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Development of an Innovative Thermal Cracking of Mixed-Plastic Waste Without Catalyst as an Alternative Fuel Source

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Abstract. The growing consumption of plastic is a major contributor to the substantial increase in waste, emphasizing the urgent need for the adoption of sustainable and effective practices in management and resource reclamation. Therefore, this study aimed to investigate the production of pyrolytic distilled oil (PDO) from five types of mixed-plastic waste using pyrolysis and distillation process without catalysts. Pyrolysis was conducted at a temperature range of 300 °C to 450 °C with a fixed heating rate of 10 °C/min for 60 minutes, while distillation was performed at 120 °C to 350 °C. The distilled pyrolytic oil obtained was characterized using analytical techniques with gas chromatography-mass spectrometry (GC-MS) and Carbon/Hydrogen/Nitrogen (CHN) analyses to determine its chemical content and heating value. The effect of pyrolysis and distillation temperature on product yield and hydrocarbon content was also examined. The results showed that the predominant compounds obtained were aromatic hydrocarbon groups, including styrene, benzene, naphthalene, and xylene. Other compounds included non-aromatic hydrocarbon groups, such as alkane and alkene. In addition, the chemical content was found to be comparable to that of product obtained from the use of common pyrolysis using catalysts. In this study, the heating values observed were in the range of 30.835 – 33.004 MJ/kg. However, the heating value of the product was still low and needed to be improved using another treatment. Comparison with previous reports showed that temperature in this study was not significant to the chemical content and heating value of the obtained distillate oil.

Keywords: Low temperature; Non-catalyst; Plastic waste; Pyrolysis; Pyrolytic oil

1. Introduction

Fossil fuels, such as coal, oil, and gas, are primary global energy sources that have been widely used for over a century. Despite the importance of these fuels, their extensive use has led to several detrimental effects, including climate change, air pollution, and resource depletion. Consequently, there is a pressing need to reduce their usage and support

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initiatives aiming for zero fossil fuels by 2050 (Holechek *et al.,* 2022). These detrimental effects are likely to persist on Earth when decisive actions are not taken to mitigate human dependence. Climate change, caused by the greenhouse gases emitted from burning fossil fuel-derived energy, has significantly impacted the planet. The impacts include rising sea levels, more frequent and intense weather events, and increased extinction rates for plants and animals (Haines *et al.,* 2006).

According to previous studies, mitigating human reliance on fossil fuels necessitates a transformative shift towards cleaner and more sustainable energy sources, including wind, solar, and geothermal power (Owusu and Asumadu-Sarkodie, 2016). This transition is expected to reduce environmental impact and create new economic opportunities and jobs within the renewable energy sector. Several studies have shown that continued dependence on the use of fossil fuels is unsustainable and poses a significant threat to Earth.

In line with several reports, plastic waste has become a significant environmental concern due to its widespread use, improper disposal, and slow degradation (Vriend *et al.,* 2021). In addition, it comprises discarded plastic, such as packaging, bags, bottles, straws, and other single-use variants. The waste typically causes several environmental problems, including land and water pollution, where it ends up in landfills, taking centuries to degrade (Sari *et al.,* 2022). This condition often leads to its accumulation in landfills, contributing to soil contamination and the release of harmful chemicals. The consequences extend to marine pollution, where it accumulates in the oceans and water bodies, posing a severe threat to marine life.

Compared to other countries, Indonesia also needs a mix of energy sources to meet its future demands while reducing its dependence on fossil fuels. This approach requires increasing the use of renewable sources, improving energy efficiency, and exploring new technologies to support a more sustainable system of producing liquid biofuel (Simanjuntak, Tambunan, and Sihombing, 2023; Simanjuntak *et al.,* 2022). In Indonesia, two crucial sources of liquid fuel production are organic and inorganic materials. Organic materials, typically sourced from plants or biomass, serve as a highly environmentally friendly alternative and are often referred to as green fuel.

