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Fuel Oil Production from Municipal Plastic Wastes in Sequential Pyrolysis and Catalytic Reforming Reactors

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Abstract

The aim of this research was to study fuel oil production from municipal plastic wastes by sequential pyrolysis and catalytic reforming processes. Three kinds of municipal plastic wastes were collected from the final disposal site and the small recycling company in Yogyakarta city, Indonesia. Commercial Y-zeolite and natural zeolite catalysts were used in this study. The results show that the feedstock types strongly affect the product yields and the quality of liquid and solid products. HDPE waste produced the highest liquid fraction. The catalyst presences reduced the liquid fraction and increased the gaseous fraction. Furthermore, municipal plastic wastes pyrolysis produced higher heating value solid products than those of biomass and low rank coal.

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Keywords : pyrolysis; catalytic reforming; municipal plastic wastes; catalyst; natural zeolite

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Nomenclature

FCC	fluid catalytic cracking
HDPE	high density polyethylene
MPW	municipal plastic wastes
NZ	natural zeolite
PE	polyethylene
PET	polyethylene therephthalate
PP	polypropylene
PS	polystyrene
WPO	waste plastics oil

1. Introduction

The growth of the plastic consumption has been occuring rapidly in the last six decades due to their ability to be simply formed, its light weight and non-corrosive behavior. These excellent properties have been used to replace the use of wood and metals. The world’s annual plastic consumption has increased about 20 times from 5 million tons in 1950s to nearly 100 million tons [1]. In Indonesia, like many developing nations, plastic consumption has also increased due to economic development and rapid urbanization. In 2011, Indonesia has consumed 10 kg plastics per capita per year [2]. However, the great number of consumptions would increase the product of plastics wastes which led to the environmental problems. Landfilling is not a suitable option for disposing plastic wastes because of their slow degradation rates. The use of incinerator generates some pollutants to the air, which also cause environmental issues. Therefore, recycling and recovering methods have been used to minimize the environmental impacts and to reduce the damage of plastic wastes.

Chemical recycling via pyrolysis process is one of the promising method to recycle waste plastics which involves thermochemical decomposition of organic and synthetic materials at elevated temperatures in the absence of oxygen to produce fuels. The process is usually conducted at temperatures between 500-800^{o}C [3]. These pyrolytic products can be divided into liquid fraction, gaseous fraction and solid residues [4]. Pyrolysis or thermal degradation of plastics have been investigated by many researchers. There are four types of mechanisms of plastics pyrolysis [4] i.e. end-chain scission or depolymerization, random-chain scission, chain stripping and cross-linking. Thermal degradation behaviour of plastics has been investigated by Aboulkas et al. [5]. The activation energy and the reaction model of the pyrolysis of polyethylene (PE) and polypropylene (PP) have been estimated for non-isothermal kinetic results. The pyrolysis reaction models of polyethylene can be described by the “contracting sphere” model, whereas that of polypropylene can be described by the “contracting cylinder” model.

The low thermal conductivity and high viscosity of plastics are the major challenges for designing the cracking reactor. Several reactor systems have been developed and used such as batch/semi batch [6], fixed bed, fluidized bed, spouted bed, microwave [7] and screw kiln. Batch or semi-batch reactors have been used by many researchers because of its simple design and easy operation.

However, the thermal degradation of plastics has a major drawback such as very broad product range and requirement of high temperature. Catalytic degradation therefore provides a means to address these problems. The use of catalyst is expected to reduce the reaction temperature, to promote decomposition reaction, and to improve the quality of the products. The direct catalytic cracking has been used widely due to several advantages, mostly in terms of the energy efficiency, with regards to the use of the reactor, the reaction temperature and the residence time. However, the direct catalytic cracking of plastic wastes have a number of drawbacks which has prevented its commercial success. The first relates to difficulty to recover the catalyst after use, which increases the operational cost. Furthermore, direct contact with plastic wastes will make catalyst deactivate rapidly due to the deposition of carbonaceous matter and the poisoning effect of extraneous elements and impurities such as chlorine, sulfur and nitrogen containing species that may be present in the plastic wastes [3].

