Pyrolysis of plastic waste for liquid fuel production as prospective energy resource

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Abstract. The worldwide plastic generation expanded over years because of the variety applications of plastics in numerous sectors that caused the accumulation of plastic waste in the landfill. The growing of plastics demand definitely affected the petroleum resources availability as non-renewable fossil fuel since plastics were the petroleum-based material. A few options that have been considered for plastic waste management were recycling and energy recovery technique. Nevertheless, several obstacles of recycling technique such as the needs of sorting process that was labour intensive and water pollution that lessened the process sustainability. As a result, the plastic waste conversion into energy was developed through innovation advancement and extensive research. Since plastics were part of petroleum, the oil produced through the pyrolysis process was said to have high caloric value that could be used as an alternative fuel. This paper reviewed the thermal and catalytic degradation of plastics through pyrolysis process and the key factors that affected the final end product, for instance, oil, gaseous and char. Additionally, the liquid fuel properties and a discussion on several perspectives regarding the optimization of the liquid oil yield for every plastic were also included in this paper.

1. Introduction

Plastic plays an important role in improving our lifestyles in numerous sectors such as healthcare, construction, packaging, electronic, automotive and many more. The rise of the world population has caused the demand of commodity plastics to further increase. According to statistic, the global production of plastic has reached about 299 million tons in 2013 and has increased by 4% over 2012. Since most of the plastics were thrown out after single use, the amount of plastic waste accumulated in the environment each year was at alarming level. In Europe, 25 million tons of plastic ended up in waste stream during the year of 2012. Based on the statistic, about 38% of the plastic waste still went to the landfill, 26% were

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recycled while 36% were utilized for energy recovery [1]. This signifies that the percentage of plastic waste dumped in the landfill was high and it occupied a large space. Besides, the degradation of plastics may take up billions of years, thus the continuous disposal of plastic in the landfill would definitely impact the environment negatively.

In order to overcome the challenges faced for recycling such as the needs of sorting that is labour intensive, a much more reliable method is established. Nowadays, converting the waste into valuable energy resource has been a brilliant way to fully utilize the waste in order to meet the increased energy demand. Plastic wastes can be turned into valuable energy since they are derived from petrochemical source which having significant calorific value. The conversion can be made possible through several thermal treatments technologies such as gasification, pyrolysis, plasma process and incineration [2]. Among all these methods, pyrolysis is the most desirable process since the initial volume of the waste is significantly reduced, more energy can be recovered from the plastic waste by producing varieties of products, requires lower decomposition temperature and low capital cost [2, 3].

Pyrolysis is the process of thermally degrading long chain polymer molecules into smaller, less complex molecules through heat. The process requires intense heat with shorter duration and in absence of oxygen. The three major products that are produced during pyrolysis are oil, gas and char which are valuable for industries especially production and refineries. Pyrolysis was chosen by many researchers since the process able to produce high amount of liquid oil up to 80 wt% at moderate temperature around 500°C [4]. In addition, pyrolysis is also very flexible since the process parameters can be manipulated to optimize the product yield based on preferences. The liquid oil produced can be used in multiple applications such as furnaces, boilers, turbines and diesel engines without the needs of upgrading or treatment [5]. The oil produced from biomass pyrolysis has received positive notice as a more environmentally friendly fuel because it contributes to reducing the amount of CO₂ in the atmosphere [6]. Unlike recycling, the process handling is also much easier and flexible than the common recycling method since it does not need an intense sorting process, thus less labour intensive.

Several research papers have been published regarding the potential of different types of plastics in pyrolysis processes for liquid production. It has to be noted that the product yield and quality mainly relies upon the set up parameters. Therefore, this paper reviewed the potential of thermal degradation of plastic with the important parameters in pyrolysis process that need to be considered to obtain an optimal liquid oil production and improve the oil quality. Selective catalysts and their influences to the product yield were briefly discussed. Additionally, some relevant discussion regarding the optimization of liquid oil yield and the potential fuel quality were also presented in this paper.

2. Process parameters influence on product yield

Parameters play major role in optimizing the product yield and composition in pyrolysis. There are several parameters that influence liquid oil production in pyrolysis such as temperature, type of reactors, pressure, residence time, type and rate of fluidizing gas, and catalyst selection. In thermal degradation of plastics, temperature is one of the most significant operating parameters in pyrolysis since it controls the cracking reaction of the polymer chain. Different plastics have different degradation temperature depending on the chemical structure. For common plastics such as PET, HDPE, LDPE, PP and PS, the thermal degradation temperature started at 350°C except for PVC which the degradation started at lower temperature of 220°C. Additionally, the operating temperature required relies strongly on the product preference. If gaseous or char product was preferred, higher temperature more than 500°C was suggested. If liquid was preferred instead, lower temperature in the range of 300-500°C was recommended and this condition is applicable for all plastics [7, 8].

