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Staged Catalytic Co-Pyrolysis of Polypropylene and High-Density Polyethylene: Optimizing Liquid Fuel Yield and Composition

Mohamad H. H. Deifalla[1], Yousif A. A.[2*], Muhab Hassanien S. S.[3], Ibrahim Y.I. Elgady[4]

[1]Chemical Engineering Department, Sudan University of Science and Technology, Khartoum, Sudan.

[2,3,4] Polymer Engineering Department, Sudan University of Science and Technology, Khartoum, Sudan.

ABSTRACT: The escalating global production of plastics and the depletion of fossil fuel reserves underscore the urgency of sustainable waste-to-energy strategies. This study investigates the staged catalytic pyrolysis of polypropylene (PP), high-density polyethylene (HDPE), and their blends for the production of liquid fuels. Experiments were conducted in a semi-batch reactor at 450 °C (Stage A) and 500 °C (Stage B), with bentonite as catalyst. Product yields and compositions were quantified via mass balance and GC–FID analysis. Results revealed strong feedstock-dependent behaviors: HDPE exhibited superior liquid recovery (81.96% in Stage A, 88.24% in Stage B) with minimal char, whereas PP was prone to higher char and gas formation. Co-pyrolysis demonstrated synergistic effects, with asymmetric mixtures outperforming single-polymer systems. Notably, the 70% PP–30% HDPE blend achieved the highest liquid recovery (95.07%) and lowest gas fraction (4.92%) during secondary cracking, while the 30% PP–70% HDPE blend enhanced diesel- and kerosene-range fractions. GC–FID analysis confirmed that PP favored gasoline-range hydrocarbons, while HDPE enriched middle distillates. The tunability of hydrocarbon distribution through feed composition highlights staged pyrolysis as a robust pathway for transforming mixed plastic waste into targeted fuel-range hydrocarbons. These findings provide actionable insights into optimizing product selectivity and yield, advancing the integration of polyolefin pyrolysis into circular economy and sustainable energy frameworks.

KEYWORDS: Catalytic; Thermal Pyrolysis; Co-pyrolysis; Fuel; Polyethylene; High-Density Polyethylene; Bentonite Catalyst.

INTRODUCTION

The rising global demand for energy, combined with the growing challenge of plastic waste management, has generated strong interest in sustainable strategies for converting discarded plastics into valuable products, particularly liquid fuels. Such an approach tackles the dual issues of plastic accumulation in the environment and the urgent need for alternative energy sources as fossil fuel reserves continue to decline [1]. Globally, plastic production has surged from about 1.5 million tonnes in 1950 to nearly 400 million tonnes by 2020, representing an increase of around 266% over seven decades [2]. Among the proposed solutions, pyrolysis has emerged as a promising thermo-chemical process, in which long-chain hydrocarbons are broken down into shorter molecules at elevated temperatures under oxygen-free conditions, enabling the conversion of plastics such as polypropylene and high-density polyethylene into liquid fuels [3, 4]. This method not only reduces the environmental footprint of plastic waste but also supports the circular economy by turning a persistent pollutant into a usable energy resource [5, 6]. In particular, polypropylene and high-density polyethylene, which are among the most widely used plastics, can undergo pyrolysis to produce liquid hydrocarbons with fuel characteristics similar to diesel and gasoline [7, 8]. These liquid products are typically rich in calorific value, making them suitable for both transportation fuels and chemical feedstocks [9]. The broad utilization of these plastics further enhances the potential of their conversion into fuels, offering a practical solution for waste reduction and energy recovery [10, 11]. Moreover, the flexibility of pyrolysis allows for fine-tuning of operational parameters such as temperature and heating rate, which can be optimized to improve product distribution and maximize yields of targeted fuel fractions [12].

The conversion of polymeric waste materials, such as polypropylene and high-density polyethylene, into valuable liquid fuels via catalytic thermal pyrolysis has garnered significant attention as a sustainable waste management strategy and a means of circularizing plastic economies [13]. This thermochemical approach effectively mitigates the environmental impact of plastic accumulation while simultaneously addressing the growing demand for alternative energy sources [14]. Pyrolysis, the thermal cracking of long hydrocarbon chains into shorter ones at elevated temperatures (300–800 °C) in the absence of oxygen, represents a promising pathway for transforming plastic waste into useful products such as liquid fuels and high-calorific-value gases [3]. The introduction of catalysts into the pyrolysis process can significantly enhance reaction rates, improve product selectivity, and lower activation

energies, leading to more economically viable solutions for large quantities of waste [15]. This catalytic approach facilitates the production of gasoline and diesel-like fuels, offering a direct pathway to valorize plastic waste into high-value energy carriers [3, 13]. However, the economic feasibility of pyrolysis is often constrained by high energy consumption, a challenge that induction heating has shown promise in mitigating for biomass pyrolysis, though its application in plastic pyrolysis remains less explored [16]. Despite this, catalytic pyrolysis is considered an efficient method to convert plastic waste into high-value products like hydrogen and jet fuel, demonstrating significant potential for waste recovery and utilization [17].

