EFFECT OF VARIOUS CATALYSTS ON FUEL OIL PYROLYSIS PROCESS OF MIXED PLASTIC WASTES

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ABSTRACT

In this study, mixed plastic wastes were converted into liquid fuels. Mixed plastic wastes used were high density polyethylene (HDPE), low density polyethylene (LDPE), polystyrene (PS), polypropylene (PP) and polyethylene-terephthalate (PET). The pyrolysis processes were carried out with fixed bed reactor. Natural catalyst such as Shwedaung clay, Mabisan clay, Bentonite clay, Dolomite and synthetic catalyst, zinc oxide, were used as catalyst for pyrolysis of mixed plastic wastes. These natural catslysts were naturally found in Myanmar and zinc oxide was collected from local market. In this study, sample was used 200g (HDPE, PP, LDPE, PS, PET = 40, 30, 22.5, 5, 2.5 wt %) and 20g of catalysts was used to accelerate the reaction. The experiments were carried out at temperature between 32 to 380 °C by using various catalysts. From this study, yields of liquid fuel were found to be 63.76 - 67.06% and yields of gas were 19.92 – 28.52%. The highest yield (67.06%) of liquid fuel was obtained from mixed plastic wastes pyrolysis process by using Mabisan clay at the range of temperature 220 to 370 °C for 1.5 hour. Fuel density was 0.7016 g/ml and kinematic viscosity was 2.26 mm2/sec. FTIR spectrum results of product crude liquid fuel showed significantly in alkane group.

KEYWORDS: Mixed Plastic Wastes, Product Fuel, Pyrolysis, FTIR, Catalysts

I. Introduction

The rapid rate of plastic consumption throughout the world has led to the creation of increasing amounts of waste and this in turn poses greater difficulties for disposal. As a brief introduction to plastics, it can be said that plastics are synthetic organic materials produced by polymerization. Plastics are also called polymers. It is non-biodegradable polymers mostly containing carbon, hydrogen, and other elements such as chlorine, nitrogen, etc. Plastics are light weight and durability. Plastic wastes can be classified as industrial and municipal waste plastic according to their origins. They have different qualities and properties. Waste plastic represents a considerable part of municipal wastes; furthermore huge amount of waste plastic arise as a byproduct or faulty product in industry and agriculture. The plastic wastes are mainly two types of plastics such as thermo plastics and thermosetting plastics. Thermo plastics are composed of polyolefin such as high density polyethylene (HDPE), low density polyethylene (LDPE), polypropylene (PP), polystyrene (PS), polyethylene terephthalate (PET) and polyvinyl chloride (PVC) and can be recycled. Thermosetting plastics mainly include epoxy resins and polyurethanes and cannot be recycled [1]. The quantity of thermoplastics found in waste is increasing corresponding.

Nowadays, the largest amount of plastic wastes is disposed of by land filling, and incineration. So, Environmental pollution increases day by day in everywhere. Plastic wastes are the main causes of environmental pollution. There are three ways to management of plastic wastes. They are land filling, incineration and recycling. The problem of wastes cannot be solved by land filling and incineration, because suitable and safe depots are expensive, and incineration stimulates the growing emission of harmful, greenhouse gases, e.g. NO_x, SO_x, CO_x. Therefore, recycling method was used in this study. Recycling has many types of techniques including pyrolysis, gasification, hydrolysis and etc. Among them, the most attractive technique of recycling is pyrolysis or thermal cracking. Pyrolysis refers to

the thermal decomposition of the matter under an inert gas like nitrogen. In pyrolysis process, macromolecular structures of polymer are break down into smaller molecules. It can be reduced the reaction temperature. The pyrolytic products can be divided into a gas fraction, a liquid fraction consisting of paraffins, olefins, naphthenes and aromatics, and solid residues. There are two types of pyrolysis or cracking process such as thermal cracking and catalytic cracking. Thermal cracking involves the degradation of the polymeric materials by heating in the absence of oxygen. In thermal cracking, the temperature is about 350°C to 900°C. In catalytic cracking, a suitable catalyst is used to carry out the cracking. The present of catalyst lowers the reaction temperature and time. Addition of catalysts enhances the conversion and fuel quality [2]. If the polymers have lower activation energy, which have considerable lower against degradation. Due to the low activation energy of polymers can be degraded easily. So, higher amount of liquid and gases could be obtained the cracking of polymer result [3].

