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Research Paper

The Production of Renewable Fuels Sago Dregs and Low-Density Polyethylene by Pyrolysis and its Characterization

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Abstract
Biomass has been suggested as a sustainable alternative to substitute fossil fuels. Based on the pyrolysis method, the biomass would be converted into energy through decomposition by thermal degradation under an inert atmosphere, resulting in charcoal, liquid, and gas products. The quality of oils is effectively enhanced through the pyrolysis of lignocellulosic biomass and plastic due to the facilitation of deoxygenation by plastics. This study investigates the impact of incorporating low-density polyethylene (LDPE) plastic in co-pyrolysis with sago dregs (SDs) waste. Pyrolysis of SDs and LDPE mixtures with ratios of 5:1, 4:2, 3:3, 2:4, and 1:5 at various temperatures of 375°C, 425°C, and 475°C. The maximum oil yield obtained for SDs and LDPE pyrolysis was 44.94%. The calorific value (CV) of all observed compositions is a minimum of 10,579.57 kcal kg^{-1} and a maximum of 11,545.21 kcal kg^{-1}. The gas chromatography-mass spectroscopy (GC-MS) analysis confirmed the interaction between SDs and LDPE on co-pyrolysis. The addition of LDPE will produce rich aliphatic and aromatic compounds, like the proportions of alkanes (45.53%), alkenes (30.62%), alcohol (0.4%), and benzene (17.68%). Co-pyrolysis of SDs and LDPE promotes enhanced oil production by reducing oxygenated compounds and increasing hydrocarbon compounds.

Keywords
Co-Pyrolysis, Fuel, Hydrocarbon, Low-Density Polyethylene, Sago Dreg

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1. INTRODUCTION

The human need for fossil fuels is increasing every year. What is more, fossil fuels are materials that take thousands of years to form. Because it is constantly in use, more of it will likely be required, which makes sense, given the population growth. It means that energy demand will continue to rise, and people will always need more energy. Almost all the energy people need comes from converting fossil fuels, such as those used to make electricity and fuel for cars. Since fossil fuels have run out, scientists have been looking for other energy sources. The estimated coal reserves in 2112 and crude oil reserves in 2042 will be exhausted, while the world's energy needs in 2040 will continue to increase by 56% (Hwang et al., 2019). According to Hwang et al. (2019), the escalating utilization of fossil fuels has led to environmental challenges, including the emission of greenhouse gases and a decline in air quality attributed to pollutants such as NO_x, SO_x, and delicate particulate matter. The world economy is in danger because fossil fuels are running out, increasing prices (Ryu et al., 2020).

Renewable energy sources (RES) such as wind, biomass, solar, and geothermal energy, along with perceptions regarding their practicality, have been employed globally (Clauser and Ewert, 2018). During these three decades, research about RES focused on biomass (56%) (Ochoa et al., 2019). The three most important things to consider when deciding which alternative energy source to use are availability, cost, and environmental effect. Biomass has been developed and holds the potential to serve as a solid, liquid, or gaseous fuel. Some benefits are that it is flexible, there is a lot of it, and it cuts down on wood waste, agricultural waste, energy-specific crops, and solid waste in cities.

The rapid pyrolysis of biomass produces two kinds of output: liquid and solid. The liquid product is referred to as bio-oil/oil, while the solid product is known as biochar/char. Pyrolysis of biomass is a technology used to make liquid fuels to replace or add to liquid petroleum or other fossil fuels (Roy and Dias, 2017). The pyrolysis method can produce about 50–75% liquid fuel under moderate operating conditions to make biofuels (Djandja et al., 2020). It involves heating the biomass raw material components to a temperature of ~ 500°C and cooling the smoke/gases to obtain oil (Roy and Dias, 2017).

The product of biomass pyrolysis is a brownish, viscous oil with a higher calorific value (CV) than the original biomass (Singh et al., 2020). Although it provides benefits related to environmental safety over fossil fuels and a High Heating Value (HHV ≈ 17 MJ kg^{-1}) (Han et al., 2017; Remón et al., 2016), using bio-oil as a 'drop-in' transport biofuel is not possible. The consequences of a high oxygen/carbon (O/C) ratio, high viscosity, high acidity, and easily corrosive (Onokwai et al., 2022; Remón et al., 2021). It concerns how the reactive oxygen in bio-oil, which comes directly from biomass, works. Because of these problems, bio-oil needs to undergo a process of upgrading that includes hydrotreating, hydrocracking, and hydrodeoxygenation (Lahijani et al., 2022).

Biofuel made from pyrolysis is good for the environment, so one way to solve this problem is to use plastic waste and biomass as renewable fuel sources. The co-pyrolysis of biomass with various types of plastics, including polypropylene (PP), low-density polyethylene (LDPE), and polystyrene (PS), has been suggested by Dyer et al. (2021) and Wang et al. (2021). The integration of biomass and plastics to enhance the quality of the pyrolysis oil demonstrates positive potential (Dewangan et al., 2016). The sago plant, indigenous to Indonesia, stands as one of the most abundant biomass sources, covering approximately 1.128 million hectares, which accounts for 51.3% of the world's sago area. The sago agriculture and plantation industry produce solid, liquid, and gas by-products. One of the agricultural wastes in solid form is sago dregs (SDs) obtained from the final process of harvesting sago trees (Sahupala and Kakerissa, 2022). Due to its potential and versatility, sago is recognized as one of the commodities suitable for use as a raw material in fuel production.