Recent studies have demonstrated that pyrolysis of PP and HDPE at optimized temperatures between 350°C and 650°C effectively breaks polymer chains into hydrocarbon liquids, gases, and minimal char residues, with liquid oil yields reaching up to 90 wt% under certain conditions. The liquid fuels produced consist mainly of C_5–C_{20} hydrocarbons, including aromatics, alkanes, and alkenes, making them potential alternatives for diesel and gasoline substitutes. Furthermore, co-pyrolysis of PP and HDPE or blends with other biomass materials can enhance liquid yield and fuel quality through synergistic effects between different feedstocks [12, 18-23].

However, catalyst-assisted pyrolysis has been explored to improve the selectivity and properties of the liquid fuels, steering the product distribution towards high-value chemicals such as benzene, toluene, and xylene (BTX) and reducing unwanted by-products [24-26]. The influence of process parameters including temperature, reaction time, feedstock composition, and heating rate has been systematically evaluated, showing that a balance between these variables is crucial to maximize liquid fuel production while minimizing gas and char formation.

Park and Lee [27] demonstrated that the thermal degradation of polypropylene (PP) is strongly governed by molecular weight distribution, where lower molecular weight fractions favor fuel-range hydrocarbon formation due to enhanced chain scission. Similarly, Aremanda and Singh [28] confirmed that non-catalytic pyrolysis of high-density polyethylene (HDPE) at 450–600 °C yields diesel-range hydrocarbons suitable for blending without extensive pretreatment. Tsang et al. [2] further emphasized the importance of feedstock control, showing that optimized PP molecular weight enhances selectivity toward liquid fuels. Ghodke et al. [29] reported that pyrolysis of HDPE, LDPE, PP, mixed plastics, and municipal waste at 773 K produced liquid yields of 62–68 wt%, predominantly hydrocarbons in the C8–C20 range, with properties comparable to conventional fuels. Wang et al. [30] highlighted that PP pyrolysis in a double-fluidized-bed reactor can generate BTX aromatics without catalysts, underscoring temperature as a critical parameter for product distribution. Dhaniswara et al. [16] also showed that integrating pyrolysis with fractionation enables temperature-driven control of yield and composition, with C1–C13 aromatic fractions exhibiting calorific values similar to commercial fuels.

Additionally, simulation software such as Aspen HYSYS has been widely utilized for modeling chemical reactions in continuous stirred tank reactors (CSTRs), offering reliable predictive capabilities for process improvement [31].

In the present work, the role of feedstock composition in staged catalytic pyrolysis of PP, HDPE, and their blends is examined, with emphasis placed on maximizing liquid fuel yield, minimizing char formation, and elucidating the synergistic effects governing fuel-range hydrocarbon distribution.

MATERIALS AND METHODS

Materials:

The feedstock employed in this study was waste plastic composed of polypropylene (PP) and high-density polyethylene (HDPE). These were tested both as individual polymers and as binary blends with weight proportions of 100% PP, 100% HDPE, 50:50 PP/HDPE, 30:70 PP/HDPE, and 70:30 PP/HDPE.

Bentonite Catalyst

Bentonite powder was used as a catalyst, with a fixed mass of 5 g applied in the experiments.

Experimental setup

The pyrolysis trials were performed using a laboratory-scale semi-batch reactor with a total volume of 1.0 L. External heating was supplied by a Glas-Col mantle capable of reaching temperatures up to 1000 °C. Reactor temperature was measured and controlled with a thermocouple (range: 0–1300 °C), providing precise thermal management during the experiments. The gaseous products generated were passed through a water-cooled condenser, where the condensable portion was collected in a receiving flask, as illustrated in Figs. 1 and 2. A recirculating water pump was employed to maintain a steady coolant flow, ensuring efficient condensation. All experimental procedures were carried out inside a fume hood for safety purposes. Mass and time measurements were taken using a high-accuracy analytical balance (AMPUT) and a digital stopwatch, respectively. A schematic diagram of the complete experimental arrangement is shown in Fig. 3.