Besides temperature, right reactor selection for the process also crucial to increase efficiency of the reaction towards achieving the final desired product. Most plastic pyrolysis in the lab scale were
performed in batch, semi-batch or continuous-flow reactors such as fluidized bed, fixed-bed reactor and conical spouted bed reactor (CSBR). Each reactor may have its own advantages and disadvantages depending on the application. Batch or semi-batch reactors were likely used in thermal pyrolysis since the parameters could be easily controlled. Nevertheless, these reactors might not be suitable for catalytic pyrolysis because of the potential of coke formation on the catalyst outer surface when they were mixed together with the plastics that would reduce the efficiency of the catalyst. Fluidized bed reactor is concluded to be the best reactor to perform catalytic plastic pyrolysis since the catalyst can be reused many times without the need of discharging, considering catalyst is a very expensive substance in the industry. On the other hand, CSBR also provides good mixing with the ability to handle large particle size distribution and low bed segregation than the bubbling fluidized bed as claimed by Olazar and Lopez [9]. However, a variety of technical challenges during operation of this reactor have been encountered such as its complicated design with many pumps, catalyst feeding, catalyst entrainment and product (solid and liquid) collection that make it less favourable [10].

Furthermore, pressure and residence time also governed the performance of pyrolysis process. Pressure and residence time are both temperature dependence factors that may have potential influence on product distribution of the plastic pyrolysis at lower temperature. Higher pressure increased the gaseous product yield and affected the molecular weight distribution for both liquid and gaseous products but only apparent at high temperatures. Based on the literature, most researchers conducted their plastic pyrolysis studies at atmospheric pressure and focused more on the temperature factor. The residence time was not brought up to attention while carrying out the experiment since the effect would become less apparent at higher temperatures. Moreover, in terms of economic viewpoint, additional units such as compressor and pressure transmitter need to be added into the overall system, thus increase the operation cost if the factor of pressure is considered. However, it should be noted that these two factors should be put under consideration based on the product distribution preference especially when running at temperature below 450°C.

Apart from that, type and rate of fluidizing gas used during pyrolysis also influenced the pyrolysis product. Fluidizing gas is an inert gas (also known as carrier gas) which only engaged in transportation of vaporized products without taking part in the pyrolysis. There are many type of fluidizing gas that can be used for the plastic pyrolysis such as nitrogen, helium, argon, ethylene, propylene and hydrogen. Each type of fluidizing gas has different reactivity based on its molecular weight as reported by Abbas-Abadi et al. [11]. Nevertheless, of all those gases, nitrogen was commonly used by most researchers as fluidizing gas in plastic pyrolysis since it was easier and safer to handle than the high reactivity gas like hydrogen and propylene due to their flammability hazard. In terms of fluidizing gas flow rate, Lin and Yen [12] found that the rate of degradation dropped instantly at the lowest fluidizing flow rate of 300 ml/min. The contact time for primary product is high at lower flow rate, causing the formation of coke precursor (BTX) to increase with the secondary product obtained even though the overall degradation rate is slower [13]. At the highest flow rate of 900 ml/min, the gasoline and hydrocarbon gases fraction were maximized. This clearly shows that the flow rate of fluidizing gas also affected the final product distribution.

As for the catalytic degradation, catalyst is used in the thermal process to speed up chemical reaction and improves the hydrocarbon distribution in order to obtain pyrolysis liquid that had similar properties to the conventional fuel such as gasoline and diesel. The three types of catalysts that are widely used in plastic pyrolysis are zeolites, fluid cracking catalyst (FCC) and silica-alumina catalysts. The usage of zeolite catalyst in pyrolysis of real municipal plastic waste may also help to reduce the impurities in the oil produced and this was proven in the study conducted by Miskolczi et al. [14]. Besides zeolite, the usage of spent FCC catalyst could also improve the plastic conversion in pyrolysis. According to Kyong et al. [15], the amount of solid residue left reduced drastically from 4.5 to 0.9 wt% and this indicates that the conversion was maximized. Other than these two catalysts, some researchers also chose to use silica-
alumina catalyst that is an amorphous acid type. The acid concentration of silica alumina catalyst is determined by the mole ratio of SiO$_2$/Al$_2$O$_3$. Different strength of acidity in catalyst has great influence in the final end product of plastic pyrolysis. Sakata et al. [16] explored the effect of catalysts acidity (SA-1, SA-2, ZSM-5) on the product distribution of HDPE pyrolysis and found that SA-2 catalyst with lower acidity produced higher amount of liquid oil (74.3 wt%), followed by SA-1 (67.8 wt%) and ZSM-5 (49.8 wt%). ZSM-5 possessed strong acid sites, thus produced more gaseous products than the other two acid catalysts but very less liquid yield. Therefore, it is worth noting that that the usage of zeolite catalyst in plastic pyrolysis only maximized the production of volatile hydrocarbon while FCC and silica-alumina maximized the liquid oil production. However, for higher efficiency and longer cycle time usage, HZSM was recommended since the deactivation rate of the catalyst was extremely low and thus, more efficient for regeneration.