In order to understand and optimise the pyrolysis of waste plastic and to investigate the impacts of different types of the plastics, extensive research has been conducted in the past decade [4]. Kaminsky, Scheirs and their colleagues [5-9] investigated the effects of reaction conditions on the pyrolysis product. Williams studied the products from pyrolysis of different individual and mixed plastics [10-15].

Aguado *et.al.* investigated the effect of catalysts on the pyrolysis reactions [16-20]. Most studies focused on the effects of operation temperature, heating rate, and catalysts on the product yield. However, there are few researches in the literature review investigating the cracking process of the pyrolysis products during the pyrolysis which is believed to be complex. In addition, the final products are also very complicated. There could be over a hundred of components in the hydrocarbon products including paraffin, olefin and their isomers. Paraffins are saturated hydrocarbons with straight or branched carbon chain, which are also called "alkane". Olefins have similar chain as paraffins, but they have one or more multiple bonds between carbon atoms in their chains. Naphthenes are saturated hydrocarbons like paraffins but their chains merge to a ring in their structure. Aromatics contain a benzene ring in the structure. Another common way to describe the hydrocarbons is based on the carbon numbers in their molecule structure. It is particularly applied to petroleum fuels. The complex pyrolysis products may also be grouped as petroleum gases petrol, kerosene, diesel and wax [4, 21].

Catalytic degradation of plastics is found to have greatest potential to be developed into a commercialized process. One of the most successful examples being the Alka Zadgaonkar's Unique Waste Plastic Management and Research Company plant in India which could produce fuel oil from waste plastics at par with the regular gasoline [22].

There are two types of catalysts for cracking of waste plastics. They are natural catalyst and synthetic catalyst. Natural catalysts are such as natural silica-alumina composition, treated bentonite clay, aluminum hydrosilicates and so on. Synthetic catalysts are synthetic silica-alumina, silica-magnesia, alumina - boria, zeolite, activated carbon and Alkaline compounds. Natural catalysts are cheaper than synthetic catalysts and they are naturally found in Myanmar instead of synthetic catalysts. The useful catalysts are available in the pellets form or powder form [23].

In this study, liquid fuel was produced from mixed plastic wastes (HDPE, LDPE, PP, PS and PET) by using natural catalyst such as Kaolin clay, Mabisan clay, Bentonite clay Dolomite and by synthesis catalyst as Zinc oxide in fixed bed reactor. The objective of this study is to produce liquid fuel from mixed plastic wastes by using pyrolysis process with various catalysts.

II. MATERIALS AND EXPERIMENTAL STUDY

2.1. Materials

In this study, equipments and chemicals were used for preparation and natural catalysts. Equipments and apparatus were also used for preparation of raw materials.

2.1.1. Mixed Plastics

High density polyethylene (HDPE), low density polyethylene (LDPE), polystyrene (PS), polypropylene (PP) and polyethylene-terephthalate (PET) wastes were used as raw materials. These

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plastics were collected locally as used laundry bottles, shampoo bottles, soft drink bottles, water bottles, juices cups, various foam, shopping bags and plastic bags.

2.1.2. Catalysts

Four different natural catalysts such as Shwedaung clay, Mabisan clay, Bentonite clay and Dolomite from different places were used to pyrolyze the mixed plastic wastes. Natural catalysts such as clays occur in nature in our country. Zinc oxide was used as a synthesis catalyst and it was collected from local market.

2.2. Experimental Study

In this experimental work, sample preparation, preparation of catalysts and experimental process were included.

2.2.1. Sample Preparation

For calculation of bulk densities of various plastics wastes, firstly, they were prepared in the laboratory by collecting from household wastes, washing by liquid detergent and drying. Then, they were chopped by crushing machine for the experiments. Selected particle size range to set was between 6mm and 10 mm. Prepared raw materials were weighted on basis one unit volume by using digital balance. Finally, bulk densities of HDPE, PP, LDPE, PS and PET were 419 kg/m 3 , 386 kg/m 3 , 52 kg/m 3 , 17 kg/m 3 and 286 kg/m 3 respectively.

