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ORIGINAL PAPER

Pyrolysis of Polyolefins into Chemicals Using Low-Cost Natural Zeolites

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Abstract

An experimental study of pyrolysis from polyethylene (PE) and polypropylene (PP) has been done by catalytic method and utilization of low-cost natural zeolite in a semi-batch reactor. A low-cost natural zeolite made from acid-treated zeolite with 1 M HCl solution was denoted as HZ. The prepared HZ catalyst was then carried out under pyrolysis reaction at 350, 400, 450 °C for 60 min. The chemical products composition from this reaction was analyzed using Gas Chromatography-Mass Spectrometry (GC–MS). According to the analysis result, a low-temperature reaction of pyrolysis could produce wax in all variant types of feed. Utilization of HZ catalyst could reduce formation wax while also increasing the yield of oil products after the reaction. Catalytic pyrolysis using HZ for PE and PP plastic feed yields oil products up to 67% and 70%, respectively. The composition of oil products mainly consisted of paraffin, olefin, and alcohol compounds. The temperature optimum for the catalytic reaction could produce the highest paraffin and olefin products at 400 °C. Additionally, utilization of low-cost natural zeolite could improve pyrolysis reaction's performance from PP to produce yield olefin products from 39 to 62%.

Graphical Abstract
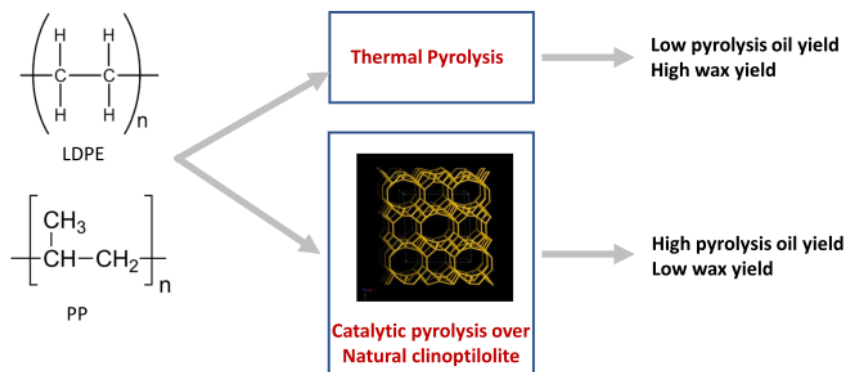
Thermal Pyrolysis LDPE and PP versus Catalytic Pyrolysis over Natural Clinoptilolite

Statement of Novelty

Plastics waste is threatening the environment due to its difficulty in degradation. Pyrolysis of plastic by using catalysts would improve the oil products. Natural zeolites are low-cost minerals and are abundantly available throughout the world. The novelty of this study is to investigate the

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local natural zeolites in Indonesia as a cost-effective catalyst for application in polyolefins pyrolysis. The oil yield and products selectivity improved significantly after using the natural zeolites. The oil composition identified would be potentially utilized as chemicals and fuels. Finally, the catalytic pyrolysis of plastic waste using low-cost local catalysts will overcome the environmental problem and create the circular economy of plastic.

Introduction

Plastic is essential in supporting our modern life. Plastic waste production globally reached 275 million tons per year [1]. In 2017, Indonesia produced about 6.8 million tonnes, with annual growth estimated at 5% [2]. Massive production of disposable plastic that is not supported with proper plastic waste management has created an environmental problem. It was estimated that 70% of Indonesia's plastic waste was mismanaged, i.e., openly burned (48%), dumped on land (13%), or leaking into waterways and the ocean (9%) [2]. This improper plastic waste treatment has been threatening environmental and ecological life. Recent studies from several researchers report that microplastics from plastic waste are already found in a small organisms in Jakarta, Wonorejo, Lampung, and Sumbawa [3, 4]. Additionally, another report predicts Banten Bay will also be covered with plastic waste by 2028 [5].

Most plastic came from petroleum derivatives such as polyethylene (PE) and polypropylene (PP) that promising to be converted into other beneficial chemical compounds. One method that preemptively could address the negative effect of plastic waste is the pyrolysis process. Pyrolysis is a method of transforming the molecules into other chemical components induced by the thermal process without oxygen (O_2). Pyrolysis can be carried out thermally by involving only one feed or multiple feeds which is called co-pyrolysis. Thermal pyrolysis in the presence of additional feed from biomass (co-pyrolysis) can reduce the activation energy so that the energy consumption required in co-pyrolysis is lower than pyrolysis with single feed [6, 7, 8]. Thermal pyrolysis has the advantage of obtaining certain products without worrying about problems with poisoning of the catalyst caused by various product feeds including co-pyrolysis processes. Catalytic pyrolysis can produce more specific and high quality products with a faster residence time [9]. Pyrolysis of polyolefins such as PE and PP could be carried out through the thermal and catalytic processes [10]. Thermal pyrolysis of PE produced mostly C_4–C_{35} paraffin with linear alkenes, while thermal pyrolysis of PP produced branch or cyclic alkenes [11].

A PE and PP waste mixture was reported successfully thermally pyrolyzed at 350 °C with a product yield of 81%

of liquid oil, 12% of residual solid material, and 7% gases in a small scale externally heated pyrolysis batch reactor [12]. It is reported that the conversion of plastic under thermal pyrolysis showed lower conversion compared with catalytic pyrolysis [13]. Tiwari et al. [14] reported that the oil obtained at a lower temperature (350 °C) of thermal pyrolysis has superior fuel properties like high calorific value, low viscosity, pour point, and flashpoint. However, the thermal pyrolysis has to be accomplished for a long duration to support polymer scission reaction and the production of lighter hydrocarbons. Thermal pyrolysis has some disadvantages, such as operating at high temperatures, high energy consumption, and low oil quality.

On the other hand, catalytic pyrolysis could be beneficial to overcome those drawbacks of thermal pyrolysis. The catalyst could lower the temperature reaction as it provides the active sites on the surface to enhance the pyrolysis reaction and reduce the activation energy. Recent investigation reported that the thermal pyrolysis of plastic mainly produced waxes and after contacting the pyrolysis vapours with ZSM-5 catalyst, the product distribution changed significantly with main products oil and gases [15]. However, the catalytic process also has disadvantages, such as the high price of catalysts and rapid catalyst deactivation [10]. The deactivated catalyst is required to be regenerated or replaced. Another thing is that the catalyst used in a high amount ratio between plastic and the catalysts within the range of 10–50% wt. [16, 17]. Hence, it is needed to find an inexpensive catalyst. Some researchers investigated the cheap catalysts for polyolefin pyrolyzes, such as spent fluid catalytic cracking (FCC) catalyst [16, 18] and natural zeolites [13, 19, 20, 21].