A typical example of organic materials is palm oil, which serves as an ideal and highly productive source of biofuel for internal combustion engines (Prihadiyono *et al.,* 2022). Algae plants also have the potential to serve as a renewable energy source due to their high productivity and ability to grow in diverse environment (Sardi *et al.,* 2022; Jamilatun *et al.,* 2020). In addition, inorganic materials, such as plastic waste, have high productivity and can be used for industrial and construction purposes. Various plastic materials can be processed into liquid fuels with properties similar to diesel (Suhartono *et al.,* 2023). The quality of liquid fuels can be improved by mixing organic and inorganic materials (Kusrini *et al.,* 2018). Plastic has been reported to possess the potential to be used as building material, thereby reducing concrete production costs. Previous reports showed that it could also be used as aggregate substitute for cement in concrete. The aggregate acts as filling materials that contribute to the structural strength of concrete (Purnomo, Baskoro, and Muslim, 2023).

Plastic waste can be a potential source for future energy mixes through the use of pyrolysis. This method comprises heating the waste to high temperatures in the absence of oxygen, leading to the breakdown of the constituent long polymer chains and the production of a liquid fuel known as pyrolysis oil (Sharma *et al.,* 2014). The liquid product obtained can be used as a fuel substitute or blended with bio-oil for internal combustion engines (Awang *et al.,* 2021), electricity generation, or feedstock for the production of chemicals. Another effective method of using plastic waste is gasification (Ahmed and

Gupta, 2010), which comprises heating in the presence of oxygen and steam to produce a gas mixture called syngas. This product can be burned for heat and electricity or processed into biofuels, such as ethanol or methanol.

Based on findings, pyrolysis and gasification have the potential to convert plastic into useful energy sources while also reducing the amount of waste being accumulated in landfills or the environment. However, there are also some challenges associated with its use as an energy source. These include potential emissions of harmful pollutants during the conversion process, the need for careful handling to prevent contamination with other materials, and the cost of building and operating the necessary infrastructure (Verma *et al.,* 2016).

According to previous studies, pyrolysis is a promising solution for addressing the issue of mixed-plastic waste. This process comprises the thermal decomposition of organic materials in an oxygen-deprived environment, leading to the production of valuable by-products, including fuel, gases, and char (Papari, Bamdad, and Berruti, 2021). Pyrolysis of mixed-plastic waste holds significant potential as it can generate a valuable energy source, serving as a sustainable alternative to fossil fuels. This transition not only mitigates greenhouse gas emissions but also contributes to the advancement of the circular economy (Siddiqui and Redhwi, 2009). However, it presents inherent challenges, such as the substantial variability in substrate composition and the imperative need for optimal operating conditions to ensure the production of high-quality fuels (Kasar, Sharma, and Ahmaruzzaman, 2020; Al-Salem *et al.,* 2017). The complexity of managing diverse waste streams necessitates comprehensive study efforts to explore the technical feasibility, economic viability, and environmental impact associated with the pyrolysis process.

Plastic waste holds great promise in the petrochemical industry through the pyrolysis process, leading to the release of several valuable industrial materials. In this study, data from previous research by Soni *et al*., (2021) is referenced, which demonstrates that plastic waste, categorized by its recycling code, contains a range of chemical components.

The values presented represent the proximate analysis of a single sample obtained from different types. The high volatile fraction and low ash content showed that co-pyrolysis process can contribute to optimal pyrolysis production, yielding a high heating value for fuels.

The current trend in plastic pyrolysis studies focuses on achieving high yields of products with high quality and low energy consumption, collectively referred to as high efficiency. This efficiency can be enhanced by using catalysts, which allow for lower operational temperatures (Peng *et al.,* 2022). However, most studies employing catalysts have been conducted on individual types of plastic. The use of these materials presents challenges, such as feeding issues and difficulties in process control. Despite the drawbacks, the primary benefit of using catalysts is the higher production of bio-oil (Kim, 2004). Previous reports have explored pyrolysis without catalysts, particularly when processing single types of plastic. These reports have demonstrated the possibility of achieving comparable yields at moderate temperatures (Miskolczi *et al.,* 2004).

