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IOP Conference Series: Earth and Environmental Science

PAPER • OPEN ACCESS

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To cite this article: H Wijayanti *et al* 2022 *IOP Conf. Ser.: Earth Environ. Sci.* **963** 012012

IOP Conf. Series: Earth and Environmental Science **963** (2022) 012012 doi:10.1088/1755-1315/963/1/012012

Copyrolysis of rice husk and plastic bags waste from low-density polyethylene (LDPE) for improving pyrolysis liquid product

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Abstract. Rice husk is an agricultural waste from the rice milling process that results in environmental problems during its handling. Biomass (such as rice husk) can be transformed into biofuel via pyrolysis. Pyrolysis is a thermal decomposition of organic compounds in biomass at high temperatures in the absence of oxygen. The main difference of pyrolysis liquid compared to fossil fuel is the significant amount of oxygen (O) content instead of its carbon (C) and hydrogen (H). Thus, making pyrolysis liquid has a lower heating value and inferior properties limiting its direct fuel application. On the other hand, plastic bag waste from low-density polyethylene (LDPE) contains a high C and H with almost no O content. This study conducted co-pyrolysis of rice husk and LDPE at a different percentage (%weight) of LDPE (0, 5, 15, 25, 50, and 75). The pyrolysis temperature was measured using a comparison of thermogravimetric analysis (TGA) curves of rice husk powder and LDPE. The pyrolysis liquid resulted was collected and analyzed. The results show that the increase of LDPE content caused the yield of the organic phase (desired as fuel) of pyrolysis liquid to increase. The properties in the term of heating value also increased while the viscosity and density of pyrolysis liquid decreased. These properties were almost similar to diesel fuel with a slightly lower heating value. However, at 50 % LDPE composition, the wax was formed. Therefore, the optimum composition was 25% of LDPE, resulting in 41.69 MJ/kg of heating value.

1. Introduction

The lack of fossil fuel sources has changed the trend to develop renewable energy sources as alternatives for fossil fuel substitution. Biomass is one of the promising renewable energy sources. Its abundant and sustainable sources make it is more interesting. Moreover, overall, it does not contribute to the greenhouse effect due to its utilization as biofuel performs zero net CO_2 emissions in the atmosphere [1].

The world's three biggest food crops are maize, rice, and wheat. These crops provide calories of the total world population for more than 42% [2]. Global rice consumption in 2016 reached 518 million tonnes, and it is also predicted will be 570 million tonnes in 2025 [3]. The increase in rice consumption is followed by a large amount of waste from the rice milling process. Prasara and Grant [4] reported that the rice husk from the milling process reaches 23% of the total rice produced.

Biomass (such as rice husk) can be transformed into biofuel via pyrolysis. Pyrolysis is a thermal decomposition of organic compounds in biomass at a high temperature in the absence of oxygen. The pyrolysis products are in the form of char (solid), pyrolysis liquid, and gas [5,6]. The yield and chemical composition of pyrolysis products were affected by the temperature, reactor configuration, type of biomass, catalyst, and heating rate [7]

The main difference of pyrolysis liquid compared to fossil fuel is the significant amount of oxygen (O) content instead of its carbon (C) and hydrogen (H) composition. Thus, make pyrolysis liquid has lower heating value and unstable properties, limiting its direct application for fuel [8]. The previous research about rice husk pyrolysis found that O content in the rice husk pyrolysis liquid was in the range of 23,38-61,72%, and its heating value was about 28 MJ/kg [9, 10, 11].

Nowadays, the utilization of materials from plastics cannot be avoided in daily life. This increase is followed by a large amount of plastic waste resulted. The most used plastics variant was plastic bags made from low-density polyethylene (LDPE). Pinto et al. [12] investigated that polyethylene contained a high amount of C and H with almost no O content. Therefore, its heating value was approximately 46 MJ/kg, higher than diesel oil at 42-43 MJ/kg [13].