Natural zeolites are inexpensive and abundant natural minerals in Indonesia. Hence, the utilization of local low-cost natural zeolites to overcome the plastic problem by catalytic pyrolysis could give high benefits for the country and the environment. Polypropylene pyrolysis over Indonesian natural zeolites was reported within a narrow temperature range 400–430 °C [19]. The type and properties of the natural zeolites were not characterized and not correlated with the chemical product's yield and distribution. Miandad et al. [13] used natural zeolites from Harrat-Shama, the Kingdom of Saudi Arabia, for Plastic waste, which was pyrolyzed at 450 °C. The type of natural zeolites was not reported, and the milled sample was used in the pyrolysis reactor without any further modification. While, Syamsiro et al. [20] used natural zeolites from Klaten (Indonesia), which identified the natural zeolites as mordenite dominant type for polyolefins waste pyrolysis. The natural mordenite was calcined before being used as a catalyst.

The study of natural zeolites as catalysts for polyolefins pyrolysis is still open in studying the temperature effect, modification of natural zeolites, and correlation between the natural zeolite properties and the pyrolysis yield, product

distribution, and targeting the products as chemicals. Thus, the research aimed to study thermal and catalytic pyrolysis of polyethylene and polypropylene plastic using local inexpensive natural zeolites as a catalyst to produce chemicals.

Experimental Section

Materials

Natural zeolites were obtained from Bayah, Indonesia. Polypropylene (PP) and low-density polyethylene (PE) were used as feedstock for pyrolysis. The PP and PE plastics were purchased from local supermarkets. Both plastics were cut into small rectangular sizes 1 cm by 1 cm.

Acid Treatment

180 g natural zeolites was soaked in 360 mL HCl 1 M for 24 h at room temperature. After that, the sample was filtered and washed with distilled water until neutral. Then, the sample was dried at 100 °C for 4 h, followed by calcination for 2 h at 300 °C using oven. The catalyst was labeled as HZ. X-ray diffraction (XRD, Pan Analytical) was performed to identify the natural zeolites phases and study acid treatment effects. The increment of the angle 2Θ was 0.02°. Acid treatment was performed to create mesopore, extract metals impurities, and provide protonated zeolites [22].

Pyrolysis and Product Analysis

Figure 1 presents the reactor setup for the pyrolysis of plastics into oil. 300 g plastics and zeolites catalyst (10% wt) was introduced into the reactor. First, the plastic is put into the reactor, then followed by zeolites catalyst. After that the mixture was stirred in the reactor until it was well mixed between the plastic and the zeolites. Samples were heated in the reactor with a constant heating rate 10 °C/min from

room temperature to the desired temperature, i.e., 350, 400, and 450 °C. The heating rate is controlled by a thermocouple located outside of the reactor. During the pyrolysis process, the temperature was maintained constant for 1 h. Hot gas produced from plastic pyrolysis was condensed by using water as a cooling media. The liquid product comes out and collects in the accumulator. Products of pyrolysis of plastics were oil, gas, and solid residue (wax and char). All of the products were weighed, except the gas determined by mass balance calculation.
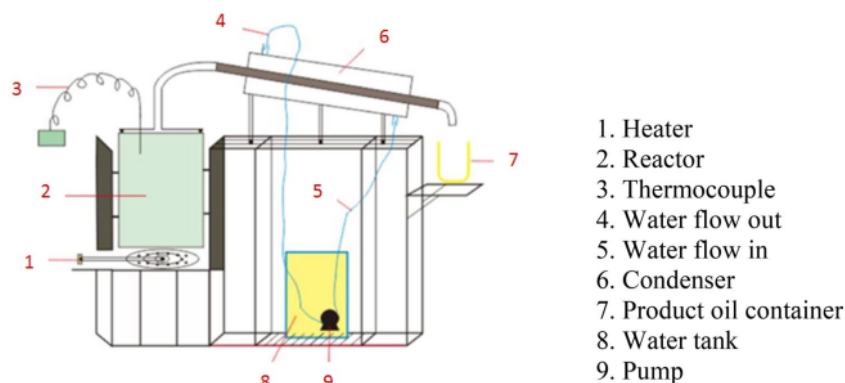
The pyrolysis oil was analyzed using Gas Chromatography-Mass Spectrometry (GC–MS QP2010 Ultra, Shimadzu Corp., Japan). The density was determined experimentally by a pycnometer. Oil viscosity was analyzed by using viscometer Ostwald. Flashpoint of oil was analyzed by flash point tester (SYD-261 PMCC).

Results and Discussion

Natural Zeolites Characterization

Figure 2 presents the the X-ray diffraction pattern of as received Bayah natural zeolites compared with the standard pattern referred to by the International Zeolite Association [23]. The as-received Bayah natural zeolites diffraction pattern identified as clinoptilolite dominant type (Fig. 2a) with mordenite (Fig. 2b) and quartz (Fig. 2c) as phase competitors. The result is aligned with another group that reported that Bayah natural zeolites were clinoptilolite dominant type, which was applied for converting glycerol to glycerol carbonate [24]. However, the present work's dominant zeolite phase is different from our previous investigation, which identified that Bayah natural zeolites were mordenite dominant type [25]. The diffraction pattern of natural zeolites was varied even samples obtained from the same mining district due to uncontrolled conditions during the zeolite synthesis in nature [26].

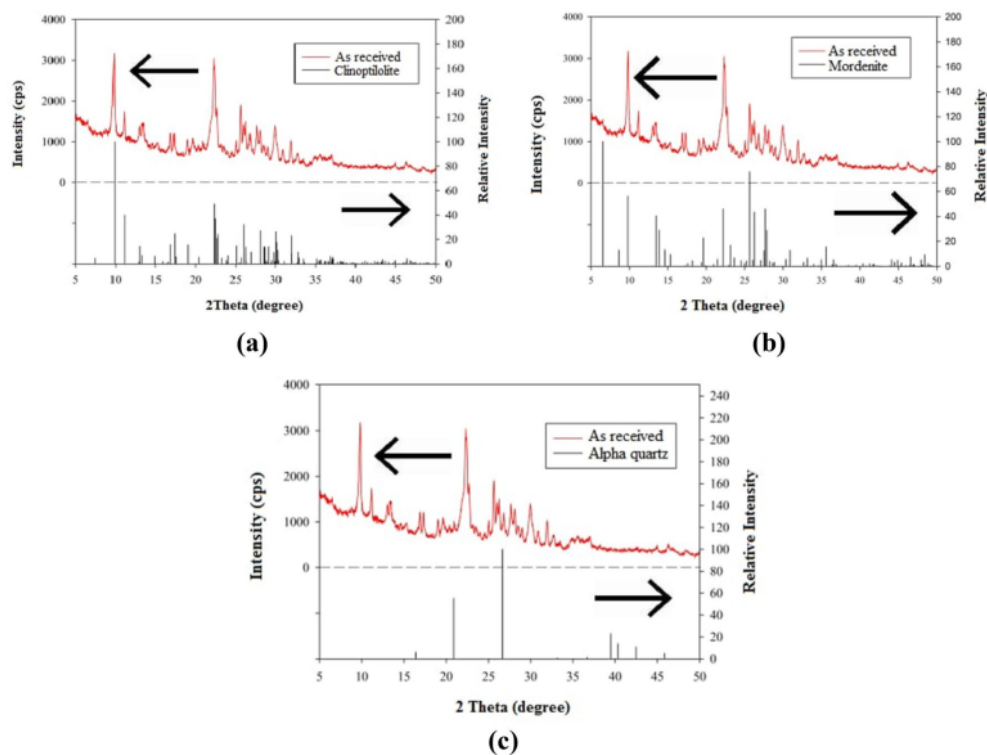Fig. 1 Pyrolysis reactor for plastic conversion into oil

