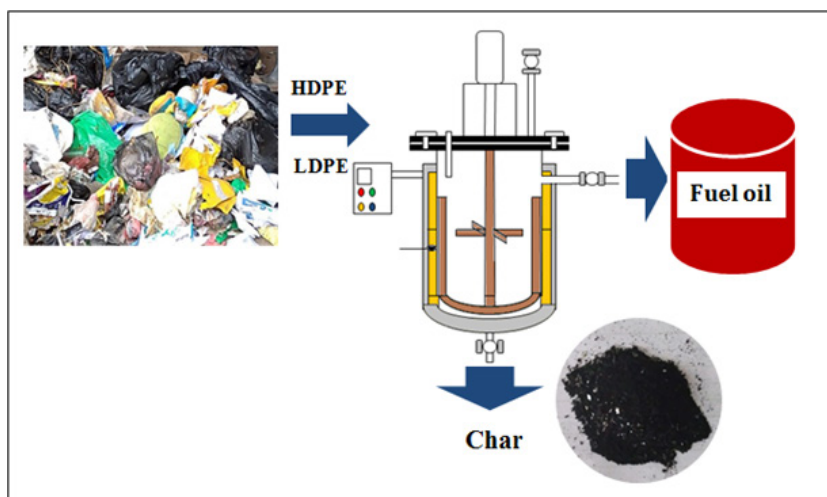


REVIEW ARTICLE

Pyrolysis as a value added method for plastic waste management: A review on converting LDPE and HDPE waste into fuel

P. G. I. Uthpalani, J. K. Premachandra, D. S. M. De Silva* and V. P. A. Weerasinghe

**Highlights**

- Systemic reviews on pyrolysis processes developed globally for HDPE and LDPE waste are lacking.
- This review emphasizes the effectiveness of catalysts and reactor types employed.
- Provides a platform to discover more efficient pyrolysis technologies.
- Emphasizes pyrolysis as an improved solution for plastic waste management.

Pyrolysis as a value added method for plastic waste management: A review on converting LDPE and HDPE waste into fuel

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Abstract: The global demand for plastic is increasing year by year due to its indispensable uses and excellent properties. Plastic wastes persist for many years due to their slow deterioration and cause severe environmental problems. Therefore, there is a growing focus worldwide on plastic waste disposal methods to overcome adverse environmental impacts. As plastics are petroleum-based materials, the pyrolysis of plastics to fuel oil, gases, and char, has more concern than the other plastic waste management methods of recycling and landfilling. A yield of 70-80 wt.% of liquid fuel from pyrolysis waste has been reported elsewhere, emerging the importance and aptness of this method in plastic waste management. The common reactor types for the pyrolysis process are batch reactor, semi-batch reactor, spouted bed reactor, and fluidized bed reactor. The common catalysts employed in plastic pyrolysis were zeolites, including ZSM-5, HUSY, Zeolite X, and Y. The pore structure and the catalyst's acidity are the most influencing parameters in increasing the liquid yield and the quality of the oil produced in the pyrolysis process. This paper reviews the existing literature on pyrolysis processes developed for HDPE and LDPE wastes globally and their governing factors. Furthermore, emissions in the pyrolysis process and engine combustion of the fuel oil, performance, and emission characteristics were discussed. Although plastic waste separation prior to its management is a challenging process, this review highlights the conversion of waste plastic into energy as a smart way to meet the rising demands.

Keywords: Catalysts; fuel; plastics; pyrolysis; waste management

INTRODUCTION

Plastic plays a vital role in enhancing the human lifestyle in various sectors such as construction, automotive, healthcare, electronics, and packaging due to its excellent properties like lightweight, high strength, durability, non-corrosive and economic feasibility (Sharuddin *et al.*, 2016). Plastics can be classified into several groups based on their chemical structure, synthesis process, and properties. To assist in the recycling of waste plastics, the Society of Plastic Industry (SPI) has defined a resin identification code system. Therein, the plastics have been divided into seven groups based on the types of plastics used as raw materials for manufacturing articles (ASTM International, 2013), and those groups are given code numbers from 1 to 7 (Alabi *et al.*, 2019). The global production of plastic has been estimated to reach 335 million tons by 2020 (Lee *et*

al., 2020). As plastic consumption is increasing worldwide, plastic waste has become a significant component in municipal solid waste.

Adverse effects of mismanagement of plastic waste

Mismanagement of plastic waste adversely affects the natural environment. The most common plastic waste disposal methods practiced in developing countries are open dumping on empty lands, collecting for recycling, and burning in an open fire (Maheshi *et al.*, 2015). When plastic waste is discarded into open dumps, mainly the lightweight materials can spread over the open dumping sites into other lands resulting in an unpleasant environment in the cities. Animals near the open dump areas, especially wild animals, happen to eat those plastic waste with food waste and are susceptible to painful death. On the other hand, plastic waste clogging drainage systems in urban areas causes flooding even in light precipitates. Also, the hollow plastic articles act as water containers, and after precipitation, they create breeding sites for mosquitos and spread epidemic diseases such as Dengue in the tropical region.

Furthermore, when plastic waste is dumped in open lands or landfills, the hazardous chemicals embedded may leach out into soil, contaminating ground and surface water. The leachates may contain toxic chemicals, including Bisphenol A (BPA), phthalates, and chlorinated organic compounds released during the degradation of plastic materials (Alabi *et al.*, 2019; Asakura *et al.*, 2004). In addition to the degradation, the toxic chemicals in additives used to enhance the properties of the plastics, such as alkylphenol additives and phthalate plasticizers, heavy metals in pigments (e.g., Pb, Zn, Cu, Co, Cr, and Cd) can migrate to the soil after disposal of plastic waste in open dumps and landfills (Rafey & Siddiqui, 2021; Campanale *et al.*, 2020; Teuten *et al.*, 2009). The migration of the additives in the plastic mainly depends on the degree of the crystallinity of the plastic (Hansen *et al.*, 2013) and the interaction of the additives with the polymer (Bejgarn *et al.*, 2015). Moreover, the open burning of plastic waste can emit hazardous pollutants such as dioxins, polychlorinated biphenyls (PCBs), brominated compounds, furans, and

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heavy metals (i.e., Cu, Cr, Co, Pb, and Hg), causing severe damage to the respiratory system in both humans and animals (Alabi *et al.*, 2019; Verma *et al.*, 2016; Alam *et al.*, 2019; Filella & Turner, 2018). Further, these heavy metals and chemical compounds may destroy the helpful bacteria in the soil leading to soil infertility.

Plastic waste put down in open dumps, rivers, and waterways harms the marine organisms and animals habitats and eventually ends up in the ocean (Alabi *et al.*, 2019). Scientists have estimated that the weight of plastic waste in the ocean would be increased more than the weight of the live fish in the ocean by 2050 (Kaplan, 2016). More than 260 species of marine organisms were found to be ingested or entangled in plastic debris, ending their lives in fatalities (Alabi *et al.*, 2019; Gregory *et al.*, 2013; Purba *et al.*, 2019). This plastic waste in the ocean can be categorized into different sizes such as macroplastics (> 200 mm), mesoplastics (5-200 mm), microplastics (1 μm -5 mm), and nanoplastics (< 1 μm) (Worm *et al.*, 2017). Microplastic has been identified as the primary pollutant of these four types. Microplastics may be created in the production process or may be formed after plastics' degradation (Alomar *et al.*, 2016). Microplastics may infiltrate through living tissues in the food chain and can cause severe health problems (Alabi *et al.*, 2019; Jambeck *et al.*, 2015).

