World Chemical Engineering Journal Vol.8, No.1, (2024), pp. 26 – 33

WCEJ

WORLD CHEMICAL ENGINEERING JOURNAL

Journal homepage: http://jurnal.untirta.ac.id/index.php/WCEJ

The Influence of Natural Bayah Zeolite on the Pyrolysis Process of Liquid Fuel Based on HDPE and PP Plastic Waste

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ARTICLE HISTORY

Received 23 June 2024
Received in revised form 25 June 2024
Accepted 26 June 2024
Available online 29 June 2024

ABSTRACT

Pyrolysis is a decomposition reaction method involving the heating of a material with little or no oxygen. The objectives of this research are to utilize plastic waste for the production of liquid fuel and determine the optimal conditions for maximizing liquid fuel yield. The pyrolysis method was used at a temperature of 350°C for 300 minutes with High-Density Polyethylene (HDPE) and Polypropylene (PP) plastic as raw materials in composition variations of 7:3, 5:5, and 3:7, and the Bayah natural zeolite catalyst was activated and varied in amounts of 0%, 3%, and 5%. The analysis included yield tests, density tests, viscosity tests, calorific value tests, and the composition analysis of the liquid product yield. The results of this research indicated that the highest liquid product yield was obtained with a composition of 30% HDPE, 70% PP, and 0% catalyst, achieving a yield of 66.4%. It was concluded that the activated Bayah natural zeolite catalyst was not sufficiently effective in the pyrolysis process at a temperature of 300°C. The highest density and viscosity values were obtained with a composition of 70% HDPE, 30% PP, and 0% catalyst, which were 0.764 g/cm^3 and 0.789 cP, respectively. The highest calorific value was obtained with a 50% HDPE and 50% PP composition, reaching 10,978.8 Cal/g. The composition analysis of the liquid product yield for a 70% HDPE and 30% PP composition resulted in 42% gasoline and 58% kerosene. For a 30% HDPE and 70% PP composition, the yield was 30% gasoline, 62% kerosene, and 8% diesel.

Keywords: *Liquid Fuel, HDPE, Pyrolysis, Polypropylene, Zeolite.*

1. INTRODUCTION

Plastic has become one of the most commonly used materials in both industrial and commercial needs. Its properties, such as being lightweight, waterproof, rust-resistant, shatterproof, strong, and relatively economical, make it attractive to both producers and consumers. Due to its massive usage, plastic has become the second-largest waste after food waste, accounting for 18.68% of household waste in Indonesia (Ministry of Environment and Forestry, 2022).

The government's efforts to reduce plastic waste have included implementing regulations to ban the use of plastic bags in modern retail outlets. As a result, by 2020, 41 local governments had implemented this policy, notably reducing plastic waste in Banjarmasin (Khairunisa, 2020). However, this result has not been sufficient to significantly reduce the total plastic waste polluting the environment. Approximately 60-80% of marine debris consists of plastic waste (Leahy, 2004).

In 2022, to manage plastic waste, Tahdid and colleagues conducted research on plastic waste processing using the pyrolysis method. The results showed a liquid fuel yield of 19.01% using LDPE plastic and a zeolite catalyst in a 1:15 ratio, producing specifications similar to gasoline and diesel (Tahdid, Harlivia, et al., 2022). Berlian and colleagues conducted similar research, resulting in a yield of 25.76% with liquid fuel compositions including gasoline, kerosene, diesel, and lubricating oil using a mixture of PP and PS plastics as raw materials (Berlian et al., 2022). Kartika conducted the same research, yielding oil quality that met diesel fuel standards using a mixture of HDPE and PET plastics with a zeolite catalyst ratio of 2:5 (200 grams) (Kartika, 2022). Furthermore, Liu et al. (2024) studied the pyrolysis process using microwave methods to convert organic waste, such as plastic waste, into products with added value. The research results showed that microwave-assisted plastic pyrolysis produces liquid fuel with high added value. The advantages of microwaves include the ability to heat materials evenly, achieve rapid and selective heating, and offer high production efficiency as well as low energy consumption (Liu et al., 2024). In addition, Hegedüs et al., (2024) studied the pyrolysis process in a batch reactor equipped with reflux and a purification process through distillation. The results showed that the mass ratio of plastic materials used in the pyrolysis process significantly affects the feasibility of oil products as a transportation fuel. An optimal mixture of plastic waste combined with olefin content saturation through catalytic hydrogenation can produce standard-quality gasoline (Hegedüs et al., 2024). Based on these findings, further research is necessary to achieve a better oil yield.

