Review Article

High Yield Oil from Catalytic Pyrolysis of Polyethylene Terephthalate Using Natural Zeolite: A Review

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Abstract
Polyethylene terephthalate (PET) waste has emerged as a critical environmental issue due to its widespread use, particularly in food and beverage packaging. Catalytic pyrolysis with natural zeolite presents a promising approach to convert PET into valuable benzene-rich oil, providing a sustainable pathway for plastic waste management. However, significant challenges persist, notably the generation of acidic byproducts such as terephthalic acid, which may lead to reactor blockages and catalyst deactivation. This review explores the role of natural zeolite catalysts in enhancing PET pyrolysis, facilitating the breakdown of PET into shorter-chain hydrocarbons, and improving oil yield and quality. Various modifications of natural zeolite, including activation, acid treatment, and metal impregnation, are assessed for their effects on catalytic performance. The review further examines mechanistic insights into the reaction pathways, including C–C bond scission, decarboxylation, and aromatization, supported by the acidic sites within zeolite. A bibliometric analysis of studies from 2014 to 2024 identifies research trends and existing gaps in PET pyrolysis, underscoring the need for innovative catalyst designs to reduce energy demands and mitigate unwanted byproducts. Recommendations are also provided for optimizing reaction conditions, including temperature, residence time, and catalyst composition, to enable scalable and energy-efficient PET pyrolysis processes. These findings emphasize the significant potential of natural zeolite as a cost-effective catalyst in transforming PET waste into alternative fuels, contributing to both environmental sustainability and advancements in waste-to-energy catalysis.

Keywords: Catalyst, Catalytic pyrolysis, Natural zeolite, Polyethylene terephthalate, Reaction mechanism

1 Introduction

Plastic materials play a critical role in modern industries, including medical devices [1], electronics, automotive manufacturing [2], and packaging [3], due to their lightweight, durable, and versatile nature. Global plastic production surpassed 460 million tons in 2019 and is projected to reach 2 billion tons by 2050, with Asian regions contributing significantly to this increase [4], [5]. Despite their utility, plastics are predominantly non-biodegradable, leading to severe environmental issues as they accumulate in landfills and ecosystems, posing threats to wildlife and human health. Conventional waste management methods, such as recycling, landfilling, and incineration, have limitations in scalability and environmental sustainability. Pyrolysis, a thermochemical process, has emerged as a promising alternative, converting plastic waste into valuable byproducts like liquid fuels, gases, and solid residues [6], [7].

Polyethylene terephthalate (PET), widely used in single-use packaging such as food containers and beverage bottles, poses unique challenges. Studies indicate that up to 95% of PET products are discarded within a year, contributing significantly to the global waste crisis [8]-[10]. Pyrolysis of PET is energy-intensive, requiring high temperatures (450–700 °C) and significant activation energy (~197 kJ/mol), resulting in elevated costs and scalability limitations [9], [10]. Moreover, non-catalytic pyrolysis often produces undesirable byproducts, including acidic compounds like terephthalic and benzoic acid, which can corrode equipment and hinder operational efficiency [11], [12]. Non-catalytic processes also yield higher amounts of gas, char, and wax, with limited production of liquid fuels [13], [14].

To address these issues, catalysis has gained attention for its ability to lower activation energy, enhance product selectivity, and reduce undesirable byproducts. Zeolite-based catalysts, particularly natural zeolites, offer an eco-friendly and cost-effective solution for PET pyrolysis. Natural zeolites are abundant aluminosilicate minerals with global production exceeding 1.1 million tons in 2023, highlighting their industrial significance [15]. Through activation processes like acid treatment or metal impregnation, natural zeolites can improve porosity, acidity, and thermal stability, optimizing catalytic performance [16], [17]. Their Brønsted acid sites facilitate critical reactions such as dehydration and decarboxylation, enhancing the yield and quality of hydrocarbon products [18]–[20].

This study aims to evaluate the catalytic pyrolysis of PET using natural zeolites, focusing on their performance in optimizing liquid fuel yields and enhancing product quality. Further investigation aims to uncover the interaction mechanisms between PET and zeolites, identify factors contributing to catalyst deactivation, and explore the potential of modified zeolites to improve process efficiency. These findings aim to support sustainable waste management practices and foster the development of a circular economy to address the environmental challenges of plastic waste.

2 Methods

2.1 *Searching method and criteria*

This study examines the development of plastic pyrolysis research through bibliometric analysis, utilizing VOSviewer for topic mapping. This approach allows for a systematic overview of the literature in this field, as demonstrated in previous studies [21].

Data Sources: The primary keywords for the literature search were "pyrolysis" and "plastic," which guided the article search within the Scopus database. Scopus was chosen due to its comprehensive coverage, reputable indexing of high-quality journals, and detailed citation analysis, which are essential for conducting bibliometric studies effectively.

Search Strategy: To ensure relevance, the search focused on articles published within the last 10 years, reflecting the most current research trends and findings. The search targeted article titles, abstracts, and keywords, as these fields are most representative of the research themes. This strategy aids in filtering relevant literature while excluding outdated or less relevant studies.

Selection Criteria: The criteria for including articles in the review were strict. Only peer-reviewed articles that fit within the specified scope were selected. Excluded articles comprised those from discontinued coverage journals, non-original research papers, and articles that were not accessible through institutional subscriptions or other means. Clarifying these exclusion criteria is crucial for maintaining the integrity of the review process.

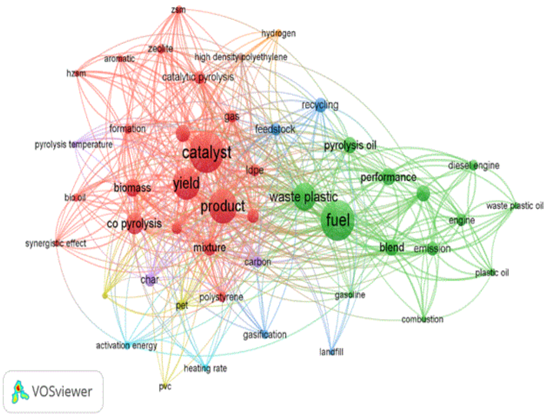


Figure 1: The clusters of the keywords ‘plastic’ and ‘pyrolysis’ contained in the Scopus database in 2014–2024.

The bibliometric analysis performed using VOSviewer is illustrated in Figure 1, which identifies six distinct research clusters: catalyst, fuel, gasification, activation energy, pyrolysis temperature, and hydrogen. The keyword analysis presented in Figure 1 highlights the dominant topics in plastic pyrolysis research, including elastomers, plastic recycling, catalysts, waste incineration, and co-pyrolysis. Notably, plastic recycling continues to be a significant method of managing plastic waste. In the realm of plastic pyrolysis, research on catalysts has gained more prominence than areas such as product pyrolysis, bio-oil, biofuel, plastic oil, gasoline, and diesel. Frequently studied catalysts include zeolite, ZSM, and HZSM. The term “plastic pyrolysis” is commonly linked to feedstocks such as polypropylene, PET, polystyrene, HDPE, and biomass, reflecting ongoing interests in co-pyrolysis. The cluster analysis indicates that keywords like “zeolite” and “pyrolysis oil” belong to separate clusters, suggesting a need for more integrated research in these areas.

Data Visualization: VOSviewer was utilized to visualize the development trends of the keywords “pyrolysis” and “plastic,” as well as to analyze frequently cited authors in the field. The visual outputs included network and overlay mappings, which help illustrate the connections between different research themes and highlight emerging trends.

2.2 *Screening for eligibility*

Figure 2 illustrates the systematic review process, detailing the number of articles identified, screened, and included in the analysis. The PRISMA flowchart effectively visualizes the steps taken throughout this process.

1) Identification

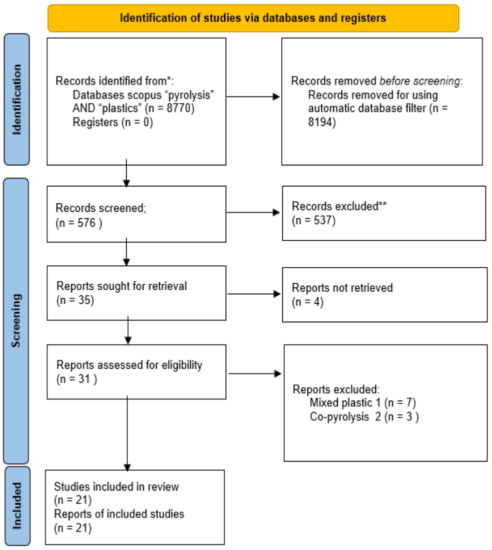
The review process identified a total of 8,770 records from the Scopus database using the search terms “pyrolysis” and “plastics.” Notably, no records were found in any registers. An automatic database filter removed 8,194 irrelevant or duplicate records, resulting in a refined dataset for evaluation.

