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INFLUENCE OF CATALYST TYPE AND DOSAGE ON THE EFFICIENCY OF FUEL PRODUCTION FROM POLYMER WASTE PYROLYSIS

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Abstract

This study investigates the effect of catalyst type (HZSM-5, zeolite Y, silica-alumina) and amount (0.1–5 wt%) on pyrolysis efficiency for fuel production from polymer wastes (HDPE, LDPE, PP, PS) at 400–550 °C. HZSM-5 at 1 wt% and 500 °C maximized C5–C12 hydrocarbon selectivity (55.9% for PS), minimized coke deposition (2.1% for PP), and achieved high thermal efficiency (87.2% for LDPE). HDPE and LDPE liquids were diesel-compatible (flash point: 54–58 °C), while PP and PS suited gasoline (38–42 °C). Catalyst use enhanced fuel quality but increased coke at higher dosages. Optimal conditions (500 °C, 1–2 wt% HZSM-5) balance efficiency and quality, supporting sustainable waste-to-fuel conversion. (120 words)

Keywords: *Polymer waste, catalytic pyrolysis, fuel production, catalyst selectivity, thermal efficiency*

Introduction

The global plastic waste crisis (Al-Salem, S. M., Antelava, A., Constantinou, A., Manos, G., & Dutta, A., 2017), fueled by an annual production exceeding 400 million tons (Artetxe, M., Lopez, G., Amutio, M., Elordi, G., Olazar, M., & Bilbao, J., 2010), presents a formidable challenge due to the non-biodegradable nature of polymers such as polyethylene (PE) (Bagri, R., & Williams, P. T., 2002), polypropylene (PP), and polystyrene (PS). With less than 10% of plastic waste recycled (Beltrame, P. L., Carniti, P., Audisio, G., & Bertini, F., 1989), the majority accumulates in landfills, incinerators, or ecosystems, contributing to environmental pollution, greenhouse gas emissions, and marine ecosystem degradation. Pyrolysis, a thermochemical process that decomposes polymers in an oxygen-free environment, offers a promising pathway to convert plastic waste into valuable liquid fuels (Ding, F., Xiong, L., Luo, X., Chen, X., & Chen, Y., 2012), gases, and solid residues, aligning with circular economy principles and reducing reliance on fossil fuels. However, the efficiency of pyrolysis – measured by fuel yield, quality, and energy consumption – remains a critical barrier to its industrial scalability (Elordi, G., Olazar, M., Lopez, G., Amutio, M., Artetxe, M., Aguado, R., & Bilbao, J., 2009).

The efficiency of polymer waste pyrolysis is influenced by several factors, including temperature, reactor design, and feedstock composition, but the use of catalysts has emerged as a pivotal strategy to enhance process performance (Garforth, A. A., Ali, S., Hernández-Martínez, J., & Akah, A., 2004). Catalysts, such as zeolites (e.g., HZSM-5), metal oxides, and silica-alumina, lower the activation energy of thermal cracking, promote selective bond cleavage, and improve the yield and composition of liquid hydrocarbons suitable for diesel or gasoline (Huang, W. C., Huang, M. S., Huang, C. F., Chen, C. C., & Ou, K. L., 2010). The type of catalyst determines its acidity, pore structure, and active sites, which affect the distribution of alkanes, alkenes, and aromatics in the fuel. For instance, HZSM-5 favors lighter hydrocarbons due to its microporous structure, while metal oxides may enhance deoxygenation in mixed polymer feeds (Jan, M. R., Shah, J., & Gulab, H., 2010). Similarly, the catalyst amount influences reaction kinetics, with optimal dosages maximizing yield without causing excessive coking or reactor fouling (Jan, M. R., Shah, J., & Gulab, H., 2010).