Therefore, separation of catalytic reforming reaction from pyrolysis stage can be applied to overcome these problems. This method has been studied by some researchers [8, 9] for PE and polystyrene (PS) using zeolite-Y and ZSM-5 catalysts. The results showed an increase in the gas yield and reduction in the oil yield. The use of other catalysts such as silica alumina and Al-MCM-41 have also been investigated by others [10, 11]. Catalytic reforming of waste agricultural polyethylene film over Al-MCM-41 catalyst produced heavier hydrocarbon products than HZSM-5 catalyst.

However, the use of catalyst is the main cost burden for recycling of plastic wastes by pyrolysis. Reducing the catalyst cost for small scale application in developing countries like Indonesia is very interesting challenges. Natural zeolites which can be found in many places worldwide including Indonesia might be used as a candidate for this purpose instead of the commercial catalysts.

A large number of papers have been published describing the pyrolysis of plastics. There are only few papers utilizing real plastic wastes as feedstocks. Plastic wastes can be classified as industrial and municipal plastic wastes according to their origins [4]. These groups have different qualities and properties. Industrial plastic wastes (IPW) are those arising from the plastics manufacturing and processing industry. Municipal plastic wastes (MPW) normally remain a part of municipal solid wastes as they are discarded and collected as household wastes. In Yogyakarta city-Indonesia, plastic wastes contributed 9.96% to the municipal solid wastes [12].

Pyrolysis and catalytic reforming of MPW which comprise PE, PP and PS have been studied by Wang and Wang [10] over nickel-loaded silica alumina catalysts. Bhaskar et al. [13] have compared the thermal degradation products from MPW and model mixed plastics. The presence of polyethylene therephthalate (PET) in model mixed plastics and MPW increased the formation of new chlorinated hydrocarbons in liquid products and also drastic decrease in the formation of inorganic chlorine content. The role of impurities in MPW was also significant. The impurities were toxic for acidic catalyst and led to easy deactivation of catalyst in the case of conversion of MPW [10].

Upgrading of pyrolytic oil produced from MPW has also been investigated using FCC catalyst as a cracking catalyst [14]. The addition of FCC catalyst in degradation process showed the improvement of liquid and gas yields and also high fraction of heavy hydrocarbons in oil product due to more cracking residue. Non-catalytic pyrolysis process has also been studied using waste PE, PP and PS [15]. The results showed that waste PS produced higher liquid while waste PE and PP produced higher gaseous products.

In this paper, we studied a sequential pyrolysis and catalytic reforming system for municipal plastic wastes degradation using commercial and Indonesian natural zeolite catalysts. Our system will utilize all kind of products as fuels including liquid, gaseous and solid products. This novel proposed system will utilize liquid product for fueling a diesel engine as a single fuel or blending with commercial diesel fuel. This oil can also be used in a pressurized cooking stove. The gaseous product can be used either as a heating source for the reactors or cooking gas stove application. The solid products will be used for co-firing with coal and biomass which can be utilized as a fuel for several applications.

2. Materials and methods

2.1. Materials

The feedstocks used for these experiments were three kinds of municipal plastic wastes, i.e. polyethylene bag with (PE bag 2) and without (PE bag 1) crushing and washing, and high density polyethylene (HDPE) waste after crushing and washing. They were obtained from the final disposal site and the small plastic recycling company in Yogyakarta city, Indonesia. The appearance of the feedstocks are shown in Fig. 1. The catalysts used for these works were commercial Y zeolite and natural zeolite.

The Y zeolite (CBV 780 CY) was obtained from Zeolyst International. It has SiO_2/Al_2O_3 mole ratio of 80, the unit cell size of 24.24 ⊕ and the surface area of 780 m^2/g in the powder form. The diameter of the pellet was 1.6 mm which contains 20% of aluminum oxide as a binder. Natural zeolite was collected from Klaten, Indonesia. The natural zeolite was calcined at 500°C for 3 hours to remove some volatile substances.