Although previous findings still consider the operating temperature for plastic pyrolysis to be high, there is potential for synergy between different types of plastic when mixed. This synergy could allow for lower operating temperatures and the production of a significant amount of hydrocarbons. However, determining the optimum operational temperature for pyrolyzing a mixture of five types without using catalysts remains a challenge. This issue arises because their cost is prohibitively expensive for large-scale production. Therefore, this study aimed to explore the potential of five mixed-plastic waste as feedstock for alternative distilled oil production through pyrolysis and distillation

processes without using catalysts at a low-temperature process. The influence of operating pyrolysis and distillation temperature on the pyrolytic oil yield and properties of oil was also examined.

2. Materials and Methods

2.1. Materials

This study involved the utilization of a mixed feedstock comprising five types of plastic waste. The plastic materials were collected from the trash bin around the study site, which was sorted, cleaned, and dried before being put into the pyrolysis apparatus. The feedstock included polyethylene terephthalate (PET), low-density polyethylene (LDPE), high-density polyethylene (HDPE), polypropylene (PP), and polystyrene (PS) derived from mineral beverage water bottles, grocery plastic bags, detergent bottle caps, beverage pipettes, and food wrapping of Styrofoam respectively. The feedstock was chopped manually to achieve a standardized size of approximately 3 cm x 3 cm. The proximate analysis of the material used was presented in Table 1.

Table 1 Proximate Analysis of Plastic (%)

Plastics	C	H	N	S	O	Cl
HDPE	85.40	15.04	0.00	0.52	2.77	0.14
PVC	38.53	5.04	0.013	0.176	0.00	56.25
PS	92.59	8.13	0.00	0.00	2.24	0.00
LDPE	77.60	21.55	0.00	0.003	0.00	0.00
PET	77.00	13.00	0.20	n.a	5.00	n.a

2.2. Method

Approximately 200 grams of each type of plastic was introduced into the pyrolysis reactor and heated to the desired temperature without oxygen using an LPG combustor. During the heating process, the mixed plastic was degraded into lighter molecules in the form of vapor. After passing through the condenser, the vapor reached its saturation temperature and condensed into liquid. Subsequently, a distillation process was used to separate the liquids according to the density of molecules which resulted in pyrolytic distilled oil (PDO), exhibiting properties similar to fossil-based liquid fuels.

Operating temperature for this process ranged from 300 to 450 °C, while the heating rate and residence time were 10 °C and 60 minutes, respectively. The schematic of the process is presented in Figure 1. The procedure began with pyrolysis, followed by the cooling of the pyrolytic vapor in the condenser until all vapor transformed into condensate. After pyrolysis process was completed, the reactor was allowed to cool down and the condensate was extracted from the condenser. In addition, distillation was conducted within the same reactor, generating pyrolytic oil as the final product.

The type of distillate or pyrolytic oil obtained was based on the operating temperature utilized during the process. A total of six samples were prepared for characteristic testing using only CHN and GC-MS methods to analyze the product yield in terms of chemical and molecule components. According to Ghodke (2021), there were six methods for characterizing pyrolytic oil. However, in this study, the CHN-GCMS was selected due to the cost-effectiveness and time consumed during the characterization process using multiple methods.

Pyrolysis process comprised the thermal decomposition of inorganic materials in the absence of oxygen. During the process, the plastic feedstock was subjected to decomposition based on the application of heat. The absence of oxygen prevented combustion, leading to the formation of various products, including gases, liquids or PPO,

and solids. These products could then be further processed or refined for various applications, such as fuel production, chemical synthesis, or waste management. The equation (1) below shows a simplified representation of pyrolysis reaction.

