

**WHITEPAPER**

**TOWARDS A SUSTAINABLE VALUE CHAIN  
OF LIGNIN-BASED BIO-AROMATICS IN FLANDERS**

**DISCLAIMER:**

*THIS WHITEPAPER WAS PURSUED AS PART OF THE BAFTA PROJECT,<sup>1</sup> AN INNOVATION PROJECT UNDER THE UMBRELLA OF CATALISTI, THE SPEARHEAD CLUSTER THAT ACCELERATES INNOVATION INTO BUSINESS IN FLANDERS' CHEMICAL AND PLASTICS SECTOR. THE PROJECT IS SUPPORTED BY FLANDERS INNOVATION & ENTREPRENEURSHIP (VLAIO) AND INCLUDED THE PARTNERS VITO (SCT – SEPARATION AND CONVERSION TECHNOLOGY) AND KU LEUVEN (CSCE – CENTER FOR SUSTAINABLE CATALYSIS AND ENGINEERING).*

*THIS WHITEPAPER WAS DEVELOPED WITHIN THE FRAMEWORK OF THE CATALISTI INNOVATION PROGRAM BIO-BASED VALUE CHAINS (BVC), WHICH REVOLVES AROUND USING BIO-BASED RESOURCES AS FEEDSTOCK TO DEVELOP AND PRODUCE BIO-BASED CHEMICAL PRODUCTS.*

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<sup>1</sup> <https://catalisti.be/project/renewable-chemicals/bafta/>

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## Setting the scene

### The problem: fossil carbon

Aromatic hydrocarbons, such as benzene, toluene and xylene isomers (BTX), play a crucial role in the petrochemical industry since a broad range of petrochemicals are derived from BTX aromatics. Most BTX aromatics are currently made from fossil carbon (e.g., petroleum, coal, natural gas). In 2020, the global market size of BTX aromatics is estimated at 122 million metric tons and is projected to witness a compound annual growth rate (CAGR) of 3.8% during the forecast period 2021-2026.

However, the exploitation of fossil resources as materials, chemicals and, in particular, fuels and energy results in CO<sub>2</sub> emissions (ca. 75% of the total global CO<sub>2</sub> emissions in 2016), which is a main driver of global warming. To limit global warming, and hence minimize its impact on climate change, Europe targets to reduce net greenhouse gas emissions by at least 55% by 2030. By 2050, Europe aims to become the world's first climate-neutral continent.<sup>2</sup>

Next to climate change, fossil carbon usage also has a geo-political dimension due to the uncertainty of access to easily accessible fossil resources in the short to medium term (e.g. Russo-Ukrainian War) and the limited number of countries with oil reserves, leading to raw material dependencies.

### A solution: renewable biosphere carbon

A sustainable solution for carbon-based chemicals and materials (especially for long-lived products) – not for fuels and energy – involves the transition from fossil carbon to **renewable carbon**, so-called **defossilization**. Renewable carbon entails all carbon sources that avoid or substitute the use of any additional fossil carbon from the geosphere. Renewable carbon can come from the atmosphere (CO<sub>2</sub>), biosphere (biomass) or technosphere (recycling) – but not from the geosphere (fossil carbon). Renewable carbon circulates between biosphere, atmosphere or technosphere, creating a carbon circular economy based on sustainable chemistry.<sup>3,4,5</sup> In general, unlike fossil carbon, sustainable exploitation of renewable carbon not only has the potential to achieve (i) net-zero CO<sub>2</sub> emissions, but it is also (ii) more readily accessible and more evenly distributed, creating limited dependence, and thus less geo-political frictions,

In 2020, the global renewable carbon demand predominantly entailed bio- (65%) and technospheric carbon (32%) and little to no atmospheric carbon (≤3%). Even though recycling and CO<sub>2</sub>-based products are expected to eventually overtake bio-based routes, **biospheric carbon will remain an important route towards carbon-based chemicals and materials**.<sup>6</sup> The unique value proposition of biospheric (biomass) carbon lies in the carbon skeleton that contains naturally built-in functionalities. Preserving this functionality as much as possible when processing biomass into chemicals and materials can provide significant techno-economic and life-cycle benefits compared to processing both fossil carbon as well as renewable techno- and atmospheric carbon.

For instance, lignocellulosic biomass, also referred to as lignocellulose, is an interesting feedstock to produce alternative sustainable aromatic hydrocarbons, so-called bio-aromatics. In addition to the abovementioned general benefits of biomass, additional advantages of lignocellulose are that it is abundant and non-edible, hence minimizing direct competition with primary food production. Lignocellulose is the main constituent of plant cell walls (Figure 1, left), protecting plants against pathogens and providing structural strength. In general, **lignocellulose consists of three important biopolymers**, being cellulose, hemicellulose and lignin. Cellulose (40-60 wt.%) and hemicellulose (10-40 wt.%) are composed of C<sub>6</sub> and C<sub>5</sub> sugars, respectively, whereas lignin (15-30 wt.%) is made up of bio-aromatics.\* Lignin is the richest natural source of renewable aromatics on the planet.

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\* Although the authors are aware that innovative technologies exist to convert sugars into-bio-aromatics, this falls outside the scope of this whitepaper.

<sup>2</sup> [https://ec.europa.eu/clima/eu-action/european-green-deal\\_en](https://ec.europa.eu/clima/eu-action/european-green-deal_en)

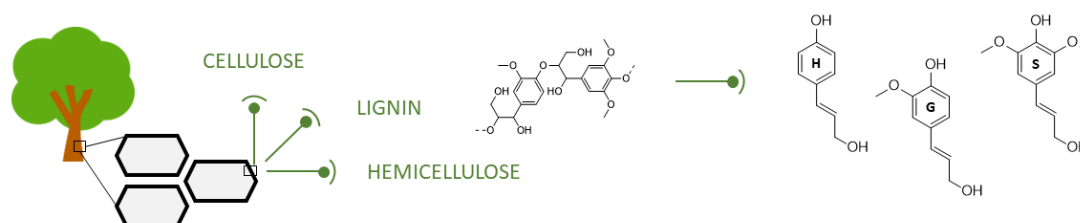
<sup>3</sup> <https://renewable-carbon-initiative.com/renewable-carbon/>

<sup>4</sup> [https://ec.europa.eu/commission/presscorner/detail/en/ip\\_21\\_6687](https://ec.europa.eu/commission/presscorner/detail/en/ip_21_6687)

<sup>5</sup> [https://ec.europa.eu/clima/eu-action/forests-and-agriculture/sustainable-carbon-cycles\\_en](https://ec.europa.eu/clima/eu-action/forests-and-agriculture/sustainable-carbon-cycles_en)

<sup>6</sup> <https://renewable-carbon.eu/publications/product/global-carbon-demand-for-chemicals-and-derived-materials-png/>

Briefly and simply explained, the **native lignin composition is composed of three common aromatic monolignols** (H, G and S, see Figure 1, right) that occur in a certain proportion depending on the type of lignocellulose, *i.e.* softwood (exclusively G), hardwood (both G and S), and grasses (G and S with little H). The natural design of all three molecules centralizes around a phenol with a propenol group in *para* position (relative to the hydroxy group). The only difference among them is the number of methoxy groups in *ortho* position, *i.e.* zero (H), one (G) or two (S). Within the lignin bio-polymer, the monolignols are predominantly connected via two main type of inter-unit linkages, *i.e.* ether (C-O-C) and carbon-carbon (C-C) linkages.



**Figure 1** | Simplified representation of the native lignin structure (here: ether linkage) as present in the cell wall of lignocellulosic biomass. The three common building blocks of lignin, so-called monolignols (indicated by H, G and S), are also shown.

Hence, native lignin represents an excellent alternative feedstock to liberate bio-aromatics that can be further applied as chemicals and materials. However, the general consensus was that (isolated) lignin, being a main waste product of the pulp and paper industry, could only be valorized in low-value applications (*e.g.* energy recovery) because of its low reactivity and degraded nature. Since the pulp and paper industry mainly aims to valorize cellulose, native lignin is indeed typically degraded upon pretreating lignocellulose into pulp and paper. In recent years, though, substantial and increasing efforts are and have been devoted to valorizing lignin into higher value applications (*e.g.* chemicals and materials) rather than just burning it as a source of process energy.

Mono-, di- and/or oligomeric bio-aromatics can be liberated by selective cleavage/depolymerization of one or both types of lignin inter-unit linkages. Typically the C-O-C linkages can be easily cleaved, whereas this is more difficult for the C-C linkages. Note that, in reality, there are even more types of linkages which add to the complexity of the depolymerization. It is important to highlight that the quality and quantity of the native lignin, including the way that the monolignol units are interconnected, depends on the type of lignocellulose (*i.e.*, softwood, hardwood, grasses), which ultimately will also have a downstream impact on refinement, processing and application.