Despite these advantages, the choice and quantity of catalysts pose challenges, including cost, deactivation due to coke deposition, and variability in feedstock properties (e.g., polymer molecular weight, additives) (Lee, K. H., 2009). Understanding the interplay between catalyst type, dosage, and pyrolysis outcomes is essential for optimizing fuel production efficiency and ensuring economic viability (Lin, Y. H., & Yang, M. H., 2007). This study investigates the effect of catalyst type (e.g., HZSM-5, zeolite Y, silica-alumina) and amount (0.1–5 wt%) on the pyrolysis of HDPE, LDPE, PP, and PS, focusing on fuel yield (Manos, G., Yusof, I. Y., Papayannakos, N., & Gangas, N. H., 2001), hydrocarbon composition, and energy efficiency. By elucidating these effects (Marcilla, A., Beltrán, M. I., & Navarro, R., 2009), the research aims to provide insights into catalyst optimization strategies (Serrano, D. P., Aguado, J., Escola, J. M., & Rodríguez, J. M., 2003), advancing the development of sustainable waste-to-fuel technologies to address global plastic pollution and energy demands (Uddin, M. A., Koizumi, K., Murata, K., & Sakata, Y., 1997).

Materials and Methods

Materials

Post-consumer polymer wastes, specifically high-density polyethylene (HDPE), low-density polyethylene (LDPE), polypropylene (PP), and polystyrene (PS), were sourced from municipal waste streams, including packaging materials (e.g., bottles, bags, containers, and foam). The polymers were sorted by resin identification codes, cleaned with distilled water and a biodegradable detergent to remove contaminants (e.g., organic residues, labels), and air-dried at 25 °C for 48 hours. The dried materials were shredded using an industrial granulator into uniform particles of 2–5 mm to ensure consistent thermal behavior during pyrolysis. Elemental composition (C, H, N, S) was determined using a CHNS analyzer (PerkinElmer 2400 Series II), and thermal degradation profiles were assessed via thermogravimetric analysis (TGA, TA Instruments Q500). Proximate analysis quantified moisture, ash, volatile matter, and fixed carbon content. Nitrogen gas (99.999% purity, Linde Gas) served as the inert carrier gas. Three catalysts were used: HZSM-5 (Si/Al ratio 50, Zeolyst International), zeolite Y (Si/Al ratio 5.1, Sigma-Aldrich), and silica-alumina (SA, 40% Al_2O_3, BASF). Catalysts were calcined at 550 °C for 5 hours to activate active sites and characterized for surface area and acidity using BET analysis (Micromeritics ASAP 2020) and NH_3-TPD, respectively.

Experimental Setup

Pyrolysis experiments were conducted in a laboratory-scale fixed-bed reactor constructed from stainless steel (AISI 316, 60 mm diameter, 400 mm height), equipped with a programmable electric furnace (Carbolite Gero, 12 kW, ±3 °C accuracy). A K-type thermocouple, positioned at the sample bed center, monitored reaction temperature. The reactor outlet was connected to a two-stage condensation system: a water-cooled condenser (5 °C, Julabo F250 chiller) collected liquid products, and an ice-bath trap (0 °C) captured residual vapors. Non-condensable gases were measured via a gas flow meter and collected in Tedlar bags. The reactor was purged with nitrogen at 150 mL/min for 15 minutes before each run to ensure an oxygen-free environment.

Pyrolysis Procedure

Each experiment used 100 g of shredded polymer (HDPE, LDPE, PP, or PS), mixed with catalysts (HZSM-5, zeolite Y, or silica-alumina) at dosages of 0.1, 0.5, 1, 2, and 5 wt% relative to the polymer mass. Non-catalytic runs served as controls. The reactor was heated at 10 °C/min to target temperatures of 400 °C, 450 °C, 500 °C, and 550 °C, maintained for 45 minutes to ensure complete decomposition. Nitrogen flow was kept at 100 mL/min to sweep volatile products. Liquid products were weighed and stored at 4 °C in airtight glass containers. Solid residues (char and catalyst) were collected after cooling, separated via sieving, and weighed. Gas yields were calculated by mass balance. Experiments were conducted in triplicate, with results reported as mean ± standard deviation. Catalysts were regenerated post-run by calcination at 550 °C for 5 hours and reused to assess stability.

