Investigating the effect of pyro-catalytic temperature on gasoline production from low density polyethylene (LDPE) plastics waste

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Abstract. This research aims to produce hydrocarbon fuels from LDPE plastic waste by the pyro-catalytic method. Pyro-catalytic of LDPE samples was carried out at 400, 425, 450, 475, 500, 525 and 550 °C, heating flow rate of 15 °C/min with zeolite catalyst. Fuel-oil quality analysis includes: density, viscosity, heat value, and hydrocarbon compounds using Gas Chromatography-Mass Spectrometry (GC-MS). The pyro-catalytic process at temperatures of 400, 425 and 450 ºC did not produce fuel oil, while at 550 ºC it produced wax, so that the analysis of the quality of fuel-oil was not carried out. At pyrolysis temperatures of 475, 500 and 525 °C, respectively: bio-oil yield 8.4; 25.6; 46.5%; viscosity 0.81; 0.75; 0.84cP, density 0.750; 0.764; 0.756 g/mL and the caloric value of 11,403; 11,421; 11,338 kcal/kg. GC-MS spectrogram analysis of fuel-oil obtained the highest gasoline (C₅-C₁₂) content at pyrolysis temperature of 500 ºC which was 81.31%. Based on the physical characteristics and percentage of the gasoline compounds content, this study recommends the production of fuel-oil from pyrolysis LDPE with zeolite catalytic done at a temperature of 500 ºC because it is in accordance with fuel characteristics standards according to the American Standard and Testing Materials (ASTM).

1. Introduction

Non-renewable energy reserves such as petroleum, natural gas and coal are increasingly limited in availability. Indonesia is one of the world’s oil-producing countries [1]. Indonesia has oil reserves of 3.6 billion barrels which are expected to run out in the next 13 years [2]. Therefore in the world today there is a need for efforts to find new sources of energy other than petroleum as a substitute for fossil fuels.

One of the efforts made to create alternative energy is to convert Low Density Polyethylene (LDPE) plastic waste into fuel oil as a substitute for fossil fuels. LDPE plastics are composed of ethylene polymers whose main constituent elements are hydrogen, carbon and some additional oxygen, nitrogen, chlorine, fluorine and sulfur atoms [3]. Plastic is derived from petroleum derivatives so that in the decomposition process it can be returned to hydrocarbons as a basis for energy fuels [4]. The process of recycling plastic waste aims to convert long-chain hydrocarbon polymers into shorter chains as raw materials for the chemical industry or fuel production [5].
The increasing amount of plastic waste causes the need for management of such plastic waste. The behavior of burning plastic waste in the environment will produce hydrogen sulfide gas (H$_2$S) which is toxic to the environment. Plastic waste containing chloride compounds (Cl) can produce dioxins (cancer-causing) if burned at low temperatures [6,7].

One of the methods that can convert LDPE plastic waste into fuel-oil is pyrolysis [8]. Pyrolysis is the thermochemical decomposition process of organic and synthetic materials through the process of heating without or little oxygen, where the raw material will undergo chemical structure breakdown into a gas phase [9]. The main product produced by pyrolysis consists of liquid, gas and solid residue fractions [8,10-12].

The nature of LDPE plastics has low thermal conductivity and high viscosity, so pyrolysis must be carried out at high temperatures 500-800 ºC [9,13]. Therefore, the pyrolysis method combined with a catalyst (pyro-catalytic) is expected to be able to overcome this problem. The use of catalysts can reduce the reaction temperature, increase the decomposition reaction and the quality of the product of pyrolysis products that produce a liquid product with a lower amount of carbon [14].

The use of LDPE plastic waste as raw material for alternative fuel production using the pyro-catalytic method is expected to be a renewable fuel equivalent to conventional fuels. The use of zeolite catalyst in the pyrolysis process of polyethylene and polystyrene (PS) has been carried out by several researchers [6,15,16]. In the process of pyrolysis, the molecular chain cutting reaction occurs randomly so as to produce molecular fractions with different molecular weights (wide distribution of molecular variations). This has the consequence of low target molecular yield so it is recommended to involve a catalyst in the pyrolysis process [17].