### A value chain of bio-aromatics: similar yet different

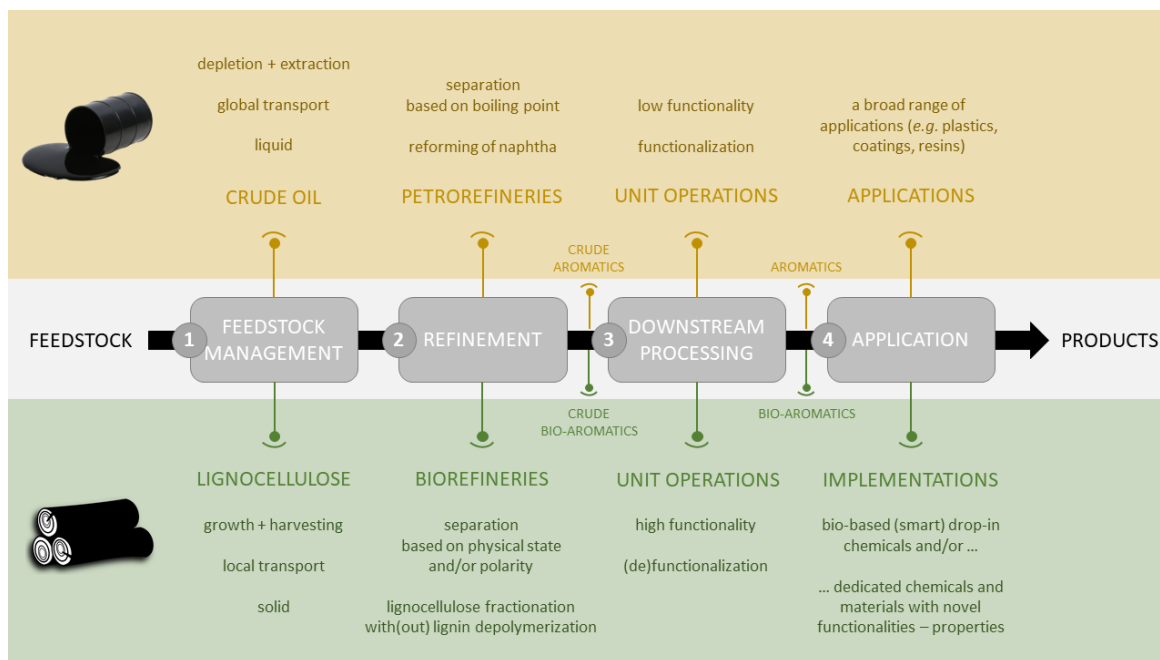
In general, the value chain of (bio-)aromatics, from feedstock to product (Figure 2, middle), encompasses **four main steps**:

- (1) **Feedstock management**: the harvest/extraction, collection, transportation and storage of suitable raw materials (*e.g.*, crude oil, lignocellulose) to (bio-)refineries, which in case of biorefining also includes the biomass growth phase;
- (2) **Refinement**: the fractionation of raw materials into useful fractions of crude (bio-)aromatics (*e.g.*, BTX, bio-BTX, isolated lignin, black liquor, bio-BTX, lignin oil), which in case of oil refining also includes the naphtha reforming and in case of biorefining typically includes lignin depolymerization (to obtain lower molecular weight bio-aromatics).
- (3) **Downstream processing**: the recovery and purification of targeted (bio-)aromatic molecules out of crude (bio-)aromatics *via* a battery of thermal, physical and (bio-)chemical unit operations and processes; and
- (4) **Application**: the implementation of the resulting processed (bio-)aromatics in a broad range of end-user products.

The traditional route (Figure 2, top) uses fossil crude oil as the feedstock (step 1). Crude oil is fractionated into useful oil fractions by distillation, after which the low-boiling naphtha fraction is reformed into crude BTX aromatics (step 2). Subsequent downstream processing of crude BTX aromatics produces an array of processed aromatics (step 3) that can be implemented in a broad range of applications (step 4).

Alternatively, the bio-based route (Figure 2, bottom) starts from renewable lignocellulose as the feedstock (step 1). Lignocellulose is fractionated into (a) useful fraction(s), after which the crude bio-aromatics

contained in the lignin fraction can be isolated, either as non-depolymerized (solid) lignin or as depolymerized (liquid) lignin (step 2). Subsequent downstream processing of crude bio-aromatics can target an array of processed bio-aromatics (step 3) that can be implemented in a broad range of applications (step 4). Even though the value chain of bio-aromatics has similarities, and can be described by such a four-step value chain as well, the practical completion and execution at every stage are different.



**Figure 2 |** General representation of the four-step value chain of (bio-)aromatics, going from feedstock to products (middle). A comparison between the traditional route based on petro-based aromatics from fossil carbon (top) and the envisioned route based on bio-based aromatics from renewable biosphere carbon (bottom).

In the next four chapters, the similarities and, in particular, the differences between the traditional fossil route and the envisioned renewable route are discussed in more detail for each step. Moreover, the main challenges and requirements, respectively, that must/should be tackled and fulfilled will be highlighted. For each step a non-limiting list of potential partners for further establishing the value chain of bio-aromatics will be suggested as well.

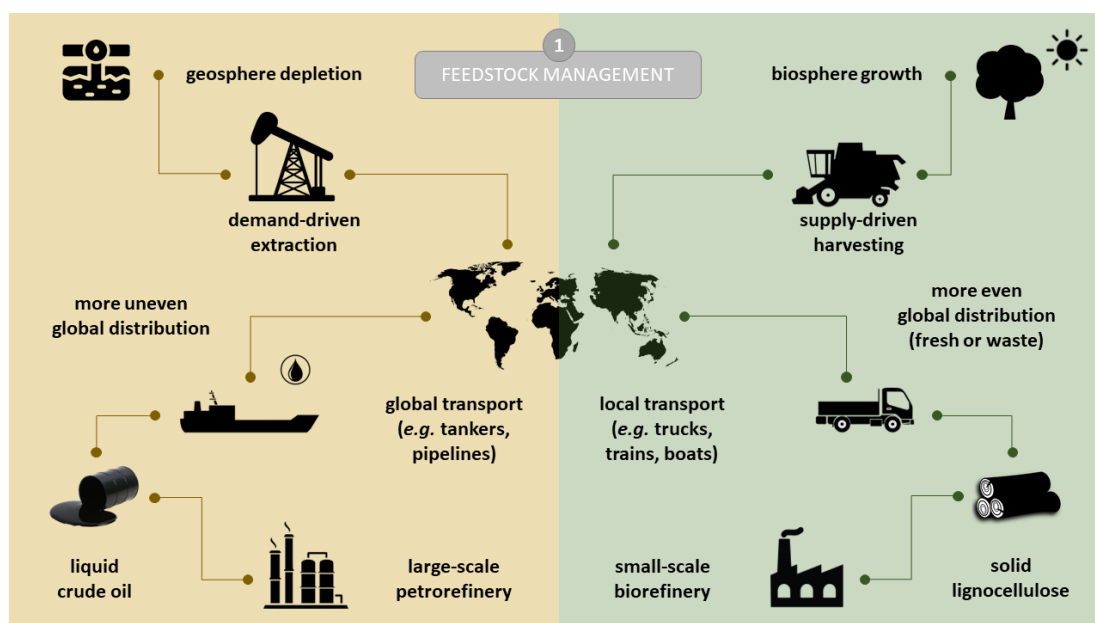
## 1. Feedstock management

### Similarities and differences

In general, feedstock management includes the harvest/extraction, collection, transportation and storage of suitable raw materials (e.g., crude oil, lignocellulose) to (bio-)refineries, which in case of biorefining **also includes a lignocellulose growth phase** (in case primary wood/plants are used) (Figure 3). For managed forestry/plant culture to be sustainable, it must address 'green' issues, such as biodiversity loss (related to controlled and limited crop variability), water use, and/or the crop protection products (fertilizers, pesticides, etc.).

Another aspect that is different for lignocellulose concerns the **harvest and storage of feedstock before refining**. With wood, this is not such a problem, particularly if the wood is stored water saturated or dried (<10% moisture content) – both of which can be a problem. However, since wood can be harvested from forests on a continuous year-round basis, stocks in the refinery yards can be held to a minimum to avoid rotting. In contrast, agricultural waste is usually only harvested twice per year, and hence the feedstock needs to be stored up to six months (for efficient working of the refining unit). Meanwhile, the feedstock can rot both aerobically and non-aerobically (by white or brown rots) and will be increasingly consumed by rodents, which will not only reduce the quantity, but more importantly, the quality of the feedstock.

Fossil resources are extracted only in a limited number of countries around the world. The largest proven crude oil reserves are located in Venezuela, Saudi Arabia, Canada, Iran, Iraq and Kuwait, and 72% of the total world oil in 2021 was produced by only 10 countries.<sup>7</sup> By consequence, due to this uneven global distribution, many countries depend on these countries to deliver sufficient feedstock. In contrast, lignocellulosic biomass is **more evenly distributed** around the globe. Lignocellulosic feedstock can be fresh wood, agricultural residues or even pre- and post-consumer waste streams. So, instead of liquid crude oil, the bio-case will use **solid lignocellulose**, which has implications on feedstock management.



**Figure 3** | A comparison between the traditional fossil-based (left) and the envisioned bio-based (right) route to produce (bio-)aromatics, focusing on feedstock management.

While global transportation of liquid crude oil is typically organized over longer distances (e.g., tankers, pipelines), solid lignocellulose/lignin streams are more likely to be **locally transported in a multimodal fashion** over shorter distances (e.g., trucks, train, river boats). Since wood transportation typically is only efficient if branches/twigs are removed from the trunk of trees, adapted methods are required to increase the usable crop yield. The transported volume of dry (and especially wet) biomass compared to fossil resources (e.g. liquid oil) per unit of aromatics is indeed estimated to be larger. Aforementioned boundary

<sup>7</sup> <https://www.eia.gov/international/data/world/petroleum-and-other-liquids/annual-petroleum-and-other-liquids-production>



conditions imply that both the location and the size as well as the logistics of biorefineries will most likely differ from current petrorefineries. Ideally, the location of a biorefinery is close to the source of the lignocellulose. As a result, the throughput of a more decentralized **small-scale biorefinery** (e.g. 1,000 ktonnes/year) is expected to be much lower compared to a traditional centralized large-scale petrorefinery (e.g. 20,000 ktonnes/year). In this regard, important lessons can be drawn from the current pulp and paper industry, which up to now sustain a successful business with the same feedstock (management).

Independent of the route, either fossil or bio, feedstock stability is key to guarantee and safeguard the quality of the end products, and thus **feedstock variability** should be as small as possible. In other words, the feedstock's composition and level of impurities should lie between certain specification limits to assure easy, consistent and efficient refining and hence reproducible production of the targeted bio-aromatics. Important challenges in this regard are the solid (and hence intrinsically more heterogeneous) nature, seasonal variability, feedstock storage (e.g. rotting) type (i.e., softwood, hardwood or grasses) and origin of biomass.

