



Mass Balance of Hydrocarbons Fuel from Waste Cooking Oil and Plastic by Co-Pyrolysis

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Received 16 May 2025, Revised 24 June 2025, Accepted 04 July 2025

doi: [10.22487/j24775185.2025.v14.i3.pp132-137](https://doi.org/10.22487/j24775185.2025.v14.i3.pp132-137)

Abstract

Indonesia has committed to climate change mitigation through a sustainable energy transition. Currently, the share of renewable energy in Indonesia's energy mix only reaches 12%. One type of renewable energy that the Indonesian government is focusing on is biofuel to reduce consumption and dependence on fossil fuels. Bio-oil produced through pyrolysis has the potential to be used as a biofuel. The chosen pyrolysis process is Co-Pyrolysis, which involves two or more feedstocks. In this study, the process was carried out using waste cooking oil (WCO) and HDPE waste as feedstocks. The process was conducted at 450°C for 30 minutes, with varying masses of both feedstocks. The data obtained were analyzed using mass balance and yield analysis to determine the optimal conditions for producing bio-oil as a hydrocarbon fuel. The optimal condition was found at a mass ratio of HDPE: WCO = 1:4, with a bio-oil yield of 78%. Under these optimal conditions, the conversion of HDPE plastic into heptadecane was 29.39%, and the conversion of oleic acid in waste cooking oil into heptadecane was 38.8%. This also serves as an initial step toward developing a pilot plant-scale process using HDPE plastic waste and waste cooking oil.

Keywords: Bio oil; copyrolysis; HDPE; Not biofuel; WCO

Introduction

The Government of Indonesia is committed to climate change mitigation through Net-Zero Emissions (NZE) in 2060, through an energy transition to sustainable, clean energy. In the energy supply sector, emissions are expected to decrease by 358 million tons of CO₂ (Adiwardhana, 2023). Currently, Indonesia's energy supply is still dominated by fossil fuels, namely coal and petroleum. According to the Republic of Indonesia (2023), the primary energy mix, including coal and petroleum, accounts for 74%, while new renewable energy accounts for only 12%. To support the government's commitment, research and development of new renewable energy is required.

Biofuel is one of the programs implemented to achieve NZE and sustainable clean energy utilization. Biofuel is any fuel derived from vegetable oils, such as biodiesel, bioethanol, and bio oil (Helbawanti et al., 2023). Vegetable oils can be derived from fruits, grains, vegetables, nuts, and plant roots. Indonesia is a country with a wealth of natural resources, including abundant vegetable oil-producing plants (Ramdhoani & Javandira, 2023).

Bio-oil is a liquid product produced by condensing the pyrolysis process and can be used as

fuel. The raw material for making bio-oil is biomass widely available in the surrounding environment, such as agricultural waste, forestry waste, and urban waste (e.g., plastics, bottles, and packaging) (Ahmed & Kishore, 2023; Khan et al., 2018; Sudalyandi & Jeyakumar, 2022). Pyrolysis can be carried out using slow, fast, or flash methods, with fast pyrolysis producing better bio-oil than the other methods. However, fast pyrolysis has been modified to improve the quality of bio-oil produced, such as catalytic pyrolysis and co-pyrolysis.

Plastic is one of the products derived from petroleum derivatives. Plastic contains high energy like other fuels such as gasoline, diesel, and kerosene (Wahyudi et al., 2018). HDPE (High-Density Polyethylene) waste is one type of plastic that is inherently difficult to decompose and takes a very long time to break down. Plastic waste is a material that cannot be naturally decomposed (non-biodegradable), so after use it becomes waste that soil microbes cannot decompose and pollutes the environment (Kulkarni et al., 2022; Rahmayani & Aminah, 2021).

Waste cooking oil (WCO) is cooking oil that has been used for frying, cooking, and other processing in restaurants, food processing industries, and households. Waste cooking oil

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(WCO) is composed of glycerol, which is linked by ester bonds to carboxylic acids. Waste cooking oil has a calorific value sufficient for direct combustion, enabling its use as a raw material in the manufacture of bio-oil (Nuraini et al., 2022).

