Co-pyrolysis of oil palm empty fruit bunches (EFB) biochar with high-density polyethylene (HDPE) for liquid fuel production

Sunarno Sunarno[1], *Padil* Padil[1], *Anisa* Mutamima[1*], *Nurfatihayati* Nurfatihayati[1], *Johan* Sitanggang[1], *Viccy Wando* Aritonang[1] and *Panca Setia* Utama[1]

[1]Department of Chemical Engineering, Faculty of Engineering, Universitas Riau, Pekanbaru 28293, Indonesia

Abstract. The increasing number of vehicles has led to elevated demand for petroleum-based fuels, which are non-renewable energy sources. Consequently, the development of alternative energy sources has become crucial. In this study, the co-pyrolysis process was conducted to produce bio-oil as an alternative energy source. Co-pyrolysis is a thermal degradation process performed in the absence of oxygen that involves biomass and other materials. This study investigates the use of empty fruit bunches (EFB) biochar and high-density polyethylene (HDPE) with a high hydrogen-to-carbon effective ratio $(H/C)_{eff}$ as raw materials for co-pyrolysis. This study aims to examine the effects of torrefaction pretreatment on EFB, the impact of varying the EFB biochar to HDPE ratio (100:0, 75:25, and 55:45), and the influence of co-pyrolysis temperature (400, 450, and 500 ^{o}C) on the yield and characteristics of produced bio-oil. Experiment results indicated that torrefaction pretreatment reduced ash content, while the addition of HDPE and temperature increase improved bio-oil yield. The best operating condition with feed EFB biochar to HDPE ratio of 55:45 at a temperature of 500°C which produced the highest yield of 28.2% and optimum characteristic bio-oil with density of 0.88 g/mL, viscosity of 1.28 cP, pH of 3.7, and a calorific value of 47.34 MJ/kg.

1. Introduction

The rapid increase in global vehicle numbers has increased greater dependence on non-renewable petroleum-based fuels. According to the International Energy Agency (IEA) in 2021, the transportation sector consumes approximately 64% of the world's oil, underscoring the critical need for renewable energy alternatives [1]. Bio-oil has emerged as a promising substitute for traditional petroleum fuels due to its potential to reduce carbon emissions, biodegrade, and effectively utilize biomass waste streams [2,3].

Bio-oil can be produced through thermochemical processes such as pyrolysis. During pyrolysis, biomass is converted into liquid, solid, and gaseous products under anaerobic or anoxic conditions [4]. Co-pyrolysis is an advanced variation of pyrolysis that involves the

*Corresponding author : anisamutamima@eng.unri.ac.id

simultaneous thermal decomposition of biomass and a secondary feedstock with a high hydrogen-to-carbon effective ratio $(H/C)_{eff}$. Some studies have added plastic into the co-pyrolysis process [5-7]. This process enhances the quality and yield of bio-oil by increasing the hydrogen content and reducing the oxygen content of produced bio-oil [8].

Empty fruit bunches (EFB) are biomass residues from palm oil mills. Indonesia is one of the largest palm oil producers in the world that generates a significant amount of EFB waste. Riau province produces approximately 6,663,956 tons of EFB annually [9]. If not managed properly, this biomass residue can cause environmental problems including air pollution, also land and water contamination [10,11]. According to Haryanto et al. [12], EFB has high moisture and oxygen content which can affect the pyrolysis process and the quality of the produced bio-oil. To overcome these issues, torrefaction with a mild thermal pretreatment process conducted at 200–300 °C can be employed. Torrefaction can reduce moisture content and increase the energy density of the biomass by partially decomposing its hemicellulose content. This leads to improvement of the physicochemical properties of EFB that make it more suitable for pyrolysis [13]. This process results in a more stable, hydrophobic, and energy-dense biomass, which can improve the efficiency and yield of the co-pyrolysis process.