1. Heater
2. Reactor
3. Thermocouple
4. Water flow out
5. Water flow in
6. Condenser
7. Product oil container
8. Water tank
9. Pump

Fig. 2 Comparison of natural zeolites diffraction with standard pattern **a** clinoptilolite, **b** mordenite, **c** alpha quartz
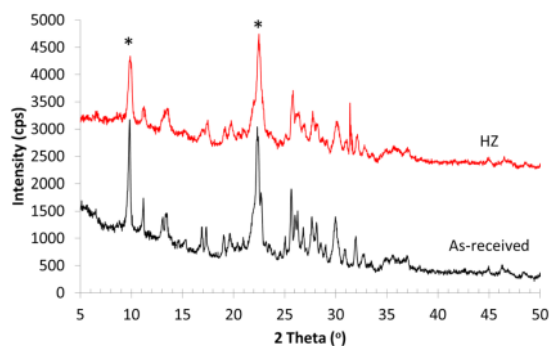

Fig. 3 Diffraction pattern of the as received and HZ natural zeolites (offset 2000 cps)

Effect of Acid Treatment on Crystallinity

Figure 3 presents the X-ray diffraction pattern of as-received and acid-treated natural zeolites (HZ). Acid treatment using hydrochloric acid 1 M reduced the intensity of peaks which shown clearly at 2Θ 9.77° and 22.32° indicated with the asterisks. This peak indicates that aluminum was extracted from the framework of zeolites during acid treatment for 24 h at room temperature. Despite the dealumination, the acid-treated natural zeolite structure was not collapsing. Further acid treatment with high acid concentration, high temperature, and or long treatment could lead to the zeolites' destruction and end the zeolites as an amorphous phase [27, 28, 29]. Acid treatment was also reported by removing aluminum and cation metals, i.e., Mg, Fe, and Ca, particularly at high-temperature acid treatment [29]. Acid treatment by using nitric acid to natural zeolites improved surface area as reported in our previous work [27]. The increase of surface area was contributed from both of micropore and mesopore. The micropore area increment was much higher than then the mesopore. HCl treatment on natural clinoptilolite was reported increased the surface area significantly which attributed to opening the windows formed by the substitution of exchangeable cations by H^+, partial delaumination and breakdown of structure clinoptilolite [30].

Effect of Temperature, Polyolefin Types, and Natural Zeolites on Pyrolysis Yield

The pyrolysis temperature significantly affected the distribution of the product: oil, wax, and gas, as presented in Fig. 4. It can be seen from Fig. 4a that the oil yield of
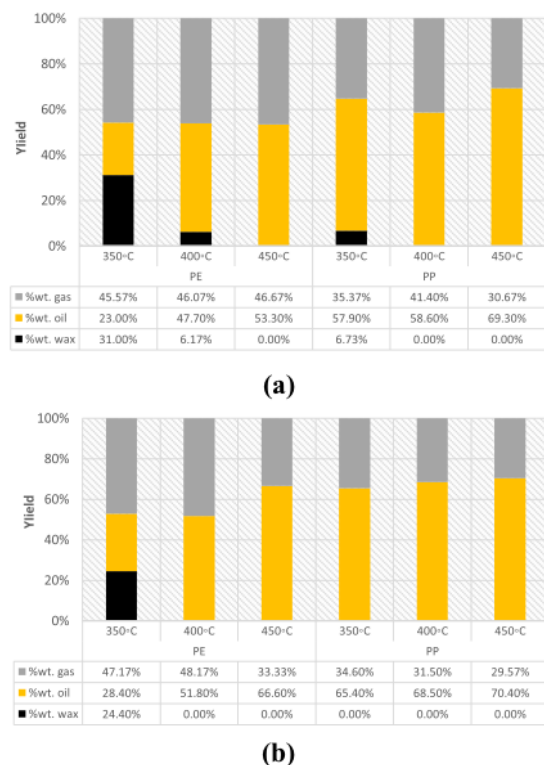
	350°C	400°C	450°C	350°C	400°C	450°C
		PE			PP	
■ %wt. gas	45.57%	46.07%	46.67%	35.37%	41.40%	30.67%
■ %wt. oil	23.00%	47.70%	53.30%	57.90%	58.60%	69.30%
■ %wt. wax	31.00%	6.17%	0.00%	6.73%	0.00%	0.00%

(a)

	350°C	400°C	450°C	350°C	400°C	450°C
		PE			PP	
■ %wt. gas	47.17%	48.17%	33.33%	34.60%	31.50%	29.57%
■ %wt. oil	28.40%	51.80%	66.60%	65.40%	68.50%	70.40%
■ %wt. wax	24.40%	0.00%	0.00%	0.00%	0.00%	0.00%

(b)

Fig. 4 Effect of temperature on yield of solid residual, oil, and gas for pyrolysis of **a** PE and PP via thermal pyrolysis **b** PE and PP over HZ catalyst

PE pyrolysis at temperature 350 °C was low. The oil yield increased with temperature until it reached a maximum oil yield at 450 °C. The same trend showed in PP pyrolysis, as seen in Fig. 4b. Gas products yield was decreased when temperature increased from 400 to 450 °C. The trend also agrees with the literature[14, 18, 19, 31], which reported that the increase in the pyrolysis temperature could increase the oil yield and decrease the gas yield in PP pyrolysis. Wax produced at temperature 350 °C and no-wax produced at temperature 450 °C. The wax is converted into liquid at high temperature as the products distribution change with dominated by the oil yield. However, the oil yield decreases and gas increases when pyrolysis operates at a temperature above 450 °C, as reported elsewhere [32, 33]. The plastics cracked into more gas, as the result the percentage of oil yield decreased. The pyrolysis temperature is critical to obtain the desired products, whether oil or gas, to be maximized. The pyrolysis oil color was slightly changed with the pyrolysis temperature from bright yellow at low temperature to dark yellow at high temperature (Figs. 5, 6).

