

Liquid Fuel from Plastic Waste using Pyrolysis: A Review

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Abstract:

Due to rapid industrialization as well as increase in medical waste, plastic waste which is non-biodegradable is increasing day by day. Plastic waste management was found to be done by several methods such as plasma process, incineration and pyrolysis out of which catalytic pyrolysis was found to be most economical as it gives alternative for non-renewable sources of energy. Fuel oil can be produced from several raw materials such as biomass, plastic, tyres etc. here polypropylene was used as raw material due to its low melting point also it gives higher calorific value fuel oil. Batch reactor has certain drawbacks as compared to continuous reactors such as manpower requirement, char getting accumulated inside reactor, higher residence time, less pure oil, higher operating temperature etc. in this review various reactors used for pyrolysis have been discussed along with effect of parameters affecting the pyrolysis. Non-condensable gases can be used to heat reactor and minimize waste heat loss.

Keywords: *Batch reactor, conical Spouted bed reactor, fuel oil, HZSM-5, polypropylene.*

1. Introduction:

Plastic waste management can be done in many ways like recycling, landfilling but recycling is labour intensive process and landfilling needs several years for plastic to get decomposed. Plastic is petroleum based long chain hydrocarbon compound and petroleum resources are non-renewable in nature therefore plastic can be converted into fuel by cracking it at some desired temperature using different types of plastics such as low density polyethylene, high density polyethylene, polypropylene, polyvinyl chloride, polystyrene etc. as a raw material. Plastic waste management can be done by Plasma Process, Incineration and Pyrolysis. In this review pyrolysis process is discussed in detail.

Pyrolysis is a process in which raw material is exposed to thermal degradation in absence of oxygen, using catalyst to increase the rate of reaction that's to obtain desired product at less temperature and in less time. Pyrolysis was found to be more economical process as it requires less temperature (500⁰C to 800⁰C) for plastic waste management as well as it emits less greenhouse gases than that of Incineration as it takes place in absence of oxygen and it is less expensive than Plasma process which require temperature of 5000⁰C-10000⁰C.

S D A Sharuddin et al. (2018) reported that during pyrolysis oil, gas, char are the three major products formed that are valuable for production and refinery industries. Pyrolysis is flexible process since the parameters can be changed according to the final product requirement. Parameters to be considered during this process are temperature, type of reactor, pressure, residence time, catalyst selection, and type and rate of fluidizing gas, if used.[1]

Feng Gao (2010) and Imtiaz Ahmad et al. (2014) reported that pyrolysis takes place in three steps:

1) Initiation: In this step larger polymers are converted into free radicals. (thermolysis)[2]



2) Propagation: In this step free radicals, molecules are converted into smaller radicals and molecules. Beta scission is a dominant reaction in pyrolysis of polyethylene.



A radical may fragment as under giving a small chain radical and aliphatic hydrocarbon:



3) Termination: In this step radicals combine and form a stable molecule.

Hydrogen abstraction (produced by shuttling) leading to formation of aliphatic hydrocarbons.



Retrogressive Recombination



2. Selection of raw material:

Fuel oil can be produced from several raw materials such as biomass, plastic, tyres but biomass is biodegradable and requires less time to get decomposed therefore here plastic was chosen as a raw material which is non-biodegradable.

Following are the two types of plastic-

1. Thermoplastics: This type of plastic can be melted by providing enough heat, hardened on cooling and they can be transformed into new plastic products. eg. Low density polyethylene, high density polyethylene, Polypropylene, polyvinylchloride, polystyrene etc.
2. Thermosets: this type of plastic are melted and then solidified but once they are solidified, they can not be reshaped. eg. Phenol, formaldehyde.

