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Analysis of Natural Zeolite Ratio Variables and Process Temperature for Increasing Yield and Heating Value of Liquid Fuel from Plastic Bags Waste Using Mini Plant Pyrolysis

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Abstract
The demand for fuel oil derived from fossils is increasing day by day, causing the depletion of oil and natural gas reserves. According to Dudley (2015), world oil reserves at the end of 2014 was around 1700.1 billion barrels, whereas Indonesia only had proven oil reserves of 3.7 billion barrels and this figure was only 0.2% of the total world oil reserves. Meanwhile, plastic waste material is an appropriate alternative as a starting material for hydrocarbon sources and can be a potential source of hydrocarbons with a relatively high hydrogen/carbon ratio compared to coal (Antal *et al*., 2000) [1]. The type of plastic waste that is often a problem in various cities in Indonesia is the type of plastic bag (LDPE), this is because of waste plastic bags are not saleable in the market. From the various existing literatures, the research is still on a laboratory scale, so further studies and tests are needed at a larger scale level in the form of a prototype. This research is intended to carry out various analyzes of the Reactor Temperature at (130, 140, 150, 160, 170 ºC) and the Ratio of Zeolite to LDPE raw material at (0.00, 0.04, 0.08, 0.12, 0,16). The Pyrolysis Reactor Prototype Unit can be used to convert plastic bag waste (Low Density Polyethylene or LDPE) into liquid fuel with a percent yield of 57.43% or 7.67 kg of the total mass of plastic waste raw material of 10 kg. In this research found parameter optimal of Zeolite Ratio at 0.12 and the process temperature at 160 ºC with a calorific value of 19750 Btu/lb.

Keywords: LDPE, Plastic, BBC

1. Introduction

The community's need for fuel oil (BBM) derived from fossils is increasing day by day, causing the depletion of oil and natural gas reserves. According to Dudley (2015) [5], world oil reserves at the end of 2014 was around 1700.1 billion barrels, while Indonesia only had reserves proven oil around 3.7 billion barrels which is only 0.2% of the total oil reserves in the world. Total oil production is 852 thousand barrels/day with consumption of 1.641 million barrels/day. From the data above, there is an imbalance between production and consumption, resulting in availability fuel in Indonesia will enter a critical stage. The issue of the depletion of world oil reserves (Marcilly, 2003) [7], is increasingly demanding a tactic and strategy for the main source of these hydrocarbons in an economical and efficient manner. Also considering that the process of its formation in a geological time frame requires thousands of years, these fossil energy sources are referred to as non-renewable resources.

On the other hand, plastic waste material is an appropriate alternative as a starting material for hydrocarbon sources and can be a potential source of hydrocarbons with a relatively high hydrogen/carbon ratio compared to coal (Antal *et al*., 2000) [1]. The type of plastic waste that is often a problem in various cities in Indonesia is the type of plastic bag (LDPE) which can still be sold at a price of 2000 to 3000 Rupiah per kilogram. So that plastic bag waste have really become hazardous waste and are difficult to manage. It is spread across city so that they become a very urgent problem to overcome.

Indonesia Ministry of Environment and Forestry notes that the use of plastic bags classified as (LDPE) in Indonesia, is more than 1 million sheets per minute. Every year, the production of plastic bags consumes around 8% of world oil production or around 12 million barrels of petroleum fuel.

The reliability of plastic bags belonging to the type of Low-Density Polyethylene (LDPE) to be used as fuel is not only limited to the amount that can be consumed continuously as a raw material, but also the component formulas inside have great potential. The main constituent component consists of polyethylene which is free from other chemical compositions such asterephthalate which is present in PET plastic types (polyethylene terephthalate), PVC (Polyvinyl Chloride) and other types so that during the conversion process into fuel it does not contain much residue which can reduce the percent yield and becomes a factor of scale or impurities in the piping system used. Another advantage of plastic bag waste is that the melting point is relatively much lower than other types of plastic. The melting point of LDPE plastic is around 115ºC, while for HDPE (High Density Polyethilene) it is around 135ºC. The melting temperature is low from plastic waste will provide benefits in terms of process because the heating temperature in the reactor will be lower than other plastic waste raw materials.