The type of polyolefins determined the yield of pyrolysis products. Pyrolysis of PP generated less wax, more oil, and less gas than PE within the temperature of 350 to 450 °C, as presented in Fig. 4. The results agreed with Aguado et al. [34], which reported that the thermal pyrolysis conversion of PP was higher than PE. The apparent activation energy of thermal pyrolysis PP and PE was reported 362.6 and 413 kJ/mol, respectively [35], suggesting that PP is easier to be pyrolyzed than PE. It is shown clearly in Figs. 5 and 6 that the wax production in PE was high at temperature 350 °C and still produced wax at 400 °C for the thermal pyrolysis.
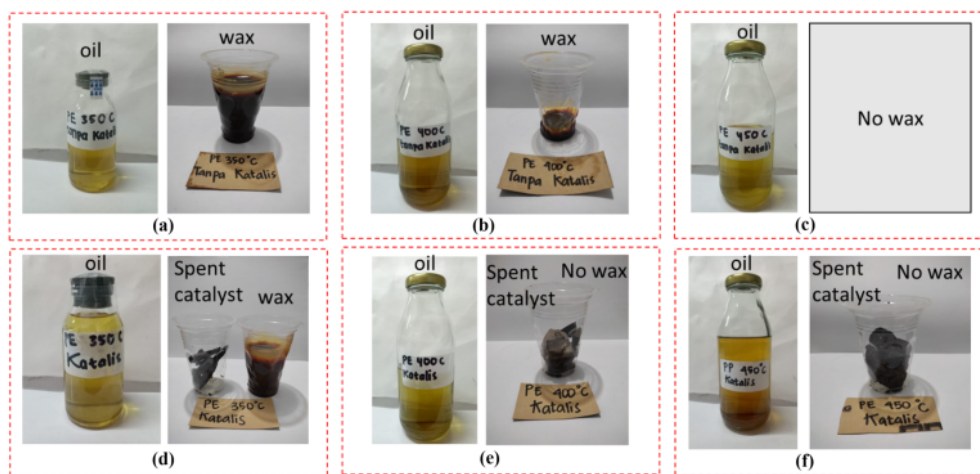

Fig. 5 PE pyrolysis product oil and wax **a** 350 °C thermal, **b** 400 °C thermal, **c** 450 °C thermal, **d** 350 °C with HZ, **e** 400 °C with HZ, **f** 450 °C with HZ

Fig. 6 PP pyrolysis product oil and wax **a** 350 °C thermal, **b** 400 °C thermal, **c** 450 °C thermal, **d** 350 °C with HZ, **e** 400 °C with HZ, **f** 450 °C with HZ
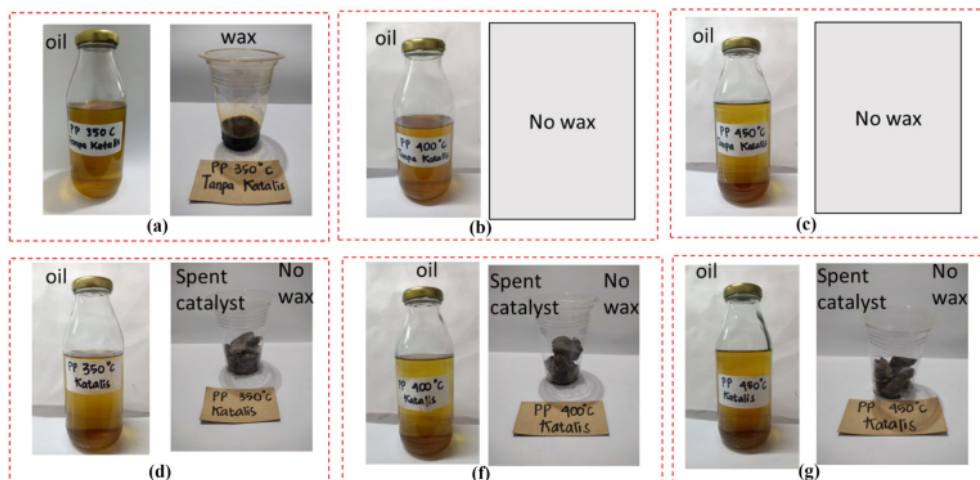
In contrast, no wax was produced for PP thermal pyrolysis at 400 °C. Thermal pyrolysis PP at temperature 350 °C yielded 6.73% wax, far less than PE with 31% wax. According to Aguado et al. [36], the higher number of tertiary carbons present in the PP chains promotes the thermal cleavage of C–C bonds. Nonetheless, the PP could be pyrolyzed at a lower temperature than PE. The result follows Miandad et al. [13], which reported that PP showed a lower temperature decomposition than PE through thermogravimetric analysis (TGA). The thermal degradation of PP started at 240 °C and achieved the maximum decomposition at 425 °C, while PE started at a higher temperature of 270 °C and achieved maximum decomposition at 480 °C.

Pyrolysis polyolefins over natural zeolites improved the oil yield and reduced wax yield, as presented in Fig. 7. At temperature 350 °C, the wax yield of catalytic PE pyrolysis reduced from 31 to 24.4% and from 6.73 to 0% for catalytic PP pyrolysis. The oil yield increased from 23 to 28% and from 58 to 65% in PE and PP catalytic pyrolysis. The results show that natural zeolites significantly affected the yield products of the pyrolysis reaction. After acid treatment, the natural zeolites had acid sites that became the place for the polyolefins cracking. The Bayah natural zeolites were dominated as clinoptilolite types. Despite the pore size of clinoptilolite being relatively small, excluding molecules larger than 0.4 nm [37], the catalyst showed high activity for pyrolysis plastic. The mesoporous features created after acid-treated in the natural zeolite pore provide access to acid sites for reactants. Many reports, including our previous work [22, 27, 38], showed that the acid treatment created the mesopore, which facilitated the high diffusion of bulky molecules. The acid treatment over ZSM-5 was also performed
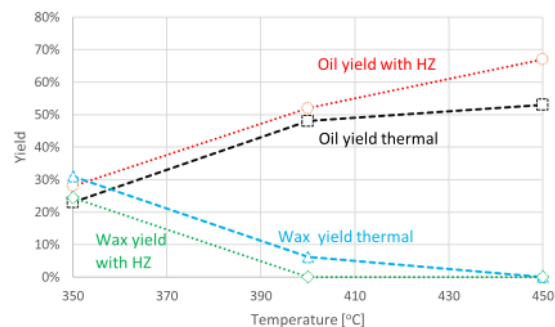
(a)
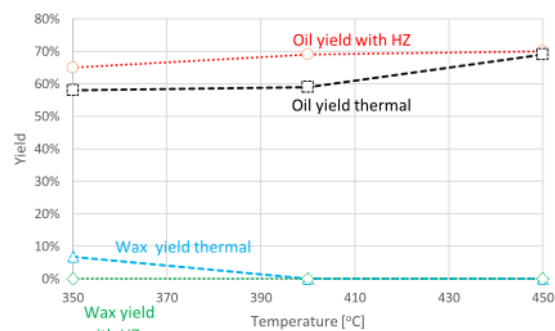
(b)