Rohit kumar singh et al. (2019) stated that the waste was characterized having proportion of PE (LDPE and HDPE) 58.8%, PP 26.9%, PS 8.7% and PET 5.6%. PVC was excluded from the waste as it produces HCL during the process affecting the product and reactor due to its corrosive nature. The process uses 200 g of sample per batch to represent the homogenous mixture and represent a real mix waste.[3]

P. Senthil Kumar et al. (2015) used PE, PS, PP in individual without using catalyst as well as in mixed manner with and without using catalyst in a batch reactor. Catalyst used here were activated carbon, granulated charcoal, calcium oxide. Temperature was maintained at 240°C and vacuum pressure of approximately

300mmHg. polystyrene at 550 mmHg and reaction time 30 min gave 80% yield. PP at 500 mmHg for reaction time 35 min gave 60.7% yield. PE at 300mmHg for 45 min reaction time gave 75% yield.[4]

Miriam Arabiourrutia et al. (2017) reported pyrolysis in CSBR at 500-700⁰C (thermal) and at 450-500⁰C (catalytic) using HDPE, LDPE, PP using sand bed of 0.6-1.2 mm diameter particle size. At 450-500⁰C PP gave better yield than HDPE and LDPE.[5]

R. Saravana Sathiya Prabhakar et al. (2018) investigated pyrolysis in batch reactor using HDPE and polypropylene (PP) as a raw material which had calorific value 3580.41kcal/kg and catalysts used were Nickel Nano particles and Nickel Oxide Nano particles. Without using catalyst 15g of HDPE gave 8.2ml oil and 15g of PP gave 11. 3ml oil. With use of catalysts following results were obtained at 450⁰C[20]. The results obtained by author reported in table 1.

Table (1) :Oil yield using HDPE and different catalysts [20]

MATERIAL	QUANTITY OF CATALYST	OIL OBTAINED WITH	
		Ni NANO PARTICLES (ml)	NiO NANO PARTOCLES (ml)
15g HDPE	0.25	12.2	10.2
15 g HDPE	0.5	12.3	11.2
15 g HDPE	0.75	12.3	12.2

Following factors were considered for choosing Polypropylene (PP) over Polyethylene (PE)

- PP has low carbon footprint than PE.
- PP is abundantly used in plastic bags, plastic bottles so raw material is easy to get.
- The crystallinity of PP is less than PE.
- PP fuel has more calorific value than all fuels.

3. Selection of Catalyst:

To improve the quality of the product and reduce the temperature and retention time catalysts such as Zeolites, FCC, Silica Alumina catalyst were found to be used. Brunauer-Emmett-Teller (BET) surface area. Pore size, volume and acidity should be high as these factors affects the characteristics of catalyst. Heterogeneous catalysts are used instead of homogeneous because solid and liquid mixture from the product can be easily separated.

Hoang Vu Ly et al. (2019) reported that by using bamboo as a raw material in a bubbling fluidized bed, MOC stainless steel reactor bed used was silica sand (100g). For thermal pyrolysis heating rate was kept 10⁰C/min from 25 to 700⁰C, the yield obtained was highest (53.94wt%). When catalytic pyrolysis with HZSM-5 and red mud catalyst it gave 49.14wt% and 50.34wt% yield respectively better yield then thermal. HZSM- 5 catalysed pyrolysis gave more methoxy phenolic and aromatic compounds than red mud catalysed pyrolysis that gave more saturated phenol and furan derivatives.[6]

Miriam Arabiourrutia et al. (2017) reported pyrolysis in CSBR at 500-700⁰C (thermal) and at 450-500⁰C (catalytic) using HDPE, LDPE, PP. With increase in temperature gas yield increased while wax decreased. With catalysts (HZSM-5, H Gamma, H Beta), 30 grams HZSM-5 gave more yield of olefin fractions while other two gave more C₅ - C₁₁ fractions. With catalyst wax yield decreased. At 450-500⁰C PP gave better yield than HDPE and LDPE. Fig. 5.6 shows process flow diagram for pyrolysis in CSBR.[5]

