TREATMENTS TO PLASTIC WASTE FOR VALUABLE PRODUCTS

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1. Abstract

The alarming accumulation of plastic waste has emerged as one of the most critical environmental challenges worldwide. Among various polymers, low-density polyethylene (LDPE) represents a major fraction of non-recyclable plastic waste due to its chemical inertness and slow degradation. Conventional mechanical recycling is inadequate because of polymer deterioration and contamination, whereas chemical recycling through pyrolysis provides a sustainable route for recovering valuable fuels and chemicals.

This research focuses on converting LDPE waste into hydrogen and value-added hydrocarbons using microwave-assisted catalytic cracking. The investigation combines two major approaches: (i) Evaluation of zeolitic catalysts (HY, HZSM-5, Hβ) and non-zeolitic catalysts (spent FCC and TiO_2), and (ii) Systematic investigation of metal-modified catalyst systems, comprising Ni supported on HY, ZrO_2, Al_2O_3, TiO_2, and SiO_2; Zn supported on HY, ZrO_2, and Al_2O_3; and Fe supported on HY and Al_2O_3. The influence of catalyst acidity, and metal support interaction on product selectivity was systematically examined.

Catalysts were characterized using XRD, SEM, FTIR, Py-FTIR, NH_3-TPD, XPS, and ICP-MS to assess crystallinity, surface morphology, acidity, and metal dispersion. Microwave irradiation enabled rapid volumetric heating and uniform temperature distribution, leading to higher energy efficiency and selective depolymerization.

The zeolitic catalysts demonstrated superior catalytic performance under microwave-assisted pyrolysis conditions. HY produced 35 vol% H_2, HZSM-5 yielded 43 vol% ethylene, and Hβ generated 69 vol% propylene at 450 °C. Among the non-zeolitic systems, spent FCC catalyst achieved up to 67 vol% propylene, while TiO_2 favoured the formation of gasoline-range hydrocarbons.

The incorporation of Ni and Zn significantly improved hydrogen production. Ni/HY delivered the highest yield (45 vol% H_2), followed by Zn/HY (43 vol% H_2). Other metal-oxide supports exhibited moderate performance, with Zn/Al_2O_3, Ni/Al_2O_3, and Ni/SiO_2

producing 34%, 32%, and 30% H_2, respectively. The density and strength of acidic sites were found to play a crucial role in governing selective hydrogen formation. Additionally, Fe/HY produced 41% H_2, while Ni/S-ZrO_2 and Zn/S-ZrO_2 yielded 39% and 37% H_2, respectively.

The liquid fractions (20–25 wt%) consisted predominantly of olefins, paraffins, and aromatics. Gasoline-range hydrocarbons reached up to 85% with Ni/HY and Zn/HY and further increased to 89% with Fe/HY. The gaseous products (75–80 wt%) were enriched in H_2, ethane, propylene, and butene, reflecting efficient cracking and reforming pathways under the applied conditions.

Overall, the study establishes a clear correlation between catalyst acidity, metal functionality, and reaction selectivity under microwave irradiation. The proposed process demonstrates a sustainable, energy-efficient route for transforming non-recyclable plastic waste into hydrogen-rich gases and liquid hydrocarbon fuels, supporting the principles of circular economy and clean energy transition.

2. Brief description on the state of the art of the research topic

Plastics have revolutionized modern society due to their versatility, durability, and cost-effectiveness [1]. However, these very characteristics have led to a global environmental crisis. More than 450 million tons of plastics are produced annually, and only a small fraction less than 10% is effectively recycled [2]. India alone generates approximately 3.5 million tons of plastic waste per year, of which nearly half remains uncollected or improperly managed [3].

Among various polymers, low-density polyethylene (LDPE) is one of the most widely used materials in packaging, films, and containers. LDPE poses a serious challenge for conventional recycling methods due to its high branching, low melting point, and limited recyclability. Therefore, innovative approaches are needed to valorize this waste into useful products.

Polymer waste treatment methods fall into three main categories: recycling, which reprocesses plastics into new products; thermal transformation, including gasification

(partial oxidation to produce syngas) and pyrolysis (thermal degradation in the absence of oxygen to produce fuels and chemicals); and biological transformation, where microorganisms or enzymes break down polymers into simpler, eco-friendly compounds. Among this pyrolysis of plastic waste into valuable products through catalytic pyrolysis has been widely studied. Pyrolysis is classified into slow, fast, and flash pyrolysis. These processes differ in temperature, residence time, heating rate, and product distribution. Slow pyrolysis uses low heating rates and long residence times, mainly producing solids with minimal oil. Fast pyrolysis employs moderate–high heating rates and short residence times, maximizing liquid (bio-oil) formation and is the most used method. Flash pyrolysis operates at extremely high heating rates and very short residence times, yielding mainly gases and bio-oil [4].