High-density polyethylene (HDPE) is plastic with a high H/C ratio that is ideal to be employed as co-feedstock for biomass co-pyrolysis [14,15]. The addition of HDPE into the co-pyrolysis process offers dual benefits: improving bio-oil yield and quality, and addressing the environmental issue of plastic waste. HDPE is hard to degrade and contributes significantly to plastic pollution [16]. This approach provides a sustainable method to recycle plastic waste while generating a valuable energy resource.

Previous research has significantly enhanced bio-oil yield and properties from biomass and plastic waste. Sembiring et al. [17] studied the fast pyrolysis of EFB at varying temperatures and found that 500 °C was optimal for bio-oil yield, but resulted in bio-oil with high moisture and oxygen content. Rachmawati et al. [18] investigated the pyrolysis of various plastics and garden waste, identifying HDPE as the feedstock that produced the highest wax yield. Sunarno et al. [19] examined the co-pyrolysis of EFB and LDPE, achieving high bio-oil yield but raising concerns over the high plastic ratio used. In another study, Sunarno et al. [20] produced bio-oil from EFB and polypropylene (PP), achieving an optimum bio-oil yield of 41.6% at an EFB and PP ratio of 80:20 and a temperature of 450 °C. The bio-oil had a high hydrocarbon content (alkanes and alkenes) of 44.27% and a calorific value of 43.2 MJ/kg. However, the bio-oil still contained a significant amount of acid, which could result in corrosive properties during application.

Zhang et al. [21] successfully increased bio-oil yield through torrefaction pretreatment, although the resulting bio-oil still contained a high acid content. Therefore, this study investigates the effects of torrefaction pretreatment on the pyrolysis process to improve bio-oil yield, and the implementation of co-pyrolysis with the addition of HDPE. By varying feedstock ratios and co-pyrolysis temperatures, the research aims to enhance both yield and bio-oil characteristics. This study seeks to optimize the co-pyrolysis process, offering a more efficient method for producing high-quality bio-oil while promoting sustainable waste management.

2. Materials and Method

2.1 Materials

The materials used in this study were empty fruit bunches (EFB), HDPE, distilled water, and NaOH.

2.2 Pretreatment of Empty Fruit Bunches (EFB)

Pretreatment of EFB consists of several steps including torrefaction water production, soaking EFB with torrefaction water, and EFB torrefaction. Firstly, EFB were shredded into fiber-like form, followed by soaking in a NaOH solution with a ratio of 1:2 for 48 hours. Then the EFB was filtered, washed with distilled water, and dried in the oven at 110 °C until it reached a constant weight. The torrefaction pretreatment process was conducted for 60 minutes at 210 °C. From the torrefaction process, a liquid product (torrefaction water) was produced and used for EFB pretreatment in the next stage. Then the shredded EFB was soaked in torrefaction water, rinsed with distilled water, and dried at 110 °C until it reached a constant weight. Lastly, the dried EFB was torrefied for 60 minutes at 210 °C to produce EFB biochar used in co-pyrolysis.

2.3 Pretreatment of HDPE Plastic

HDPE plastic waste was collected, cleaned, sun-dried, and then cut into smaller pieces (with an approximate size of 2 × 2 cm).

2.4 Co-Pyrolysis

Referring to the study by Sunarno et al. [20] with modifications to the feedstock ratios, the production of liquid fuel was carried out by mixing EFB biochar and HDPE plastic waste in weight ratios of 100:0, 75:25, and 55:45. The prepared feedstocks were then placed in a reactor and heated under a nitrogen flow of 200 mL/min for 45 minutes at varying temperatures of 400, 450, and 500 °C. The co-pyrolysis process produced liquid, char, and gaseous compounds. The vapors formed during co-pyrolysis were condensed using a condenser to produce a liquid product (bio-oil). Physical properties (density, viscosity, pH, and calorific value) of the resulting bio-oil were analyzed. The experimental setup for the co-pyrolysis of EFB biochar with HDPE can be seen in Figure 1.

