residue mass/feedstock mass}) \times 100 \quad (7)$$

$$PYROLYSIS_{yield} \quad (\text{wt. } \%) = W_{Liquid}/(W_{Tot.}) \times 100\% \quad (8)$$

Operating Parameters During Thermal Conversion Reaction

Parameters play a prominent role in optimizing the product yield and composition during several thermal conversion reactions. There are diverse parameters that influence liquid oil generation in pyrolysis and HTL such as temperature, type of reactors, pressure, residence time, type and rate of fluidizing gas, and catalyst selection. But according to literature and previous journals, temperature and retention time were found to be the most influential parameters that affect crude yield. This article briefly discusses these two operating parameters here.

Temperature

Temperature is one of the most presiding operating parameters in thermal conversion techniques and it strongly influences biocrude yields and properties (Dimitriadis et al.,2017). According to the cited literature, the HTL temperature ranges between 200 °C and 450 °C and the Pyrolysis temperature ranges between 400°C to 800°C. In the thermal degradation of plastics, the temperature is one of the most notable operating parameters in pyrolysis since it regulates the cracking reaction of the polymer chain. Different plastics have diverse degradation temperature which is dependent upon the chemical structure. (Sharuddin et al., 2018).

Retention Time

Many researchers have studied the consequence of residence time on several thermal conversions, which is defined as the period during which the maximum temperature is maintained for certain thermal reactions not accounting for the heating and cooling periods. It has been observed that residence time influences the overall conversion of plastic biomass, product yields, and residue yields. Increasing residence time heightens the biocrude yields, however, beyond a certain threshold, further increment in residence time can have adverse results (Xu j et al.,2000). This residence time threshold is dependent upon the biomass feed, catalyst type, and operating conditions (Ye L et al.,2014).

RESULTS AND DISCUSSION

Biomass Characterization

The proximate and ultimate composition of different plastic biomass is presented in Table 1. From the above-presented data, it can be analyzed that the volatile matter content is the highest in LDPE (99.85% and is due to the pyrolysis process) and the lowest is in PC (81.51% and is also due to the pyrolysis process). High volatile matter always involves an increased amount of bio-oil production. But due to the higher amount of initial moisture content, this phenomenon can alter. The amount of fixed carbon ranges from 0 (LDPE, PP, and PS [all due to pyrolysis]) -18.1% (PC [due to HTL]). Among the biomasses, PVC has the highest ash content value of 9.11% (in the Pyrolysis process), and also another PVC has the lowest ash content value of 0% which can be found from the elemental analysis value of co-pyrolysis of microalgae EN and PVC. It is known that a lower ash content value is much more advantageous because it is requisite to acquire a lower ash content value from the yield as the higher the ash content value, the lower the biocrude yield. According to Zhang et al., biomass with an H/Ceff ratio <0.2 has a higher tendency to generate coke. LDPE contains the highest H/Ceff ratio of 1.87 (in the Pyrolysis process) but the H/Ceff ratio obtained from co-pyrolysis of EN and PVC is 0. So, only PET (0.11) contains the H/Ceff value closest to 0.2 which can be found from the pyrolysis process and this percentage implies the best feedstocks to generate higher yields. PS contains the highest percentage of carbon (91.57%) [in the Pyrolysis process] and PVC contains the lowest (37.24%) [in the Pyrolysis process]. Also, HDPE contains the highest percentage of hydrogen (14.2%) which can be found from the co-HTL of spirulina, and HDPE and PET contain the lowest (4.52%) [in the Pyrolysis process]. An elevated portion of carbon and hydrogen is requisite in the obtained yield due to incrementing percentage of carbon and hydrogen in the biomass influences higher production of biocrude yield. Only HDPE [in the Pyrolysis process], LDPE [in the Pyrolysis process], PP [in the Pyrolysis process], and PS [in CO-HTL of DT and PP] contain higher HHV values of 46.6, 47.9, 47.1, and 42 MJ/kg respectively. But all other biomasses have HHV values approximately in the range of 9-32 MJ/kg.

Energy Feasibility Assessment

The following table consists of the elemental composition of Bio-oil of different plastic biomass and different ratios of plastic biomass and additional feedstocks.

Table 2 Elemental Analysis of Biocrude of different Plastic Biomass generated due to several Thermal Conversion Techniques.