4. Reactors used for pyrolysis

4.1. Pyrolysis of plastic waste in batch reactor:

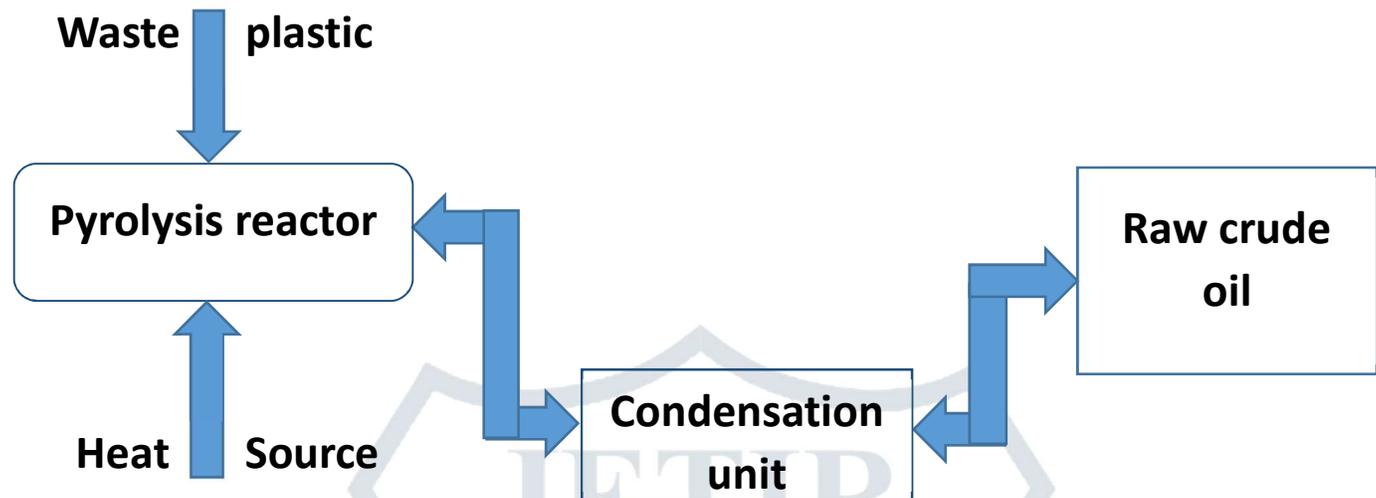


Fig. (1): Box flow diagram for pyrolysis of plastic waste in batch reactor investigated by **Samuel Kofi Tulasiea et al. (2019)**

Properly washed and dried plastic is subjected to temperature of approximately 600-650⁰C in absence of oxygen in a reactor so as to obtain end products namely oil, gas and char. Here plastic starts melting and the melting point depends on type of plastic chosen, lower the melting point higher will be the yield since secondary cracking reactions would be terminated. As the plastic is heated the vapours will start coming out from reactor which will contain hydrocarbons, they are condensed at 20-30⁰C and our desired product will be obtained. This process requires 1-2 hours and the oil is impure and hence can be used as feed for refineries or hydrocracking. If catalyst is added this process will take less time and by terminating secondary cracking reactions and minimizing vapour residence time the yield obtained would be more at lesser temperature. The gases which are collected that's non condensable gases can be flammable and can be used for energy production. Here char gets accumulated inside the reactor as it is not a continuous operation.

4.2. Continuous reactors for plastic waste pyrolysis:

Khan Muhammad Qureshi et al. (2018) reported that in **Bubbling Fluidized Bed Reactor (BFB)** the fluidizing gas will rise through distributor plate and will burst on the bed surface when velocity is reached minimum fluidization velocity. Bubbles are formed of small amount of solid. The system is stable for particle size of 0.5-2mm. Uniform heat transfer is achieved here. Optimum operating temperature for BFB is 500-550⁰C and vapour residence time of approximately 2-3 s. In case of **circulating Fluidized Bed Reactor (CFB)** particles circulate in a constant loop that forms cyclone and therefore here there will be good gas-solid contact that will offer good mixing. Here vapour residence time will be lesser tat's 0.5-1 sec but particle size required would be less than that of required in BFB.[7]