In previous studies, pyrolysis of waste cooking oil as a feedstock has yielded relatively high bio-oil yields of 60–70%. The fuel has a viscosity of 9.53 mm²/s, an acid number of 124.56 mg KOH/g, and a flash point of 21 °C. In contrast, pyrolysis of waste cooking oil yields a high acid content of 48% (Kraiem et al., 2016). Meanwhile, HDPE pyrolysis tends to produce more char and gas than bio-oil, with a bio-oil yield of 40–50% (Dutta et al., 2022). Other studies show that HDPE pyrolysis fuel has a viscosity of 1.18 mm²/s and a density of 777 kg/m³ (Ardianti et al., 2019). These values do not comply with fuel specifications, specifically SNI Biodiesel 2015, which requires a viscosity of 2.3 – 6.0 mm²/s, a density of 850 – 890 kg/m³, a maximum acid number of 0.4 mg KOH/g, and a minimum flash point of 130 °C. Therefore, waste cooking oil and HDPE plastic are treated in a single system, which is expected to overcome the disadvantages of each as fuel.

In support of Indonesia's commitment to climate change mitigation through a renewable energy mix and the processing of HDPE plastic and cooking oil waste, the co-pyrolysis process emerges as a promising method. Although both materials have been extensively studied individually, the combination of these two feedstocks in a single system to produce hydrocarbon fuel has not yet been thoroughly explored. This study uses mass balance analysis and product yield calculations to evaluate the efficiency of converting raw materials into hydrocarbon fuel. It is expected that this research can serve as a foundation for designing a production process on a pilot plant scale or for scale-up.

Methods

Instruments

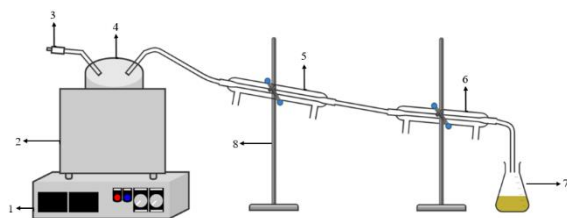


Figure 1. Set of Pyrolysis Instruments

The instruments employed are a set of pyrolysis instruments as illustrated in Figure 1. This set of instruments includes a Temperature Controller (1), Furnace (2), Nitrogen Gas Line (3), Pyrolysis Reactor (4), Condenser 1 and 2 (5,6), Erlenmeyer Flask (7), and Clamp Stand (8)

Materials

The Waste cooking oil (WCO) utilized in this study was collected from vendors in the Rungkut area of Surabaya. The HDPE plastic is a Titanvene™ HD5401GA product from PT. Lotte Chemical Titan Nusantara

Co-pyrolysis process

The masses of waste cooking oil and HDPE plastic were varied at 0 g, 25 g, 50 g, 75 g, and 100 g. Before being fed into the pyrolysis reactor, the two feedstocks were weighed according to the variables and then mixed. The co-pyrolysis process was carried out for 30 minutes at 450 °C. Subsequently, the gas produced in the pyrolysis reactor was condensed twice in succession using condensers. The condensed gas, which turned into a liquid phase, is referred to as bio-oil or hydrocarbon fuel and was then collected. The hydrocarbon fuel was analyzed through mass balance and yield calculations to determine the conversion of feedstocks into hydrocarbon fuel (product).

Mass balance

The mass balance analysis was conducted to evaluate the efficiency of the co-pyrolysis process. The analysis was performed by considering the balance between the feedstock and product masses. According to the book by Himmelblau & Riggs (2022), it can be systematically expressed using equations (1) and (2).

$$I + G - O + C = A$$

Keterangan:

- I = Input
- G = Generation
- O = Output
- C = Consumption
- A = Accumulation

(1)

$$I = O$$

(2)

Yield

Yield analysis was carried out to determine the percentage of feedstocks converted into the desired product. The calculation of the yield percentage analysis can be a systematic formula by (Doran, 1995) As shown in equation (3).

$$\text{Yield} = \frac{\text{mass product (gram)}}{\text{mass total (gram)}} \times 100\%$$

(3)

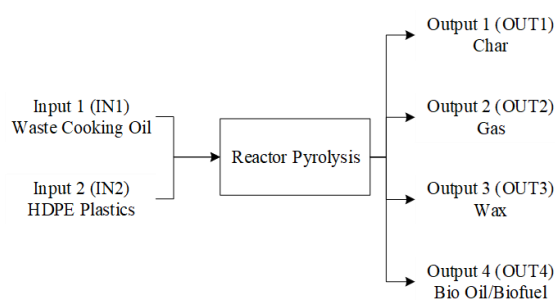


Figure 2. Co-pyrolysis process

Results and Discussion

Mass Balance Analysis

Mass balance analysis was performed based on the process mass flow diagram shown in **Figure 2**, which illustrates the flow of raw materials, conversion processes, and product outputs. Detailed mass balance calculations are presented in **Tables 1-4**. **Tables 1** and **2** present calculations to evaluate the effect of adding waste cooking oil (WCO) on product distribution. The addition of WCO, rich in fatty acids such as oleic acid, tends to increase the liquid fraction (bio-oil) and the content of long-chain hydrocarbons, such as heptadecane, while reducing the solid residue. Meanwhile, **Tables 3** and **4** present mass-balance calculations focused on the effect of adding HDPE on product outcomes.