Thermal pyrolysis process

In each experimental run, 50 g of plastic feed was placed inside the reactor and heated up to 450 °C. The vapors generated were condensed to obtain liquid oil, while the solid residue (char) remained within the reactor. The amount of gaseous products was calculated through a mass balance approach. To assess the effect of secondary cracking, the wax fraction collected from stage A was fed back into the reactor

and subjected to a higher temperature of 500 °C (stage B), leading to the production of additional liquid hydrocarbons.

Experimental Design

A total of five pyrolysis runs were conducted using varying polypropylene (PP) and high-density polyethylene (HDPE) feed ratios: (i) pure PP, (ii) pure HDPE, (iii) equal mixture of PP and HDPE (50:50), (iv) 30% PP with 70% HDPE, and (v) 70% PP with 30% HDPE. Each run was carried out in two consecutive stages.

- Stage A (Primary Pyrolysis): The polymer feedstocks were thermally decomposed in an inert atmosphere to generate volatile vapors, waxes, and char.
- Stage B (Catalytic Cracking): The heavy waxes and long-chain hydrocarbons from Stage A were passed through a secondary catalytic reactor containing Bentonite as the catalyst. Bentonite, a naturally occurring aluminosilicate, provides strong acidic sites and a layered structure that promote cracking, deoxygenation, and aromatization reactions. Its use enhanced the yield of lighter liquid hydrocarbons, reduced wax accumulation, and improved overall fuel quality.

Bentonite was chosen due to its low cost, natural abundance, thermal stability, and environmentally benign nature, making it a practical alternative to synthetic zeolites for catalytic pyrolysis.

1.1. Product analysis

The composition of the liquid products obtained from pyrolysis was analyzed using gas chromatography equipped with a flame ionization detector (GC–FID, Agilent Technologies). This technique allowed efficient separation of the hydrocarbon constituents and enabled quantitative evaluation of fuel-like fractions, including those corresponding to gasoline, kerosene, diesel, and heavy oil present in the pyrolysis oil.

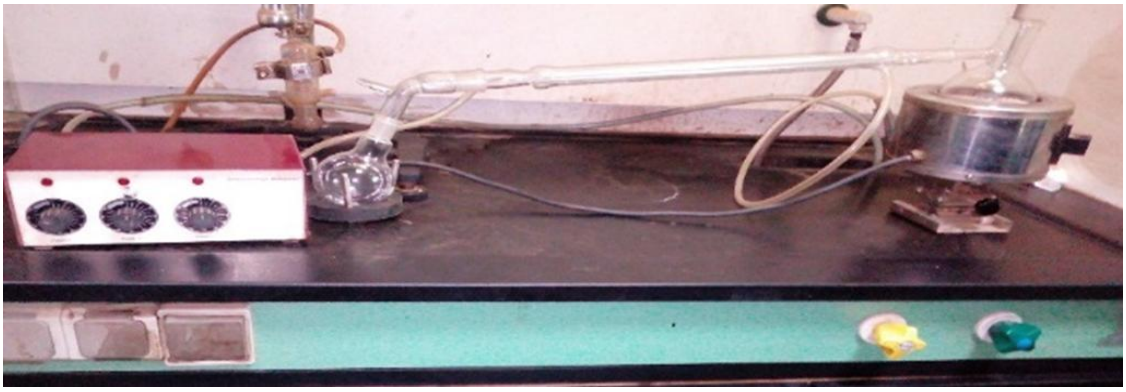

Fig. 1. The co-pyrolysis process.