2.2.2. Preparation of Catalysts

Prior to use, the four natural catalysts and synthetic ZnO catalysts were pelletized by moulding. The advantages of pelletizing the catalyst powder are to put easily in catalytic bed and to reduce the pressure drop. In this study, catalyst pellets were used by mixing with starch powder. By blending with starch powder, void fractions between the inter-particles of catalyst pellets for various catalysts were increased. To prepare catalyst pellets, twenty grams of catalysts with one gram of starch powder was done by blending. Starch powder binder was added 1% by weight of catalysts and 10 ml of water was added in catalyst mixture for catalysts pelletizing. The catalyst pellets were activated by heating (800-850°C) for 3 hours. Average size of applied natural catalyst pellets were 0.8 cm diameter and 0.3 cm thickness. Various types of catalyst pellets were shown in Figure 1.



Figure 1. Natural catalyst pellets

2.2.3. Experimental Process

The necessary equipments for waste plastics cracking were mainly consisted of electric coil furnace, reactor, electric sensor with connected temperature control box, nitrogen gas cylinder, condenser units, water, fuel container and Teflon bag for collection of gases. Reactor and condensers are significant role in pyrolysis process. The function of reactor is to break down the long chain hydrocarbon composed in various plastics. The reactor has been designed as a cylindrical shell for the solid waste pyrolysis. The reactor is fixed bed reactor. The condenser is provided to cool and condense the gases evolved out from the pyrolyser. As a result of cooling, the condensable gases become liquid which is called pyrolytic oil. The condenser is a type of shell and tube heat exchanger. The evolved gases from the pyrolyser pass through the tubes and the cooling water is circulated on the shell side. It has 1-shell pass and 1-tube pass counter flow type of arrangement. The required units for reactors and condensers were constructed in the local workshop. In this study, two stages condensers were used. Product crude fuel and wax would be flowed out in first condenser and the residual liquid fuel vapour condensed in second condenser.

The mixture of plastic wastes and catalysts were placed into the reactor. This reactor was placed in the electric coil furnace. The reactor and its cover were tightly closed with bolts, nuts and rubber seal to prevent from gas leakage. After that, reactor and condenser were connected with flanges, First and second condensers were joined by using elbow pipe. Boiler was used for hot water in first stage condenser and water cooler was used for cold water in second condenser. Two conical flasks were fitted on the elbow pipe to collect the crude fuel oil. Experimental setup was shown in Figure 2. To do catalytic cracking, 200 g of mixed plastic wastes (HDPE, PP, LDPE, PS, PET = 40, 30, 22.5, 5, 2.5 wt %) and 20 g of each catalysts pellets were placed into the reactor. Nitrogen was passed through before starting experiment to remove oxygen from the reaction medium. Heat applied between 32°C and 400°C. Firstly, the reactor was heated and temperature was slowly raised with the rate of 5°C min ¹.Plastics were started to melt when decomposition temperature was reached. When mixed plastic wastes were complete melted and turn to liquid slurry via liquid slurry to vapour. The organic vapour passed through the catalytic bed for catalytic cracking. After that, the vapours were condensed as liquid and wax in first condenser. In all experiments, wax was not produce much. The first dropped of liquid fuel when temperature goes above 210°C. The residual liquid fuel vapours were condensed when passed through second condenser. Crude liquid fuel was collected at the end of each condenser. During fuel production, non-condensable gases were collected into Teflon bag. The whole experiment time was 1.5 hour. After experiment finished, solid residues (chars) were collected in the reactor.



Figure 2. Experimental setup for fuel oil production process

III. RESULTS AND DISCUSSION

3.1. Analysis of Natural Catalysts

Four different natural catalysts were identified by X-ray fluorescence (XRF). Chemical compositions of four natural catalysts are shown in Table 1.

Component	Mabisan Clay (%)	Shwedaung Clay (%)	Bentonite Clay (%)	Dolomite (%)
0	40.66	39.63	38.18	36.6
Na	0.381	0.085	0.954	-
Mg	0.154	0.163	1.059	10.14
Al	15.06	19.23	8.748	0.669
Si	22.78	25.16	24.51	0.985
Cl	-	-	0.052	-
P	0.011	0.018	-	0.025
S	0.028	0.016	-	0.0544
K	0.973	1.32	1.19	-
Ca	2.957	0.0998	1.79	32.75
Ti	0.33	0.496	0.421	0.034
Cd	-	-	-	6.11

Table 1. Chemical composition of natural catalysts

It could be seen that the highest amount of Si and Al contented in Mabisan clay and Kaolin clay. These compounds could increase the cracking efficiency. The composed of Si and Al in dolomite were smaller than other natural catalysts. But the largest amount of Ca and Mg can be found in dolomite.

