# Experimental study the influence of zeolite size on low-temperature pyrolysis of low-density polyethylene plastic waste

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# Experimental study the influence of zeolite size on low-temperature pyrolysis of low-density polyethylene plastic waste

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A B S T R A C T

The plastic wastes may need billions of years to degrade naturally due to their slow degradation rate. The accumulation of the plastic wastes becomes a serious problem. Since a plastic is polymers of hydrocarbon, it has a potential to be converted into oil fuel. In the present work, zeolite based catalytic pyrolysis of low-density polyethylene (LDPE) plastic waste is performed with different zeolite sizes (i.e. 1, 2, and 3 mm in diameter) at low-temperature. The aim of the work is to investigate the effect of zeolite sizes on pyrolysis of LDPE plastic. The results show that smaller zeolite size increases heat transfer rate, pyrolysis temperature, reaction rate, and oil yield. From 1000 g of LDPE plastic, the oil yields are 138, 134, 126 mL for the use of 1, 2, and 3 mm in diameter of the zeolite, respectively. In order to improve the conversion of vapor into oil, the performance of the condenser have to be improved by lowering the temperature of cooling water or increasing heat transfer surface area of the condenser.

## 1. Introduction

According to Ministry of Environmental and Forestry of Republic of Indonesia [1], plastic waste is the second largest waste after organic waste in Indonesia. As shown in Fig. 1, plastic waste grasps 14% of total waste in 2017. The plastic waste mainly comes from bottles of mineral water, food plastic packages, and other plastic packages. The plastic wastes may take billions of years to degrade naturally [2] because their degradation rate is very slow [3]. This leads a huge accumulation in landfill and various natural habitats, i.e. rivers and oceans [4]. This accumulation becomes a problem in the future [5]. Since a plastic is a polymers of hydrocarbon, it has a potential to be converted into oil fuel. One of the promising ways of taking profit of the energetic of the plastic waste is pyrolysis [6]. Pyrolysis is potential process in waste management sector [7]. Many researchers have worked on plastic pyrolysis and have demonstrated the use of the technology to encounter the plastic waste problem [8-11].

Pyrolysis is a thermochemical conversion of solid biomass into useful energy in the absence of oxygen, resulting a char, a condensable liquids or tars, and a trace amount of gaseous products [12]. Stated by Dhyani et al. [13] and Guedes et al. [14], the main advantage of pyrolysis is that liquid fuel produced can be easily stored and transported. Depending on the process conditions, the pyrolysis process can be classified into following categories: fast pyrolysis, intermediate pyrolysis, slow pyrolysis, and hydropyrolysis [13]. Table 1 shows the difference in parameter and major product between slow and fast pyrolysis. Fast pyrolysis is recommended when liquid fuel is the goal of the pyrolysis.

In order to enhance the process and product of pyrolysis, a catalyst is used to increase reaction rate and hydrocarbon distribution in the liquid product and to reduce optimum pyrolysis temperature. Three common catalysts used in the plastic pyrolysis are Zeolite, FCC, and Silica-Alumina catalyst. Among those three, the zeolite catalyst is extensively applied in the plastic pyrolysis [2]. Catalytic pyrolysis of plastic waste using natural and synthetic zeolite catalysts was conducted by Miandad, et al. [3]. They obtained that the use of both natural and synthetic zeolite catalysts improved the quality of liquid oil by increasing the light hydrocarbon compounds. Other works on zeolite catalytic pyrolysis have been also reported by other researchers [12,15].

Several works on the use of zeolite as a catalyst in plastic pyrolysis have been reported, but none of those works discussed the effect of zeolite's particle size on characteristics and product yield of low-density polyethylene (LDPE) pyrolysis. In the present work, zeolite based catalytic pyrolysis of LDPE wastes is performed with different zeolite sizes (i.e. 1, 2, and 3 mm in diameter) at low-temperature. The effect of the particle sizes on characteristics and product yield of LDPE pyrolysis are investigated and discussed. In addition, performance of the condenser is also discussed.