In the research, the conversion of LDPE waste into fuel-oil uses the pyro-catalytic method with the zeolite catalyst, where the pyro-catalytic process is carried out at a temperature of 400-550ºC. Zeolite has been widely used as a catalyst in the pyro-catalytic process of plastic waste because it is acidic, deoxygenated, and has micro-pores so that it can absorb water and oxygen content in the pyrolysis process [14,18]. The fuel-oil quality test was carried out with several analyzes namely, density, viscosity, heating value and hydrocarbon content with GC-MS instruments. Several researchers have conducted analyzes of hydrocarbon content in fuel-oil using GC-MS instruments [19,20]. This research will study the effect of temperature on the production and quality of gasoline produced from the LDPE plastic waste pyro-catalytic process.

### 2. Materials and Methods

#### 2.1. Materials and Instrumentations

The material used in this study is a sample of LDPE plastic waste that has been collected from various waste disposal places from the Kendari region, Southeast Sulawesi. The tools used in this study are a series of pyrolysis devices, Gas Chromatography-Mass Spectrometry (GC-MS), filter paper (whatman), pycnometer, viscometer and measure glass.

#### 2.2. Sample preparation

Sample preparation includes washing the LDPE plastic waste to remove the remaining dirt, then the moisture content is removed by drying in the sun then chopped with a size of 3-5 cm.

#### 2.3. LDPE pyro-catalytic at temperatures 400, 425, 450, 475, 500, 525 and 550 ºC

The process of converting LDPE plastic waste into fuel-oil is carried out at temperatures of 400, 425, 450, 475, 500, 525 and 550 ºC using a zeolite catalyst at a ratio of 2.5%. The pyrolysis reactor is designed to have a 500 gram capacity, with a cylindrical reactor made of stainless steel and using electricity as a source of heat energy with 1000 watts of power. Then the LDPE plastic waste is put into the reactor and heated for each temperature variation. The next step is to calculate the rate of burning of the plastic until it is converted into fuel-oil. The pyrolysis steam is channeled into a condenser circuit
that is drained by cooling water, then the liquid from the condensation results is collected in a reservoir.

2.4. The density analysis of fuel-oil
The fuel oil density was analyzed using a 25 ml picometer, then calculated by equation (1) and measurements are in accordance with ISO 4787 standards [21,22]. To minimize measurement errors, all measurements are carried out three times for each sample and the results are averaged:

\[ \rho = \frac{m}{\nu_p} (\text{g/mL}) \]  

(1)

m: mass (pycnometer + sample) - mass pycnometer is empty.

\[ \nu_p: \text{Pycnometer volume (25 mL).} \]

2.5. The viscosity analysis of fuel-oil
The fuel-oil viscosity is determined by comparing the viscosity of the fuel-oil and the viscosity of the comparative liquid in the form of water using an ostwald viscometer which involves each of the fuel-oil density and water density variables, then calculated by equation 2 [21].

\[ \frac{\eta_1}{\eta_2} = \frac{\rho_1 t_1}{\rho_2 t_2} \]  

(2)

\[ \eta_1: \text{Viscosity of Plastic Fuel (cP)} \]

\[ \eta_2: \text{Water viscosity (cP)} \]

\[ \rho_1: \text{Density of plastic fuel (g/mL)} \]

\[ \rho_2: \text{Water density (g/mL)} \]

\[ t_1: \text{Flow time of plastic fuel (s)} \]

\[ t_2: \text{Flow time of water (s)} \]

2.6. The analysis calorific value of fuel-oil
The calorific value is analyzed by involving the variable density, specific gravity and API (American Petroleum Institute) gravity. Specific gravity (SG) is the ratio between the density of fuel-oil with the density of water, while the value of API gravity is calculated using equation 3 and 4 [21,23].

\[ ^oAPI = 141,5 + SG - 131,5 \]  

