



Bio-based aromatics for chemicals and materials: Advances in renewable drop-in and functional alternatives

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Abstract

Benzene, toluene, and xylenes (BTX), as well as their downstream products, are a fundamental part of numerous processes in the chemical industry. However, by now, aromatics are still yielded from fossil resources like naphtha, coal, and natural gas. Thus, to push the chemical industry further toward renewability, the production of bio-based aromatics is an essential step to take. The implementation of bio-based aromatics to replace petrochemical aromatics can proceed in two main ways: as direct replacement *via* renewable drop-in or as replacement by renewable functional alternatives. However, the implementation of both pathways still requires significant process optimization toward large-scale application in industrial processes. In this work, renewable drop-in is mainly discussed in the context of pyrolysis and Diels–Alder reactions. Furthermore, renewable functional alternatives discussed here focus on furan derivatives and lignin-based building blocks.

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Current Opinion in Green and Sustainable Chemistry 2024, 47:100931

This review comes from a themed issue on **Biobased Aromatic Chemicals and Materials - A Roadmap For Sustainable Development**

Edited by **Fabio Arico, Andreia F. Sousa, Tobias Stern and Nathanaël Guigo**

Available online 30 April 2024

For complete overview of the section, please refer the article collection - [Biobased Aromatic Chemicals and Materials - A Roadmap For Sustainable Development](#)

<https://doi.org/10.1016/j.cogsc.2024.100931>

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Keywords

Bio-based aromatics, Renewable aromatics, Renewable drop-in, FDCA, HMF, Lignin.

Introduction

Currently, the chemical industry is still strongly dependent on fossil fuels. This industry sector is not only a significant energy consumer, but also uses large amounts of fossil resources to produce chemicals and materials. Thus, changes in the availability or price of fossil fuels have a significant impact on the chemical industry and consumers. Since it is long known that fossil resources are not produced at the same rate as they are consumed, and thus far from circular, the energy sector and feedstock supply need to be shifted toward renewable alternatives. In the long term, this cannot only stabilize cost fluctuations, but also ensures a more reliable and independent feedstock supply. Due to their high value and extensive application in the chemical industry, the production of bio-based aromatics will have major economic and ecologic impact. Among these aromatics, the commodity chemicals benzene, toluene, and xylenes (BTX) are of particular interest, as they are precursors to many downstream products. Conventionally, these aromatics are produced in multiple steps from fossil resources, namely naphtha, coal, or natural gas. To replace these fossil resources in the production of versatile platform molecules, lignocellulosic biomass is considered a promising and abundant alternative. Its three main components are lignin, hemicellulose, and cellulose. Some types of biomass also contain starch and other carbohydrates. For a successful replacement of fossil resources by bio-based alternatives, efficient pathways of biomass valorization need to be investigated. These pathways can utilize various approaches that allow optimization toward a variety of target molecules. Efforts toward renewable aromatics can be divided into two main approaches: the direct replacement of petrochemicals by their renewable equivalents, so-called drop-in solutions, and renewable functional alternatives to commodity aromatics. In the following paragraphs, the focus is set on current developments in the manufacture of aromatics from biomass and their further utilization. Due to their rather limited applications, if compared to monoaromatic molecules, polycyclic aromatic molecules will not be discussed in the scope of this work.

Direct replacements of petrochemicals *via* renewable drop-in

Over the past 50 years, the global plastic production has increased eightfold [1,2] and is expected to increase

further in the future. In 2022, 400.3 Mt of plastic were produced in total, of which only 0.5% were bio-based [2]. In the same year, poly(ethylene terephthalate) (PET) and polystyrene (PS) accounted for a total of 11.4% of the global plastic production from fossil resources [2]. PET and PS are commodity plastics, containing terephthalic acid and styrene as aromatic building blocks, respectively. Both of these building blocks are downstream products of BTX aromatics. Since BTX downstream products have already found suitable wide range purpose due to their established applications, a direct replacement of these *via* renewable drop-in would be highly advantageous in terms of the easier implementation into already existing production lines and consumer products. This way of implementing bio-based chemicals avoids the necessity of completely new or strongly adapted downstream processes and is in this regard less cost and labor intensive, making them economically easier to implement. Various pathways toward bio-based BTX aromatics are recently investigated. These include upgrading of bio-fuels from biomass *via* catalytic fast pyrolysis, as well as cycloaddition reactions of bio-based building blocks (Figure 1) and more.