SDs waste biomass, according to Hammado et al. (2020), contains 15.18% hemicellulose, 36.32% cellulose, and 12.34% lignin. It contains phenol derivatives, alcohols, aldehydes, acids, ketones, and furans, which have higher acidity polarity, viscosity, low CV, and low stability, thus limiting their use in liquid fuels and other platform chemicals. The main criterion for using plastic as a co-feed in biomass pyrolysis is that biomass is typically deficient in hydrogen, which is one of the key factors contributing to low bio-oil yield. Co-feeding with a hydrogen-rich feedstock can enhance the oil yield's quantity and quality.

The characteristics of plastic are polymers rich in carbon and hydrogen, which are more likely to produce oil with a CV close to that of conventional fuels, which is 40 MJ kg^{-1} (Fulgencio-Medrano et al., 2022). Meanwhile, biomass is a raw material that lacks hydrogen. Plastic waste is made by polymerizing olefins with a ratio of 2 of hydrogen to carbon. LDPE is a polyethylene plastic with a low density made from ethylene monomers (C_2H_4) (Dewangan et al., 2016; Samal et al., 2021; Yang et al., 2016). Dewangan's study focused on the co-pyrolysis of sugarcane bagasse (SCB) and LDPE in different ratios, conducted at temperatures from 350 to 600°C with a heating rate of 20 ± 1°C per minute. Notably, when a 1:1 mixture of SCB and LDPE is co-pyrolyzed at 500°C, the resultant oil exhibits a greater liquid yield, reduced water content, and an elevated heating value of 40 MJ kg^{-1} (Dewangan et al., 2016). Furthermore, (Yang et al., 2016) investigated the co-pyrolysis of cedar wood and LDPE with a 1:1 ratio at temperatures ranging from 500 to 600°C (Yang et al., 2016). When combined with SDs in the pyrolysis process, they can enhance the properties and yields of the resulting oil (Zhang et al., 2014). LDPE, characterized by a higher H/C ratio, elevated CV, and lower oxygen content, emerges as a suitable candidate for co-pyrolysis with SDs, given its advantageous characteristics (Dewangan et al., 2016; Samal et al., 2021).

In the investigations conducted by Dewangan et al. (2016) and (Samal et al., 2021), it was reported that the thermal degradation of LDPE into hydrocarbon fuels was effective. Consequently, this study incorporates LDPE as a component in the pyrolysis process. The utilization of SDs/LDPE co-pyrolysis represents a recent advancement in research to enhance the yield of higher-quality bio-oil. The determination of the physical and chemical properties of the bio-oil was carried out under the specified conditions.

2. EXPERIMENTAL SECTION

2.1 Materials and Instruments

The waste material from a plantation in South Konawe, Southeast Sulawesi, Indonesia, was used to produce SDs. The plastic utilized in the study is LDPE. The SDs were dried in an oven at 40°C for 24 hours and ground into a fine powder using a grinder. The SDs were sieved to maintain a consistent size range of 100 mesh, while LDPE pellets had a diameter of approximately ±2 mm. The co-pyrolysis of SDs and LDPE used a laboratory-scale, semi-batch reactor made of stainless steel (45 cm high and 60 cm in diameter) (Figure 1). Analysis of chemical compounds using gas chromatography-mass spectrometry (GC-MS) in a Shimadzu QP 2010 SE equipped with an Rtx-5MS column. The flowchart summarizing the research process is depicted in Figure 1.

2.2 Pyrolysis Methods

The reactor system consists of a temperature controller and a condenser, which will condense the condensed gas with the help of a cooling medium. The reaction started when it reached the right temperature, making a liquid of heavy oil (tar) and light oil (hydrocarbon oil). The pyrolysis process there is a modified method that has been done by Dewangan et al. (2016); Ryu et al. (2020). The method was fast pyrolysis at temperature variations of 375, 425, and 475°C with varying SDs and LDPE ratios of 5:1, 4:2, 3:3, 2:4, and 1:5 at a heating rate of 12 ± 1°C with a duration of 20 minutes. The byproducts of pyrolysis included oil, char, and gas or volatiles. The oils were filtered using filter paper to separate them from the tar. The product yield of bio-oil is determined by measuring the volume (mL) produced by multiplying the bio-oil's density (g/ml). So, the product results are obtained in gram units. Carbon yield is calculated by initial weight/final weight × 100%.

2.3 Physical Analysis of the Oil Product

The American Society for Testing and Materials (ASTM) standard method assessed the physical properties of both SDs and LDPE. These properties include moisture content, pH value, viscosity, density, specific gravity (SG), API gravity, and CV.

The moisture content of the bio-oils was determined according to IP 74/82 and ASTM D9583 using the Dean and Stark apparatus through distillation. A sample of 10 mL was placed into a 250 mL round bottom flask with approximately 30 mL of toluene. The water was separated into a reservoir under azeotropic conditions. Following the separation of water from the oil, subsequent physical analyses were conducted.

The pH value of the oil was determined using a pH meter (WT61) calibrated with pH buffer solutions of 4, 7, and 10. The oil's viscosity was measured using an Ostwald viscometer, which determines the fluid flow rate through a tube.

The densities, SG, and API gravities of the oils were determined by IS 1448 [P:32]:1992 and ASTM D1298-12B (2017). They compare distilled water and oil weights at the same temperature. SG and API gravity are calculated based on Equations 1 and 2:

$$\mathrm{SG} = \frac{\text{Density of sample}}{\text{Density of } \mathrm{H_2O}} \tag{1}$$

$$\text{API Gravity} = \frac{141.5}{\mathrm{SG}} - 131.5 \tag{2}$$

The determination of CV is calculated based on the following Equation 3:

$$\mathrm{CV} = \frac{2.2046226}{3.9673727} \times ((18,650 + 40) \times (\text{API Gravity} - 10)) \tag{3}$$

2.4 Analysis of GC-MS

The GC-MS temperature was programmed to start at 70°C for 3 minutes and then ramped up to 300°C at a rate of 10°C/min, with a total runtime of 25 minutes. A 1 μL injection of the oil sample was introduced into the column using helium as the carrier gas at a flow rate of 1.5 mL/min. Chemical compounds present in the oil were ionized at an energy level of 70 electron volts (eV) within an ion source temperature of 230°C and analyzed across an electron mass range (m/z) of 40–700. Chromatograms displaying chemical compounds at various retention times and their respective mass spectra were generated and compared with spectral data from the NIST MS library for identification purposes.