Many studies have been carried out on converting LDPE plastic waste, especially plastic bags, and have obtained various operating conditions and yield values produced. It has been recorded since 2012 that Moinuddin Sarker from the United States Department of Research and Development has carried out a degradation process in a pyrolysis reactor at a temperature of 250 ºC and the resulting yield reached 40%. In 2014, P. Premkumar from Annamalainagar University in India, converted LDPE plastic waste in a pyrolysis reactor at 250 ºC to obtain 35% liquid fuel formation. Stepping on in 2017, S.L. Wong from the University of Technology Malaysia carried out pyrolysis of plastic LDPE types of plastic bags to obtain optimal degradation temperatures at 252.3 ºC with a yield percentage of 38%.

The three studies that have been conducted to convert plastic bag waste into liquid fuel, all refer to the pyrolysis process. Pyrolysis is a process of fractionating materials by temperature. When components are thermally unstable, and volatile matters in plastic waste will break and evaporate along with other components. The evaporating liquid product contains tar and hydrocarbons. The speed of the decomposition reaction in pyrolysis is based on the change in mass or mass fraction per unit time. The pyrolysis product will be greatly affected by temperature and time. Temperature gain in the pyrolysis reactor will be greatly influenced by the temperature setting in the reactor (Rodiansono *et al* 2007) [14].

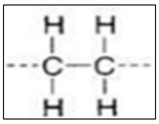
If studied further, various studies that have been carried out, have obtained a variety of good operating conditions with the achievement of a yield percentage that is already above 40%. However, from the various existing literature, the research is still on a laboratory scale, so further studies and tests are needed at a larger scale level in the form of a prototype.

Based on the description above, the scope of this study aims to develop a process for converting plastic bag waste into liquid fuel which is carried out on a prototype scale equipment unit using various operating conditions from research that has been done before.

2. Materials and Methods

2.1 Material

This research using waste plastic bag which is LDPE type as raw material. LDPE is a thermoplastic made from the monomer ethylene. LDPE is the first class of polyethylene, produced in 1933 by Imperial Chemical Industries (ICI) using a high-pressure process by free radical polymerization. Despite competition from more modern polymers, LDPE continues to be an important plastic class. In 2013, the worldwide LDPE market reached a volume of approximately US$ 33 billion. LDPE is a plastic that is easily shaped when heated, which is made from petroleum. its molecular formula is (-CH2-CH2-)n. LDPE has a density between 0.92-0.94 g/mL and has a melting point of 115 ºC. The molecular formula is (C2H4) n as shown in Fig 1.



Source: wikipedia, accessed 17 July 2018

Fig 1: LDPE Molecular Structure

Produced by free radical polymerization, LDPE has long and short chain branches of various forms of PE, resulting in a lower density. Branching keeps the molecular chains from packing tightly in their crystalline form, so LDPE has less tensile strength but greater ductility. That extraordinary "formatability" makes LDPE very useful.

LDPE is a resin that is hard, strong and does not react to other chemicals, is the highest quality plastic. Usually used for food containers, plastic packaging, soft bottles, plastic bags, etc. An example of LDPE plastic material can be seen in Fig 2.