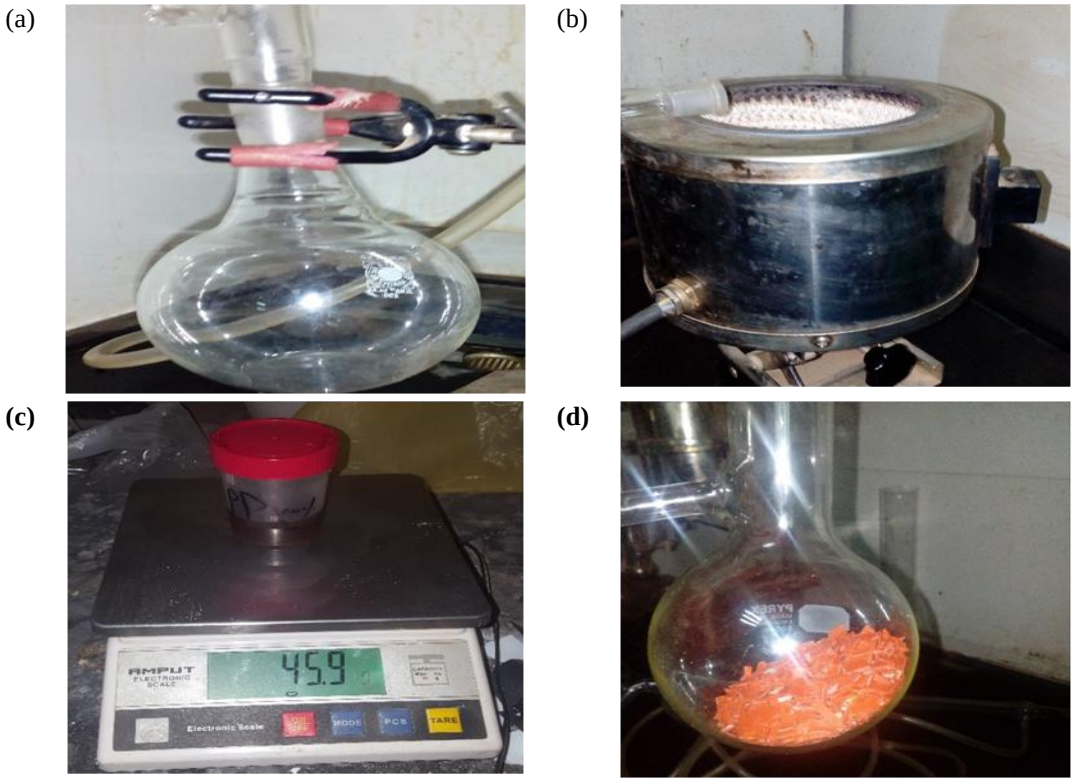

Fig. 2. Experimental apparatus of the co-pyrolysis process (a) receiving flask, (b) a heating mantle, (c) an analytical balance, (d) a flask containing PP and HDPE blend.

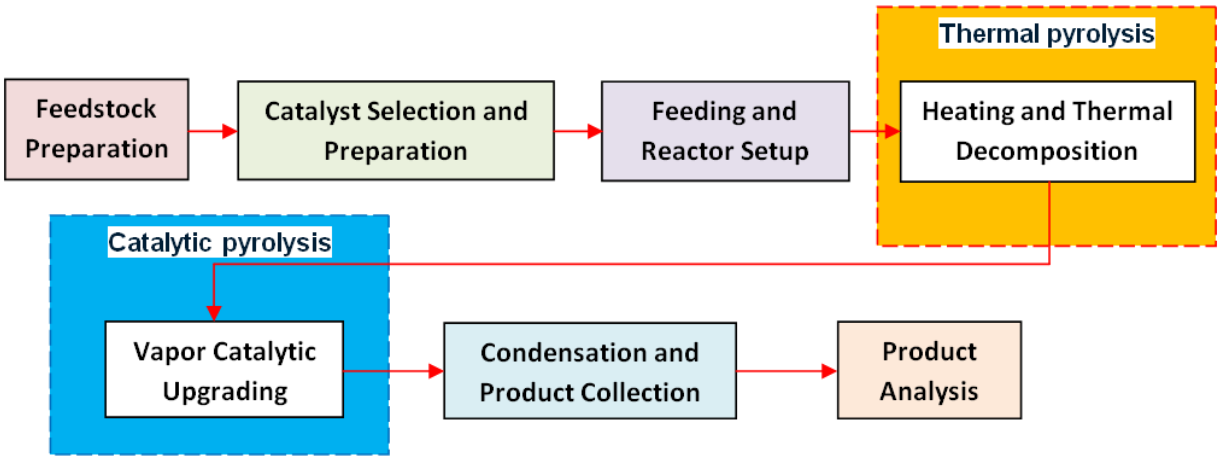

Fig.3. Schematic workflow of the co-pyrolysis study.

RESULTS AND DISCUSSION

The staged catalytic pyrolysis of PP, HDPE, and their blends revealed significant differences in degradation behavior and product selectivity as shown in Tables 1 and 2. In Stage A (450 °C), pure PP generated 26.14 g of liquid, accompanied by elevated gas (6.76 g) and char (9.92 g) formation. Fig. 4 shows the final products of PP, HDPE, and their blends obtained from the co-pyrolysis process. This product spectrum reflects the random chain scission of tertiary carbons in PP, which promotes radical recombination, gas evolution, and coke deposition. Increasing the proportion of PP in the feed was correlated with higher char yields (up to 9.5 g for the 30% PP–70% HDPE blend) and reduced reaction temperature requirements, consistent with the enhanced cracking reactivity of PP.