Feedstock Samples	Conversion Technique	Ultimate Composition of Oils (wt%)							References
		C(wt%)	H(wt%)	O(wt%)	H/C	O/C	H/Ceff	HHV (MJ/kg)	
Polycarbonate (PC)	HTL	75	8	17	1.28	0.17	0.94	33.73	(Pedersen et al.,2017)
Poly (methyl methacrylate) (PMMA)	HTL	63.6	9	27.4	1.7	0.32	1.05	29.44	(Pedersen et al.,2017)
Poly(oxymethylene) (POM)	HTL	78.7	7.7	13.6	1.17	0.13	0.91	35.16	(Pedersen et al.,2017)
Poly (phenylene oxide) (PPO)	HTL	85.7	8.2	6.1	1.15	0.053	1.04	39.57	(Pedersen et al.,2017)
Poly (vinyl alcohol) (PVA)	HTL	81.5	9	9.5	1.33	0.087	1.15	38.68	(Pedersen et al.,2017)
Styrenebutadiene (SB)	HTL	90.5	8.6	0.9	1.14	0.0075	1.13	42.69	(Pedersen et al.,2017)
Ng+PC	CO-HTL	75.2	6.9	17	1.1	0.17	0.76	32.63	(Li, N., et al.,2022)
Ng+PS	CO-HTL	78.6	8.3	8.7	1.27	0.083	1.1	36.61	(Li, N., et al.,2022)
50% PE and L. digitata Blend	CO-HTL	78.92	11	N/A	1.67	N/A	N/A	39.3	(Raikova et al.,2019)

25% PP and L. digitata Blend	CO-HTL	75	8.5	N/A	1.36	N/A	N/A	34.9	(Raikova et al.,2019)
25% NY and L. digitata Blend	CO-HTL	65.4	8.2	N/A	1.5	N/A	N/A	30.3	(Raikova et al.,2019)
Spirulina and HDPE (50:50 ratio)	CO-HTL	67.97	20.2	3.04	0.3	0.04	3.5	48.35	(Pei et al.,2011)
10% PE and Pistachio Hull Blend (same for 20%)	CO-HTL	71.2	7.2	12.7	1.21	0.13	0.946	38	(Hongthong et al.,2019)
10% PP and Pistachio Hull Blend (same for 20%)	CO-HTL	68.8	5.4	16.2	0.94	0.18	0.59	33.5	(Hongthong et al.,2019)
10% PET and Pistachio Hull Blend (same for 20%)	CO-HTL	70	4.8	19.4	0.82	0.21	0.41	34.7	(Hongthong et al.,2019)
10% NY and Pistachio Hull Blend (same for 20%)	CO-HTL	68.8	3.7	20	0.64 5	0.22	0.21	32.8	(Hongthong et al.,2019)
PP Fraction (746°C)	Pyrolysis	91.5	8.3	N/A	1.09	N/A	N/A	N/A	(Jung et al.,2009)
PE Fraction	Pyrolysis	87.2	11	N/A	1.51	N/A	N/A	N/A	(Jung et al.,2009)
50 PS: 50 Pine	Co-Pyrolysis	66.27	6.72	7.02	1.22	0.079	1.06	28.42	(Bhattacharya et al.,2008)
50 HDPE: 50 Pine	Co-Pyrolysis	74.38	9.11	6.33	1.47	0.064	1.34	27.68	(Bhattacharya et al.,2008)
50 PP: 50 Pine	Co-Pyrolysis	79.7	10.64	9.5	1.6	0.089	1.42	36.94	(Bhattacharya et al.,2008)