Fig. 1. The feedstock used in the experiments : (a) PE bag 1; (b) HDPE waste; and (c) PE bag 2.

The chemical properties and BET surface area of natural zeolite is shown in Table 1. In order to examine the crystalline structure in the natural zeolite, the XRD patterns of sample is shown in Fig. 2. It vary depending on their mining sites. It can be seen that the main structure of the natural zeolite catalyst was identified to be mordenite. Most of the peaks observed at 2θ (degree) = 5-35 for the natural zeolite samples can be assigned to be those of mordenite type crystalline matter as reported by Trisunaryanti et al. [16]. The samples showed relatively broad base lines. This suggests that the samples contain amorphous and crystalline impurities.

Table 1. Chemical composition and BET surface area of natural zeolite (NZ) .

	Si/Al ratio	Na/Si	Mg/Si	K/Si	Ca/Si	Fe/Si	S_{BET} ($m^2.g^{-1}$)
Natural zeolite	4.21	0.007	0.009	0.021	0.238	0.093	91.146

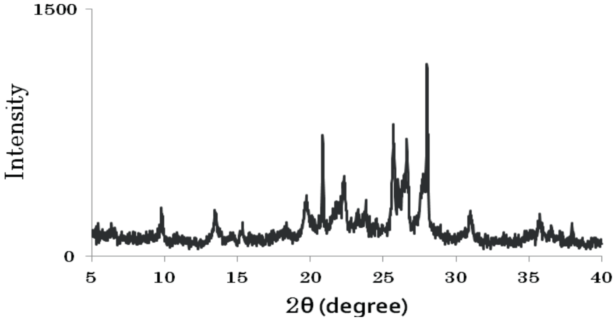

Fig. 2. X-Ray powder diffraction pattern of natural zeolite samples.

2.2. Pyrolysis and catalytic reforming experiments

Pyrolysis and catalytic reforming experiments were carried out in a pilot scale two stage reactor using batch system. It consists of the pyrolysis reactor and the catalytic reforming reactor. The snapshot of the experimental apparatus was shown in Fig. 3. The pyrolysis reactor and the reformer were made of stainless steel and covered with an electric heater. The pyrolyzer`s inner diameter and height are 200 mm and 400 mm, respectively. The reformer`s inner diameter and height are 100 mm and 400 mm, respectively. A shell and tube type condenser was installed at the outlet of the reformer to separate gas and liquid products.

In these experiments, 1.6-2.6 kg of the feedstock was fed into the pyrolysis reactor. The pyrolyzer and the reformer were then heated up to the preset temperatures. The catalyst (100 g) was loaded in the catalytic reforming reactor, where the pyrolysis gas generated in the first reactor was reformed. After having the reforming reaction, the gas was condensed into liquid products in the condenser. Liquid products were then collected and weighed for mass balance calculation. The experiments were carried out at the pyrolyzer temperature of 450°C and the reforming temperature of 450°C. The gaseous products were burned off to prevent emission from hydrocarbon gases.

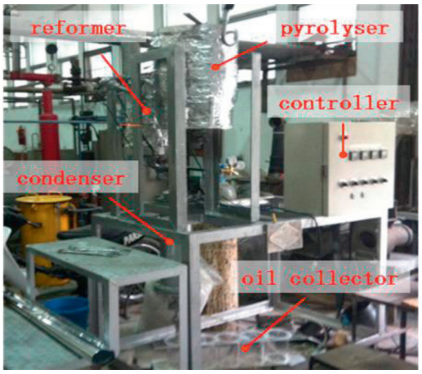

Fig. 3. The snapshot of experimental apparatus.

2.3. Liquid product analysis

The fraction of liquid products were analysed by using a gas chromatography-mass spectrometry (GC-MS, QP2010S Shimadzu). The column was DB-1 (Crossbond R 100% dimethylpolysiloxane) capillary column, 30m length with 0.25 mm diameter and 0.2 µm film thickness. Helium was used as the carrier gas. The temperature program used was, initial temperature 80°C for 5 min followed by a heating rate of 8 °C /min to 305 °C and then held at 305°C for 17 min. The other properties were analyzed based on ASTM methods as shown in Table 2.