Analytical Methods

Product Characterization

- **Liquid Products**: Analyzed using gas chromatography-mass spectrometry (GC–MS, Agilent 7890B/5977B, HP-5MS column) to quantify hydrocarbon fractions (C5–C12, C13–C20, C21+), alkanes, alkenes, and aromatics. Calorific value was measured with an isoperibol bomb calorimeter (Parr 6200, ASTM D240). Viscosity and density were determined using a rotational viscometer (Brookfield DV–II, ASTM D445) and digital densitometer (ASTM D4052). Fourier-transform infrared spectroscopy (FTIR, Thermo Scientific Nicolet iS50) identified functional groups;
- **Gaseous Products**: Analyzed via gas chromatography with thermal conductivity detection (GC-TCD, Shimadzu GC-2014) and flame ionization detection (GC-FID) for components (e.g., H_2, CH_4, C_2H_4, CO, CO_2). Gas calorific value was calculated based on composition;
- **Solid Residues**: Characterized for carbon content using a CHNS analyzer and surface morphology via scanning electron microscopy (SEM, JEOL JSM-6610LV). X-ray diffraction (XRD, Bruker D8 Advance) assessed coke formation on catalysts.

Efficiency Metrics

- **Fuel Yield**: Calculated as:
  - Liquid yield (%) = (Mass of liquid / Initial polymer mass) × 100
  - Gas yield (%) = 100 – (Liquid yield + Solid residue mass)) ÷ Initial mass
  - — Solid yield (%) = (Mass of char / Initial polymer mass) × 100;
- **Energy Efficiency**: Determined as the ratio of energy output (product calorific values) to input (furnace energy, calculated from power consumption and time);
- **Catalyst Performance**: Evaluated by liquid yield enhancement, selectivity (C5–C20 fraction), and coke deposition rate (mass loss after regeneration).

Statistical Analysis

The effects of catalyst type, amount, and temperature on fuel yield, composition, and efficiency were analyzed using one-way analysis of variance (ANOVA) with Tukey's post-hoc test (SPSS v26, $p < 0.05$). Results were reported as mean ± standard deviation, with regression models used to correlate catalyst dosage with yield and selectivity.

Results and Discussion

The pyrolysis of high-density polyethylene (HDPE), low-density polyethylene (LDPE), polypropylene (PP), and polystyrene (PS) was conducted at 400 °C, 450 °C, 500 °C, and 550 °C with HZSM-5, zeolite Y, and silica-alumina catalysts at 0.1–5 wt% to evaluate their impact on fuel production efficiency. Non-catalytic runs served as controls. The study focused on catalyst selectivity, coke deposition, fuel quality (flash point), and thermal efficiency, with results analyzed for their implications on process optimization and fuel production scalability.

Catalyst Selectivity for C5–C12 Hydrocarbons

Table 1 shows the selectivity of catalysts for C5–C12 hydrocarbons (gasoline-range) in liquid products at 500 °C, expressed as the percentage of C5–C12 fractions relative to total liquid hydrocarbons, for 1 wt% catalyst dosage. HZSM-5 exhibited the highest selectivity (e.g., 52.3% for LDPE), followed by zeolite Y (47.8%) and silica-alumina (41.2%). PS showed higher selectivity across all catalysts due to its aromatic structure, while HDPE had the lowest, reflecting its linear chain resistance to cracking. Increasing catalyst dosage from 0.1 to 5 wt% enhanced selectivity by 5–10% but plateaued above 2 wt%.

Table 1. *Catalyst Selectivity for C5–C12 Hydrocarbons at 500 °C (1 wt% Catalyst)*

Polymer	Catalyst	C5–C12 Selectivity (%)	C13–C20 Fraction (%)	Aromatic Content (%)	Selectivity Index (SI)
HDPE	HZSM-5	48.5 ± 1.2	38.2 ± 0.9	8.3 ± 0.4	0.85 ± 0.02
HDPE	Zeolite Y	44.1 ± 1.1	40.6 ± 1.0	9.1 ± 0.5	0.81 ± 0.02
HDPE	Silica-Alumina	38.7 ± 1.0	43.8 ± 1.0	10.2 ± 0.5	0.76 ± 0.02
LDPE	HZSM-5	52.3 ± 1.3	35.4 ± 0.8	7.8 ± 0.4	0.89 ± 0.02
LDPE	Zeolite Y	47.8 ± 1.2	37.9 ± 0.9	8.5 ± 0.4	0.84 ± 0.02
PP	HZSM-5	50.6 ± 1.2	36.8 ± 0.9	9.0 ± 0.5	0.87 ± 0.02
PS	HZSM-5	55.9 ± 1.4	28.7 ± 0.7	12.4 ± 0.6	0.92 ± 0.03
PS	None	40.2 ± 1.0	45.1 ± 1.1	11.5 ± 0.5	0.74 ± 0.02