3.2. Analysis and Yield Percentages of Product Liquid Fuels

The physical properties of liquid fuel such as density, kinematic viscosity and experiment reaction temperature were described in Table 2. According to Table 2, liquid fuel density obtained by using Shwedaung, Mabisan and Dolomite were not significantly different. Density of fuel obtained by using bentonite clay and Zinc oxide catalysts was lower than that of other. In analysis of kinematic viscosity, fuel obtained by using Mabisan clay was lowest in all. It can be seen that Mabisan clay provided good cracking activity.

Catalysts used	Liquid fuel density at 19°C (g/cm³)	Kinematic viscosity of liquid fuel at 19°C (mm²/s)	Reaction temperature of crude fuel start to drop to finished Temperature°C
Shwedaung clay	0.7031	2.82	210 – 365
Mabisan clay	0.7016	2.26	220 – 370
Bentonite clay	0.7016	2.49	218 – 377

2.65

2.56

Table 2. Physical properties of crude liquid fuels

The product yield such as oil, gas and char from pyrolysis process were presented in Table 3. Crude liquid fuel obtained two layers by using various catalysts. The upper layer was crude liquid fuel and the bottom layer contained wax. It can be seen that some wax contained in crude liquid fuel. However, the yield % of wax contained small amount in product fuel.

Table 3. Comparison of product yields by using various catalysts

Mixed plastic wastes (HDPE, LDPE, PP, PS and PET): 200 g Reaction Temperature : 210 - 380 $^{\circ}$ C Reaction Time : 1.5 hour

0.7036

0.7021

Dolomite

ZnO

Catalyst used	Yield (wt%) based on feed					
	Liquid fuel	Wax	Char	Gas fuel	Colour	
Shwedaung clay	65.81	1.07	11.34	21.78	Brownish Yellow	
Mabisan clay	67.06	0.59	12.43	19.92	Brownish Yellow	
Bentonite clay	64.92	1.11	12.11	21.86	Brownish Yellow	
Dolomite	63.96	0.65	6.87	28.52	Brownish Yellow	
Zinc Oxide	63.76	No wax	8.23	28.01	Yellow	

According to Table 3, the yield % of wax obtained from cracking process with Mabisan Clay was lowest in all catalysts. According to Table 3, the yield % of product liquid fuels using Mabisan clay was higher than that of other catalysts. Besides Shwedaung clay gave the second highest yield compared to other three catalysts. Because Mabisan clay and Shwedaung clays were possess good cracking properties because they have been composed the large amount of the chemical components such as Si, Al, Mg and Ca and these compound could promote the cracking efficiency. The yield % of product liquid fuels using synthetic catalyst as ZnO was lower than product oils by using natural catalysts. According to these results, natural catalysts were suitable catalysts for cracking process of mixed plastic wastes because the highest oils yield % was obtained by using natural catalysts and they are naturally occur in Myanmar instead of synthetic catalyst as ZnO.

224 - 358

223 - 370

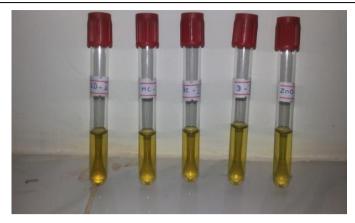


Figure 3. Product fuel from mixed plastic wastes by using various catalysts

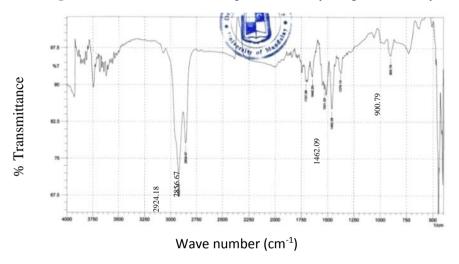


Figure 4.a. FTIR spectrum of mixed plastic wastes to liquid fuel with Shwedaung clay