3. Results and discussions

3.1. Effect of different types of feedstocks

The product yields as the effect of different types of the feedstocks can be seen in Fig. 4(a). Commercial Y zeolite was used in these experiments. PE bag 1 obtained from the final disposal site still produced water and highest portion of solid residue because of uncrushed and unwashed sample. It means that very high impurities were exist in the sample. The water might be obtained from organic material impurities which normally have high moisture content.

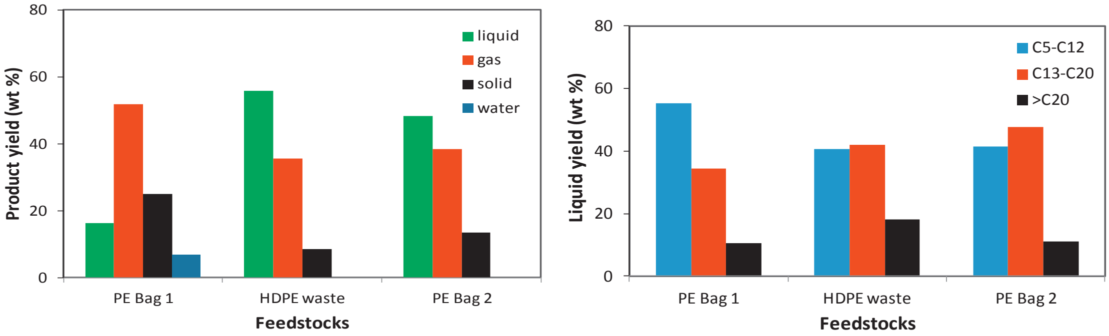

Fig. 4. Effect of different types of feedstocks on (a) the product yields; and (b) the liquid fraction composition.

These impurities have also led to a high percentage of gaseous products. The organic materials such as biomass produced high gaseous fraction up to 50% during pyrolysis process [17, 18]. All samples produced higher solid residues compared with those of pure plastics as reported by others which produce less than 5% of residues [19, 20]. It means that the impurities were dominated in the solid residues. HDPE waste produced highest liquid fraction and lowest gaseous fraction. The strong structure of HDPE made it more difficult to crack the hydrocarbon chains into lighter hydrocarbons.

However, the heavy oil fraction was still high in the oil from HDPE waste as shown in Fig. 4(b) indicating the low quality of the oil. The liquid products have been classified into three groups i.e. the gasoline fraction (C_5-C_{12}), diesel fuel fraction (C_{13}-C_{20}) and heavy oil (>C_{20}). As can be seen in Fig. 4(b), PE bag 2 produced highest diesel fraction while PE bag 1 produced highest gasoline fraction. As mentioned previously, the organic materials presence in PE bag 1 contributed to the high fraction of gasoline as reported by Lei et al. [21] for biofuel production. It was found that C_6-C_{14} chemical compounds were up to 95% of bio-oils. HDPE waste yielded lower diesel fuel fraction than that of PE bag 2. This is due to the different materials in PE bag 2 which consist mostly of low density polyethylene.

Table 2. Properties of liquid products for various feedstocks.

Properties	units	PE Bag 1	HDPE waste	PE Bag 2	Test method
Density @ 15°C	g/cm3	0.8544	0.7991	0.824	ASTM D1298
Kinematic viscosity @ 40°C	cSt	1.739	2.319	1.838	ASTM D 445
Flash point	°C	<10	<10	<10	ASTM D 93
Pour point	°C	24	27	24	ASTM D 97
Water content	%vol	0.1	0.5	trace	ASTM D95
Heating value	MJ/kg	41.45	42.82	46.67	ASTM D240

The properties of the liquid products from the pyrolysis and catalytic reforming of MPW are shown in Table 2. The properties of commercial diesel fuels in Indonesia are also shown in Table 3 for comparison. Indonesia produced two kinds of diesel fuels viz. Diesel 48 (Solar) and Diesel 51 (Pertamina Dex). As can be seen in Table 2 and 3, the density of waste plastics oils (WPO) is acceptable for substituting commercial diesel fuels.