(3)

\[ CV = \frac{2,2046226}{3,9673727} \times (18,650 + 40 \times \left( ^oAPI - 10 \right)) \text{kcal/kg} \]  

(4)

2.7. Gas chromatography-mass spectrometry (GC-MS) analysis
Identification of fuel-oil hydrocarbon chemical compounds was carried out by GC-MS instruments using DB-624 agilent capillary column, the injector temperature was 250 °C, and the carrier gas separation ratio was 69.4 the carrier gas was helium in a controlled flow of 3 mL/min. Initially, the oven temperature is held at 60 °C for 42 minutes, then the temperature rises from 60 to 200 °C at 5 °C/minute and held for 5 minutes [24,25].

3. Results and Discussion

3.1 The Effects pyro-catalytic temperature on fuel-oil yield and decomposition time of LDPE plastic waste pyro-catalytic
Pyro-catalytic is one method that can be used to convert plastic waste into fuel oil with addition of catalyst in the form of zeolite. Research by adding catalysts has been carried out by several researchers such as Beltrame and Carniti [13] comparing various types of catalysts used in polyethylene pyrolysis.
They found that zeolite catalysts were the most effective catalysts, then Ishihara et al [26] reported that polyethylene pyrolysis using silica-alumina catalysts could shorten polymer chains and increase chain branches, and Manos et al [27] discovered the process of pyrolysis with zeolite catalysts producing a product liquid in the form of hydrocarbons with range C₃-C₁₅.

Zeolite in pyrolysis process will provide a fast and stable temperature propagation on the biomass due to the presence of alumina in zeolite, so that with this process the temperature will break the chemical structure of biomass. The process of hydrocarbon breakdown occurs because the role of pore structure in zeolites. Because these pores have a role to sort specifically in absorbing certain molecules and rejecting other molecules [28]. Zeolite catalysts cannot reduce amount of oxygenated compounds in fuel-oil and also increase amount of hydrocarbons (aliphatic and aromatic), therefore, increasing carbon content in fuel-oil and making it more efficient to be used as transportation fuel [29, 30].

In this research the sample used was LDPE plastic waste, LDPE plastic is composed of ethylene monomers with a molecular structure of C₂H₄ and has a low density of 0.910-0.940 gr/ml. LDPE waste of 400 grams was pyrolysed at temperatures of 400, 425, 450, 475, 500, 525 and 550ºC with a ratio of zeolites and samples was 2.5%. At that temperature plastic will melt and then turn into gas. During pyrolysis process, long chain of hydrocarbons will be cut into shorter chains. Furthermore, cooling is carried out on the gas so that it will condensate and form a liquid. This liquid will later be accommodated in an erlemenyer glass and can be used as fuel as a substitute for fossil fuels (Figure 1).

The increase in pyro-catalytic temperature affects the yield of pyro-catalytic produced, at temperatures of 475, 500 and 525 ºC, respectively8.4;25.6 and 46.5%. This is because LDPE plastic has a high boiling point, so that the higher the combustion temperature produces more plastic fuel volume. Also at high temperature the carbon chain will be more easily swelled than at low temperatures [24], according to Aprian Ramadhan [31] with increasing heating temperatures the substances contained in plastics will decompose completely. This is consistent with Arhenius's theory that temperature greatly influences the product produced, where the higher the temperature the greater the energy produced as a result the pyrolysis rate increases and the yield increases [24,32]. While at other temperatures namely 400, 425 and 450 no fuel-oil yield is produced, this is because at that temperature the plastic has not been decomposed into fuel-oil. In addition, at that temperature the thermal energy required cannot break the carbon chain and change plastic from solid phase to steam, while at 550 ºC rate of hydrocarbon decomposition is faster than condensation capability so that

![Figure 1](image-url)
decomposition product is colloidal. This is consistent with what is explained by Wanchai and Chaisuwan [33] that higher temperature, more oil conversion results are produced. But at even higher temperatures the gas yield will be more than liquid yield, and resulting product has like a wax form.