Source: ljplastindo.com, accessed 17 July 2018

Fig 2: LDPE material

2.2 Methods

2.2.1 Simple Static Treatment and Analysis

In this study, the variables to be taken consist of fixed and Changed variables. This research was conducted with the data obtained from the measurement results arranged in tabular form to be used as material for study in determining the performance of the plastic bag waste conversion prototype unit in terms of calorific value, density and yield produced by the Pyrolysis Reactor unit. So that it can be used as fuel for combustion engines.

a. Fixed variables

- Mass of plastic bag: 10 kg
- Reactor volume: 0.371 m^3
- Coconut shell mass: 260 kg

b. Changed Variables

- Ratio Catalyst to Mass of plastic bag: 0.00, 0.04, 0.08, 0.12, 0,16
- Process Temperature Reactor: 130 ºC, 140 ºC, 150 ºC, 160 ºC, 170 ºC

c. Process of Converting Plastic Bag Waste (LDPE) into Liquid Fuel

Block diagrams for the Plastic Waste Bag Conversion using Pyrolysis process can be seen in Fig 3 below:

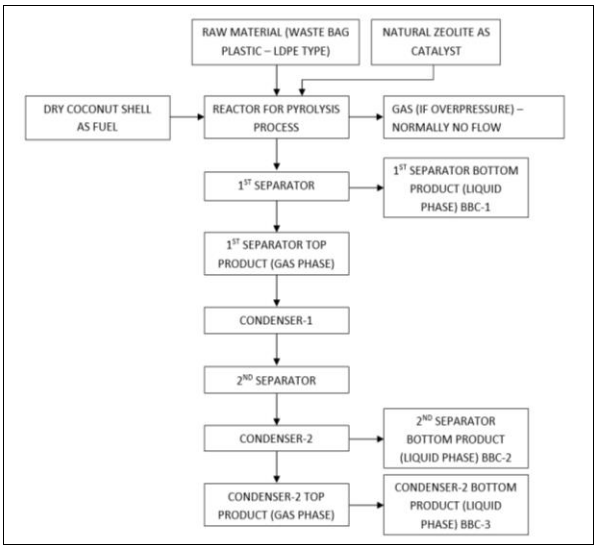


Fig 3: Block Diagram

Process of the Plastic Waste Bag Conversion Unit, it begins with cleaning plastic waste from dirt and then drying it in the sun. After drying, the plastic bag waste is rolled up by squeezing and folding it to reduce of its volume. The total mass of raw material for plastic bag waste feed to the reactor is 10 kg and then the pyrolysis process is carried out by heating the reactor in the furnace using coconut shell as fuel with variation of catalyst and temperature reactor as changed variables. Then the product in the form of gas from the top reactor is sent to 1st Separator (SP1) to be separated in the form of gas and liquid. In 1st Separator (SP1) the bottom product is obtained as 1st product (BBC-1) which is still mixed with wax and heavy fuel oil, the top gas phase product is fed to Condenser-1 (C1). Outlet of C1 will be two phase and send to 2nd Separator (SP2) to separate the liquid and vapor products again. 2nd Separator bottom product is obtained as 2nd Product (BBC-2), which is similar to gasoline. 2nd Separator overhead still in the form of gas will be fed to further condensation process in the Condenser-2. The bottom product of Condenser-2 is obtained in the form of a liquid which will be combined with BBC-2 and overhead Condenser-2 is in the form of a gas.

After data collection and calculation using below matrix, then the products tested for composition, characteristics, calorific value and yield. The characteristics, calorific value and composition of the resulting fuel oil will be compared with refined gasoline products from Indonesia Oil and Gas Company (Pertamina).

No. of Expriment	Ratio Zeolit	Mass Zeolit (Kg)	T Reactor (°C)	Mass (Kg)			Product 1 (BBC-1)			Product 2 (BBC-2)			Total Product (BBC-1 + BBC-2)
				Residue	Coconut Shell	Wax	Vol (L)	Mass (Kg)	GHV (Cal/Mol)	Vol (L)	Mass (Kg)	GHV (Cal/Mol)	
1													
2													
3			130										
4													
5													
6													
7													
8			140										
9													
10													
11													
12													
13			150										
14													
15													
16													
17													
18			160										
19													
20													
21													
22													
23			170										
24													
25													

Fig 4: Research Matrix

3. Results and Discussion

3.1 Effect of Zeolite Ratio and Temperature Reactor for Yield Percent and Product Calorific Value

The ratio of the amount of Zeolite to LDPE feedstock and the temperature of the degradation process will affect the yield of liquid fuel BBC-1 and BBC-2 produced. Fig 5 shows the effect of the ratio of the amount of Zeolite to raw material (LDPE) and process temperature to the yield of liquid fuel produced. The effect of the ratio of Zeolite to raw material (LDPE) and process temperature to the calorific value of the resulting liquid fuel (GHV) can be seen on Fig 6.