However, the kinematic viscosity of WPO was lower than those of commercial diesel fuels for PE bag 1 and PE bag 2. The higher fraction of gasoline and the lower fraction of heavy oil contributed to the lower kinematic viscosity. Viscosity play significant role in the lubrication of fuel injection systems, particularly those incorporating rotary distributor injection pumps that rely fully on the fuel for lubrication within the high pressure pumping mechanism [22]. Lower fuel viscosity lead to greater pump and injector leakage reducing maximum fuel delivery and ultimately power output. The flash points were lower than those of diesel fuels. The flash point is an important parameter in relation to fuel storage. Higher flash point will be more safe for storing and transporting the fuels.

Table 3. Properties of commercial diesel fuels according to Indonesian Government regulation [23].

Properties	units	Diesel 48 (Solar)	Diesel 51 (Pertamina Dex)
Cetane number		48	51
Density @ 15°C	g/cm3	0.815-0.870	0.820-0.860
Kinematic viscosity @ 40°C	cSt	2.0-5.0	2.0-4.5
Flash point	°C	min 60	min 55
Pour point	°C	max 18	max 18
Water content	mg/kg	max 500	max 500
Sulfur content	%wt	max 0.35	max 0.05
Ash content	%wt	max 0.01	max 0.01

WPO produced higher pour point than those of diesel fuels since the presence of heavy oil which normally has high pour point. The property becomes very important when diesel engine running in very low temperature condition especially in subtropical countries. The water content presence in WPO was high. It will affect to the performance of diesel engine.

3.2. Effect of catalysts

Fig. 5(a) shows the product yields obtained from the sequential pyrolysis and catalytic reforming of municipal plastic wastes as the effect of catalysts. PE bag 2 has been used as a feedstock in these experiments. It can be seen that the thermal pyrolysis (without catalyst) produced highest liquid fraction. The presence of catalyst reduced the liquid fraction and increased the gaseous fraction. Theoretically, the catalyst can enhance the cracking reaction of the pyrolysis gas. Long chain hydrocarbons have been cracked into lighter hydrocarbon gases. Pyrolysis over natural zeolite catalyst produced higher liquid product compared with Y zeolite catalyst. This is due to different activity between natural zeolite and Y zeolite. As can be seen in Table 1, NZ has lower BET surface area than that of Y zeolite. Higher surface area will give more contact between catalyst and pyrolysis gas which means more gas will be cracked to produce shorter chain hydrocarbons.

However, the presence of catalysts has slightly effect to the product yields. This might be due to the presence of impurities as mentioned previously. The impurity which contains some toxic materials will deactivate the catalysts. Thus, the catalysts will have the activity in the beginning of the reforming process and deactivate in the end of the process.

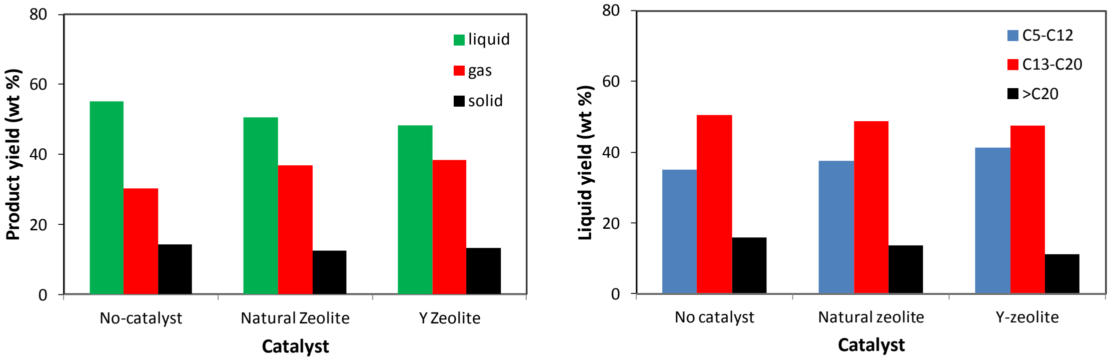

Fig. 5. Effect of catalysts on (a) the product yields; and (b) the liquid fraction composition.