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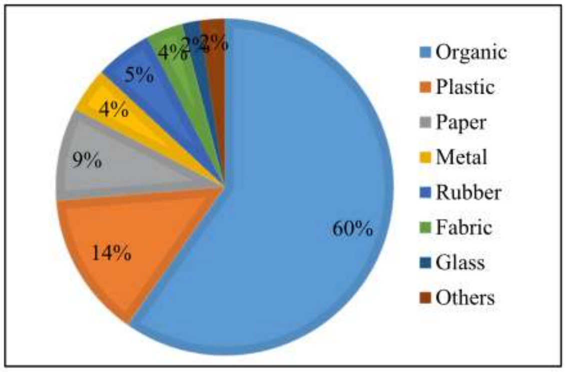


**Fig. 1.** Percentage of wastes in Indonesia [1].

**Table 1**
Parameter and major product of the slow, fast, and flash pyrolysis.

| Parameter | Pyrolysis | |
|---|---|---|
| | Slow | Fast |
| Temperature | Low | Moderate |
| Heating rate | Low | High |
| Residence time | Long | Short |
| Major product | Char | Liquid |

## 2. Materials and method

### *2.1. Materials*

LDPE plastic is widely applied as plastic bags [2]. Thus, only waste of plastic bags are collected from local waste disposal site in Yogyakarta to ensure the plastic is LDPE. The plastic bag is washed and sun dried for two days and crushed into smaller pieces as the feedstock of the pyrolyzer. Natural zeolite catalyst is bought from local market and crushed into smaller size of 1, 2, and 3 mm in diameter. Fig. 2 presents the photograph of the LPDE plastic and the zeolite.

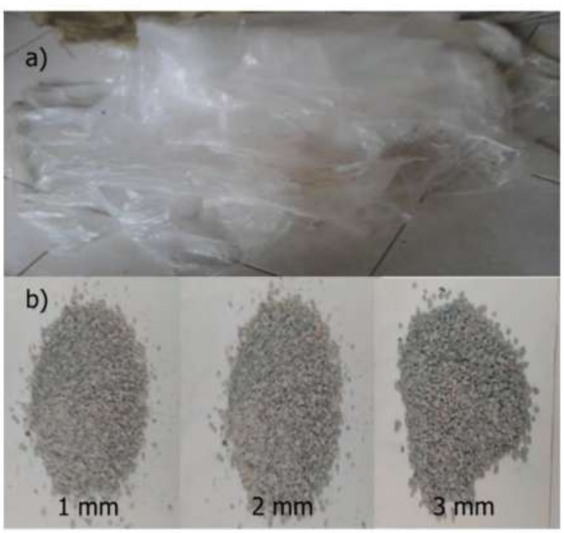


**Fig. 2.** Photograph of the plastic waste and the zeolite catalyst.

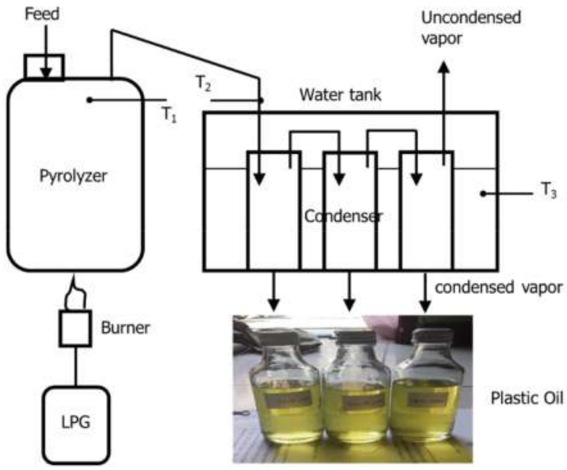


**Fig. 3.** Experimental setup and photograph of the plastic oil.

### *2.2. Experimental work*

Fig. 3 shows the experimental setup in the present work. The setup consists of a pyrolyzer, a condenser, a water tank, an LPG burner, K-type thermocouples and a data logger "GRAPHTEC GL240". The pyrolyzer, condenser, and water tank are hand-made from Stainless Steel plate with the thickness of 3 mm. The LPG burner and K-type thermocouples are bought from local market in Yogyakarta. The experimental work is performed in batch mode. For 180 min batch operation, 1000 g of the LDPE plastic and 500 g of the zeolite catalyst are fed into the pyrolyzer. The pyrolyzer is heated up using an LPG burner. Temperature at upper part of the pyrolyzer ($T_1$), temperature of the vapor at the condenser inlet ($T_2$), and temperature of the cooling water ($T_3$) are measured using the thermocouples and logged into the data logger. The plastic oil is collected every 15 min and weighted. Data of the temperatures and the plastic oil yield for the use of 1, 2, and 3 mm zeolite are analyzed and discussed. Higher heating value (HHV) of the oil is analyzed using Bomb Calorimeter. In addition, performance of the hand-made condenser is also discussed.

