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Sustainable Materials: Production Methods and End-of-life Strategies

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Abstract: All three natural polymers of biomass and the monomer platforms derived from them present multiple avenues to develop products from specialty to bulk markets, which could serve as entry points into the industry for bio-based sustainable materials. However, several roadblocks still exist in the pathway of technology development of these materials due to challenges related to cost-competitiveness, scalability, performance and sustainability. This review outlines these major technical challenges as four key checkpoints (cost-competitive, scalability, sustainability, performance) to be addressed for successful market entry of a new sustainable material.

Keywords: Bio-based · Biomass fractionation · Circular economy · End-of-life · Performance · Scalable process · Sustainable materials

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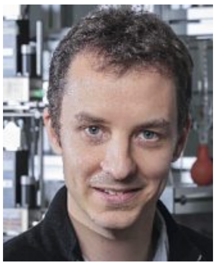

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1. Introduction

Billions of tons of carbon dioxide (CO_2) are released endlessly into the environment by non-renewable and/or fossil-based industries fueling our society's economy and lifestyle. In light of growing concern for global climate change due to rising levels of CO_2 in the atmosphere, the Paris Agreement has called for a target of net-zero CO_2 emissions by 2050 across the world. Moreover, the recent report by the Intergovernmental Panel on Climate Change (IPCC) highlights that the reduction of CO_2 emissions by decarbonization of industries has to be a key mitigation strategy to help limit the global temperature rise to 1.5–2 °C above pre-industrial levels and achieve a 'net-zero' goal urgently.[1] To decarbonize industries and steer away from fossil-based economy rapidly, significant effort is required to cut down carbon emissions at the source, that is the manufacturing process itself. In this regard, circular manufacturing strategies can be applied to the development of carbon-based products.

A circular manufacturing can serve as the backbone of a circular economy. Although several countries have actively taken part in developing new green economy strategies in the wake of climate crisis, we still have a long way to go to reach circular economy.[2] For instance, it is recently reported that Swiss economy is only 6.9% circular.[3] The report further highlights that an average Swiss citizen consumes 19 tons of new materials per year (estimated sustainable level is 8 tons), of which 40% is attributed to manufacturing. Furthermore, everyday fossil-based materials such as plastics, which increasingly contribute to greenhouse gas

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(GHG) emissions and microplastics pollution, was recycled only at 18% in 2015 globally.[4] A significant effort has ensued from this urgent need for circular manufacturing in developing materials from CO_2 emissions in power plants and recycling of waste plastics.[5,6] However, these processes do not fully omit the use of fossil-based carbon sources and also may have to undergo series of complicated steps to generate value-added, green materials.

Alternatively, use of renewable, non-edible, carbon-rich biomass can be a great sustainable platform for production of these value-added materials in the drive toward circular economy due to its low price, high abundance and possibility to make a range of products by utilizing its natural carbohydrates and phenolic structures in a variety of ways.[7] Many recent studies have suggested that the plastics industry could even be turned into a carbon negative sector by the use of biomass as the feedstock combined with recycling.[8–10] In fact, since 2005, startups all over the world involved in circular manufacturing, have been able to raise a total of $7.1 billion, with 80% of this investment flowing into bio-based technologies.[11] On this background, the scope of our review is also focused on the production of sustainable materials from biomass only.

Despite tremendous research interests, technology development initiatives and funding availability over the last few years, several roadblocks still exist in the pathway of technology development for making greener materials from biomass. Major challenges related to cost-competitiveness, scalability, performance and sustainability must be overcome to take these products to the market, which is still dominated by petroleum-based products. This review paper outlines the recent advances in production methods of sustainable materials from biomass and attempts to address key challenges to help in planning a successful market entry of a new sustainable material.

2. Valorization of Lignocellulosic Biomass for Production of Sustainable Materials

One of the biggest challenges in making sustainable carbon-intensive materials is finding an efficient pathway for the production of these materials. The main goal of valorizing bio-based resources is to produce renewable and sustainable materials having equal or better properties compared to petroleum-based products. However, it is highly difficult to design green, efficient and economic conversion pathways to transform lignocellulosic biomass into hydrocarbon-rich materials similar to petro-derivatives because biomass has a highly oxygenated, complex, polymeric structure. Nevertheless, if engineered in an intelligent way, biomass valorization in principle could offer some of the most promising routes for production of sustainable materials.