2) Screening

In the screening phase, the remaining 576 records were carefully reviewed for relevance. Of these, 537 records were excluded for not meeting the established inclusion criteria, which focused on the relevance of the studies to plastic pyrolysis and their methodological rigor. Subsequently, 35 reports were selected for retrieval, although four of these could not be accessed due to potential accessibility issues, which may include subscription barriers or publication restrictions. Ultimately, 31 reports were assessed for eligibility based on the predefined inclusion and exclusion criteria.

3) Included

The review resulted in the exclusion of specific reports for distinct reasons. Seven reports were excluded due to the focus on mixed plastics, while three were excluded for addressing co-pyrolysis, which did not align with the review's primary focus. Consequently, 21 studies were included in the review, representing those reports that fulfilled all inclusion criteria and were relevant to the study objectives, as presented in Table 1.



n: represents the number of studies at each phase of the systematic literature review process.

Figure 2: PRISMA flowchart.

This systematic approach to screening for eligibility ensures a thorough and comprehensive review of the literature on plastic pyrolysis, laying a solid foundation for the subsequent analysis.

3 Catalyst Optimization and Product Yield in PET Pyrolysis

3.1 *Data synthesis*

This section synthesizes findings from previous studies related to the pyrolysis of plastic PET, catalysts, and the yield of pyrolysis products. The studies included in this synthesis were selected following the PRISMA statement, as detailed in Figure 2, which outlines the systematic selection process in accordance with PRISMA guidelines [22].

The development of PET pyrolysis presents several challenges. During the pyrolysis process, the formation of acids such as terephthalic acid (TPA) and benzoic acid can occur, significantly impacting the quality of the resulting products. These byproducts not only degrade the final yield but also necessitate additional processing steps for their removal, which can complicate and increase the cost of the pyrolysis process.

Furthermore, the synthesis of the data indicates that various catalysts have been explored to mitigate these issues and enhance product yield. Commonly studied catalysts include zeolites, which have shown potential in promoting the breakdown of PET while reducing the formation of undesirable byproducts. The implications of these findings suggest a need for further research into optimizing catalyst performance and addressing the challenges associated with acid formation to improve the efficiency and viability of PET pyrolysis.

3.2 *Pyrolysis of plastic*

Pyrolysis products can be categorized into liquid, gas, and solid fractions [23]. The yield of the liquid fraction from plastic pyrolysis is influenced by both the type of plastic and the pyrolysis temperature. For example, pyrolysis of polyethylene (PE) at 350 °C yields a minimal amount of oil, which significantly increases at 450 °C, resulting in the highest oil yield. Conversely, polypropylene (PP) achieves optimal liquid product yield at 450 °C, with no wax formation observed. Notably, pyrolysis above 450 °C tends to reduce oil yield while increasing gas production [24].

The decomposition of PET begins at 270 °C and is complete by 480 °C, primarily through the thermal cleavage of C-C bonds [24], [25]. The liquid products from plastic pyrolysis are complex mixtures, containing various compounds such as paraffins, olefins, and naphthenes. The specific composition is influenced by the plastic feedstock. For instance, PET at 350 °C produces a high quantity of wax, which decreases at higher temperatures [24]. The primary compounds from PET pyrolysis include terephthalic acid, benzoic acid, carbon monoxide (CO), and carbon dioxide (CO_2) due to its linear structure, along with byproducts such as vinyl terephthalate, acetaldehyde (CH_3CHO), and divinyl terephthalate [26]. Catalytic pyrolysis is typically carried out at temperatures ranging from 300 °C to 500 °C for a duration of 20 to 60 min, utilizing acid catalysts to facilitate the breakdown of plastic chains [9].

The application of catalysts in PET pyrolysis significantly enhances product yields, especially in liquid and gas forms. Catalysts such as natural zeolite, HZSM-5, and MgO-ZSM-23 effectively reduce solid residues and facilitate the conversion of hydrocarbons into valuable compounds, including benzene and other aromatic hydrocarbons [27]. These catalysts shift the reaction pathway, promoting the formation of smaller, more desirable compounds such as benzoic acid and light hydrocarbons [28], [29]. Furthermore, catalysts improve reaction efficiency, enabling the breakdown of hydrocarbon chains at lower temperatures [20], [30]. This advancement leads to increased yields of lighter hydrocarbons and gas, optimizing the overall process [30], [31].

Catalysts in PET pyrolysis have been proven to effectively minimize the production of undesirable byproducts, including acidic compounds and tar. Zeolites, particularly HZSM-5, reduce levels of terephthalic acid and aromatic compounds in the tar, resulting in cleaner products and minimizing the risk of reactor equipment corrosion. This improves the overall efficiency of the pyrolysis process [30], [31]. Furthermore, the HZSM-5 catalyst enhances product selectivity, especially in producing aromatic hydrocarbons like benzene, toluene, ethylbenzene, and xylene (BTEX), with yields of up to 14.9%. This catalyst also converts terephthalic acid into more valuable hydrocarbons, making it suitable for advanced applications [32], [33]. In the pyrolysis of PET, catalyst implementation in reactors can occur through in situ and *ex situ* approaches, each with distinct advantages and challenges impacting process efficiency and effectiveness [34], [35]. Additionally, catalysts not only improve product yields but also reduce the temperature required for PET decomposition, making the process more energy-efficient [36]. They further aid in reducing carbon residue, resulting in a cleaner and more efficient pyrolysis process [37].

Pyrolysis of plastics, such as polyethylene (PE), polypropylene (PP), and polyethylene terephthalate (PET), yields liquid, gas, and solid fractions, with temperature and plastic type significantly influencing these outcomes. For example, PE produces minimal oil at 350 °C but reaches the highest oil yield at 450 °C, while PP's optimal oil yield occurs at 450 °C with no wax formation. Catalysts like HZSM-5 improve PET pyrolysis, enhancing liquid and gas yields by reducing tar and solid residues. However, while catalysts enhance efficiency and product selectivity, their high cost and environmental impact are not fully addressed. Additionally, the potential challenges in scaling catalytic pyrolysis for large-scale plastic waste processing and the sustainability of catalyst use in commercial applications warrant further investigation.

Table 1: Review of data synthesis from pyrolysis of PET.

No.	Title	Purpose	Main finding	Ref.
1.	Benzoic acid recovery via waste poly-(ethylene terephthalate) (PET) catalytic pyrolysis using sulfated zirconia catalyst	Measures the yields of three main products from PET pyrolysis: solid residue, wax, and gas	▪ Benzoic acid recovery from PET pyrolysis is 27–32 wt% at 450–600 °C with a 20 s residence time. ▪ A 10 wt% catalyst-to-plastic ratio lowers benzoic acid yield to 26 wt% but increases light hydrocarbons in the gas. ▪ The SZ catalyst deactivates at temperatures above 525 °C due to coke and decomposition, but this doesn't happen at lower temperatures.	[38]
2.	Monocyclic aromatic hydrocarbons production from NaOH pretreatment metalized food plastic packaging waste through microwave pyrolysis coupled with ex-situ catalytic reforming	▪ Yields of pyrolysis products: liquid oil, wax, residue, and gas. ▪ Chemical composition of the liquid oil, including MAHs, NAP+, and PAHs.	▪ NaOH pretreatment broke the bonds between plastic and aluminum, simplifying the pyrolysis process and improving microwave energy efficiency. ▪ The best MAHs yield of 80.50% was achieved with HZSM-5 (Si/Al = 50) catalyst, pyrolysis temperature of 550 °C, and catalysis temperature of 550 °C	[39]
3.	Resource recovery from discarded COVID-19 PPE kit through catalytic fast pyrolysis	Yields and composition of pyrolysis products from PPE kit components using non-catalytic and catalytic fast pyrolysis.	▪ Catalytic fast pyrolysis using HY and HZSM-5 zeolite catalysts can convert these components into valuable products like BTEX and naphthalene. ▪ The best results for BTEX and naphthalene yield were achieved at 600 °C with a 1:4 feed-to-catalyst ratio for both HY and HZSM-5 catalysts.	[37]
4.	Catalytic pyrolysis of polyolefin and multilayer packaging based waste plastics: A pilot scale study	▪ Yields of oil, gas, and char from catalytic pyrolysis of packaging plastics. ▪ Physicochemical and thermal properties of the pyrolysis oil.	▪ PET-based MLPs plastic waste yields more oil 17.8% oil yield, and a higher calorific value of 30 KJ/g.	[40]
5.	Catalytic Pyrolysis of Plastic Waste: Moving Toward Pyrolysis Based Biorefineries	▪ Yields of liquid oil, gas, and char from catalytic pyrolysis PET using modified natural zeolite (NZ) catalysts. ▪ Chemical composition of the liquid oil was analyzed by GC-MS, HHV and FT-IR.	▪ The oil had high aromatic content along with aliphatic hydrocarbons. ▪ The oil's heating value is similar to fuel, showing potential as an alternative fuel after refining.	[41].
6.	Catalytic pyrolysis of polyethylene terephthalate over zeolite catalyst: Characteristics of coke and the products	▪ Zeolite catalyst's effect on primary and secondary reactions during PET pyrolysis. ▪ Coke formation on the catalyst and potential metal transfer from PET to the catalyst. ▪ Properties of liquid products	▪ Zeolite catalyst boosted cracking, increasing tar and gas yields while reducing char and wax. ▪ It raised hydrogen and light hydrocarbons in the gas through enhanced dehydrogenation and cracking. ▪ Zeolite reduced acid and aromatic content in tar via decarboxylation and dearomatization.	[31]
7.	Catalytic upcycling of post-consumer multilayered plastic packaging wastes for the selective production of monoaromatic hydrocarbons	Evaluation of the catalytic activity of zeolites in the fast pyrolysis of MLPWs to produce value-added chemicals, particularly monoaromatic hydrocarbons (MAHs)	▪ Using zeolite for fast pyrolysis of MLPW produces monoaromatic hydrocarbons (MAHs) like benzene, toluene, ethylbenzene, and xylene (BTEX). HZSM-5 zeolite gives the highest yield at 14.9%. ▪ HZSM-5 is more effective due to its acidity and larger pore size compared to HY and Hβ zeolites.	[32]
8.	Effect of catalyst contact mode and gas atmosphere during catalytic pyrolysis of waste plastics	Measures the yields and selectivity of products from the catalytic pyrolysis of plastics PET, under various conditions (catalyst contact mode, such as in-situ vs. ex-situ, and carrier gas type, such as He vs. H2). The yields and selectivity were reported on a carbon basis.	In-Situ vs. Ex-Situ Pyrolysis: In-situ catalytic pyrolysis produced higher yields of aromatics than ex-situ pyrolysis for PET. CO_2 vs. CO: In-situ catalytic pyrolysis of PET resulted in more CO_2 and less CO compared to ex-situ pyrolysis.	[34]