The previous research concluded that the co-pyrolysis of wood and plastics improved pyrolysis liquid properties [14]. In addition, the ratio of C/H and H/C from co-pyrolysis switchgrass and plastic increased compared to pyrolysis of switchgrass itself [15]. However, the study about the amount of plastics composition during co-pyrolysis with biomass is rare published and still interesting to be investigated further. Therefore, this study was conducted to investigate the prospect of rice husk as a biomass source for pyrolysis liquid that focused on the effect of LDPE addition to the rice husk-LDPE co-pyrolysis products on the pyrolysis liquid product.

2. Materials and method

2.1. Materials preparation

Rice husk was collected from the local rice milling plant at Banjar district, South Kalimantan, Indonesia. It was crushed and sieved to 0.25-1 mm of particle size. Then the powder was dried in an electric oven. The waste of LDPE plastic bags was pelletized to 2-3 mm of pellets diameter. Then, both samples were characterized through proximate and ultimate analysis. The proximate analysis was conducted to determine the moisture content (ASTM D3173), ash content (ASTM E1755), and volatile matter (ASTM D3175) in the rice husk. Meanwhile, the ultimate analysis was performed using a LECO model 628 series CHN 2000 elemental analyzer. The heating value of them also is measured by using an IKA C 6000 isoperibol oxygen bomb calorimeter.

2.2. Pyrolysis reaction

The pyrolysis temperature was measured using a comparison of thermogravimetric analysis (TGA) curves of rice husk powder and LDPE pellets using thermogravimetry analyzer Linseis STA PT 1600. Approximately 15 mg of sample was heated at a rate of 10 ^{0}C/min up to 600^{0}C under 50 ml/min of nitrogen. Then this selected pyrolysis temperature was used to conduct the pyrolysis reaction using a pyrolysis reactor. Pyrolysis reaction was conducted in a fixed bed tubular reactor with 16 and 25 cm of diameter and height, respectively. 500 gram of feed (rice husk and LDPE pellet) was used for each run. The weight percentage of LDPE pellets to the total feed was 0 (rice husk), 5, 15, 25, 50, 75, and 100%. This sample was heated up at the selected pyrolysis temperature and held at that temperature for 1 hour. The pyrolysis vapors produced were condensed in the condenser then the result was collected as a liquid product. After 1 hour of reaction, the reactor was cooled down to room temperature to be opened to collect the char (solid product). Gas product was calculated from the mass balance. Furthermore, this liquid product was separated into the organic phase (heavy phase) and aqueous phase (liquid phase). The organic phase would be analyzed to measure its physical properties such as heating value, viscosity, and density.

3. Result and discussion

3.1. Characteristics of raw materials

The proximate and ultimate analysis of both rice husk and LDPE and their heating value are presented in table 1. To maintain the effectiveness as a fuel, the moisture content of raw materials should be

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lower than 10 wt% [8]. From table 1 also can be seen that rice husk has a low heating value due to its high oxygen content, which lowers the heating value [16]. On the other hand, LDPE contains a high heating value resulting from its high carbon and hydrogen content.

Table 1. Characteristics of rice husk and low-density polyethylene (LDPE)

Analysis		Value	
		Rice husk	*LDPE*
Proximate analysis (% wt)	Volatile matter	68.02	95.61
	Ash	8.29	3.37
	Fixed carbon	15.35	0.68
	Moisture	8.34	0.34
Ultimate analysis (dry and ash free,% wt)	Carbon	34.4	85.7 [12]
	Hydrogen	5.07	14.3 [12]
	Nitrogen	0.35	-
	Oxygen	37.28	-
	Sulfur	0.04	-
	Ash	22.86	-
Heating Value (MJ/kg)		15.48	46.50

3.2. Thermogravimetric analysis of raw materials

Thermogravimetric analysis (TGA) was used to determine the pyrolysis temperature of rice husk and LDPE. This analysis determines the change in weight concerning the change in temperature [17]. From the TGA curve, the weight loss of the sample with the increase in temperature could be obtained. The TGA curves of rice husk and LDPE are shown in figure 1. The results show that overall, the decomposition curves consist of three regions. Due to water removal, the first region was 100-200°C for rice husk and 100-300 °C for LDPE, followed by the second region, which shows the highest weight loss at 270-350°C and 400-500°C for rice husk and LDPE, respectively. Rice husk began to slowly decompose at 380°C, while LDPE was at 500°C. The maximum weight loss due to decomposition of sample is constant at 500°C for LDPE. The obtained result was in good agreement with that of Papuga et al. [18], whereas rice husk continues to decline to an almost constant decomposition rate at 600°C. As a consequence of both curves, the selected pyrolysis temperature for pyrolysis reaction of rice husk and LDPE mixing was 500°C.

