The effect of plastic type on the yield and quality of lignite and plastic waste pyrolysis products

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Abstract. Lignite is a low rank coal which has great potential in South Kalimantan. However, it has not been used optimally due to its low quality. One of the ways to improve it is pyrolysis. Pyrolysis is the thermal decomposition of organic material in the absence of oxygen which will produce three products (char, tar and gas). Pyrolysis can make lignite into liquid fuel (pyrolysis liquid), but it still requires improvement due to the need for pure hydrogen donors. Plastic waste has a higher hydrogen/carbon ratio than coal. This material can be used as an additive in the pyrolysis process because it is rich in hydrogen. The samples of plastic waste used were polyethylene (PE), polypropylene (PP), and polystyrene (PS). Samples of lignite and plastic (plastic composition was 25 wt%) were used for every experiment, and pyrolysis was carried out with a holding time of 60 minutes at 500°C. The pyrolysis liquid obtained is then analyzed for its yields and properties (density, kinematic viscosity, heating value). The most feasible result for fuel alternative was obtained with the addition of PE.

1. Introduction

Indonesia has abundant potency of coal. The data from ESDM in 2018 [1] mentioned that the coal deposit in Indonesia is approximately 26.2 billion tons. The deposit in Kalimantan island leads as the highest with the amount 14.9 billion tons, followed by Sumatera and Sulawesi with the total amount of 11.2 and 0.12 billion tons, respectively. However, coal from Kalimantan island including to lignite and sub-bituminous coal [2] which is considered as a low grade (low rank) coal due to its low energy content and high moisture [3].

Some previous research have been conducted to optimize the utilization of lignite as an efficient energy source namely gasification, hydothermal liquefaction [4] and pyrolysis [5]. Pyrolysis is an alternative method for clean and efficient utilization of lignite. This technology breaks the sample (such as lignite) structure into smaller species [6] similar to the technique for treating petroleum products and its derivatives [7]. In pyrolysis, biomass is decomposed into liquid, solid (char) and gas in the absence of oxygen. However, the oxygen content of lignite is considerably high, so it is become a drawback in the pyrolysis process, since oxygen contributes in lowering calorific value of its product [8]. The addition of hydrogen can improve the quality of pyrolysis product. However, pure gas hydrogen is still expensive.
Plastic waste is still a big issue for the environment. Not only it is unable to be decomposed by nature, but it causes air pollution if burned [9]. Three main types most used of plastics waste are made of polyethylene, polypropylene and polystyrene [10]. Baofeng et al. [4] found that the composition of carbon and hydrogen in plastics is very high, even higher than that of in coal. They also observed that there is an interaction between plastics and lignite during the pyrolysis process. It is proposed that plastic waste will take part as the hydrogen “donor” during the lignite pyrolysis process. Zhou et al [11] observed that pyrolysis of coal with polyethylene (PE) and polypropylene (PP) addition at various conditions result in the improvement of the liquid yields and the coal conversion. Moreover, Ozsina and Putun [12] discovered the pyrolysis of walnut shell and peach stones with polystyrene decreased activation energy value. The research about pyrolysis lignite mixed with plastics is interesting to be conducted, in order to increase the quality of its pyrolysis product and at the same time also contribute to solve plastic waste problem issue. In this work, pyrolysis lignite (low rank coal) with three common types of plastics, those are polyethylene, polypropylene and polystyrene, is conducted. The effect of plastic type on pyrolysis product yields and also its quality are investigated. Furthermore, this study could contribute to develop lowrank coal as an efficient energy source.

2. Materials and Method
Lignite was obtained from Satui Region at Tanah Bumbu district, South Kalimantan. It was ground and sieved to the size of 0.5-1 mm to homogenize the sample. The lignite powder was dried in an oven. Whereas polyethylene (PE), polypropylene (PP) and polystyrene (PS) plastics were pelleted to the size of 2-4 mm. Then, these samples were characterized using proximate analysis. The result of proximate analysis for the feedstocks is presented in Table 1.

| Sample  | Moisture | Ash | Volatile Matter | Fixed Carbon |
|---------|----------|-----|----------------|-------------|
| Lignite | 17.10    | 6.51| 59.15          | 17.24       |
| PE      | 0.34     | 3.37| 95.61          | 0.68        |
| PP      | 0.30     | 5.48| 93.40          | 0.82        |
| PS      | 0.24     | 0.48| 96.18          | 3.10        |

Pyrolysis reaction was investigated in a stainless steel tubular reactor with 16 cm of diameter and 25 cm of height. For every run, 500 grams of lignite was used. This sample was heated up to 500°C at the heating rate of 8°C/min and held at that temperature for 1 hour. The pyrolysis temperature (500°C) was chosen as observed by Kojic et al.[13] that at that temperature showed the positive synergetic effect. The vapor that produced from the reactor was flown through the connecting pipe and condensed through the condenser then the result was collected as pyrolysis oil in the flask. When the reaction was completed, the reactor was cooled down to room temperature so it could be opened to collect the char (solid product). The yields of product were obtained from the weight percentage of liquid, gas and char products to the total weight of sample used for every reaction. Furthermore, the pyrolysis oil was separated from its aqueous phase (contains much water) to get the organic phase that would be analyzed to measure its physical properties such as: heating value, viscosity and density. Furthermore, the pyrolysis of lignite and polyethylene (L/PE), polypropylene (L/PP) and polystyrene (L/PS) plastics were conducted as the same method as pyrolysis of lignite. The weight ratio of lignite:plastics used was 3:1.
3. Results and Discussion

3.1. Effect of plastic type on pyrolysis products yields

The yield of product is highly influenced by temperature, reactor configuration, type of biomass, catalyst and heating rate [14]. The pyrolysis of lignite will produce products in the form of liquid pyrolysis (tar and water), gas and char. The addition of plastic waste to lignite pyrolysis is expected to affect the yield of the resulting liquid pyrolysis. The yield char, gas and pyrolysis liquid of each type of plastic addition can be seen in Figure 1.