3. Liquid fuel properties from pyrolysis of plastics

Table 1 summarized the fuel properties of the liquid oil produced in pyrolysis process. The experimental calorific value of HDPE, PP and LDPE are all above 40 MJ/kg and were considered high for energy utilization. The calorific value of PS was commonly lower than the polyolefin plastic due to the existence of the aromatic ring in the chemical structure which had lesser combustion energy than the aliphatic hydrocarbon [17]. Overall, PET and PVC had the lowest calorific value below 30 MJ/kg due to the presence of benzoic acid in PET and chlorine compound in PVC that deteriorated the fuel quality. Benzoic acid also consisted of aromatic ring that explained the low calorific value in PET. Overall, Table 1 clearly depicted that the physical properties of plastics pyrolysis oil were very close to the properties of commercial gasoline and diesel. Therefore, plastic pyrolysis oil has very high potential to be used as new energy resources.

4. Discussion on plastic pyrolysis scenarios
Table 2 summarized the optimum temperature required to optimize liquid oil yield in thermal and catalytic pyrolysis at different conditions. Other affected parameters include the type of reactors, pressure, heating rate and pyrolysis duration for each type of plastics. All experiments carried out were using nitrogen gas as the fluidizing medium. Based on table 2, PET and PVC are two plastics that produced very low yield of liquid oil in comparison with other plastic types, which made these plastics infrequently explored by researchers. It also should be noted that not all plastic types are recommended for pyrolysis. PVC was not preferred in pyrolysis since it produced the major product of harmful hydrochloric acid and very low yield of liquid oil. Additionally, the pyrolysis oil also contained chlorinated compound that would degrade the oil quality and also toxic to the environment.

As summarized in table 2, it can be concluded that the most effective temperature to optimize the liquid oil yield in plastic pyrolysis would be in the range of 500-550°C for thermal pyrolysis. However, with the usage of catalyst in the pyrolysis, the optimum temperature could be lowered down to 450°C and higher liquid yield was obtained. In most plastics, the usage of catalyst in the process might improve the liquid oil yield, but PS was exceptional. This is because PS degraded very easily without the needs of any catalysts to speed up the reaction and yet 97 wt% of oil was produced [17]. Therefore, PS was the best plastic for pyrolysis since it produced the highest amount of liquid oil production among all the plastics. As for the polyolefin plastic type, LDPE produced the highest liquid oil yield (93.1 wt%), followed by HDPE (84.7 wt%) and PP (82.12 wt%) in thermal pyrolysis. However, with addition of catalyst such as FCC and at the right operating temperature, the liquid yield could be further maximized to above 90 wt%. Besides, the energy potential of pyrolysis oil can also be calculated based on total amount of plastic waste available in Malaysia. For instance, the amount of PS waste available in Malaysia is 280,000 tons/year [25]. Pyrolysis oil obtained from PS manage to contribute 11.68 PJ/year to Malaysia’s energy supply based on the HHV of 43 MJ/kg as shown in Table 1 and the liquid oil production of 97wt% as reported by Onwudili, Insura [17], indicated in table 2.