In contrast, HDPE exhibited superior performance, producing the highest liquid fraction (40.98 g) with the lowest char residue (5.13 g). This outcome is attributed to HDPE's linear structure, where β-scission dominates and favors the formation of long-chain hydrocarbons and waxes with limited secondary condensation. Binary mixtures yielded intermediate values, yet evidence of synergism was apparent. For example, the 70% PP–30% HDPE blend generated 39.3 g of liquid and only 7.5 g of char, demonstrating that HDPE partially suppresses PP's tendency for solid residue formation. Similarly, the 50:50 mixture balanced the characteristics of the two polymers, producing 38.7 g of liquid with moderate gas and char fractions.

Stage B (500 °C) secondary pyrolysis further transformed the wax-rich intermediates into liquid oils, with negligible char formation across all formulations. For PP, the liquid yield increased to 27.4 g, accompanied by 9.1 g of gases, confirming that secondary cracking volatilizes waxes and reduces solid residues. HDPE maintained the highest overall performance, generating 28.0 g of liquid with minimal gas (3.4 g), underscoring its ability to produce stable fuel-range hydrocarbons under severe thermal conditions. Binary blends revealed complex interactions. The 70% HDPE–30% PP mixture delivered 26.5 g of liquid with only 3.2 g of gas, closely resembling pure HDPE and confirming the role of HDPE in stabilizing liquid production. Conversely, the 70% PP–30% HDPE blend, though initially more char- and gas-prone, produced 25.1 g of secondary liquid, demonstrating that HDPE moderates PP's gasification pathways during secondary cracking.

Taken together, these results indicate that HDPE-rich systems maximize liquid recovery with minimal char and gas, while PP-rich systems require secondary processing to enhance oil yield. The synergistic effects observed in co-pyrolysis highlight that feedstock composition provides a tunable lever for selective product distribution. Among the tested formulations, the 70% HDPE–30% PP blend consistently offered the most favorable balance, high wax yield in Stage A and substantial oil recovery in Stage B, making it the most promising candidate for liquid fuel production from mixed plastic waste.

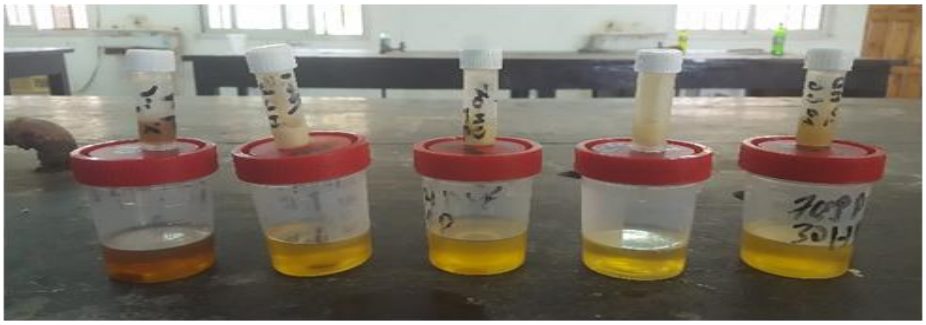

Fig. 4. Final products of PP, HDPE, and their blends from the co-pyrolysis process.

Table 1 Pyrolysis of PP, HDPE, and their mixtures (Stage A, 450 °C)

Run	Feed Composition (%)	Temperature (°C)	Reaction Time (min)	Final Composition (g)	Gases (g)	Char (g)
1	100% PP	260	26.14	38.32	6.76	9.92
2	100% HDPE	376	25.00	40.98	8.72	5.13
3	50% PP-50% HDPE	238	24.02	38.70	7.70	8.60
4	30% PP-70% HDPE	364	28.27	37.00	8.50	9.50
5	70% PP-30% HDPE	280	27.00	39.30	8.20	7.50

Table 2 Experimental results of thermal pyrolysis of PP, HDPE, and their blends (Stage B, 500 °C).

Run	Feed Composition (%)	Temperature (°C)	Initial Amount (g)	Reaction Time (min)	Final Liquid (g)	Gases (g)
1	100% PP	175	36.50	14.23	27.40	9.10
2	100% HDPE	178	28.90	28.00	25.50	3.40
3	50% PP-50% HDPE	125	32.20	11.09	21.50	10.70
4	30% PP-70% HDPE	124	29.70	11.07	26.50	3.20
5	70% PP-30% HDPE	126	26.40	13.21	25.10	13.00

