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Article

Mini-Review on Renewable Production of Green p-Xylene

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Abstract

The sustainable and renewable production of p-xylene (PX), a crucial component for polyethylene terephthalate (PET), is increasingly important as an alternative to fossil-based processes. This review examines biomass-derived routes for PX synthesis, emphasizing the use of bio-px production pathways to feasible for commercialization. While bio-PX production offers reduced greenhouse gas emissions, challenges remain in cost, catalyst stability, and energy requirements. Recent innovations in catalyst regeneration and hierarchical structures enhance stability and minimize coke formation. Life-cycle assessments confirm bio-PX's environmental advantages, suggesting that further research into biomass sources and catalyst efficiency will advance bio-based PX production toward commercial viability in a sustainable bioeconomy.

Keywords: p-xylene, bio-based production, green chemistry

1. Introduction

In the past decade, the petrochemical industry has continued to rely heavily on fossil-based resources, posing environmental challenges and depleting reserves. This dependency has sparked a growing interest in alternative sources such as biomass, which offers an economically viable carbon source for energy, fuel, and chemical feedstocks. Biomass-derived feedstocks have emerged as a promising solution, as they can replace fossil-based raw materials in the production of high-demand polymers like polyethylene terephthalate (PET), which is widely used in the textile, plastics, and electronics industries [1]; [2];.

PET production relies on purified terephthalic acid (PTA) and ethylene glycol (EG), with PTA itself being synthesized from para-xylene (PX), a component of BTX (benzene, toluene, and xylene) mixtures traditionally obtained through catalytic reforming and alkylation processes. Achieving high PX selectivity is crucial, and it typically involves specific catalysts such as modified zeolites (e.g., ZSM-5) designed to optimize reaction pathways [3]; [4]; [2]. Given the current market demand for PX, projected to reach 62.66 million tons in 2024 and 80.78 million tons by 2029 with a compound annual growth rate (CAGR) of 5.21%, the Asia-Pacific region remains a focal point of the PX market expansion [5]

The increasing emphasis on sustainable production methods has led to intensive research into renewable PET through biomass-based PTA and EG. Bio-based EG is commonly produced by dehydrating bio-ethanol, while bio-derived PX can potentially fulfill PTA requirements. Four main approaches for bio-based PX synthesis have been explored: bio-ethanol synthesis, isobutanol reforming, biomass pyrolysis, and the conversion

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of biomass-derived 2,5-dimethylfuran (DMF). Among these, DMF has gained substantial attention due to its high yield and economic feasibility [3]; [6];[7],.

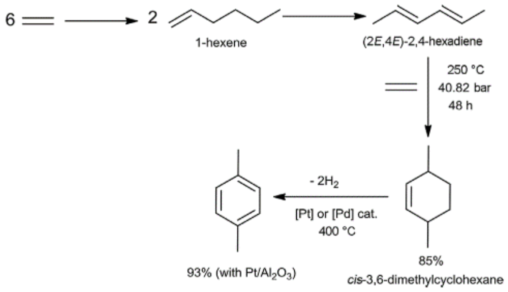

Fig. 1. Reaction pathways P-xylene from ethylene [22]

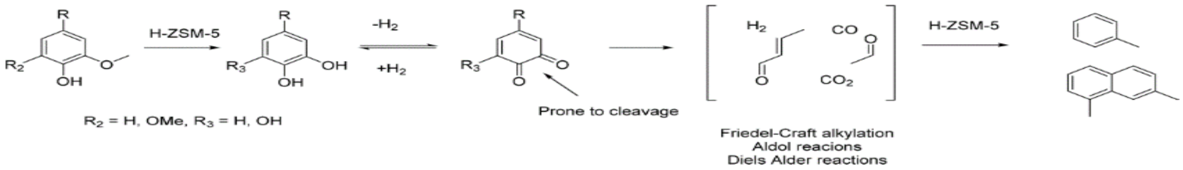

Fig. 2. Proposed mechanism of reaction catalytic pyrolysis [19]

This paper reviews the biomass-derived routes to p-xylene that recent years has been attention. As the push toward sustainable production intensifies, these biomass-derived technologies hold the promise of reshaping the PX market and reducing the petrochemical industry's reliance on fossil fuels, paving the way for a more sustainable future in polymer production and beyond.