Pyrolysis

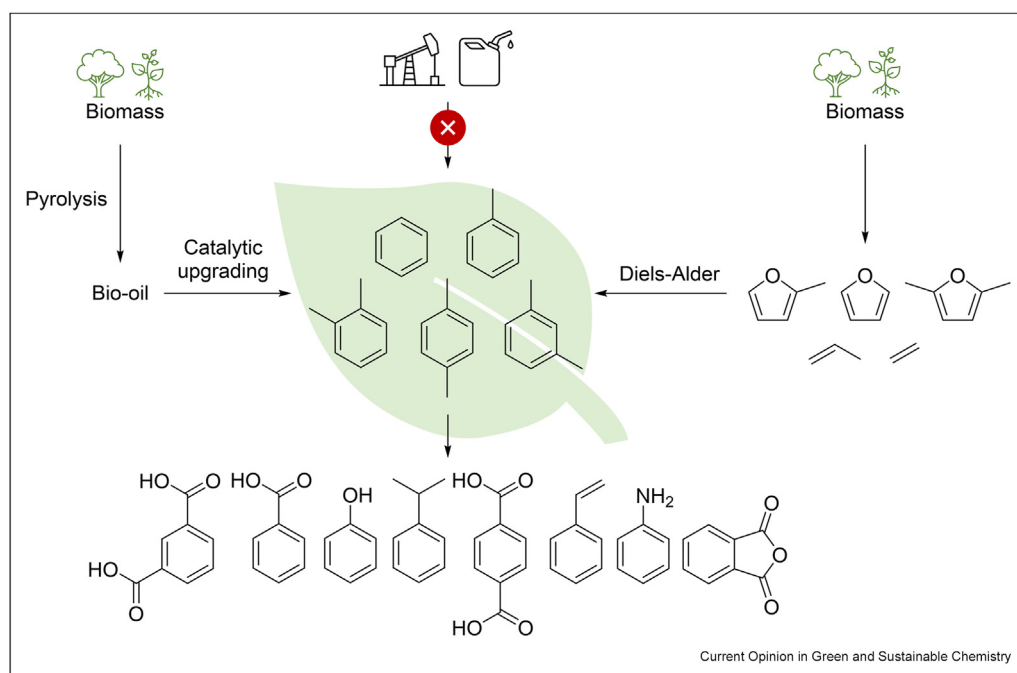
Fast pyrolysis is a thermochemical procedure by which the feedstock decomposes thermally in the absence of oxygen to form fractions of solid char, gases, and most important for the discussion herein: liquid bio-oil (Figure 1, left) [3]. For fast pyrolysis, bio-oil is the

main product with typical yields around 40%–60% [4,5]. However, the exact distribution of the three main product fractions of fast pyrolysis varies strongly with the reaction conditions, experimental setup and the type of biomass that is used [5]. Due to the high content of oxygen in biomass, the obtained bio-oil shows undesired properties like high acidity and low stability [6]. Thus, the bio-oil must be upgraded to higher value compositions either in a separate, subsequent process or directly during catalytic fast (hydro)pyrolysis [3,6]. Other techniques to lower the oxygen content of bio-oil cover various pre- and in-process treatments of the used biomass [7].

Catalytic fast pyrolysis under the right conditions is considered a suitable technique for BTX production, since the higher quality bio-oil obtained *via* this process usually has higher hydrocarbon content due to the removal of oxygen from the bio-oil [8]. Different catalytic systems based on zeolites or metal oxides, other metal based catalysts and waste products like red mud have been reported to be suitable for improving BTX selectivity during catalytic fast pyrolysis [3,9,10].

Besides the used catalysts, also feedstock choice has a great impact on the product distribution after catalytic fast pyrolysis [11]. Since biomass feedstock usually contains high fractions of oxygen, which is rather disruptive for optimal BTX yield, the addition of materials that help to increase hydrogen fractions during pyrolysis can improve the process [11,12]. Based on this,

Figure 1



Fast pyrolysis and Diels–Alder cycloaddition approaches toward renewable BTX aromatics and some discussed downstream products.

catalytic co-pyrolysis of biomass with waste materials like tires, plastic (PVC, LDPE) or oil sludge has been reported recently to increase the hydrocarbon fractions and BTX selectivity during catalytic fast pyrolysis [12–15]. This is especially interesting as it combines the use of biomass as feedstock with the valorization of accumulating waste.