3. RESULTS AND DISCUSSIONS

3.1 Yield of Pyrolysis Products

This research was conducted by directly mixing SDs and LDPE waste. The pyrolysis method converted SDs and LDPE into fuel or hydrocarbon oils, where the temperature settings varied, namely 375, 425, and 475°C. Heating in the pyrolysis reactor causes SDs and LDPE to decompose into a gas phase, then that gas is condensed into the oil. The direct mixing of biomass and plastic in pyrolysis is called co-pyrolysis. This technique leverages the synergistic effect between biomass and plastic throughout the process. Several studies have described cracking polymer waste into valuable compounds with lower molecular weights that can be used as fuels. Hence, biomass and plastic produce more stable and homogeneous oil than pyrolysis without mixing. Meanwhile, bio-oil from biomass is polar and unstable because it phase separation in a short time. Pyrolysis of LDPE at a temperature of 475°C resulted in an oil yield of 18.70%. Meanwhile, SDs were reported to yield an oil content of 44.94%. The study concluded that the oil yield from biomass increased with the severity of heating but decreased with the presence of small particles (Montoya et al., 2015). Therefore, the positive effect of mixing SDs and LDPE enhances the yield of pyrolysis products (Figure 2).

3.1.1 Effect of Composition on Pyrolysis Yield

The pyrolysis of the SDs: LDPE mixture was conducted within a temperature range of 375-475°C, utilizing various mixture ratios, including 5:1, 4:2, 3:3, 2:4, and 1:5, aimed at enhancing both the quality and quantity of the resulting oil. The outcomes of the pyrolysis process for the SDs: LDPE mixture are depicted in Figure 2. The effect of material composition on the yield of pyrolysis results shows that if pyrolysis by adding more plastic and less biomass, the amount of oil produced will be less. Figure 2a depicts the trend for the 1:5 composition ratio, where the oil yield initially measured at 375°C is only 16.40%. However, this yield exhibits a continuous increase with rising pyrolysis temperatures. This phenomenon can be attributed to the escalated gas production resulting from the increased temperature. More plastic and less biomass will affect the amount of carbon produced, evidenced by carbon yield at 375°C (Figure 2b), where the carbon produced is only 13.50% and continues to decrease with increasing temperature. If pure plastic is pyrolyzed, it will not produce carbon because it burns out at the appropriate pyrolysis temperature. The addition of biomass causes the carbon produced in pyrolysis.

According to Dyer et al. (2021), the augmentation in pyrolysis oil is attributed to the breakdown of solid molecules through cracking during the pyrolysis of biomass mixed with plastic. In the 5:1 variation, the carbon content produced is 46.17%, which is anticipated to decrease as the pyrolysis temperature rises due to the more significant proportion of biomass utilized. On the other hand, SDs exhibit a relatively high water content of 22.85%. When the temperature increases, incorporating plastic into the biomass enhances pyrolysis oil production. In cases where biomass or LDPE undergoes self-pyrolysis, the oil originates from the decomposition of lignocellulosic material or polyethylene, respectively (Chin et al., 2014; Yang et al., 2016). During pyrolysis, the oil is produced through the decomposition of lignocellulosic material and polyethylene