### State of the art

Feedstock details such as availability, location and prices can be consulted in literature. Moreover, in the completed *Bio-Aromatics Feedstock and Technology Assessment* (BAFTA) VIS project, which ran from 2018 until mid-2019, a feedstock database was constructed, listing the most crucial parameters for selected feedstock classes (i.e., fresh wood, pre- and post-consumer waste wood and isolated lignin).

An important first feedstock class to be considered is **fresh wood**. Worldwide fresh wood is mainly used in material applications, such as pulp and paper industry and construction. In Flanders, forests consist of coniferous (27%), deciduous (54%) and mixed wood (19%). This corresponds with a total stock of 44.52 Mm<sup>3</sup> wood, of which 0.85 Mm<sup>3</sup> is harvested every year. Currently, the annual increment of wood is 1.42 M<sup>3</sup>/year.

The availability of woody resources is much more extensive if also **waste wood** is taken into account. Two major subclasses can be distinguished, being pre-consumer and post-consumer waste wood. Large amounts of **pre-consumer waste wood** are available from the wood processing industry (e.g., sawdust, chips, bark, twigs and off-cuts), the pulp and paper industry (e.g. bark) and agricultural industry (e.g., straw and other crop culms/stems). Serious challenges with regard to refining pre-consumer waste residues include knots, insect damaged or killed wood, bark (e.g. extractives, suberin), root content, mud contamination, the waxy outer layer of the culms and the low density pith present in the lumen. In contrast to pre-consumer streams, **post-consumer waste wood** (e.g. construction waste, wood from recycling centers) typically contains a certain degree of impurities. While some resources can be quite clean, others can contain paints, glues, heavy metals, fire retardants and/or wood preservatives. The market of waste wood is present in the European union and often these streams are transported between EU countries. Waste wood streams are to some extent reused and/or recycled into material applications (e.g. particle board), but, in contrast to fresh wood, mainly consumed as energy carrier. In 2015, the quantity of waste wood in Flanders was approximately 1,500 ktonnes.

Whilst it is correct to consider these materials as waste compared to the primary crops, some are currently used extensively as, for instance, energy/fuel, animal feed and bedding, and natural re-fertilization. If such feedstocks are to be used to produce potentially higher-value chemicals and materials, then **solutions for the loss of feedstock for the current purposes** will need to be realized. Also, the current use of 'waste' wood for energy/fuel is important for a number of reasons. Firstly, paper and pulp factories are often use these feedstocks to be self-sufficient in energy, to such an extent that they are not even connected to the energy grid (in particular when they are in remote regions). Secondly, the current cost of energy/fuel is extremely high, and hence the option towards chemical and materials needs to be valued against the cost of energy/fuel.

### Value chain requirements

Feedstocks to produce lignin-derived bio-aromatics should be harvested in a sustainable manner, and ideally:

- in sufficient amounts all year round;
- display a stable and origin-independent composition with a tolerable degree of impurities; and
- be efficiently transported to a local biorefinery.



### **Possible partners**

A broad range of partners can be valuable to manage the feedstock. A non-limiting list of potential partners is provided below:

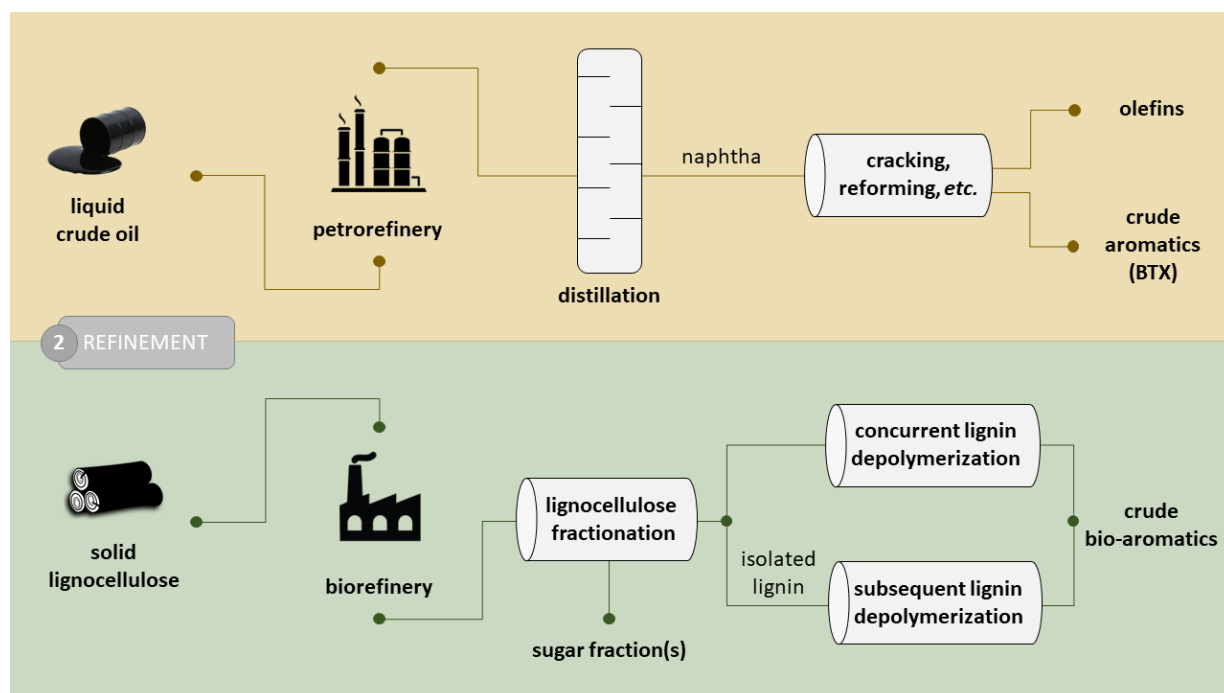
- forest owners (fresh wood);
- sustainable land and forest management organizations (fresh wood);
- agricultural industry (lignocellulosic residues);
- feed/food (processing) industry (lignocellulosic residues);
- material companies (e.g. pulp and paper industry) at different levels of wood processing (pre-consumer streams); and/or
- waste wood collecting companies (post-consumer streams).

## 2. Refinement

### Similarities and differences

In general, refinement includes the fractionation of raw materials (e.g., crude oil, lignocellulose) into useful fractions of crude (bio-)aromatics (e.g., BTX, bio-BTX, isolated lignin, black liquor, bio-BTX, lignin oil), which in case of oil refining also includes naphtha reforming and in case of biorefining **typically includes lignin depolymerization**.

The petrorefinery fractionates liquid crude oil into several streams, going from lighter to heavier fractions **based on boiling point**, by using a distillation unit (Figure 4, top). One of the lighter fractions is called naphtha, which has a chain length ranging from 5-12 carbon atoms. Cracking and reforming of naphtha creates unsaturated fractions of which olefins and crude BTX aromatics are the most important ones.



**Figure 4** | A comparison between the traditional fossil-based (top) and the envisioned bio-based (bottom) route to produce crude (bio-)aromatics, focusing on feedstock refinement.

The aim of a biorefinery is similar to that of a petrorefinery (Figure 4, bottom). However, the fractionation of lignocellulose is completely different, and is, in first instance, **not relying on differences in boiling point**. This is because lignocellulose is a quite complex matrix of multiple solid (*i.e.* non-volatile) biopolymers with various chemical structures and properties. As a result, it is much more challenging to find a well-balanced fractionation technology that simultaneously keeps degradation of the original chemical structure at a minimum. After fractionation, the lignin fraction is typically depolymerized into bio-aromatics with lower molecular weights, but this is not compulsory since isolated lignin with higher molecular weight can be directly applied as well. Besides the refinement technology, the corresponding reactor concept needs to be well-designed, taking into account the various aspects of lignocellulose refinement, such as the handling of solid feedstocks, the process conditions used, whether or not in presence of solvents, additives and/or catalyst (see section *State of the art* for more details).

In the end, **each fraction generated should have a well-defined value chain and a solid business case to contribute to the overall economic feasibility**. The petrorefinery, for instance, valorizes light hydrocarbons into LPG, naphtha into olefins, aromatics and gasoline, and heavier hydrocarbons into diesel and kerosene. The same goes for a biorefinery. In other words, the renewable character of the feed will not be enough as such. The transition towards a value chain of bio-aromatics can only be sustainable if each fraction generated is valorized with a maximal retention of carbon.

Importantly, in addition, a match should be found between feedstock availability (step 1 of the value chain) and the market of bio-aromatics (step 4), taking into account the process efficiencies of intermediate refinement (step 2) and downstream processing (step 3).

### State of the art

In literature, **more than 10 types of refining technologies** to fractionate lignocellulose into different intermediate product streams are described based on thermal and/or (bio-)catalytic conversion strategies. All of them have advantages, but also drawbacks. While thermal strategies (e.g. pyrolysis) also yield aromatics, (bio-)catalytic strategies are preferred if the aim is to preserve functionality and narrow the product scope. A key distinction is that some (one-step) refining technologies apply **simultaneous depolymerization** during fractionation, while other (two-step) refining technologies apply **subsequent depolymerization** on isolated lignin after fractionation to produce crude bio-aromatics (Figure 4, bottom).

#### Lignocellulose fractionation with(out) simultaneous depolymerization

The most important **lignocellulose refining technologies** are highlighted in Table 1. Lignocellulose refinement typically yields both lignin and carbohydrate (cellulose and/or hemicellulose) products.