Maidier Amutio et al. (2015) reported fast pyrolysis of eucalyptus waste in CSBR. Raw material was in size range 1-2mm and for feeding feeder of capacity 200g/h and 0.12kg/h feed rate gave 75.4wt% liquid yield at 500⁰C which was maximum along with 6.2wt% gas and 18.4wt% char. As compared to previous literatures of

pyrolysis in fluidized bed reactor which gave average 65wt% yield was obtained which was less than CSBR.[9]

Conical Spouted Bed Reactors (CSBR) are alternative to fluidized bed reactors. Here counter current flow of solid and gas phase takes place. Secondary reactions which lead to low yield are minimized here since low residence time (<1sec) of volatile products and gives higher yield. Heat and mass transfer rates are higher in case of CSBR as compared to fluidized bed reactor due to excellent gas-solid mixing. In lab scale grinding of particles is needed but when it comes to large scale grinding is not necessary and therefore grinding cost is decreased unlike fluidized bed reactors which require equal and lesser diameter particles otherwise clogging of distributor plate. Here char removal becomes easy due to density difference between char and solid particles in bed. Even if CSBR give less yield than that of in Fluidized bed reactor and fixed bed reactor but the quality of oil was better in CSBR in terms of higher calorific value as well as gasoline range oil with valuable compounds such as limonene were present in it when biomass such as orange peel is used.

In **Screw /Auger reactors** the screw in a rotational motion mixes and transports the biomass through the tube. It offers effective mixing and transport. The product output depends on residence time and reactor temperature of the reactor. Liquid yield is from 18 to 25% and charcoal yield is from 50 to 60%. These are applicable for both slow and fast pyrolysis. Separation of solid and liquid is easy. [10]. Higher residence time, Risk of moving parts, Difficulties of heat transfer at large scale, poor mixing are the reason for selecting CSBR over Screw reactors.

In **Continuous Rotary Kiln Reactor** explained by **S. Q. Li (2004)** particles of 13-15mm size can be used. operating temperature range from 450 – 650⁰C. Adjustable residence time. Used for both batch and continuous process.

Reason for selecting CSBR over CRKR are:

1. Low heat efficiency.
2. Formation of agglomerates.
3. Yield – 45.1wt%.[4]

R.W.J. Westerhout et al. (1998) used a **Rotary Cone Reactor** which consisted of following:

Operating temperature is 275-302⁰C. Sand diameter is 0.1-0.3 mm. grinding of particle is not necessary. External gas is not required.

Reason for selecting CSBR over rotary cone reactor are:

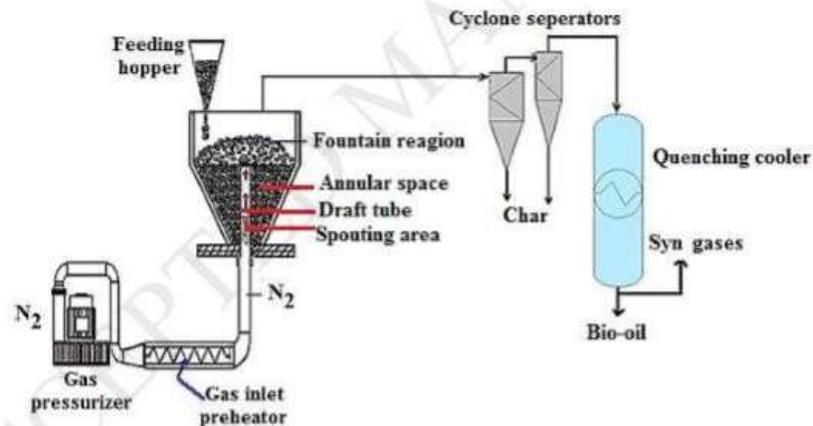
1. High rotational speeds due to which particle inclusion may occur.
2. Yield is 60 – 70wt%.[11]

Fixed Bed Reactors are batch type of reactors where it becomes difficult to remove char and requires temperature from 600-650⁰C in case of catalytic pyrolysis and till 800⁰C if thermal pyrolysis.