Municipal plastic waste management strategies in developing countries

Due to enormous problems created by plastic waste, many countries worldwide are struggling to find solutions for its management. Developing countries that have not implemented a feasible plastic waste management system are facing critical environmental and social problems. The studies carried out in India (Rafey & Siddiqui, 2021), Bangladesh (Masud *et al.*, 2019), Malaysia (Chen *et al.*, 2021), Vietnam (Salhofer *et al.*, 2021), and Thailand (Wichai-utcha & Chavalparit, 2019) have reported the efforts taken to address the issues of the rapid accumulation

and mismanagement of plastic waste in their countries. Therein, the reasons identified for the improper plastic waste management in developing countries were the lack of capital investment and infrastructure, migration of population to urban areas, dearth of awareness in the society, lack of adequate technical instruments, dearth of strict restrictions on plastic waste disposal, low rate of recycling, lack of separation of household plastic waste, and practice of improper disposal methods (Figure 1) (Rafey & Siddiqui, 2021; Purba *et al.*, 2019; Hossain *et al.*, 2020; Evode *et al.*, 2021; Padgelwar *et al.*, 2021). As a positive approach to plastic waste management, China has banned imports of plastic waste from western countries (Brooks *et al.*, 2018; Vollmer *et al.*, 2020; Marks, 2019). This action has intensified the impetus of developed countries with sufficient infrastructure to reconsider plastic usage and implement recycling programs without sending their plastic waste to other countries. Consequently, many developed countries such as the United Kingdom, Canada, United States, Japan, Ireland, and Taiwan have introduced bans on single-use plastic bags and bottles or collected a tax from customers or retailers to promote environmentally friendly alternatives for single-use plastics (Table 1) (Wichai-utcha & Chavalparit, 2019; Palugaswewa, 2018). National action plans have been launched in Indonesia to attempt a 70% reduction in marine plastic waste by 2025 (Purba *et al.*, 2019). Moreover, manufacturers are forced by the rules and restrictions to make plastic products that can be recycled (Wichai-utcha & Chavalparit, 2019). However, economically developing countries are challenged by plastic waste management issues due to the lack of sustainable methods of reducing or recovering plastic waste. The fish-bone diagram (Figure 1) shows the above discussed common causes affecting the overall improper plastic waste management in developing countries, mainly under management, method, material, machine, community, and nature.

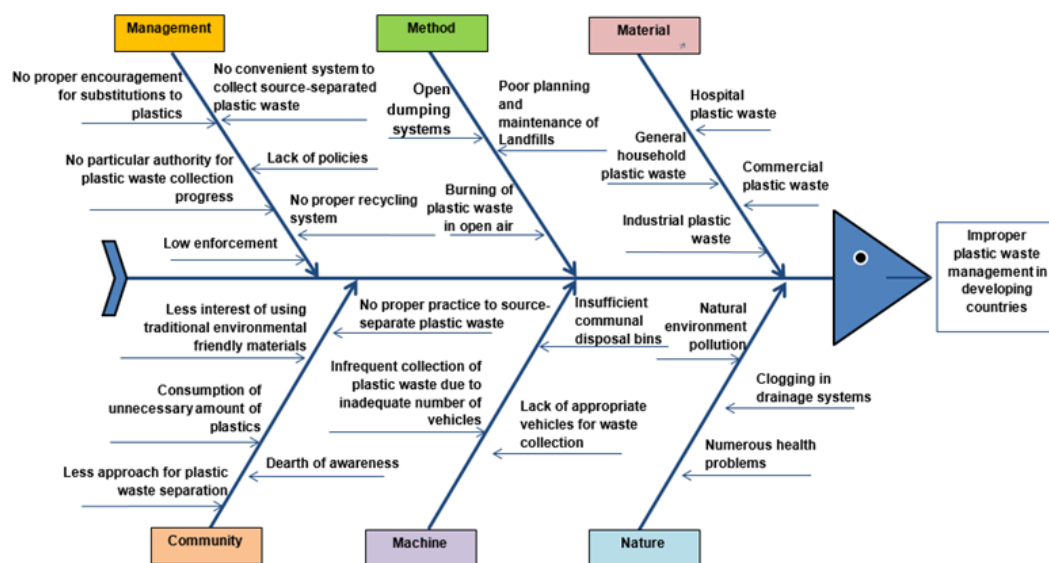


Figure 1: Fish-bone diagram of improper waste management in developing countries.

which has been implemented in different modes such as sink-float separation (Bauer *et al.*, 2018), froth flotation (Wang *et al.*, 2015), hydrocycloning (Serranti & Bonifazi, 2019). The separation of filler-containing and hollow waste plastic products and composites is difficult using wet separation techniques (Vollmer *et al.*, 2020). Also, additional energy is required for the drying process prior to the extrusion after the wet separation is another drawback (Arachchige *et al.*, 2019). The 'density separation' is not applicable for many plastics due almost nearer densities of different types ($\rho_{HDPE} = 0.941$, $\rho_{LDPE} = 0.915-0.925$, $\rho_{LLDPE} = 0.91-0.94$, $\rho_{PP} = 0.90-0.94$, $\rho_{PET} = 1.35-1.40$, $\rho_{PVC} = 1.34-1.43$ g/cc) (Al-Salem *et al.*, 2009; Schyns & Shaver, 2021). These shortcomings can be minimized during the incineration process, as even composite plastic waste can be used in the incineration process, as no detailed source separation is required.

In the incineration process, plastic waste is burned at high temperatures (> 1000 °C) (Dodbiba & Fujita, 2004) in the oxygen environment and the released energy can be recovered as heat and transformed into steam and electricity (Gradus *et al.*, 2017). However, the associated cost related to the investment, maintenance, and reducing environmental impacts (CO_2 emission, release of dioxins, other polychlorinated biphenyls, and furans) is high in the incineration process (Gradus *et al.*, 2017; Hopewell *et al.*, 2009). Process handling in incineration is more complicated than the pyrolysis process.

Among the recycling methods available, pyrolysis is the most effective and sustainable method for plastic waste management because of its viability of converting plastics to fuel oil (gasoline, kerosene, diesel, furnace oil), char, and gases. These end products can be used as value-added products (Verma *et al.*, 2016; Budsareechai *et al.*, 2019). In the pyrolysis process, long-chain hydrocarbons are degraded into small chain hydrocarbons or less complex molecules upon heating in an oxygen-free environment (Ragaert *et al.*, 2017; Sharuddin *et al.*, 2018; Panda *et al.*, 2010). Many research articles claim that the pyrolysis of plastics produces a high amount of fuel oil (up to 80 wt.%) at moderate temperatures around 500 °C (Sharuddin *et al.*,

2018; Sharuddin *et al.*, 2016; Wróblewska & Rydzkowski, 2020; Eze *et al.*, 2021).

There were many research papers published on the subject of the pyrolysis of plastic waste. Therein, to maximize the oil production from plastics, various types of pyrolysis processes were developed and the yield and quality of the product of each were found to be dependent on the set-up parameters. Therefore, considering the effectiveness of the pyrolysis process and its adaptability to the local context, this review discusses the pyrolysis of High-density polyethylene (HDPE) and Low-density polyethylene (LDPE), the most abundant waste plastics in the environment. Further, the effects of process control parameters such as applied temperature & pressure, type of reactor, residence time, types of catalysts, and the type of fluidizing gas and its flow rate on the pyrolysis process are discussed. Herein, a special emphasis is made on the effect of the different catalysts, silica-alumina catalysts, zeolite catalysts, fluid catalytic cracking (FCC) catalyst, and others used in pyrolysis. Additionally, some pertinent discussion on the type of emissions evolved in pyrolysis, engine combustions of the resultant fuel oil and emission characteristics were also presented.

The HDPE and LDPE are thermoplastic materials of the polyolefin family, which are of petrochemical origin. Polyethylene is the most common and well-known plastic material used to manufacture many products. The properties of HDPE and LDPE are tabulated in Table 2 (Sam *et al.*, 2014; Najafi, 2013; Mendes *et al.*, 2011; Kwon *et al.*, 2002). HDPE is a linear polymer with a high degree of crystallinity. It is widely used to manufacture containers/ bottles for detergent, milk, oil, shampoo, conditioner, and bleach (Adrados *et al.*, 2012). LDPE has a low degree of crystallinity due to its branched structure. The branches make it more flexible than HDPE (Salih *et al.*, 2013). Hence, LDPE can be applied for a wide range of available products in the packaging industry such as plastic bags, wrapping foils for packaging, trash bags, *etc.* (Sharuddin *et al.*, 2016). Therefore, the amount of waste LDPE present in municipal solid waste is very high compared to that of other plastics.

Table 2: Properties of LDPE and HDPE.