Diels–Alder cycloaddition

Diels–Alder reactions are atom efficient cycloadditions of a diene and a dienophile, in which a six-membered, unsaturated carbon ring is formed, thus offering an attractive pathway for the production of BTX aromatics (Figure 1, right) [16]. The general method for the manufacture of bio-based aromatics *via* Diels–Alder cycloadditions involves the reaction of a bio-based furan derivative as diene with a bio-based short chain olefin like ethylene or propylene as dienophile [16,17]. Suitable furan derivatives are for example furan, 2-methylfuran and 2,5-dimethylfuran, which can all be produced from biomass [17]. Diels–Alder reactions of these building blocks are also an important pathway for BTX formation during catalytic fast pyrolysis of biomass [18]. Hence, one way to produce BTX aromatics *via* Diels–Alder cycloadditions is to expose the mentioned furan derivatives and olefins to conditions similar to those used in catalytic fast pyrolysis, i.e. high temperatures (around 500 °C) and catalysts like HZSM-5 or β -zeolites [19]. Under these conditions, a complex product mixture of benzene, toluene, xylenes, but also polycyclic aromatics is obtained [19,20]. Recently, it was also shown that by using this Diels–Alder approach, platform chemicals like furfural and ethanol can be used directly instead of the alkylated furan derivatives and olefins, due to the *in-situ* decarbonylation of furfural to furan and dehydration of ethanol to ethylene under the harsh conditions [21].

A different approach toward BTX production *via* Diels–Alder cycloaddition uses lower temperatures and olefin pressure, while working with catalysts that contain Brønsted acid sites (BAS) and Lewis acid sites (LAS) [22]. The implementation of both, BAS and LAS, relates to the two step mechanism of the BTX formation *via* Diels–Alder reactions of furans and olefins. While the first step, the Diels–Alder reaction, is typically catalyzed by LAS, the second step, which is the dehydration aromatization of the formed oxanorbornene derivative, is more effectively catalyzed by BAS [17,22,23]. Depending on the furan derivative and olefin used, different products and side products form [23]. With regard to the synthesis of renewable terephthalic acid for PET production, the synthesis of *p*-xylene from 5-hydroxymethylfurfural (HMF) derived 2,5-dimethylfuran and ethylene is particularly interesting [24]. A techno-economic analysis showed that, compared to other bio-based *p*-xylene production routes, this is indeed an attractive process that can be further optimized toward cheaper prices, mainly by lowering HMF

cost [25]. For further insights, a life cycle assessment for this method was performed on the production of bio-based *p*-xylene and compared the environmental impact of fossil-based *p*-xylene and bio-based *p*-xylene from edible biomass (starch) and non-edible biomass (red oak) [26]. While bio-based *p*-xylene from non-edible biomass was found to be similar to its fossil-based equivalent in terms of environmental friendliness, the bio-based *p*-xylene from edible biomass was found to be less environmentally friendly than fossil-based *p*-xylene [26]. Thus, in order to make bio-based *p*-xylene competitive to its fossil-based equivalent, several improvements along the whole process from supply-chain to process optimization are necessary.

Functional alternatives to petrochemicals and their application

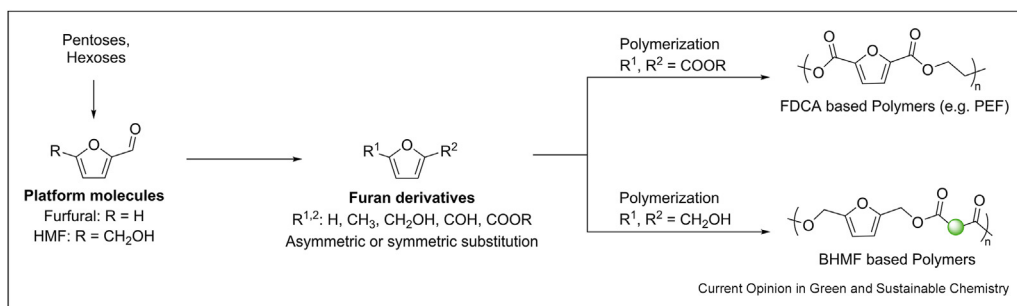
Besides the efforts made to produce already established aromatics from renewable sources, there is also great enthusiasm toward the exploration of bio-based functional alternatives. Inevitably, the implementation of new building blocks into the basis of production in the chemical industry will initially entail higher costs and efforts due to the requirement of new concepts, production lines, and consumer products. Nevertheless, the investigation of novel bio-based functional building blocks opens up new possibilities and provides room for innovative improvements. Furthermore, new functional alternatives could exhibit desired properties upon their discovery. Currently, many novel building blocks are discovered. Among these, furan derivatives and lignin-based aromatics have attracted particular attention.