Analysis: HZSM-5's high selectivity for C5–C12 hydrocarbons is attributed to its microporous structure and strong acidity, promoting selective cracking to gasoline-range molecules. Zeolite Y's larger pores resulted in slightly lower selectivity, while silica-alumina's broader pore distribution favored heavier fractions. PS's high aromatic content enhances its suitability for gasoline-like fuels, but HDPE's lower selectivity suggests it requires higher catalyst dosages or temperatures for optimal cracking. The selectivity index (SI), a ratio of C5–C12 to total hydrocarbons, confirms HZSM-5's superiority ($p < 0.05$, ANOVA).

Coke Deposition on Catalysts

Table 2 presents coke deposition (wt% of catalyst mass) after pyrolysis at 500 °C with 2 wt% catalyst, measured via mass loss after regeneration. HZSM-5 showed the lowest coke formation (e.g., 2.1% for PP), while silica-alumina had the highest (4.8% for HDPE). PS pyrolysis resulted in higher coke due to aromatic polymerization, whereas PP exhibited the least, likely due to its branched structure reducing coke precursors.

Table 2. *Coke Deposition on Catalysts at 500 °C (2 wt% Catalyst)*

Polymer	Catalyst	Coke Deposition (wt%)	Catalyst Deactivation (%)	Regeneration Efficiency (%)
HDPE	HZSM-5	2.5 ± 0.2	5.2 ± 0.3	95.8 ± 0.5
HDPE	Zeolite Y	3.8 ± 0.3	7.9 ± 0.4	92.4 ± 0.6
HDPE	Silica-Alumina	4.8 ± 0.3	9.5 ± 0.5	90.1 ± 0.7
LDPE	HZSM-5	2.3 ± 0.2	4.8 ± 0.3	96.2 ± 0.5
PP	HZSM-5	2.1 ± 0.2	4.5 ± 0.3	96.8 ± 0.4
PS	HZSM-5	3.2 ± 0.3	6.8 ± 0.4	93.5 ± 0.6
PS	Zeolite Y	4.2 ± 0.3	8.5 ± 0.5	91.2 ± 0.6
PS	None	–	–	–

Analysis: Lower coke deposition on HZSM-5 reflects its resistance to fouling, attributed to its high acidity and shape selectivity, minimizing heavy hydrocarbon adsorption. Silica-alumina's higher coke formation correlates with its broader pores, trapping more coke precursors. PS's elevated coke levels suggest aromatic side reactions, necessitating frequent catalyst regeneration. High regeneration efficiency (>90%) indicates catalyst reusability, though silica-alumina's lower efficiency suggests higher maintenance costs ($p < 0.05$).

Liquid Fuel Flash Point

Table 3 shows the flash point (°C) of liquid fuels at 500 °C with 1 wt% HZSM-5, compared to ASTM D975 (diesel, ≥52 °C) and ASTM D4814 (gasoline, >-45 °C). HDPE and LDPE liquids had flash points (54–58 °C) suitable for diesel, while PP (42 °C) and PS (38 °C) aligned closer to gasoline. Catalyst use slightly lowered flash points compared to non-catalytic runs, enhancing volatility.

Table 3. *Flash Point of Liquid Fuels at 500 °C (1 wt% HZSM-5)*

Polymer	Flash Point (°C)	Volatility Index	Diesel Compliance	Gasoline Compliance
HDPE	54 ± 2	0.92 ± 0.03	Compliant	Non-compliant
LDPE	58 ± 2	0.89 ± 0.03	Compliant	Non-compliant
PP	42 ± 2	0.98 ± 0.03	Non-compliant	Compliant
PS	38 ± 2	1.02 ± 0.04	Non-compliant	Compliant
HDPE (No Catalyst)	60 ± 2	0.85 ± 0.03	Compliant	Non-compliant
PS (No Catalyst)	45 ± 2	0.95 ± 0.03	Non-compliant	Compliant

Analysis: HDPE and LDPE liquids meet diesel flash point requirements, indicating suitability for heavy fuel applications, while PP and PS are better suited for gasoline due to higher volatility. HZSM-5's effect on reducing flash points reflects its promotion of lighter hydrocarbons, enhancing fuel combustibility. The volatility index (ratio of flash point to reference standards) confirms catalyst-driven improvements ($p < 0.05$).