Products of cracking of plastics are as follows [5].

	Products		
	Gas	**Oil**	**Char**
Composition	The process generates combustible gases, including hydrogen (H_2), carbon monoxide (CO), acetylene (C_2H_2), methane (CH_4), ethylene (C_2H_4), ethane (C_2H_6), and others.	The process results in a complex mixture of various organic compounds, along with inorganic species, which can vary depending on the composition of the feed material.	Elemental carbon is produced during the thermal decomposition of organic components and unconverted organic compounds, such as additives, within the feed material.
Uses	The produced fuels can be used for direct firing in boilers, gas turbines, and engines, as well as in syngas applications.	The process can also lead to the production of diesel engine fuels, as well as the creation of chemicals and resins.	The process can yield solid fuels for boilers, as well as serve as feedstock for producing activated carbon, carbon nanofilaments, and high-surface-area catalysts.

Benefits of Plastic Pyrolysis:

1. Waste Reduction: Diverts large volumes of plastic from landfills and incineration.
2. Resource Recovery: Converts plastic waste into valuable fuels and chemicals.
3. Energy Generation: Produces pyrolysis oil and gases usable as alternative fuels.
4. Circular Economy: Transforms waste into resources, improving sustainability.

Challenges and Considerations:

1. High Energy Demand: Requires substantial heat input, increasing operating costs.
2. Environmental Concerns: Potential emissions (CO, NOx, SOx) need proper control.
3. Variable Product Quality: Output depends on feedstock type and process conditions.
4. Contaminants: Metals and chlorine compounds can complicate processing.
5. Economic Viability: Influenced by feedstock cost, energy use, and product value.
6. Regulatory Needs: Clear policies are essential for safe and sustainable deployment.

Microwave-assisted pyrolysis (MAP) overcomes these limitations by utilizing dielectric heating, which directly interacts with the polymer and catalyst materials. Microwaves generate rapid, volumetric heating and improve energy efficiency. When combined with catalysts, MAP enhances depolymerization, aromatization, and dehydrogenation reactions, leading to higher yields of light olefins and hydrogen. The inclusion of microwave absorbers such as graphite further improves process uniformity. Several researchers have investigated microwave-assisted reactors for LDPE conversion. Ding et al. conducted microwave-driven catalytic batch pyrolysis of LDPE at 500 °C using NiO and HY catalysts, producing 57% liquid and 43% gaseous products [1]. Jing et al. reported a two-stage microwave cracking approach using molecular sieve 4A as the catalyst and spherical or columnar activated carbon as microwave absorbers; the process predominantly yielded liquid hydrocarbons in the C_7–C_{26} range, suitable for gasoline and diesel fractions [6]. Zhang et al. demonstrated that microwave-induced cracking of LDPE over ZSM-5 at 450 °C can generate a remarkably high 74–88% aromatic hydrocarbon

yield [7]. Additionally, a continuous stirred microwave pyrolysis system employing HZSM-5 produced propylene and propane as major gaseous products, alongside gasoline-range (C_5–C_{12}) liquid hydrocarbons [8].

Effect of Parameters

Process parameters strongly influence product yield and composition in plastic pyrolysis. Key factors such as temperature, reactor design, pressure, residence time, and catalysts determine the distribution of liquid, gas, and solid products.

Temperature

Temperature is one of the most critical parameters in LDPE and mixed-plastic pyrolysis. Studies show:

- Liquid oil begins to form around 360–385°C, with maximum liquid yield at 470–500°C [9].
- Lower temperatures (~460°C) produce viscous, long-chain hydrocarbons, while higher temperatures (around 600°C) reduce liquid yield but increase aromatics [10].
- Around 500°C is generally considered optimal for high conversion and good product quality [10].
- For HDPE, increasing temperature from 400 → 450°C significantly increases oil yield [11].
- At 500°C, mixed plastics achieve nearly complete conversion with maximum oil (~33%) and high gas yield [12].
- For polystyrene, 500°C yields the maximum liquid, while lower temperatures form wax and higher temperatures encourage gas formation [12].