2. METHODS

2.1 Material Preparation

The first procedure in the study on the influence of catalyst variations in the pyrolysis of plastic for fuel production is material preparation. The materials used are waste plastics obtained from the Cilegon area. The initial step involves sorting the plastic waste to obtain PP (polypropylene) and HDPE (high-density polyethylene) plastics. Next, the plastic waste is cleaned of any dirt and then dried by exposing it to sunlight for 2 days. After drying, the plastic waste is shredded into pieces approximately 2-5 cm in size.

2.2 Activation of Bayah Natural Zeolite Catalyst

Natural Bayah Zeolite is activated by grinding and sieving the zeolite until it reaches a size of 60 mesh. Subsequently, the sample is immersed in distilled water and stirred at room temperature for 8 hours, then allowed to settle for one day. The formed sediment is then filtered, followed by drying in an oven at 120°C for 2 hours. Next, the zeolite is soaked for 24 hours at room temperature in a 1M NH4Cl solution (Hendrawati et al., 2022). Afterward, the zeolite is filtered and washed with distilled water. Then, the zeolite is dried again in an oven at 120°C. Once cooled, place the zeolite in a porcelain crucible and calcine it at 450°C for 8 hours in a furnace.

2.3 *Pyrolysis*

In the pyrolysis process of plastic waste, a mixture of PP (polypropylene) and HDPE (high-density polyethylene) plastics is used, totaling 250 grams of plastic waste. The pyrolysis is conducted in a closed system (no air ingress) using a batch reactor at a temperature of 350°C for 300 minutes. After the pyrolysis process, the resulting product is in the form of gases, thus requiring condensation. During the condensation process, the gases produced are converted into liquid form.

2.4 Yield Test

The yield test is used to determine the percentage of the total products obtained from the material used. The percentage yield of the pyrolysis products can be calculated using the following equations (Azubuike A. et al., 2018):

$$\boldsymbol{Yliquid = \frac{M2}{M1} \times 100\% \ldots (1)}$$

$$\boldsymbol{Yresidu = \frac{M3}{M1} \times 100\% \ldots (2)}$$

$$\boldsymbol{Ygas = 100\% - (Yliquid + Yresidu) \ldots (3)}$$

Where:
M1: Mass of the sample,
M2: Mass of liquid products,
M3: Mass of residue.

2.5 Density Test

In density testing, the first step involves weighing the mass of the pyrolysis oil using a balance and measuring its volume. Density can be calculated using the following equation (Badrul W., 2020):

$$\boldsymbol{D = \frac{m}{v} \ldots (4)}$$

Where,
D = Density of pyrolysis oil (g/mL),
m = Mass of pyrolysis oil (g),
v = Volume of pyrolysis oil (mL).

2.6 Viscosity Test

In viscosity testing, the first step is to pour the sample solution into an Ostwald viscometer. Next, the sample is drawn into the bulb up to the upper mark. Then, the bulb is slowly released and the other hole on the Ostwald viscometer is covered. Subsequently, the hole is opened, and the time taken for the sample to descend from the upper mark to the lower mark is recorded. Viscosity can be calculated using the equation (Cantika R., et al. 2022).

$$\boldsymbol{Kinematic\ Viscosity\ (cSt) = C\left(\frac{mm^2}{sec^2}\right) \times t(\mathbf{sec}) \ldots (5)}$$

$$\boldsymbol{Dynamic\ Viscosity\ (cP) = Kinematic\ Viscosity(cSt) \times density \ldots (6)}$$

2.7 Calorific Value Test

The calorific value of a fuel refers to the total heat energy that can be released by the fuel through complete combustion per unit mass or volume of the fuel (Elwina, et al., 2022). This test is conducted to determine the calorific value generated from the pyrolysis process. The characteristics of the pyrolysis products are tested using a bomb calorimeter to measure this value. In calorific value testing, we provide samples to a third party to determine the calorific value of the tested samples.