Furan derivatives

Furan derivatives can not only be used for the Diels–Alder pathways toward BTX aromatics, but are also versatile and promising building blocks on their own. They maintain a central role in sustainable chemistry, as they can be prepared from abundantly available, non-edible biomass. Renewable HMF and furfural are mainly produced from hexoses and pentoses, respectively [27,28]. Both furan derivatives are used as starting materials to produce many useful building blocks (Figure 2) [27,28].

In the face of a growing plastic demand, one furan derivative that has been intensely researched in the past years is 2,5-furandicarboxylic acid (FDCA). FDCA can be produced by oxidation of HMF *via* different biocatalytic [29], chemocatalytic [30], electrocatalytic [31,32], or photocatalytic [32,33] routes. One advantage that is regularly mentioned for the electrooxidation of HMF to FDCA is the opportunity to couple it with an electroreduction that results in other value-added chemicals, like hydrogen [32]. For instance, a coupling of HMF electrooxidation to the reduction of 4-nitrophenol to 4-aminophenol was recently reported [34].

Figure 2



Bio-based platform molecules furfural and HMF and their downstream furan derivatives, some of which can be used in polymer synthesis.

FDCA has mainly gained popularity as potential alternative to terephthalic acid in the PET substitution polymer named poly(ethylene furanoate) (PEF) (Figure 2) [35]. Indeed, PEF shows properties that make it suitable for applications similar to those of PET. Compared to PET, PEF shows enhanced carbon-dioxide barrier properties accompanied by attractive thermal and mechanical properties [35,36]. These properties make PEF attractive for food and beverage packaging. In this regard, the Dutch company Avantium has put major effort into the commercialization of PEF with their YXY® technology [36,37]. Life cycle assessments and techno-economic analyses regarding HMF and FDCA have shown them to be promising, greenhouse gas emission reducing intermediates for polymer production [38]. Environmental impacts of both intermediates are expected to further decrease with advances in catalytic systems, solvent systems and the development of direct synthetic routes from lignocellulosic biomass [38]. Recent applications of FDCA also include its use in fully bio-based aromatic polyester polyols for the synthesis of polyisocyanurate rigid foams [39]. Furthermore, copolyester synthesis using FDCA and other bio-based dicarboxylic acids is frequently reported [40–42].

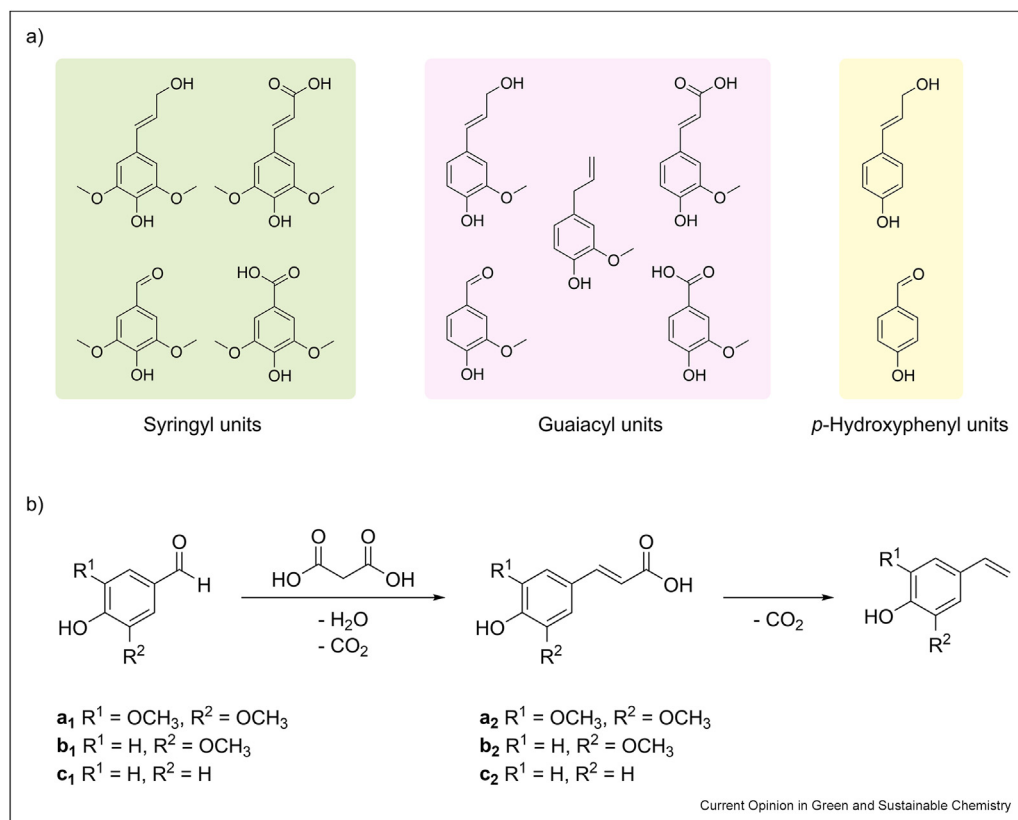
Instead of incorporating the furan building block into polyesters as dicarboxylic acid, it can also be used as a diol in the form of 2,5-bis(hydroxymethyl)furfural (BHMf). BHMf can be used as a monomer to synthesize polymers like polyesters, polycarbonates, or polyurethanes [43,44]. Furthermore, polyesters that are similar to those from FDCA with diols can be synthesized from BHMf by using the corresponding dicarboxylic acid to the diols as second monomer [44]. This way, structural isomers to FDCA derived polyesters can be obtained (Figure 2). However, it must be noted that BHMf exhibits low thermal stability, making it less suitable for high-temperature bulk polymerization [44].