**Table 1** | Overview of different technologies for lignocellulose fractionation with(out) depolymerization.

Technology*	TRL	Lignin** (physical state, product yield)	Cellulose	Hemicellulose
Kraft process	9	C (solid, >90%)	in pulp (good quality fibers)	-
Sulfite process	9	C (solid, >70%)	in pulp	dissolved and depolymerized
Soda process	8-9	C (solid, >80%)	in pulp	extracted and depolymerized
Hydrolysis	6-9	PP (solid, up to 60%)	removed as soluble sugars	removed as soluble sugars
Pyrolysis	3-8	C/D (solid/liquid, n.a.)	-	-
AFEX	6-7	PP (solid, up to 65%)	in pulp	mainly in pulp
Organosolv	3-7	PP (solid, 70-85%)	in pulp	in pulp or removed as soluble sugars
AAP	3-5	PP (solid, <65%)	in pulp	partially extracted and depolymerized
ARP	3-5	PP (solid, 75-85%)	in pulp	partially extracted and depolymerized
Acidic flow-through	3-5	PP (solid, 70-85%)	in pulp	extracted, depolymerized and partially degraded
AAF	3-5	P (solid, 70-80%)	in pulp	extracted and depolymerized
RCF	3-5	D (liquid, 50-100%)	in pulp	in pulp or removed as soluble sugars
OCF	3-5	D (liquid, n.a.)	in pulp	-

\* AFEX, ammonia fiber expansion; AAP, anhydrous ammonia pre-treatment; ARP, ammonia recycle percolation; AAF, aldehyde-assisted fractionation; RCF, reductive catalytic fractionation; OCF, oxidative catalytic fractionation. \*\* C, condensed; PP, partially preserved, P, preserved; D, depolymerized. Abbreviations: n.a., not available.

For the **lignin-derived products**, although significant variations between different technologies exist, product yields in general are relatively high (>50-100% based on the initial lignin content). Moreover, important chemical and physical differences (e.g., ether linkage content, physical state) are found among the end-products obtained by each technology:

- (solid) condensed lignin with limited structural preservation and limited residual ether linkages (i.e., Kraft, sulfite and soda processes);
- (solid) lignin with partial structural preservation (i.e., AFEX, AAP, ARP, acidic-flow through, (mild) organosolv and hydrolysis processes);
- (solid) lignin with full preservation of ether linkages (i.e., AAF);
- (solid and/or liquid) pyrolysis products consisting of oxygenated bio-aromatics (i.e., pyrolysis-related processes); and
- (liquid) lignin oil containing monomeric and oligomeric bio-aromatics obtained by one-step simultaneous fractionation-depolymerization (i.e., RCF and OCF processes).

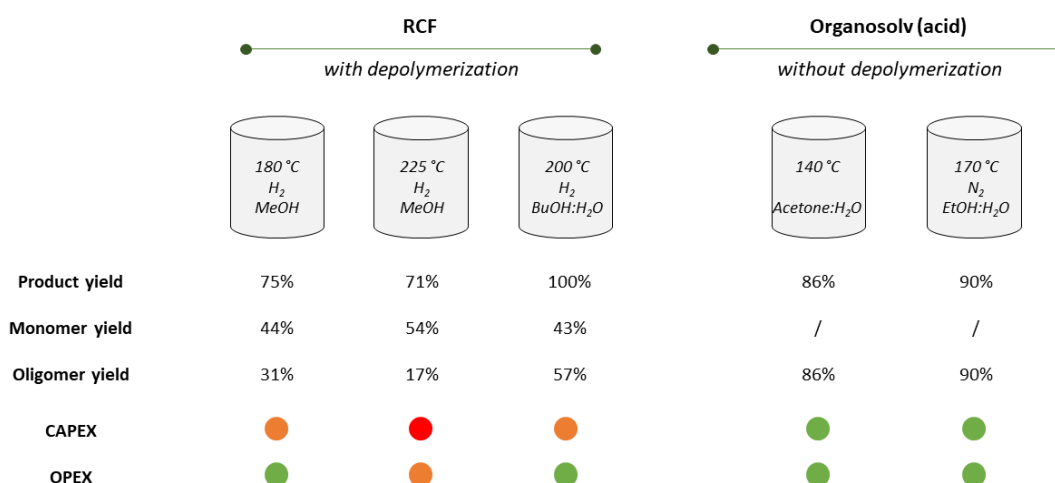
For the **carbohydrate fraction**, in general, these technologies provide a high retention of cellulose in the pulp as well, with the hemicellulose either being retained in the pulp or extracted(-depolymerized) into soluble sugars. Overall, major differences in cellulose and hemicellulose structure exist amongst different technologies. Pulp properties should therefore always be taken into account since establishing bio-based value chains also requires valorization of the (hemi)cellulose, which accounts for 70-85% of the mass.

For subsequent downstream processing steps in the value chain, it is important to highlight that some fractionation technologies introduce **impurities** (e.g. sulfur in case of Kraft or sulfite pulping, nitrogen in case of ammonia-based techniques). Besides, some impurities arise from the starting feedstock itself, being either originally present or contaminated through, for instance, glues or paints in post-consumer wood. Such impurities can have an adverse impact on the overall process efficiency, and thus the overall cost, of the entire value chain and/or the final product quality.

**Well-established technologies**, which operate at high technology readiness level (TRL; 8 or 9), mainly focus on high cellulose yield with good fiber quality after fractionation. For instance, the Kraft process – operating at 155-175 °C for 1-3 h under 5-10 bar with water as solvent and Na<sub>2</sub>S - NaOH as catalysts – is the benchmark process to produce high quality fibers for the paper industry. Although the product yield is quite high, the major drawback of the Kraft process, as well as others high-TRL technologies (e.g., sulfite and soda process), is the production of a **highly condensed lignin structure**. In other words, the generated isolated lignin contains only limited residual ether linkages (through depolymerization – repolymerization mechanism towards mainly C-C linkages), which hampers subsequent selective lignin depolymerization. Selective lignin depolymerization is of paramount importance to liberate crude bio-aromatics suitable for downstream processing into targeted bio-aromatics (step 3).

Recently, several **new technologies** were developed to either **preserve the native lignin structure** (and hence the C-O-C linkages) as much as possible during fractionation (e.g., AAF) or to **avoid repolymerization after depolymerization** through active stabilization (e.g., RCF or OCF). This way of working is known as a lignin-first biorefining, targeting the maximal valorization of lignin. From a chemical point of view, both type of technologies enable production of functionalized bio-aromatics. Due to the novelty of these processes, these strategies are currently operating at lower TRLs, ranging from 3 to 5. In the future, important upscaling steps should be taken in order to operate at the same TRL as the established processes, mainly focusing on cellulose valorization.

Within the BAFTA project,<sup>8</sup> two dedicated lignocellulose refinement strategies were selected: (i) one resulting in high yields of crude bio-aromatic monomers, i.e. (one-step) RCF under H<sub>2</sub> atmosphere; and (ii) one providing high yields of crude bio-aromatic oligomers, so-called organosolv lignin, i.e. (two-step) mild acidic organosolv processing. Within each strategy, three (RCF) or two (organosolv) variations were selected and evaluated on several key performance indicators (KPIs; Figure 5). The technological KPIs evaluated in BAFTA were yield, CAPEX, and OPEX, as well as the process conditions.



**Figure 5** | Schematic representation of the evaluated and scored KPIs for two selected lignocellulose refining technologies within the BAFTA project. Yields are calculated and expressed on basis of initial lignin content.

<sup>8</sup> <https://catalisti.be/project/renewable-chemicals/bafta/>

As stated above, the full potential of the selected lignocellulose refining technologies will depend on the valorization of the sugar-related streams as well. Hence, the quality of the obtained carbohydrate pulp should either be sufficient to serve as a drop-in material for traditional pulp and paper industry, or should be amenable for chemical conversion towards valuable drop-in (e.g., ethanol) or dedicated chemicals. In other words, rather than ambitioning either a cellulose-first or a lignin-first approach, preferably a carbon-first biorefinery must be envisioned. Moreover, the economic feasibility of such a biorefinery, to **maximally valorize each individual stream by matching feedstock availability with applications** (on properties, scale, price, etc.), should be evaluated, however, without ignoring the techno-economic requirements of the derived products.

### Isolated lignin depolymerization

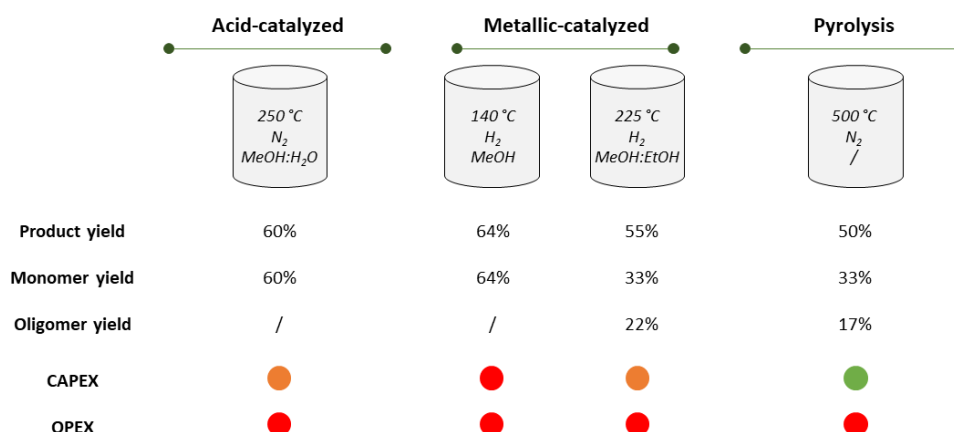
If depolymerization of lignin does not occur during lignocellulose fractionation, the **(solid) isolated lignin**, such as obtained from the pulp and paper industry and upcoming second-generation cellulosic ethanol plants, could be further depolymerized in a subsequent step to obtain lignin-derived bio-aromatics with lower molecular weight with a potentially broader application scope (and mostly higher value) than the starting material.

Some kinds of isolated lignin are already produced at an industrial scale after a refining step (step 2 of the here described value chain; Table 2 **Fout! Verwijzingsbron niet gevonden.**). Here, only a limited number of companies are generating this kind of lignin streams and typically they are mainly used as energy carrier.