Property	LDPE	HDPE
Chemical structure	More branching	Less branching, more linear
Density	0.91-0.94 g/cm ³	0.95-0.97 g/cm ³
Flexibility	More flexible due to low degree of crystallinity (50-60%)	More tough and rigid due to high degree of crystallinity (>90%)
Melting point	101-115 °C	135-145 °C
Chemical resistance	Resistant to most alcohols, acids, and alkalis; low resistance to oxidizing agents and selected hydrocarbons	Superior resistance to solvents, alcohols, acids, and alkalis; low resistance to most hydrocarbons
Strength	Relatively increased impact strength in cold conditions	High tensile and specific strength
Transparency	High, due to amorphous condition	Low, due to increased level of crystallinity
Tensile strength at 20 °C	6–17 MPa	14–32 MPa

Pyrolysis of HDPE

Much research on the pyrolysis of HDPE have been conducted during the last decade (Budsaereechai *et al.*, 2019; Adeniyi *et al.*, 2019; Shukla *et al.*, 2016). Budsaereechai *et al.* (2019) investigated the pyrolysis of HDPE using a bench-scale fixed-bed batch reactor. The pyrolysis was performed at 500 °C under an optimum heating rate of 10 °C/min in the presence of a nitrogen medium. Moreover, highly mesoporous bentonite clay catalysts with BET surface area of 47 m²/g were used in this research. The results revealed that the highest total conversion occurred, yielding 88.9 wt.% fuel oil at 3:20 of catalyst to polymer ratio.

Adeniyi *et al.* (2019) have investigated thermal cracking pyrolysis of HDPE in a simple batch reactor at 425 °C under the heating rate of 7 °C/min which resulted in a high liquid yield (51.84%), a high char yield (45.33%), and a low gaseous yield (2.83%). It also reported that the possibility of cracking the resulted char into the liquid fuel with further heating above 550 °C. Although the designing and process controlling of a simple batch reactor are easier than those of a fixed-bed batch reactor, the simple batch reactor could not be used for high-scale conversions. This is due to the high operation cost associated with batch-wise operations and the tendency of high coke formation in the batch process (Sharuddin *et al.*, 2016).

Al-Salem (2019) has studied the thermal pyrolysis of HDPE in a novel fixed bed reactor system to produce gasoline range hydrocarbons. The pyrolysis trials were conducted at different temperatures between 500 °C to 800 °C while purging (20 ml/min) nitrogen carrier gas. Therein, the optimum liquid yield obtained was 70 wt.% at 550 °C temperature. The novelty of this fixed bed reactor was the presence of two gas/liquid separators (GLS) used in the system. This pyrolysis system consisted of a collection hopper with a nut and bolt at the bottom of the reactor for collecting ash/char. The resulting liquid yield of this study is moderately higher than that of the typical pyrolysis systems which employ fixed bed reactors (Adeniyi *et al.*, 2019; Marcilla *et al.*, 2009b; Patil *et al.*, 2018; Kumar *et al.*, 2013). This high liquid yield was credited to the design of

the reactor, which has three heating elements that maintain uniform temperature along with the profile of the reactor.

Further, Elordi *et al.* (2012) studied the pyrolysis of HDPE in a conical spouted bed reactor (Figure 3) at the temperature of 500 °C using two types of HZSM-5 zeolite catalysts. One of the catalysts is composed of SiO₂/Al₂O₃ with a ratio of 80 and the other has been composed of SiO₂/Al₂O₃ with a ratio of 30. When the catalyst with SiO₂/Al₂O₃ ratio of 80 was used, the system produced a 59.8 wt.% yield of olefin (C₂-C₄), a 25.4 wt.% yield of non-aromatic C₅-C₁₁ fraction, a 5.6 wt.% yield of light alkane fraction, and a 6.7 wt.% yield of monoaromatics. At the same time, the system has produced a 57.0 wt.% yield of olefin (C₂-C₄), a 15.5 wt.% yield of non-aromatic C₅-C₁₁ fraction, a 14.8 wt.% yield of light alkane fraction, and a 10.9 wt.% yield of monoaromatics with the catalyst that SiO₂/Al₂O₃ ratio of 30. The coke formation on the catalyst was found to be low when the SiO₂/Al₂O₃ ratio of the catalyst was high. It has been demonstrated that conical spouted bed reactor has special properties for avoiding the agglomeration of particles when they collide, as it facilitates the cyclic movement to the catalytic particles. Generally, the feed particle size of the a conical spouted bed reactor is 1-4 mm (Sriifa *et al.*, 2019). The main difference between this reactor and the fluidized bed reactor is that the conical spouted bed reactor allows for a continuous operation with higher plastic flows into the reactor unit offering a higher versatility. The schematic diagram of the conical spouted bed reactor is shown in Figure 3. The main drawbacks of the conical spouted bed reactor are the difficulties in catalyst feeding and product collection (solid and liquid).

According to the literature, most of the laboratory scale plastic pyrolysis were carried out in batch reactors, semi-batch reactors, or continuous flow reactors such as fixed bed, fluidized bed, and conical spouted bed reactors. There are advantages and disadvantages in using each type of reactor in the plastic pyrolysis process. Table 3 summarizes a comparison between the uses of the batch reactor and the fluidized bed reactor in pyrolysis (Figure 3) (Sharuddin *et al.*, 2016; Kaminsky, 2021; Scheirs & Kaminsky, 2006).

Table 3: Advantages and limitations of the batch reactor and fluidized-bed reactor in the plastic pyrolysis process.

Batch Reactor	Fluidized-bed reactor
Parameters can be easily controlled in the thermal pyrolysis process.	Difficult to control the parameters in the thermal pyrolysis process.
Offer a high liquid yield in the thermal pyrolysis process.	Offer a high gaseous yield in the thermal pyrolysis process.
Not suitable for the catalytic pyrolysis process due to the high tendency of coke formation on the catalyst surface.	Best reactor for the catalytic pyrolysis process due to the ability of reuse the catalyst many times without the forming of coke on the catalyst surface.
Operational cost is high for large-scale production due to frequent repeated recharging and restarting.	Operational cost is low due to low repeated feedstock recharging and no need of frequent restarting.
Suitable for lab-scale experiments.	Suitable for pilot scale operations.

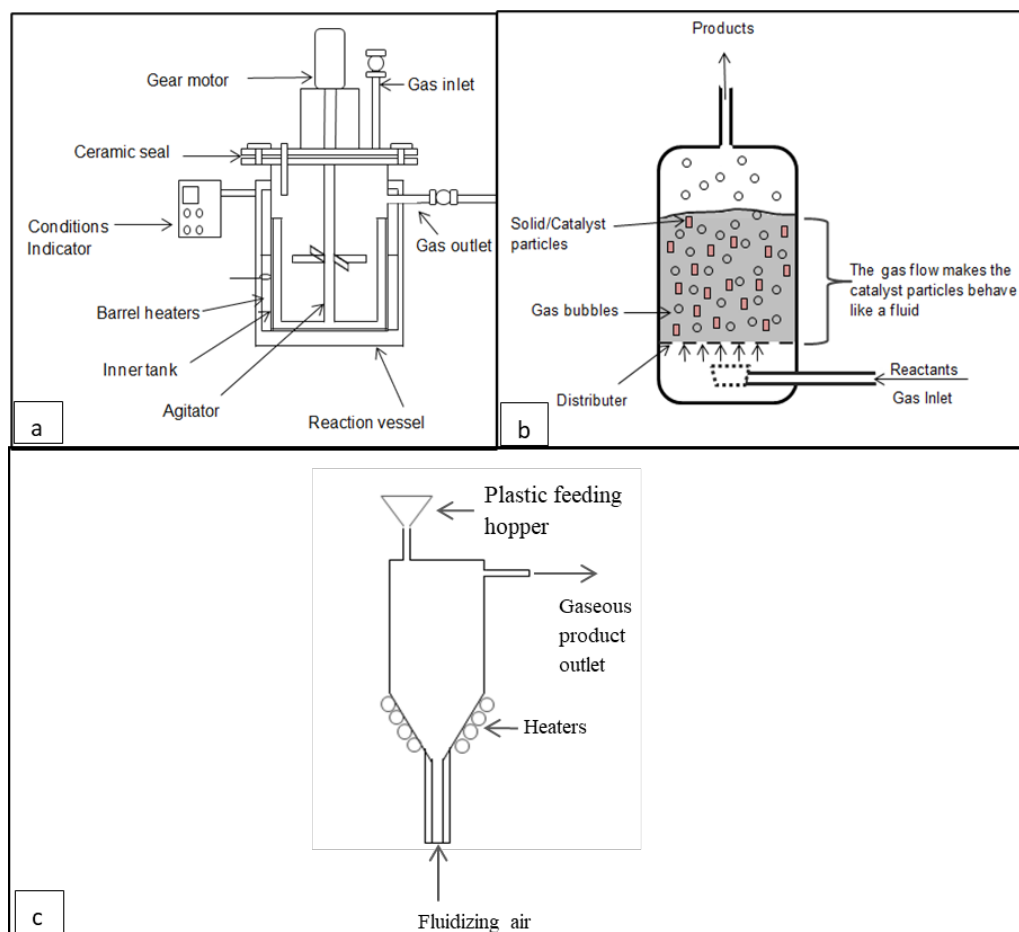


Figure 3: A schematic diagram of (a) batch reactor with the agitator, (b) fluidized-bed reactor, and (c) conical spouted bed reactor.