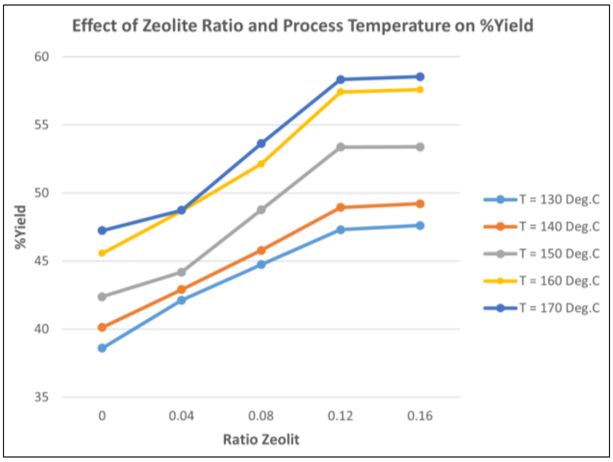


Fig 5: Effect of Zeolite Ratio and Process Temperature on %Yield

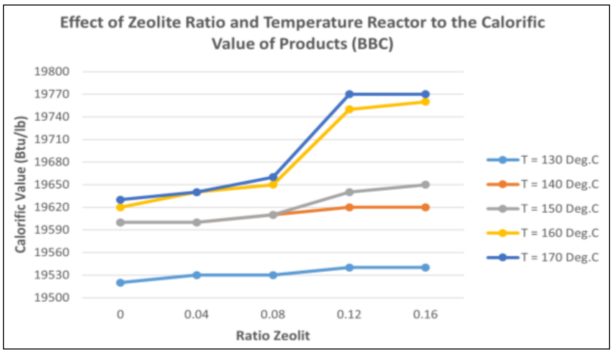


Fig 6: Effect of Zeolite Ratio and Temperature Reactor on Calorific Value of Products (BBC)

From Fig 5 the effect of the ratio zeolite to the percent yield in this study gave a significant condition to the zeolite ratio from the ratio range of 0.04 to 0.12. However, the yield value tends to be constant at the zeolite ratio at 0.16. This gives a clear indication that the optimal zeolite ratio is at a ratio of 0.12 (in experiment number 19). Whereas without zeolite, the yield value tends to be smaller compared to the use of zeolite as catalyst. The pattern or trend in the zeolite ratio occurs in the same way for each change in reactor temperature starting from 130 ºC to 170 ºC. The yield value at the optimal zeolite ratio conditions reaches 57.43% or in other words, 10 kg of LDPE plastic waste raw material is degraded as much as 5.743 kg into liquid fuel (BBC-1 = 1.2843 kg and BBC-2 = 4.4584 kg). If related to the total volume of liquid fuel produced, it gives a volume value of liquid fuel of 7.7612 liters from the conversion of 10 kg of raw material for LDPE plastic waste.

The increase in percent yield with the increasing amount of zeolite use is caused by a decrease in the value of the activation energy from the degradation reaction process of the chemical components in the LDPE plastic waste raw material into liquid fuel. The addition of the amount of zeolite will affect the percent yield of liquid fuel which is produced as a result of a number of residues or heavy fractions in the Bottom Reactor experiencing a process of breaking the chain into shorter chains to become liquid fuel products and can evaporate to the Top Reactor. This can be seen from the research data with the addition of zeolite, the amount of residue decreases drastically. The residue without catalyst reached 5.5892 kg and the use of catalyst decreased to 3.0220 kg or reduced the amount of residue to 46.018%.