Fig. 7 Effect of catalyst HZ on oil and wax yield from pyrolysis of **a** PE, **b** PP

by Aguado et al. [36], which showed that the ZSM-5 was more active for polyethylene pyrolysis than MCM-41. They suggested that zeolites' acidity also plays a crucial factor in the PE cracking beside the pore size. The results were different with Syamsiro et al. [20], who reported that the oil yield of pyrolysis PE over natural zeolites is lower than the thermal pyrolysis. The difference result is caused by the configuration of the reactor which used two series reactors. The first reactor for thermal pyrolysis and the second reactor for the catalytic reaction for the product from the first reactor. It is most likely that the catalyst active in one reactor (in situ) converts the large solid polymer molecules into the liquid while two reactors tend to convert the condensable gas into a shorter molecular chain using active catalysts.

Main mechanism of thermal decomposition of polyolefin was proposed by Yan et al. [35] as presented in Fig. 8. In the thermal pyrolysis, radicals formation breaking C–C bonds to produce wax, oil, char and gas. In the catalytic process, beside the thermal mechanism with radicals, the cracking of long molecular chain also performed due to the acid catalysts on the external and or mesopore surface
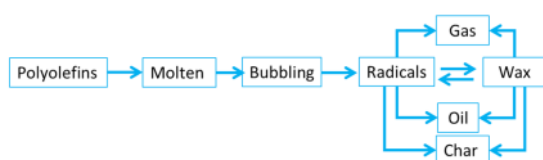
of the catalyst (Fig. 9). Thus, the conversion of wax into the oil in catalytic pyrolysis was high as compared with the thermal pyrolysis. The fact that percent yield of gas production was reduced in the catalytic PP pyrolysis suggested that the micropore was not accessible for most of large molecules.

Almost all samples showed similar weight after and before pyrolysis, which suggested that some natural zeolites dissolved or carried over and the cokes formation within the zeolites pores balanced the weight of the spent catalyst. The topology of the zeolite controls the product distribution and coke deposition [39]. For example, the coke precursors have a tendency to grow up to a point where they have steric hindrance in zeolite Y and stuck in the cages pore network. However, the ZSM-5 with its 3D pore architectural network improves the stripping of coke precursors, as the results increases the life of the catalyst. In our investigation, cokes were observed to cover the natural zeolite's surface. The Bayah natural zeolites are mainly clinoptilolite with main pore size 5.5 Å × 3.1 Å, which is much smaller than the commercial catalysts. Thus, the coke deposition could be much higher. The oligomerization and cyclization are the main reaction scheme inside the pore zeolites to form cokes, which are mainly polyaromatic structure. The cokes covering the catalyst reduce its activity; hence, the regeneration procedure should be performed before reusing it for the next batch. Improving catalyst stability by impregnating with metal and increasing the mesopore could also reduce coke formation and prolong the catalyst's lifetime.

Fig. 8 Main routes for thermal decomposition of polyolefins (adapted from [35])

Fig. 9 Mechanism of PE and PP thermal and catalytic pyrolysis over the natural zeolites

Effect of Temperature on Physical Properties of Pyrolysis Oil

Table 1 presents the oil properties: density, viscosity, and flash point under conditions, i.e., temperature, polyolefins type, and catalyst. Generally, the temperature is not significantly affected the oil density. Singh et al. [40] suggested that the increased heating rate reduced the volatile retention time, produced long-chain hydrocarbon oil as the oil density increased. It can be seen that the oil density from the PE pyrolysis is within the range of 0.788–0.798 g/mL, while the oil density of PP plastic pyrolysis is within the range of 0.786–0.796 g/mL. Based on product density data and fuel standards, the oil from the pyrolysis of PE and PP plastics is close to the density of kerosene and diesel fuel.

The results showed that the viscosity of the PE type was within the range of 0.83–1.04 cP, and for the PP type, it was within the range of 0.95–1.14 cP (Table 1). The viscosity of each type of plastic increased with increasing temperature. Viscosity was also affected by density, where the heavier the density of the liquid, the higher the viscosity. These phenomena are caused by the heavier the density, the more particles contained in the oil, which will hinder the fluid flow. The oil viscosity of PE pyrolysis was 0.8–1.04 cP, while the viscosity of PP pyrolysis oil was in the range of 0.96–1.14 cP. Based on the table above, it can be seen that the oil produced by the pyrolysis of PE and PP plastics was close to the viscosity of diesel fuel.

The flashpoint values for PP and PE plastics pyrolysis are almost similar. The flashpoint of the oils from PE pyrolysis was in the range of 44 to 48 °C and for PP were 40 to 46 °C. The results above showed that the highest flash point value was produced by uncatalyzed PE plastic with a temperature of 350 °C, and the lowest flash point value was produced by uncatalyzed PP plastic with a temperature of 400 °C and 450 °C. From the overall flash point results on PP and PP plastics, it can be seen that those using a catalyst have a lower flash point, which more suitable to use as chemicals instead of fuel.

Effect of Catalyst Natural Zeolites on Products Composition of Pyrolysis Oil

Functional groups of pyrolysis oil thermal and catalytic are presented in Fig. 10. The oil products mainly consist of paraffin, olefins, and alcohol. Other traces were also identified, such as ketone and ether. The composition of functional groups of PE is almost similar between thermal and catalytic pyrolysis. There was a slight increase in olefinic products and a slight decrease in paraffinic products. This result may not be statistically significant and could be due to inherent variation in the process and feedstock.