**Table 2** | Overview of global availability of different types of isolated lignin.

Feedstock subtype	Ligno-sulfonate	Hydrolysis lignin	Kraft lignin	Soda lignin	High purity lignin	Organosolv lignin
Global availability (kton/year)	1,098	274	96.5	10	8.5	3

In literature, the following **isolated lignin depolymerization strategies** are described: base-catalyzed, acid-catalyzed, metal-catalyzed, ionic liquid-assisted, supercritical fluid-assisted, solvolytic, fast and slow pyrolysis, catalytic fast pyrolysis and biological depolymerization technologies. While some technologies are already commercialized (e.g., pyrolysis), most available depolymerization strategies are currently operating at an intermediate TRL of 3 to 5. In line with the two selected lignocellulose refining technologies (**Fout! Verwijzingsbron niet gevonden.** and Figure 5), three promising lignin refining technologies (with two variations for metallic-catalyzed) were selected in the BAFTA project as well (Figure 6). Again, the technological KPIs evaluated were yields, CAPEX, and OPEX, as well as the process conditions.



**Figure 6** | Schematic representation of the evaluated and scored KPIs for three selected lignin depolymerization technologies within the BAFTA project. Yields are calculated and expressed on basis of initial lignin content.

### Value chain requirements

Ideally, biorefineries converting lignocellulose into crude bio-aromatics are based on:

- technologies to efficiently extract, and fractionate/purify lignin (with high product yields and low amounts of unwanted side streams and/or impurities) with or without direct depolymerization into lignin-derived bio-aromatics; and
- technologies generating product classes (lignin as well as sugar-related products) for which relevant value chains and business cases already exist or could be established.

### **Possible partners**

Whereas companies fractionating lignocellulose into isolated lignin [and sugar fraction(s)] are established or in development, companies (in)directly depolymerizing (isolated) lignin into crude bio-aromatics are scarce. A non-limiting list of potential partners is provided below:

- pulp and paper industry;
- 2<sup>nd</sup> generation ethanol plants;
- companies producing chemicals and/or fuels from biomass resources; and/or
- companies producing chemicals and/or fuels from fossil resources.

Based on the complementary expertise of these partners (*e.g.*, feedstock management, solids processing, refining, downstream processing, catalysis, *etc.*) – as highlighted throughout this white paper, a joint effort of all these partners should be encouraged to realize future biorefineries.

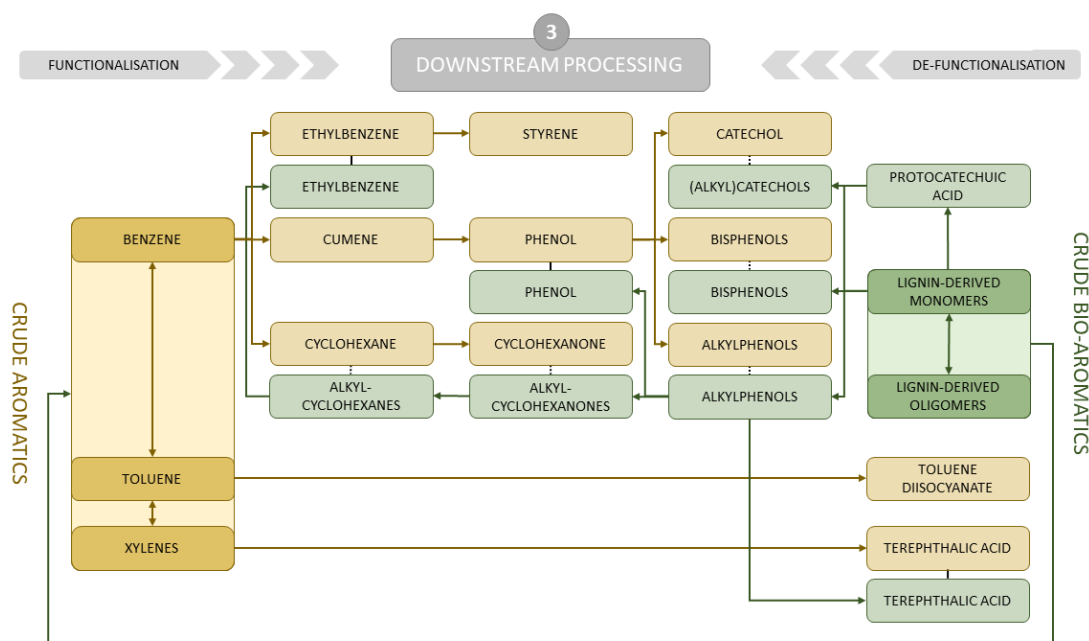
### 3. Downstream processing

#### Similarities and differences

In general, downstream processing includes the recovery and purification of targeted (bio-)aromatic molecules out of crude (bio-)aromatics via a battery of thermal, physical and (bio-)chemical unit operations and processes.

After traditional refining of fossil oils, the resulting **mixtures of crude aromatics** undergo downstream processing into a wide range of fossil-based aromatics (Figure 7, brown), which serve as important precursors for numerous applications (see section 4). Since crude BTX aromatics are typically **simple and bear limited functional groups**, the **chemical unit processes are aimed towards functionalization** (e.g., oxidation, alkylation, (de)hydrogenation). For example, benzene can be converted into alkylphenols in three steps by successive alkylation to cumene, oxidation to phenol and (a second) alkylation to alkylphenols, whereas *p*-xylene can be converted into terephthalic acid by repetitive catalytic oxidation. In addition, **boiling points are low** (<200 °C), which enables gas-phase processing and thermal unit operations (e.g., distillation).

In contrast to crude BTX aromatics, depending on the type of refinement, the resulting crude bio-aromatics are typically **more complex and bear extensive functional groups** and comprise a **broad family of molecules**, ranging from aromatic monomers to oligomers. Also, depending on the type of refinement, either a plethora of defunctionalized molecules (closely resembling BTX aromatics) or a handful of functionalized molecules (closely resembling monolignols) are obtained. In strong analogy, downstream processing of crude bio-aromatics into specific bio-aromatic fractions (Figure 7, green) also requires both thermal, physical and chemical unit operations, but with a different focus. Next to functionalization (e.g., coupling), **chemical unit operations should also be directed towards de-functionalization** (e.g., cleavage). For instance, lignin-derived monomers can be defunctionalized into bio-phenol by subsequent demethoxylation (to alkylphenols) and dealkylation (to phenol). Also, in general, **boiling points are higher** (>200 °C), which favors liquid-phase processing and physical unit operations (e.g., extraction, membrane filtration) over thermal unit operations.



**Figure 7** | A comparison between the traditional fossil-based (brown) and the envisioned bio-based (green) routes to downstream process crude (bio-)aromatics into similar or identical aromatic bulk chemicals by (de-)functionalization. Note that the examples are non-limitative and incomplete and, for reasons of clarity, mainly focus on processing lignin-derived monomers. In addition, lignin-derived di-/oligomers can be (i) processed into monomers by breaking down the interunit C-C linkages, (ii) processed as such by identical or similar unit operations/processes or (iii) applied directly in selected applications.



In other words, a successful transition from fossil to bio-based aromatics implies the development and implementation of new knowledge and technology on downstream processing of crude bio-aromatics in the traditional refining industry.

To give bio-based aromatics an edge over fossil-based aromatics, and to go beyond mere renewability, it might be most alluring to retain the intrinsic functionality as much as possible, which can eventually facilitate downstream processing. Depending on the envisaged application, making use of this functionality, it can (i) reduce the number of reaction steps, (ii) increase product selectivity, and/or (iii) limit process severity towards existing aromatics compared to traditional routes. For example, bio-based alkylphenols can be selectively produced in one safe step from lignin-derived monomers while the synthesis of traditional alkylphenols from benzene requires multiple steps of which some are more dangerous and/or non-selective.

### State of the art

Crude bio-aromatics – being either mono-, di- and/or oligomers – in which the functionality is maximally preserved upon refinement can in general be classified as **substituted methoxyphenols**. The type of substituent (e.g., aldehyde, alkyl, alcohol, acid, ether) depends on the applied refinement technology. Examples of monomeric substituted methoxyphenols include, but are not limited to, vanillin and/or syringaldehyde; alkylguaiacols and/or -syringols; alkanolguaiacols and/or -syringols; vanillic and/or syringic acid. More stringent refinement conditions typically lead to a loss of functionality, producing related but slight different molecules, such as guaiacol and/or syringol or even less functionalized molecules (e.g., cresols, phenol, benzene, toluene, xylenes).

Downstream processing of (functionalized) crude bio-aromatics is a topical and relatively young field of research. The resulting bio-aromatics can be identical (drop-in), similar (lookalikes) or rather different (novelties) compared to existing aromatics. Some promising examples of thermal, physical and (bio-)chemical unit operations/processes towards targeted bio-aromatics are described below. Note that the examples are non-limitative and incomplete.

For reasons of clarity, the (bio-)chemical unit operations are mainly described for processing of crude bio-aromatic monomers (Figure 7). Still, identical or similar unit operations/processes can be directly applicable to the structurally related crude di-/oligomeric fraction as well, which might reduce the total number of unit operations. Alternatively, di-/oligomeric fractions can also be further broken down into additional crude bio-aromatic monomers by cleavage of the remaining more stable interunit C-C linkages.