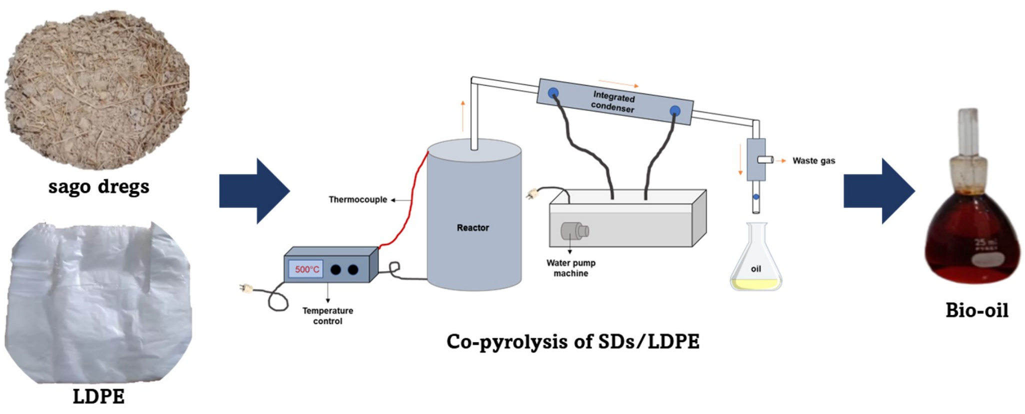

Figure 1. Flowchart of Research

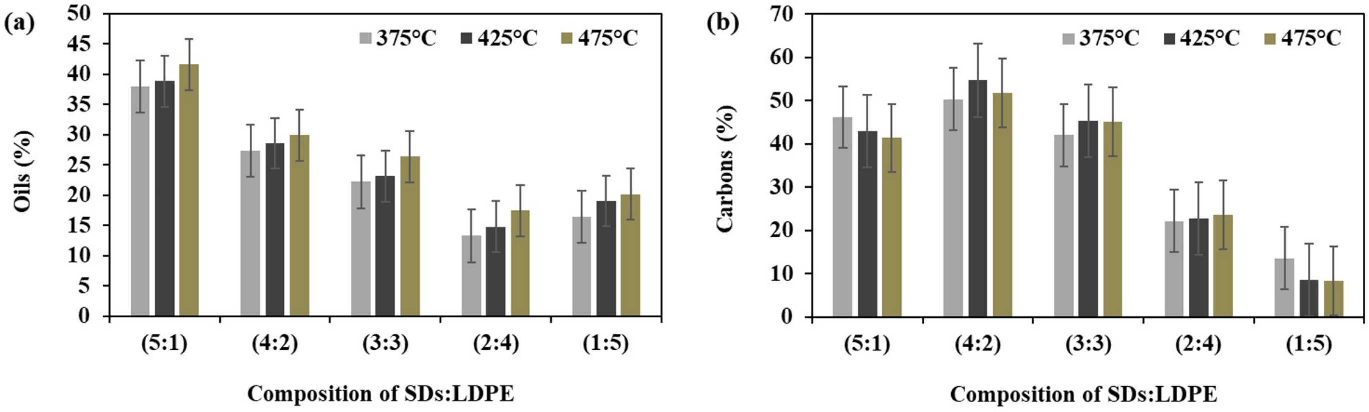

Figure 2. Effect of Composition and Temperature on Pyrolysis Yield (a) Oils and (b) Carbons

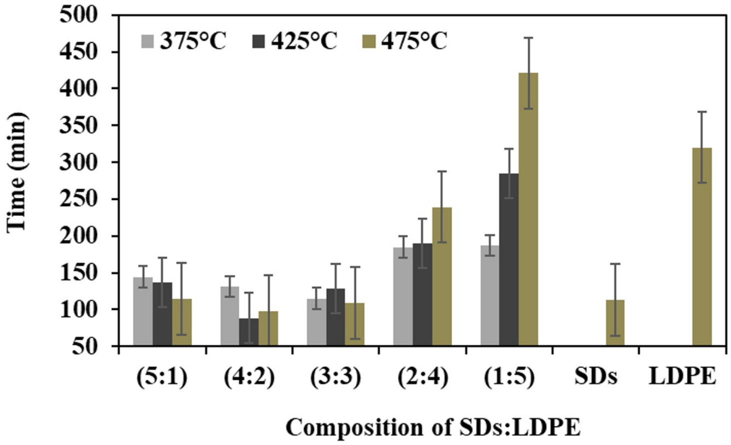

Figure 3. Effect of Temperature and Composition SDs and LDPE on Pyrolysis Time

and through reactions between their derivative products (Chin et al., 2014). In this case, higher temperatures generally promote this reaction. In the experiment, the 2:4 mixture made many waxes, which was considered more significant at a lower pyrolysis temperature. So, it is almost impossible to see how great co-pyrolysis is for making oil at low temperatures.