Table 1 *(Continued)*

No.	Title	Purpose	Main finding	Ref.
9.	Integrating PET chemical recycling with pyrolysis of mixed plastic waste via pressureless alkaline depolymerization in a hydrocarbon solvent	▪ PET Conversion: To measure how PET is converted into terephthalic acid (TPA) and how much TPA is recovered. ▪ Depolymerization Impact: To assess how pre-treating PET affects.	PET was fully converted and over 98% of terephthalic acid (TPA) was recovered by alkaline depolymerization using a high boiling point solvent and NaOH at 260 °C with a NaOH/PET ratio of 2.0.	[42]
10.	Enhanced energy recovery from polyethylene terephthalate via pyrolysis in CO2 atmosphere while suppressing acidic chemical species	▪ CO Production: Compare the amount of carbon monoxide (CO) generated during the pyrolysis of PET waste in CO_2 versus N_2 atmospheres. ▪ Product Composition: Compare the types and amounts of benzene derivatives, polycyclic aromatic hydrocarbons (PAHs), and acidic compounds (like benzoic acid) in the pyrolysis products from CO_2 versus N_2 atmospheres.	▪ CO_2 speeds up the breakdown of volatile organic compounds (VOCs) from PET pyrolysis, increasing CO production. ▪ CO_2 reduces benzene derivatives, PAHs, and acids (e.g., benzoic acid) while increasing linear hydrocarbons.	[43]
11	Catalytic Pyrolysis of Waste Plastics over Industrial Organic Solid-Waste-Derived Activated Carbon: Impacts of Activation Agents	▪ Pyrolysis Characteristics: Examines how different carbon materials (AC-ST, AC-HP, AC-ZN) affect the pyrolysis temperatures and characteristics of PET. ▪ Product Yields: Measures the yields of gas, liquid, and solid products from the catalytic pyrolysis of PET. ▪ Liquid Composition: Analyze the composition and carbon number distribution of the liquid products.	▪ Chemical Activation: Using H_3PO_4 and $ZnCl_2$ results in carbons with higher surface area and porosity than steam activation. ▪ Reaction Pathways: Acid sites from P and Zn on AC-HP and AC-ZN carbons alter the reaction pathways, enhancing cracking, aromatization, and decarboxylation of plastics. ▪ Product Yields: AC-ZN carbon yields more long-chain alkylbenzenes, which are valuable, compared to AC-HP carbon.	[44]
12.	Slow pyrolysis of waste polyethylene terephthalate yielding paraldehyde, ethylene glycol, benzoic acid and clean fuel	▪ Yield pyrolysis of waste PET. ▪ The composition of the oil fraction, including the identification and quantification of specific compounds	▪ Pyrolysis Yields: The slow pyrolysis of waste PET produces gas (13.3%), oil (46.7%), and solid carbon residue (39.7%). ▪ Oil Composition: The oil contains compounds like terephthalic acid, benzoic acid, aldehydes, ethylene glycol (23.7%), paraldehyde (54.7%), and other aromatic hydrocarbons.	[45]
13.	Slow pyrolysis of polyethylene terephthalate: Online monitoring of gas production and quantitative analysis of waxy product	Study on the slow pyrolysis of polyethylene terephthalate (PET), focusing on the online monitoring of gas production and the quantitative analysis of waxy products.	▪ Gas Production: The types and amounts of gases produced during the slow pyrolysis of PET, including acetaldehyde, CO, CO_2, C_2H_4, C_2H_6, benzene, and other aromatics. ▪ Waxy Products: The amounts of waxy products from PET pyrolysis, including benzoic acid and its derivatives, monovinyl terephthalate, divinyl terephthalate, and other compounds.	[46]
14.	Insight into the competitive reaction mechanism of polyethylene terephthalate (PET) pyrolysis by ReaxFF-based reactive molecular dynamics simulation	How PET pyrolysis works by using detailed simulations and Pyrolysis-GC-MS measurements to measure the competition between different reaction types during PET breakdown.	▪ PET pyrolysis mainly involves random chain scissions. ▪ Initially, homolytic cleavage is more common, while concerted reactions increase over time. ▪ Crosslinking reactions become more significant in the later stages of pyrolysis.	[47]

Table 1 *(Continued)*

No.	Title	Purpose	Main finding	Ref.
15.	Characterization of products obtained from waste polyethylene terephthalate by pyrolysis	▪ Total conversion of the waste polyethylene terephthalate (wPET) during pyrolysis ▪ Oil and gas yield from the pyrolysis of wPET	▪ The highest total conversion (62%) was achieved with microwave pretreatment and catalytic conditions. ▪ The highest yield of oil and gas (45%) was at 425 °C for 30 min. ▪ Under catalytic conditions, the solid products were mostly crystalline terephthalic acid and boron compounds. Without a catalyst, only terephthalic acid was present.	[48]
16.	Effects of MgO-ZSM-23 Zeolite Catalyst on the Pyrolysis of PET Bottle Waste	Measured in this study were the yields of the different products (gas, liquid/wax, and char) from the pyrolysis of PET bottle waste, both with and without the MgO-ZSM-23 zeolite catalyst	▪ The MgO-ZSM-23 catalyst increased gas yields and decreased solid char compared to pyrolysis without a catalyst. ▪ Catalytic pyrolysis produced less CO_2 and more lighter hydrocarbon gases (C_1–C_5) than non-catalytic pyrolysis. ▪ The catalyst shifted the liquid/wax products from high molecular weight compounds (like biphenyl) to lower molecular weight benzene derivatives, particularly benzoic acid.	[28]
17.	Low-Temperature Treatment of Waste PET	Mass Balance: Measures the yields of solid residue, liquid mixture, and gas from the pyrolysis process. Liquid Composition: Analyzes the liquid fraction for the presence and amount of ethylene glycol and other compounds.	▪ At 400 °C, the pyrolysis produced solid fuel with a high heating value (HHV) of 31–32 MJ/kg and a low heating value (LHV) of 30–31 MJ/kg. ▪ Ethylene glycol and aldehydes were obtained as useful chemicals, particularly with slow pyrolysis at 400 °C. ▪ Quinones were also found in the decomposition products.	[49]
18.	Analysis of Products Obtained from Slow Pyrolysis of Poly(ethylene terephthalate) by Fourier Transform Ion Cyclotron Resonance Mass Spectrometry Coupled to Electrospray Ionization (ESI) and Laser Desorption Ionization (LDI)	analysis of products from slow pyrolysis of PET using advanced techniques: Fourier transform ion cyclotron resonance mass spectrometry (FT-ICR MS)	▪ The waxy products from PET pyrolysis were a complex mixture, with terephthalic acid (TPA) being the most prominent. ▪ These waxy products included various oxygen-containing compounds. ▪ The solid residue from PET pyrolysis had three main types of compounds: oxygenated compounds, hydrocarbons, and pure carbon. The amount of carbonaceous materials increased with higher pyrolysis temperatures.	[50]
19.	Conversion of Polyethylene Terephthalate Based Waste Carpet to Benzene-Rich Oils through Thermal, Catalytic, and Catalytic Steam Pyrolysis	The paper measures the yields and selectivity of liquid, gas, and solid products from pyrolysis of waste PET under various conditions, including temperature, heating rate, catalysts, and steam addition.	▪ Thermal pyrolysis of PET carpet waste produces high amounts of benzoic acid and acetylbenzoic acid. Catalytic pyrolysis converts these acids into aromatic hydrocarbons more effectively. ▪ Catalytic steam pyrolysis using a CaO catalyst has the highest selectivity for benzene, thanks to enhanced hydrolysis reactions. ▪ The benzene-rich liquid from steam pyrolysis of PET carpet waste offers a promising option for reusing this waste.	[33]
20.	Catalytic Fast Pyrolysis of Poly (Ethylene Terephthalate) (PET) with Zeolite and Nickel Chloride	Studies the catalytic fast pyrolysis of PET using ZSM-5 zeolite and nickel chloride, examining how different catalyst amounts and temperatures affect the yields and composition of the pyrolysis products.	▪ Adding ZSM-5 zeolite and $NiCl_2$ catalysts reduced waxy product yields but increased gas yields. ▪ ZSM-5 zeolite mainly affected secondary reactions, while $NiCl_2$ enhanced primary decomposition, leading to more liquid products. ▪ ZSM-5 zeolite removed 42% of carbonyl groups and 20% of aliphatic C-O bonds in the waxy products compared to pyrolysis without a catalyst.	[30]