Thermal and physical unit operations/processes:

- Solvent **extraction**: mono/mono- and/or mono/oligo-separation based on green solvent affinity
- Vacuum, extractive and/or reactive **distillation**: mono/mono- and/or mono/oligo-separation based on boiling point<sup>9</sup>
- Membrane **filtration**: mono/mono- and/or mono/oligo-separation based on molecular size<sup>10,11,12</sup>
- Evaporative, cooling and/or reactive **crystallization** (or precipitation): mono/mono- and/or mono/oligo-separation based on (in)solubility and intermolecular forces
- **Centrifugation**: mono/oligo-separation based on density

Chemocatalytic unit operations/processes:

- **Demethylation** to 4-alkylcatechols and/or 4-alkylpyrogallols, which can be coupled to:
  - dealkylation to catechol and/or pyrogallol<sup>13</sup>
  - de-2-carboxyvinilation to catechol and/or pyrogallol<sup>14</sup>
- **Demethoxylation** to 4-alkylphenols, which can be coupled to:
  - dealkylation to phenol<sup>15</sup>

<sup>9</sup> <https://doi.org/10.1039/C7GC02989F>

<sup>10</sup> <https://doi.org/10.1016/j.seppur.2016.12.033>

<sup>11</sup> <https://doi.org/10.1515/hf-2018-0303>

<sup>12</sup> <https://doi.org/10.1016/j.indcrop.2016.10.005>

<sup>13</sup> <https://doi.org/10.1002/cssc.202102248>

<sup>14</sup> <https://doi.org/10.1002/anie.201913023>

<sup>15</sup> <https://doi.org/10.1126/science.aau1567>

- isomerization to 3-alkylphenols<sup>16</sup>
- full hydrogenation to 4-alkylcyclohexanols<sup>17</sup>/-hexanones<sup>18</sup>
- full hydrodeoxygenation to 4-alkylcyclohexanes<sup>19</sup>
- full hydrodeoxygenation and dehydrogenation to 4-alkylbenzenes<sup>20</sup>
- carbonylation and oxidation to terephthalic acid<sup>21</sup>
- **C-C coupling** to new bisphenols<sup>22</sup>
- Dehydrogenative decarbonylation and deoxygenation to 4-alkylbenzenes<sup>23</sup>

Biocatalytic unit operations/processes:<sup>24,25</sup>

- **Benzylic oxidation** to 4-hydroxybenzoic, vanillic and/or syringic acid, which can be coupled to:
  - Hydroxylation and/or demethylation to protocatechuic acid
    - Decarboxylation, hydroxylation or demethylation to catechol
- **Hydroxylation and/or demethylation** to catechol

Also, it is noteworthy to mention **biocatalytic and/or chemocatalytic funneling** as a tool to process crude bio-aromatic mixtures into specific products or fractions. Funneling relies on product simplification rather than product separation by converting complex mixtures into single chemical species based on central biochemical metabolites (e.g., protocatechuic acid) and/or chemical intermediates (e.g., phenol).

Note that the resulting bio-aromatics are mostly linked to the value chain of phenol, which is a logical consequence of the fact that lignin biopolymer is composed of phenolic units. Still, processing towards non-phenolic bio-aromatics, such as styrene, terephthalic acid, and aniline derivatives, which are not part of the value chain of phenol, is also possible.

As soon as the crude and/or processed bio-aromatic monomers, dimers and/or oligomers – either pure or as a mixture – are available, in some specific cases, they can be used as such. If not, the necessary additional processing steps must be performed to match the properties with the selected applications.

Finally, it will be key to perform techno-economic analyses and life cycle assessments to evaluate the impact on implementing additional downstream processing steps on the economic feasibility and environmental impact to produce certain bio-aromatics from the crude mixture, compared to the traditional fossil-based routes. Indeed, every additional unit operation/process adds costs to the production of bio-aromatics, greatly influencing the economic viability of the transition towards bio-based aromatics.

### Value chain requirements

Ideally, the thermal, physical and chemical unit operations/processes applied in downstream processing of lignin-derived crude bio-aromatics are:

- economic and cost-efficient;
- robust and applicable on mixtures instead of pure substrates (*i.e.* funneling);
- adhering to the principles of green chemistry; and
- exploit and benefit of the existing natural structure and functional groups (*i.e.* atom efficiency).

### Possible partners

The potential partners for downstream processing seem to be straightforward and involve established chemical producers as well as new companies and start-ups. Besides that, companies with expertise in fermentation and (bio-, chemo- or photo-)catalysis could prove to be valuable.

<sup>16</sup> <https://patents.google.com/patent/WO2016187678A1/de>

<sup>17</sup> <https://doi.org/10.1021/acssuschemeng.6b01580>

<sup>18</sup> <https://doi.org/10.1002/cssc.201403375>

<sup>19</sup> <https://doi.org/10.1038/ncomms11162>

<sup>20</sup> <https://doi.org/10.1039/C9GC04246F>

<sup>21</sup> <https://doi.org/10.1002/anie.201814284>

<sup>22</sup> <https://doi.org/10.1039/D1PY00909E>

<sup>23</sup> <https://doi.org/10.1016/j.apcatb.2019.118143>

<sup>24</sup> <https://doi.org/10.1073/pnas.1410657111>

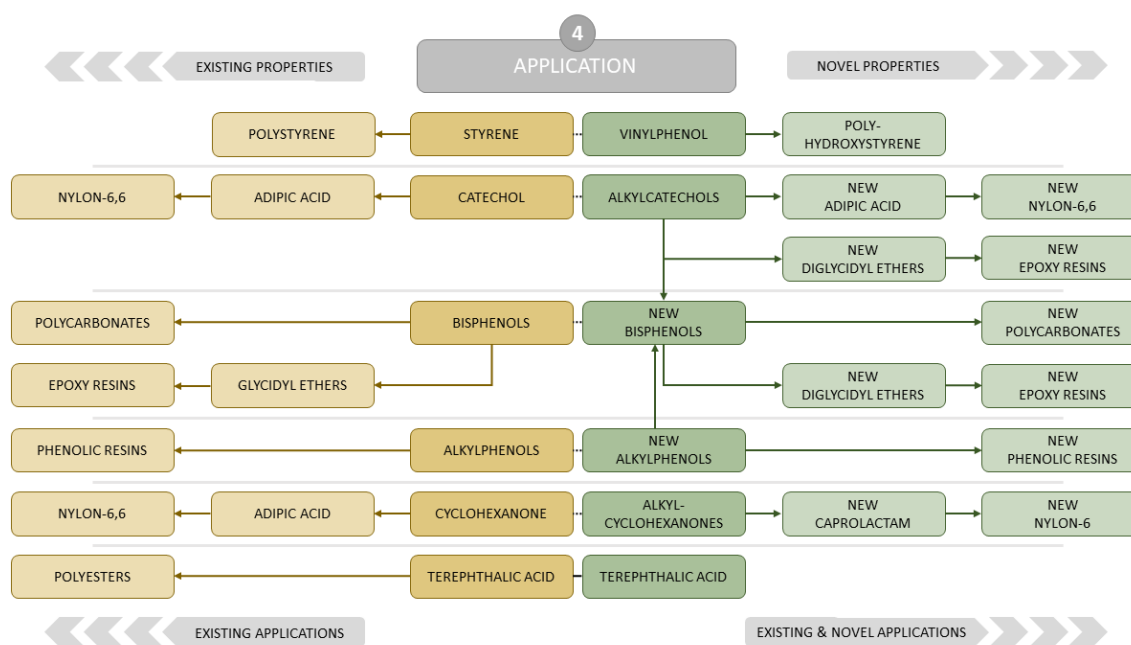
<sup>25</sup> <https://doi.org/10.1038/s41467-021-22556-8>

## 4. Application

### Similarities and differences

In general, application includes the implementation of the resulting processed (bio-)aromatics in a broad range of end-user products (Figure 8). Selective production of (smart) drop-in chemicals (e.g., phenol, terephthalic acid; see Figure 7) can be seen as an easy route to replace fossil aromatics by bio-aromatics with **identical properties in existing applications**. However, for drop-in applications it is crucial to produce bio-based twins *via* downstream processing (step 3 in the value chain), which might (often) be complex and cumbersome. From a green chemistry point of view, this route doesn't seem to be the best option since it downgrades the functionality (*i.e.* low atom efficiency).

In some cases, a more convenient and complementary route might be to apply new bio-aromatics with **novel properties in either existing or novel applications** (Figure 8). In existing applications, such chemical structures, containing typical bio-based functionalities, can potentially improve existing favorable properties and/or ameliorate existing unfavorable characteristics (e.g., recyclability, biodegradability, toxicity) leading to bio-based applications with improved performance compared to their fossil-based counterparts. Going beyond existing applications, in novel applications, bio-aromatics might prompt innovative products with non-existing properties, which can only be achieved due to the natural functionalities in bio-aromatics. Also, the available range of molecular weight within the bio-aromatics, being monomers, dimers and/or oligomers, enables to tailor the bio-products for a broad range of applications. In other words, the value added to applications can, and preferably should, extend beyond merely switching to a renewable carbon source.



**Figure 8** | A comparison between the traditional fossil-based (brown) and the envisioned bio-based (green) routes to apply (bio-)aromatics into existing/novel polymeric applications with existing/novel properties. Note that the examples are non-limitative and incomplete and, for reasons of clarity, mainly focus on applying processed monomers. In addition, processed di-/oligomers as well as unprocessed mono-, di- and/or oligomers can be applied in certain applications.

Indeed, the fossil-to-renewable transition can provide a window of opportunity to substitute hazardous substances by less dangerous ones, enabling companies to comply with strict EU regulation (REACH). For instance, bio-based functional groups might be exploited to design endocrine disruption out of the next generation of chemicals (e.g., alkylphenols and bisphenols). In this regard, in particular the higher molecular weight bio-aromatic oligomers are expected to be advantageous. In the long run, this will improve the protection of human health and the environment from the risks that can be posed by chemicals, while enhancing the competitiveness of the chemical industry of the EU.

Still, before implementing bio-aromatics in existing and/or novel applications, the performance of such bio-molecules should be thoroughly evaluated. For instance, the presence of (unknown) impurities or side streams can limit their applicability.