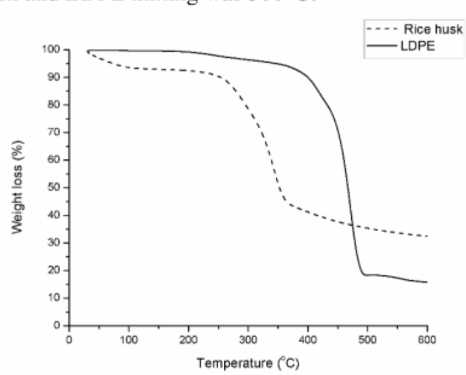

Figure 1. Thermogravimetry curves of rice husk and LDPE

3.3. Effect of LDPE composition on products yields and composition

To study the effect of LDPE addition on product yields and composition, the co-pyrolysis of rice husk and LDPE was conducted at the previously selected temperature (500°C) with the various components of LDPE (in wt%): 0, 5, 15, 25, 50, 75 and 100. Pyrolysis of biomass will produce three products (i.e., solids (char), liquid (pyrolysis liquid) and, gas, so does in co-pyrolysis of biomass and plastics. These products' composition will vary depending on the pyrolysis conditions, such as raw material composition [19]. The effect of LDPE addition in co-pyrolysis of rice husk-LDPE on the yield of the products is shown in figure 2.

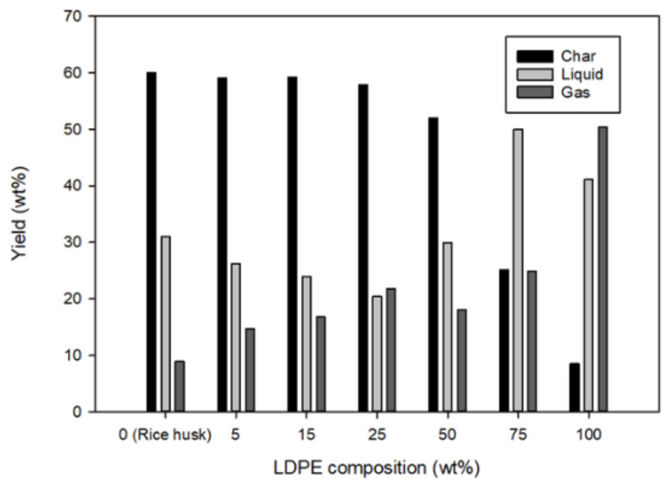

Figure 2. Effect of LDPE composition during co-pyrolysis of rice husk-LDPE on the yields of the products

It can be seen in figure 2 that LDPE addition in pyrolysis of rice husk decreased the yield of char resulted. The increase of LDPE content was followed by the decline of char. Because from the TGA result, the weight loss at 500°C was lower than that of rice husk. In contrast, at the range of 5 to 25 %, the liquid content was slightly decreased as the gas production increased. Possibly this condition prefers to decompose the sample to gas. However, the liquid product increased as the LDPE content rose from 50 to 75% because more H is supplied from LDPE, which leads to an increase in liquid production [20]. Furthermore, the liquid product was separated from the aqueous phase (containing much water) to get the organic phase desired and further analyzed due to its potency as fuel. The effect of LDPE addition to organic and aqueous phase yields is presented in figure 3.