Pyrolysis of LDPE

Wan and coworkers (2020) have studied the catalytic pyrolysis of LDPE at 500 °C using biomass derived active carbon as the catalyst and obtained jet-fuel range alkanes (C_8 - C_{16}) and aromatics ($<C_{16}$). Herein, 75.3 wt.% of liquid yield, 23.4 wt.% of gaseous yield, and 1.3 wt.% of char yield have been obtained. This study further revealed that the activated carbon with high acidity and high catalytic temperature parameters are beneficial to the generation of aromatics and in contrast, activated carbon with weak acidity and low catalytic temperature parameters is favorable for the generation of alkanes.

Fan *et al.* (2017) and Zhang *et al.* (2015) have studied the pyrolysis of LDPE using a microwave-assisted system. In this system, LDPE waste has been thermally cracked at 500 °C of temperature using SiC as the microwave absorbent to enhance the heating of LDPE powder. The resulting vapor has been sent through an ex-situ catalytic system composed of a quartz column. This column was filled with the catalyst sandwiched between a layer of quartz wool and a polyporous (pore size was 90–150 mm) fritted disc. The quartz wool and fritted disc help secure the catalyst powder and facilitate the uniform passage of the resultant vapors through the column. A heating tape was used to heat the catalytic bed and the temperature of the reaction was measured using a K-type thermocouple. By using microwave-assisted thermal cracking without a catalyst, Fan and coworkers and Zhang and coworkers

obtained 38.5 wt.% of liquid yield and 32.58 wt.% of a liquid yield, respectively (Fan *et al.*, 2017; Zhang *et al.*, 2015). In microwave-assisted thermal cracking followed by catalytic cracking, Fan and coworkers used MgO as the catalyst whereas, Zhang and coworkers have used ZSM-5 catalyst. In that effort, both research groups have upgraded the liquid yield up to around 45 wt.%.

Bagri & Williams (2002) investigated polyethylene pyrolysis in a fixed-bed reactor at 500 °C with a heating rate of 10 °C/min. The pyrolysis was carried out for 20 min using nitrogen as fluidizing gas. Without employing the catalyst, this has resulted in a high liquid yield of 95 wt.% with low gaseous yield and negligible char formation in the thermal pyrolysis process. When Y-zeolite was used as the catalyst, this liquid yield reduced to 85 wt.% and the gaseous yield increased to 10 wt.%.

In another study of LDPE pyrolysis, Marcilla *et al.* (2009a) observed that the thermal degradation of LDPE occurred between the temperature of 469–494 °C, whereas in the HDPE, thermal degradation occurred between the temperature of 490–510 °C. Furthermore, Onwudili *et al.* (2009) have observed that the LDPE thermally decomposes into oil at the temperature of 425 °C. A brown waxy material formed at a temperature below 410 °C, indicating the incomplete combustion of the material. They concluded that the optimum temperature to obtain the highest liquid yield from LDPE was 425 °C.

Comparison between the pyrolysis processes of HDPE and LDPE

The chemical composition of the HDPE and LDPE plastic obtained through proximate analysis studies are shown in Table 4 (Sharuddin *et al.*, 2016; Abnisa *et al.*, 2014). According to the proximate analysis, the volatile matter of the plastic is high while the ash and the moisture content are low.

This shows that HDPE and LDPE have higher potential to transform into liquid oil and gaseous fuel products in the plastic pyrolysis process than the biomaterials which contain high moisture content in the composition.

The most effective temperature range to optimize the liquid oil yield would be 500-550 °C for the thermal pyrolysis process of HDPE and LDPE as shown in Table 5. However, in the presence of catalysts, the optimum temperature for this pyrolysis could be lowered to 450 °C with a further increase in the liquid yield.

In thermal pyrolysis, LDPE can offer a high liquid yield of around 93.1 wt.%, whereas HDPE can offer around 84.7 wt.%. However, with the addition of catalysts such as Fluid Catalytic Cracking (FCC) catalyst at the proper operating temperature, the liquid yield of the HDPE could be further maximized to above 90 wt.%.

Table 4: Proximate analysis of HDPE and LDPE plastics in various studies.

Type of plastic	Moisture content wt.%	Fixed carbon wt.%	Volatile matter wt.%	Ash content wt.%	References
HDPE	0.00	0.01	99.81	0.18	Vijayakumar & Sebastian (2018); Aboulkas <i>et al.</i> (2010)
	0.00	0.03	98.57	1.40	Heikkinen <i>et al.</i> (2004)
LDPE	0.30	0.00	99.70	0.00	Aboulkas <i>et al.</i> (2010);
	-	-	99.60	0.40	Sharuddin <i>et al.</i> (2016)

Table 5: Summary of the process parameters of HDPE and LDPE in the pyrolysis studies (Vijayakumar & Sebastian, 2018).

Type of plastic	Reactor	Process parameters				Yield			Other details	References
		Temperature (°C)	Pressure (atm)	Heating rate (°C/min)	Duration (min)	Liquid fraction Wt.%	Gaseous fraction Wt.%	Char Wt.%		
HDPE	Batch	350	-	20	30	80.88	17.24	1.88		Ahmad <i>et al.</i> (2015)
HDPE	Semi batch	400	1	7	-	82	16	2	Stirring rate 200 rpm FCC catalyst 10 wt.%	Sharuddin <i>et al.</i> (2016)
HDPE	Batch	450	-	-	60	74.5	5.8	19.7		Borsodi (2014)
HDPE	Semi batch	450	1	25	-	91.2	4.1	4.7	Stirring rate 50 rpm FCC catalyst 20 wt.%	Abbas-Abadi <i>et al.</i> (2013)
HDPE	Fluidized bed	500	-	-	60	85	10	5	Silica alumina catalysts	Luo <i>et al.</i> (2000)
HDPE	Batch	550	-	5	-	84.7	16.3	0		Marcilla <i>et al.</i> (2009b)
HDPE	Fluidized bed	650	-	-	20-25	68.5	31.5	0		Rezvanipour <i>et al.</i> (2014)
LDPE	Batch	425	-	10	60	89.5	10	0.5		Escola <i>et al.</i> (2011)
LDPE	Batch	430	-	3	-	75.6	8.2	7.5	Also yield wax = 8.7wt.%	Onwudili <i>et al.</i> (2009)
LDPE	-	500	1	6	-	80.41	19.43	0.16		Choi <i>et al.</i> (2010)
LDPE	Fixed bed	500	-	10	20	95	5	0		Fakhrhoseini & Dastanian (2013)
LDPE	Batch	550	-	5	-	93.1	14.6	0		Lee <i>et al.</i> (2003)
LDPE	Fluidized bed	600	1	-	-	51.0	24.2	0	Also yield wax = 24.8 wt.%	Williams & Williams (1999)

These fractions are defined based on the number of the carbon in the carbon chain such as Gaseous fraction; C_1 - C_5 , Liquid fraction; C_6 - C_{28} , Char fraction; $>C_{30}$ (Wang *et al.*, 2016).