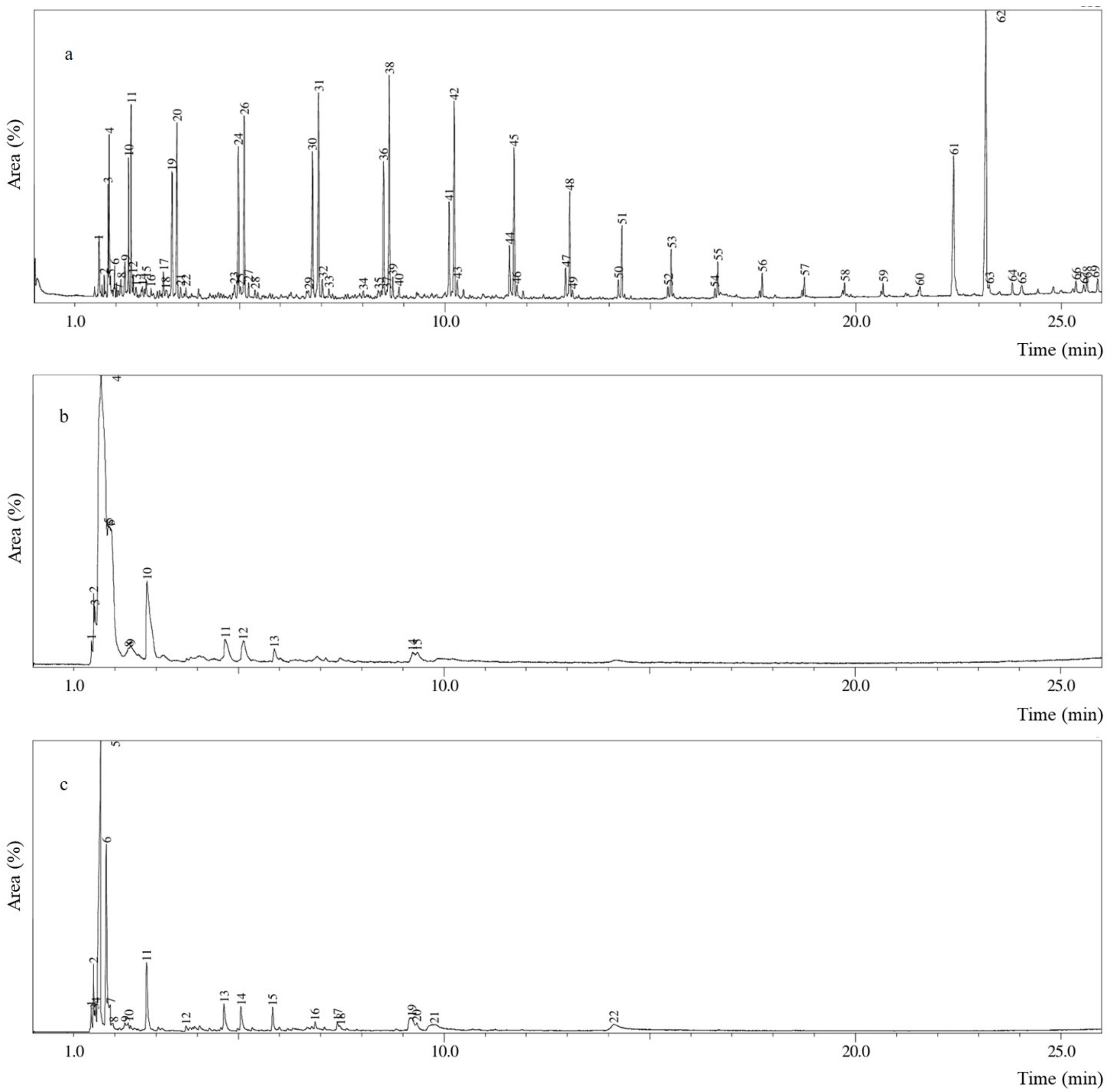

Figure 4. Chromatogram of Hydrocarbon-Oil Pyrolysis at Ratios of (a) 5:1, (b) 3:3, and (c) 1:5 (T, 475°C)

3.1.2 Effect of Temperature on Pyrolysis Yield

The results of the pyrolysis products, as depicted in Figure 2, indicate that the oil produced in each sample increases as the temperature rises from 375°C to 475°C. It suggests that higher temperatures lead to increased oil production. Similar observations have been reported in several other studies, indicating that cracking solid reactions occur at elevated temperatures, leading to the secondary decomposition of charcoal residue and maximizing oil production (Dewangan et al., 2016). The percentage increase of oil produced for the composition of 1:5 at 475°C was 21.36% higher than at 425°C and 375°C, respectively 19% and 16.4%. The remaining carbon produced at a temperature of 475°C is lower than 375-425°C and has a large percentage of gas. Research on biomass pyrolysis with plastics has indeed been conducted, demonstrating that the operation temperature, typically within the range of 400-600°C, is crucial to maximizing oil production. However, it is worth noting that the optimum temperature for achieving maximum oil production may vary depending on the raw materials' characteristics, as indicated by other researchers (Uzoejinwa et al., 2018).

To investigate the synergistic effect of co-pyrolysis of LDPE and biomass, cedar, sunflower, and Fallopia japonica stems were initially pyrolyzed at a 1:1 ratio of 600°C. The resulting organic condensation products included tar from the pyrolysis of biomass and oil from the pyrolysis of LDPE. In the studies conducted by Yang et al. (2016), "oil" referred to the mixture of tar and oil produced through rapid co-pyrolysis. Their research indicates that each mixture of biomass and LDPE yields 64.08%, 57.17%, and 58.96% by weight, respectively (Yang et al., 2016). In our study, however, tar and oil were separated by filtering, which showed a slight difference in the yield. Oil produced at a composition of 5:1 at temperatures of 425 and 475°C increased, and the carbon produced decreased. Plastic in the pyrolysis process does not produce much carbon, while the carbon produced comes from the rest of the burner from SDs biomass. If we review the research of Terry et al. (2021), it is explained that the oil from the pyrolysis of plastics and biomass will increase to a specific temperature. The carbon yield will also decrease when the plastic composition and temperature increase. It is because when pure plastic is pyrolyzed, it will burn out and leave no solids. Meanwhile, gas will increase with increasing temperature.

The characteristic of LDPE has oil and the difference in the thermal stability of the polymer chain causes a large amount of gas that is not fully condensed, resulting in a low oil percentage. Both linear and branched hydrocarbons exhibit a decrease in thermal stability as the temperature increases. Consequently, at 475°C, the C=C bond undergoes more complete decomposition compared to lower temperatures, leading to a higher yield of volatile products (Zhao et al., 2021). As temperatures increase, the percentage of charcoal and gas yield becomes irregular. The findings confirm that the optimal temperature for the pyrolysis of SDs and LDPE is 475°C, as it maximizes the yield of oil products. The primary goal of pyrolysis is to enhance the quantity and quality of oil produced from biomass. LDPE serves as an additive in the pyrolysis process, and numerous studies have highlighted the improved quality of pyrolysis oil with higher volumes of additives. Therefore, the pyrolysis of SDs and LDPE in a 5:1 composition ratio at 475°C represents the most suitable condition for maximizing the production of hydrocarbon oil.

3.2 Physical Properties of Hydrocarbon-Oil Pyrolysis

The synergistic effect is essential in improving the quality of hydrocarbon oil. The physical properties of hydrocarbon oil, such as moisture content, pH value, viscosity, density, specific gravity (SG), API gravity, and CV, are presented in Table 1.

3.2.1 The Moisture Content

Moisture content is one factor affecting the quality of the pyrolysis product. The raw materials' moisture content can affect the yield, oil quality, and reduce the resulting product's level (Dewangan et al., 2016; Mei et al., 2020; Ryu et al., 2020). As a result, materials with high water content require high energy for the pyrolysis process, or in other words, at the same energy, materials with high water content produce less gas than materials with low water content.

Table 1 demonstrates that the water content in the pyrolysis products of SDs and LDPE plastic decreases as the pyrolysis temperature increases. It is due to a large amount of water lost at higher pyrolysis temperatures. Some of the products obtained have met the quality standards of fuel oil in Indonesia, which have a maximum water content of 0.05%. The high moisture content will produce more oil. However, it will reduce the quality of the oil produced because the condensed water vapor mixes with the resulting oil, thereby reducing acid levels. The high moisture content will cause a decrease in the CV.