3.2 Effect of pyro-catalytic temperature on the density and viscosity of fuel-oil product

Density and viscosity is one of important parameters in determining quality of fuel oil. Density is an indicator of the amount of impurities produced by the reaction. Density is one of important parameters in determining fuel quality [22]. The relative density of oil is ratio of oil density and water density at a certain temperature, for petroleum the calibration temperature used is 15 °C [23]. Results of the analysis of fuel-oil density of pyro-catalytic low-density polyethylene (LDPE) plastic waste are shown in Figure 2.

Based on the Figure 2, pyrolysis density obtained at 475°C, 500°C and 525°C were 0.7479 gr/ml, respectively. 0.7446 gr/ml and 0.7505 gr/mL. The value of density or density in the range of 0.7446-0.7505 gr/mL is the range of density values close to the value of conventional gasoline density that is 0.7100 - 0.7700 gr/mL [32, 34]. This is not much different from what was done by Taufan Landi [35] by converting same material, LDPE, the density produced from three variations of temperature 350 °C, 500 °C, and 700 °C which is 0.7291 gr/mL, 0.7563 gr/mL, and 0.7336 gr/mL. Density values obtained are close to the range of gasoline density values. Inequality of conventional gasoline density with LDPE plastic fuel is due to the fact that LDPE plastic fuel is not 100% pure and still contains another fraction namely kerosene fraction and other compounds.

Density is an indicator of the amount of impurities produced by reaction [32]. The more percentage of heavy substances or impurities contained in the fuel, the higher specific gravity of the fuel. Because these heavy substances are difficult to evaporate and tend to become smoke or soot that does not burn completely. These heavy substances are also the main elements of carbon residues that can pollute the engine and this affects combustion and exhaust emissions [36,37]. Based on research that has been done, the best results from LDPE plastic waste pyrolysis are at a temperature of 500 °C with the lowest density value compared to temperature of 475 °C and 525 °C which is 0.7446 gr/mL. The density value is fulfilled standard range density of premium fuels [32].

Viscosity is one of important parameters in determining quality of fuel oil. Viscosity affects the atomization of fuel when it is injected into combustion chamber. Fuel injection can affect the stage of mixing fuel with air and affect occurrence of complete combustion [38]. If oil is too thick, it will be
difficult for the pumping process, difficult to turn on the burner, and difficult to flow. Poor atomization will result in formation of carbon deposits on engine walls. The lower viscosity, the better fuel is [39,40]. Analysis of viscosity plastic fuel from pyrolysis of Low Density Polyethylene waste plastic is shown in Figure 3. Viscosity value of LDPE plastic waste pyrolysis results at 475 °C, 500 °C and 525 °C were 0.81 cP, 0.75 cP and 0.84 cP. The value of viscosity produced in this study approached the value of viscosity of gasoline type of fuel that is equal to 0.6520 cP.

3.3 Effect pyro-catalytic temperature on the calorific value of fuel oil product

The calorific value is maximum amount of heat energy released by a fuel through the combustion reaction of mass or volume of fuel. Determination of calorific value can be done by measuring using a calorimeter bomb instrument [41] and a calculation method [42]. In this study, determination of calorific value is done by means of calculations involving variable density, API Gravity and Specific Gravity. The equation used in calculating calorific value can be seen in Eq. 4. The calorific value of fuel oil generally ranges from 10,160 kcal/kg-11,000 kcal/kg [43]. The results of analysis of calorific value of plastic fuel from pyrolysis of low density polyethylene plastic waste are shown in Figure 4.

![Figure 4. Effect of pyro-catalytic temperature on the fuel-oil calorific value](image)

Figure 4. shows the results of analysis of heating value of LDPE plastic waste fuel, where the calorific value produced at 475 °C, 500 °C and 525 °C respectively 11,113.433 kcal/kg, 11,421.474 kcal / kg, and 11,388.601 kcal/kg. Calorific value obtained from LDPE plastic waste pyrolysis results is not much different and is in accordance with quality standard of the calorific value of fuel oil which is generally marketed domestically, between 18,300-19,800 BTU/lb or 10,160-11,000 kcal/kg. The calorific value obtained in the study is close to the calorific value of premium/gasoline which is 47,080 MJ/kg or equivalent to 11,244,862 kcal/kg.