### Table 2. Summary of studies on plastic pyrolysis.

| Ref. | Type of plastic | Reactor     | Temp (°C) | Pressure | Heating rate (°C/min) | Duratio on (min) | Oil   | Gas   | Solid | Additional information                          |
|------|----------------|-------------|-----------|----------|-----------------------|------------------|-------|-------|-------|-----------------------------------------------|
| [18] | PET            | Fixed bed   | 500       | -        | 10                    | -                | 23.1  | 76.9  | 0     | Stiring rate 200 RPM, FCC catalyst 10 wt%      |
| [4]  | PET            | -           | 500       | 1 atm    | 6                     | -                | 38.9  | 52.1  | 8.9   |                                               |
| [20] | HDPE           | Horizontal steel | 350  | -        | 20                    | 30               | 80.9  | 17.2  | 1.9   |                                               |
| [26] | HDPE           | Semi-batch  | 400       | 1 atm    | 7                     | -                | 82    | 16    | 2     |                                               |
| [27] | HDPE           | Batch       | 450       | -        | -                     | 60               | 74.5  | 5.8   | 19.7  |                                               |
| [28] | HDPE           | Semi-batch  | 450       | 1 atm    | 25                    | -                | 91.2  | 4.1   | 4.7   | Stiring rate 50 RPM, FCC catalyst 20 wt%       |
| [29] | HDPE           | Fluidized bed | 500   | -        | -                     | 60               | 83    | 10    | 5     | Silica alumina catalyst                        |
| [30] | HDPE           | Batch       | 550       | -        | 5                     | -                | 84.7  | 16.3  | 0     |                                               |
| [31] | HDPE           | Fluidized bed | 650   | -        | -                     | 20-25            | 68.5  | 35.1  | 0     |                                               |
| [18] | PVC            | Fixed bed   | 500       | -        | 10                    | -                | 12.3  | 87.7  | 0     |                                               |
| [32] | PVC            | Vacuum batch | 520    | 2 kPa    | 10                    | -                | 12.8  | 0.34  | 28.1  | Also yield HCl=58.2 wt%                        |
| [17] | LDPE           | Pressurized batch | 425 | 0.8-4.3 Mpa | 10                  | 60               | 89.5  | 10    | 0.5   |                                               |
| [33] | LDPE           | Batch       | 430       | -        | 3                     | -                | 75.6  | 8.2   | 7.5   |                                               |
| [4]  | LDPE           | -           | 500       | 1 atm    | 6                     | -                | 80.4  | 19.4  | 0.2   | Also yield wax= 8.7 wt%                        |
| [34] | LDPE           | Fixed bed   | 500       | -        | 10                    | 20               | 95    | 5     | 0     |                                               |
| [30] | LDPE           | Batch       | 550       | -        | 5                     | -                | 93.1  | 14.6  | 0     |                                               |
| [35] | LDPE           | Fluidized bed | 600  | 1 atm    | -                     | -                | 51.0  | 43.2  | 0     | Also yield wax=24.8 wt%                        |
| [20] | PP             | Horizontal steel | 300  | -        | 20                    | 30               | 69.8  | 28.8  | 1.3   |                                               |
| [36] | PP             | Batch       | 380       | 1 atm    | 3                     | -                | 80.1  | 6.6   | 13.3  |                                               |
5. Conclusion

Pyrolysis process was chosen by most researchers among other thermal treatment technologies mainly because of its potential to convert the most energy from plastic waste to valuable liquid oil, gaseous and char. The flexibility that it provides in terms of product preference could be achieved by adjusting the parameters accordingly. The pyrolysis could be done in both thermal and catalytic process. However, the catalytic process provided lower operating temperature with greater yield of liquid oil for most plastics with the right catalyst selection. The sustainability of the process is unquestionable since the amount of plastic wastes available in every country is reaching millions of tons. With the pyrolysis method, the waste management becomes more efficient, less capacity of landfill needed, less pollution and also cost effective. Moreover, with the existence of pyrolysis method to decompose plastic into valuable energy fuel, the dependence on fossil fuel as the non-renewable energy can be reduced and this solves the rise in energy demand.

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| Experiment | Type          | Temperature | Pressure | Time | Yield (wt%) | Oil (wt%) | Coke (wt%) | Gas (vol%) | Reaction Conditions |
|------------|---------------|-------------|----------|------|-------------|-----------|------------|------------|---------------------|
| [26] PP    | Semi-batch    | 400         | 1 atm    | 7    | -           | 85        | 13         | 2          | Stirring rate 200 RPM, used FCC catalyst 10 wt% |
| [11] PP    | Semi-batch    | 450         | 1 atm    | 25   | -           | 92.3      | 4.1        | 3.6        | Stirring rate 50 RPM, used FCC catalyst 10 wt% |
| [4] PP     | Batch         | 740         | -        | -    | -           | 48.8      | 49.6       | 1.6        | Stirring rate 200 RPM, used FCC catalyst, cat/poly=10 w/w |
| [26] PS    | Semi-batch    | 400         | 1 atm    | 7    | -           | 90        | 6          | 4          | Stirring rate 200 RPM, used FCC catalyst, cat/poly=10 w/w |
| [17] PS    | Pressurized batch | 425         | 0.31-1.6 MPa | 10 | 60 | 97        | 2.50       | 0.5        | Used Zn catalyst, Cat/poly=5 w/w |
| [38] PS    | Batch         | 500         | -        | -    | -           | 150       | 96.7       | 3.27       | Used Zn catalyst, Cat/poly=5 w/w |
| [37] PS    | Batch         | 581         | -        | -    | -           | 89.5      | 9.9        | 0.6        | Used Zn catalyst, Cat/poly=5 w/w |

*All experiments used nitrogen gas as fluidizing medium.
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