Plastic Waste Pyrolysis Optimization to Produce Fuel Grade Using Factorial Design

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Abstract. Indonesia is one of the biggest plastic waste production in the world. The government targets to reach 20% for recycling plastic waste in 2019. One alternative to manage plastic waste is using pyrolysis to produce fuel. Pyrolysis is used to degrade the plastic long chain of polymer molecules into smaller molecules. All type of plastic except polyvinyl chloride (PVC) were used in this study to produce fuel. For the purpose, experiment factorial design was used for the optimisation plastic type, residence time, and temperature to maximise the yield of liquid products of the pyrolysis process. In this study 2k factorial design was used for each factor. The result shows that the pyrolysis process used is able to produce diesel like fuel in low temperature.

Keywords: **Plastic Waste; Pyrolysis; DoE; Factorial Design; Fuel**

1 Introduction

Due to its chemical bond making plastic resistant to the degradation process [1]. Plastics have a very long life span. Every year human beings produce close to 280 million tons of plastic. Most of the plastics end up in the environment, damaging marine life and other ecosystems. Plastic waste has damaging environmental and operational problems in landfill because of the slow rate of degradation and toxic dyes and additives. Plastic waste causes fires and releases pollutants that spread to the air and water or acts as a habitat for mice and disease-causing flies [2].

The World Bank What a Waste Study in 2012 estimated the percentage of plastic waste in Indonesia is 12%. The composition of waste in Jakarta has a plastic content including rubber and artificial leather. As illustrated in Figure 1, plastics in municipal water streams has an average composition of plastics in urban waste flows above 31%, ranging from 20% bags, packaging and other types of plastic waste, such as rubber slippers, toys, and glass identified in the waste sample taken from the flow of water, with the most types of plastic found in the form of plastic bags, an average of 16% in all cities. The expert's assessment determined that 30-50% of the collected waste is discharge into the water stream. With the percentage of plastic in the waste mix around 11-14% for urban residents in Indonesia, then the amount of plastic waste present in the water flow is estimated to reach 400,000 tons / year [3].

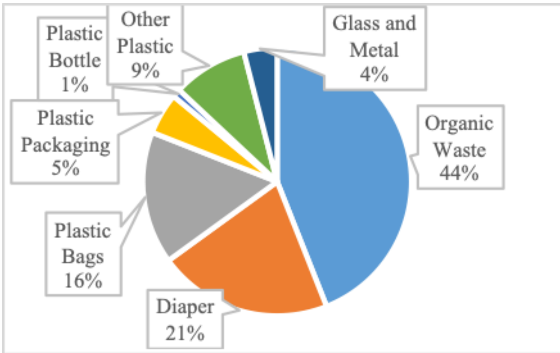

Fig. 1. Average Waste Composition in Water Flow on Sample Hotspots in the City. [3]

There are many studies conducted to convert waste, especially plastic waste into fuel. As done by [1] uses a process of thermal cracking without catalyst or pyrolysis in converting plastic waste into liquid fuel using two types of plastic waste, namely PP and LDPE which are washed and then cut by 5 cm. The results obtained using a temperature of 250 ºC obtained 44.9% oil residue and 53% gas products. The pyrolysis process was chosen by most researchers because of its potential to convert most of the energy from plastic waste into liquid oil, gas and char. According to research conducted by [4] PS is the best plastic for pyrolysis because it produces the highest amount of liquid oil production among all plastics. PS produces liquid oil by 97% by weight. As for the type of polyolefin plastic, LDPE produces the highest liquid oil yield (93.1% by weight), followed by HDPE (84.7% by weight) and PP (82.12% by weight) in pyrolysis. While PET and PP produce the least liquid oil, which is 23.1% by weight and 12.3% by weight. The pyrolysis process is more sustainable because liquid oil can still be produced from plastic blends in raw materials. This has been found by several researchers who conducted pyrolysis research with mixed plastic raw materials. Polyvinyl Chloride (PVC) is not used because it can produce

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chlorine gas and is able to undergo chlorination at a temperature of 250-300 ºC and disrupts the pyrolysis process against other types of plastic [5].