Table 1 *(Continued)*

No.	Title	Purpose	Main finding	Ref.
21.	Conversion of plastic waste into fuel oil using zeolite catalysts in a bench-scale pyrolysis reactor	Measures the yields of oil, gas, and char from the catalytic pyrolysis of various types of plastic waste, expressed as weight percentages.	▪ PET pyrolysis mainly produces gases and solids, with only a small amount of liquid. ▪ At lower temperatures, the main products were oxygen-containing compounds like benzoic acid, terephthalic acid, and monomethyl terephthalate. ▪ The catalyst's strong acidity and microporous structure enhanced cracking and isomerization, increasing oil yield by breaking down larger molecules into smaller ones.	[51]

3.3 *Catalytic pyrolysis*

Plastic pyrolysis has emerged as a promising approach to address plastic waste issues, yet it faces significant challenges, including the diverse nature of pyrolysis products and the high temperature requirements. Catalysts, particularly zeolites, are instrumental in enhancing the efficiency and selectivity of the pyrolysis process. By using catalysts, it becomes possible to increase reaction rates, lower the required temperature, reduce reaction times, and produce more targeted, higher-quality outputs, making the pyrolysis process more viable on an industrial scale [36], [52]. Commonly used catalysts in plastic pyrolysis include ZSM-5, Y-zeolite, FCC, MCM-41, and USY [9]. These catalysts significantly improve yield and selectivity by targeting specific reactions that reduce unwanted by-products and optimize oil production. For instance, Ni/ZSM-5 in the pyrolysis of polyethylene terephthalate (PET) has been shown to increase oil yield up to 52.9% at 600 °C, compared to 34.4% without a catalyst, demonstrating a marked improvement in efficiency and output [53]. Such enhancements underscore the role of catalysts in making plastic pyrolysis more economically feasible and environmentally sustainable.

Catalyst efficacy varies across different types, with microporous catalysts like HZSM-5 gaining recognition for boosting the yield of aromatic and iso-alkane compounds, which are crucial for producing high-energy fuels. The strong acidic sites in HZSM-5 facilitate the cracking of polymer chains, leading to a higher yield of desirable hydrocarbons, particularly aromatics [41]. When combined with mesoporous catalysts such as MCM-41, HZSM-5 shows even greater potential. MCM-41's larger pore size allows for enhanced diffusion, facilitating interactions between larger polymer molecules and the catalyst. This combination has been shown to yield higher-quality liquid oils with lighter hydrocarbons, such as C_3–C_4 compounds, which are suitable for fuel applications [54], [55]. This synergy not only increases the overall efficiency of the pyrolysis process but also produces a favorable product composition.

Recent advances in catalyst modification aim to further enhance pyrolysis efficiency. Modifying HZSM-5 through metal doping or hierarchical structuring, or blending it with SiO_2-Al_2O_3 and MCM-41, has proven effective in reducing gas by-products while maximizing liquid yields. Metal doping, in particular, has been found to enhance catalyst stability and provide greater active surface area, further optimizing the process [54]. Some types of synthetic zeolite used for plastic pyrolysis are shown in Table 2. These modifications not only improve the catalytic activity but also enable pyrolysis at lower temperatures, which is beneficial from an environmental perspective as it reduces energy consumption.

Mixed oxide catalysts, such as Al_2O_3 and SiO_2 blends, have also shown promising results in pyrolysis, especially at moderate temperatures of 400–550 °C. Research indicates that increasing the SiO_2 content enhances liquid product yields, as SiO_2 supports cracking reactions that favor liquid formation [56], [57]. Using a two-stage catalytic system, such as MCM-41 for initial pyrolysis and ZSM-5 for further cracking, can yield up to 83.15 wt.% of pyrolysis oil [58]. This setup allows the MCM-41 catalyst to break down the plastic into smaller hydrocarbons, which ZSM-5 then processes into high-value liquid fuels, including C_2 (ethene), C_3 (propene), and C_4 (butene and butadiene) [59]. The use of large-pore Y-zeolite further enhances the production of aromatics [60], valuable for fuel due to their high energy density [61].

Catalytic hydrocracking has also shown notable improvements, especially when using catalysts such as NiMo/ Al_2O_3 and Pt/Al_2O_3. Gas chromatography-mass spectrometry (GC-MS) analysis reveals that these catalysts can increase the gasoline-range fraction significantly, from 15.94 wt% to 34.45 wt% with NiMo/ Al_2O_3 and up to 49.85 wt% with Pt/Al_2O_3, indicating high selectivity in producing fuel-grade hydrocarbons [62]. Calcium oxide (CaO) catalysts have been shown to reduce benzoic acid production while improving the quality of the pyrolytic liquid, during PET pyrolysis [63]. These results suggest that catalyst choice and configuration can allow for tailored product distributions, thus optimizing the pyrolysis output based on specific requirements for fuel or chemical applications.

Catalytic pyrolysis offers a promising solution for plastic waste recycling by improving efficiency and product selectivity. Catalysts, especially zeolites like ZSM-5 and MCM-41, enhance the reaction rates, reduce temperature requirements, and produce higher-quality liquid products, including valuable hydrocarbons such as aromatic compounds and iso-alkanes. For example, Ni/ZSM-5 significantly increases oil yields from PET, showcasing the potential for industrial scalability. Recent advancements, such as catalyst modification and the use of mixed oxide systems, further optimize yields and reduce undesirable by-products. However, the challenges of catalyst deactivation, high cost, and energy consumption in large-scale operations remain significant barriers. The balance between catalytic efficiency and economic feasibility, especially in long-term applications, warrants further attention.

Table 2: Performance of different types of zeolite catalysts in plastic pyrolysis.

Types of Zeolites	Activation Method	Feedstocks Type	Key Findings	Ref.
Natural and synthetic zeolite	NZ particle size 15–35 µm without modification/activation. Temperature 450 °C for 75 min	PS, PE, PP, PET	PE plastic highest oil yield reached 54% NZ, 50% synthetic zeolite. Diesel equivalent oil with HHV 40.2–45 MJ/kg	[55]
MCM-41 & zeolite ZSM-5	2 layers of catalyst mixed 1:1 (4gr in total). Feedstock and catalyst ratio 1:2. Temperature 500 °C and heating rate 10 °C/min.	HDPE	Yield oil 83.15 wt%, with main gases of C_2 ethene, C_3 propene, C_4 butene & butadine. Oil with 95.85% aromatic and 97.72% hydrocarbon range gasoline.	[59]
Natural zeolite (NZ)	NZ was heated at 120 °C for 5 h, and calcined at 500 °C for 5 h.	Waste Compact disc (CD) case	Highest oil yield >90% with NZ catalyst at 450 °C, HHV 41.75 MJ/kg	[64]
NZ & HY zeolite	NZ was treated with HCl (20gr NZ: 500 mL HCl (0.5, 1, 6 M). Calcined at 500 °C for 3 h.	PP, PS	HCl increases the SI/Al ratio and surface area, and HCl concentration increases oil yield.	[65]
ZSM-5	Optimization of cristalin ZSM-5 by baking at 150 °C for 24 h, stirrer at 100 °C for 6 h and calcined at 550 °C for 6 h.	LDPE	Zeolite ZSM-5 catalyst produced 70% oil, 16% gas, and 14% charcoal.	[53]

3.4 *Characterization natural zeolite*

Zeolites are crystalline aluminosilicate materials renowned for their microporous structure and are widely applied as catalysts, adsorbents, and ion exchangers due to their unique physicochemical properties [66], [67]. The general chemical formula of zeolites is given by Mx/n[AlxSiyO(x+y)].pH_2O, where M represents exchangeable cations such as Na, K, Li, Ca, Mg, Ba, or Sr, and n denotes the cation's charge. In zeolites, the Si/Al ratio is a key factor influencing acidity, stability, and catalytic performance. The ratio y/x ranges from 1 to 6, p/x ranges from 1 to 4, and n is the cation charge [68]. This ratio ranges from 1 to 6, categorizing natural zeolites into low (1–2), intermediate (2–5), and high Si/Al ratios (>5) [69].