## 3. Results and discussion

Fig. 4 presents the temperature profile at upper part of the pyrolyzer ($T_1$) during 180 min batch operation time. The curves indicate that the

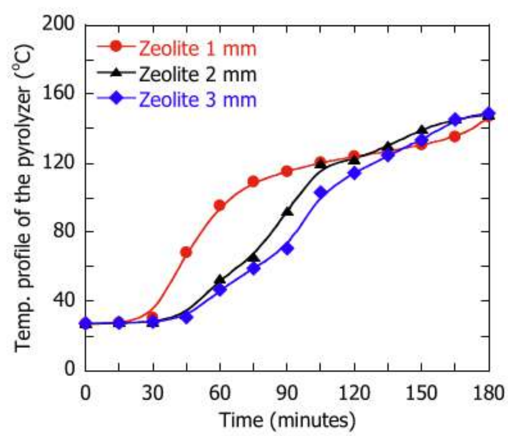


**Fig. 4.** Temperature profile at upper part of the pyrolyzer.

temperature at upper part of the pyrolyzer increases faster with the use of the 1 mm zeolite. Heat transfer rate to the feedstock is higher when using the 1 mm zeolite. This is due to larger heat transfer area with the use of 1 mm zeolite than with the use of 2 and 3 mm zeolite. Increasing heat transfer rate to the feedstock causes the pyrolysis temperature increases faster. The temperature at upper part of the pyrolyzer reaches 120 °C within 90 min for the use of the 1 mm zeolite. At the same minutes, the temperatures observed are 90 °C and 50 °C for the use of 2 mm and 3 mm zeolite, respectively. At 180th minute, the temperatures at that location are nearly similar for all zeolite sizes, such that 155 °C. According to Aboulkas et al. [16], LPDE pyrolysis starts to produce oil at temperature of 300 °C. The temperature does not change significantly by adding a catalyst [17]. In the present work, the heat for pyrolysis is obtained from LPG burner. Thus, it is difficult to maintain a proper pyrolysis temperature. However, the temperature at upper part of the pyrolyzer ($T_1$) can be used to predict the pyrolysis temperature at the bottom of the pyrolyzer. The pyrolysis temperature should be higher than 155 °C. Conduction heat transfer occur in the pyrolyzer from bottom to the top (i.e. from higher temperature to lower temperature). Theoretically, the temperature at the bottom part of the pyrolyzer (i.e. pyrolysis temperature) is higher than 155 °C and probably near to 300 °C.

In general, the trend of oil yield is similar to the pyrolysis temperatures trend for all zeolite size as shown in Figs. 5–7. Comparing all three curves in Fig. 5, it can be noticed that oil production starts after 80, 90, and 100 min for the use of 1, 2, and 3 mm zeolite, accordingly. The oil production begin when the temperature at upper part of the pyrolysis reaches 100 °C. Since the pyrolysis temperature increases faster for the use of 1 mm zeolite, shorter time is required to start the production of the plastic oil. The increase in temperature affects the oil production positively. The temperature in the pyrolysis process indicates the necessary heat for the decomposition of the plastic bonds. The conversion efficiency increases with increasing temperature [13]. However, very high temperatures give negative effect on oil production. It is because at very high temperature there is a secondary cracking of the volatiles, which results in a higher gas yield. Pyrolysis temperature of LDPE is considered high when the process was conducted at temperature higher than 550 °C. Marcilla et al. [18] conducted LDPE pyrolysis at temperature of 550 °C. No other reported work on LPDE pyrolysis was performed at temperature higher than 550 °C. Besides increasing heat transfer area, reducing size of the zeolite also improves contact area between the LDPE and the zeolite. Improving contact area leads in enhancing the pyrolysis reaction rate.Fig. 6.