The elemental analysis of biocrude samples was inspected to signify the quality and energy density of produced bio-oil through the detection of heating value, atomic molar ratio, and energy analysis. From the elemental composition data of crude oil produced from plastic waste biomass (presented in table 2), it can be demonstrated that the Carbon content percentage range has excelled in the case of pyrolysis and co-pyrolysis (66.27-91.5%) than the HTL and Co-HTL (63.6-90.5%) whereas in the case of Hydrogen content percentage range, the reversal phenomenon has occurred and the HTL and Co-HTL process (3.7-20.2%) has stood out. The additional closer inspection of the above-tabulated values evaluates that the average carbon content percentage (79.81%) in the HTL and Co-HTL is higher than the average carbon content in the pyrolysis and co-pyrolysis process (74.68%). The lowest oxygen content percentage (0.9%) can be found in SB due to HTL. The lower the oxygen content, the more elevated will be the crude yield percentage. Average H/C is found to be highest in the case of pyrolysis (1.38) [Though the highest H/C ratio of 1.70 can be found to be in the case of PMMA, due to the HTL process]. Whereas in the case of average O/C, the reverse phenomenon has occurred and the HTL process reigns (0.14). As it is known that biomass with H/Ceff <0.2, has a higher tendency to generate coke, only the H/Ceff value obtained from the Co-HTL of 10% NY & pistachio hull blend and 10% PET and pistachio hull blend are closest to 0.2 (0.21 and 0.41 respectively). From the analysis, it is clear that the HHV values of the crude oil obtained from the Co-HTL of spirulina and HDPE 50:50 (48.35 Mj/kg), the HTL of SB (42.69 Mj/kg), the HTL of PPO (39.57 Mj/kg), the Co-HTL of 50% PE and L. digitata blend (39.3 Mj/kg) and the Co-HTL of 10% PE and pistachio hull blend (38 Mj/kg) are the closest to the HHV of the Petro crude specification (42-49 Mj/kg) and the HHV values acquired due to the HTL and Co-HTL are elevated than the HHV values obtained due to the pyrolysis and co-pyrolysis process. According to (Koley et al., 2018) various components of Petro-crude through elemental analysis comprised of C (83-87%), H (10-14%), O (0.05- 1.5%), HHV (42-49 MJ/kg), etc. The elemental composition components value range of crude oil samples mainly included C (48.5- 86.23%), H (6.28-12.37%), O (1.05- 42.7%),

HHV (17.8-45.22 MJ/kg) respectively. From the above-tabulated value, it can be detected that the crude oil produced from the HTL of Styrenebutadien provides the elemental composition values closest to the Petro-crude specification [C (90.5%), H (8.6%), O (0.9%), HHV (42.69 Mj/kg)]. The analysis of the above-presented elemental composition values certainly demonstrates that the HTL and Co-HTL process is far better than the pyrolysis and co-pyrolysis process to generate biocrude due to higher carbon and hydrogen content percentage and higher HHV values.

Analysis of Bio-Oil by Van Krevelan Diagram

Van Krevelen Diagram plotted against H/C and O/C ratio of biocrude to assess the reaction mechanism and the quality and characteristics of the crude oil. From the above-represented graph, it can be demonstrated that the molar atomic ratio H/C and O/C of the crude oil obtained due to different thermal conversion mechanisms ranges from (0.30-1.70) and (0.0075-0.32). Most of the plastic biomass meets the specification of Petro-crude for H/C atomic ratio (1.5-2.0) or close to this range (above 1.1) except the Co-HTL of Spirulina and HDPE (50:50 ratio), 10% PP, and pistachio hull blend, 10% PET and pistachio hull blend, 10% NY and pistachio hull blend and the pyrolysis of PP fraction. Coincidentally the Highest H/C ratio (1.7) and O/C (0.32) ratio both can be found due to the HTL of PMMA. As the Petro crude specification is determined to maintain the O/C ratio < 0.02, the crude oil generated from most of the plastic biomass according to the above-presented table can't sustain the Petro crude stipulations, except for SB (O/C ratio=0.0075). The crude oil generated from the HTL of PPO, PVA, the Co-HTL of Ng+PS, and the Co-pyrolysis of 50 PS:50 Pine, 50 HDPE:50 Pine, and 50 PP:50 Pine can produce O/C ratios of 0.053, 0.087, 0.083, 0.079, 0.064 and 0.089 and these values are quite close to the Petro crude specification (O/C ratio <0.02). It is well-renowned that Higher H/C ratios and lower O/C ratios present in the biocrude sample produced from different plastic biomass and additional feedstocks compared to digested samples also indicate the plastic biomass sample decarboxylation and deoxygenation during different thermal conversion reactions.