2. BTX Production

The method adopted in this study involves a review and compare approach to evaluate the state-of-the-art in biomass-derived routes to p-xylene, with a particular focus on pathways offering high selectivity through the use of bio-derived compounds and selective reactions, as well as their potential for commercialization. A comparative analysis, both qualitative and quantitative, was conducted to assess the strengths and weaknesses of each pathway, focusing on metrics such as energy efficiency, greenhouse gas emissions, and industrial feasibility. Findings were presented in tables and visualized through comparative graphs to facilitate understanding and highlight key technological trends.

2.1. Conventional Method

Conventional BTX production typically involves catalytic reforming, fluid catalytic cracking (FCC), and pyrolysis processes applied to petroleum-derived feedstocks. Catalytic reforming of naphtha, one of the primary methods, uses catalysts like platinum on alumina to promote the formation of aromatic compounds such as benzene, toluene, and xylene. This method is widely applied due to its high efficiency in producing aromatics and by-products like hydrogen, which can be used elsewhere in refining operations [8]

Fluid catalytic cracking (FCC), originally developed to maximize gasoline production, has been adapted to increase the yields of light olefins and BTX aromatics. Modified FCC units enable higher BTX production by cracking heavier hydrocarbons in the presence of zeolite catalysts, an effective approach for petrochemical applications [9]. Conventional BTX production processes, which have been validated through many years of successful commercial application. However, there is an urgent necessity to replace these with bio-based BTX solutions to support sustainable green chemistry initiatives.

2.2. Bio-Based Method

2.2.1. Ethanol to PX route

Routes to p-xylene derived from ethanol are interesting by many known of using ethanol as high-volume and easy to produce to any method with economic cost, this become one of most favorable utilization of ethanol for BTX production. The production of bioethanol from biomass waste or sugar cane has been establish. Upgrade ethanol to ethylene The initial step is the dehydration of ethanol to ethylene, typically achieved using alumina or silica-based catalysts at temperatures around 300–400°C. This reaction is critical, as ethylene serves as the primary intermediate for further transformations. Following dehydration, ethylene undergoes oligomerization to form C4 (butenes) and C8 (octenes) compounds. This step generally employs acidic catalysts, such as zeolite Y or supported metal catalysts (e.g., Ni or Pd), which operate effectively within a temperature range of 200–350°C and atmospheric to moderate pressures, promoting the formation of desired intermediates.

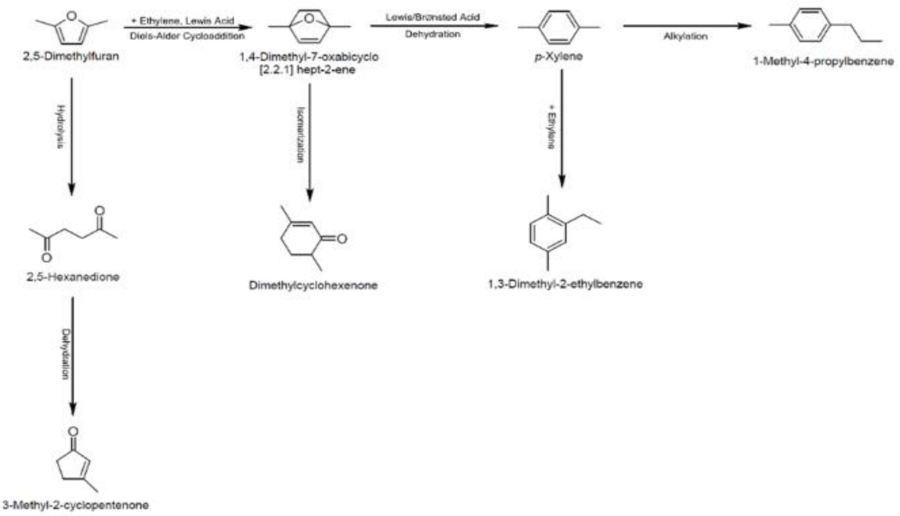

Fig. 2. Diels-Alder reaction of DMF and ethylene [17]