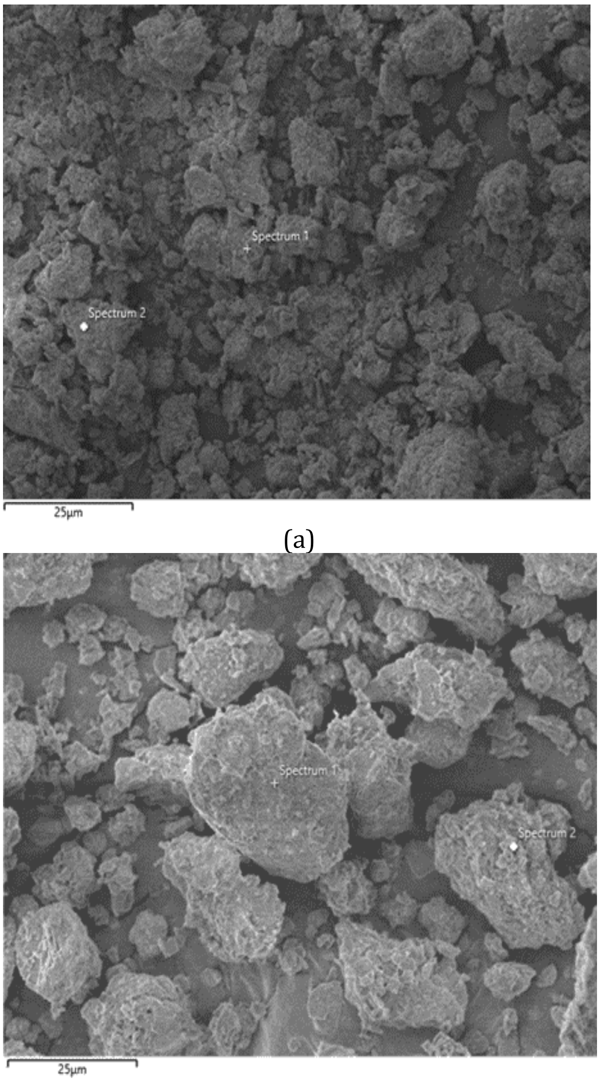
2.8 GCMS (Gas Chromatography Mass Spectrometry) Test

Analysis of the content of pyrolysis oil using GC-MS (Gas Chromatography-Mass Spectrometry) instrument. The GC (Gas Chromatography) method involves separating a mixture into its components based on interactions between the mobile phase and stationary phase. Subsequently, the sample in vapor form is carried by the gas flow into the separating column. The results of the separation are analyzed using a chromatogram (Mues L., 2019). In GC-MS testing, we provide samples to a third party to determine the content analysis of the tested samples.

3. RESULT AND DISCUSSION

3.1 The Influence of Activating Natural Bayah Zeolite Catalyst on Morphology

A comparison of the catalyst surface before and after activation observed using the SEM method is presented in Figure 1.



(a)

(b)

Fig, 1. Surface of Catalyst Before Activation (a) and After Activation (b).