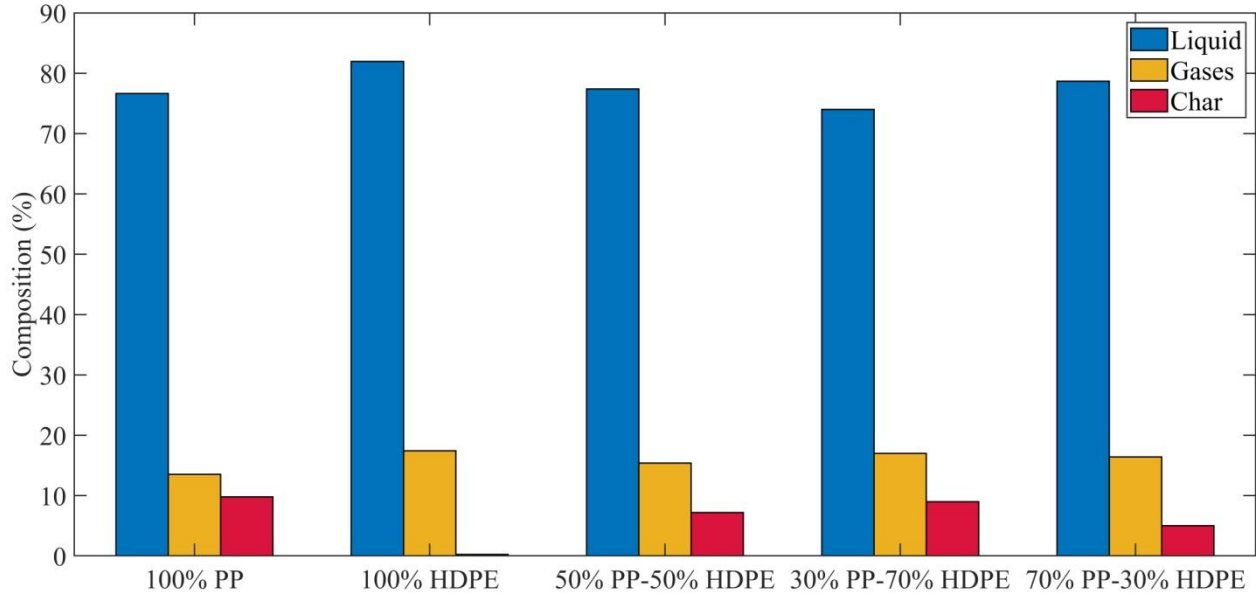

Fig. 5. GC–FID analysis of the co-pyrolysis process at Stage A.

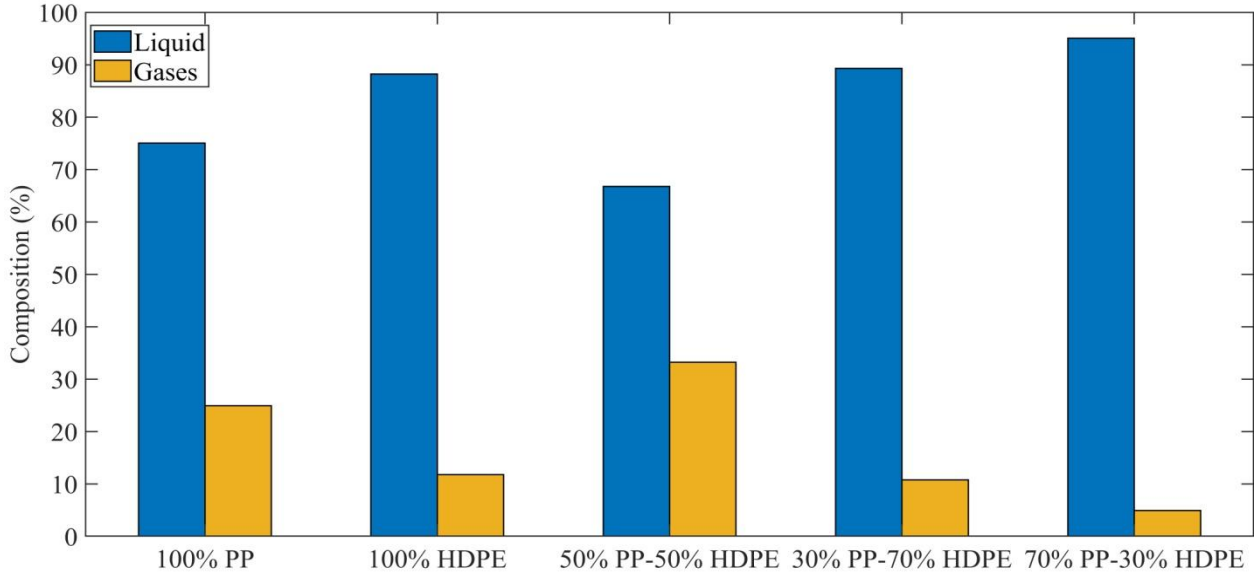

Fig. 6. GC–FID analysis of the co-pyrolysis process at Stage B.