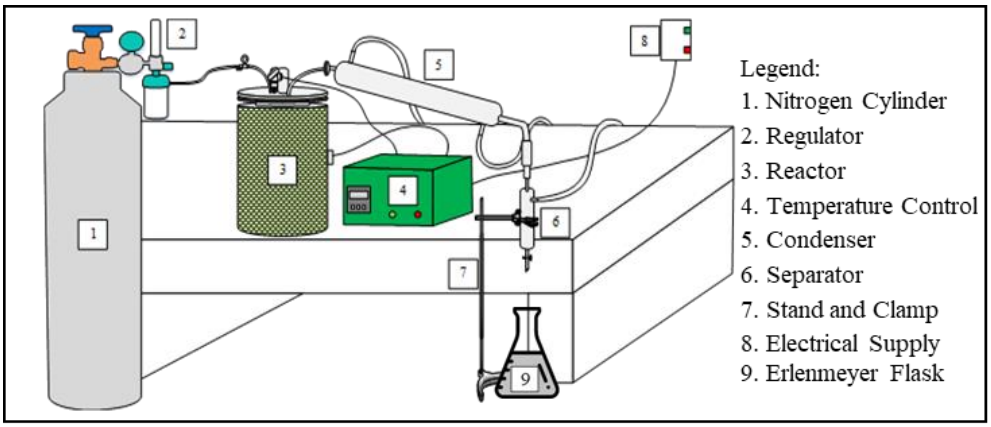

Fig. 1. Co-Pyrolysis Experimental Setup

2.5 Liquid Products Characterization

The produced fuel was characterized by its physical properties, including pH, density, viscosity, and calorific value.

2.5.1. pH Test (SNI 06-6989.11-2004)

pH measures the concentration of hydrogen ions that indicate acid content in a solution. The pH measurement was conducted using a pH meter.

2.5.2. Density Test (ASTM D 1298)

Density indicates the compactness or determined as ratio of the weight to a given volume of a material. Density was calculated using the Equation 1.

$$\rho = \frac{m}{v} \quad (1)$$

The variables are defined as follows: ρ represents the density in kilograms per cubic meter (kg/m³), m denotes the mass in kilograms (kg), and v refers to the volume in cubic meters (m³).

2.5.3. Viscosity Test (ASTM D 445)

Viscosity refers to the ability of a substance to flow in a particular medium, also known as thickness. The viscosity of the liquid was measured using an Otswald viscometer and was calculated using Equation 2.

$$\eta_x = \frac{t_x \rho_x \eta_w}{t_w \rho_w} \quad (2)$$

The variables are defined as follows: η_w represents water viscosity in centipoise (cP), η_x is the bio-oil viscosity in centipoise (cP), t_w denotes the flow time of water in seconds (s), t_x refers to the flow time of the tested liquid in seconds (s), ρ_w is the water density (kg/m³), and ρ_x represents the bio-oil density (kg/m³).

2.5.4. Calorific Value (SNI 01-6235-2000)

The calorific value of a fuel indicates the amount of energy produced during the combustion process per unit mass of the fuel. The calorific value was measured using a bomb calorimeter and calculated using Equation 3.

$$\text{Hg (cal/g)} = \frac{\Delta t \times w}{m} \quad (3)$$

The variables are defined as follows: Hg represents the calories per gram (cal/g), Δt is the temperature increase in degrees Celsius (°C), w denotes the calorimeter's heat capacity, which is 2565.446 cal/°C based on calibration, and m refers to the sample mass in grams (g).

3. Results and discussions

3.1 Influence of EFB Pretreatment on Bio-oil Yield and Physical Properties

Pretreatment was an initial treatment performed on EFB biomass before the pyrolysis process. In this study, the pretreatment involved reducing the size of EFB, drying, and soaking it in torrefaction liquid. Reducing the size of EFB increased contact between the raw material mixture and the reactor walls, which enhanced the heat transfer rate during the co-pyrolysis process [19]. The analysis of the EFB composition before and after soaking in torrefaction liquid is shown in Table 1.