In Figure 1, it is shown that the catalyst before activation has a denser surface compared to the catalyst after activation. This indicates that the activation method applied has achieved its intended purpose of enlarging the catalyst's pore surface area. This observation is further supported by the composition of compounds present in the zeolite. The compositions of the compounds contained in the zeolite before and after activation are presented in Table 1 and Table 2:

Table 1. Compound Composition in Zeolite Before Activation

Spektrum 1				
Element	***Line Type***	***Weight %***	***Weight% Sigma***	***Atomic%***
O	*K Series*	43.32	0.86	58.17
Na	*K Series*	0.17	0.12	0.16
Mg	*K Series*	0.64	0.13	0.57
Al	*K Series*	9.13	0.26	7.27
Si	*K Series*	39.65	0.80	30.33
K	*K Series*	1.51	0.81	0.83
Ca	*K Series*	3.51	1.48	1.88
Fe	*K Series*	2.07	0.69	0.80
Ni	*K Series*	0.00	0.29	0.00
Total		100.00		100.00

Source: Results of EDX Test from Integrated Laboratory of Sultan Ageng Tirtayasa University

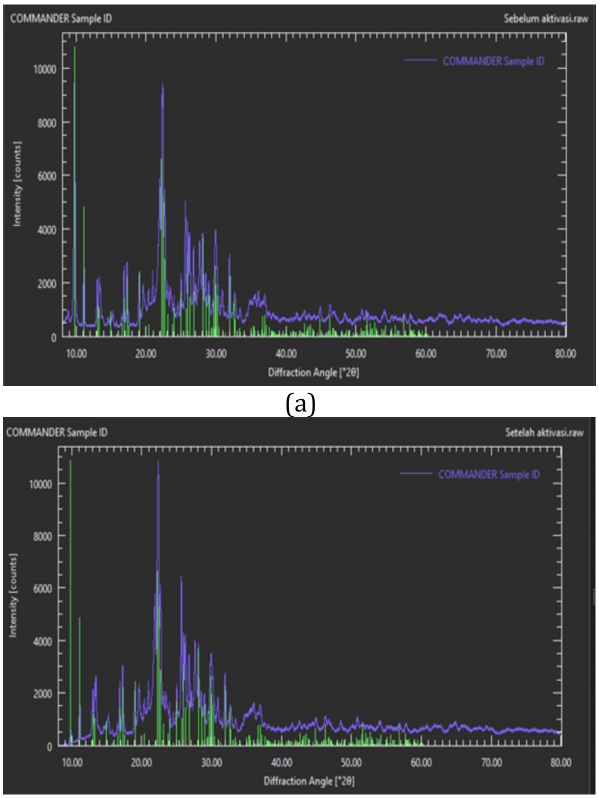
Table 2. Compound Composition in Zeolite After Activation

Spektrum 1				
Element	***Line Type***	***Weight %***	***Weight% Sigma***	***Atomic%***
C	*K Series*	3.38	0.43	5.82
O	*K Series*	44.14	0.77	57.17
Na	*K Series*	0.02	0.09	0.02
Mg	*K Series*	0.49	0.10	0.42
Al	*K Series*	6.63	0.19	5.09
Si	*K Series*	38.22	0.68	28.19
K	*K Series*	0.80	0.67	0.42
Ca	*K Series*	3.54	1.26	1.83
Fe	*K Series*	2.73	0.56	1.01
Ni	*K Series*	0.05	0.23	0.02
Total		100.00		100.00

Source: Results of EDX Test from Integrated Laboratory of Sultan Ageng Tirtayasa University

The table above shows that the Si/Al content decreased by 3% for Al content and 1% for Si content. This indicates that the activation method can reduce the Al and Si content in zeolite, thereby effectively enlarging the pores in zeolite after activation. In this study, natural Bayah zeolite was used. XRD analysis was conducted to determine the type of zeolite used. The results of the XRD analysis processed using Profex 5.0 software can be seen in Figure 2.

Based on Figure 2, it can be observed that natural Bayah zeolite exhibits crystallization patterns similar to those of clinoptilolite type. This indicates that natural Bayah zeolite belongs to the category of clinoptilolite zeolites.



(a)

(b)

Fig.2. Results of XRD Test Before Activation (a) and After Activation (b).

3.2 The Effect of Catalyst Quantity on Liquid Yield Yield.

The results of liquid yield yield with variations in the amount of natural Bayah zeolite catalyst at 0%, 3%, and 5% are presented in Figure 3.