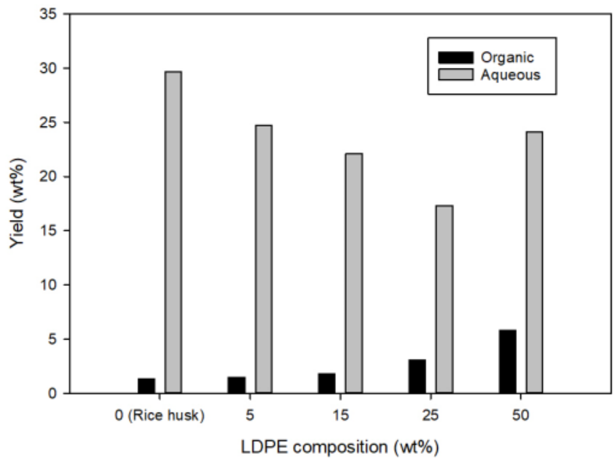

Figure 3. Effect of LDPE composition during co-pyrolysis of rice husk-LDPE on the yields of the organic and aqueous phase in pyrolysis liquid

From figure 3, the yield of the organic phase was increased significantly as LDPE content raised. However, at 50 % LDPE, wax formed in the organic phase product, and its quantity became more significant as the LDPE content increased. Figure 3 shows that the organic phase rose as the LDPE content increased. This implies that at the same time, the rise of LDPE would enlarge the percentage of C and H but also reduce the O content.

3.4. Effect of LDPE composition on the pyrolysis liquid properties

The pyrolysis liquid properties analysis was carried out to measure its viscosity, density, and heating value. Table 2 presents the result of pyrolysis oil analysis. In addition, this table also shows the conventional fuel from fossil fuel (Indonesian-based companies) as a comparison.

Table 2. Properties of pyrolysis liquid (organic phase) compared with fuels

LDPE (wt%)	Viscosity (mm^2/s) (40 °C)	Density (g/cm^3) (20 °C)	Heating Value (MJ/kg)
0	6.0518	0.8913	39.39
5	1.8268	0.8339	40.13
15	1.1744	0.7979	40.26
25	1.5251	0.8074	41.69
50	2.1689	0.8231	39.69
75	*Wax*	*Wax*	Not measured
100	*Wax*	*Wax*	Not measured
Fossil (conventional) fuels [21]			
Diesel fuel (Pertamina)	2.0-4.5	0.815-0.860	44.15
Diesel fuel (Total)	2.5-11	0.900-0.920	44.97

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As shown in table 2, the viscosity of pyrolysis liquid from rice husk was 6.0518 mm^2/s. It was lowering to the range of 1.1744 – 2.1689 mm^2/s with the addition of 5 to 50 % LDPE. The same trend was observed in pyrolysis liquid density. It implies that decomposition reaction during pyrolysis tends to produce more water through dehydration and cracking reaction (figure 3 shows the water content represented by the aqueous phase composition). Zadgaonkar [22] found that water contributed to lowering density and viscosity. Both of these properties were not far from fossil-based fuels. Those are presented in table 2.

Compared to the heating value of fuels from fossils, the heating value resulting from this study was slightly lower. However, the addition of 5 to 25 % of LDPE during pyrolysis of rice husk was successfully increasing the heating value of pyrolysis liquid resulted. The heating value increased significantly, indicating that more hydrocarbon was produced in the liquid product due to LDPE addition (figure 4). The increase of hydrocarbon contributes to the increase of heating value [23]. Even though it further decrease in 50 % LDPE due to the wax production.

Table 2 also indicates that the properties of pyrolysis liquid from co-pyrolysis rice husk and LDPE (25% LDPE) in this study almost similar to diesel fuel, however, the heating value are slightly lower possibly due to the oxygenated compounds those still contained in the pyrolysis liquid (figure 4) [24].

4. Conclusion

The yield of the organic phase, which is desired as fuel substitution, was increased significantly as LDPE content raised. However, at 50 % LDPE, wax formed in the organic phase product, and its quantity became more significant as the LDPE content increased. Rice husk pyrolysis resulted in more oxygenated compounds than hydrocarbons. The addition of LDPE lowered these compounds but increased hydrocarbons. Finally, the oxygenated compounds were not detected at higher LDPE content, 50%. This study's properties of pyrolysis liquid from co-pyrolysis rice husk and LDPE (25% LDPE) are almost similar to diesel fuel. However, the heating value is slightly lower, possibly due to the oxygenated compounds still contained in the pyrolysis liquid. Thus, study about reducing oxygenated compounds in the pyrolysis liquid should be further investigated.

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Acknowledgments

The authors wish to thank the Directorate of Research and Community Service, The Ministry of Research, Technology and Higher Education of Indonesia for Research University Grant Year 2019.

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