3.2.2 The pH Value

Table 1 shows that the pH of the oil exhibits minimal variation, remaining within the range of 3-4, indicative of its acidic nature. This acidity is attributed to the predominant conversion of lignocellulose in the raw materials, producing higher amounts of acid and phenolic compounds. The oil's elevated content of acids and phenols contributes to its acidic pH value. According to Permana et al. (2021), bio-oil typically exhibits acidity with a pH ranging from 2.5 to 3.0, akin to the acidity of vinegar. The pH value of the pyrolysis products from SDs and LDPE, as indicated by the increasing oil moisture content, confirms that the high moisture content in the oil influences the resultant pH value.

3.2.3 The Density Value

Density, a measure of mass per unit volume of a substance, indicates the number of impurities resulting from a reaction (Davidson et al., 2020). The density of SDs is reported as 0.985 g cm^{-3} and LDPE is 0.7479 g cm^{-3} in Table 2. Table 1 presents the density analysis results for SDs and LDPE oils, with values falling within the range of 0.7446-0.7505 g cm^{-3}. These values are close to the density range of conventional gasoline, which typically ranges from 0.7100-0.7700 g cm^{-3} (Jahiding et al., 2021a).

Density serves as an indicator of the presence of impurities resulting from a reaction. According to Yusuf et al. (2023), the higher the percentage of heavy substances or impurities contained in the fuel, the greater the density of the fuel. These heavy substances, challenging to evaporate and prone to forming smoke or soot that does not combust entirely at temperatures ranging from 375 to 475°C, contribute to the density of the fuel. Consequently, as the proportion of plastic increases and the amount of biomass decreases, the density of the pyrolysis oil decreases. It is because the density of plastic pyrolysis oil, reported as 0.74 g cm^{-3} (Jahiding et al., 2021a), is lower than that of biomass, which is 0.99 g cm^{-3} (Jahiding et al., 2021a; Mashuni and Jahiding, 2021). Table 1 shows that more plastic and less biomass will get a lower density. Heavy substances in the pyrolysis oil are also the central element in the occurrence of carbon residues that can contaminate the engine and cause erosion of the vehicle fuel injectors. The density value has met the standard range density for premium fuel, which is 0.700-0.750 g cm^{-3} (Directorate General of Oil and Gas, 2018).

Table 1. Physical Properties of Hydrocarbon-Oil Pyrolysis

Parameters	Composition of SDs: LDPE				
	(5:1)	(4:2)	(3:3)	(2:4)	(1:5)
Pyrolysis temperature, 375°C					
Moisture content (%)	17.14	11.84	11.43	5.71	6.28
pH value	3.23	3.03	3.03	3.20	3.17
Density (g cm-3)	0.9	0.92	0.92	0.83	0.74
Viscosity (cP)	1.17	1.23	1.23	0.84	0.42
Specific Gravity	0.91	0.92	0.92	0.83	0.74
API Gravity	24.4	22.36	22.36	38.71	58.95
CV (kcal kg^{-1})	10,683.67	10,638.28	10,638.28	11,001.74	11,451.65
Pyrolysis temperature, 425°C					
Moisture content (%)	16.20	11.34	9.07	5.21	5.48
pH value	3.20	3.03	3.00	3.10	2.83
Density (g cm^{-3})	0.92	0.93	0.92	0.84	0.76
Viscosity (cP)	1.28	1.24	1.05	0.9	0.51
Specific Gravity	0.93	0.93	0.93	0.84	0.76
API Gravity	21.03	20.37	21.03	37.08	53.94
CV (kcal kg^{-1})	10,608.67	10,594.05	10,608.67	10,965.54	11,340.25
Pyrolysis temperature, 475°C					
Moisture content (%)	14.29	10.11	8.57	4.89	2.86
pH value	3.13	3.00	3.03	3.27	3.70
Density (g cm^{-3})	0.93	0.92	0.92	0.81	0.72
Viscosity (cP)	1.3	1.18	1.16	0.85	0.41
Specific Gravity	0.94	0.93	0.92	0.82	0.73
API Gravity	19.72	21.03	21.69	42.06	63.16
CV (kcal kg^{-1})	10,579.57	10,608.67	10,623.41	11,076.29	11,545.21

Table 2. Physical Properties of SDs and LDPE Oil Pyrolysis (T = 475°C)

Parameters	SDs	LDPE
Moisture content (%)	22.86	0.00
Density (g cm^{-3})	0.98	0.75
Viscosity (cP)	1.33	0.81
Specific Gravity	0.99	0.75
API Gravity	11.73	56.94
CV (kcal kg^{-1})	10,401.94	11,406.94

3.2.4 The Viscosity Value

Viscosity as a crucial parameter in determining the quality of fuel oil. As the Directorate General of Oil and Gas (2018) reported, viscosity values typically fall within the range of 0.5-4 cP. Viscosity directly impacts fuel atomization when injected into the combustion chamber, as noted by Sun et al. (2021). If the oil is sufficiently thick, it will be easy to pump, ignite, and flow. Conversely, poor atomization can lead to carbon deposits on the engine walls. As indicated by Saravanan et al. (2019) and (Sujesh et al., 2020), lower viscosity results in better fuel performance. The viscosity of SDs is reported as 1.33 cP, while LDPE has a viscosity of 0.81 cP in Table 2.