$$C_nH_mO_lN_kS_j + Heat \rightarrow Pyrolysis\ Product \quad (1)$$

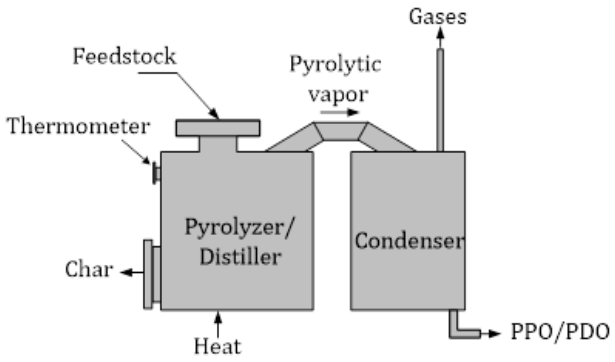

Figure 1 Schematic pyrolysis system of five types of plastic waste

3. Results and Discussion

The plastic pyrolytic oil (PPO) product, derived from pyrolysis of a mixture of five types of plastic, occurred within a temperature range of 300 to 450 °C. The procedure yielded predominantly liquid oil, constituting an intricate amalgamation of hydrocarbons, along with a solid residue identified as char. This char primarily consisted of mixed carbon, and the quantity generated was dependent on the type of plastic and the specific pyrolysis conditions employed. The yield of each product was intricately linked to several factors, including the composition of the mixed plastic, the temperature range applied, the heating rate, and various other parameters. In addition, the choice of the pyrolysis reactor and the conditions under which the procedure was conducted significantly influenced the final product outcomes.

In this study, the yield of PPO from the five mixed plastics reached 45% by weight of the feedstock at a temperature of approximately 400 °C. This finding was consistent with the results of other reports, who achieved a yield of up to 69% when pyrolyzing mixed plastic at approximately 500 °C (Genuino *et al.,* 2022). Figure 2 shows the appearance of pyrolytic distilled products (PDO), revealing a strong dependence on the applied distillation temperature. A total of six samples (S1-S6) each with a volume of 10 ml, and subjected to different pyrolysis and distillation temperature, were tested. The specific conditions for each sample were 1) S1 pyrolyzed at 350 ^{o}C and distilled at 120 ^{o}C, 2) S2 pyrolyzed at 400 ^{o}C and distilled at 250 ^{o}C, 3) S3 pyrolized at 400 ^{o}C and distilled at 350 ^{o}C, 4) S4 pyrolyzed at 300 ^{o}C without distillation from the gasoline-like port, 5) S5 pyrolyzed at 300 ^{o}C without distillation from the diesel-like port, and 6) S6 pyrolyzed at 380 ^{o}C and distilled at 180 ^{o}C. Lower distillation temperature (<200 °C) led to the production of products with bright colors, while higher temperature (>200 °C) produced a yellowish hue. This observation was in line with the findings of a previous report (Wiriyaumpaiwong and Jamradloedluk, 2017).

Figure 2 Six samples of PDO of mixed five types of plastic pyrolysis and distillation

3.1. Chemical characterization of plastic distilled oil

The chemical characterization of the plastic distilled oil was conducted at the National Study and Innovation Agency, Indonesia (BRIN). The samples were subjected to total CHN (carbon/hydrogen/nitrogen) and analyzed using an Exeter Analytical (Chelmsford, MA) CE-440 Elemental Analyzer. Subsequently, the oxygen content was determined through mass balance closure. Table 2 presented the elemental composition and heating value of the PDO obtained from the five mixed types of plastic. Table 3 provided a comparison of the results with those from another study that used a catalyst. The chemical content was found to be comparable to the common pyrolysis processes using catalysts. However, it was noteworthy that the heating value was lower compared to the report by Sharma *et al.* (2014). This disparity could be attributed to the higher oxygen content in the PDO produced in this study.