Factorial design has the ability to process several variables and evaluate the interactions between factors, in order to maximize a dependent variable which is function of independent variables. The factorial design also allowed development of mathematical empirical models that can describe the interactive effect of key parameters [6]. This is why factorial design used, to check what if we use variable that different from other study, such as low temperature that only 175 ºC and residence time that over than 180 minute. Using low temperature and long residence time is novelty because just few people study about slow pyrolysis with low temperature. In this study, factorial design was used to determine what factors that have most affect in optimization pyrolysis of the plastic.

2 Literature review

2.1 Type of Plastics

Fundamentally, types of plastics have different compositions that normally reported in terms of their analysis. Type of plastic sold in society that can be reused or recycled with their value in chemical properties is as followed:

2.1.1. Polyethylene Terepthalate (PET)

PET that sold in market has the great choice in packaging for various food products, drinks such as mineral water, soft drink bottle and fruit juice containers. This is due to its properties that are suitable for large-capacity production, lightweight and pressure-resistant containers [7]. [8] Found that higher liquid oil yield could produce at the same operating temperature and pressure. The liquid yield obtained was 39.89 wt%, gaseous was 52.13 wt% and solid residue was 8.98 wt%. Therefore, it can concluded that the liquid from the PET pyrolysis obtained in the ranges of 23–40 wt% while gaseous yield in the ranges of 52–77 wt%.

2.1.2. High-Density Polyethylene (HDPE)

HDPE wastes have a great potential to used in pyrolysis process since it can produce high liquid yield depends on the parameters. Many studies have been conducted on HDPE pyrolysis at different operating parameters to investigate the product yield obtained such as[4]. [9] found that the highest conversion using HDPE happened at 350 ºC with liquid was the dominant product yield (80.88 wt%). The solid was very high at 300 ºC (33.05 wt%) but the amount was reducing to 0.54 wt% at the highest temperature of 400 ºC.

2.1.3. Polyvinyl Chloride (PVC)

Unlike other thermoplastics such as polyethylene (PE), polystyrene (PS) and polypropylene (PP) which can softened by heating and solely derived from oil, PVC is exceptional since it is manufactured from the mixture of 57% chlorine that derived from industrial grade salt and 43% carbon that derived from hydrocarbon feedstock [10]. The research done on the PVC pyrolysis found in the literature was very less due to the dangerous substance that it tend to release when heated at low or high temperature.

2.1.4. Low-desnisty Polyethylene (LDPE)

LDPE has characteristics in long branching that results in weaker intermolecular force, thus lower tensile strength and hardness than HDPE. However, LDPE has better ductility than HDPE since the side branching causes the structure to be less crystalline and easy to molded [5]. [11} Investigated the LDPE pyrolysis in fixed-bed reactor at temperature 500 ºC with heating rate of 10 ºC/min. The experiment done for duration of 20 min and nitrogen used as fluidizing gas. It was observe with high liquid yield of 95 wt% was obtaine with low gas yield and negligible char.

2.1.5. Polypropylene (PP)

PP is a polymer with linear hydrocarbon chain that has a good chemical and heat resistance. PP has a lower density than HDPE but higher hardness and rigidity that makes it preferable in plastic industry [4]. Study conducted by [9] shows that PP pyrolysis within 250-400 ºC produce the highest liquid oil at temperature of 300 ºC around 69.82 wt% with total conversion of 98.66%. The increase in temperature to 400 ºC only reduced the total product conversion to 94.3% and increased solid residue from 1.34 to 5.7 wt%.