The use of a catalyst also affects the calorific value of the resulting liquid fuel products as shown in Fig 6. The improvement in the calorific value is more due to the improvement of the hydrocarbon chain. The hydrocarbon chains become shorter as a result of the zeolite work by contributing to reducing the value of the activation energy in the reaction process that takes place in the pyrolysis reactor.

The effect of temperature has a very good impact on the process as a result of increasing the distance between the molecules of the chemical elements contained in the raw material for LDPE plastic waste, making it easier to break long chains into short chains. On the other hand, the effect of temperature gives an increase in the kinetic energy between molecules so that when the collisions between molecules will provide momentum for the molecules to interact with the zeolite catalyst so that chain breaking occurs more easily to form liquid fuel. From the arhenius equation Temperature gives effect to the increase in the value of the reaction rate constant. Temperature can be said to be a heat aid in the pyrolysis process or thermal cracking process in the form of breaking long hydrocarbon chains into smaller chain hydrocarbons. In the LDPE pyrolysis reaction at the beginningp of the process there will be a breaking of the C-C and C-O bonds in the hydrocarbon chain because these two bonds have a very low bond energy of only around 350 kJ/mol compared to other atomic bonds such as C=C which reaches 837 kJ/mol. So that the breakdown of polyethylene into shorter chains will occur at temperatures that are not too high. The pyrolysis process of plastic bags (Low Density Polyethylene) (LDPE) in this study took place as optimal at 160 °C. Fig 7 below is overall data collected from the expriment.