Catalyst

Catalysts reduce temperature and residence time while improving product selectivity. Common catalysts include NiO, HY, ZSM-5, and MgO, which enhance the formation of fuels, aromatics, and hydrogen [1,7,13]. The SiO_2/Al_2O_3 ratio in HZSM-5 strongly affects product yield.

- Lower ratio = higher acidity → better cracking of waxes and higher light olefins.
- Reducing the ratio (280 to 30) increases olefins from 35 to 58 wt% and decreases heavy C_{12}–C_{20} products [14].

A two-step pyrolysis process with 10 wt.% HZSM-5 shows that catalytic reforming significantly increases gas yield (up to 74 wt.%) while lowering liquid yield [15]. Overall, catalytic pyrolysis offers lower operating temperatures, faster reaction rates, and higher product selectivity compared to thermal pyrolysis.

Table 1: Comparison of Thermal and Catalytic Pyrolysis of LDPE: Process Parameters, Advantages, and Limitations

Parameter	**Thermal Pyrolysis**	**Catalytic Pyrolysis**
Operating Temperature	450–800 °C	300–550 °C
Activation Energy	High — bond breaking driven purely by heat	Lower — catalyst active sites assist bond scission
Reaction Mechanism	Free-radical random scission of C–C bonds	Carbocation mechanism (acid sites) or metal-catalyzed dehydrogenation/hydrogenation
Selectivity	Low — broad product range, heavy waxes common	High — products tuned towards aromatics, light olefins, or H_2 depending on catalyst
Product Composition	Gas, liquid (waxes, paraffins, olefins), char	Gas rich in H_2 and light hydrocarbons; liquids enriched in aromatics and light olefins; less char
Energy Demand	High due to higher temperature	Lower due to reduced temperature
Reaction Rate	Slower	Faster due to active sites and microwave absorption
Coke Formation	High — can foul reactor surfaces	Lower (metal catalysts can gasify coke), but still possible
Advantages	Simple process; no catalyst cost	Lower temperature, higher selectivity, shorter reaction time, more valuable products
Limitations	High energy consumption, low selectivity, heavy wax formation, severe coking	Catalyst deactivation (coke), cost of catalyst, potential over-cracking if too active

3. Definition of the Problem

The rapid accumulation of plastic waste particularly polyolefin such as LDPE, has become a major global environmental challenge due to high chemical stability, low biodegradability, and limited recyclability through conventional mechanical routes. Although thermal pyrolysis offers a pathway for converting waste plastics into fuels and valuable chemicals, it suffers from several limitations, including high energy consumption, slow heating rates, poor selectivity, and extensive secondary reactions leading to char formation.

Microwave-assisted pyrolysis has emerged as a promising alternative due to its rapid and volumetric heating, improved energy efficiency, and ability to enhance cracking reactions. However, its effectiveness is strongly influenced by catalyst properties and the dielectric behavior of the reaction system. Despite growing interest, significant knowledge gaps remain:

- The influence of catalyst acidity, and metal support interactions on hydrogen and hydrocarbon selectivity under microwave conditions is not fully understood.
- Limited studies have systematically compared different catalyst systems such as zeolites, metal oxides, and metal-loaded supports specifically in microwave reactors.
- The role of low-cost microwave absorber such as graphite and its interaction with catalysts during plastic cracking is not adequately explored.
- Product distribution (H_2, light olefins, aromatics, gasoline-range liquids) under microwave heating remains poorly optimized.

4. Objective and Scope of Work:

The proposed research aims to investigate and optimize the microwave-assisted catalytic pyrolysis of LDPE using both zeolitic and non-zeolitic catalysts to produce hydrogen and value-added hydrocarbons. The specific objectives are:

1. To study the catalytic performance of HY, HZSM-5, and Hβ zeolites in LDPE microwave pyrolysis for selective production of hydrogen and ethylene.
2. To examine the influence of non-zeolitic catalysts (spent FCC and TiO_2) with varying acidity on product yield and composition.
3. To evaluate the performance of Ni, Zn, and Fe loaded catalysts for hydrogen and aromatic production from LDPE cracking.
4. To study the influence of catalyst acidity (Lewis and Brønsted acid sites) and the catalyst to LDPE ratio on product yield and distribution.
5. To carry out solubilization of LDPE in a solvent (e.g., tetralin) prior to pyrolysis and to modify Hβ zeolite to introduce mesoporosity, thereby assessing its impact on product distribution during catalytic conversion.