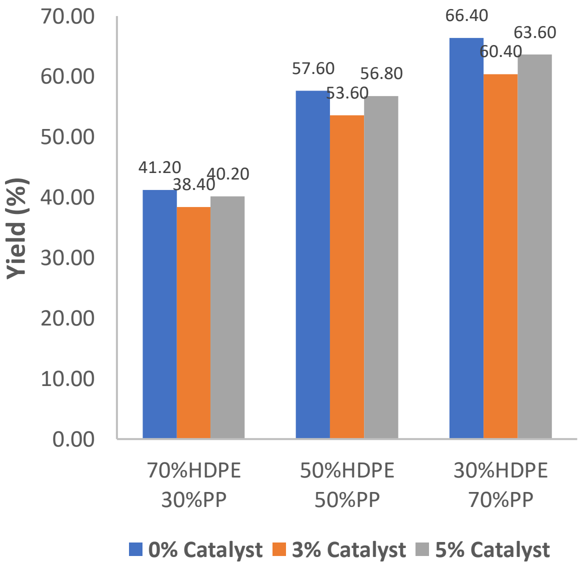


Fig. 3. The Effect of Catalyst Quantity on Liquid Yield Yield.

Figure 3 shows that each composition condition of plastic exhibits the highest yield variation at 0% catalyst, 5% catalyst, and 3% catalyst. This indicates that the activated natural Bayah zeolite catalyst is not sufficiently effective in the pyrolysis process at a temperature of 300°C. This could be attributed to the specific properties of catalysts, where catalysts have specific characteristics that can influence their type and amount of their usage. A catalyst that is not suitable for the reaction can become a toxic catalyst that inhibits the reaction process (Herizal, 2020).

The decrease in liquid yield after using the catalyst in the reaction is due to the mismatch of the type and amount of catalyst in the pyrolysis process conducted, where catalysts have specific properties. Therefore, it can be concluded that the varied amount of natural Bayah zeolite catalyst used was not suitable for the pyrolysis reaction.

3.3 The Effect of Plastic Composition on Liquid Yield Yield

The results of liquid yield yield with variations in plastic composition are presented in Figure 4.

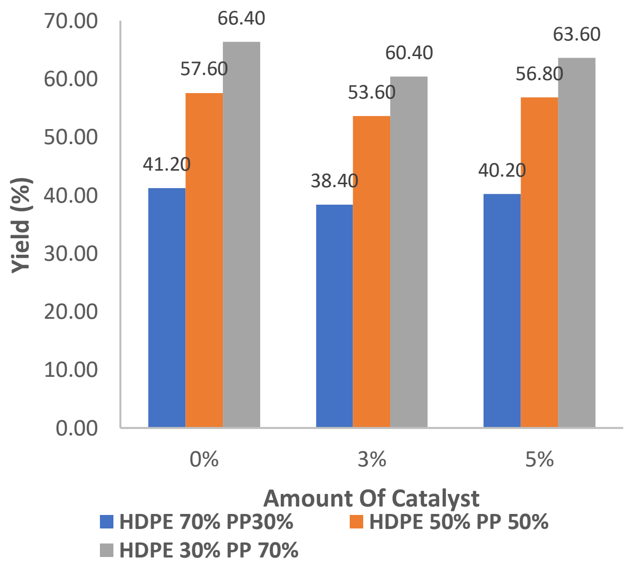


Fig. 4. The Influence of Plastic Composition on Liquid Yield Yield

Figure 4 shows that in each variation of catalyst amount, a similar trend is observed, where plastic compositions with a higher proportion of HDPE yield less liquid yield compared to those with a higher proportion of PP in the pyrolysis process. This could be attributed to the different melting points of the two materials. According to Xometry (2022), HDPE has a melting point of 180-205°C, while PP has a melting point of 160-168°C. From this data, it is evident that HDPE has a higher melting point compared to PP. Plastics with lower melting points will melt earlier than those with higher melting points when heated together. In this case, PP plastic will start to melt before HDPE reaches its melting point, resulting in a higher yield of liquid products in the plastic composition of 70% PP and 30% HDPE.