Effect of catalysts on pyrolysis of HDPE and LDPE

Catalysts are widely used in the pyrolysis process of HDPE and LDPE to reduce the reaction temperature, improve the yield of volatile content and provide selectivity in the resulting hydrocarbon ranges (Bagri & Williams, 2002; Garcia *et al.*, 2005). It is very beneficial in selectively producing gasoline and diesel range fuel oil fractions from volatile products (Sharuddin *et al.*, 2016; Miandad *et al.*, 2019a). Catalysts reduce the activation energy of pyrolysis reactions and increase the speed of reactions. Thereby, catalysts are very useful in minimizing energy usage in the pyrolysis process (Elordi *et al.*, 2009).

Catalysts can be classified into two groups, homogeneous catalysts and heterogeneous catalysts. In the case of homogeneous catalysts, the catalyst takes the same single phase as the reaction components. Heterogeneous catalysts do not take the same phase as the reaction components (Sharuddin *et al.*, 2016; Miandad *et al.*, 2019a). According to the literature, heterogeneous catalysts are the most commonly used catalysts in the pyrolysis process because of the easiness of separation from the liquid product, low cost, and reusable property of the catalysts (Budsaerechai *et al.*, 2019). Heterogeneous catalysts can be classified into several types including nanocrystalline zeolites, conventional acid solids, mesostructured catalysts, metal-supported on carbon, and basic oxides (Sharuddin *et al.*, 2016; Elordi *et al.*, 2009).

Silica-alumina catalysts used in the pyrolysis of HDPE and LDPE

The silica-alumina catalysts are acid catalysts that contain amorphous structures with Bronsted acid sites and Lewis acid sites (Sharuddin *et al.*, 2016; Busca, 2020). It does not contain a stable crystalline structure compared to zeolite catalysts. Generally, the total pore volume of the amorphous silica-alumina catalysts is higher than those of crystalline zeolite catalysts (Klaimy *et al.*, 2020). This is because the silica-alumina catalyst has larger pore sizes whereas the crystalline zeolite catalyst has micro pore sizes (Pourjafar *et al.*, 2018; Ishihara *et al.*, 2010).

Klaimy *et al.* (2020) have investigated the effect of the acidity, pore size distribution, and specific surface area of silica-alumina catalysts on the cracking of LDPE in a pilot reactor at 450 °C of temperature. Four different catalysts,

silicate-1(Si-MFI), ZSM-5(Si/Al-MFI), amorphous SiO_2 and amorphous silica-alumina(Si/Al) have been used in this cracking process. Table 6 illustrates the textural properties and the acidity of these catalysts.

Table 6: Textural properties and acidity of SiO_2 , Si/Al, Si-MFI and Si/Al-MFI catalysts.

Catalyst	Acidity ^[a] mmol/g		
	Total	Weak	Strong
SiO_2	0.045	0.030	0.015
Si/Al	0.209	0.071 0.138	-
Si-MFI	0.017	-	0.017
Si/Al-MFI	0.639	0.193	0.445

[a] quantity of NH_3 desorbed (Klaimy *et al.*, 2020)

According to Table 6, the amorphous silica-based catalysts have weak acid sites and exhibit a low acidity resulted by the presence of silanol groups on their external structure. A higher total acidity in silica-alumina catalysts (Si/Al and Si/Al-MFI) than in silica catalysts due to the presence of aluminum in the catalytic structure which can produce Bronsted and Lewis acid sites. In addition, the crystalline silica-alumina catalyst (Si/Al-MFI) contains a higher density of both strong and weak acid sites.

When the silica-alumina catalysts (Si/Al and Si/Al-MFI) having high acidity were employed in pyrolysis, that have produced high gas and liquid yields without the formation of char residue. In contrast, the low acid activity catalysts (SiO_2 and Si-MFI) have produced low gas and liquid yields with wax and char residue (Klaimy *et al.*, 2020; Elordi *et al.*, 2009; Ma *et al.*, 2013). When the Si/Al-MFI was used, it has produced 65-64 wt.% and 35-36 wt.% yields of gas and liquid oil fraction, respectively. It has also been reported that the Si/Al-MFI zeolite catalyst increases the formation of aromatics and cycled hydrocarbons whereas the mesoporous silica-alumina catalyst increases the formation of olefins in the liquid fraction (Klaimy *et al.*, 2020).

In another study, pyrolysis of HDPE over mesoporous silica, silica-alumina (SA-1 and SA-2), and ZSM-5 zeolite catalysts have been investigated by Sakata *et al.* (Sakata *et al.*, 1997) in a semi-batch reactor at 430 °C. Properties of the catalysts and the yields of the resultant products from both thermal and catalytic pyrolysis processes are shown in Table 7.

Table 7: Properties of the catalysts and the yields of the resultant products in the pyrolysis process (Sakata *et al.*, 1997).

Property	Thermal	SA-1	SA-2	ZSM-5	Silica
Surface area (m^2/g)	-	420	270	360	900
SiO_2/Al_2O_3 ratio	-	4.99	0.267	75.9	-
Yield					
Liquid oil wt.%	63.3	67.8	74.3	49.8	71.1
Gas wt.%	9.6	23.7	13.4	44.3	11.0
Char wt.%	21.1	8.5	12.3	5.8	17.9

It has been reported that the acidity of these four catalysts is in the order of SA-1>ZSM-5>SA-2>>silica=0. According to Table 6, the ZSM-5 catalyst which contains both weak and strong acid sites has produced a higher amount of gaseous product (44.3 wt.%) when compared to the silica-alumina catalysts (SA-1, 23.7 wt.%; SA-2, 13.4 wt.%) that contain only weak acid sites. The moderately acidic SA-2 catalyst has produced a higher yield of liquid oil than those of SA-1 and ZSM-5 catalysts. A slightly higher coke formation has been observed in both silica-alumina catalysts than that of the ZSM-5 catalyst.

Zeolite catalysts used in the pyrolysis of HDPE and LDPE

Mainly Zeolite catalysts have been used in the plastic pyrolysis process due to their desirable properties in the selective recovery of gasoline and diesel range hydrocarbons as final products. They are aluminosilicate crystalline materials having a pore structure and ion exchange capabilities (Marcilla *et al.*, 2009b; Elordi *et al.*, 2012). The ratio of $\text{SiO}_2/\text{Al}_2\text{O}_3$ determines the zeolite type and its reactivity. The structure of the zeolite is formed by a three-dimensional framework where oxygen atoms link the tetrahedral sides with aluminum and silicon atoms (Sharuddin *et al.*, 2016; Nwankwor *et al.*, 2021; Verdoliva *et al.*, 2019).

Many zeolites have been synthetically manufactured by chemical processes to obtain a uniform chemical composition (high purity), uniform pore size, and better ion-exchange abilities (Xu *et al.*, 2010). These synthetic zeolites contain higher pore size and higher acidity than those of natural zeolites according to the purpose of commercial applications (Flanagan & Crangle, 2017).

The main types of synthetic zeolites are zeolites A, X, Y, ZSM-5, omega, and beta (Xu *et al.*, 2010; Kulprathipanja, 2010). HUSY zeolite has been modified from the zeolite Y to obtain an ultra-stable form of zeolites (Ma *et al.*, 2013; Xu *et al.*, 2010). The pore size and Si/Al ratio of various types of synthetic zeolites have been summarized in Tables 8 and 9.

Table 8: The pore diameter of various types of synthetic zeolites (Petrov & Michalev, 2012).

Type of the zeolite	Pore diameter (nm)	Examples
Small pore zeolites	0.30-0.45	Zeolite A
Medium pore zeolites	0.45-0.60	ZSM-5
Large pore zeolites	0.60-0.80	Zeolite X, Zeolite Y

Table 9: The Si/Al ratio of various types of synthetic zeolites (Xu *et al.*, 2010; Kulprathipanja, 2010).

Si/Al ratio	Examples of the zeolite
1-1.5	Zeolite A, X
2-5	Zeolite Y, L, omega
10-100	ZSM-5, beta

Framework diagrams of Zeolite types are shown in Figure 4 according to the database of the International Zeolite Association (IZA) structure commission.