Table 1. Mass balance of HDPE: WCO=1:2

Component	Input		Output			
	IN1 (g)	IN2 (g)	OUT1 (g)	OUT2 (g)	OUT3 (g)	OUT4 (g)
HDPE	-	25	-	-	-	-
WCO	50	-	-	-	-	-
Char	-	-	8.4	-	-	-
Bio Oil	-	-	-	-	-	57.5
Wax	-	-	-	-	7.3	-
Gas	-	-	-	1.8	-	-
Subtotal	50	25	8.4	1.8	7.3	57.5
Total	75		75			

Table 2. Mass balance of HDPE: WCO=1:4

Component	Input		Output			
	IN1 (g)	IN2 (g)	OUT1 (g)	OUT2 (g)	OUT3 (g)	OUT4 (g)
HDPE	-	25	-	-	-	-
WCO	100	-	-	-	-	-
Char	-	-	10.3	-	-	-
Bio Oil	-	-	-	-	-	97.5
Wax	-	-	-	-	12.4	-
Gas	-	-	-	4.8	-	-
Subtotal	100	25	10.3	4.8	12.4	97.5
Total	125		125			

Yield Analysis

The percentage yield of products from the pyrolysis process using mixed feedstocks of HDPE plastics and waste cooking oil is shown in **Figure 3**. The results obtained are more liquid products, such as bio-oil, than char and gas products. This is supported by previous research, which indicates that fast pyrolysis is the optimal pyrolysis method for producing liquid products, as high temperatures and short residence times facilitate the

decomposition of organic compounds into volatile vapors (Ridhuan & Irawan, 2020).

Table 3. Mass balance of HDPE: WCO=2:1

Component	Input		Output			
	IN1 (g)	IN2 (g)	OUT1 (g)	OUT2 (g)	OUT3 (g)	OUT4 (g)
HDPE	-	50	-	-	-	-
WCO	25	-	-	-	-	-
Char	-	-	8.6	-	-	-
Bio Oil	-	-	-	-	-	54.9
Wax	-	-	-	-	7.9	-
Gas	-	-	-	3.6	-	-
Subtotal	25	50	8.6	3.6	7.9	54.9
Total	75		75			

Table 4. Mass balance of HDPE: WCO=4:1

Component	Input		Output			
	IN1 (g)	IN2 (g)	OUT1 (g)	OUT2 (g)	OUT3 (g)	OUT4 (g)
HDPE	-	100	-	-	-	-
WCO	25	-	-	-	-	-
Char	-	-	13.7	-	-	-
Bio Oil	-	-	-	-	-	89
Wax	-	-	-	-	14.4	-
Gas	-	-	-	7.9	-	-
Subtotal	25	100	13.7	7.9	14.4	89
Total	125		125			

Since HDPE is a saturated hydrocarbon polymer, its addition increases the saturated hydrocarbon fraction (alkanes/alkenes) in the bio-oil. However, it may also yield more gas and residue if the cracking process is uncontrolled. Using these four tables, it is possible to analyze how the composition of the raw materials affects conversion efficiency and the quality and quantity of the product fractions produced.

Figure 3 shows that the char yield at the 1:2 ratio decreases at the 1:4 ratio, then increases again until the 4:1 ratio. This indicates that adding WCO mass can reduce char formation, whereas adding HDPE mass can increase it. This is caused by the higher fixed carbon and ash content in HDPE than in WCO, as reported: HDPE plastics have 3.7% ash content and 4% fixed carbon (Habtewold et al., 2020). At the same time, WCO has very low fixed carbon and ash content of only 0.5 % (Kedir & Asere, 2022).

Meanwhile, the bio-oil yield is inversely related to the char yield, with a ratio of 1:2 increasing to 1:4, then decreasing again to 4:1. This indicates that adding WCO can increase bio-oil yield. The addition of HDPE plastics can reduce the bio-oil yield. This occurs because the HDPE volatile matter content is higher at 99 % than the WCO volatile matter content of 97 % (Rodríguez-Luna et al.,

2021; Sharma et al., 2021). The high volatile matter content of a material will be more volatile and released when heating occurs, so that it is likely to form a greater amount of non-condensable gas (Banchapattanasakda et al., 2023).