The analysis of EFB composition pre- and post-soaking in torrefaction liquid showed significant changes. EFB soaking in torrefaction liquid increased lignin from 25% to 29% and cellulose from 27.50% to 40.50%, while hot-water-soluble compounds and hemicellulose decreased from 23.50% to 13.00% and 15% to 12.50%, respectively. Ash content also decreased from 9% to 5%. These changes indicated the effective reduction of non-structural components like hemicellulose and hot-water-soluble compounds, enriching structural components such as cellulose and lignin. Increased lignin content likely resulted from partial degradation and re-polymerization during soaking, consistent with findings by Kristiani et al. [22]. Removal of hemicellulose and hot-water-soluble compounds facilitated cellulose thermal decomposition, enhancing bio-oil yield in pyrolysis.

Table 1. EFB composition before and after soaking in torrefaction liquid

Composition	**Before Soaking (%)**	**After Soaking in Torrefaction Liquid (%)**
Hot-Water-Soluble Compounds	23.50	13.00
Hemicellulose	15	12.50
Cellulose	27.50	40.50
Lignin	25	29
Ash	9	5

Reduced ash content further supported pretreatment efficiency, minimizing mineral catalytic effects that could lead to undesired reactions during pyrolysis. This aligned with Chen et al. [23], who found torrefaction water effective in removing alkali and alkaline earth metals, improving biomass quality. In contrast, Sunarno et al. [20] observed decreased lignin content post-soaking in their study on catalytic co-pyrolysis of EFB and plastic waste. Variations in soaking medium and conditions highlighted the need to optimize pretreatment parameters for specific biomass types and desired outcomes.

Table 2. Yield masses from EFB with and without torrefaction pretreatment

EFB Pretreatment	**Sample Mass (g)**	**Mass Yield (%)**		
		Bio-oil	**Char**	**Gas**
Without torrefaction	30	44	52	4
With torrefaction	30	8	68	24

Table 2 shows varying yields of bio-oil, char, and gas from EFB samples treated with and without torrefaction pretreatment. The yield of bio-oil was notably higher without torrefaction at 44% compared to 8% after torrefaction. Conversely, torrefaction pretreatment significantly increased char and gas yield to 68% and 24% respectively. These results were consistent with Zhang et al. [21], who investigated catalytic bio-oil upgrading from torrefied rice husk using Fe-modified zeolite ZSM-5. They reported a 39% bio-oil yield without torrefaction and 32% with torrefaction using water as the pretreatment medium. The decrease in bio-oil yield due to torrefaction occurred as a result of decomposition during the heating process. This process removed moisture and volatile components. Hemicellulose and some

cellulose, which are important for bio-oil formation, were particularly affected [24]. Torrefaction altered biomass composition by preferentially decomposing hemicellulose and some cellulose while increasing residual lignin content, as noted by Meng et al. [25] and Ren et al. [24].

The increase in char and non-condensable gas from torrefaction matched the results of Srinivasan et al. [26], who studied the catalytic pyrolysis of torrefied biomass for hydrocarbon production. Their research showed that the fibrous structure of biomass was lost due to the breakdown of hemicellulose and cellulose during torrefaction. The findings pointed out the balance between bio-oil yield and the production of char and gas, while highlighting the need to optimize torrefaction condition to improve bio-oil production efficiency in pyrolysis.

The physical properties analysis of bio-oil in this study was summarized in Table 3. The analysis includes density, viscosity, and pH of bio-oil. Analysis results show that bio-oil density slightly decreased after the pretreatment process, with 1.14 g/mL for EFB without torrefaction and 1.11 g/mL for EFB after torrefaction. This result aligns with that of Chang [27]. Fresh biomass-derived bio-oil contained denser organic compounds like lignin, cellulose, and hemicellulose, whereas bio-oil from biochar was richer in aromatic and lighter carbonated compounds.