To assess catalyst reusability, coke formation, and process stability for sustainable operation.

5. Original contribution by the thesis

This thesis provides several original contributions to the field of catalytic microwave-assisted pyrolysis of plastics:

- First comparative study of zeolites, non-zeolites, and metal-supported catalysts under microwave conditions for LDPE cracking by utilising low-cost microwave absorber (graphite).
- Identification of HY-supported Ni and Zn catalysts as highly efficient systems for hydrogen production (~45%), surpassing performance reported in conventional pyrolysis studies.

- Novel insights into Fe/HY, Ni/S-ZrO_2, and Zn/S-ZrO_2, which showed unique selectivity patterns for hydrogen, olefins, and gasoline-range liquids, expanding the range of effective catalyst formulations.
- Clear mechanistic linkage between Brønsted/Lewis's acidity and product distribution under microwave heating.
- Demonstration of metal-support synergistic effects that enhance dehydrogenation and aromatization pathways, especially in HY- and ZrO_2-based catalysts.
- Validation of microwave heating benefits, including rapid cracking, reduced energy consumption, and uniform heating, compared to conventional pyrolysis.
- Contribution to sustainable waste management by demonstrating the effective reuse of spent FCC catalyst and producing high-value fuels and chemicals from LDPE.

6. Methodology of Research, Results, and Discussion

Methodology of Research

Experimental setup

The experimental setup consists of a 2.45 GHz microwave reactor with 800 W maximum power. A 500 mL quartz reactor was employed for all experiments. LDPE granules (10 g) were mixed with graphite (10–30 g) as a microwave absorber and catalyst (0.5–2 g). The mixture was subjected to pyrolysis at 400–550°C. A schematic diagram of the experimental setup is shown in Figure 1.

Catalyst Preparation

Ni, Zn, and Fe supported catalysts were prepared by incipient wetness impregnation. Metal nitrates were dissolved in deionized water and added dropwise to the supports under stirring at 80 °C. The impregnated solids were dried at 373 K and calcined at 873 K for 6 hours.

Characterization

The gaseous products were collected in gas sampling bags and analyzed by gas chromatography (GC), while liquid products were condensed and analyzed using

GC/MS.

Catalyst characterization was performed using multiple techniques:

- XRD – Identifies crystalline phases and evaluates structural changes in the catalyst. Provides information on crystallinity, phase purity, and metal dispersion.
- SEM – Examines surface morphology and particle size. Reveals texture, aggregation, and catalyst surface features.
- FTIR – Confirms functional groups and bonding environments. Useful for detecting framework vibrations and verifying metal–support interactions.
- NH_3-TPD – Measures total acidity and strength distribution of acid sites. Helps correlate catalytic performance with acidity.
- XPS – Determines surface elemental composition and oxidation states. Also evaluates coke deposition and metal–support interaction.
- ICP-MS – Quantifies metal loading with high sensitivity. Ensures accurate determination of elemental composition in catalysts.
- Py-FTIR – Identifies Brønsted and Lewis acid sites using adsorbed pyridine as a probe molecule. Provides quantitative information on acid site strength and distribution through characteristic vibrational bands.