3.4 The GC-MS analysis of fuel-oil

The hydrocarbon content of LDPE pyro-catalytic plastic waste was analyzed using a gas chromatography-mass spectrometry (GC-MS) instrument. Characterization technique using gas chromatography aims as a separator of components in a compound then proceed with mass spectrometry to detect each molecule of components that have been separated [44]. Pyro-catalytic plastics generally produce mixed liquids with complex compounds, gasoline range hydrocarbons (C4-C12), diesel range hydrocarbons (C11-C18) and including wax (C10 +) [30].
Based on the results of analysis that has been carried out produced several hydrocarbon fraction compositions, namely gasoline fraction (C₅-C₁₂) and the long chain hydrocarbon fraction (C₁₃-C₆₀). The percentages of gasoline fractions at temperatures of 475 °C, 500 °C and 525 °C respectively were 64.52, 81.31 and 60.59% while the rest are kerosine, diesel and long chain hydrocarbon fractions (≥C₅₀). As a result, pyro-catalytic fuel oil needs to be processed further (such as distillation) before it can be used as a substitute for conventional fuels (Figure 5).

Figure5. The spectro gram of LDPE fuel-oil at pyro-catalytic temperature (a) 475 °C (b) 500 °C (c) 525 °C

The highest percentage of gasoline is obtained at a temperature of 500, namely 81.31%. The main chemical compounds were identified to be present of the bio-oil at pyro-catalytic temperature 500°C as shown in Table 1 and Figure5(b).

Table 1. The analysis GC-MS of LDPE fuel-oil at pyro-catalytic temperature 500 °C