Table 3. Characterization results of bio-oil physical properties

Pretreatment	Density (g/mL)	Viscosity (cP)	pH
Without torrefaction	1.14	2.07	3.1
With torrefaction	1.11	2.92	3.7

Additionally, pretreatment of EFB increased bio-oil viscosity, with values of 2.92 cP and 2.07 cP for torrefaction and non-torrefaction pretreatments, respectively. These viscosity levels were comparable to standard diesel oil viscosity (2-4.5 cP) [28]. The difference in viscosity was attributed to water content in non-torrefaction-derived bio-oil, which reduced viscosity due to water's lower viscosity. Lower viscosity bio-oil benefits include reduced flow resistance, enhancing its suitability as a fuel. The pH values of 3.7 for torrefaction-treated EFB and 3.1 for non-torrefaction-treated EFB indicated improved bio-oil quality post-pretreatment, aligning with findings by Khor et al. [29] for EFB derived bio-oil. Fresh biomass-derived bio-oil typically contains more organic acids, while bio-oil from biochar tends to have more basic compounds like carbonate and potassium [30].

3.2 The Impact of EFB Biochar to HDPE Ratio on Yield

The co-pyrolysis of EFB biochar and HDPE at varying ratios (100:0, 75:25, and 55:45) at 500 °C produced bio-oil, char, and gas, as shown in Figure 2. Figure 2 illustrated that the addition of plastic increased bio-oil and gas yields compared to the 100:0 ratio, while decreasing char yield. This phenomenon was attributed to the higher decomposition rate of plastic into gases and liquids as opposed to solid char. The addition of plastic improved hydrogen donation during co-pyrolysis. This helped break down cellulose and hemicellulose, leading to an increase in the production of condensed gases. At a ratio of 100:0, the bio-oil yield was lowest due to the limited availability of hydrogen.

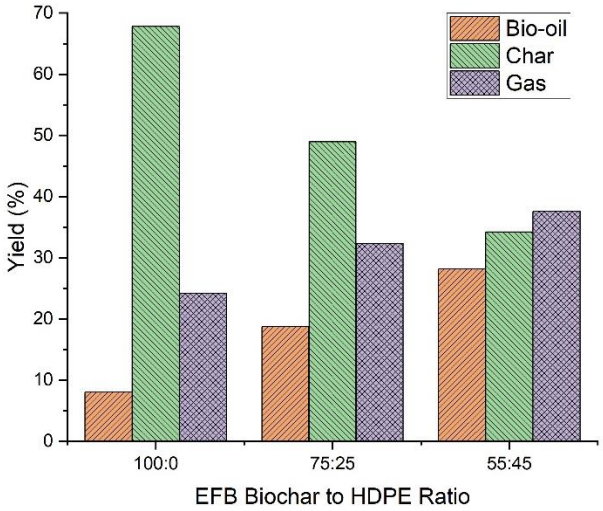

Fig. 2. Influence of EFB Biochar to HDPE Ratio on Product Composition

Specifically, the bio-oil yield increased significantly from 8.03% at a ratio of 100:0 to 18.73% at a ratio of 75:25, and further to 28.2% at a ratio of 55:45. This trend indicates that the presence of plastic promotes the production of bio-oil. Conversely, the char yield decreased from 67.8% at a ratio of 100:0 to 48.93% at a ratio of 75:25, and further to 34.2% at a ratio of 55:45, highlighting that plastic addition reduces the solid residue. Similarly, the gas yield increased from 24.17% at a ratio of 100:0 to 32.33% at a ratio of 75:25, and further to 37.6% at a ratio of 55:45, demonstrating that higher plastic content leads to more gaseous products.