Elordi *et al.* (2012) have investigated the effect of the acidity of HZSM-5 zeolite catalysts on the cracking of

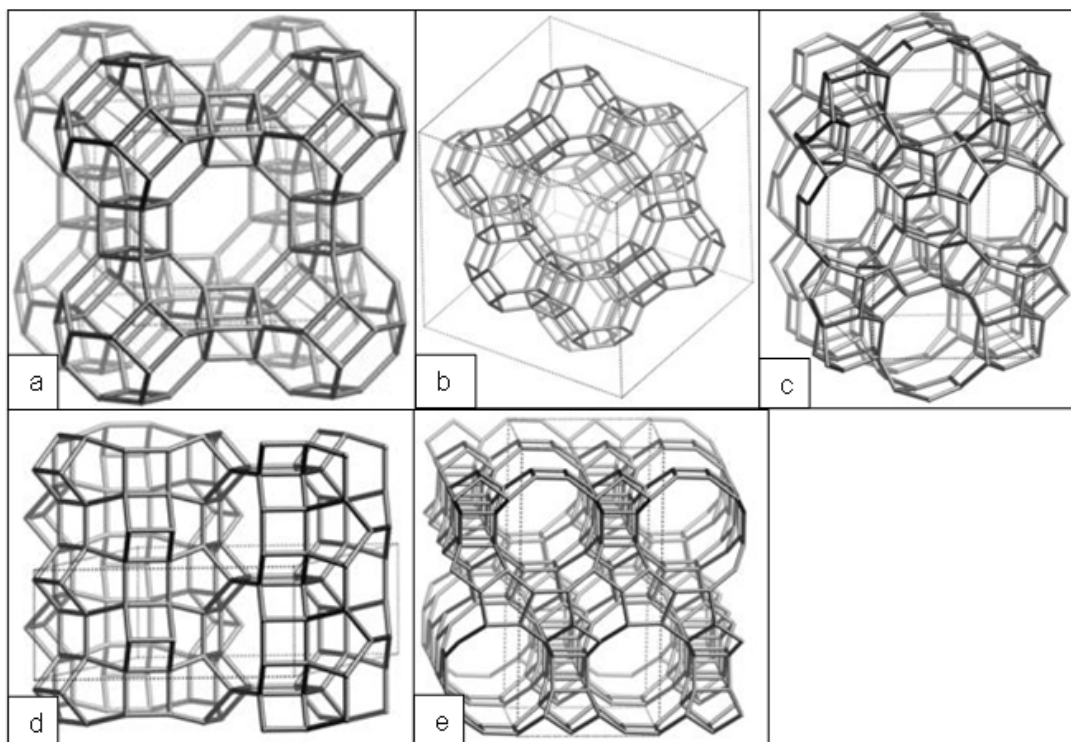


Figure 4: Framework of (a) Zeolite A (b) Zeolite X and Y (c) ZSM-5 (d) omega (e) beta catalysts (Resource – Database of International Zeolite Association (IZA) structure commission).

HDPE in a conical spouted bed reactor at 500 °C. Two different Zeolite catalysts with SiO₂/Al₂O₃ ratio of 30 and 80 and having different acid strength have been used in this cracking process. It has been observed that the increase in SiO₂/Al₂O₃ ratio decreases the total acidity thus decreases the acid strength of these catalysts. As a result of this, when the zeolite with SiO₂/Al₂O₃ ratio of 80 was used it has been able to obtain a high yield of C₂-C₄ olefins and non-aromatic C₅-C₁₁ fraction with a low yield of aromatic components and C₁-C₄ paraffin. In addition, using the same catalyst it has been able to obtain 59.8 wt.% and 32.1 wt.% yields of C₂-C₄ olefins and gasoline fraction (C₅-C₁₁), respectively. However, the development of the coke was increased as the SiO₂/Al₂O₃ ratio of the zeolite was increased.

The catalytic pyrolysis of HDPE and LDPE over HZSM-5 and HUSY zeolite catalysts have been investigated by Marcilla *et al.* (2009b) in a batch reactor. The resultant products of this thermal cracking and the catalytic cracking processes can be summarized as shown in Table 10.

In this study, the gas yield has been drastically increased when the HZSM-5 catalyst is used. In addition, the liquid/wax yield has been significantly decreased when the HZSM-5 catalyst is used. In the case of the HUSY catalyst, the resulting gas yield has been moderately increased while the liquid/wax yield has been moderately decreased when compared to the thermal cracking process. Also, a slight formation of coke was observed in both of these catalytic cracking processes of LDPE and HDPE.

According to the literature, it has been confirmed that polyethylene molecules are able to diffuse into the narrow pores of the HZSM-5 catalyst unlike the other polymers such as polypropylene (PP) (Zhou *et al.*, 2004). In another study, Marcilla *et al.* (2009b) have suggested that the branches or chain ends of polyethylene may penetrate the pores of HZSM-5 zeolite and contact with the acid sites located there, thus increase the catalytic reactivity. HZSM-5 has both strong and weak acid sites, whereas HUSY only has weak acid sites. When compared to weak acid sites, strong acid sites have a higher ability to degrade or crack the heavier hydrocarbon chains in the polymers (Marcilla *et al.*, 2009b).

It has been observed a slight formation of coke with HUSY catalyst than that of HZSM-5 catalyst. The surface area and the pore volume are higher in HUSY when compared to the HZSM-5 as shown in Table 11. In addition, the HUSY catalyst contains only weak acid sites, and the catalytic cracking process is not effective as in the HZSM-5 catalyst

which contains both strong and weak acid sites. Therefore, polymer chains that are not degraded completely can be deposited as the coke yield.

Table 11: Properties of the catalysts (Marcilla *et al.*, 2009b).

Property	HZSM-5	HUSY
Particle size (μm)	3	1
BET surface area (m ² /g)	341	614
Micropore volume (cm ³ /g)	0.16	0.29

Miskolczi *et al.* (2016) have studied the catalytic effect of activated carbon, HZSM-5, and MCM-41 separately and their mixtures on the pyrolysis of a mixture of waste HDPE and LDPE. This pyrolysis has been carried out in a batch reactor at 530-540 °C of temperature. In the absence of the catalysts, it has been able to obtain 42.7 wt.% and 5.1 wt.% yields of pyrolysis oil and gaseous products, respectively. When activated carbon catalyst was used, the resulting pyrolysis oil and gas yields have been increased only to 49.2 wt.% and 7.2 wt.%, respectively. This shows that activated carbon is not a highly effective catalyst for this pyrolysis process. However, the sulfur content of pyrolysis oil has been significantly decreased in this process.

In the presence of MCM-41 catalyst, it has been able to obtain 63.9 wt.% and about 10 wt.% yields of pyrolysis oil and gaseous fraction, respectively while HZSM-5 catalyst yielded 61.4 wt.% and 21.1 wt.% of pyrolysis oil and gaseous fraction, respectively. The reason for the increment of the gaseous yield could be the differences in pore sizes and the acid strength of these two catalysts (Table 12). As the Si/Al ratio of MCM-41 is higher than that of HZSM-5 (40 and 25, respectively), the acidity of the MCM-41 is lower than that of the HZSM-5 (Miskolczi *et al.*, 2016). Also, the MCM-41 catalyst contains a structure with a higher average pore size than the HZSM-5. Therefore, the MCM-41 is more catalytically active and increases the pyrolysis oil yield. As the pore size of HZSM-5 is smaller, it can increase the yield of the gaseous product in the plastic pyrolysis process (Miskolczi *et al.*, 2016).

Table 12: Properties of the catalysts (Miskolczi *et al.*, 2016).

Property	Activated carbon	MCM-41	HZSM-5
Si/Al ratio	-	40	25
Acidity (NH ₃ /g)	-	0.15	0.60
BET area (m ² /g)	859	824	298

Table 10: Yield of the different fractions obtained during the polyethylene pyrolysis process using two types of zeolite catalysts (Marcilla *et al.*, 2009b).

Yield (mg/100 mg of polyethylene)	LDPE Thermal cracking	HDPE Thermal cracking	LDPE – HZSM5	HDPE – HZSM5	LDPE - HUSY	HDPE - HUSY
Gases	14.6	16.3	70.7	72.6	34.5	39.5
Liquid/waxes	93.1	84.3	18.3	17.3	61.6	41.0
Coke	-	-	0.5	0.7	1.9	1.9

FCC catalyst used in the pyrolysis of HDPE and LDPE

The FCC catalyst has three main parts including zeolite crystals and a non-zeolite acid matrix made of silica-alumina with a binder in the catalyst structure (Sharuddin *et al.*, 2016; Lin *et al.*, 2010). In the case of the plastic pyrolysis process, both fresh FCC catalyst and deactivated FCC catalyst named as spent or equilibrium catalyst has been used for the catalytic cracking process. Achilias *et al.* (2007) carried out catalytic pyrolysis of both virgin and waste HDPE and LDPE in a fixed bed reactor with FCC catalyst at 450 °C. The product yields obtained herein are shown in Table 13.