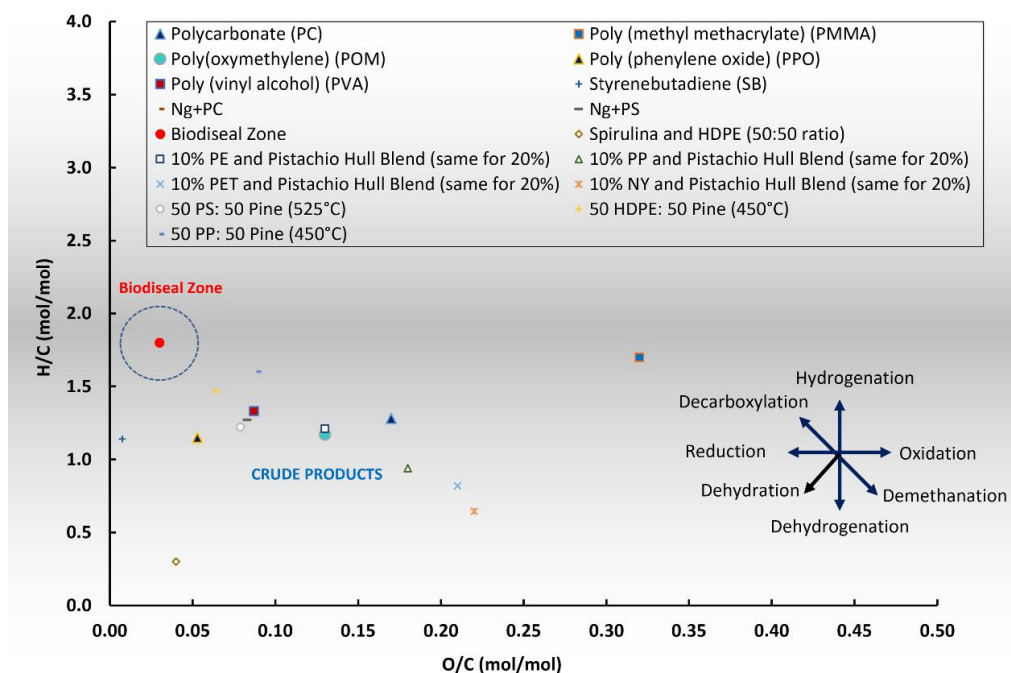


Figure 1: Reaction mechanisms of biomass to biocrude conversion using Van Krevelan diagram

Variations in Yield Percentage due to Different Operating Parameters

The above-illustrated graphs demarcate the influence of different operating parameters on the production of crude-oil percentage. From figure 2, it can be evaluated that the average bio-oil yield obtained from PVC (due to the pyrolysis process) and PP (due to the HTL process), at a temperature of about 500-520°C and 350-450°C are the lowest (12.55% and 16.125%). Coincidentally the average bio-oil yield is the highest in PS [(93.25% in the pyrolysis process, in the presence of 400-581°C) and (58% in the HTL process, in the presence of 350-450°C)] in both the HTL and Pyrolysis process. The inspection of figure 2 concludes that the HTL of PS at 350°C, the pyrolysis of HDPE, LDPE, PP, and PS at 450°C, 500°C, 450°C, and 425°C produce the maximum yield of 86%, 91.2%, 95%, 92.3%, and 97%.

Though the pyrolysis process prevails here in terms of temperature, this process requires different types of reactor beds and diversity in temperature to produce the elevated yield percentage, which seems to