Khan Muhammad Qureshi et al. (2018) reported that Batch mode pyrolysis requires high vapor residence time which lowers the yield of final product. Batch process doesn't require any fluidizing gas, high pressure pump or compressor and is simple at lab scale but when laboratory batch setup is proposed in industrial scale it requires high labour cost and shows difficulty in operation since it requires high reaction time, shows inconsistency in each batch product, it needs more heat up and cool down time. The major difference is fuel oil obtained from continuous pyrolysis has higher amount of lighter hydrocarbons than that of obtained in batch mode which gives more tar and heavy liquid fractions. In case of semi-continuous mode reactors

accumulation of feed inside the reactor takes place which leads to unsteady operation. In case of continuous mode reactors feeding of reactors, product withdrawal as well as by-product removal takes place continuously for which it needs additional separating units such as cyclone separator, filters, electrostatic precipitator. Continuous mode reactors give lower labour cost and consistent product quality.[7]

Fig. (2): Process flow diagram for pyrolysis in Conical Spouted Bed Reactor investigated by **Khan Muhammad Qureshi et al. (2018)**



5. Effect of Parameters on catalytic pyrolysis:

5.1. Temperature:

Temperature is an important parameter in thermal cracking. It affects the amount of gaseous and liquid product produced. At low temperature long chain hydrocarbons are produced and at high temperatures small chain hydrocarbons are produced. Therefore, an operation should be conducted at optimum temperature.

A pyrolysis conducted by **Miandad R et al. (2016)** observed following effect of temperature on the different plastic types.

Table (2): Type of plastic and optimum temperature at which it will give better yield [12]

Type of Plastic	Temperature range (°C)
PET	350-520
HDPE	380-540
PVC	220-520
LDPE	360-550
PP	400-500
PS	350-500

If gaseous or char product is required then process should be operated at more than 500⁰ and if liquid product

is required then about 300-600°C temperature is applied.

S. M. Al- Salem (2019) reported pyrolysis of HDPE in novel fixed bed reactor, with raw material of size 3mm and melting point 131°C. The experiment was conducted between 500-800°C and N₂ flow. At 550°C maximum yield of oil was obtained that's 70% and at 750°C gas of 36wt%. [14]

Jon Alvarez et al. (2014) reported pyrolysis of rice husk for bio- oil (BO) production in CSBR. Experiment was conducted at 4 runs that is 400, 450, 500, 600°C, sand bed of 100g with particle size 0.3-0.63 mm was used, N₂ flow rate was kept 10NL/min. At 450°C maximum bio-oil yield was obtained that's 70 wt %. Gas yield was very low below 500°C (less than 6%). [15]

5.2 Residence Time:

Lower the vapour residence time higher will be the yield and better quality fuel would be obtained since secondary cracking reactions will be terminated. For higher residence time liquid yield will be less and gas yield will be more. Residence time can change with type of reactor used, catalyst, operating temperature and fluidization gas velocity.

Amer Inayat et al. (2020) compared parameters with respect to thermo-catalytic pyrolysis of polystyrene in batch and semi batch reactor. At 500°C catalytic batch reactor configuration performed better than one of the semi batch configuration since the N₂ rate was kept constant and residence time decreased. At 400°C without N₂ flow part of volatiles was unable to leave reaction zone which went under further degradation and minor compounds and residue was formed and with high amount of catalyst the residence time decreased which gave rise to incomplete reaction and resulted in more oligomers. [16]

5.3. Particle size:

Particle size requirement will vary as per the type of reactor.