2.1 *Biomass as a Feedstock for Sustainable Materials*

As a non-edible and waste lignocellulosic biomass can serve as a key feedstock for production of sustainable materials due to several advantages:[12]

- Terrestrially, lignocellulose is the most abundant, low-cost feedstock and a massive source of renewable reduced carbon.
- Contains no sulfur and other impurities.
- Supplied as a concentrated stream of carbon, hydrogen and oxygen.
- Consists of carbohydrate polymers (cellulose and hemicellulose) and phenolic polymer (lignin) that can be converted to intermediate precursors to replace petrochemicals.
- Offers the potential to tune chemistries with bio-privileged molecules to make a new set of materials that have enhanced performance and are not available in petroleum industry presently.
- Biodegradability of products is likely possible as biomass contains nature's building blocks.
- Has no competition with food and land use.

2.2 *Structural and Thermodynamic Advantages of Biomass Valorization*

As shown in Fig 1, petroleum feedstocks and building block chemicals are built on mostly C_2–C_4 alkenes and aromatic intermediates with low content of oxygen or other heteroatoms compared to carbon and hydrogen. Therefore, it becomes very difficult to go up the thermodynamic hill to create a new product from petroleum derivatives containing especially, C–O or C–N bonds such as poly-lactic acid (PLA), epoxies and polyurethanes *etc*. Similarly, converting CO_2 molecules into the above types of polymers also seems thermodynamically inefficient. On the contrary, a low thermodynamic penalty to make these materials from biomass (wood) using nature's building blocks such as lignin, cellulose or hemicellulose and their derivatives, offers a clear advantage by requiring a much lower degree of chemical transformations to final bioproducts.[13]

In fact, it has been suggested that highly oxygenated feedstocks with additional functional groups such as biomass are more suitable to create materials with specific chemical properties.[14] As per the above theory, biomass utilization efficiency (BUE), which indicates the percentage of initial biomass that ends in the final product based on molar mass of reactant and target product, can help with designing the right processes for efficient conversion of biomass to performance materials.

2.3 *Chemical Pathways for Production of Sustainable Lignocellulosic Materials*

All three natural polymers of biomass and the monomer chemicals derived from them present multiple avenues to develop chemicals and materials from niches to bulk applications, as illustrated in Fig. 2. Herein, we present a few emerging pathways for the production of bio-based materials that have recently received great attention in academia and industry for commercialization. These bioproducts could potentially serve as entry points into the industry of sustainable materials including plastic bottles, packaging films, resins, adhesives, thermal insulation foams, dispersants, and plasticizers.

2.3.1 *Production of Cellulose-based Materials*

Application of PLA for the production of single-use plastics has gained particular interest in the last several years. With a growing market demand, researchers have attempted to produce PLA from waste biomass through utilization of its platform monomer, lactic acid, to avoid any competition with edible sugars used as raw materials. Cellulose from pretreated biomass is typically converted to glucose by enzymatic hydrolysis which can then be fermented to lactic acid for PLA production.[15,16] A great advantage of this route is that no harsh thermochemical conditions are

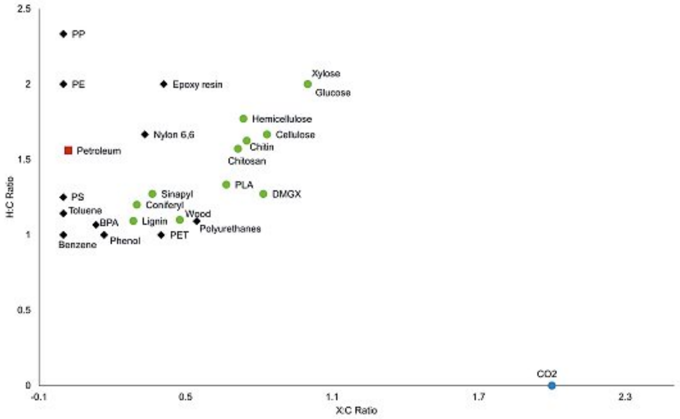


Fig 1. Van Krevelen diagram of CO_2, fossil-based and bio-based feedstocks and target materials. Reproduced from ref. [13].

employed and a high yield (up to 86%) and concentrated stream of lactic acid (up to 70 g L^{-1}) are achievable from a cellulose-rich feedstock.[17] Impressively, 100% of biomass gets utilized (glucose basis) in the synthesis of lactic acid making this process highly atom-efficient.