2.1.6. Polystyrene (PS)

PS is make of styrene monomers obtained from the liquid petrochemical. The structure consists of a long hydrocarbon chain with phenyl group attached to every other carbon atom. PS is naturally colorless but it can be colored by colorants. It is heat resilience and it offers reasonable durability, strength and lightness that make this polymer desirable to used in variety of sectors such as in food packaging, electronics, construction, medical, appliances and toys [5]. Study of PS by [12] shows that the pyrolysis of PS in a batch pressurized autoclave reactor at 300–500 ºC for one hour duration. The heating rate used was 10 ºC/min and the experimental pressure given was 0.31 MPa up to 1.6 MPa. They found that the PS pyrolysis produced a very high liquid oil yield around 97.0 wt% at optimum temperature of 425 ºC. The largest amount of gas produced was only 2.5 wt%.

2.1.7. Mixed Plastic

The potential of polyolefins mixed plastics in pyrolysis explored by [13] with composition of 46 wt% LDPE, 30 wt% HDPE and 24 wt% PP. The results showed that the liquid obtained was higher at lower temperature of 650 ºC which was around 48 wt%. However, this oil fraction consisted of 52% heavy fraction such as heavy oil, wax and carbon black. There is also studied of mixed plastic in pyrolysis by [5] that shows that addition of PS with PP further reduced the liquid oil yield down to 25% from their individual yields of 80.8 and 42% respectively. Similarly, mixing of PS with PE reduced the liquid yield in comparison to each PS pyrolysis.

2.2 Process Parameter Conditions

Parameters play the major role in optimizing the product yield and composition in pyrolysis process. In plastic pyrolysis, the key process parameters may influence production of final products such as liquid oil, gaseous and char. Those important parameters may summarized as temperature, type of reactors, pressure, residence time, catalysts, type of plastic.

2.2.1 Temperature

Temperature is one of the most significant operating parameters in pyrolysis since it controls the cracking reaction of the polymer chain. When temperature in the system increases, the vibration molecules inside the system will be greater and molecules tend to evaporate away from the surface of the object. This happens when the energy induced by force along the polymer chains is greater than the enthalpy of the C–C bond in the chain, resulted in the broken of carbon chain [14].

It was proven that the temperature has the greatest impact on reaction rate that may influence product composition of liquid, gaseous and char for all plastics. The operating temperature required relies strongly on the product preference. [4] state that if gaseous or char product preferred, higher temperature more than 500 ^{o}C suggested. If liquid was prefer instead, lower temperature in the range of 300–500 ^{o}C was recommended and this condition is applicable for all plastics.

2.2.2 Type of Reactor

The type of reactors has important impact in the mixing of the plastics and catalysts, residence time, heat transfer and efficiency of the reaction towards achieving the final desired product. Most plastic pyrolysis in the lab scales performed in batch, semi-batch or continuous-flow reactors such as fluidized bed, fixed-bed reactor and conical spouted bed reactor (CSBR).

2.2.3 Pressure

Pressure affected the carbon number distribution of the liquid product by shifting the lower molecular weight side when it was high. Pressure also had a significant effect on the rate of double bond formation. the rate of double bond formation decreased when pressure increased and this suggested that pressure directly affected the scission rate of C–C links in polymer [15].

2.2.4 Residence Time

Residence time can defined as average amount of time that the particle spends in the reactor and influence product distribution. Longer residence time increases the conversion primary product, thus more thermal stable product is yield such as light molecular weight hydrocarbons and non-condensable gas [4]. It seems that the longer feed or plastic intact with heat the product will be higher in yield of product.

2.2.5 Catalysts

Catalyst speeds up chemical reaction but remains unchanged in the end of the process. Catalysts are widely used in industries and researches to optimize product distribution and increase the product selectivity. When catalyst used, the energy of the process is lowered, thus speeds up the rate of reaction. Therefore, catalyst reduces the temperature that required in the process and this is very crucial since the pyrolysis process requires high energy that hinders its commercial application. The usage of catalyst may help in saving energy as heat is one of the most expensive costs in industry [4].

2.3 Design of Experiment (DoE)

Design of Experiment is the process of planning an experiment to produce the proper data and analyzed using statistical methods to produce valid, objective, and meaningful conclusions.