This aligns with the theory that plastics have different compositions of constituent compounds. The differences in these compound compositions affect the melting points and boiling points of the liquid phase of the reacted plastics (Lanang et al., 2020). This influences how quickly plastics undergo the reaction to become liquid yield. At the same reaction time and temperature, the plastic composition affects the liquid yield produced from the pyrolysis process. Furthermore, the liquid yield can also be influenced by the thickness of both types of plastics. HDPE typically has a thicker thickness compared to PP (Lanang et al., 2020). The thickness of the material affects the time required to reach the desired melting temperature because thicker materials tend to have uneven heat distribution. Thicker materials require more time to reach the melting temperature due to the greater heat energy needed to heat thicker materials (Anwar et al., 2020).

3.4 The Effect of Plastic Composition on the Density of Liquid Yield

The density values obtained for each variation of plastic composition can be seen in Figure 5.

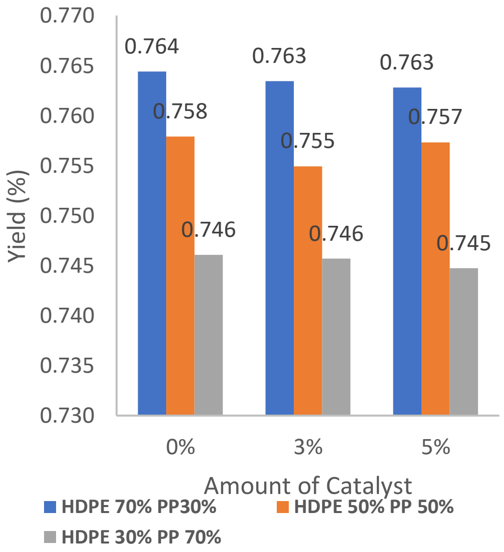


Fig.5. The Effect of Plastic Composition on the Density of Liquid Yield

Based on Figure 5, it can be seen that the density values obtained for each variation of catalyst amount show a similar trend, where compositions with more HDPE compared to PP have higher density values. This aligns with the fundamental properties of HDPE and PP, where HDPE plastic (0.94 g/cm^3) has a higher density value compared to PP plastic (0.90 g/cm^3) (Deglas, 2023).

Density represents the specific gravity of a compound, and in the context of fuels, it has standard values that categorize it as a liquid fuel. According to the decision of the Director General of Oil and Gas Number 0177-K/10/DJM.T.2018, the density value for gasoline fuel ranges from 0.715 to 0.770 g/cm^3. Based on the results obtained, the density values of the pyrolysis products meet the standard.

3.5 The Effect of Plastic Composition on the Viscosity of Liquid Yield

The viscosity values obtained for each variation of plastic composition can be seen in Figure 6. Based on

Figure 6, it can be observed that the viscosity values obtained show that compositions with more HDPE have higher viscosity compared to compositions with more PP. The highest viscosity is obtained with the variation of HDPE 70%, PP 30%, and 0% catalyst, which is 0.789 cP. On the other hand, the lowest viscosity is observed with the variation of HDPE 30%, PP 70%, and 5% catalyst, which is 0.689 cP. According to the literature, natural gasoline has a viscosity of 0.65± cP, and kerosene has a viscosity of approximately 0.79 cP (Purwanti L., 2015). From this data, it can be inferred that the pyrolysis oil falls within the viscosity range of gasoline and kerosene.