No. of Expriment	Ratio Zeolit	Mass Zeolit (Kg)	T Reactor (°C)	P Reactor (Bar)	Mass (Kg)			Product 1 (BBC-1)				Product 2 (BBC-2)				Total Product (BBC-1 + BBC-2)	% Rendeman	Volume Product BBC-1 + BBC+2
					Residue	Coconut Shell	Wax	Vol (L)	Mass (Kg)	Density (gr/mL)	GHV (Cal/Mol)	Vol (L)	Mass (Kg)	Density (gr/mL)	GHV (Cal/Mol)			
1	0	0		1	5.5982	260	0.5083	0.88352	0.856	0.9693	18400	3.661	3.0051	0.8208	19520	3.8615	38.615	4.54472
2	0.04	0.4		1	5.1044	260	0.5378	0.95489	0.925	0.9688	18420	4.007	3.2876	0.8204	19530	4.2127	42.127	4.96219
3	0.08	0.8	T = 130 Deg.C	1	4.8304	260	0.5788	1.02399	0.99	0.9672	18470	4.25	3.4841	0.8198	19530	4.4745	44.745	5.27389
4	0.12	1.2		1	4.579	260	0.5808	1.10203	1.064	0.9654	18500	4.475	3.6673	0.8195	19540	4.7312	47.312	5.57703
5	0.16	1.6		1	4.5498	260	0.5851	1.10951	1.071	0.9652	18510	4.504	3.6903	0.8194	19540	4.7612	47.612	5.61321
6	0	0		1.2	5.3902	260	0.5104	0.99907	0.968	0.9687	18450	3.717	3.045	0.8192	19600	4.0128	40.128	4.716111935
7	0.04	0.4		1.2	5.008	260	0.5564	1.0314	0.998	0.968	18450	4.021	3.293	0.819	19600	4.2914	42.914	5.052161979
8	0.08	0.8	T = 140 Deg.C	1.2	4.6877	260	0.5998	1.0818	1.046	0.967	18450	4.315	3.5331	0.8188	19610	4.5792	45.792	5.396772511
9	0.12	1.2		1.2	4.3483	260	0.6281	1.23067	1.188	0.965	18500	4.529	3.7067	0.8185	19620	4.8943	48.943	5.759323545
10	0.16	1.6		1.2	4.2785	260	0.6577	1.2357	1.192	0.9648	18500	4.556	3.7298	0.8186	19620	4.922	49.22	5.792012178
11	0	0		1.4	5.2248	260	0.5372	1.03456	1	0.9664	18450	3.954	3.2382	0.819	19600	4.238	42.38	4.988407412
12	0.04	0.4		1.4	4.993	260	0.5883	1.06732	1.031	0.9655	18500	4.137	3.3882	0.819	19600	4.4187	44.187	5.204318968
13	0.08	0.8	T = 150 Deg.C	1.4	4.4959	260	0.6282	1.17036	1.129	0.965	18500	4.576	3.7465	0.8188	19610	4.8759	48.759	5.745961131
14	0.12	1.2		1.4	4.0007	260	0.6622	1.2849	1.24	0.9649	18500	5.009	4.0973	0.818	19640	5.3371	53.371	6.293824195
15	0.16	1.6		1.4	3.9681	260	0.6928	1.28552	1.24	0.9649	18500	5.011	4.0987	0.8179	19650	5.3391	53.391	6.296770135
16	0	0		1.6	4.7089	260	0.5556	1.42926	1.119	0.9654	18500	4.649	3.4368	0.8185	19620	4.5562	45.562	6.078615205
17	0.04	0.4		1.6	4.4192	260	0.5937	1.49183	1.168	0.9652	18500	5.003	3.6985	0.818	19640	4.8669	48.669	6.495210431
18	0.08	0.8	T = 160 Deg.C	1.6	4.0332	260	0.6455	1.55516	1.218	0.965	18500	5.404	3.9948	0.8179	19650	5.2128	52.128	6.959379104
19	0.12	1.2		1.6	3.3848	260	0.6794	1.63981	1.284	0.9647	18550	6.031	4.4584	0.8172	19750	5.7427	57.427	7.671196313
20	0.16	1.6		1.6	3.4038	260	0.7309	1.64607	1.289	0.9646	18550	6.046	4.4694	0.8171	19760	5.7586	57.586	7.692333649
21	0	0		1.7	4.5573	260	0.5738	1.44218	1.13	0.9653	18500	4.86	3.5943	0.8184	19630	4.7241	47.241	6.302621381
22	0.04	0.4		1.7	4.4673	260	0.5803	1.49209	1.169	0.965	18500	5.009	3.7043	0.818	19640	4.8732	48.732	6.501281182
23	0.08	0.8	T = 170 Deg.C	1.7	3.8576	260	0.6743	1.60799	1.26	0.965	18500	5.549	4.1032	0.8178	19660	5.3629	53.629	7.156604738
24	0.12	1.2		1.7	3.3089	260	0.7443	1.64641	1.29	0.9645	18550	6.145	4.5439	0.817	19770	5.8337	58.337	7.790970204
25	0.16	1.6		1.7	3.022	260	0.7589	1.64692	1.29	0.9644	18550	6.171	4.5637	0.817	19770	5.8539	58.539	7.818255647

Fig 7: Effect of Zeolite Ratio and Temperature Reactor on Calorific Value of Products (BBC)

4. Conclusions and Suggestions

4.1 Conclusion

1. The Pyrolysis Reactor Prototype Unit can be used to convert plastic bag waste (Low Density Polyethylene) (LDPE) into liquid fuel with a percent yield of 57.43% or 57.74 kg or a volume of 7.67 liters of the total mass of bag waste raw material 10 kg of plastic.
2. The ratio of Zeolite to LDPE plastic waste raw material is optimal at 0.12 with a pyrolysis process temperature in the reactor of 160 °C.
   Under optimal conditions, the calorific value of liquid fuel produced for BBC-1 is 18550 Btu/lb equivalent to diesel oil. And BBC-2 is a premium equivalent with a heating value of 19750 Btu/lb.

4.2 Suggestion

It is necessary to conduct research on the possibility of breaking down the resulting residual heavy hydrocarbons which amount to 33.85% in order to increase the yield of the resulting liquid fuel.

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