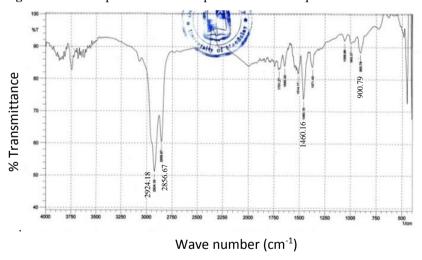


Figure 4.b. FTIR spectrum of mixed plastic wastes to liquid fuel with Mabisan Clay catalyst

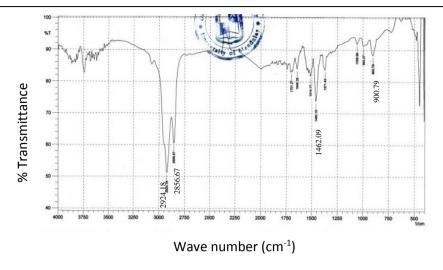


Figure 4.c. FTIR spectrum of mixed plastic wastes to liquid fuel with Bentonite Clay catalyst

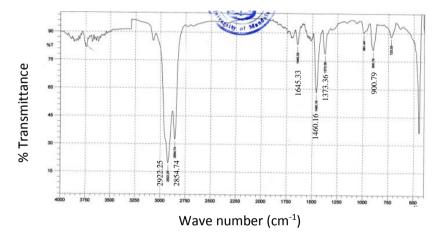


Figure 4.d. FTIR spectrum of mixed plastic wastes to liquid fuel with Dolomite catalyst

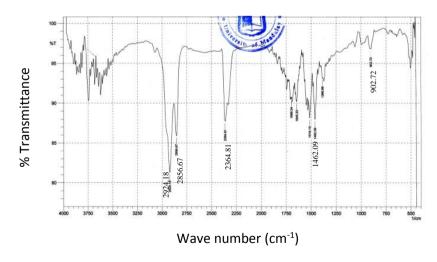


Figure 4.e. FTIR spectrum of mixed plastic wastes to liquid fuel with Zinc Oxide Catalyst

FTIR analyses of mixed plastic wastes to liquid fuel by using various catalysts were shown in Figure 4a, 4b, 4c, 4d and 4e. According to Figure 4a, the following types of functional groups in wave number were significant appeared in the same analyses. In wave number 2924.18 cm⁻¹ and 2856.67 cm⁻¹ functional group is C-CH₃, wave number 1462.09 cm⁻¹ functional group is CH₂, wave number 1379.15 cm⁻¹ functional group is CH₃, wave number 900.79 cm⁻¹ functional group -CH=CH-

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respectively. According to Figure 4b, 4c, 4d and 4e, functional groups of wave numbers were similar to Figure 4a in the analysis of FTIR spectrum. In the various analyses, some groups are emerged single and double bonded functional groups. Methyl and methylene groups are significantly seen in the same analysis spectrum. Therefore, alkane (paraffins) group were significantly appeared in all product fuel oils according to all figures. The product fuels might be gasoline range. Moreover, all product fuels from mixed plastic wastes did not contain aromatic compounds producing smoke because this compound can be appeared in wave number 3000 to 3100 cm⁻¹.

IV. CONCLUSIONS

In this study, Shwedaung clay, Mabisan clay, Bentonite clay, Dolomite and zinc oxide were used as catalyst for pyrolysis of mixed plastic wastes. Mixed plastic wastes include HDPE, LDPE, PP, PS and PET. Pyrolysis processes were carried out at the temperature range of 32 - 380°C for 1.5 hour. By comparing the catalytic activity, the yield (%) of sample by using Mabisan clay is the highest yield. In this study, the maximum crude liquid fuel yield based on the feed mixed plastic wastes was 67.06% and the maximum gas fuel yield was 28.52 %. Fuels yield % obtained by using natural catalysts and by using synthesis catalyst were not significantly different. So, it was concluded that natural catalysts should be used because they can be occurred in nature and they possess good cracking efficiency due to their chemical composition. Product fuels were analyzed by FTIR. The results of product fuel were significantly appeared as alkane and alkene groups. Product fuels were mainly paraffins and composed of more alkane group. Fuel can be used internal combustion engine after distillation. In addition, waxes obtained from plastic cracking can be used as petroleum waxes and char can be used as solid fuel.

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