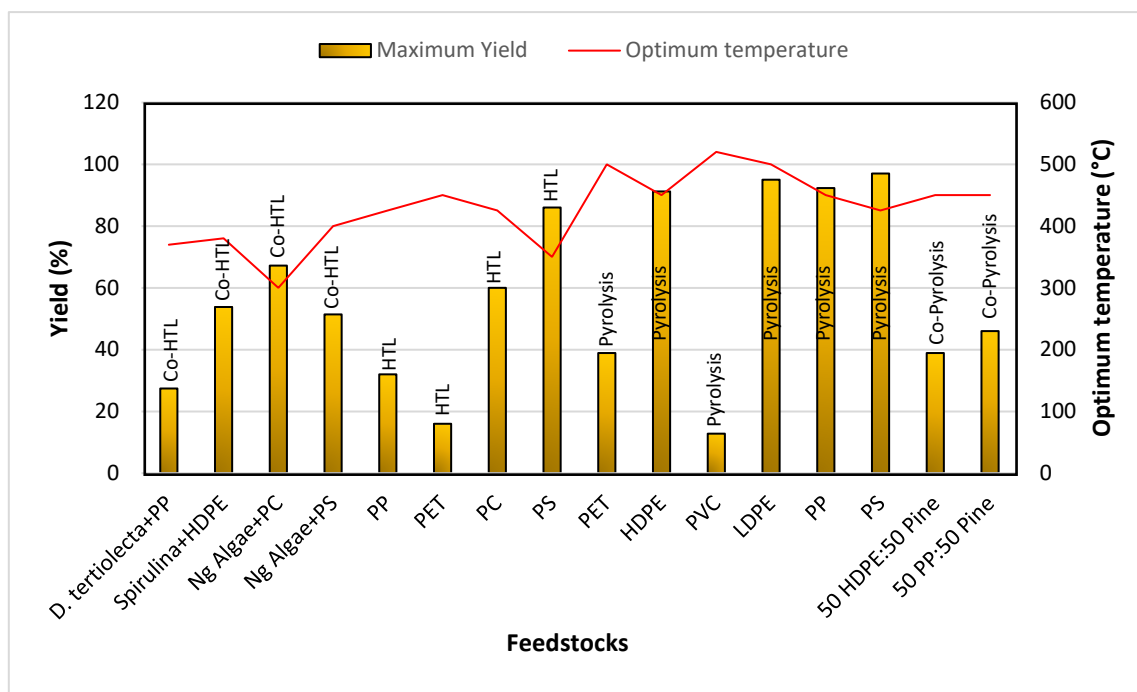


Figure 2: Effect of Temperature on the Maximum Yield (wt%). references: (Yao et al., 2017; Pei et al., 2011; Guo et al.,2021; Subramanya et al.,2020; Aragaw et al.,2013; Arregi et al.,2013; Miranda et al.,1998; Williams et al., 2004; Onwudili et al.,2009; Bhattacharya et al.,2009)

be quite extravagant for the completion of the reaction process. Though the yield percentage produced during the HTL process is a little bit ancillary in terms of the yield produced in the pyrolysis process, the HTL process requires a pretty much lower temperature than the pyrolysis process and, there is almost no diversity in the category of bed reactor. So, with lesser expense, a generous amount of oil yield can be produced.

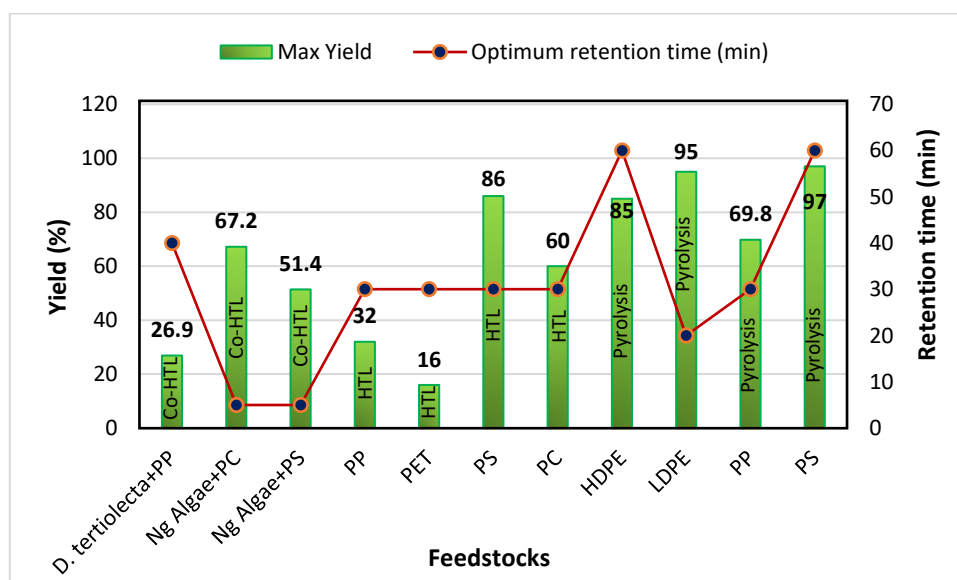


Figure 3: Effect of Retention Time on the Maximum Yield (wt%). References: (Wu et al.,2017; Subramanya et al.,2020; Das et al.,2018; Luo et al.,2000; Bagri et al.,2001; Williams et al., 2004)

Figure 3 depicts the impact of retention time on the maximum yield range. The average bio-oil yield obtained from PP (due to the pyrolysis process) and PET (due to the HTL process), at a residence time of about 390-460 min and 30-60 min are the lowest (6.56% and 10%). Coincidentally the average bio-oil yield is the highest in PS [(93.25% in the pyrolysis process, in 60-150 min retention time) and (58% in the HTL process, in 30-60 min retention time)] in both the HTL and Pyrolysis process. The proper assessment of figure 3 culminates that the HTL of PS at a retention time of about 30 min, the pyrolysis of HDPE, LDPE, and PS at a retention time of about 60 min, 20 min, and 60 min produce the maximum yield of 86%, 85%, 95%, and 97%.