In contrast, there was a significant increase in olefinic products from 39% in the thermal pyrolysis to 62% in the catalytic pyrolysis of PP. Additionally, Alcohols decrease from 32 to 16% for PP's thermal and catalytic pyrolysis, respectively. The paraffinic product slightly decreased from 20 to 17% which was occurred due to the inherent variation. The type of zeolites affected the pyrolysis products [42, 43]. Due to the shape of selectivity, the ZSM-5 produced more aromatics products than zeolite-Y and mordenite. In our work, the zeolite was clinoptilolite dominant type and mordenite with the impurities, which showed non-aromatics products. Clinoptilolite has a small pore aperture ca. 4 Å,

Table 1 Effect of temperature, plastics material, and catalyst on pyrolysis oil properties

Oil	350 °C			400 °C			450 °C		
	Density (g/mL)	Viscosity (cP)	Flash point (°C)	Density (g/mL)	Viscosity (cP)	Flash point (°C)	Density (g/mL)	Viscosity (cP)	Flash point (°C)
PE thermal	0.789	1.045	48	0.792	1.096	46	0.788	1.139	46
PE with catalyst HZ	0.789	0.949	46	0.797	1.055	44	0.798	1.153	44
PP thermal	0.786	1.135	46	0.794	1.148	44	0.796	1.149	44
PP with catalyst HZ	0.792	1.144	42	0.793	1.145	40	0.794	1.194	40

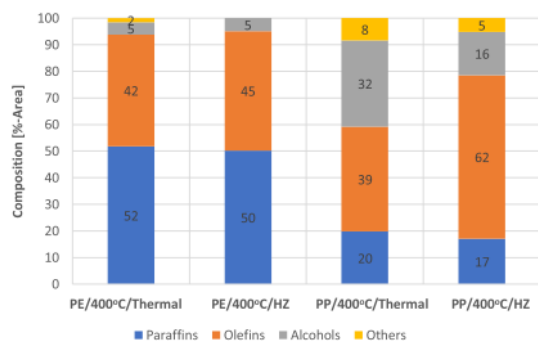|	Commercial fuel properties [41]		
	Density (g/mL)	Viscosity (cP)	Flash point (°C)
Gasoline	0.715–0.770	0.65	–
Kerosene	0.790–0.835	0.79	38
Diesel	0.815–0.870	0.83–1.04	52

Fig. 10 Functional groups of oil composition from thermal and catalytic pyrolysis of PE and PP

while mordenite 1-D zeolite hence aromatization was difficult to occur in both zeolite types.

Figure 11 presents an oil chromatogram for PE over the thermal and catalytic process. More details of compound and formula are presented in Table 2. As we can see from GC–MS results in Fig. 11 and Table 2, the large hydrocarbons increased in the catalytically pyrolysed oil. It most likely, some wax which was not converted into oil in the thermal pyrolysis of PE, after using natural zeolites converted into oil on the external and mesopore surface, hence the molecules obtained were still large hydrocarbons. Yan et al. [35] proposed a mechanism for thermal pyrolysis of PE and PP. For PE mechanism was dominated by intermolecular hydrogen transfer and β-disproportionate reactions. Hence, the main product was paraffin. For the catalytic pyrolysis of PE, Miandad et al. [44] reported the mechanism reaction over natural catalyst zeolites. Two possible mechanisms may involve the degradation of PE in the presence of a catalyst; hybrid ion abstraction due to the presence of Lewis sites or, due to the carbenium ion mechanism via the addition of a proton by Brønsted acid to the C–C bonds of the PE molecule followed by PE carbocation cracking into lower
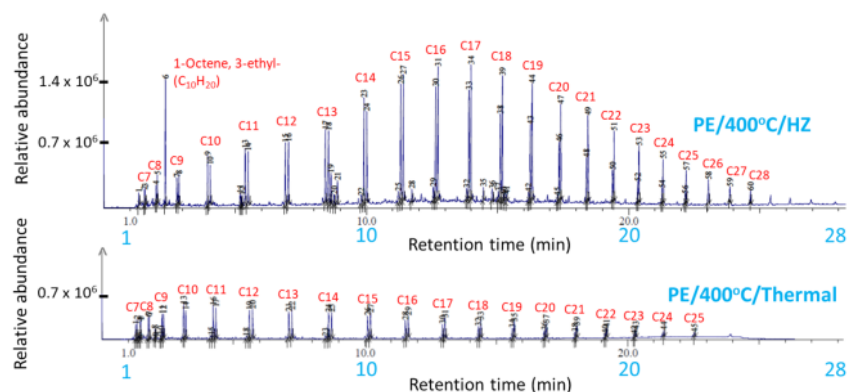
molecular weight hydrocarbons [45]. In our case, the acid treatment with HCl could lead to dominant Brønsted acid. Hence, the carbenium ion mechanism via a proton could be the dominant mechanism of pyrolysis. Since the polyolefin has a large molecular size, it can not enter the natural zeolite's pore. Thus, the cracking, either thermally or catalytically on the catalyst's external surface and mesopore surface, occurred at the beginning of the reaction. After that, the lower molecular can now enter zeolite's pore to further reaction. The type of zeolites drives the product selectivity, such as H-ZSM-5, which has a 3-dimensional pore with shape selectivity favored 18% naphthene and 77% aromatic products such as benzene, toluene, xylene [46].

Figure 12 presents the oil chromatogram for PP over thermal and catalytic processes. More details of compound and formula are presented in Table 3. The short chain hydrocarbons more observed in the catalytically pyrolyzed oil as compared with the thermal pyrolyzed oil of PP. It is most likely that the PP provide more reactive radicals which resulted on the conversion of all wax at 400 °C both in the thermal and catalytic process. It is most likely that the molecules was not only reacted on the external surface and mesopore but also in the micropore of zeolites. This make the oil from catalytic pyrolysis observed with short chain hydrocarbons, while the oil from thermal pyrolysis contain more large hydrocarbons. The highest chemical component was 2,4-Dimethyl-1-heptene both in thermal and catalytic pyrolysis. Zhang et al. [11] was found 2,4-Dimethyl-1-heptene as the highest component in the thermal pyrolysis of PP. They proposed the mechanism for thermal pyrolysis of PE and PP. According to Yan et al. [35] the higher content of olefins in PP thermal cracking products instead of alkane shows the intramolecular hydrogen transfer and β-scission reactions are the main mechanisms of pyrolysis. The catalytic degradation of PP is reported to proceed by carbenium ion mechanism [45, 47]. According to Kim et al. [47] The degradation reaction has initially occurred through

Fig. 11 Chromatogram of oil for PE at 400 °C from thermal and catalytic pyrolysis according to GC–MS

Table 2 GC–MS analysis of oil from thermal and catalytic pyrolysis PE at T 400 °C