## State of the art

It is clear that the direct integration of crude and/or processed (mono-, di and/or oligomeric) bio-aromatics into the current chemical and plastic industry is having strong potential. Some external factors can have a major positive or negative influence on the overall evaluation:

- Variation and tuneability of bio-aromatics functionalities (e.g., highly purified or mixtures);
- Physical properties (e.g., color, scent, viscosity)
- Presence of impurities;
- Drop-ins vs. additional functionalities;
- Price;
- Market potential;
- Environmental impact; and/or
- Circularity.

Until now, the implementation of crude and/or processed bio-aromatics is already explored and demonstrated in many applications (on small scale). A non-limitative list of selected applications, ranging from high-volume/low-value to low-volume/high-value chemicals, is:

- **Fuels** (e.g., cyclic hydrocarbons)<sup>26</sup>
- **Polymer additives** (e.g., plasticizers, antioxidants, UV stabilizers, flame retardants)
- **Polymers** (see Figure 8)
  - New polystyrene alternatives (made from bio-vinylphenols)<sup>27</sup>
  - New phenolic resins (made from bio-aromatic oligomers) for bio-based printing ink<sup>28</sup>
  - New polycarbonates (made from new bisphenols)<sup>29</sup>
  - New epoxy resins from new diglycidyl ethers (made from alkylcatechol, new bisphenols and/or bio-aromatic oligomers)<sup>30</sup>
  - New polyurethanes (made from bio-aromatic oligomers)<sup>31,32</sup>
  - New nylon-6 from new caprolactams (made from alkylcyclohexanones)
  - New nylon-6,6 from new adipic acids (made from alkylcatechols)<sup>33</sup>
  - Drop-in poly(ethylene terephthalate) (made from bio-terephthalic acid)<sup>34</sup>
- **Fragrances** from bio-aromatic monomers
  - Drop-in vanillin and 4-*n*-propylguaiacol (made from bio-aromatic monomers)
  - Smart drop-in aldolone (made from 4-alkylcatechol)<sup>35</sup>
- **Lubricants** (made from crude or refined bio-aromatics *via* esterification)<sup>36</sup>
- **Protective films** (made from crude bio-aromatics by tandem thiol-yne chemistry/Claisen rearrangement)<sup>37</sup>
- **Pharmaceuticals**<sup>38</sup>
  - Smart drop-in substituted anilines (made from bio-aromatic monomers)<sup>39</sup>
  - Smart drop-in aminoalkyl-phenol derivatives and seven-membered *N*-heterocycles<sup>40</sup>

In the following steps, the applications with the highest potential, identified through preliminary screening at lab scale, should be further evaluated and developed, which, for most applications, requires the production of larger amounts (*i.e.* kg-scale instead of g-scale) of lignin-derived bio-aromatics. A close

<sup>26</sup> <https://doi.org/10.1039/C5GC01534K>

<sup>27</sup> <https://doi.org/10.1021/acs.macromol.7b00970>

<sup>28</sup> <https://doi.org/10.1126/science.aau1567>

<sup>29</sup> <https://doi.org/10.1039/C9GC02619C>

<sup>30</sup> <https://doi.org/10.1039/D1CC02263F>

<sup>31</sup> <https://doi.org/10.1021/acs.biomac.0c00927>

<sup>32</sup> <https://doi.org/10.1016/j.indcrop.2022.114703>

<sup>33</sup> <https://doi.org/10.1039/D0GC02210A>

<sup>34</sup> <https://doi.org/10.1002/anie.201814284>

<sup>35</sup> <https://doi.org/10.1039/D0GC04268D>

<sup>36</sup> <https://doi.org/10.1021/acssuschemeng.1c02799>

<sup>37</sup> <https://doi.org/10.1002/macp.202100461>

<sup>38</sup> <https://doi.org/10.1016/j.isci.2021.102211>

<sup>39</sup> <https://doi.org/10.1021/acssuschemeng.8b06467>

<sup>40</sup> <https://doi.org/10.1021/acscentsci.9b00781>

collaboration with industry is encouraged to accelerate the adoption rate of bio-aromatics in applications at commercial levels, following increasing availability/production capacity of lignin-derived bio-aromatic platform molecules.

### **Value chain requirements**

Ideally, with regard to applications, bio-aromatics:

- are able to replace current fossil-based aromatics in a cost-efficient way;
- or if the costs of the bio-route are remarkably higher, bio-aromatics must have advantages compared to traditional aromatics (e.g., improved characteristics, better performance, lower toxicity, lower environmental impact).

### **Possible partners**

Currently, many aromatics end up in various high-volume polymer applications (*i.e.*, polystyrene, polycarbonate, polyurethane, phenolic resins, epoxy resins, nylon, *etc.*). Chemical companies that produce, use and sell aromatic-derived polymers are potential partners. Nevertheless, also other more high-value sectors (pharma, agro, life sciences, *etc.*) can benefit from the use of bio-aromatics, especially since these sectors typically apply tailor-made and highly-functionalized molecules.

## Complexity of the entire value chain and steps to debottleneck the major hurdles

As described in the previous sections, Catalisti has been and remains involved in a portfolio of projects investigating certain key aspects related to a value chain of bio-aromatics, being (i) feedstock diversity and availability, (ii) technology assessment, (iii) upscaling assessment, and (iv) application development. At this moment, valuable insights are available and possible solutions are already defined with respect to remaining hurdles (e.g., sufficient demonstrator samples and de-risked process technology). Figure 9 illustrates this portfolio of bio-aromatic projects in which Catalisti is a stakeholder, revealing a clear and ongoing development of the bio-aromatics value chain (in terms of TRL).

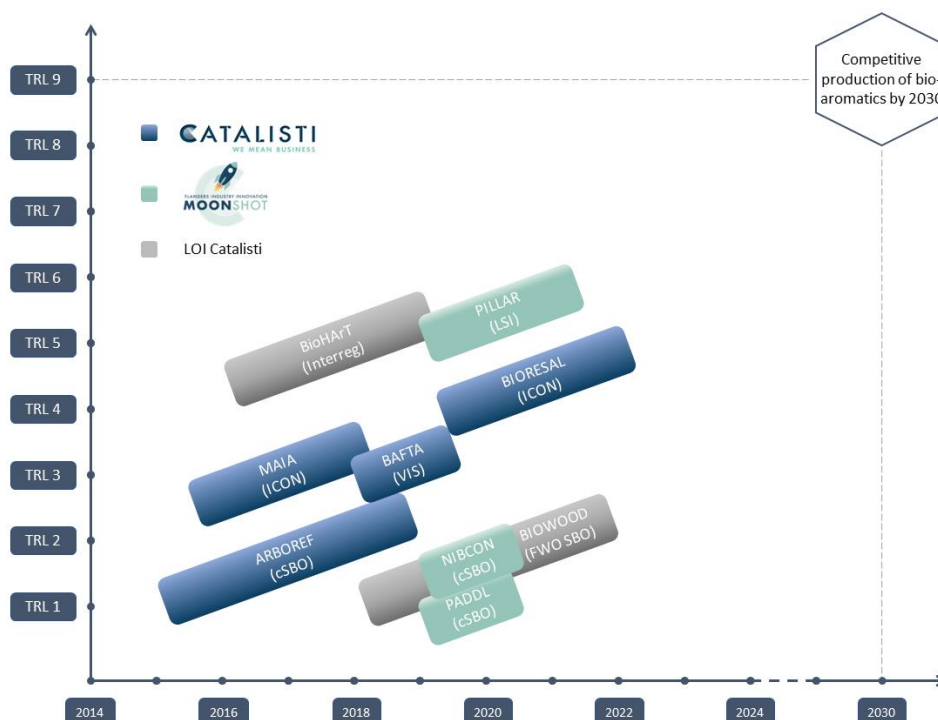


Figure 9 | Schematic representation of past and present projects on bio-aromatics in which Catalisti is stakeholder.

### ARBOREF | Refining of Wood to Aromatics | cluster Strategic Basic Research project (cSBO)

01/04/2015 – 31/03/2019

Partners: KU Leuven, UGent, UAntwerpen, VITO, VIB

ARBOREF intends to propose a biorefinery concept for bio-aromatics from raw biomass (e.g. grasses and wood) as well as lignocellulosic-derived streams. The key challenges are (i) to understand the relationship between plant structure and its convertibility towards a lignin and sugar pulp fraction [feedstock management]; (ii) to identify interesting synthetic routes towards bio-aromatics from these fractions [technology assessment], and (iii) to address the physical separation of chemicals after conversion by downstream processing (i.e., extraction, membrane filtration) [technology assessment].

### MAIA | Manufacturing of Advanced & Innovative Bio-Aromatics | Interdisciplinary Collaborative Research project (ICON)

01/09/2015 – 31/08/2017

Partners: Suez, Cobalin, ChemStream, TransFurans Chemicals, Lawter, Beaulieu, KU Leuven, VITO

MAIA aims to evaluate the usability of waste wood and flax shives [feedstock management] as renewable feedstock to produce advanced and innovative bio-aromatics, preserving and utilizing their natural functionality. Different unit operations (i.e., membrane filtration, vacuum distillation) are evaluated to separate/purify specific bio-aromatic fractions [technology assessment]. In addition, [application development] is also a major part of MAIA, evaluating the synthesized bio-aromatics as adhesives, UV-stabilizers, flavors, inks, etc.



### **BioHArT | Up-scaling of Innovative Technologies for the Production of Renewable Aromatics | Interreg**

06/03/2016 – 05/03/2019

Partners: Avantium, Bio Base Europe Pilot Plant, Chemelot InSciTe, DSM, KU Leuven, Maastricht University, TNO, TU/e, Universiteit Antwerpen, VITO

The purpose of the BioHArT project is to realize the upscaling of bio-aromatics [**upscaling assessment**] and produce sufficient quantities. Within the BioHArT-project, the entire value chain, going from raw materials suppliers over converting technologies towards end-users, is cooperating together and this in the border region between Belgium and the Netherlands.