According to Dyer et al. [31] and Sunarno et al. [32], the higher volatile content of plastics facilitates hydrogen donation during co-pyrolysis. Sunarno et al. [20] found that adding polypropylene (PP) reduced char yield but increased gas yield due to the decreased lignin content. These findings align with those of Aboelela et al. [33], who noted that a higher volatile content in biomass correlates with increased bio-oil fractions in pyrolysis, as the thermally degraded biomass forms pyrolysis vapors that condense.

3.3 The Impact of Co-pyrolysis Temperature on Co-Pyrolysis Yield

This study evaluated the variation of co-pyrolysis temperature (400, 450, and 500 °C) on bio-oil yield. As illustrated in Figure 3, the results showed that raising the co-pyrolysis temperature resulted in higher bio-oil and gas yields, while reducing the char yield. At temperatures of 400, 450, and 500 °C, the bio-oil yields were 26.1%, 27.17%, and 28.2%, respectively.

At higher temperatures, more heat energy is supplied to the material, leading to the breakdown of components like hemicellulose, cellulose, and lignin. This additional thermal energy promotes gas formation, and cellulose decomposes into condensable gases, resulting in increased bio-oil and gas production [21]. The ideal co-pyrolysis temperature was identified as 500 °C. At temperatures below 400 °C, the material undergoes dehydration and forms char, whereas temperatures above 500 °C result in higher gas yields due to the secondary cracking of pyrolysis vapors and further char decomposition.

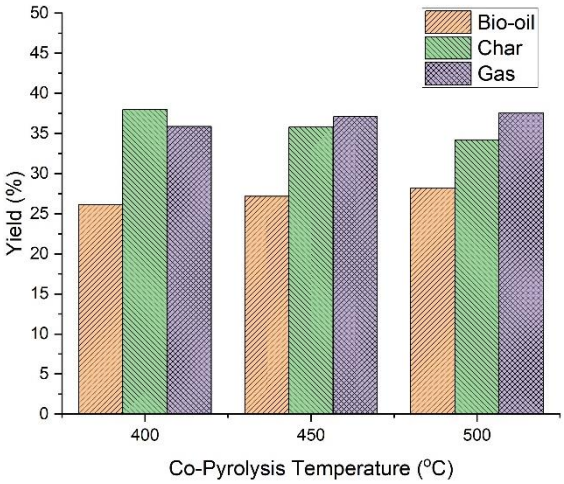

Fig. 3. Impact of Co-Pyrolysis Temperature on Co-Pyrolysis Yield

These results were consistent with the bio-oil yield from EFB and waste tires, which increases between 400 °C and 500 °C [34]. However, at temperatures exceeding 550 °C, bio-oil yield declines due to the thermal cracking of volatile compounds, converting pyrolysis vapors into low molecular weight organics or non-condensable gases [35-37].

3.4 The Impact of Co-pyrolysis Feed Ratio and Temperature on Bio-Oil Physical Properties

The bio-oil produced from the co-pyrolysis of EFB biochar and HDPE was analyzed. Table 4 shows how the density, viscosity, and pH of the bio-oil changed with different ratios of EFB biochar and co-pyrolysis temperatures. At a co-pyrolysis temperature of 550 °C, the data revealed that as the HDPE content increased, the bio-oil density dropped from 1.11 g/mL to 0.88 g/mL. Although the bio-oil density is higher than that of diesel, it is lower than bio-oil from other feedstock combinations. For example, the bio-oil produced in this study with a feedstock ratio of 55:45 had a density of 0.88 g/mL, which is slightly lower than the 0.891 g/mL density from co-pyrolysis of EFB and polypropylene (PP) [20], and also lower than the bio-oil from co-pyrolysis of newspaper and HDPE, which ranged from 1.14 to 1.2 g/mL [23]. In line with density, viscosity decreased from 2.92 cP to 1.28 cP as HDPE content increased. This indicates that HDPE plays a role in viscosity reductions. This study's findings are aligned with those of Chen et al [23], who showed a decline in bio-oil viscosity as the HDPE ratio in the feedstock increased. According to Wang et al. [38], the beneficial synergistic impact of plastic and biomass co-pyrolysis reduces water and oxygen content, lowering the density, total acid number, and viscosity. Meanwhile, pH values ranged from 3.7 to 4.2, indicating HDPE's effect on bio-oil acidity levels.