In the present work, the pyrolysis temperature at the bottom part of the pyrolyzer should be higher than 300 °C According to Aboulkas et al. [16], pyrolysis temperature of LDPE at which oil production begin was

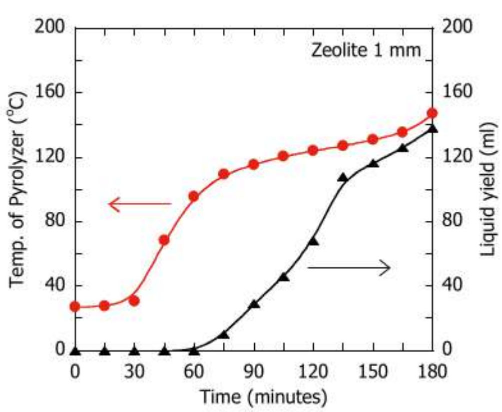


**Fig. 5.** An effect of pyrolysis temperature on oil production using 1 mm zeolite.

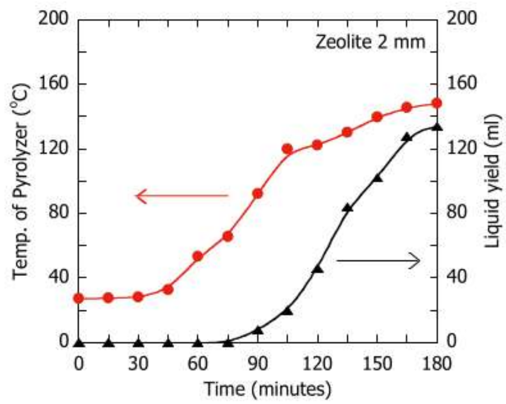


**Fig. 6.** An effect of pyrolysis temperature on oil production using 2 mm zeolite.

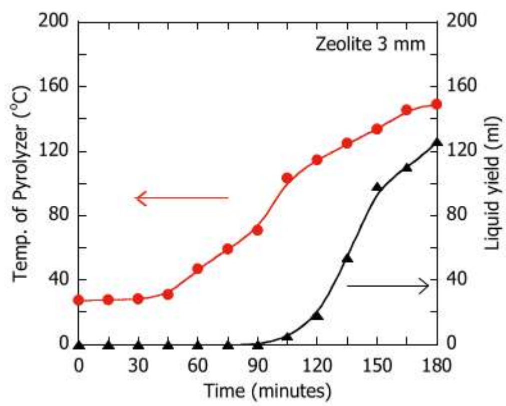


**Fig. 7.** An effect of pyrolysis temperature on oil production using 3 mm zeolite.

300 °C. Pyrolysis temperature range at which conversion of the plastic begin can be indicated by their thermal degradation temperature which is depended on plastic type [2]. Various pyrolysis temperatures of specific plastic have been investigated. For high-density polyethylene (HDPE), the pyrolysis temperatures observed are 300–400° [8], 450–550 [9]. Meanwhile for low-density polyethylene (LDPE), Onwudili et al. [19] performed LDPE pyrolysis at temperature of 425 °C, Bagri and William [20] conducted LDPE pyrolysis at temperature of 500 °C, and LDPE pyrolysis was conducted at temperature of 550 °C by Marcilla et al. [18].

Figs. 8 and 9 display the total oil yield and weight percentage of the product and residue after 180 min pyrolysis. From Fig. 8, the oil yields are 138, 134, 126 mL for the use of 1, 2, and 3 mm zeolite, respectively. Those volume of oils have a weight of 103.5, 100.5, and 94.5 g, accordingly. In weight percentage, the highest oil yield is 6.9 wt% which is obtained for the use 1 mm zeolite as shown in Fig. 9. The highest oil yield obtained at 1 mm zeolite is due to the highest reaction rate with the use of 1 mm zeolite. The oil yield in the present work is much lower than that obtained by Marcilla et al. [18] who obtained oil yield of 93.1 wt% when carried out LDPE pyrolysis in a batch reactor at 550 °C with heating rate of 5 °C/min. This may due to uncontrolled heating rate and much lower of pyrolysis temperature in the present work. To obtain more precise measurement of oil yield, the digital measurement device should be used. This may reduce uncertainty in device reading during the experimental work.

Meanwhile, Fig. 10 presents higher heating value (HHV) of the plastic oil. The use of 1 mm zeolite produces plastic oil with the highest