### **BAFTA | Bio-aromatics feedstock and technology Assessment | Collective Research project (VIS)**

01/01/2018 – 30/06/2019

Partners: Catalisti, VITO, KU Leuven

BAFTA takes the first step from research towards industrial scale. The main goal of BAFTA is to evaluate different aspects of the value chain:

- Feedstock overview [**feedstock management**]; and
- Technology mapping (*i.e.*, IP landscape, freedom-to-operate) based on scientific and patent literature [**technology assessment**].

By doing this, the most promising technologies for upscaling each feedstock are selected [**upscaling assessment**].

### **BIORESAL | Bio-based resins from aldehydes and lignin | Interdisciplinary Collaborative Research project (ICON)**

01/05/2019 – 30/04/2022

Partners: INEOS Phenol, Sumitomo Bakelite High Performance Plastics, Kingspan, VITO, UGent

BIORESAL aims to produce bio-based lignin-phenol-formaldehyde (LPF) resins by replacing phenol with (modified) oligomeric lignin fractions, as potentially less hazardous and sustainable building blocks for their application in insulation materials and moulding compounds [**application development**]. Most importantly, this replacement is needed in a technologically proven and economically viable way. Additionally, BIORESAL will include the evaluation of a series of aldehydes as co-reactant in the synthesis of bio-based LPF resins.

### **BIOWOOD | Modelling and optimization of future wood supply for bioeconomy development in Flanders, Belgium | Strategic Basic Research project (FWO SBO)**

01/01/2018 – 31/12/2021

Partners: KU Leuven, VITO, UAntwerpen

The goal of the BioWood project is to:

- develop a detailed inventory of the currently available biomass from forests, landscapes, imports and waste flows in Flanders and the determination of their current value [**feedstock management**];
- design a flexible refining process for complex and variable lignocellulosic input to realize new organic products for agro-industrial applications [**technology assessment**];
- deliver a proof-of-concept of new products for agro-industrial applications that are equivalent to existing agro-industrial products [**technology assessment**];
- perform a techno-economic and real option analysis of the full value chain from bio-based production of woody biomass to agro-industrial products. Analysis of uncertainty and flexibility is central to this [**technology assessment**].

### **PADDL | Polymer additives from lignin building blocks | cluster Strategic Basic Research project (Moonshot cSBO)**

01/01/2019 – 30/06/2020



Partners: KU Leuven, UGent, UAntwerpen, VITO, Centexbel

Additives (e.g., plasticizers, antioxidants, UV stabilizers and flame retardants) are often added to improve the characteristics of polymeric materials. PADDL seeks to design new and safe bio-based additives for polymeric materials [**application development**] and, in this way, contributes to the shift away from non-renewable resources.

**NIBCON | Novel integrated biorefinery concepts for a carbon neutral bio-economy | cluster Strategic Basic Research project (Moonshot cSBO)**

01/01/2019 – 30/06/2020

Partners: KU Leuven, UGent, VITO, VUB

The interdisciplinary NIBCON project aims at the successful introduction of unique bio-based primary building blocks in future Flemish and European chemical industries. In this way, the project will promote the transition towards a carbon neutral circular bio-economy. It is built around inventive lignocellulose-based RCF biorefining and downstream separation technology. [**Feedstock management**] is optimized by matching local biomass feedstock with biorefinery demand (cf. MooV tool). The ambition of NIBCON is to intensify [**technology development**] in an integrated way (including TEA and LCA) and to achieve environmental sustainability, technical scalability and economic feasibility.

**PILLAR | Unique pilot infrastructure for innovative catalytic biorefinery of lignocellulosic material to functional bio-aromatics | Later Stage Innovation (Moonshot LSI)**

01/01/2019 – 30/06/2020

Partners: KU Leuven, VITO

PILLAR aims to realise a unique pilot infrastructure platform [**upscaling assessment**] for innovative catalytic biorefining of wood, lignin and by extension any lignocellulosic material to produce functional bio-based aromatics. PILLAR will focus on:

- a 100 L batch reactor (TRL 5) for a one-step lignin-first biorefinery technology (PILLAR I; KU Leuven); and
- an extension of the LignoValue Pilot catalytic reactor with a multi-purpose feed dissolver and solid feed injection (TRL 6) able to treat/liquify lignin (PILLAR II; VITO).

## **A future roadmap for bio-aromatics in Flanders**

It should be clear that a multidisciplinary approach, which brings together all the stakeholders operating at different stages of the value chain, is a very complex and intensive process. Nevertheless, this multidisciplinary approach will be vital to create a viable bio-aromatics ecosystem. Just looking at the Catalisti and Moonshot portfolio, many projects related to bio-aromatics are facilitated in the past years. These projects are typically focusing on one or more key aspects of the value chain (*i.e.*, feedstock management, technology assessment, upscaling assessment and/or application development). On top of these projects within Catalisti and Moonshot, many more projects with a focus on lignin (and thus bio-aromatics) valorization have been running, or are currently being executed. Some of them are funded directly by the Flemish government or *via* different funding frameworks (Interreg, Horizon 2020, BBI, *etc.*).

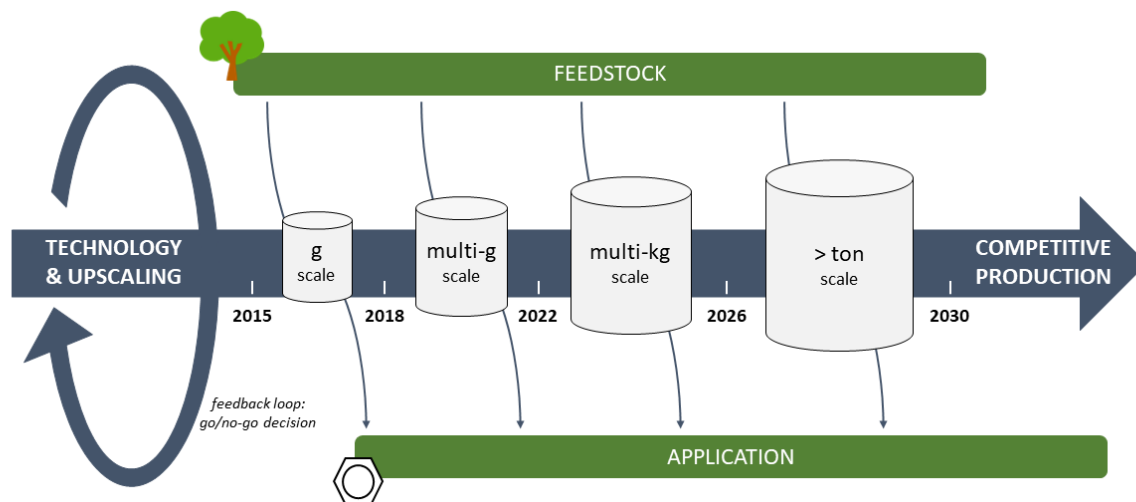
Considering all bio-aromatic projects, the TRL already increased from 1 (fundamental research) in 2015 towards 5-6 (pilot infrastructure) in 2022. In the Flemish region, two complementary pilot infrastructures – operating at multi-liter scale – are hosted by knowledge institutions and will be operational in 2022. The LignoValue Pilot catalytic reactor (VITO, Balmatt site) will focus on technology to depolymerize isolated lignin. The BioCon Pilot facilities (KU Leuven, TRANSfarm site) will focus on upscaling RCF technology to refine lignocellulose and downstream unit operations such as separations and chemocatalytic (de)functionalisations. Even though multiple examples of potential lignin-based bio-aromatic value chains have been demonstrated at lab scale and upscaling initiatives are ongoing, we are still far from a full-fledged commercial-scale value chain, which is the ultimate goal.

Catalisti is convinced that the new pilot infrastructure will further debottleneck the main hurdles by making multi-kg lignin-derived demonstrator samples available while simultaneously de-risking process technology at higher TRL. In other words, the refining technology at pilot scale will bridge the gap between available feedstocks and potential applications. If the potential of lignin-based bio-aromatics (business case, improved properties, sustainable character, *etc.*) is confirmed at intermediate TRL (5-6), investments towards industrial scale will be further de-risked. This might create an attractive window of opportunity for private investors, feedstock suppliers (*e.g.*, pulp/paper industry, cellulosic ethanol plants), technology providers and/or bio-based product developers to seriously invest in establishing (one or more key steps of) a full value chain of lignin-based bio-aromatics, which is technically and economically feasible. In this way, the full value chain can further mature, thereby gradually shifting from the knowledge institutions to the companies, and becoming financially self-sufficient.

In the upcoming years, the full potential of lignin-based bio-aromatics should be further explored at multi-g and multi-kg scale by using the new pilot infrastructure. Ideally, further investments will follow resulting in an operational refining capacity exceeding the ton scale by 2026. Different decisions should be taken/finetuned in the meantime with regard to:

- selecting a group of reliable feedstocks that can be easily handled by the biorefinery, with the following selection criteria:
  - good availability of resources, preferentially near the biorefinery location;
  - resources at low cost;
  - manageable resource composition and impurities;
- developing and de-risking the selected technologies in order to be easily operated within the biorefinery (*i.e.* technological proof of concept), with the following selection criteria and points of attention:
  - reactor design;
  - product reproducibility (*i.e.* yield, selectivity) and separation;
  - solvent recycling;
  - catalyst development (*i.e.* activity) and recycling (*i.e.* stability);
  - integration of unit operations;
  - process modelling and optimization.
- selecting a group of valuable applications that can be easily delivered by the biorefinery, with the following selection criteria:
  - matching scale;
  - valuable properties;
  - low environmental footprint;
  - applications with high added value.

Each individual aspect (feedstock, technology and application) should be fulfilled and will act as go/no-go decisions in the upcoming 10 years. A tentative roadmap towards competitive production of lignin-based bio-aromatics in Flanders by 2030 is represented in Figure 10.



**Figure 10** | Roadmap towards competitive production of lignin-based bio-aromatics in Flanders.