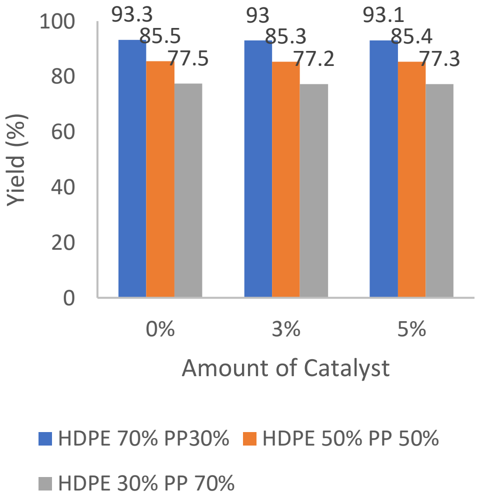


Fig.6. The Effect of Plastic Composition on the Viscosity of Liquid Yield

Viscosity, as described by Dominggus G.H., et al. (2016), is also influenced by density, where higher density leads to higher viscosity. This occurs because higher density means more particles are present in the fluid, which impedes flow due to inter-particle friction. Based on Figure 4.4 and Figure 4.5, it can also be seen that density and viscosity are directly proportional. This correlation aligns with the theory that viscosity is affected by density; higher-density fluids tend to have higher viscosity (Dominggus G.H., et al., 2016).

3.6 The Effect of Plastic Composition on the Calorific Value of Liquid Yield

The calorific values obtained for various plastic composition variations can be seen in Table 3.

Table 3. Compound Composition in Zeolite Before Activation

Variations	Heating Value (Cal/g)
HDPE 70% PP 30%	10863.9
HDPE 50% PP 50%	10978.8
HDPE 30% PP 70%	10961.4

Based on Table 3, the calorific values obtained are not significantly different from the range of calorific values for gasoline, which typically range from 10050.1 to 11441.5 Cal/g (Novandi, 2018). It can be concluded that the calorific value of the pyrolysis products still falls within the range of gasoline calorific values. The calorific value of gasoline itself represents the amount of energy released by burning gasoline. The higher the calorific value of gasoline, the more energy is released during combustion (Novandi, 2018). Therefore, gasoline with a higher calorific value can provide better engine performance and higher fuel efficiency. The calorific value testing of the pyrolysis products was analyzed by the Technical Implementation Unit (UPT) Laboratory of the Department of Engineering at the University of Indonesia using qualitative methods with a Parr Bomb Calorimeter 6400. In addition, The hydrocracking of LDPE plastic into fuel using bifunctional catalysts was studied by Sriningsih et al., (2014). The study uses zeolite from Sukabumi, Indonesia. It was activated by refluxing in 6N HCl and drying, resulting in mordenite-type zeolite. Bifunctional catalysts were created by loading Ni, Ni-Mo, Co, and Co-Mo metals into the activated zeolite. These catalysts were heated in a microwave and reduced with hydrogen gas to produce Ni/Z, Ni-Mo/Z, Co/Z, and Co-Mo/Z. The hydrocracking process was conducted at 350 °C with a hydrogen gas flow of 20 mL/min for 1 hour. The conversion of LDPE with Co-Mo/Z yielded the highest gasoline selectivity, reaching 71.49%. GCMS data showed that the liquid product consisted of hydrocarbon compounds with carbon chains ranging from C6 to C19, including paraffins, olefins, and naphthenes. This technology is expected to reduce environmental pollution, support land use, and increase energy storage (Sriningsih et al., 2014). In addition, Hendrawati et al., (2023) studied the pyrolysis of PP and HDPE samples with the addition of catalysts at 7% and 3% (w/w), yielding oil with liquid fractions of 69.69% and 65.60%, respectively. The use of natural zeolite catalysts in the pyrolysis of PP and HDPE from plastic waste offers a simple method and demonstrates reliable catalyst performance (Hendrawati et al., 2023).

3.7 Composition of Liquid Yield in Plastic Pyrolysis Results.

The composition of compounds in the liquid yield obtained from various plastic composition variations can be seen in Table 4.