The final and most crucial step in PX production involves the cyclization and aromatization of C8 intermediates to form aromatic compounds, specifically p-xylene. This step is typically catalyzed by modified ZSM-5 zeolites, which are renowned for their shape-selective properties that favor the production of p-xylene over other xylene isomers. Metal doping, such as incorporating gallium (Ga) or zinc (Zn) into the ZSM-5 framework, enhances the catalytic efficiency and selectivity towards PX. These catalysts operate optimally at temperatures ranging from 400 to 500°C, with slight pressures maintained to improve aromatic yields. Under these conditions, yields of p-xylene can reach approximately 93%, using palladium catalyst and 400^0C condition temperature[10]

In terms of economic and commercial viability this step has been proven industrially established as 1-hexene is produced via oligomerization of ethylene at high yield. According to meuwese (2013) every 1 kg of biomass from ethanol produce 0.39 of CO_2 emission and biomass BTX produce 0.72 kg of every kg production of product. It is very promising than of fossil BTX that produce 1.4 kg of every kg BTX that produce using ration (67:5:28).[11]

2.2.2. Biomass Catalytic route

Catalytic pyrolysis is another significant method, particularly suited for co-processing biomass and hydrocarbon feedstocks. Recent studies highlight the effectiveness of using zeolite catalysts, such as ZSM-5, to enhance BTX selectivity. For example, Yi et al. (2019) demonstrated that Org-CaO/Nano-ZSM-5 catalysis during pyrolysis could improve aromatic hydrocarbon yields, providing a sustainable alternative pathway for BTX production [12]

Innovative catalyst modifications are being explored to further optimize BTX yields. Shape-selective catalytic pyrolysis of lignin, a renewable biomass source, using modified zeolite catalysts has shown promise in increasing the efficiency of BTX production from biomass. Ma et al. (2021) investigated various zeolite catalysts for bio-BTX production and found that shape-selectivity enhances the conversion of biomass intermediates into aromatic products [13]

Another approach involves using hierarchical HZSM-5 zeolites modified through NaOH treatment, which enhances the catalytic cracking of renewable feedstocks like methyl oleate into BTX components. Feng et al. (2021) demonstrated that these modified zeolites could improve BTX yields while maintaining eco-friendly production standards [14]

Potential of green aromatics production pathways using catalytic pyrolysis method has shows promising ways, using HZSM-5 impregnated using gallium has shows 5-7% of organic compound has been reported by Heeres (2018) it is lead that the usefulness of acidic catalyst is significance. However, the mechanism of catalytic reaction pathways toward green aromatics still in under debate. Fig 2, proposed mechanism pathways of catalytic.

2.2.3. DMF to PX Rotute

Conversion of DMF to PX involves a Diels-Alder reaction with ethylene, followed by the selective dehydration of the oxanorbornene intermediate to yield PX this is show in Fig 3. A schematic diagram of Diels-Alder reaction of DMF and ethylene. This route provides higher PX exclusivity compared to traditional naphtha catalytic reforming, which relies on petroleum feedstocks [6]; [15]; [16];[17]. Biomass-derived DMF is typically synthesized via catalytic hydrogenation of HMF (5-hydroxymethylfurfural), a compound obtained from biomass carbohydrates. The hydrogenation of HMF to DMF can be effectively carried out using various metal catalysts such as palladium, ruthenium, copper, and nickel, supported on materials like activated carbon, silica, or alumina [4]; [1]. By employing this bio-based route, the synthesis of PX aligns more closely with green chemistry principles, reducing dependency on fossil resources.

Considerable advances have been made in developing catalysts and optimizing conditions to maximize the yield of PX from DMF. Modified zeolites, especially ZSM-5 doped with elements such as phosphorus and magnesium, have been effective due to their shape-selective properties, which enhance PX selectivity by facilitating selective diffusion and transformation of smaller aromatic molecules within the zeolite framework ([18]; [2]). Studies have shown that doping ZSM-5 with zinc or gallium leads to improved yields of BTX (benzene, toluene, and xylenes) due to the promoted dehydrogenation and aromatization activities of the modified catalyst. [19];[20]; [21]Moreover, reaction conditions such as temperature and pressure have been refined to improve the catalytic performance of ZSM-5-based systems, yielding promising results in the selective production of PX [22]; [6].