Zeolites with a lower Si/Al ratio exhibit higher ion-exchange capacity and are typically less stable in acidic environments. Conversely, those with a higher Si/Al ratio, such as mordenite-type zeolite, are more acid-resistant, making them suitable for catalytic applications that require acid stability [69]. Natural zeolites generally have a Si/Al ratio of around 5, similar to Y-zeolite, indicating moderate acidity and structural stability. However, certain types like clinoptilolite and mordenite—often found in Indonesian deposits—exhibit enhanced resistance to acidic conditions and are promising for catalytic pyrolysis applications due to their structural robustness [69].

Another fundamental characteristic of zeolites is porosity, determined by their microporous framework, with pore sizes typically ranging from 0.3 to 1 nm. This microporosity contributes to high internal surface areas, generally between 300 and 700 m^2/g, which is beneficial for catalytic reactions by providing extensive active sites for reactant adsorption [69]. The pore volume of natural zeolite typically lies within 0.1 to 0.35 cm^3/g, accommodating a variety of molecular sizes, making them suitable for the selective adsorption of hydrocarbon compounds during plastic pyrolysis. Natural zeolites also possess notable cation-exchange properties, which are beneficial in low-temperature catalytic processes. This ion-exchange capability is ecologically advantageous as it releases non-toxic exchangeable cations, such as K^+, Na^+, Ca^{2+}, and Mg^{2+}, into the environment, adding a layer of sustainability to their use [70].

Natural zeolite from Indonesia of the clinoptilolite and mordenite type has been used for pyrolysis [71]. To enhance the catalytic properties of natural zeolite for pyrolysis, physical and chemical activation processes are essential, as presented in Table 3. In this study, natural zeolite underwent a series of preparation stages. Initially, the zeolite was crushed and sieved to achieve a particle size of 2–5 mm, which improves surface area exposure and aids in reaction efficiency [72]. The sieved zeolite was then washed with demineralized water to remove surface impurities, an essential step to prevent contamination during catalytic processes. Following the initial washing, the zeolite was subjected to activation using 1 M NH_4Cl solution for 24 h. This ammonium ion exchange process replaces native cations in the zeolite structure with NH_4^+, enhancing the acidity of the zeolite, which is beneficial for catalytic cracking reactions in pyrolysis [71]. After soaking, the zeolite was filtered and washed with distilled water, followed by a rinse in silver nitrate ($AgNO_3$) solution to remove residual Cl- ions, ensuring the zeolite's purity. The next steps included oven drying at 120 °C and calcination at 450 °C for eight hours, which serves to remove the NH_4^+ ions, thus creating active acid sites necessary for hydrocarbon cracking [71]. The activated zeolite was then used in varying concentrations (1–9% w/w) during pyrolysis at 400 °C, a temperature optimized for the thermal degradation of plastics like polypropylene (PP) and high-density polyethylene (HDPE).

Table 3: Si/Al ratio and surface area composition of various natural zeolites.

Natural Zeolite (NZ)	Silica (%)	Alumina (%)	Si/Al	Surface Area (m^2/g)	Pore Volume (cm^3/g)	Micropore Volume (cm^3/g)	Ref.
NZ Bayah, INA	64.54	10.62	6.08	47.45	-	-	[72]
NZ Lampung, INA	79.91	11.67	6.85	49.85	-	-	[72]
Modified NZ Bayah, INA	78.2	8.5	9.2	86.24	-	-	[72]
Modified NZ Lampung, INA	78.2	7.8	10.03	102.34	-	-	[72]
NZ Bayah, INA	-	-	4.6	19	0.06	0.003	[73]
NZ Sukabumi, INA	-	-	4.6	31	0.089	-	[74]
NZ Ponorogo, INA	-	-	4.5	30	0.116	0.002	[75]
NZ Klaten, INA	-	-	6.0	113	0.009	0.055	76]
NZ Kucin, Slovakia	-	-	4.1	33	0.135	0.003	[77]
NZ Demirci, Turkey	-	-	4.7	112	0.33	0.045	[78]
NZ Harrat, KSA	43.39	6.90	6.28	5.65	0.022	0.018	[79]

This structured preparation process enhances zeolite's catalytic activity by increasing its surface area and active site density, thus improving its efficiency in breaking down polymers. The increased acidity and optimized pore structure allow the zeolite to facilitate selective cracking of long-chain hydrocarbons into lighter fractions, which are valuable as fuel components. Such improvements underscore the potential of Indonesian natural zeolites in sustainable pyrolysis applications, offering a cost-effective alternative to synthetic catalysts. Therefore, the characterization and activation of natural zeolite emphasize its suitability as a catalyst in plastic pyrolysis. By adjusting the Si/Al ratio and enhancing acidity through ion exchange and calcination, the zeolite's catalytic performance can be tailored to maximize efficiency in hydrocarbon cracking. This study demonstrates that activated natural zeolite is a viable catalyst for pyrolysis, providing an eco-friendly, scalable solution for plastic waste valorization in fuel production.

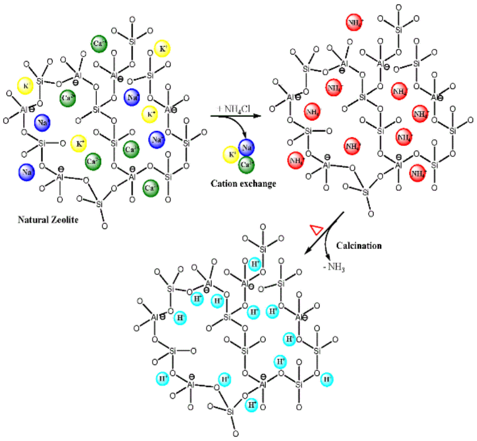


Figure 3: Schematic of ion exchange on natural zeolite [71].

The activation of natural zeolite is a crucial step in enhancing its catalytic properties for various applications, including catalytic pyrolysis. Zeolites, which are crystalline aluminosilicates, typically exhibit weak electrostatic bonds with alkali and alkaline earth ions such as Ca^{2+}, Na^{+}, K^{+}, Mg^{2+}, Ba^{2+}, Li^{+}, and Sr^{2+} on their outer surfaces as shown in Figure 3 [80], [81]. To improve the catalytic efficiency of natural zeolite, soaking in ammonium chloride (NH_4Cl) solution is employed. This treatment facilitates the exchange of NH_4^+ ions for the cations in natural zeolite, effectively displacing some of the alkali and alkaline earth ions and thus enhancing the pore structure of the zeolite [71], [82]. The implications of this process can be observed in the increased porosity and surface area, which are vital for catalytic applications.

During the calcination process, NH_4^+ ions are thermally decomposed into ammonia (NH_3) vapor, which is released from the zeolite structure, leaving behind H^+ ions. This transformation is critical as it results in the formation of H-zeolite, which increases the density of Bronsted acid sites, thereby improving the catalyst's ability to facilitate acid-catalyzed reactions [82]. As evidenced by X-ray diffraction (XRD) analysis, treatment with hydrochloric acid at 1 M concentration effectively reduces specific peaks at $2\theta = 9.8°$ and 22.4°, indicative of a successful dealumination process that does not compromise the overall crystalline structure of the zeolite [82]. After activation, the crystal purity of natural zeolite is markedly enhanced as impurities are expelled, notably seen in XRD results where peaks at $2\theta = 28.1°$ and 28.4° diminish significantly which can be observed in the XRD test results shown in Figure 4 [71]. The acidity of the activated zeolite, measured through ammonia temperature-programmed desorption (NH_3-TPD), was found to increase to 0.4704 mmol NH_3/g, confirming the successful activation and enhancement of catalytic properties [83]. The transition from microporous to mesoporous zeolite is significant in catalytic processes. Mesoporous zeolites, characterized by larger pore sizes, facilitate the accommodation of bulky reactants derived from plastic pyrolysis, thus preventing blockages and promoting the production of valuable aromatic hydrocarbons [84], [85]. The higher surface area of mesoporous zeolites enhances accessibility to active sites, which is pivotal in improving the catalytic conversion rates of oxygenates to aromatic compounds [84], [86].