Lignin based building blocks

On the subject of bio-based aromatics, lignin is considered a promising raw material. Its chemical structure is composed of a network rich in aromatic building blocks [45,46]. To yield small molecules from lignin, its highly branched network must be depolymerized. Lignin depolymerization can be performed by various thermal, chemical, and biochemical methods [45]. Thermal depolymerization of lignin is mainly carried out *via* pyrolysis, but also by combustion, gasification, or liquefaction [45]. The chemical depolymerization of lignin can be performed using acid or base catalyzed hydrolysis, oxidative or reductive methods and ionic liquids or deep eutectic solvents [47,48]. Biological methods for lignin depolymerization include the application of various bacteria, fungi, or enzymes [48]. Depending on which method of depolymerization is performed, the main products can vary [49]. Building blocks yielded from chemical and biochemical lignin depolymerization can be categorized in three groups, i.e. syringyl, guaiacyl and *p*-hydroxyphenyl units (Figure 3a) [47]. Due to unstable intermediates during lignin depolymerization, stabilization techniques are required to increase monomer yields [50]. Another challenge faced for lignin depolymerization is to find efficient separation methods that lead to pure products. Here, methods like extraction, filtration, chromatography, distillation or a combination of these techniques can be applied [49]. Nevertheless, when successfully depolymerized and purified, lignin monomers find various applications in polymer synthesis and medical or cosmetic applications [51–53]. However, since the separation and purification of the depolymerization products are quite time-consuming and cause considerable costs [49], it would also be interesting to develop applications for unseparated lignin depolymerization products.

Certain building blocks from lignin depolymerization are also interesting for producing monomers that are structurally similar to styrene (Figure 3b). One attempt

Figure 3



(a) Types of functional monomers yielded from lignin depolymerization: syringyl, guaiacyl and *p*-hydroxyphenyl units. (b) Synthesis of functional styrene alternatives from lignin derived *p*-hydroxybenzaldehydes.

here is to first produce α,β -unsaturated carboxylic acids by Knoevenagel reaction of malonic acid with different lignin derived 4-hydroxybenzaldehyde derivatives. Subsequent decarboxylation of these building blocks creates a terminal double bond that can be radically polymerized after protection of the phenol groups [54].

Conclusion and outlook

The synthesis of bio-based aromatics is an essential step for the chemical industry to combat its dependence on fossil resources. Recent literature has shown a variety of different ways to produce bio-based aromatics and to use them for further applications like building block or polymer synthesis. While methods that focus on the direct replacement of petrochemicals like BTX aromatics *via* renewable drop-in can take advantage of easier downstream utilization, new functional alternatives offer great potential that still needs to be explored. Especially on the example of PEF for replacing PET, the advantage of functional alternatives to enable innovative, and possibly better performing products and materials is emphasized. Nevertheless, the production of bio-based

aromatics still requires further innovation and progress to make it viable for large-scale industrial production.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this article.

Data availability

No data was used for the research described in the article.

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