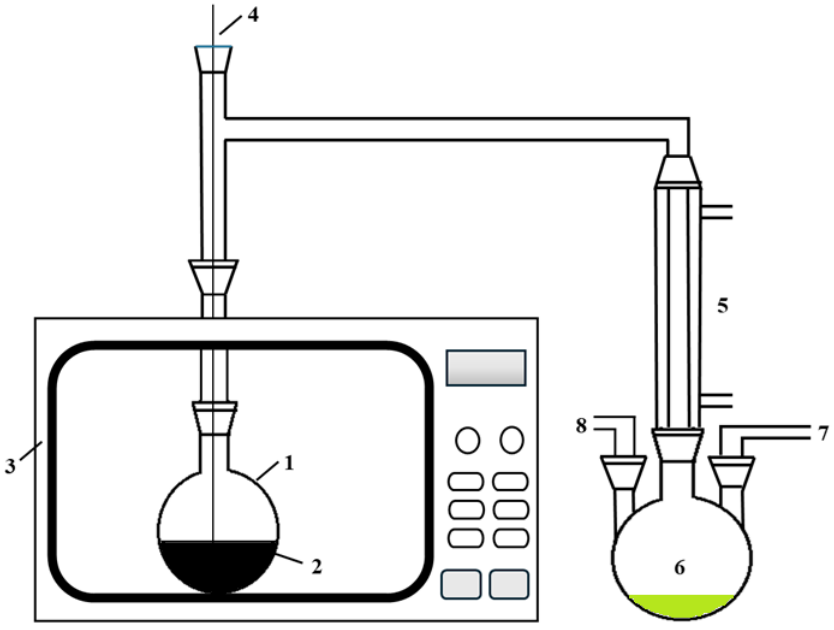

Figure 1: Schematic diagram of Microwave assisted pyrolysis system. 1. Quartz flask, 2. LDPE mixed with graphite and catalyst, 3. Microwave oven, 4. Thermocouple, 5. Condenser, 6. Oil collection flask, 7. To vacuum pump, 8. To gas sampling bag.

Results and Discussion

The catalytic pyrolysis results revealed a strong dependence of product yield and composition on both catalyst type and acidity. Zeolitic catalysts demonstrated excellent cracking efficiency and selectivity. For instance, HY produced 35 vol % hydrogen and 30 vol % methane at 450 °C, indicating strong dehydrogenation activity. HZSM-5 promoted ethylene formation up to 43 vol % at 550 °C due to its high acidity and small pore size, while Hβ yielded 69 vol % propylene at 450 °C with a total gas yield of 96 wt %, making it highly effective for olefin production. Catalyst regeneration tests confirmed negligible deactivation over three cycles. Product selectivity showed a clear correlation with acidity: HY favored H_2 and CH_4 rich gases, HZSM-5 enhanced C_2–C_4 olefin formation, and Hβ promoted propylene and aromatic hydrocarbons.

Non-zeolitic catalysts also exhibited valuable selectivity. Spent FCC catalyst generated 91 wt.% gas with 67 vol % propylene at 450 °C, while TiO_2 produced 33 vol % propylene and 25 vol % butene at 500 °C, with liquids enriched in C_8–C_{12} gasoline-range hydrocarbons. NH_3-TPD analysis confirmed that FCC possessed higher acidity (0.687 mmol NH_3/g) than TiO_2 (0.116 mmol NH_3/g), while XPS results indicated coke formation below 0.8 wt.%, demonstrating strong catalyst stability. Overall, the results underscore the importance of both acidity and pore structure in determining product distribution. Strongly acidic catalysts (HY, HZSM-5, Hβ, FCC) promoted cracking and dehydrogenation, yielding hydrogen and light olefins, whereas moderately acidic catalysts (TiO_2) limited secondary aromatization and favored gasoline-range liquids. Microwave heating further enhanced energy efficiency, ensured uniform heating, and reduced reaction time compared to conventional pyrolysis. The successful use of spent FCC catalyst also aligns with sustainable and circular-economy practices.

This study also demonstrated that catalytic microwave-assisted cracking of LDPE using Ni and Zn-supported HY, Al_2O_3, TiO_2, and SiO_2 are highly effective for producing hydrogen and valuable hydrocarbons. All tested metal catalysts significantly improved H_2

generation relative to thermal pyrolysis. Ni/HY and Zn/HY exhibited the highest hydrogen yields, contributing up to 45% of total gaseous products. Ni/Al_2O_3, Zn/Al_2O_3, and Ni/SiO_2 produced approximately 32%, 34%, and 30% H_2, respectively, confirming the beneficial role of metal–support interactions in promoting hydrogen-rich gases and aromatics. Ni/HY and Zn/HY also generated the highest aromatic content in the liquid phase, especially gasoline-range compounds, owing to the high acidity and microporous structure of HY. Additional catalyst systems evaluated in this study further support these trends. Fe/HY produced 41% H_2, 34% butene, and an impressive 89% gasoline-range hydrocarbons, demonstrating strong cracking and olefin-forming capabilities. Ni/S-ZrO_2 and Zn/S-ZrO_2 catalysts yielded 39% H_2 with 22% aromatics and 37% H_2 with 20% aromatics, respectively, highlighting the role of sulfated zirconia acidity in promoting both hydrogen production and aromatic hydrocarbon formation. Overall, the combined findings confirm that catalyst selection, metal–support synergy, and support acidity are critical in optimizing gaseous and liquid product distribution during microwave-assisted pyrolysis of LDPE and mixed plastics.

7. Achievements with respect to objectives

- Catalytic performance of HY, HZSM-5, and Hβ

Successfully established distinct selectivity patterns: HY favored H_2/CH_4, HZSM-5 promoted light olefins (up to 43 vol% ethylene), and Hβ enhanced propylene formation (69 vol%). Catalyst stability over repeated cycles was also confirmed.