Table 2 Elemental composition and heating value of PDO of five mixed plastics (wt %)

Sample Name	Carbon (%)	Hydrogen (%)	Nitrogen (%)	Oxygen (%)	HHV (MJ/kg)
S1	85.633	10.540	0.019	3.808	33.004
	85.171	10.385	0.077	4.367	32.768
S2	80.302	10.123	0.061	9.514	31.221
	79.029	10.081	0.041	10.849	30.835
S3	82.951	11.521	0.052	5.476	32.903
	83.325	11.545	0.054	5.076	33.025
S4	82.190	10.032	0.078	7.700	31.692
	82.133	10.244	0.060	7.563	31.818
S5	81.278	10.680	0.057	7.985	31.869
	81.607	10.785	0.067	7.541	32.032
S6	83.266	11.328	0.061	5.345	32.863
	81.719	11.071	0.043	7.167	32.255
Average	82.384	10.695	0.056	6.866	32.190

Table 3 Elemental composition (wt. %) and heating value of PDO, catalyst and non-catalyst process

Carbon (%)	Hydrogen (%)	Nitrogen (%)	Oxygen (%)	HHV (MJ/kg)	Reference
81.24	14.69	0.13	3.95	34.54	Quesada *et al.* (2019)
74.90	1.70	2.78	8.6	32.12	Santella *et al.* (2016)
82.38	10.69	0.06	6.87	32.19	This work (Non-catalyst)

3.2. Gas chromatography-mass spectroscopy (GC–MS)

Figures 3 and 4 depicted the molecular fraction successfully recorded by GC-MS for the six samples (S1–S6). The data exhibited a consistent and relatively similar pattern, suggesting that the impact of temperature on pyrolysis products was not highly significant. The GC-MS analysis revealed two distinct hydrocarbon groups, which were aromatics, including benzene, toluene, xylene, styrene, and naphthalene, and a non-aromatic group

comprising C1-C10 alkanes, C11-C20 alkanes, and C1-C10 alkenes. Table 4 provided comparative data on pyrolysis results with and without the use of a catalyst. From Table 4 it was evident that the pyrolysis process without a catalyst had a similar product of liquid hydrocarbon with pyrolysis without a catalyst.

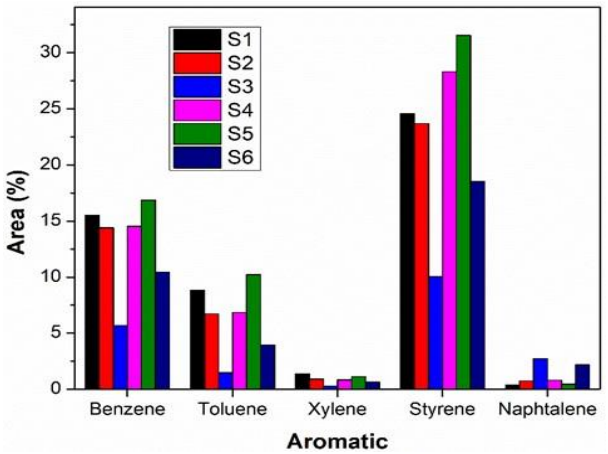

Figure 3 Selectivity of compounds produced by S1-S6 samples against aromatic hydrocarbons

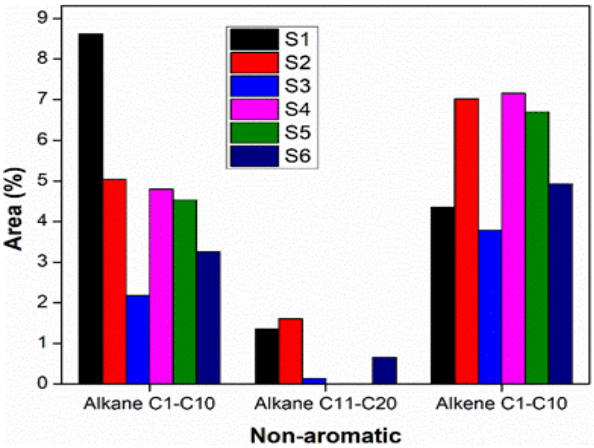

Figure 4 Selectivity of compounds produced by S1-S6 samples against non-aromatic hydrocarbons