Fig. 5(b) shows the carbon atom number distribution of WPO over different catalysts. The heavy oil fraction ($>C_{20}$) could be slightly reduced which affect to the quality of the oils. On the other hand, the gasoline fractions (C_5-C_{12}) were increase because of cracking of longer chain hydrocarbons into lighter chain hydrocarbons. The diesel fraction was almost similar in all conditions. This is because the balance between the addition of diesel fraction from heavy oil cracking and the reduction of diesel fraction cracked into gasoline fraction.

Table 4 shows the properties of liquid products over different catalysts. Similar results have been obtained for all properties. The presence of the catalysts slightly decreased the pour point. However, the values were still higher than those of commercial diesel fuels. This condition will make WPO become solid in low temperature condition. The problems with high pour point of WPO can be overcome by using additional heater before injecting the fuel to ensure fluidity and keep the viscosity of the fuels. The heating value of WPO was similar to the common commercial fuels due to the same origin of plastics and commercial fuels which are produced from petroleum oil.

Table 4. Properties of liquid products for different catalysts.

Properties	units	No catalyst	Y Zeolite	Natural Zeolite
Density @ 15°C	g/cm3	0.8719	0.824	0.868
Kinematic viscosity @ 40°C	cSt	1.999	1.838	2.191
Flash point	°C	<10	<10	<10
Pour point	°C	27	24	24
Water content	%vol	0.5	trace	trace
Heating value	MJ/kg	46.74	46.67	45.58

3.3. Solid residues

The proximate analysis and heating value of the solid residue produced are shown in Table 5. The results show that higher ash content of solid residues produced from MPW compared with raw plastics as reported in previous works [23]. PE bag 1 produced higher ash content of solid product than others due to the high impurities which reduce the heating value of the product. PE bag 2 has lowest ash content and highest heating value. In waste plastics, fixed carbon and volatile matter contributed to the high heating value of solid products. Unlike biomass, volatile matters in waste plastics contain mostly hydrocarbon gases which have high energy content. Therefore, plastic pyrolysis produced higher heating value solid products than those of biomass and low rank coal as reported by others [24, 25]. Therefore, they can be used either for blending with biomass and coal or for single fuel.

Table 5. Proximate analysis of solid residues.

	Composition (wt.%)		
	PE Bag 1	HDPE waste	PE Bag 2
Moisture	1.12	1.74	2.32
Volatile matter	35.29	58.56	44.47
Fixed carbon	14.13	8.59	25.88
Ash	49.47	31.11	27.33
Heating value (MJ/kg)	19.80	26.35	31.53

4. Conclusion

Sequential pyrolysis and catalytic reforming of Indonesian municipal plastic wastes have been done over Y-zeolite and natural zeolite catalysts. The results show that the feedstock types strongly affect the product yields and the quality of liquid and solid products. HDPE waste produced the highest liquid fraction. However, the heavy oil fraction was still high in the oil from HDPE waste. The highest diesel fraction has been produced in PE bag 2 while PE bag 1 produced highest gasoline fraction. The catalyst presences reduced the liquid fraction and increased the gaseous fraction. Pyrolysis with natural zeolite catalyst produced higher liquid product compared with Y zeolite catalyst. However, the presence of catalysts has slightly effect to the product yields. This might be due to the presence of impurities in MPW. The quality of WPO was still lower than those of commercial diesel fuels according to the oil properties. Blending of WPO and diesel fuels will obtain better quality of oil. MPW pyrolysis produced higher heating value solid products than those of biomass and low rank coal, so that they can be used either for blending with biomass and coal or for single fuel.

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