Table 4. Composition of Compounds in Liquid Yield

Sample	Carbon Chain	Type	Amount (%)
HDPE 70% PP 30%	C9-C11	Premium	30%
	C12-C20	Kerosene	62%
	C21-C24	Solar	8%
HDPE 30% PP 70%	C9-C11	Bensin	42%
	C12-C20	Kerosene	58%
	C21-C24	Solar	0%

In this study, the composition of the liquid products obtained from pyrolysis was identified using the GC-MS (Gas Chromatography-Mass Spectrometry) method. This identification was carried out to determine the distribution of components in the liquid products resulting from pyrolysis. The pyrolysis products tested using GC-MS included samples with 70% HDPE and 30% PP without catalyst, and 30% HDPE and 70% PP without catalyst. The calorific value test of the pyrolysis products was conducted by the Technical Implementation Unit (UPT) of the Integrated Laboratory, Diponegoro University, using GC-MS equipment. Below are the results of the GC-MS spectra breakdown of the pyrolysis liquid products.

Based on Table 4, it can be seen that the main component of the pyrolysis liquid products is kerosene. The sample with 70% HDPE and 30% PP is found to have a higher content of kerosene compared to the sample with 30% HDPE and 70% PP. Meanwhile, gasoline content is highest in the sample with 30% HDPE and 70% PP, amounting to 42%. According to Proxsis East (2015), gasoline is a distillation product of crude oil at temperatures between 35-75°C, while kerosene (kerosene) and aviation fuel are distilled at temperatures between 170-250°C. The temperature used in this study was 300°C, which aligns with the results of the pyrolysis, showing liquid yield compositions including gasoline, kerosene, and aviation fuel.

3.8 Economic Analysis of Plastic Pyrolysis on the Produced Liquid Yield Products.

The process of producing liquid fuel from plastic waste appears to be highly profitable. However, it is also important to analyze the production costs incurred. Production costs are assumed to include fuel requirements for the reactor, material preparation costs, and catalyst activation costs. The reactor fuel uses LPG priced at 25,000 Rupiah per 3 kg cylinder refill, totaling 50,000 Rupiah for 9 production cycles. Hence, the fuel cost per production cycle is 5,556 Rupiah.

Material preparation costs, including accommodation and others, are assumed to be 10,000 Rupiah for 9 production cycles, amounting to 1,111 Rupiah per production cycle. Catalyst activation costs for acetic acid, NH4Cl, and electricity total 37,250 Rupiah for 9 production cycles. Thus, catalyst activation costs per production cycle are 0 Rupiah for 0% catalyst variation, 375 Rupiah for 3% catalyst use, and 625 Rupiah for 5% catalyst use.

The common market selling price is 10,000 Rupiah per liter. However, the capital required to produce 130-225 ml ranges from 6,000 to 11,000 Rupiah. This indicates that the required capital is significantly higher compared to the existing market price, making the techno-economic analysis of the synthetic liquid fuel synthesis innovation from HDPE and PP plastic waste using catalytic pyrolysis with natural catalysts economically unfeasible. The main reason for this is the high production costs.

The fuel used in the reactor is very expensive in this study, leading to inflated production costs. 100% of the capital cost for variations without catalysts is in the production cost, while for variations with catalysts, about 60% of the capital cost is in the production cost and 40% is for catalyst activation. The high production costs indicate the ineffectiveness of the process. From an economic feasibility standpoint, the main issue lies in the choice of fuel source. The use of LPG as the main reactor fuel source drives up production costs. A simple analysis was conducted assuming electricity as the fuel source for the reactor.

4. CONCLUSION

Based on the research conducted, several conclusions can be drawn as follows:

a. Utilization of plastic waste as raw material for producing liquid fuel can be achieved using pyrolysis reaction, a cracking process in a reactor at 300 °C using Bayah Natural Zeolite catalyst.
b. The optimal condition that yields the highest output with quality meeting Indonesian national standards for liquid fuel from various compositions is under operating conditions of 350 °C temperature, pressure below 1 atm, with a composition of 70% PP and 30% HDPE without using Bayah Natural Zeolite catalyst.
c. Activated Bayah Natural Zeolite catalyst is not sufficiently effective in the pyrolysis process at 300 °C.
d. The economic feasibility analysis of producing liquid fuel from plastic waste indicates infeasibility, where the cost to produce 130-225 ml ranges from 6,000 to 11,000 Rupiah, while the selling price of pertalite gasoline is 10,000 Rupiah per liter.

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