Though the pyrolysis process outshines the HTL process in terms of retention time, a much closer look at the obtained yield percentage can be able to detect that the pyrolysis process requires a much longer retention time to produce the elevated yield percentage than the HTL process. Another phenomenon in the pyrolysis process is that the elevated percentage of yield can be seen in the case of common plastic biomasses like HDPE, LDPE, PS, etc. Whereas, the HTL process is well-known to generate oil yield from the plastic biomasses which are not known for producing enough oil yield except distinctive reaction condition is fabricated (PET and PC). Though the obtained yield percentage is not so satisfactory, the yield is produced in a lot lesser time than expected. Whereas, the pyrolysis process does not produce noteworthy yields in the case of these plastic biomasses let alone the necessity of lesser retention time to lessen the reaction expenditure.

Conclusion

Bio-fuels (Biodiesel and Bio-oil) are currently popular renewable sources of energy that exhibit heightening dominance over conventional fossil fuels. However, high production costs associated with raw feedstock are a significant constraint on this technology. In this study, an abundant proportion of plastic biomass (virgin raw plastic or modified in the form of powder or pellets) can be taken into account as an alternate biomass for biofuel production due to the longer carbon and hydrogen chains present in different plastic polymers. The higher proportion of C and H percentages generates a large amount of calorific value in the bio-oil yield.

This study emphasized plastic biomass as a sustainable solution to produce third-generation biofuel to vanquish the scarcity of the non-renewable Petro-crudes. From the overall analysis, it can be evaluated that the HTL of PE, PS, and the pyrolysis of PP and PS can produce satisfactory yield percentages of about 95%, 86%, 92.3%, and 97%. Though the overall analysis indicated that the Pyrolysis process is the better solution to produce an adequate proportion of biofuel, much closer attention can furnish disparate scenarios. The pyrolysis process generates adequate yield only in the case of common plastic biomasses and in the case of rarer type plastic biomasses like PET, PVC, PC, SB, and PPO; this process falls flat even in the presence of higher reaction temperature and longer retention time. The pyrolysis process also requires different types of the reactor bed to produce notable bio-oil yield which seems to excel the expense even more. Whereas, the HTL process can produce a much more adequate yield percentage even in the case of rarer plastic biomasses (For example, PC=60%, SB=80.8%, PPO=78.9%) in the presence of much lesser temperature and this process is less costly. Also, some current studies have demonstrated that a fast pyrolysis process can cause almost 50% more global warming potential impact compared to the hydrothermal liquefaction process, due to both high energy demand in the drying process and the elevated temperature functioning of fast pyrolysis in the alteration of waste biomass product. The appraisal of other environmental effects also indicated that the hydrothermal liquefaction operation is more environmentally congenial compared to fast pyrolysis due to the truncating energy consumption. And a lot of research has already been conducted regarding the high yield percentage in the pyrolysis process. If more and more research studies can be conducted, (For example, considering diversity in the different operating parameters; also the diversity in the reactor bed; also the addition of several catalysts and solvents to enhance the bio-oil yield)the yield proportion close to the Petro-crude specification can be expected from the HTL of different types of plastic biomass as this process seems to be the more promising solution (considering all the potentialities and obstacles of both the thermal conversion techniques) to produce adequate renewable energy. This review article suggests, further optimization of different influential parameters requires to be delineated properly for higher biocrude yield. Acquired crude oil may require further upgradation for commercial production. Due to this reason, researchers should require to give much closer attention to crude upgradation. The prime intention is to produce an adequate proportion of biofuel but also to perpetuate the generation procedure beneficial and profitable. Finally, these thermal conversion techniques not only safeguard the environment but are also expected to make enough capital from the Waste to Energy (WtE) industry in the long run.

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