To develop a drop-in ready PET-substitute plastic, Origin Materials has commercialized the chloromethyl furfural (CMF) process successfully to utilize cellulosic sugars to make bio-based purified terephthalic acid (TPA) for bio-based poly(-ethylene terephthalate) (PET) in the market.[18] CMF could be produced at high yields by chemo-catalytic upgrading process and purified easily by distillation in a stable form.[19] This feature alone made the scale up of CMF process economically favorable. They received funding from investors including Nestlé and PepsiCo and are preparing two commercial plants.[20] By this strategy, it is possible to get a clean, upgradeable stream of CMF directly from lignocellulose which does not need expensive purification steps of sugars. In this process, a concentrated aqueous HCl and LiCl salt solution is used in the pretreatment to deconstruct and hydrolyze cellulose in the wood to glucose which is subsequently dehydrated to CMF (~47% yield, 61% BUE), releasing levulininc acid and furfural as by-products.[19] By upgrading CMF through hydrogenation, Diels-Alder and oxidation steps followed by separation and purification, finally TPA is obtained.[21,22]

The 2,5-furandicarboxylic acid (FDCA) process implemented by Avantium is another prominent route of manufacturing a cellulosic material at large scale.[23,24] They aim to produce a 100% plant-based, recyclable polymer, poly(ethylene furanoate) (PEF), which is superior in performance with lower carbon footprint than bio-PET plastic.[25] This process applies sequential pre-hydrolysis and hydrolysis at low temperatures to generate unmodified sugar streams (in concentrated acid) providing greater process and product flexibility.[26] The hydrolysate sugars are subsequently dehydrated to CMF (47% yield, 61% BUE), which is then upgraded to FDCA by etherification followed by oxidation.[27]

2.3.2 *Production of Hemicellulose-based Materials*

One key direction is the development of processes that enable drop-in replacement of petroleum-based materials from lignocellulosic biomass, such as the CMF process for bio-PET production, to avoid greenhouse gas emissions and our reliance on petroeconomy. In the long term, however, it would be more desired to design chemical processes for sustainable materials that undergo a minimum degree of chemical transformations to attain a high BUE. Furthermore, it is evident from the success of the pilot-scale CMF process that platform sugars, once released from biomass, have to be quickly and continuously separated without much dehydration from an aqueous reaction mixture. Recovering these sugar monomers in a stable, hydrophobic form in the course of reaction can help greatly in achieving the above goals and improve overall efficiency of the process. In this regard, the Laboratory of Sustainable and Catalytic Processing at EPFL has developed a unique strategy where an external aldehyde reagent helps stabilize the reactive xylose monomer, derived from hemicellulose, by formation of acetal groups on its side chains.[28] This not only prevents xylose degradation, but also allows for its transformation into a hydrophobic platform molecule dimethylglyoxylate xylose (DMGX) at 83% yields and 97% BUE, by acetalization and esterification at mild conditions. DMGX can serve as a building block for generating a wide variety of polyester materials.[28]

2.3.3 *Production of Lignin-based Materials*

Lignin, a highly functionalized, complex biopolymer with a phenolic backbone comprised of plenty of hydroxyl groups, is an attractive feedstock for synthesizing value-added aromatic composite materials with diverse functionalities and crosslinked rigid structures.[29] Although isolation methods of lignin from pulping processes have been in existence for many decades, it is still an underutilized by-product of the pulping industry, termed as technical lignin.[30] Mostly, technical lignin is burned as a waste material for energy production but recently it has started to receive great amount of interest from academia and industry for its wide array of application as a green material precursor.[31,32] A big advantage of scaling the production of technical type of lignin-based material is that the feedstock is easily and cheaply available from existing paper mills. In particular, technical lignin with high chemical reactivity due to abundance of free phenolic hydroxyl groups, low molecular weight and dispersity, and high purity is a desirable substrate for synthesis of bio-based adhesive resins, antioxidants, flame retardants and polyurethane (PU) foams.[33,34] Many companies and research centers such as Stora Enso, VTT, Borregaard in Europe have developed successful upgrading routes for the lignin obtained from Kraft and organosolv pulping processes to transform into hard carbon material, oxidized lignin, for applications in battery devices, concrete plasticizers, binding agents and dispersants.[35–37]

Recently, organosolv lignin has been utilized at 200 g scale to make lignin-based liquid polyols with the help of ethylene carbonate in polyethylene glycol (PG) solvent, as reported by Duval *et al*.[38]

To gain higher efficiency of lignin isolation and improve quality and properties of the extracted lignin from biomass, many researchers all over the world have attempted to develop a 'lignin-first' technology in the past decade.[39,40] This novel biomass fractionation technique enables, for the first time, extraction of lignin in an uncondensed form directly from biomass, and preferentially over the cellulosic fibers, retaining its native-like structure and functionalities.[41] While there exist many efficient pathways for such 'lignin-first' fractionation of biomass, reductive catalytic fractionation (RCF) has emerged as a promising method for scaling up in recent years. In this process, native wood is treated in the presence of a heterogeneous metal catalyst that allows for the rapid hydrogenation of lignin intermediates before they condense.[39] The main products from RCF process include lignin monomers and oligomers, which are low molecular weight and highly functional, and thus can be easily upgraded to bisphenol (BPA) replacement for making bio-based epoxy resins.[42]

Another recently developed 'lignin-first' technology, aldehyde-assisted fraction (AAF), by Luterbacher's research group, offers the unique advantage of extracting lignin from biomass in a near-native, preserved form and also being compatible with the upgrading of biomass polysaccharides.[43] Operating under mild conditions (below 150 °C for approximately 5 hours) and utilizing aldehyde protection to safeguard β-O-4 linkages in lignin, the strategy yields a substantial quantity of monomers upon depolymerization. Furthermore, the resulting oligomers are of smaller dimensions than market alternatives, facilitating convenient functionalization for diverse applications.