According to the research results in Table 1, the viscosity values of SDs and LDPE in the sample variation are 1:5 with temperatures of 475, 425, and 375°C, respectively, namely 0.41, 0.51, and 0.42 cP. They are close to the viscosity value of gasoline-type fuels. Temperature affects viscosity, indicating that a 500°C increase increases it more than 220-fold between 55 and -55°C. Based on the existing research, there is a relationship between density and viscosity (Hoang, 2018). Generally, lower viscosity corresponds to lower density values. High density and viscosity can adversely impact the atomization process during fuel combustion, leading to reduced engine performance and increased emissions of nitrogen oxides (NOx) (Hoang, 2018; Razzaq et al., 2020). The lowest viscosity value is 475°C. Reducing the amount of biomass lowers the viscosity value because biomass has a greater viscosity than water (Mishra and Mohanty, 2020). Water has a viscosity of 1 cP, while the viscosity of plastic pyrolysis oil is more petite than water. Referring to research Jahiding et al. (2021a,b); Razzaq et al. (2020), the viscosity value of pyrolysis fuel around 0.81 cP.

Table 3. GC-MS Analysis of Hydrocarbon-Oil Compounds (T = 475°C)

Compounds Name	Molecular Formula	m/z (g/mol)	Composition of SDs: LDPE (% Area)		
			5:1	3:3	1:5
Alkane					
Heksana	C_6H_{12}	84	1.57	1.18	0.98
Heptane	C_7H_{16}	100	2.12	1.02	0.96
Methylcyclohexane	C_7H_{14}	98	0.62		
Octane	C_8H_{18}	114	3.40		2.15
Ethylcyclohexane	C_8H_{16}	112	0.48		
Nonane	C_9H_{20}	128	4.18	1.27	0.65
Decane	$C_{10}H_{22}$	142	4.65	2.86	1.26
Dodecane	$C_{12}H_{26}$	170	11.25	3.11	1.95
Tridecane	$C_{13}H_{28}$	184	4.97		
Hexadecane	$C_{16}H_{34}$	226	3.75		
Pentadecane	$C_{15}H_{32}$	212	2.73		
Heptadecane	$C_{17}H_{36}$	240	3.26		
Nonadecane	$C_{19}H_{40}$	268	0.30		
Eicosane	$C_{20}H_{42}$	282	0.50		
Tetracosane	$C_{24}H_{50}$	338	0.78		
Tricosane	$C_{23}H_{48}$	324	0.33		
Pentatriacontane	$C_{35}H_{72}$	493	0.24		
Heptacosane	$C_{27}H_{56}$	380	0.40		
Alkene					
4-Methylcyclopentene	C_6H_{10}	82	0.29		
Alkene					
1-Heptene	C_7H_{14}	98	1.67	2.56	0.67
2-Methyl-1,5-hexadiene	C_7H_{12}	96	0.41		
1-Ethylcyclopentene	C_7H_{12}	96	0.57		
1-Methylcyclohexene	C_7H_{12}	96	0.91		
1-Octene	C_8H_{16}	112	2.51	2.99	1.22
(E)-4-Octene	C_8H_{16}	112	0.44		
(2,2-dimethylpropylidene)-Cyclopropane	C8H14	110	0.24		
1,2-Dimethylcyclohexene	C8H14	110	0.22		
(1-methylethenyl)-Cyclopentane	C8H14	110	0.31		
ethylidene-cyclohexane	C_8H_{14}	110	0.83		
4,5-Nonadiene	C_9H_{16}	124	0.40		
1-Nonene	C_9H_{18}	126	3.13	1.85	0.77
1-Decene	$C_{10}H_{20}$	140	3.76	0.87	1.09
1-Undecene	$C_{11}H_{22}$	154	4.08	2.01	1.77
1,11-Dodecadiene	$C_{12}H_{22}$	166	0.30		
1-Dodecene	$C_{12}H_{24}$	168	3.49		1.39
1-Tridecene	$C_{13}H_{26}$	182	2.35		
1-Pentadecene	$C_{15}H_{30}$	210	2.12	2.41	1.02
1-Hexadecene	$C_{16}H_{32}$	224	0.51	2.01	1.15
1-Heptadecene	$C_{17}H_{34}$	238	1.13	1.02	1.11
1-Nonadecene	$C_{19}H_{38}$	266	0.23	1.85	1.39
Squalene	$C_{30}H_{50}$	410	0.72		
Alcohol					
n-Undecanol	$C_{11}H_{24}O$	172	0.40		
2-Methyl-3-heptanol	$C_8H_{18}O$	130			0.63
Ketone					
2-Propanone	C_3H_6O	58		3.19	1.61

2-Hydroxy-3-methyl-2-cyclopenten-1-one	$C_6H_8O_2$	112		2.29	0.94
Cyclopentanone	C_5H_8O	84			1.49
Acetol	$C_3H_6O_2$	74		10.88	15.41
Ester					
Allyl acetate	$C_5H_8O_2$	100			7.19
Methyl acetate	$C_3H_6O_2$	74		1.12	2.60
Benzene and derivated					
Bis(2-ethylhexyl) phthalate	$C_{24}H_{38}O_4$	390	17.68		
Phenol and derivated					
Phenol	C_6H_6O	94		3.02	2.18
Acid					
Formic acid	CH_2O_2	46		1.59	
Acetic acid	$C_2H_4O_2$	60		18.55	32.88
Propionic acid	$C_3H_6O_2$	74		1.81	
Furan					
2-Furancarboxaldehyde	$C_5H_4O_2$	96		7.29	9.19

3.2.5 The Specific Gravity and API Gravity

The American Petroleum Institute (API) gravity is directly related to specific gravity and is correlated with the chemical composition and fuel quality. Many researchers have incorporated API gravity as part of the characterization of crude oil to differentiate between different types of petroleum (Guzmán-osorio et al., 2020; Nwadinigwe and Alumona, 2017). In Table 1 and 2, API gravity was obtained from 6.54 to 60 (Guzmán-osorio et al., 2020; Mousazadeh et al., 2021), with specific gravity ranging from 0.818 to 1.025 (Guzmán-osorio et al., 2020). Thus, a representative sample of each API classification level is obtained.