Hoang Vu Ly et al. (2019) reported that by using bamboo as a raw material When particle varied from 0.45 to 0.75 the oil yield decreased from 54.03 to 50.85wt%. CSBR allows uneven and wide range of particle size. If the particle size is small the yield of the product will be more. In lab scale grinding of particles is needed but at large scale grinding is not necessary and therefore grinding cost is decreased. [6]

6. Waste heat recovery from non-condensable gases:

As Pyrolysis of plastic gives 3 products namely oil, char, gas. Char is not harmful to environment, rather it has many uses like in plant growing, as a catalyst, in fuel cell etc. But the non-condensable gases that are obtained during the process can be harmful to the environment, though it is not our major product, but these gases need to be treated or used in effective way after treatment. These gases can be used as fuel. Though they won't be as efficient as conventional fuel gases, they can be used. They can be used to heat the reactor, but they cannot achieve that much temperature so external heating coils are anyways required.

Sha Wang et al. (2014) reported that as the temperature was increased there was increase in Alkene/Alkane ratio which indicated secondary cracking reactions. Oil/non condensable gases ratio decreases with increase in temperature. With increase in temperature from 430 to 520°C non condensable gases, oil yield was increased and there was decrease in char yield. [17]

Kedar Sahoo et al. (2020) non-condensable gases produced during pyrolysis were made of CO, CO₂ and H₂ mainly. Non-condensable gases produced as a by-product could have better fuel application since it contained

mainly these gases. The CO and CO₂ content was found to be high in non-condensable gases produced below 450°C unlike high temperature, maximum CO, CO₂ was obtained at approximately 300°C, CO and CO₂ content was decreasing till 650°C. Due to C=O bond present in biomass high yield of CO and CO₂ at low temperature was obtained.[18]

Andrei Veksha et al. (2017) Reported that plastics were initially pyrolyzed at 600°C producing condensable oils and non-condensable gases. The non-condensable gases were then heated in the second reactor either at 500 or 800°C. Heating at 800°C caused decomposition of the gases to intermediate products (methane, ethylene and condensable oils) and that were converted over a nickel-based catalyst into multi-walled carbon nanotubes with similar yields and properties. Heating at 500°C shown no effect on the composition of non-condensable pyrolysis gases. At 500°C, the Mixed Plastic non-condensable pyrolysis gases were converted into carbon nanocages and multi-walled carbon nanotubes having higher graphitization degree compared to carbon materials from PP and LDPE, which were mainly multi-walled carbon nanotubes.[19]

On small scale if these gases are not treated, they can not be used as it is neither they can be vented out in environment so they can be discarded in water. At large scale, the scrubber needs to be installed to dissolve these gases.

7. Limitations and Future Scope:

Limitations: The fuel oil obtained from pyrolysis has lower octane number. The engines of vehicles are not designed for such fuel so it may cause problem in ordinary engine. Even though if the fuel oil is used in vehicles since it has higher carbon footprints than conventional fuel it can cause environmental problems. Catalyst delivery in reactor should be done by taking precautions else if catalyst fails it will cost more as it is expensive. If the fuel oil obtained is impure then it can be used as feed for refineries or for hydrocracking process if it is highly viscous than conventional fuel that's it needs to be further processed.

Future Scope: All the products obtained using plastic Pyrolysis namely gas, char, oil can be used as renewable source of energy. Gases can be used for generation of electricity and hydrogen rich gases for synthesis of fuel. Char can be used to prepare activated carbon that has high demand in wastewater treatment, air pollution control by adsorption. It can be used for making renewable energy storage devices like super capacitor, Li-ion batteries, fuel cell etc.

Conclusion:

Polypropylene is suitable type of plastic for performing pyrolysis, since it is abundantly available. Also it has low crystallinity, so it becomes easy to degrade it. And also fuel obtained from it has better calorific value. Catalytic cracking was found to be better than the thermal cracking because it is economically and environmentally suitable than thermal cracking. In this review different types of reactors that can be used for pyrolysis have been discussed along with the effect of parameters on pyrolysis. Conical Spouted Bed Reactor was found to be the better one among other reactors as compared above. Though it has lower yield than some other reactors, it has better quality. The parameters that should be considered are temperature and residence time. If residence time is less then secondary reactions don't occur and the good yield is acquired.

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