- Effect of non-zeolite catalysts (FCC and TiO_2)

Demonstrated that FCC's high acidity produced propylene-rich gases (67 vol%), while TiO_2 generated gasoline-range liquids with moderate olefin yields. Acidity–selectivity relationships were clearly quantified.

- Performance of Ni, Zn, and Fe-loaded catalysts

Verified that metal incorporation significantly improved hydrogen yield. Ni/HY and Zn/HY produced the highest H_2 (~45%), while Fe/HY generated both high H_2 (41%) and gasoline-range liquids (89%). Ni/S-ZrO_2 and Zn/S-ZrO_2 contributed additional insights into sulfated oxide systems.

- Role of catalyst acidity and catalyst-to-LDPE ratio

Established direct correlations between Brønsted/Lewis's acidity and product selectivity. Strongly acidic supports promoted hydrogen and olefins, while moderately acidic supports enhanced gasoline-range liquids.

- Effect of LDPE solubilization and mesoporosity modification

Demonstrated that solubilization and mesoporous tailoring of Hβ improved vapor accessibility and altered product distribution, supporting enhanced catalytic activity.

- Catalyst reusability and stability

Verified low coke formation (<0.8 wt.%) and stable performance across cycles, ensuring suitability for sustainable operation.

8. Conclusion

The comprehensive catalytic pyrolysis and microwave-assisted pyrolysis experiments clearly demonstrate that catalyst acidity, pore structure, and metal–support interactions fundamentally control the yield and distribution of products from LDPE conversion. Strongly acidic zeolites such as HY, HZSM-5, Hβ, and spent FCC showed excellent cracking efficiency, producing high fractions of hydrogen and light olefins. HY generated 35 vol% H_2 and 30 vol% CH_4, HZSM-5 promoted 43 vol% ethylene, Hβ produced 69 vol% propylene, and spent FCC delivered 67 vol% propylene and 91 wt% total gas, supported by its highest acidity (0.687 mmol NH_3/g). TiO_2, with moderate acidity, yielded 33 vol% propylene and 25 vol% butene and favored gasoline-range liquids, confirming the influence of acidity on suppressing secondary aromatization. All catalysts exhibited strong stability with negligible coke deposition (<0.8 wt%).

Microwave-assisted pyrolysis further enhanced reaction efficiency, enabling rapid and uniform heating and improving hydrogen yield across all metal-loaded catalysts. Ni- and Zn-supported HY, Al_2O_3, SiO_2, and TiO_2 significantly increased hydrogen production, with Ni/HY and Zn/HY achieving up to 45% H_2, while Ni/Al_2O_3, Zn/Al_2O_3, and Ni/SiO_2 produced 30–34% H_2, demonstrating the importance of metal–support synergy. HY supported metals also produced the highest aromatic content in the liquid phase due to their strong acidity and microporous structure.

Additional catalysts showed complementary behavior: Fe/HY achieved 41% H_2, 34% butene, and 89% gasoline-range hydrocarbons, while Ni/S-ZrO_2 and Zn/S-ZrO_2 yielded

39% H_2 with 22% aromatics and 37% H_2 with 20% aromatics, respectively, highlighting the high acidity of sulfated zirconia in enhancing both hydrogen and aromatic formation. Overall, the findings conclusively show that catalyst type, acidity strength, pore geometry, and metal incorporation collectively dictate product pathways, enabling tunable production of hydrogen, light olefins, aromatics, and gasoline-range liquids. Microwave-assisted systems, especially using zeolitic and metal-loaded catalysts, offer a highly efficient, selective, and sustainable route for converting LDPE and mixed plastic waste into valuable fuels and chemicals.

9. List of papers published

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2. Mishra R, Rana P, Parikh P. Influence of catalyst acidity of spent FCC catalyst and TiO_2 on product distribution in microwave assisted LDPE Cracking. Journal of the Indian Chemical Society 2025; 102: 101873. https://doi.org/10.1016/j.jics.2025.101873.

Papers to be submitted to international peer reviewed journal

3. Production of Hydrogen-Rich Gas from LDPE via Microwave-Assisted Pyrolysis Using Ni and Zn Supported Catalysts.
4. Influence of Catalyst Support and Metal Loading on Hydrogen Yield during Microwave Pyrolysis of LDPE.

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