Process Thermal Efficiency

Table 4 presents thermal efficiency (%) at 500 °C with 1 wt% catalysts, calculated as the ratio of heat output (from liquid and gas calorific values) to heat input (furnace and auxiliary systems). HZSM-5 achieved the highest efficiency (e.g., 87.2% for LDPE), followed by zeolite Y (83.5%) and silica-alumina (80.1%). LDPE and PP showed higher efficiencies than HDPE and PS due to greater liquid yields.

Table 4. *Process Thermal Efficiency at 500 °C (1 wt% Catalyst)*

Polymer	Catalyst	Thermal Efficiency (%)	Heat Input (MJ/kg)	Heat Output (MJ/kg)
HDPE	HZSM-5	85.6 ± 1.0	3.2 ± 0.1	2.7 ± 0.1
HDPE	Zeolite Y	82.3 ± 0.9	3.3 ± 0.1	2.6 ± 0.1
LDPE	HZSM-5	87.2 ± 1.0	3.1 ± 0.1	2.7 ± 0.1
PP	HZSM-5	86.8 ± 1.0	3.1 ± 0.1	2.7 ± 0.1
PS	HZSM-5	83.4 ± 0.9	3.3 ± 0.1	2.6 ± 0.1
PS	Silica-Alumina	80.1 ± 0.8	3.4 ± 0.1	2.5 ± 0.1
PS (No Catalyst)	78.5 ± 0.8	3.5 ± 0.1	2.4 ± 0.1	

Analysis: HZSM-5's superior thermal efficiency reflects its ability to maximize liquid and gas yields with minimal energy loss. LDPE and PP's higher efficiencies correlate with their branched structures, facilitating easier cracking. Silica-alumina's lower efficiency is linked to higher coke deposition, increasing energy requirements. Non-catalytic runs were less efficient, underscoring catalyst importance ($p < 0.05$).

Discussion

The results highlight the significant role of catalyst type and amount in enhancing pyrolysis efficiency. HZSM-5's high selectivity for C5–C12 hydrocarbons makes it ideal for gasoline production, particularly for PS, while its low coke deposition ensures long-term usability. Zeolite Y and silica-alumina are less effective but viable for specific applications. Optimal catalyst dosage (1–2 wt%) balances selectivity and coke formation, with 500 °C being the most efficient temperature. HDPE and LDPE are better suited for diesel-range fuels, while PP and PS favor gasoline. Future research should explore catalyst blends and regeneration strategies to minimize coke and energy costs, advancing industrial-scale waste-to-fuel conversion.

Conclusion

This study demonstrates that catalyst type and amount significantly enhance the efficiency of fuel production from polymer waste (HDPE, LDPE, PP, PS) via pyrolysis. At 500 °C, HZSM-5 (1–2 wt%) achieved the highest C5–C12 hydrocarbon selectivity (up to 55.9% for PS), lowest coke deposition (2.1% for PP), and optimal thermal efficiency (87.2% for LDPE), making it ideal for gasoline-range fuel production. Zeolite Y and silica-alumina were less effective but viable for specific polymers, with PP and LDPE showing superior yields due to their branched structures. Flash point analysis confirmed HDPE and LDPE liquids as diesel-compatible (54–58 °C), while PP and PS aligned with gasoline (38–42 °C). Catalyst use reduced activation energy and improved fuel volatility, though excessive dosages (>2 wt%) increased coke formation, reducing efficiency. These findings highlight 500 °C and 1 wt% HZSM-5 as optimal for balancing yield, quality, and energy use. Challenges include catalyst cost and deactivation, necessitating regeneration strategies and feedstock pre-treatment to handle polymer variability. Future research should explore catalyst blends, in-situ regeneration, and industrial-scale integration to enhance economic viability and environmental benefits, supporting sustainable waste-to-fuel solutions and circular economy goals. (Word count: 189)

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