According to Joppert [6], Factorial Design and Response Surface Methodology (RSM) to predict the effect of experimental conditions on product yields formed from pyrolysis of waste mixtures has used to optimize reaction time, temperature and initial pressure, to maximize yield and composition of liquid yield from plastic mixtures. Use of RSM to predict the results of pyrolysis with a large reduction in the number of trials. This methodology also proved useful for supplying information to optimize experimental conditions that maximize the production predetermined gas and liquid compounds [16].

Factorial Design is a powerful analytical tool for modeling and analyzing the effect of process variables on certain variables, which are unknown functions of pyrolysis process variable [17]. The use of factorial design can integrate experimental design and data analysis that offer a results optimization approach and have the advantage of reducing the number of experiments by choosing the right experimental conditions [6]. Full Fractional Factorial Design allows checking the effects of each input factor on the test output, also allows for investigation of the interaction effects between various input factors [18].

3 Research methodology

This study uses a reactor made of stainless steel with a length, width, and height as 60 cm, 69.5 cm, and 77 cm, respectively. Operating conditions used were 175 and 250 ^{o}C and residence time of 180 and 300 minutes. Plastic waste used were homogeneous plastic of PP and heterogeneous type of plastic consisting of PET, LDPE, HDPE, PP and PS. PVC is not used as raw material because during the pyrolysis process it can produce dangerous chlorine gas. Products in the form of liquid, char or tar, and gas are then weighed to find the mass, besides that, the density, viscosity and number of the test will also be tested. Data analysis results on liquid

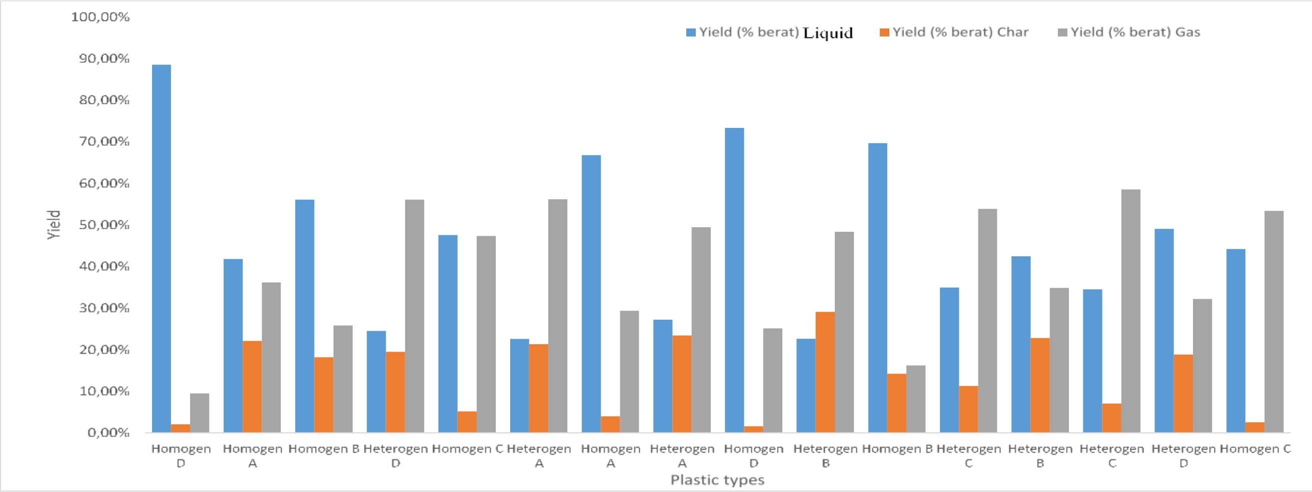

Fig. 2. Yield From Pyrolysis Plastic Waste

yields were then processed using ANOVA with the help of Minitab 17 to determine the optimum conditions for pyrolysis of plastic waste into oil.

Heterogeneous plastics or mixtures of various types of plastic used in this study were selected or taken from plastic waste in the University of Indonesia's Faculty of Engineering. Plastic waste used is a mixture of PET plastic types as much as 22%, LDPE as much as 12%, HDPE as much as 16%, PP as much as 31%, PS as much as 13%, and others that are not identified as 6%. The plastic waste that has an aluminum mixture is not used because in the pyrolysis process it interferes with the process and inhibits the plastic from reacting to oil.