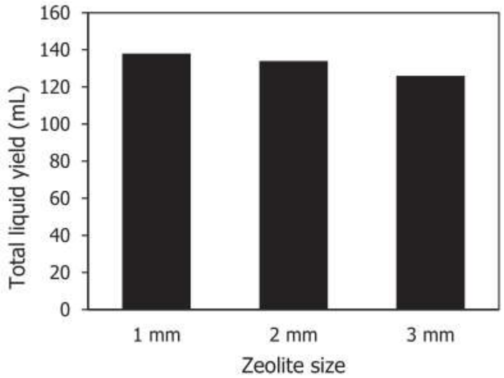


**Fig. 8.** Total liquid yield.

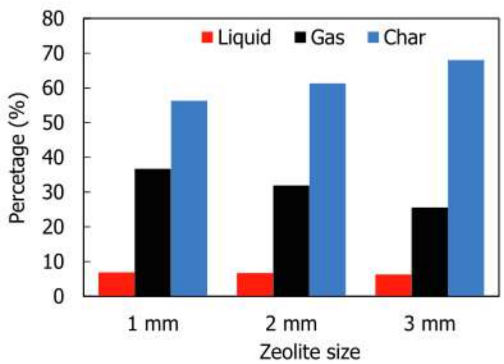


**Fig. 9.** Percentage of liquid, gas, and char.

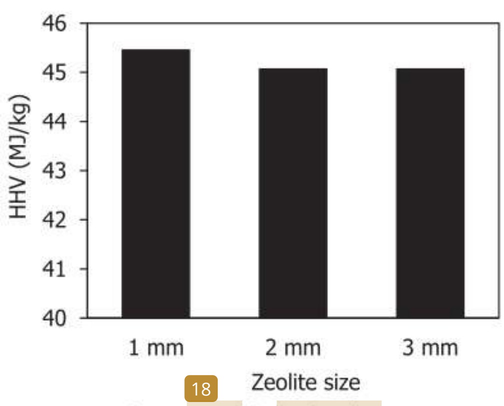


**Fig. 10.** HHV of the liquid product.

HHV. The HHV of the oil with the use of 1, 2, and 3 mm zeolite are 45.47, 45.08, and 45.08 MJ/kg. These HHVs are nearly similar to the HHV of gasoline and diesel fuel. In terms of density, oil from LDPE pyrolysis seems comparable with the commercial standard value of both gasoline and diesel [2]. Desai and Galage [21] found that LDPE oil has a density of 0.78 which is similar to that of gasoline [8]. From measurement of volume and mass of the oil yield, the density of the oil is calculated to be 0.75 in the present work. This value is nearly similar to that of gasoline. Since the HHV and density of the oil are almost similar to that of gasoline, the oil obtained from LDPE pyrolysis in the present work could be used as a fuel of spark ignition engine.

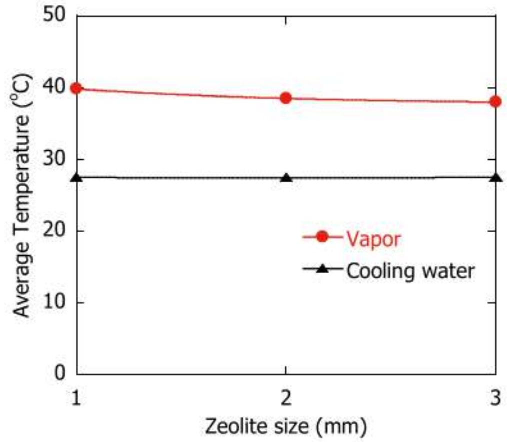


**Fig. 11.** Average temperature of vapor and cooling water of the condenser.

In addition, overall performance of the condenser is analyzed based on Fig. 9 and Fig. 11. Weight percentages of the oil are lower than that of the gas and the char for all zeolite sizes. Much lower the oil percentage is due to low condensation rate of the vapor in the condenser. Heat transfer rate from the vapor to the cooling water is low. This phenomenon is indicated by the temperature difference between vapor temperature at the condenser inlet and the temperature of cooling water which is relatively small as shown by Fig. 11. In order to increase oil yield, performance of the condenser could be improved by lowering the temperature of the cooling water to 15 °C [22] or increasing heat transfer surface area of the condenser. Improving heat transfer rate will enhance condensation rate, resulting more vapor is converted into oil.

## 4. Conclusion

Experimental work of catalytic pyrolysis of the LPDE plastic waste is been conducted in batch pyrolysis using different zeolite size (i.e. 1, 2, and 3 mm in diameter) at low-temperature. It can be concluded that zeolite size impacts the pyrolysis process and oil yield. Smaller zeolite size increases heat transfer rate, pyrolysis temperature, reaction rate, and oil yield. The pyrolysis temperature plays major role in oil yield. Oil yield increases as increasing in pyrolysis temperature. However, oil yield percentage is relatively low compared to gas yield and remaining char. Condensation rate of the vapor in the condenser is very low. The performance of the condenser have to be improved by lowering the temperature of the cooling water or increasing heat transfer surface area of the condenser.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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