In Stage A (450 °C), the primary pyrolysis yielded liquid products as the dominant fraction across all feedstocks, with values ranging from 74.0% to 81.96% as shown in Fig. 5. HDPE exhibited the highest liquid recovery (81.96%) with minimal char residue (0.26%), consistent with its linear structure that promotes β-scission and the generation of long-chain hydrocarbons while suppressing condensation to solids. In contrast, PP produced a lower liquid yield (76.64%) alongside the highest char fraction (9.8%), a reflection of its random scission pathway and the high reactivity of tertiary carbons that favor cyclization and coke formation. Binary blends displayed intermediate behaviors. The 50% PP–50% HDPE mixture produced 77.4% liquid and 7.2% char, while the 70% PP–30% HDPE blend yielded 78.68% liquid with reduced char (5.0%), indicating partial mitigation of PP's char tendency by HDPE. Conversely, the 30% PP–70% HDPE mixture delivered 74% liquid but an elevated char fraction (9.0%), resembling the PP-rich degradation route. These observations highlight that co-pyrolysis outcomes are not linear with respect to composition; rather, HDPE can either suppress or amplify PP's char-forming tendency depending on the relative blend ratio.

In Stage B (500 °C), secondary pyrolysis of the wax fraction led to near-complete elimination of char across all feedstocks, with enhanced liquid recovery as shown in Fig. 6. Pure HDPE achieved the highest liquid fraction (88.24%) with only 11.76% gases, confirming its propensity to produce fuel-range hydrocarbons at elevated temperatures with minimal solid residue. PP, by contrast, yielded 75.06% liquid and 24.93% gases, signifying its greater tendency toward gas evolution during secondary cracking. Binary blends exhibited more complex and sometimes non-linear product distributions. The 50% PP–50% HDPE mixture generated the lowest liquid fraction (66.77%) and the highest gas fraction (33.22%), implying antagonistic interactions between PP and HDPE pathways, where PP-driven radical recombination enhances gas evolution while suppressing liquid stability. In contrast, asymmetric mixtures demonstrated clear synergistic benefits. The 30% PP–70% HDPE blend achieved 89.32% liquid yield with only 10.77% gases, outperforming the individual polymers, while the 70% PP–30% HDPE blend delivered the highest overall liquid recovery (95.07%) and the lowest gas fraction (4.92%). These results underscore that HDPE not only suppresses char formation but, when combined with PP, can enhance liquid selectivity far beyond the individual contributions of each polymer.

Overall, the staged thermal pyrolysis strategy demonstrates distinct roles of feedstock composition in shaping product distributions. HDPE is intrinsically favorable for liquid recovery with minimal char, while PP is more prone to gasification and coke formation. However, co-pyrolysis enables tuning of product yields, where asymmetric PP/HDPE mixtures, particularly PP-rich blends, exhibit strong synergistic effects that maximize liquid oil recovery while virtually eliminating char. These insights emphasize that feedstock blending and staged operation are critical levers for optimizing pyrolysis toward high liquid yields and reduced solid residues, advancing the viability of waste polyolefins as feedstocks for sustainable fuel-range hydrocarbon production.