Table 13: Product yield from the catalytic pyrolysis of HDPE and LDPE (Achilias *et al.*, 2007).

Plastic type	Gas wt.%	Liquid oil wt.%	Residue wt.%
Virgin LDPE	0.5	46.6	52.9
Virgin HDPE	0.5	38.5	61.0
Waste LDPE	8.5	72.1	19.4
Waste HDPE	3.3	44.2	52.5

In the above study, it has been observed that the liquid oil fraction of waste LDPE mainly contains gasoline range (C_7 - C_{12}) hydrocarbons with *iso*-alkanes or *iso*-alkenes. In addition, the liquid oil fraction from waste HDPE mainly contains a high carbon range of hydrocarbons with normal alkenes.

In an another study, Marcilla *et al.* (2005) have studied the thermal behavior of different HDPE mixtures under three different catalysts, HZSM-5, FCC catalyst, and HUSY. The properties of these three catalysts are shown in Table 14.

Table 14: Properties of the catalysts used in pyrolysis of HDPE (Marcilla *et al.*, 2005).

Property	HZSM-5	FCC	HUSY
Composition	SiO ₂ /Al ₂ O ₃ : 30 (molar basis)	Al ₂ O ₃ : 49 (wt.%) NaO: 0.25 (wt.%) Re ₂ O ₃ : 2.7 (wt.%) ZSM-5: 18 (wt.%)	SiO ₂ /Al ₂ O ₃ : 6 (molar basis)
BET area (m ² /g)	420	135	790

In this study, it has been observed the effect on pyrolysis temperature of HDPE (uncatalyzed- 470 °C), by the presence of the 3 catalysts of HZSM-5 (374 °C), HUSY (382 °C), and FCC (415 °C).

Elordi *et al.* (2009) studied the effect of fresh FCC and equilibrium FCC catalysts on the pyrolysis of HDPE in a conical spouted bed reactor at 500 °C. After pyrolyzing HDPE under fresh FCC and obtaining the resultant yield, the catalyst was deactivated by steaming treatments to obtain the equilibrium FCC catalyst. In the case of fresh

FCC catalyst, it has been able to obtain 52 wt.%, 35 wt.%, and 13 wt.% yields of gases (C_1 - C_4), light oil fraction (C_5 - C_9), and diesel fraction (C_{10}^+ hydrocarbons), respectively. In addition, the resultant diesel fraction has been increased to 40 wt.% and 69 wt.% with the equilibrium catalysts obtained through mild and severe steaming treatments, respectively. However, the gaseous yields have been decreased when using the equilibrium FCC catalysts (mild steam: 22 wt.%; severe steam: 8 wt.%) due to the reduction of acid sites in the FCC catalysts due to steaming treatments.

Other catalysts used in the pyrolysis of HDPE and LDPE

Several researchers have investigated the catalytic properties of TiO₂ in plastic pyrolysis. TiO₂ has been identified as a suitable heterogeneous catalyst in pyrolysis due to its porous surface and product selectivity, high thermal stability and mechanical strength (Eschemann *et al.*, 2014). The Lewis acidity, as well as non-toxicity of TiO₂, were reported to be very useful for hydrocarbon cracking in plastic pyrolysis (Nwankwor *et al.*, 2021).

Nwankwor *et al.* (2021) have studied the synthesis of gasoline range fuels by the catalytic cracking of waste plastics using TiO₂ and zeolite catalysts. This experiment has demonstrated that the liquid products of the LDPE pyrolysis process were mainly aliphatic hydrocarbons in the gasoline range. The use of zeolite catalyst in pyrolysis of LDPE has produced a higher amount of liquid products 44.2 wt.% than the TiO₂ catalyst (27.2 wt.%). In another study, TiO₂ has been added as a nano additive (particle size, 30-40 nm) to liquid oil obtained after the pyrolysis of a mixture of plastic waste to investigate its ability to change the properties of liquid oil (Bharathy *et al.*, 2019). By using this TiO₂ and liquid oil mixture in a diesel engine, it has been observed that the amount of hydrocarbons and CO pollutants in the emission can be minimized.

Panda *et al.* (2019) have used sulfated zirconium hydroxide to pyrolyze HDPE and LDPE in a batch reactor at 500 °C to obtain gasoline, kerosene, and diesel range hydrocarbons (C_{10} - C_{24}) in the resultant liquid oil yield. The sulfated zirconium hydroxide catalyst contains both Bronsted and Lewis acid sites and can be easily synthesized for low cost for commercial use. It has been reported that the liquid oil yields from the pyrolysis process of HDPE and LDPE were 79.5 wt.% and 82 wt.%, respectively.

Kunwar *et al.* (2016) have carried out the pyrolysis of waste HDPE by employing MgCO₃ as the basic catalyst for the catalytic cracking at 450 °C. It has been observed that a slight reduction in process temperature (by 10 °C) with the use of MgCO₃ catalyst than that of the thermal pyrolysis process. In the presence of MgCO₃ catalyst, it has been able to obtain 80 wt.% and 18 wt.% yields of liquid oil and gaseous products, respectively. In addition, the resultant liquid oil yield was found to contain a higher amount of diesel range hydrocarbons. In the thermal pyrolysis process, 86.2 wt.% of the wax product was yielded indicating ineffective decomposition.

The extensive review of the literature reveals that the use of suitable catalysts enhanced the production of desirable

yield (liquid oil yield or gaseous yield) in the resultant product at a lower temperature compared to the thermal pyrolysis process. At around 500 °C of temperature, it has been observed that the HZSM-5 catalyst has selectivity toward the gaseous products whereas HUSY, MCM-41, silica-alumina, and other catalysts have selectivity toward the liquid oil products in plastic pyrolysis (Miandad *et al.*, 2019b; Achilias *et al.*, 2007; Li *et al.*, 2016). Table 15 summarizes several HDPE and LDPE pyrolysis experiments performed under different catalysts and temperatures.

Emissions in the pyrolysis process and engine combustion of the fuel oil, performance, and emission characteristics

The amount of non-condensable gases in the pyrolysis mainly depends on the operation temperature, the residence time, and the catalytic behavior. In the incineration or gasification processes, the product formation occurs at high temperatures in the presence of oxygen and generates harmful compounds to the environment. Whereas in the pyrolysis thermal cracking process occurs at low temperatures (400-900 °C) in the absence of oxygen, it prevents the formation of dioxins and reduces the formation of carbon monoxide(CO) and carbon dioxide(CO₂) gases (Singh & Ruj, 2016; Miskolczi *et al.*, 2009).

A very few studies have focused on the gas emissions in the pyrolysis processes of municipal plastic waste whereas many researchers have studied the gas emission from the pyrolysis process of virgin plastics and the mixtures of HDPE, LDPE, PP, and PS. It has been reported that the waste HDPE and LDPE have produced more gaseous components in the carbon range of C₃ and C₄, such as methane, ethane, ethene, n-butene in the pyrolysis at 500 °C. In addition, it has produced H₂, CO, and CO₂ gases during the pyrolysis of waste HDPE and LDPE (Singh & Ruj, 2016).

Miskolczi *et al.* (2009) have analyzed the gaseous yield of the waste HDPE pyrolysis process at 520 °C in the presence and absence of ZSM-5 catalyst. They have obtained 5.1 wt.% and 12.2 wt.% yields of gases with and without the catalyst, respectively. The composition of the gaseous product in this process is shown in Table 16.

According to Table 16, the formation of alkenes is slightly higher than the formation of alkanes in both thermal and catalytic pyrolysis processes.

The engine combustion of the liquid fuel obtained, its performance, and emission characteristics were measured in some studies using internal combustion diesel engines (Kalargaris *et al.*, 2017b; Kalargaris *et al.*, 2017c; Güngör

Table 15: Summary of the yield obtained in thermal and catalytic pyrolyzing of HDPE and LDPE.