An advanced approach in zeolite catalysis involves metal modification, where metals such as nickel (Ni), cobalt (Co), zinc (Zn), and iron (Fe) are impregnated into the zeolite framework. This modification significantly alters the texture, acid sites, and thermal stability of the zeolite, leading to reduced char formation and increased yields of pyrolysis liquids [87], [88]. The technique used was the impregnation of Ni, Co, Zn, and Fe metals into the zeolite framework, and was able to reduce char production by 50% during co-pyrolysis of wheat straw and polystyrene [87]. The introduction of phosphorus (P) and nickel (Ni) has been particularly effective in decreasing strong Bronsted acid sites, which reduces char deposition and enhances the production of olefins and monoaromatic hydrocarbons during pyrolysis [89]. The metal-modified zeolites are known to facilitate decarboxylation and decarbonylation reactions while inhibiting dehydration, thus optimizing the overall catalytic performance [87].

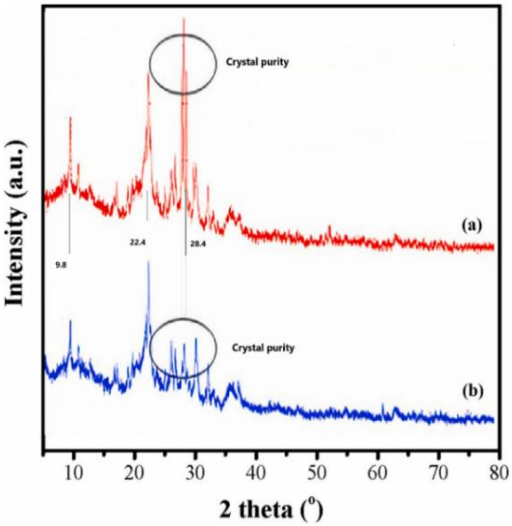


Figure 4: Comparison of XRD test results of zeolite after (a) and before activation (b) [71].

The characterization of natural zeolites focuses on their chemical composition, physical properties, and catalytic potential, particularly in pyrolysis applications. Zeolites, primarily composed of aluminosilicate structures, exhibit a microporous framework that enables high surface area and ion-exchange properties. The Si/Al ratio is crucial, as it influences the stability, acidity, and catalytic efficiency, with lower ratios offering better ion-exchange capabilities but reduced acid resistance, while higher ratios improve acid resistance. Activation of natural zeolites, particularly through ammonium ion exchange and calcination, enhances their catalytic properties by increasing acidity and improving pore structure, making them effective in breaking down long-chain hydrocarbons during plastic pyrolysis. The activation process not only increases surface area and active sites but also transitions zeolites from microporous to mesoporous, which is beneficial for accommodating larger reactants. Additionally, metal modifications, such as introducing nickel or cobalt, further enhance catalytic performance by improving thermal stability and reducing char formation. This study underscores the potential of natural zeolites as sustainable and cost-effective catalysts in pyrolysis, offering an eco-friendly solution for plastic waste management. Critically, while the activation process significantly improves catalytic performance, further research is needed to optimize the metal modification process and understand its long-term stability and scalability in industrial applications. Additionally, the reliance on ammonium chloride for ion exchange may pose environmental concerns if not properly managed, warranting further exploration into greener alternatives for activation.

Despite the advantages of zeolite catalysis, the deactivation of zeolites presents a significant challenge. The formation of coke, a carbonaceous deposit resulting from the catalytic reaction, obstructs access to active sites and diminishes catalytic efficiency [90], [91]. Several pathways contribute to zeolite deactivation, including carbon chemisorption and physisorption, encapsulation of metal particles, pore blockage, and degradation of the zeolite structure. The accumulation of coke obstructs the access of reactants to active sites, thereby diminishing the catalyst's ability to facilitate chemical reactions [92]. Coke deposition on a supported metal catalyst leads to deactivation through several pathways as shown in Figure 5: (i) carbon chemisorption or physisorption that obstructs reactant access; (ii) encapsulation of metal particles; (iii) blockage of pores; and (iv) degradation of the catalyst's structure [93]. Additionally, other factors such as sintering and the adsorption of impurities also contribute to the reduction in catalytic activity.

To mitigate these issues, effective regeneration strategies for deactivated natural zeolites must be employed. Oxidative regeneration is a common approach, wherein the deposited coke is combusted using air or oxygen to restore catalytic activity. This method necessitates a comprehensive understanding of coke oxidation kinetics to ensure efficient mass transfer and removal of carbon deposits [92], [93]. Advanced oxidation processes, such as ozonation, have also gained attention for their ability to utilize ozone's strong oxidizing properties to effectively eliminate coke without the thermal drawbacks associated with traditional methods [92], [93]. Alternative methods such as gasification, which involves reacting coke with steam or carbon dioxide (CO_2) to convert it into synthesis gas (syngas), and hydrogenation, which facilitates the removal of carbon deposits under milder conditions, can also be implemented [92], [93].

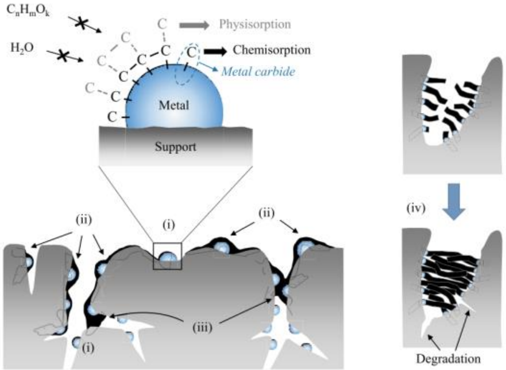


Figure 5: Deactivation pathways by coke formation [92], [93].

Zeolite catalysis, while advantageous, faces the challenge of deactivation primarily due to coke formation, which obstructs active sites and reduces catalytic efficiency. Several mechanisms contribute to this deactivation, including carbon chemisorption, metal particle encapsulation, pore blockage, and structural degradation. Regeneration strategies, such as oxidative regeneration, advanced oxidation processes like ozonation, and alternative methods like gasification or hydrogenation, are essential to restore zeolite activity. However, these methods require careful control to ensure efficiency and avoid thermal damage or other drawbacks. Effective regeneration techniques are crucial for maintaining the long-term viability of zeolite catalysts in industrial applications.

3.5 *Pyrolysis reaction mechanism of plastic and zeolite*

The catalytic pyrolysis of plastics using zeolites as catalysts is a promising approach for converting waste plastics into valuable hydrocarbons. Pyrolysis is a thermal degradation process that occurs at elevated temperatures (typically between 300 °C and 900 °C) and can be enhanced by the use of zeolites, which exhibit several advantageous properties, including high acidity, substantial specific surface area, and excellent adsorption capacity. These characteristics render zeolites highly effective catalysts for producing high-value chemicals from plastic feedstocks [94], [95].

In general, catalytic pyrolysis of plastics occurs in two primary stages: 1) depolymerization and devolatilization, followed by 2) catalytic conversion of the resulting volatiles into hydrocarbons [96]. The initial depolymerization stage involves breaking the long polymer chains of plastics into smaller fragments through a series of cracking reactions. For instance, studies utilizing HZSM-5 zeolite (with a SiO_2/Al_2O_3 ratio of 30) and HY zeolite (with a SiO_2/Al_2O_3 ratio of 23) during the co-pyrolysis of Quercus variabilis and plastic waste at 600 °C have demonstrated that HZSM-5, characterized by its higher acidity, is more effective at enhancing aromatics production due to improved cracking efficiency [97].

However, the effectiveness of zeolite catalysts can be significantly hampered by their pore structure. The HZSM-5 zeolite, which features a small pore size of less than 2 nm, can restrict the diffusion of larger plastic-derived compounds into its internal acid sites during pyrolysis. As a result, the inability of these compounds to access active sites leads to blockage and deactivation of the catalyst, ultimately reducing its lifespan and efficiency [98], [99]. To address this issue, the development of mesoporous zeolites—characterized by larger pore sizes—has become essential. Mesoporous zeolites, as heterogeneous catalysts, facilitate more efficient reactions without dissolving into the reactants. Their enhanced surface area and larger pore structures minimize the risk of blockage by large plastic-derived compounds, thereby improving access to active sites and catalytic interactions [100], [101]. In addition to enhancing the efficiency of the pyrolysis process, mesoporous zeolites can also improve product selectivity, yielding valuable hydrocarbons such as benzene and other aromatic compounds while minimizing the formation of undesirable byproducts, such as tars and acids [55], [102]. The high surface area of mesoporous zeolites enhances the conversion of oxygenates to aromatic hydrocarbons, making these materials particularly suitable for catalyzing the transformation of complex plastic waste into high-value products [84], [103].

The pyrolysis of polyethylene (PE) typically involves random scission of the polymer chains, resulting in a range of hydrocarbon fragments, including n-alkanes and alkenes [104]. Increasing the pyrolysis temperature promotes greater bond C-C cleavage, leading to higher yields of lighter gaseous products (C_1–C_4) [105]. For example, a study using a fluidized reactor for the pyrolysis of PE at 740 °C with gaseous product recycling yielded 19.2% by weight of benzene, demonstrating the effectiveness of high temperatures and long residence times in facilitating aromatic production through Diels-Alder reactions [106]. The high temperature and long residence time caused the Diels-Alder aromatization reaction resulting in aromatics as illustrated in Figure 6.