3.1 Factorial Design

The pyrolysis process is designed according to three parameters used; temperature, residence time, and type of plastic. The design was carried out according to the 2k factorial design and carried out randomly or randomized with the help of Minitab 17 as in Table I.

Table 1. Process randomization

NO.	RUN	BLOCK	PLASTIC TYPE	TEMPERATURE	RESIDENCE TIME
16	1	1	1	1	1
10	2	1	1	-1	-1
6	3	1	1	-1	1
15	4	1	-1	1	1
4	5	1	1	1	-1
9	6	1	-1	-1	-1
2	7	1	1	-1	-1
1	8	1	-1	-1	-1
8	9	1	1	1	1
5	10	1	-1	-1	1
14	11	1	1	-1	1
3	12	1	-1	1	-1
13	13	1	-1	-1	1
11	14	1	-1	1	-1
7	15	1	-1	1	1
12	16	1	1	1	-1

In the type of plastic, temperature, and residence time there are values of -1 and 1 which are variables used, both quantitative and qualitative which indicate low or high. The level of variables used in this study is shown in Table II.

Table 2. Variable level

Factor	Parameter	-1	1
A	Plastic Type	Heterogeneous	Homogeneous
B	Temperature (°C)	175	250
C	Residence Time (minute)	180	300

This research is to know what if high and low temperatures, long and short residence times, homogeneous and heterogeneous types of plastic so that the desired results obtained.

3.2 Product Characterization

By using the pyrolysis method, homogeneous and heterogeneous plastic waste materials will decompose into several products. Products produced such as liquids will look for the following characteristics.

- Mass Equilibrium
- Viscosity
- Density
- Acidic Liquid Content
- Water Content

4 Result

Figure 2. shows that the biggest oil yield is obtained at homogeneous, with temperatures of 250 °C and a residence time of 300 minutes, obtained 88.5% by weight. While for char and gas obtained 2.03% and 9.47% by weight. While in homogeneous with a temperature of 175 °C, the highest liquid yield of 69.61% was obtained at 300 minutes of residence with char and gas yields obtained at 14.20% and 16.19% by weight. As research conducted by [9] which states that in the process of plastic pyrolysis using PP plastic with an increase in temperature from 250 to 300 °C, the results of liquid products increased from 57.27 to 69.82%. This proves that in this study with increasing temperatures from 175 to 250 °C liquid yield increased 69.61 to 88.5%.

4.1 Liquid Characterization

The results of oil characterization obtained were obtained from the homogeneous and heterogeneous plastic waste pyrolysis process. Oil or liquid products at temperatures of 175 and 250 °C are then characterized based on fuel standards such as diesel and premium at the institutions. The results of the oil test are obtained as shown in table III. And then compared to the standards in Indonesia.

Table 3. Variable level

Properties	Plastic Type		Diesel Standard
	Homogenous	Homogenous	
Cetane Number	48.1	48.3	Min 48
Acidic Value (mgKOH/gr)	43.88	4.04	Max 0.6
Viscosity 40 °C (cst)	2.28	2.4896	2 – 4,5
Density 15 °C (kg/m^3)	805.4	806	Max 860
Water Content (mg/kg)	18.6	271.6	Max. 500
Ash Content (%v/v)	1	1	Max.0,01

Density is one of the important parameters for all petroleum products [5]. Because if the density value of a fuel is high then the fuel consumption will be reduced while if the density is too low it will consume more fuel which can damage the engine. Density of the type of diesel fuel produced by homogeneous pyrolysis b and d is 805.4 and 806 kg / m3. The value obtained does not exceed the maximum density value of diesel 48 (Diesel) of 869 kg / m3 according to Indonesian diesel / biodiesel standards.