No	Formula	Compound	% Area		Difference
			Thermal	Catalyst HZ	
1	$C_{16}H_{34}$	Hexadecane	3.40	5.16	1.76
2	$C_{17}H_{36}$	Heptadecane	3.33	4.66	1.33
3	$C_{15}H_{30}$	1- Pentadecene	3.51	4.47	0.96
4	$C_{18}H_{38}$	Octadecane	2.79	4.46	1.67
5	$C_{15}H_{32}$	Pentadecane	3.97	4.34	0.37
6	$C_{16}H_{32}$	1-Hexadecene	2.76	4.32	1.56
7	$C_{17}H_{34}$	1-Heptadecene	2.34	4.01	1.67
8	$C_{19}H_{40}$	Nonadecane	2.50	3.97	1.47
9	$C_{14}H_{28}$	1-Tetradecene	3.94	3.62	− 0.32
10	$C_{20}H_{42}$	Eicosane	2.16	3.35	1.19
11	$C_{18}H_{36}$	1-Octadecene	1.90	3.25	1.35
12	$C_{14}H_{30}$	Tetradecane	4.20	3.24	− 0.96
13	$C_{10}H_{20}$	1-Octene, 3-ethyl-	0.93	3.01	2.08
14	$C_{21}H_{44}$	Heneicosane	1.82	2.88	1.06
15	$C_{19}H_{38}$	1-Nonadecene	1.58	2.77	1.19
16	$C_{13}H_{28}$	Tridecane	3.90	2.43	− 1.47
17	$C_{13}H_{26}$	1-Tridecene	3.83	2.36	− 1.47
18	$C_{22}H_{46}$	Docosane	1.50	2.33	0.83
19	`$C_{20}H_{40}$	1-Eicosene	1.20	2.19	0.99
20	$C_{12}H_{24}$	1-Dodecene	4.23	2.13	− 2.1
21	$C_{12}H_{26}$	Dodecane	4.44	2.07	− 2.37
22	$C_{11}H_{22}$	1-Undecene	4.46	1.98	− 2.48
23	$C_{23}H_{48}$	Tricosane	1.23	1.83	0.6
24	$C_{11}H_{24}$	Undecane	4.15	1.78	− 2.37
25	$C_{21}H_{42}$	1-Heneicosene	0.90	1.58	0.68
26	$C_{10}H_{20}$	1-Decene	3.79	1.48	− 2.31
27	$C_{24}H_{50}$	Tetracosane	0.78	1.4	0.62
28	$C_{10}H_{22}$	Decane	3.38	1.37	− 2.01
29	$C_{22}H_{44}$	1-Docosene	0.66	1.09	0.43
30	$C_{26}H_{54}$	Hexacosane	0.00	1.06	1.06
31	$C_{25}H_{52}$	Pentacosane	0.59	1.03	0.44
32	$C_{13}H_{28}O$	1-Tridecanol	0.00	0.99	0.99
33	C_9H_{20}	Nonane	2.50	0.86	− 1.64
34	C_9H_{18}	1-Nonene	2.67	0.77	− 1.9
35	$C_{23}H_{48}$	1-Tricosene	0.45	0.72	0.27
36	C_8H_{18}	Octane	2.32	0.65	− 1.67
37	$C_{20}H_{38}$	1,19-Eicosadiene	0.00	0.56	0.56
38	C_7H_{16}	Heptane	1.44	0.52	− 0.92
39	$C_{24}H_{48}$	1-Tetracosene	0.00	0.50	0.50
40	$C_{27}H_{56}$	Heptacosane	0.00	0.49	0.49
41	$C_{14}H_{26}$	1,9-Tetradecadiene	0.50	0.48	− 0.02
42	$C_{20}H_{42}O$	1-Hexadecanol, 3,7,11,15-tetramethyl-	0.00	0.46	0.46
43	C_8H_{16}	1-Octene	2.63	0.4	− 2.23
44	$C_{28}H_{58}$	Octacosane	0.00	0.4	0.4
45	C_7H_{14}	1-Heptene	1.74	0.38	− 1.36
46	C_6H_{12}	1-Hexene	1.63	0.37	− 1.26
47	$C_{25}H_{50}$	1-Pentacosene	0.00	0.35	0.35

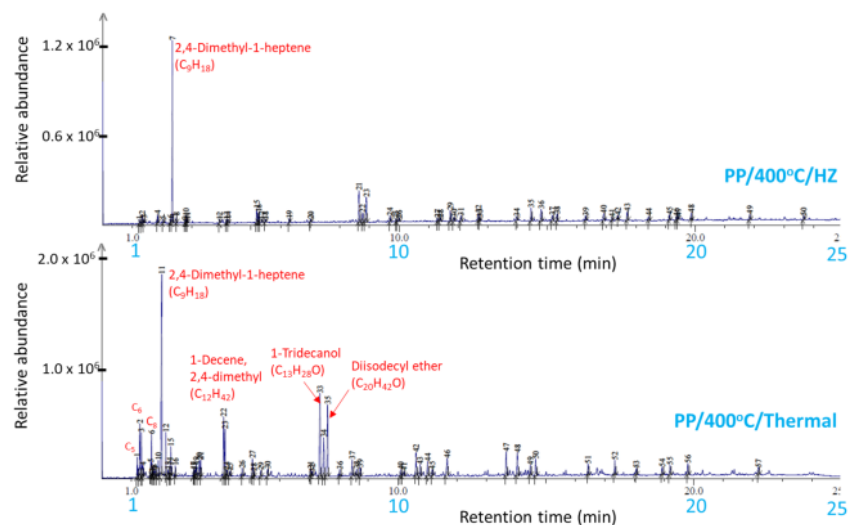**Fig. 12** Chromatogram of oil for PP at 400 °C from thermal and catalytic pyrolysis according to GC–MS

Table 3 GC–MS analysis of oil from thermal and catalytic pyrolysis PP at T 400 °C

No	Formula	Compound	Area (%)		Difference
			Thermal	Catalyst HZ	
1	C_9H_{18}	2,4-Dimethyl-1-heptene	16.32	32.98	16.66
2	$C_{12}H_{24}$	1-Undecene, 7-methyl-	0.00	6.58	6.58
3	$C_{12}H_{24}$	2,6,8-trimethyl-trans-4-nonene	0.00	5.29	5.29
4	$C_{12}H_{24}$	1-Dodecene	0.00	4.69	4.69
5	$C_{29}H_{60}O$	Nonacosanol	0.00	3.14	3.14
6	$C_{27}H_{56}O$	1-Heptacosanol	0.00	2.95	2.95
7	$C_{13}H_{26}$	2-Undecene, 4,5-dimethyl-	3.80	2.43	− 1.37
8	$C_{12}H_{24}$	1-Nonene, 4,6,8-trimethyl-	0.00	2.35	2.35
9	$C_{17}H_{34}$	4-Tetradecene, 2,3,4-trimethyl-	0.00	2.03	2.03
10	C_6H_{12}	1-Pentene, 2-methyl-	3.24	0	− 3.24
11	C_9H_{18}	Cyclohexane, 1,3,5-trimethyl-	2.53	0	− 2.53
12	$C_{12}H_{24}$	1-Decene, 2,4-dimethyl-	4.84	0	− 4.84
13	$C_{13}H_{28}O$	Tridecanol	8.11	0	− 8.11
14	$C_{19}H_{38}$	1-Nonadecene	6.86	0	− 6.86
15	$C_{20}H_{42}O$	Diisodecyl ether	4.11	0	− 4.11
16	$C_{20}H_{42}O$	1-Hexadecanol, 3,7,11,15-tetramethyl-	2.41	0	− 2.41
17	$C_{41}H_{84}O$	1-Hentetracontanol	2.57	0	− 2.57

abstraction of hydride ion by Lewis acid sites from the PP molecule or the addition of proton through Bronsted acid sites to the double bonds of the PP molecule or olefins which produced from thermal degradation. Successive scission of the main chain generated fragments with lower molecular weights than PP. The resulting decomposed fragments are further cracked in the subsequent steps. Thus, the molecular chain of PP catalytic pyrolysis is not wider than the thermal catalytic.