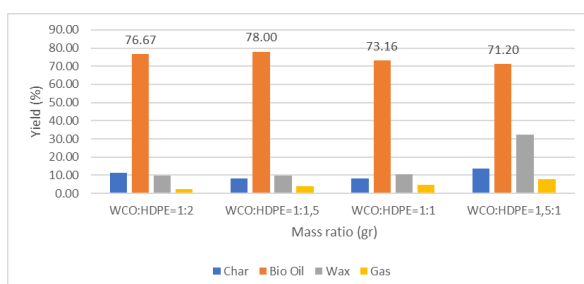


Figure 3. Yield products of co-pyrolysis WCO and HDPE plastic

In **Figure 3**, the gas yield increased with increasing feedstock mass, whether waste cooking oil (WCO) or HDPE plastics. The gas yield, which was initially only 2.4% at a mass ratio of 1:2, increased to 7.9% at a mass ratio of 4:1. This increase is attributed to a secondary reaction during cracking. According to previous research, at temperatures around 400–450 °C, it tends to produce high gas yields because secondary reactions produce highly volatile, non-condensable gases, which are generally short-chain, carbon-bonded compounds (C1–C5) (Rahman et al., 2021).

In this study, a wax product is one of the outputs from the pyrolysis of all types of plastic. Common waxes are mixtures of compounds, predominantly paraffins and olefins, that, at room temperature, form a wax-like form (Salaudeen et al., 2021). The wax yield gained in this study has increased, as shown in **Figure 3**. As previously explained, wax is a mixture of paraffin and olefin, saturated hydrocarbons derived from the cracking of HDPE (Ong et al., 2023).

The optimal condition obtained for producing hydrocarbon fuel with the highest yield is at an HDPE : WCO mass ratio of 1 : 4. Based on this result, calculations were performed to determine the percentage conversion of feedstocks into hydrocarbon fuel using stoichiometric analysis. Since the bio-oil product contains a variety of compounds, the calculation uses the compound with the highest percentage as a reference. In this case, heptadecane has an area percentage of 3.8 %. Additionally, oleic acid is used as a reference compound, as it has the highest percentage in waste cooking oil, at 45.6 %. The conversion of HDPE into heptadecane is found to be 29.39 %, and the conversion of oleic acid in waste cooking oil into heptadecane is 38.80 %. Both HDPE and WCO produce more compounds than heptadecane. This may occur because the decomposition processes of HDPE and WCO are non-selective. In the decomposition of HDPE, which is a long polymer of ethylene monomers, the decomposition products range from C5 to C22. Meanwhile, WCO contains

other fatty acids besides oleic acid, such as palmitic acid, stearic acid, and linoleic acid. When decomposed, the resulting compounds are not only hydrocarbons but also aldehydes, ketones, and other compounds.

The oleic acid content in WCO is an unsaturated fatty acid that will undergo decarboxylation to form heptadecane. Other studies report that using metal catalysts, such as Ni-Pt, in the thermal decomposition of oleic acid can increase the yield of heptadecane to 91 %. This occurs because metal catalysts such as Ni-Pt can accelerate the decarboxylation of oleic acid to heptadecane, in which the hydrogen atom on oleic acid is transferred to the Ni site (Chen et al., 2020). In the case of HDPE, the amount of monomer formed is governed by the substituent groups or atoms on the alpha carbon (i.e., the one with the unpaired electron), i.e., hydrogen atoms in HDPE. Suppose the substituent groups on the alpha-carbon are large. In that case, e.g., the methyl group in polypropylene or the phenyl group in polystyrene, the intermolecular hydrogen step is hindered (steric hindrance), and the reaction proceeds via reverse polymerization or unzipping, increasing the monomer yield (Diaz-Silvarrey et al., 2018).

Conclusions

The optimal condition for producing hydrocarbon fuel from HDPE plastic and WCO by co-pyrolysis, with the highest yield, was an HDPE:WCO mass ratio of 1:4, yielding 78%. Furthermore, stoichiometric calculations were used to determine the conversion and selectivity of each product component. The conversion of HDPE into Heptadecane was found to be 29.39 %, while the conversion of WCO into oleic acid was achieved with a selectivity of 38.80 %. Therefore, this study can serve as a foundation and a starting point for the design of a pilot plant to produce hydrocarbon fuel from HDPE plastic, specifically HDPE waste and WCO, through co-pyrolysis.

Conflict of Interest

The author states that they have no competing interests that could affect the publication of this paper.

Acknowledgment

The author would like to express sincere gratitude to all those who contributed to the success of this research. Special thanks are extended to Universitas Pembangunan Nasional “Veteran” Jawa Timur, whose invaluable guidance and support made this study.

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