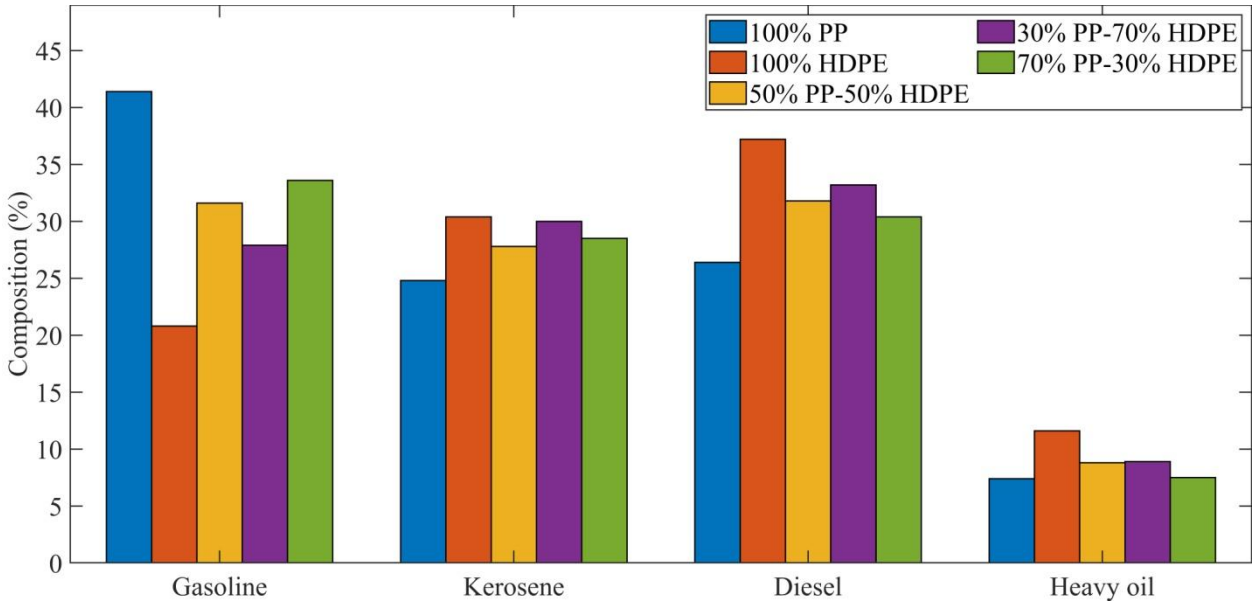

Fig. 7. Fuel-range hydrocarbon distribution of liquid products from the co-pyrolysis process of PP, HDPE and their blends by GC–FID analysis.

The compositional analysis of the liquid fractions obtained from catalytic staged pyrolysis of PP, HDPE, and their mixtures provides clear evidence of how feedstock identity governs the distribution of fuel-range hydrocarbons as shown in Fig.7. Pure PP produced the highest proportion of gasoline (41.4%), consistent with its random chain scission

mechanism that favors the formation of lighter hydrocarbons. Its kerosene (24.8%) and diesel (26.4%) fractions were comparatively lower, while heavy oil was minimal (7.4%). In contrast, HDPE exhibited a markedly different profile, with gasoline yield reduced to 20.8% and heavier fractions dominating—diesel (37.2%) and kerosene (30.4%)—reflecting the influence of its linear structure and β-scission pathway that promote long-chain hydrocarbon formation.

Co-pyrolysis of PP and HDPE yielded intermediate distributions, highlighting synergistic and competitive interactions between the two degradation mechanisms. The 50:50 PP–HDPE blend produced a balanced profile with gasoline (31.6%), kerosene (27.8%), and diesel (31.8%), suggesting that mutual moderation of PP's light product tendency and HDPE's heavy product bias can yield a versatile fuel spectrum. HDPE-rich mixtures (70% HDPE–30% PP) generated higher diesel content (33.2%) and kerosene (30.0%), whereas PP-rich mixtures (70% PP–30% HDPE) shifted the spectrum toward gasoline (33.6%) at the expense of heavier fractions.

Overall, these results demonstrate that PP strongly contributes to gasoline-range hydrocarbons, which are of high economic value for spark-ignition engines, while HDPE favors kerosene and diesel fractions suitable for compression-ignition engines and industrial fuel applications. Importantly, blending the two polymers provides a practical strategy for tuning the hydrocarbon profile: HDPE-rich blends enhance middle-distillate yields, while PP-rich blends increase light-end fuel selectivity. Such tunability underscores the flexibility of staged pyrolysis as a platform for tailoring fuel-range products from mixed plastic waste streams.

CONCLUSION

This study demonstrated that staged thermal pyrolysis of polypropylene (PP), high-density polyethylene (HDPE), and their blends provides an effective pathway for valorizing waste plastics into liquid fuels with tunable product distributions. HDPE consistently exhibited superior performance, yielding the highest liquid fractions with minimal char formation due to its linear structure and β-scission pathway. By contrast, PP was more prone to gas and char generation, although secondary cracking at elevated temperatures significantly improved its liquid yield. Importantly, co-pyrolysis revealed strong synergistic effects: while symmetric blends such as 50:50 PP–HDPE exhibited antagonistic interactions that increased gas evolution, asymmetric mixtures; particularly 70% PP–30% HDPE and 30% PP–70% HDPE, maximized liquid oil recovery while virtually eliminating char.

Fuel-range hydrocarbon analysis confirmed that PP-rich systems favor gasoline-like fractions, whereas HDPE enriches kerosene and diesel products, offering flexibility in tailoring fuels for spark-ignition and compression-ignition applications. Among all formulations, the 70% HDPE–30% PP blend provided the most balanced profile, combining high liquid recovery with favorable middle-distillate yields.

Overall, the findings highlight that feedstock composition and staged operation are critical levers for optimizing pyrolysis outcomes. The ability to fine-tune hydrocarbon distributions through PP/HDPE blending advances the potential of pyrolysis as a sustainable waste management strategy and a contributor to circular plastic economies. Future work should explore catalyst-assisted staged pyrolysis and process intensification strategies, such as induction heating and reactor design optimization, to further enhance selectivity, energy efficiency, and scalability toward industrial implementation.

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