![Figure 1. Effect of plastic type on the yields of pyrolysis products.](image)

From Figure 1, the different type of plastic used resulted in different trend of products yield compared to the result of lignite pyrolysis itself. In general, the highest char product was resulted from lignite pyrolysis (55.61%), while the lowest was produced from L/PS at 46.74%. On the other hand, L/PS resulted the highest liquid yield at 36.92% compared to L/PE, L/PP and lignite pyrolysis while the smallest resulted from L/PP. As observed by Pinto et al [10], that found pyrolysis of PS resulted the highest liquid yield compared to PE and PP. As observed by Kiran et al. [15] the PS pyrolysis yielded higher liquid than that of PE and PS. L/PP pyrolysis performed the highest gas product at 32.96%. In contrast, the gas resulted from L/PS pyrolysis was the lowest. These results imply that the addition of plastic PE, PS and PP in lignite pyrolysis will increase the production of liquid and gas, but lowering char products. These in accordance to Qian's (2014)[16] research on product yield with the addition of PE and PP at various compositions, the most products formed were tar and gas due to the high hydrocarbon content. Furthermore, the pyrolysis liquid product consists of two different phases, namely the organic phase (tar) and the aqueous phase (water). The organic phase of the liquid pyrolysis product has the potential to be used as fuel. The yields of the organic phase and the aqueous phase between the type of plastic waste used (PP, PE, PS) during lignite pyrolysis is presented in Figure 2.
As can be seen in Figure 2, the addition of three types of plastics shows significant differences in results compared to lignite alone. The order of the organic phase yield from the lowest to the highest is lignite, L/PP, L/PE and L/PS. In lignite pyrolysis, as the temperature increases, free radicals will be generated when the linking bonds between the basic structural units of lignite are broken so that hydrocarbon gases can be released from the system[17]. This causes the yield of gas and organic phase increase, while char yield decline [18-21]. In addition, Qian et al. [16] stated that free radicals from lignite participate in plastic decomposition reactions, and plastics act as a hydrogen donor for free radicals from lignite. In the co-pyrolysis process, the less active hydrogen from lignite is combined with the hydrogen from plastic, and the combination of these two can form a molecule with the same size of liquid pyrolysis [18]. Thus, the organic phase yields increased. Sinag et al.[22] observed a similar trend with the addition of LDPE plastic to Mustafa Kemal Pastia (MKP) lignite compared to the absence of LDPE. Moreover, lignite pyrolysis itself without the addition of plastic will require a higher pyrolysis temperature [22].

3.2. Effect of plastic type on pyrolysis liquid properties
Analysis of physical properties is carried out to determine the viscosity, density and also heating value of the organic phase. The results of the analysis and also the properties of some types of other fuel can be seen in Table 2.

Table 2. Properties of Pyrolysis Liquid (Organic Phase) Compared with Fuels

|                  | High Heating Value (cal/g) | Kinematic Viscosity at 20°C (mm²/s) | Density at 20°C (kg/m³) |
|------------------|----------------------------|-------------------------------------|-------------------------|
| L/PE             | 10935.859                  | 9.0478                              | 825                     |
| L/PS             | 9823.637                   | 1.3543                              | 878                     |
| L/PP             | 10338.097                  | 9.0258                              | 804                     |
| Crude oil [23]   | 11059.000                  | 5 – 20                              | 700-1000                |
| Gasoline [23]    | 10605.000                  | 0.5                                 | 720-760                 |
| Diesel [23]      | 10844.000                  | 2 – 4                               | 780-860                 |
| Biodiesel [23]   | 10079.000                  | 4 – 6                               | 860-900                 |
| Jet Fuels (kerosene based) [23] | 10461.000 | 8                                   | 760-810                 |
Based on the data in Table 2, the heating value of the organic phase resulting from pyrolysis shows a value that is not much different from other fuels. Bhattacharya et al. [24] reported the co-pyrolysis of PE mixed with pine wood caused the reduction of oxygen and water content that increase high heating rates. L/PE has the highest heating value. This is because the H element in PE is the highest at 14.3% [25] compared to other types of plastics. L/PE and L/PP meet the specifications as fuel oil because generally fuel oil has a heating value of 10079-10844 cal / g. Meanwhile, L/PS still has a heating value below the fuel oil specification. The kinematic viscosity of the organic phase obtained in the three types of plastics in this study is still between the viscosity values for fuel. When viewed from the viscosity value, L/PE and L/PP are close to the characteristics of kerosene-based jet fuels. However, the L/PS viscosity is almost not comparable with other types of oil. As for the density parameters L/PE and L/PP are included in the diesel oil range.

4. Conclusion
The different type of plastic used results different trend of products yield compared to the result of lignite pyrolysis itself. These results imply that the addition of plastic PE, PS and PP in lignite pyrolysis will increase the production of liquid and gas, but lower char products. Moreover, the addition of three types of plastics shows significant differences in results compared to lignite alone. The order of the organic phase yield from the lowest to the highest are: lignite, L/PP, L/PE and L/PS. The properties in the term of heating value, viscosity and density of the organic phase from pyrolysis of lignite and PE and PP in this study are almost similar to other fuels, however, the heating value and density of lignite and PS are not comparable with others.
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