Kinematic viscosity is the fuel characteristic that determines the spray pattern and atomization of the fuel injected in the combustion chamber. High viscosity oils provide poor atomization in the engine, which makes the engine performance less good [5]. This viscosity can also act as a lubricant from the fuel injection system. According to [19], liquids produced by pyrolysis with plastic raw materials are classified into three groups, namely gasoline fraction (C5-C12), diesel fraction (C13-C20) and heavy oil (> C20). These three fractions also determine the high and low viscosity values of a liquid oil.

Kinematic viscosity values of homogeneous pyrolysis and are 2.28 and 2.489 cst. The liquid from homogeneous plastic pyrolysis has a lower viscosity value than diesel fuel, which is the kinematic viscosity value of diesel 48 (Diesel) with a maximum of 4.5 cst in accordance with Indonesian diesel standards.

The oil liquid produced from homogeneous plastic waste has a high ash content, which is 1% v / v, which is very high when compared to the Indonesian solar standard which has a maximum ash content of 0.01% v / v. The large ash content can affect the performance of the diesel engine because it can disrupt the combustion process that occurs. High ash content indicates the possibility of metal contamination in liquid oils [4].

The acid content in the liquid oil from homogeneous plastic pyrolysis b and d is 43.88 and 4.04 mgKOH/gr, while the standard diesel 48 (diesel) is 0.6 mgKOH/gr. High acid content can accelerate corrosion that occurs in the engine. The water content in the pyrolysis liquid is 18 and 271.6 mg / kg while the maximum standard water content of 48 (diesel) diesel Indonesia is 500 mg/kg. The water content is quite good because the presence of high water content in liquids will affect the performance of diesel engines [19].

4.2 Factorial Design

The liquid yield data that has been obtained, then ANOVA test is performed using the help of the Minitab application 17. The results obtained consist of a normal probability graph, the values of F and P on the F table and significant values. Then explain the results of ANOVA so that it is known what parameters have a significant effect on the yield of liquid by using three factors or parameters, namely the type of plastic, temperature, and residence time.

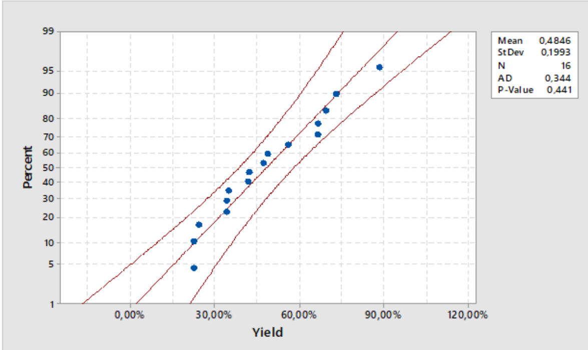

Fig. 3. Normal Probability Graphic

Figure 3 shows that all points are close to a straight line or do not come out of the outermost line or often referred to as a fat pencil test so that it can be said that all data is normal and calculations can be done by referring to the normal distribution pattern. By knowing that the data obtained is normally distributed so that it can determine the confidence interval, in this study it was 95%.

From the results of the Pareto graph illustration in Fig. 4. Shows that the length of each bar in the chart shows the standard effect of the factor on the response. Plastic type and residence time factors significantly influence the yield of liquid yields. However, the biggest influence is indicated by the type of plastic that has the greatest value from other factors. From Fig. 4. It is known that the smallest interaction is in AB which is the interaction between the type

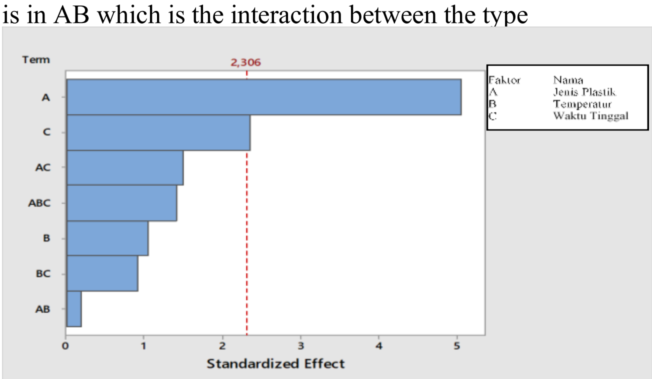

Fig. 4. Pareto Graphic of Standardized Effect

of platinum and temperature, this occurs because the temperature difference is not too different, namely at temperatures of 175 and 250 °C.