According to these data, the PDO yields were predominantly composed of aromatic hydrocarbons. The GC–MS analysis revealed that the pyrolysis liquid oils from the 6 samples mainly consisted of aromatic hydrocarbons with a minor presence of aliphatic hydrocarbon compounds similar to the findings of Miandad *et al.* (2017). Specifically, styrene, derived from the cracking of polystyrene (PS), was the dominant aromatic compound, which was consistent with the study conducted by Shah and Jan (2015). The production of aromatics from the pyrolysis of polyethylene (PE) could involve Diels-Alder reaction and dehydrogenation mechanisms (Miandad *et al.,* 2019). This study identified a high percentage of BTX components (benzene, toluene, xylene), styrene monomers, and other mono-aromatic compounds, highlighting their potential application in the petrochemical industry. In addition, these mono-aromatic compounds could be blended into the gasoline pool to enhance the fuel's octane number (Jaafar *et al.,* 2022).

Table 4 Comparison of the chemical composition of liquid products identified by GC–MS

Component name					
Naphthalene	Benzene	Toluene	Xylene	Styrene	References
17.20	28.8	28.80	n.a	n.a	Aisien, Otuya, and Aisien (2021) *)
0.21	1.10	1.49	n.a	64.31	Shah and Jan (2015) *)
0.63	4.00	15.30	8.80	24.50	Miandad *et al.* (2017) *)
2.90	4.50	24.00	3.40	54.0	Onwudili, Insura, and Williams, (2009) **)
2.5	5.28	10	1.75	30	This work**)

*) Catalyst

*) Non-catalyst

For polystyrene, the raw material's highly aromatic nature resulted in the predominant production of aromatic products during pyrolysis. Conversely, for PET, LDPE, and HDPE, the generated oil mainly comprised aliphatic hydrocarbons, including alkanes and alkenes (Budsaereechai, Hunt, and Ngernyen, 2019). The hydrocarbon groups ranging from C4–C11 represented light fraction petroleum fuels, while carbon numbers from C12–C20 indicated medium fraction diesel fuels. The thermal degradation of polyalkene mixed plastics occurred through random cutting, producing a diverse range of hydrocarbon fragments with varying carbon atom numbers. The weakest C-C bonds in the polyalkene structure underwent random cutting reactions during degradation, leading to the formation of carbon double bonds (C=C) in the resulting structure. Consequently, the pyrolysis oil obtained showed a notable concentration of alkenes. This thermal degradation process, induced by random cutting reactions, resulted in the formation of a diverse array of hydrocarbon species. However, due to the presence of CH_3 side chains in the PET structure, several hydrocarbons were also formed alongside those observed in the pyrolysis of other polyalkenes (Siddiqui and Redhwi, 2009)."

The pyrolysis oil content, dominated by aromatic monobenzene, as well as non-aromatic alkanes and C1-C10 alkenes, indicated that plastic samples had undergone a cracked process to produce short-chain hydrocarbons. Furthermore, these compounds also produced content, such as Benzoic acid, n-Pentadecanol, 1-Tetradecanol, and some >C21 components, including Behenic alcohol, and Hentriacontane (Carbonic acid, decyl undecyl ester). The heavy oil content of compounds with >C21 was primarily obtained from the degradation of HDPE, which was mostly found in sample S6.

4. Conclusions

In conclusion, valuable industrial products could be obtained from five mixed plastic waste through pyrolysis and distillation processes without a catalyst. This waste included polyethylene terephthalate (PET), low-density polyethylene (LDPE), high-density polyethylene (HDPE), polypropylene (PP), and polystyrene (PS) from mineral water bottles, grocery bags, detergent bottle caps, beverage pipette, and Styrofoam food wrapping respectively. The chemical content was found to be comparable to the common pyrolysis utilizing a catalyst. The heating values achieved ranged from 30.835 to 33.004 MJ/kg. However, this heating value was relatively low and required improvement through the oxygenation process or blending with biodiesel, kerosene, and other fuels with higher heating values.

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