| RT   | RSI  | Compound Name                  | CF    | MW  | Area (%) |
|------|------|--------------------------------|-------|-----|----------|
| 0.58 | 842  | Pentane                        | C₅H₁₂ | 72  | 0.85     |
| 0.67 | 793  | 2,3-dimethyl-butane            | C₆H₁₄ | 86  | 0.31     |
| 0.71 | 915  | 1-ethyl-2-methyl-cis-cyclopropane | C₆H₁₂ | 84  | 3.36     |
| 0.91 | 879  | 1-methyl-cyclopentane          | C₆H₁₀ | 82  | 1.03     |
| 0.96 | 943  | Benzena                        | C₆H₆  | 78  | 0.76     |
| RT  | RSI | Compound Name                          | CF   | MW  | Area (%) |
|-----|-----|----------------------------------------|------|-----|----------|
| 1.09| 932 | 1,2-dimethyl- trans-cyclopentane       | C₆H₁₄| 98  | 2.56     |
| 1.14| 9.17| Heptana                                 | C₇H₁₆| 100 | 3.32     |
| 1.27| 952 | Cycloheptane                            | C₇H₁₄| 98  | 1.05     |
| 1.5 | 939 | 2-methyl-3-hexyne                       | C₈H₁₂| 96  | 1.11     |
| 1.59| 912 | 4-methyl-heptane                        | C₈H₁₈| 114 | 4.17     |
| 1.79| 904 | 1-Octene                                | C₈H₁₆| 112 | 3.7      |
| 1.87| 885 | Octene                                  | C₈H₁₈| 114 | 4.09     |
| 1.93| 813 | 4-chloro-octene                         | C₈H₁₇Cl| 148 | 1.3      |
| 2.05| 9.16| 5-methyl-3-heptyne                      | C₉H₁₄| 110 | 1.15     |
| 2.13| 819 | 1,2,3-trimethyl-cyclohexene             | C₉H₁₈| 126 | 2.09     |
| 2.27| 870 | 2,4-dimethyl-1-heptene                  | C₉H₁₈| 126 | 7.51     |
| 2.36| 903 | 1,3,5-trimethyl-cyclohexene             | C₉H₁₈| 126 | 2.3      |
| 2.49| 865 | 2,4-octadiyne                           | C₈H₁₀| 106 | 2.97     |
| 2.7 | 9.18| 3-nonene                                | C₉H₁₈| 126 | 4.28     |
| 2.79| 902 | Nonene                                  | C₉H₂₀| 128 | 3.4      |
| 3.01| 816 | propyl-cyclohexene                      | C₉H₁₈| 126 | 0.7      |
| 3.3 | 883 | 1-(1-propynyl)-cyclohexene              | C₉H₁₂| 120 | 0.86     |
| 3.36| 856 | 2,4-dimethyl-2,3-heptadien-5-yne        | C₉H₁₂| 120 | 0.54     |
| 3.61| 916 | (Z)-2-decene,                           | C₁₀H₂₀| 140 | 4.17     |
| 3.69| 929 | Decane                                  | C₁₀H₂₂| 142 | 2.74     |
| 3.78| 862 | 4-methyl-decane,                        | C₁₀H₂₄| 156 | 1.6      |
| 4.16| 842 | 3,8,11-trioxatetrayclo                 | C₁₁H₁₅O₃| 154 | 1.05     |
| 4.37| 846 | 2,3,7-trimethyl-4-octene                | C₁₁H₂₂| 154 | 2.65     |
| 4.45| 893 | 5-undecene                              | C₁₁H₂₂| 154 | 2.82     |
| 4.53| 893 | Undecene                                | C₁₁H₂₂| 156 | 2.94     |
| 4.93| 793 | 1-methyl-4-(2-propenyl)-benzena         | C₁₀H₁₂| 132 | 0.87     |
| 5.23| 921 | 1-methyl-2-(4-methylpentyl)-cyclopentane| C₁₂H₂₄| 168 | 2.05     |
| 5.31| 920 | Dodecane                                | C₁₂H₂₆| 170 | 2.36     |
| 5.98| 927 | Cyclotridecane                          | C₁₂H₂₆| 182 | 1.4      |
| 6.05| 889 | 3,3-dimethyl-heptane                    | C₁₀H₂₀| 128 | 1.52     |
| 6.1 | 845 | 2,3,7-trimethyl-4-octene                | C₁₁H₂₂| 154 | 1.39     |
| 6.15| 849 | 2,3,7-trimethyl-4-octene                | C₁₁H₂₂| 154 | 0.71     |
| 6.21| 838 | 2,3,7-trimethyl-4-octene                | C₁₁H₂₂| 154 | 1.03     |
| 6.68| 902 | Cetene                                  | C₁₀H₃₂| 224 | 1.01     |
| 6.74| 900 | Tetradecane                             | C₁₁H₃₀| 198 | 1.11     |
| 7.34| 920 | 8-heptadecene                           | C₁₃H₃₄| 238 | 0.56     |
| 7.4 | 893 | Pentadecene                             | C₁₃H₃₂| 212 | 0.76     |
| 8.05| 863 | 10-methylnonadecane                     | C₁₂H₂₄| 282 | 1.02     |

*RT: Retention Time, RSI: Relative Similarity Index, CF: Compound Formula, MW: Molecular Weight
The results of composition analysis hydrocarbon fraction resulting from pyro-catalytic of LDPE plastic waste are shown in Figure 6.

![Figure 6](image-url)  
**Figure 6.** Analysis composition of hydrocarbon fraction resulting from LDPE pyro-catalytic method

### 4. Conclusion

Based on the results of analysis that has been done, the optimum pyrolysis temperature to produce LDPE plastic waste fuel oil which is close to conventional gasoline is obtained at a pyrolysis temperature of 500 °C. This is indicated by low density and viscosity values of 0.7446 gr/ml and 0.81cP and with the highest calorific value of 11,421,474 kcal/kg, so it can be concluded that the results obtained from LDPE plastic waste pyrolysis, resembles gasoline type fuels (premium) based on color parameters, GC-MS results, density, viscosity, API gravity, Specific gravity and calorific value. Thus LDPE plastic waste can be produced into gasoline type of fuel as an alternative fuel source as well as to overcome pollution caused by plastic waste.

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