In the absence of a catalyst, the dominant reactions during PE pyrolysis involve intermolecular hydrogen transfer and β-disproportionation, with paraffins as the main products. In contrast, the presence of natural zeolite alters the reaction pathway, promoting hybrid ion abstraction via Lewis sites or carbenium ion mechanisms through Brønsted acid catalysis. This process allows protons to add to the C-C bonds of PE molecules, leading to the cracking of PE into lower molecular weight hydrocarbons. However, it is important to note that the pore sizes of natural zeolites are often inadequate to accommodate the larger molecular sizes of polyolefins, necessitating that thermal or catalytic cracking occurs primarily on the external surface and mesopores of the catalyst [24]. Following this initial reaction, smaller hydrocarbons can enter the zeolite pores to undergo further reactions, thereby enhancing overall conversion efficiency.

Therefore, the development and application of zeolite catalysts for the pyrolysis of plastic waste represent a significant advancement in waste management and resource recovery. By enhancing catalytic activity through the design of mesoporous zeolites and optimizing reaction conditions, it is possible to improve the yield and quality of valuable hydrocarbons while minimizing byproducts. Ongoing research into the regeneration and longevity of these catalysts will be crucial in ensuring the sustainability and effectiveness of catalytic pyrolysis processes in industrial applications. Future studies should focus on innovative approaches for modifying zeolite structures and improving catalytic performance to maximize the utility of plastic waste as a feedstock for high-value chemical production.

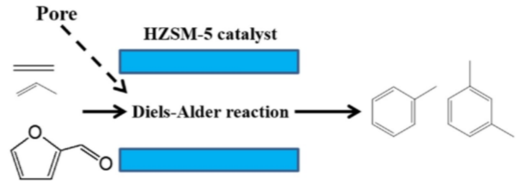


Figure 6: Diels-Alder reaction [107].

Thermal pyrolysis is a pivotal process for the conversion of polyolefins such as polypropylene (PP) and high-density polyethylene (HDPE) into valuable hydrocarbons. The primary mechanisms driving this process involve the formation of free radicals that initiate bond cleavage, resulting in the production of wax, oil, char, and gas [108]. In the presence of mesoporous zeolite catalysts, these pyrolysis mechanisms can be significantly enhanced, facilitating more efficient degradation of long polymer chains into valuable products [24], [105]. The thermal pyrolysis of polyolefins follows a defined sequence of steps. Initially, the polymer decomposes into free radicals at the initiation stage. As the reaction progresses, these radicals propagate, leading to further breakdown of larger molecules into smaller hydrocarbon fragments. This process culminates in the formation of olefin hydrocarbons, with the cracking reaction concluding when the free radicals converge to form oligomers. Notably, catalytic pyrolysis demonstrates a superior capacity for converting wax into oil compared to thermal pyrolysis conducted without zeolite catalysts [24], [109].

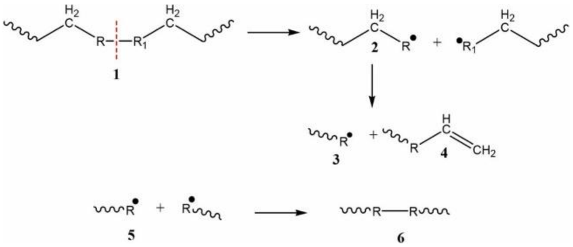


Figure 7: Proposed mechanism of thermal pyrolysis of HDPE plastic samples [71].

As illustrated in Figure 7, the thermal and catalytic processes for microwave pyrolysis of HDPE highlight the importance of zeolite catalysts in enhancing product yields. At temperatures below 500°C, HDPE degradation occurs through two distinct mechanisms: random chain scission producing long-chain hydrocarbons, and oligomeric chain-end cutting yielding lower molecular weight products. These concurrent processes generate free radicals along the carbon chain, leading to the formation of unsaturated fragments. Subsequent hydrogen transfer reactions convert these radical fragments into a variety of products, including straight-chain dienes, alkenes, and alkanes. The catalytic breakdown of volatiles occurs primarily on ZSM-5 zeolite through two carbocationic mechanisms: the classical bimolecular mechanism, known as β-scission, and a monomolecular or proteolytic mechanism utilizing carbonium ions. The unique structure of HDPE allows many chain ends to access the active sites of the ZSM-5 zeolite, facilitating the production of light olefins. Under conditions of high catalyst-to-reactant ratios and elevated catalytic temperatures, the contact between volatiles and the catalyst promotes cracking and reforming reactions. However, incomplete catalytic degradation can lead to the formation of high concentrations of intermediate olefins, which may produce branched hydrocarbons. It is important to note that slower thermal degradation intermediates often cannot penetrate the micropores of ZSM-5, limiting their interactions with the zeolite’s active acid sites [110], [111].

The catalytic activity of zeolite surfaces plays a crucial role in this process. Macromolecules primarily degrade on the outer surface of the zeolite crystallites, with only a fraction diffusing into the micropores for further reaction. The porous structure of the zeolite facilitates the degradation of intermediate vapors, thereby enhancing overall efficiency [112]. As depicted in Figure 8, the production of aromatic compounds during pyrolysis is governed by the interplay of internal acid sites, hydrocarbon pools, and Diels-Alder reactions. The presence of oxygenated compounds, primarily in the form of carboxylic acids, esters, and phenolic derivatives, is also notable. Increasing the pyrolysis temperature, coupled with the use of catalysts, significantly enhances the decomposition of these oxygenated compounds, including terephthalic acid, acetic acid, and benzoic acid, resulting in the formation of oxygen-free aromatics alongside carbon dioxide. Moreover, elevated pyrolysis temperatures favor intramolecular hydrogen transfer reactions, promoting isomerization processes.

The catalytic pyrolysis of plastics using zeolites involves a two-stage mechanism: first, depolymerization and devolatilization, where long polymer chains are broken into smaller fragments, and second, the catalytic conversion of these volatiles into hydrocarbons. Zeolites, such as HZSM-5 and HY, enhance this process by promoting cracking reactions and increasing aromatic production. However, the small pore sizes of zeolites like HZSM-5 can limit access to larger plastic-derived compounds, reducing catalyst efficiency. To address this, mesoporous zeolites with larger pores have been developed to improve catalytic performance by preventing pore blockage, enhancing surface area, and promoting more efficient cracking. These zeolites also improve product selectivity, producing valuable hydrocarbons while minimizing undesirable byproducts. Critical issues remain, such as optimizing catalyst structures and regeneration methods to ensure long-term effectiveness in industrial applications. The development of mesoporous zeolites holds promise in overcoming these challenges and enhancing the sustainability of plastic waste conversion processes.

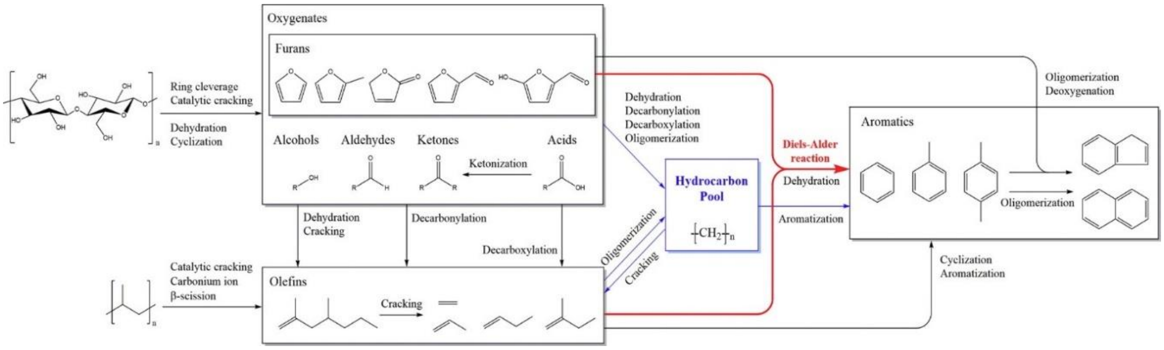


Figure 8: Mechanism of aromatics formation during pyrolysis processes [112].

4 Discussion of Findings and Future Research Directions

The pyrolysis of plastics, including polyethylene (PE), polypropylene (PP), and polyethylene terephthalate (PET), represents a critical process for converting polymer waste into useful energy carriers. The efficiency of pyrolysis depends significantly on the type of plastic, temperature, and the use of catalysts. PE achieves minimal oil production at 350 °C but reaches its optimal yield at 450 °C [24]. Similarly, PP achieves its highest oil yield at 450 °C, producing no wax at this temperature [24]. The addition of catalysts, such as HZSM-5, improves PET pyrolysis by increasing the yield of liquid and gaseous products while minimizing solid residues like tar [30], [31]. However, the high cost and environmental impact of catalysts remain unresolved challenges that hinder the commercial scalability of catalytic pyrolysis [113], [114].