3.2.6 The Calorific value

The CV is the total combustion value for a unit volume of gas fuel or one unit weight of solid fuel or liquid in the normal state. The CV will release heat energy in the fuel during the oxidation process of the chemical elements. The CV of fuel oil ranges from 42.57-46.09 MJ kg^{-1} (Directorate General of Oil and Gas, 2018). Table 2 presents the CV of the pyrolysis products of SDs and LDPE, conforming to the quality standards for fuel oil, typically ranging from 10,160 to 11,000 kcal kg^{-1} or equivalent to 42.50 to 46.02 MJ kg^{-1}. The average CV derived from the pyrolysis of SDs and LDPE is 45.49 ± 3.45 MJ kg^{-1}. It is observed that compositions with a higher proportion of biomass tend to yield lower CV values compared to those with more plastic. This is because biomass contains significant oxygen, resulting in a lower CV (Huang and Lo, 2019; Liu et al., 2018). While pyrolysis oil with more plastic produces a high CV, this is due to the presence of more heavy liquid. This heavy liquid contains hydrocarbon chains above 6, so the amount of plastic and the reduction in the heavy liquid biomass increases, and the amount of oxygen decreases and increases the CV (Dewangan et al., 2016; Huang and Lo, 2019). The increase in the CV of the oil, accompanied by the rise in pyrolysis temperature, is attributed to the enhanced evaporation of volatile matter and a reduction in water content, leading to carbon accumulation. At higher temperatures, the decomposition process occurs more efficiently, contributing to the higher CV of the oil (Dewangan et al., 2016; Ryu et al., 2020).

The CV of LDPE without adding biomass was 11,406.94 kcal kg^{-1} at 475°C. For, biomass was 10,401.94 kcal kg^{-1} in Table 2. The increase in CV is insignificant, so the reaction temperature is not too influential. Figure 3 shows the pyrolysis time of SDs and LDPE, that the longer the pyrolysis operation time, the higher of CV (Dewangan et al., 2016; Ryu et al., 2020). Oxygen can impact the CV of hydrocarbon oils, resulting in lower values. Therefore, the direct blending of SDs and LDPE increases the CV of the pyrolysis oil. Research conducted by Ryu et al. (2020), concluded that the energy content of pyrolysis oil is relatively higher in co-pyrolysis cases than single pyrolysis. Moreover, the biomass pyrolysis with plastic also contributes positively to the yield of pyrolysis products (Dyer et al., 2021; Mishra and Mohanty, 2020; Ryu et al., 2020; Uzoejinwa et al., 2018).

3.3 The GC-MS Analysis

Hydrocarbon is a compound consisting of carbon (C) and hydrogen (H). In this research, hydrocarbon oil is classified into two, namely aliphatic and aromatic. GC-MS analysis of hydrocarbon oil pyrolysis of SDs and LDPE at ratios of 5:1, 3:3, and 1:5 (T, 475°C), in Table 3 and Figure 4. However, the composition of the chemical compounds is different, where the ratio of 1:5 containing alkanes and alkenes is dominant due to the influence of LDPE. Meanwhile, the 3:3 and 5:1 ratio have hydrocarbon compounds, but many compounds include oxygen because of biomass. Biomass contains a lot of oxygen, esters, ketones, aldehydes, phenols, acids, and furans resulting from the decomposition of biomass in the pyrolysis process.

According to the research conducted by Ke et al. (2022), the findings indicate that in the pyrolysis of biomass, aromatic hydrocarbon compounds are more prevalent than aliphatic ones. The aromatic compounds obtained were phenolic (35.17%) and furans (27.62%). At the same time, aliphatic compounds include acetic acid, hexane, and other ketone compounds. Research SD and LDPE (1:5), rich in aliphatic and aromatic compounds, like alkanes (45.53%), alkenes (30.62%), alcohols (0.4%), and benzenes (17.68%). Besides that, lesser amounts of benzene were also observed. However, the highest alkanes and alkanes were obtained from pure LDPE oil, with minimal amounts of acid, alcohols, ketones, and benzene in biomass. Phenolic compounds were not found in pure LDPE pyrolysis oil. Therefore, the biomass and plastic oil mixture showed a reduced phenol content.

Alternatively, the pyrolysis oil may contain additional hydrogen and carbon compounds. The increase in the amount of LDPE causes the content of aliphatic compounds to increase. The hydrocarbon oil components contained in fuel oil will affect its quality, such as cetane number, pour point, dew point, and engine performance. Therefore, fuel oil with a high fraction indicates low quality. On the other hand, if the compounds' components are close to the range of combinations, the fuel has better quality (Ivanova et al., 2014).

4. CONCLUSIONS

The recent study on the co-pyrolysis of biomass and plastics, specifically SDs and LDPE, aims to produce high-quality pyrolysis oil while reducing oxygenated compounds and enhancing the presence of aliphatic and aromatic hydrocarbons. Including LDPE in the process generates a higher proportion of these compounds in the resulting oil, which consists of more alkanes and alkenes. The hydrogen from LDPE promotes the growth of these compounds, stabilizing free radicals. Reducing phenolic compounds and acids can further stabilize the oil as a potential fuel. A 1:1 ratio of biomass to LDPE at 400-500°C is recommended for optimal results. More research is needed to understand the mixture's reactions for better waste treatment and bioenergy use. Additional steps, such as distillation, are necessary to refine the fuel product.

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