In addition, Optimizing reaction conditions, including temperature, pressure, and feed ratios, is essential to achieve high selectivity and conversion in PX production from DMF and ethylene. The Diels-Alder reaction between DMF and ethylene to produce PX requires elevated temperatures to drive the reaction, while controlling pressure is crucial to avoid unwanted by-products. Additionally, altering the molar ratios of DMF to ethylene has a significant impact on conversion efficiency and PX yield. Research suggests that higher temperatures are beneficial for the Diels-Alder reaction itself, while moderate pressures enhance PX selectivity by balancing cycloaddition and dehydration rates. Temperature-programmed testing and reactor systems with precise control over these variables are key for optimizing PX yield and reducing by-products, thus supporting the process's efficiency and economic feasibility for large-scale implementation.[7];[23];[24];[25]

Moreover, recent advancements in catalyst development have explored the use of hierarchical zeolites and mesoporous structures to improve catalytic efficiency. Hierarchical HZSM-5, prepared through NaOH treatments, has been shown to possess increased mesoporosity, allowing for better mass transport and accessibility to active sites, which enhances PX yields. These hierarchical catalysts exhibit improved catalytic lifetimes and reduced coke formation, which are common challenges in conventional zeolite catalysts. The increased surface area and pore volume facilitate the diffusion of reactants and products, reducing reaction times and enhancing overall productivity. Additionally, the acidity profile of hierarchical catalysts can be tailored to optimize the catalytic process by enhancing the

Brønsted/Lewis acid site ratio, a factor known to influence PX selectivity in Diels-Alder reactions. [15];[26];[24]

Another approach explored in recent research is the use of bifunctional catalysts that combine acidic and metal sites, which have been shown to improve the conversion rates and selectivity of biomass-derived DMF to PX. For instance, catalysts composed of Pt-modified ZSM-5 or Ga-promoted ZSM-5 allow simultaneous hydrogenation and aromatization, critical steps in biomass conversion. The presence of metal sites enables efficient deoxygenation and stabilization of intermediate compounds, which enhances the selectivity for aromatic hydrocarbons. Such bifunctional catalysts are particularly effective in producing BTX compounds from DMF, as they reduce the likelihood of side reactions that produce unwanted heavy compounds. [27];[28] acidic metal has shown potential in industrial application, with simultaneous reaction and stability it is very likely promising use in commercial use.

Table 1. Summary of Catalytic Conversion Processes for Bio-Based Aromatics Production.

Product	**Feedstock**	**Catalyst Type**	**Operating Condition**	**Yield**	**Source**
p-Xylene (PX)	DMF and Ethylene	ZSM-5 doped with Phosphorus, Magnesium	High temperature, moderate pressure	High PX selectivity (>70%)	[34];[17]
Bio-BTX (Benzene, Toluene, Xylenes)	Lignin	Zn-modified HZSM-5, Zn-HZSM-5	500–600°C (temperature)	BTX selectivity 65.02%	[15];[27]
Aromatic hydrocarbons	Canola oil methyl ester (CME)	MoO3/ZSM-5	Torrefaction at 280°C, catalytic pyrolysis	Aromatic yield 91.38%	[23]
Green BTX	Canola oil methyl ester (CME)	Zn-modified HZSM-5	450°C, WHSV 2 hr	aromatic yield 42.6%	[20]
BTX	Paraffinic Naphtha	Pt-Ga/ZSM-5	550°C, WHSV 1 hr	Aromatics yield 60%	[28]
p-Xylene	2,5-Dimethylfuran (DMF) and Acrylic Acid	Beta Zeolite	473 K, 10.1 min residence time	PX yield 83%	[25]