Potential and Challenges for Plastics Pyrolysis to Chemicals in Indonesia

Even though plastic pyrolysis is a promising technology for overcoming the problem of plastic waste, there are some challenges to using the technology in Indonesia, such as collecting plastics from people, non separated plastic with others waste. Typically, plastic waste is mixed with food waste and other biomass due to no separation conducted in every

house. Hence it makes it difficult to get high-quality plastic raw materials. The plastic waste is frequently dumped in an open space, so it gets wet by the rain. High moisture content could reduce energy efficiency and products yield. Investigation on the effect of a mixture of plastic with various typical biomass or food waste in landfills could be interesting to perform. Ahmed et al. [48] reported the co-pyrolysis of polyethylene and biomass (cellulose, lignin), which described that the pyrolysis of plastic materials in the biomass waste provided extra hydrogen, which synergistically contributed to the hydrodeoxygenation of 5-hydroxymethylfurfural and 4-methyl-1,2-Benzenediol. With various biomass sources, Indonesia could be co-pyrolyzed the biomass waste with plastics to obtain various chemicals. Our previous works on co-pyrolysis of biomass and coal suggested that chemicals product of co pyrolysis of biomass and coal affected by the type of the feedstocks, heating rate and temperature [49, 50].

Pyrolysis oil could be potentially utilized as co-feedstock with virgin naphtha for steam naphtha crackers. However, pretreatment of the pyrolysis oil must be prepared to remove contaminants, i.e., nitrogen, oxygen, chlorine, iron, sodium, and calcium, which favor coke formation, damage reactor tubes, or deactivate catalysts [51]. The concept of the chemical circular of polyolefins is presented in Fig. 13. Investigate the effect of catalysts on the oil impurities and pretreatment of the oil by adsorption technique using porous materials to eliminate the impurities also important to be performed. Gas product of PE and PP thermal pyrolysis components were reported mainly C_2H_6, C_3H_8, C_2H_4, and C_3H_6, with olefins as the highest elements produced [11, 14]. Ethylene and propylene could be utilized as raw materials for polyethylene and propylene. Gas products could be integrated with the burner system hence reducing the cost of energy for pyrolysis.
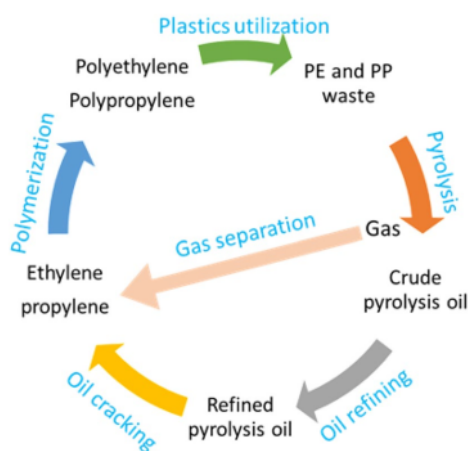

Fig. 13 Circular process diagram of polyolefins waste into chemicals

Recently, Hariadi et al. [52] suggested that the pyrolysis oil could be potentially applied as a solvent for natural asphalt, well-known as Buton asphalt in Indonesia. It has high hardness, which makes it cannot be used directly. It is necessary to modify the asphalt as one approach to improve performance. It is also necessary to add an additive to soften the asphalt. Additives that can be added include light oil, gasoline/naphtha, kerosene, diesel, or other diluents. One alternative as a solvent is oil produced by the pyrolysis of plastic waste. Better fractionation technology of pyrolysis oil for chemical raw materials could be applied to improve the quality of fuel and obtain various chemicals that can be used as raw materials for other products such as lubricant by hydroisomerization the pyrolysis oil [53].

The quality and yield of the oil could increase by using natural zeolites, which are available at low cost and easy to get in the country. The price of natural zeolites in the local shops is around 0.3 USD per kg. The pyrolysis oil product of liquid oils has been utilized by local businessmen using simple fractionation to obtain gasoline, diesel fuel, and kerosene. Some of them are sold commercially in motorcycles, cars, and small boats for the local fisherman. The energy sources for pyrolysis plastic come from biomass available in large amounts, particularly in remote areas. The quantity and quality of the pyrolysis oil could be improved by using the natural zeolites as in the present work.

Synthetic zeolite could also be investigated to enhance products' selectivity, such as ZSM-5 for the aromatic products (benzene, toluene, xylene) as reported by Lee et al. [46]. The three-dimensional pore of ZSM-5 allows cyclization and shape selectivity toward aromatics products. However, López et al. [54] reported that the ZSM-5 was easily deactivated by coke, which significantly reduced the catalyst activity. Due to the high cost of synthetic zeolites, it is needed to investigate the zeolites with high stability and or easily regenerate. Another type of synthetic zeolite, metal-impregnated zeolite-Y (nickel, iron, molybdenum, gallium, ruthenium, and cobalt), reported high selectivity to aromatics, i.e., toluene, ethylbenzene, and xylene, and the 2-ring aromatic hydrocarbons were naphthalene and alkylated naphthalenes [55].

Conclusion

The conclusions of the present work is presented below:

1. Pyrolysis polyethylene and polypropylene was successfully conducted to produce oil consisting of various hydrocarbon chemicals.
2. Wax could be generated during pyrolysis at low temperatures.

3. Catalytic pyrolysis over acid-treated natural zeolites (HZ) reduced the formation of wax and increased the oil yield compared to thermal pyrolysis.
4. The oil yield increased along with temperature for thermal and catalytic pyrolysis. PE and PP plastics pyrolysis over catalyst produced the highest oil yields at 450 °C with 67% and 70%, respectively.
5. Thermal pyrolysis of PP over catalyst increased the yield of olefins from 39 to 62% at 400 °C.
6. The oil composition mainly consisted of paraffin, olefins, and alcohols, with paraffin and olefins as the highest component of pyrolysis oil with feedstock PE and PP, respectively. Catalytic pyrolysis using low-cost natural zeolites can potentially be applied to improve the quantity of oil and increase the selectivity of olefins.

Data Availability All data generated or analysed during this study are included in this published article.

Declarations

Competing Interests The authors declare they have no financial interests.

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