Further analysis in Table 4 aimed to explore additional fuel-related properties across various co-pyrolysis temperatures for the 55:45 feed ratio. The data in Table 4 indicated that increasing pyrolysis temperature decreased bio-oil density from 0.89 g/mL to 0.88 g/mL, accompanied by reductions in viscosity (from 1.33 cP to 1.28 cP) and pH (from 3.8 to 3.7). These changes suggested enhanced thermal decomposition at higher temperatures, resulting in bio-oil containing lighter hydrocarbons and reduced acidic components.

Table 4. Characterization results of bio-oil physical properties under the influence of feedstock ratio and co-pyrolysis temperature

EFB biochar to HDPE Ratio	Co-pyrolysis Temperature (°C)	Density (g/mL)	Viscosity (cP)	pH	Calorific Value (MJ/kg)
100:0	500	1.11	2.92	3.7	-
75:25	500	0.98	2.09	4.2	-
55:45	500	0.88	1.28	3.7	47.34
55:45	450	0.89	1.32	3.8	Undetected
55:45	400	0.89	1.33	3.8	Undetected

At high co-pyrolysis temperatures, the thermal breakdown of biomass and plastic created smaller, lighter compounds like gases and light aromatic hydrocarbons. These compounds have lower densities than the larger, heavier compounds produced at lower co-pyrolysis temperatures [39]. The lowest density value from this study was still higher than the density range of diesel oil, which is 0.815-0.870 g/mL, and the density range of RON 90 gasoline, which is 0.715-0.77 g/mL.

The calorific value test was conducted at an EFB biochar to HDPE ratio of 55:45 at all co-pyrolysis temperatures (400, 450, and 500 °C). The calorific values for 400 °C and 450 °C were undetected, while only the temperature of 500 °C had a calorific value of 47.34 MJ/kg. At 500 °C, the calorific value exceeded that of diesel oil, which ranges between 42-46 MJ/kg, and approached that of RON 90 gasoline, which is 47.79 MJ/kg. The undetected calorific values at 400 °C and 450 °C may have been due to contamination or impurities such as minerals and water, preventing complete combustion in the bomb calorimeter.

These findings highlight a challenge in alternative fuel production to produce high quality bio-oil at lower temperatures. High calorific value bio-oil produced at 500 °C suggests that higher co-pyrolysis temperatures improve the breakdown of biomass and plastic. However, the undetected values at lower temperatures emphasize the need for process optimization to ensure reliable fuel production across varying temperatures.

4. Conclusion

Torrefaction pretreatment of EFB reduced the bio-oil yield from 44% to 8%. The best physical properties of bio-oil from torrefaction pretreatment included a density of 1.11 g/mL, viscosity of 2.92 cP, and pH of 3.7. Increasing the ratio of high-density polyethylene (HDPE) to EFB biochar improved the bio-oil yield to 28.2%. At an EFB biochar to HDPE ratio of 55:45, the bio-oil exhibited a density of 0.88 g/mL, viscosity of 1.28 cP, pH of 3.7, and a high heating value (HHV) of 47.34 MJ/kg. Furthermore, raising the co-pyrolysis temperature enhanced the bio-oil yield; at 500 °C with an EFB ratio of 55:45, the bio-oil yield reached 28.2%, with a density of 0.88 g/mL, pH of 3.7, and HHV of 47.34 MJ/kg. These results have significant implications for energy sustainability and plastic waste management. Integrating HDPE plastic waste into the co-pyrolysis process enhances bio-oil yield and quality while addressing plastic waste issues.

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