Moreover, catalytic pyrolysis is advantageous for improving reaction efficiency, product selectivity, and lowering reaction temperatures. Zeolites, such as ZSM-5 and MCM-41, have been instrumental in enhancing reaction rates and producing hydrocarbons, including aromatics and iso-alkanes. For instance, the Ni/ZSM-5 catalyst significantly improves oil yields from PET, underlining its potential for industrial-scale applications [53]. Modifications to catalysts, such as using mixed oxide systems, further enhance yield and reduce by-products [56], [57]. Nevertheless, catalyst deactivation due to coke formation, cost, and energy-intensive regeneration processes are barriers that need innovative solutions [58], [115]–[117].

Natural zeolites, characterized by their aluminosilicate frameworks and high ion-exchange properties, are promising candidates for sustainable catalytic applications. The Si/Al ratio in zeolites influences their stability, acidity, and catalytic efficiency. Activation techniques, such as ammonium ion exchange and calcination, enhance their acidity and pore structures, transitioning them from microporous to mesoporous states. This structural modification is particularly beneficial for processing larger molecules in plastic pyrolysis. Additionally, metal doping with elements like nickel or cobalt increases catalytic performance by enhancing thermal stability and reducing char formation. However, the environmental concerns associated with ammonium chloride-based activation necessitate further exploration of eco-friendly alternatives [113], [114].

However, zeolites face deactivation challenges primarily due to coke formation, which blocks active sites and reduces efficiency. Mechanisms of deactivation include carbon chemisorption, pore blockage, and structural degradation. Regeneration strategies such as oxidative regeneration and advanced oxidation processes, including ozonation, are crucial for maintaining catalyst longevity. However, these methods require precise control to avoid thermal damage and ensure efficiency. Innovations in regeneration techniques will be essential for sustaining the viability of zeolite catalysts in large-scale applications [58], [115], [116].

Temperature significantly impacts the yield and quality of pyrolysis products. For PET, oil yield decreases at higher temperatures, with a yield of 34.4% at 600 °C dropping to 11.4% at 900 °C [113]. However, using Ni/ZSM-5 catalysts at 600 °C increases the yield to 52.9%, demonstrating the critical role of catalysts in enhancing performance [113]. The product distribution typically includes n-olefins, n-paraffins, non-oxygenated aromatics, and oxygenated compounds. Higher temperatures favor the production of non-oxygenated aromatics, while oxygenated compounds diminish. Catalysts also influence the decomposition of carboxylic acids, promoting the formation of oxygen-free aromatic compounds and reducing undesirable by-products [48].

In addition, the pyrolysis of PET produces various acidic compounds, such as terephthalic acid, that corrode reactors and block pipelines. To mitigate these issues, the pyrolysis process is typically conducted at temperatures ranging from 510 °C to 730 °C [116]. Catalysts such as Cu-Al metals increase the yield of non-condensable gases, while non-catalytic methods yield higher liquid fractions [117]. Pre-treatment of feedstock, such as drying and size reduction, enhances reaction efficiency and improves product yields. Understanding the reaction mechanisms, including α- and β-scissions, decarboxylation, and carbonium ion formation, is essential for optimizing the pyrolysis process. The reaction pathways and mechanisms to optimize gasoline-equivalent liquid products are illustrated in Figure 9. It illustrates the reaction pathways for optimizing gasoline-equivalent liquid products in PET pyrolysis. It highlights also key reactions, including α-scission, β-scission, decarboxylation, and aromatization, facilitated by zeolite catalysts.

Therefore, Future research should focus on developing environmentally friendly activation processes for zeolites to replace ammonium chloride-based methods. Investigating the long-term stability and scalability of metal-doped zeolites will be crucial for commercial adoption. Additionally, advancements in regeneration technologies to reduce energy consumption and prevent thermal damage will enhance catalyst longevity. Exploring innovative reactor designs that mitigate the corrosive effects of acidic by-products and improve heat transfer efficiency could further optimize pyrolysis operations. Lastly, integrating machine learning and computational modeling to predict reaction pathways and optimize operating conditions offers a promising avenue for advancing the field.

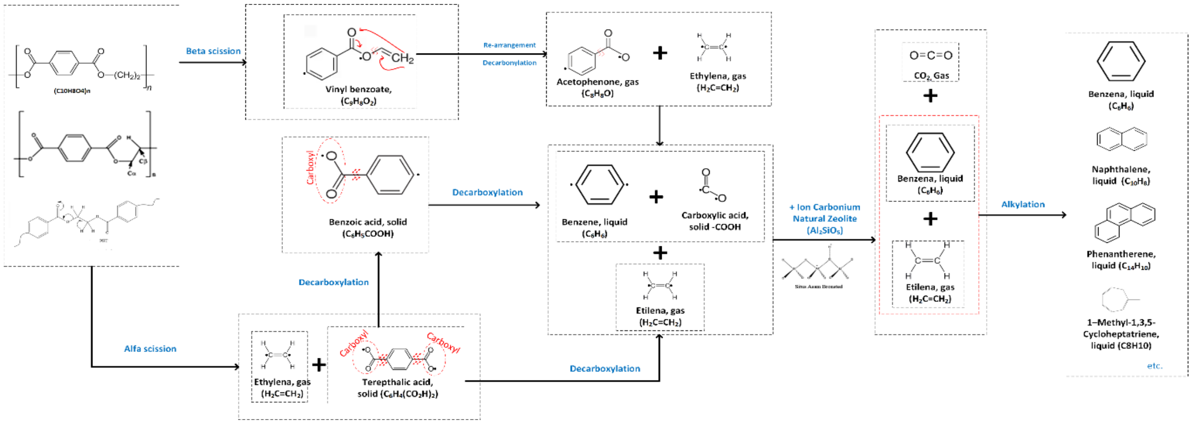


Figure 9: Reaction mechanism of PET pyrolysis with modified zeolite catalyst.

5 Conclusions

This study has demonstrated the effectiveness of natural zeolite catalysts in enhancing both the yield and quality of liquid products from the pyrolysis of PET plastic waste. By catalyzing the breakdown of long hydrocarbon chains into desirable fractions, such as non-oxygenated aromatic hydrocarbons, natural zeolites have proven to be a crucial component in maximizing the potential of PET waste for energy conversion. This research has successfully achieved its primary objective of investigating the catalytic performance of zeolites in PET pyrolysis, contributing valuable insights into how optimized catalytic strategies and pyrolysis conditions can support sustainable plastic waste management.

The findings indicate that activated natural zeolite significantly improves liquid product yield from PET pyrolysis. Through physical and chemical enhancements like increased surface area, porosity, and acidity, the activation process improves zeolite’s catalytic efficiency, allowing it to selectively produce high-energy hydrocarbons while reducing unwanted byproducts. Activated zeolite’s ability to accelerate reaction rates, reduce sintering, and enhance thermal stability ensures sustained catalytic activity during the pyrolysis process, which is essential for maintaining efficiency in industrial applications. This selective production of non-oxygenated aromatic compounds, promoted by optimized catalytic properties, contributes to an increase in the energy value of the liquid products, making them more suitable for fuel applications and supporting the circular economy by converting waste into valuable energy resources.

The influence of process parameters, such as pyrolysis temperature and reaction time, was also found to be significant in determining the composition and yield of liquid products. Higher pyrolysis temperatures, in particular, were associated with an increased proportion of non-oxygenated aromatic compounds in the final product, thus improving the quality and energy density of the pyrolysis oil. By optimizing these parameters, this study has identified conditions under which PET waste can be most effectively converted into fuel, addressing both quality and quantity improvements. These findings have the potential to inform future research and industrial-scale applications, where fine-tuning process parameters could lead to more efficient waste-to-energy conversion.

Furthermore, this research supports sustainable waste management practices by offering a practical solution for mitigating the environmental impact of PET waste. The catalytic pyrolysis of PET using natural zeolite not only demonstrates a viable route for recycling plastic waste into fuel products but also aligns with the broader goals of a circular economy. This process promotes innovative, sustainable practices that help address the global plastic waste crisis and contribute to renewable energy production.

However, a primary challenge faced during the pyrolysis process is the deactivation of the catalyst due to coke formation. Therefore, it is essential to implement effective regeneration strategies to maintain catalytic activity. Regeneration methods, such as oxidative regeneration, gasification, and hydrogenation, should be considered to ensure the sustainability of the pyrolysis process.

To advance the application of catalytic pyrolysis in PET waste management, future studies could explore additional modifications to zeolite catalysts or alternative catalysts that may further optimize product yield and reduce operational challenges. Investigating the interaction mechanisms of PET with various catalyst surfaces under different thermal conditions could provide insights that refine the pyrolysis process and expand its practical applications.

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Author Contributions

S.N.: conceptualization, investigation, methodology topic organization, writing an original draft; S.Y.: writing, reviewing, editing, and supervision; Z.A.: methodology, reviewing and editing; M.S.: writing, reviewing and editing. All authors have read and agreed to the published version of the manuscript.

Conflicts of Interest

The authors declare no conflict of interest.

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