The shift towards bio-based production methods for PX is also supported by the growing demand for polyethylene terephthalate (PET), which relies heavily on PX as a precursor. As global awareness of environmental sustainability increases, bio-based PX production aligns with efforts to reduce greenhouse gas emissions and environmental impact. Bio-PX production from DMF leverages biomass, a renewable resource, and aligns with sustainable energy goals by reducing fossil fuel dependency and minimizing carbon footprint.[29]; [11];[30]

To further enhance the economic feasibility of bio-based PX, studies are investigating the life-cycle assessment of this process. By evaluating the energy consumption, greenhouse gas emissions, and material requirements across each stage of bio-PX production, researchers can identify areas for improvement and optimization. The integration of renewable energy sources into the production process, such as using green hydrogen in hydrogenation steps, can also further lower the environmental impact. Additionally, advances in catalyst regeneration techniques, such as oxidative treatment, are being explored to extend catalyst lifetimes and reduce overall production costs, making bio-PX production more competitive with conventional petrochemical methods[26];[17];[29]

Overall, the conversion of biomass-derived DMF to PX offers a promising and sustainable alternative to traditional fossil-based PX production. Advances in catalyst design, particularly through modified and hierarchical zeolites, have significantly improved the yield and selectivity of PX, while optimized reaction conditions have further enhanced process efficiency. The development of robust, selective, and recyclable catalysts will be pivotal in advancing this technology and meeting the increasing global demand for PET in a more sustainable manner. Further studies into catalyst design and process optimization are essential to establish bio-based PX as a commercially viable alternative to conventional methods [31]; [4].

3. Results and Discussion

Catalytic Conversion Processes for Bio-Based Aromatics Production, many routes on conversion process of biomass to aromatics production using various catalyst comparative performance has been summarize to table 1.

The varying performances in bio-based aromatic hydrocarbon production, reflecting how catalyst selection, feedstock type, and reaction conditions significantly impact conversion efficiency and yield outcomes. The conversion of 2,5-dimethylfuran (DMF) to p-xylene (PX) via a Diels-Alder reaction with ethylene achieves notably high PX selectivity (>70%) when ZSM-5 doped with elements such as phosphorus and magnesium is used. This high performance can be attributed to the shape-selective properties of ZSM-5, which facilitate precise molecular diffusion and minimize by-products. Additionally, the high temperature and moderate pressure conditions optimize both cycloaddition and dehydration steps, enabling efficient PX production[32].

In contrast, the catalytic pyrolysis of lignin for bio-BTX (benzene, toluene, xylenes) production, using Zn-modified HZSM-5, achieves a BTX selectivity of around 65%. This process, operated at higher temperatures (500–600°C), benefits from the zinc modification, which enhances the acidity and dehydrogenation ability of the catalyst, essential for breaking down complex lignin structures. However, the yield remains slightly lower than that of PX due to the inherent complexity of lignin's structure, which contains multiple oxygenated and aromatic components that require additional deoxygenation steps[15]

Further differences are observed in the pyrolysis of Miscanthus, where the dual-catalyst system of MoO_3/ZSM-5 yields a high aromatic output (91.38%) with significant BTX selectivity. The use of MoO_3 in conjunction with ZSM-5 facilitates a deoxygenation process while retaining aromatic structures, essential for maximizing BTX yield. Torrefaction of the feedstock prior to catalytic pyrolysis aids in reducing oxygen content, enhancing carbon efficiency, and thereby contributing to higher aromatic yields compared to processes without feedstock pretreatment[23]

Comparatively, the conversion of canola oil methyl ester (CME) over Zn-modified HZSM-5 at 450°C results in a moderate yield of 42.6% for aromatics. The lower yield, compared to biomass-derived feeds like DMF, can be attributed to the fatty acid ester structure of CME, which has fewer direct pathways to aromatics and requires more extensive cracking and dehydrogenation reactions. Zinc-modification improves the catalyst's aromatization capacity, but additional steps are necessary to achieve complete transformation, thereby slightly limiting the yield. [20]

Lastly, the conversion of light paraffinic naphtha to BTX aromatics over Pt-Ga/ZSM-5 under elevated temperatures (550°C) and a controlled space velocity shows an impressive aromatic yield of 60%. This outcome demonstrates the effectiveness of Pt-Ga/ZSM-5's dual functionality in promoting both dehydrogenation and aromatization, essential for converting lighter hydrocarbons into valuable BTX compounds. The Ga component enhances the dehydrogenation rate, a crucial step for naphtha, which primarily consists of saturated hydrocarbons. [28].