Plastic-type	Catalytic pyrolysis						Thermal pyrolysis				References
	Temperature °C	Catalyst	Liquid Wt.%	Char Wt.%	Gas Wt.%	Polymer to catalyst ratio	Temperature °C	Liquid Wt.%	Char Wt.%	Gas Wt.%	
HDPE	430	Y-zeolite	75	6	19	10:1	460	86.2 wax	1	13	Kunwar <i>et al.</i> (2016)
	450	MgCO ₃	80	2	18	10:1					
HDPE	500	HZSM-5 (30)*	78.65	1.85	19.5	10:3	-	-	-	-	Elordi <i>et al.</i> (2012)
		HZSM-5 (80)*	72.98	1.02	26.0	10:3	-	-	-	-	
HDPE/ LDPE	530	Activated carbon	49.2	-	7.2		540	42.7	-	5.1	Miskolczi <i>et al.</i> (2016)
		MCM-41	63.9	-	10	25:1					
		HZSM-5	61.4	-	21.1						
HDPE	450	FCC	91.2	4.1	4.7	10:1	450	85 wax	-	-	Olazar <i>et al.</i> (2009)
HDPE LDPE	500	Sulfated zirconium hydroxide	79.5 82	- -	- -	10:1	-	-	-	-	Panda <i>et al.</i> (2019)
LDPE	99-198	TiO ₂	27.2	-	-	10:4	82-140	28.5	-	-	Nwankwor <i>et al.</i> (2021)

*SiO₂/Al₂O₃ ratio

Table 16: Composition of the gaseous product in the waste HDPE pyrolysis process using ZSM-5 catalyst (Miskolczi *et al.*, 2009).

Gaseous product	HDPE without a catalyst	HDPE + ZSM-5
Methane	3.1	2.5
Ethene	30.6	26.1
Ethane	21.4	19.5
Propane	8.9	8.9
Propene	4.3	6.8
Butene	17.1	14.6
Butane	14.6	10.9
<i>iso</i> -Butane	0.0	10.7

et al., 2015). Next, their properties were compared with the combustion characteristics of commercially available diesel fuel to identify the right applications for the pyrolysis product. Some studies reported that diesel engines could operate merely with liquid fuel resulting in pyrolysis with no added conventional diesel (Kalargaris *et al.*, 2017b). However, studies showed that when it used in internal combustion diesel engines, the brake thermal efficiency and cetane number were lower than in conventional diesel (Kumar *et al.*, 2013; Kalargaris *et al.*, 2017a; Kalargaris *et al.*, 2017b; Güngör *et al.*, 2015).

Kalargaris *et al.* (2017b) revealed that fuel oil from pyrolysis of waste LDPE exhibit almost identical combustion characteristics and brake thermal efficiency with conventional diesel operations. Therein, minimal NO_x, CO, and CO₂ emissions and higher unburned hydrocarbons were observed (Kalargaris *et al.*, 2017b). Gopinath *et al.* (2020) have observed that a 20% (volume) of waste LDPE fuel oil blended with diesel combination showed similar combustion performance and brake thermal efficiency as diesel. However, NO_x and smoke emissions were high compared to diesel. In another study of waste HDPE pyrolysis, Kulandaivel *et al.* (2020) reported a significant decrement in NO_x, hydrocarbons (HC), and CO emissions when using 30% (volume) of HDPE pyrolyzed fuel oil with diesel upon minimal modifications of the diesel engine as retarding injection timing.

Suggestions for future work: Research areas to complete

It is important to conduct awareness among communities and to introduce a proper waste separation method to the country to develop energy recovery practices with plastic waste. There are some issues in the mixed plastic waste pyrolysis process including difficulties in identifying the composition of the mixed waste, composite, laminated materials, and metal parts in the plastics. It is important to identify the waste composition of the mixed plastic waste because PVC and PET plastics generate hydrochloric acid and benzoic acid, respectively, during the pyrolysis process which are toxic and corrosive to the reactor (Papari *et al.*, 2021). The spectrum analysis method such as FTIR, Raman, and Near-IR spectrum analysis can be used to identify the composition of the mixed plastic waste. Different sorting

techniques such as manual sorting, sorting by density, air classifications, electrostatic separations and sensor-based sorting techniques can be used to separate plastic types (Sutar, 2015). The novel trend of plastic waste separation from municipal solid waste is the automated sorting technique (Gundupalli *et al.*, 2017).

The influence of the catalysts is a governing factor in the plastic pyrolysis process when considering resultant product selectivity and reduction of the temperature. The high cost of the catalysts in the catalytic pyrolysis process is a critical issue when considering the implementation of pyrolysis technology in the plastic waste management system. Natural origin minerals such as Bentonite clay, Mordenite, Clinoptilolite, Red mud, Shewedaung clay, and Mabisan clay that are highly available in the country can be used as catalysts to reduce the cost of the pyrolysis process. The zeolite and different derivatives of zeolites have been used by many researchers because of their abundance. In addition, the repeated catalysts regeneration method has been used to improve the catalysts' lifetime and reduce the operation cost of the plastic catalytic cracking process.

When the objective is to maximize the liquid oil fraction in the pyrolysis process, the resultant gaseous product becomes an issue. These resultant gas components have a high calorific value and can be used as a heating source for the pyrolysis process again. Also, without further treatments, the resultant gases can be used in gas turbines to generate electricity and can be fired to operate boilers. The ethane and propene formed in the pyrolysis of plastics can be used as feedstocks in the chemical industry after the separation using a proper separating technique (Honus *et al.*, 2018). In addition to that, the emission of hazardous gases in the plastic pyrolysis process can be avoided by using gas filters (Padmaja, 2016). Filters with a porous material can trap gases or may convert them to other products by chemical reactions that occurred inside the pores of the filters.

The oil yield and its quality characteristics vary depending on the process conditions of pyrolysis. In order to achieve sustainable development goals, the enacting of waste recycling policies and standards is mandatory for the establishment and operation of plastic waste processing facilities. (Namkung *et al.*, 2022). Some States of the United States, including Iowa, Ohio, and Texas, enacted legislation that allowed certain types of plastics recyclers to process crude oil, gasoline, and home heating oil (Hogue, 2022). Although some states allow the use of plastic in advanced recycling, in Kentucky, a law passed in 2021 prohibits the use of post-consumer polymers in the production of fuels (Hogue, 2022). However, companies in several countries, including Malaysia, Thailand, and India, are currently considering using pyrolysis technology in their waste management plans, and further commercialization of plastic waste pyrolysis plants is expected to continue (Wong *et al.*, 2015). Also, institutions were established to provide technical assistance to use this technology in their respective countries. Owing to the advantages of this technology, many countries will incorporate pyrolysis technology into their waste management plans in the future.

CONCLUSION

The demand for plastic waste pyrolysis technology is increasing due to growing concerns on plastic waste pollution and the need of sustainable solutions for such waste management. Pyrolysis technology is an effective and an environmentally friendly method over landfilling and incineration and create a circular economy for plastics while converting waste into valuable byproducts. This review has provided a summary of plastic pyrolysis of LDPE and HDPE with a discussion of the parameters that control the fuel oil, char, and gaseous products. The plastic pyrolysis process can be carried out in both thermal and catalytic processes. The operating parameters such as temperature and reaction time can be reduced using appropriate catalysts that increase the desired product yields. At the temperature of 400 - 500 °C, the HZSM-5 catalyst has selectivity toward the gaseous products whereas HUSY, MCM-41, silica-alumina, and other catalysts have selectivity toward the liquid oil products in plastic pyrolysis.

Adversely, there are some difficulties in the feedstock selection of plastic pyrolysis due to ineffective segregation methods of municipal waste. Although the effect of environmental pollution is lower in plastic pyrolysis than in landfilling and incineration practices, it is vital to practice filtering or trapping gaseous products evolved in pyrolysis to safeguard the environment. Regulations for environmental protection, health and safety, and waste management have been implemented and those apply to plastic waste pyrolysis too. In some countries, including European Union and United states, there are specific regulations or incentives to promote the use of pyrolysis technology for plastic waste management. Furthermore, these laws and regulations are in favor of deployment of plastic waste pyrolysis technology in their community. Further, the need of fossil fuel can be reduced to a certain extent, if there are effective means of refining the fuel oil produced in pyrolysis.

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DECLARATION OF CONFLICT OF INTEREST

The authors declare no competing interests.

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