4.3 Optimization Pyrolysis Process

From the results of the fluid yield response that has been obtained, then the optimal reaction can be sought so that the highest liquid yield can be achieved on the tool used. Optimization of the pyrolysis tool uses help from Minitab 17 as shown in Fig. 5.

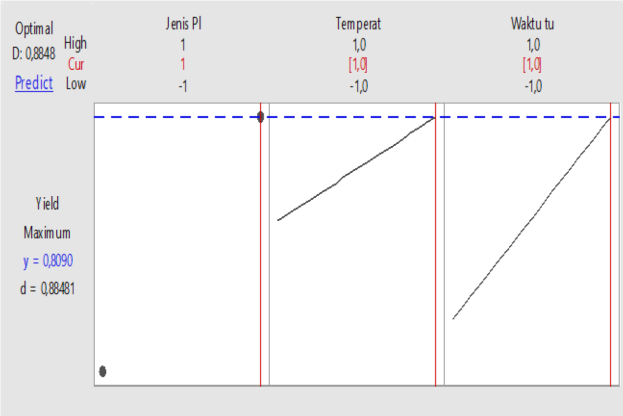

Fig. 5. Optimization Plot Pyrolysis Process

From the optimization chart above, it shows that if the expected yield is 80.9% by weight, the optimal setting for the plastic pyrolysis process to reach this amount is to use plastic types: homogeneous or PP, temperature: 250 oC and residence time: 300 minutes.

Liquid oil from pyrolysis of plastic waste contains some impurities such as sulfur, chlorine, solid residue, moisture, and acid. The presence of this dirt not only lowers the quality of liquid oil but also limits its commercial applications. Therefore, liquid oil requires further maintenance including an increase by removing charcoal particles, and acid removal and neutralization to increase liquid oil with a stable pH and low corrosivity.

The procedure for increasing liquid pyrolysis oil depends on the application being targeted. There are two ways reported for increasing liquid oil, including refining and mixing with conventional diesel to be suitable for a variety of commercial applications [5]. The ways that can be done to improve the quality of oil from plastic pyrolysis are as follows in table IV.

Table 4. Improving liquid oil quality

Problems	Proses	Methods
Acid Contamination	Amine Treating	Absorpsion
Sulfur and Water	Sweetening and Drying	Absorpsion atau Termal
Colour and Viscosity	Hydrotreating	Hidrogenasion Catalytic
Fuel Oil (*Unsaturated*)	Solvent Extraction	Absorpsion or Presipitasion
Contaminant	Desalting	Dehidrasion via absorpsion

5 Conclusion

Implementation of the proposed pyrolysis process for plastic waste could be the answer to the problem of plastic waste in most countries, including Indonesia. The biggest oil yield was obtained at homogeneous d, with a temperature of 250 °C and a waiting time of 300 minutes obtained 88.5% by weight. While for char obtained was 2.03% by weight. The biggest oil yield was found in heterogeneous d at a temperature of 250 °C and a residence time of 300 minutes at 49% by weight. While for char was obtained 18.81 by weight. The factorial design of the pyrolysis process shows that the significant factors are the type of plastic and the waiting time with a calculated F value greater than F table. Tool optimization can be done to get liquid yield of 80.90% using homogeneous plastic or PP, temperature of 250 °C, and residence time of 300 minutes. Optimization can be done by distilling and mixing with conventional diesel to be suitable for a variety of commercial applications. The resulting liquid can be used with consequences for the engine corrosion due to acid numbers and high ash content in the liquid.

This work has been supported partially by PITTA 2019 Grant funded by DRPM Universitas Indonesia under contract No: NKB-0731/UN2.R31/HKP.05.00/2019

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