4. Conclusion

Based on several literatures discussed previously, the performance differences in bio-based aromatic hydrocarbon conversions are primarily influenced by the complexity of feedstocks, catalyst composition, and process-specific operating conditions. Catalysts modified with metals such as zinc (Zn), gallium (Ga), and molybdenum (Mo) demonstrate superior selectivity and yield by enhancing critical reactions such as dehydrogenation, acidity control, and deoxygenation. Simpler feedstocks, like 2,5-dimethylfuran (DMF), tend to offer higher selectivity, whereas more complex biomasses, such as lignin and canola oil methyl ester (CME), require advanced catalysts and conditions to achieve optimal yields, highlighting the vital interaction between feedstock characteristics and catalyst design

In summary, the shift to bio-based p-xylene (PX) production not only addresses resource depletion but also brings economic and environmental challenges. Achieving economic viability is essential for bio-based PX to compete with traditional petrochemical methods. Currently, the production costs for bio-PX remain higher due to the price of renewable feedstocks like DMF, derived from 5-hydroxymethylfurfural (HMF), and the added expenses of catalyst development. For example, catalysts such as ZSM-5 modified with metals like gallium, zinc, or molybdenum, significantly improve yields and selectivity, but come at an elevated cost due to the need for high purity metals and precise doping techniques. Additionally, processes that require high temperatures (e.g., around 500–600°C for BTX production from lignin or naphtha) demand significant energy input, which can further drive up production costs if not optimized for energy efficiency.

Reaction stability and catalyst longevity are vital for the economic and environmental viability of bio-PX production. Catalyst deactivation, often due to coke buildup and by-products, limits scalability. Hierarchical and metal-doped catalysts, like Pt-Ga/ZSM-5 for BTX from light paraffinic naphtha, resist coke formation, extending active site lifetimes. MoO3/ZSM-5 with Miscanthus feedstocks also shows strong deoxygenation stability, reducing maintenance needs. While hierarchical catalysts enhance diffusion and ease regeneration, further research into catalyst durability and regeneration methods is needed to boost long-term efficiency and reduce costs.

The environmental impact, particularly in terms of greenhouse gas (GHG) emissions, is another crucial factor. Bio-based PX production is generally considered to have a lower carbon footprint compared to fossil-based processes due to the renewable nature of biomass. However, certain steps, like hydrogenation, may still require hydrogen sourced from fossil fuels, potentially offsetting the environmental benefits. Integrating green hydrogen from renewable sources, such as electrolysis powered by wind or solar energy, could further reduce emissions associated with hydrogenation steps. Life-cycle assessments (LCA) are essential to quantify emissions across each production phase, and early studies indicate that bio-based PX can offer significantly lower CO_2 emissions, especially when using non-food biomass and agricultural residues as feedstocks [33].

Finally, Future progress in bio-based PX production will depend on addressing economic, stability, and emission challenges. Lowering the cost of renewable feedstocks, advancing catalyst regeneration, and integrating renewable energy sources could make bio-based PX more competitive with petrochemical alternatives. Additionally, expanding diverse biomass sources and minimizing external energy input through process integration can enhance sustainability. Some newly introduced biomass sources for PX production present promising long-term potential, though commercial feasibility from these substrates remains distant. To accelerate short-term viability, recent technological advancements have improved biomass-derived PX production, yet most methods still rely on sustainable, abundant, and cost-effective feedstocks, and catalysts require further optimization in selectivity and operating conditions. Intensified research and industry efforts in the coming years are anticipated to address these limitations and develop robust, high-performance catalytic systems. This focus on enhancing catalytic technologies will be key to achieving affordable, scalable, and sustainable bio-aromatics production, supporting the transition toward a bioeconomy that reduces dependence on fossil fuels and mitigates environmental impacts.

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