3. Checkpoints for Market Entry of Sustainable Materials

The main factors controlling the successful market entry of bio-based materials were identified in this review as cost-competitiveness, scalability, sustainability and performance, which are discussed in detail below.

3.1 *Cost Competitiveness*

One of the most critical decisions to make in developing a new sustainable material is finding where to invest and innovate now in order to get the pay-off later. Breakthrough Energy has proposed an index, namely, The Green Premium, which can serve as a useful guideline in this principle: *"The Green Premium is the additional cost of choosing a clean technology over one that emits more greenhouse gases."*[44] Accordingly, the sustainable materials with no green premium with production scaled significantly to reduce the cost of good sold (COGS) are the ones to have successfully entered the market. Recent plant shutdown of Clariant's SunLiquid technology for cellulosic bioethanol production in Romania suggests that price parity must be achieved rapidly after commercialization at industrial scale. Even with subsidies provided for ethanol production from cellulosic biomass, the bottlenecks for valorizing lignocellulosic biomass still persist. Key contributing issues to Clariant's plant shutdown were lower-than-expected production, high consumable chemical costs, challenging on-site production of enzymes, and excessive organic material in the plant's wastewater.[45] Several of these challenges are also relevant for making biomass-derived materials due to similarity of the upstream processing steps.

3.1.1 *Performance at the Cost of Lower Material Efficiency*

In order to create drop-in petro-substitutes from biomass, current methods mainly aim for designing highly deoxygenated molecules having a lower BUE. For example, bio-PET from CMF process can likely match the performance of fossil-sourced PET as it is synthesized from biomass-derived TPA monomers, but this comes at the cost of sacrificing 58% of biomass as unutilized.[46]

3.1.2 *High BUE and Full Valorization of Biomass*

Although developing high performance materials at the cost of low material efficiency is a more practical strategy at present to rapidly shift our reliance on fossil-based resources, a bio-based circular economy could be more sustainable if the materials are designed with high biogenic content rather than retrofitting biomass chemistry into petroleum structures.[14] Further, innovating biomass-derived sustainable materials with superior performance or unique applications (*e.g.* biopharma) and high BUE, in sectors where petro-based materials have limited market, could be more helpful in the long run to be cost-competitive as greener materials.[12]

Targeting margin is highly important to make price parity for a new material. In this light, aiming to generate a product mixture or full valorization of lignocellulosic biomass would be ideal to reach maximum value per ton of biomass. Integral biomass valorization can be achieved by spreading the production costs over more mass, *i.e.* producing multiple low volume, high-value products from the biorefinery (polysaccharides and lignin to plastics, packaging films, flavor & food additives, aviation fuels) versus a single high volume, low value product (bio-oil from pyrolysis).[43,47] Ultimately, a combination of producing sustainable performance materials at high BUE and full valorization of lignocellulosic biomass to a range of useful products would be most desired for a profitable bioprocess.

3.2 *Scalability*

In this section of the review, we evaluated the representative processes for the production of sustainable materials from each biopolymer of lignocellulose, as illustrated in Fig. 2, in terms of implementation at large scale and roadblocks to technology development.

3.2.1 *Recalcitrance of Lignocellulosic Biomass – A Major Bottleneck to Fractionation*

Lignocellulosic biomass has a highly recalcitrant structure which makes its deconstruction extremely challenging. Plant cell walls have biologically evolved to resist attack by pathogens, bacteria as well as physical and chemical stressors and is thus per default difficult to break down.[48] Lignocellulose is made up of three complex biopolymers, namely, cellulose, hemicellulose and lignin. Fig. 3 shows that the plant cell wall architecture features a tough, rigid fiber reinforced composite structure.[29] At the core of this composite, the cellulose microfibrils acts as the main scaffolding framework like reinforcing steel bar and the hemicellulose and lignin matrices act as the concrete to bind with cellulose.[49] Due to this complex structure of biomass, it is highly difficult to efficiently disintegrate the polymeric matrix of lignocellulose and extract the biopolymers as individual fractions at high purity and quality, with or without further deconstruction to smaller units of monomers or oligomers. Since this is the very first and one of the

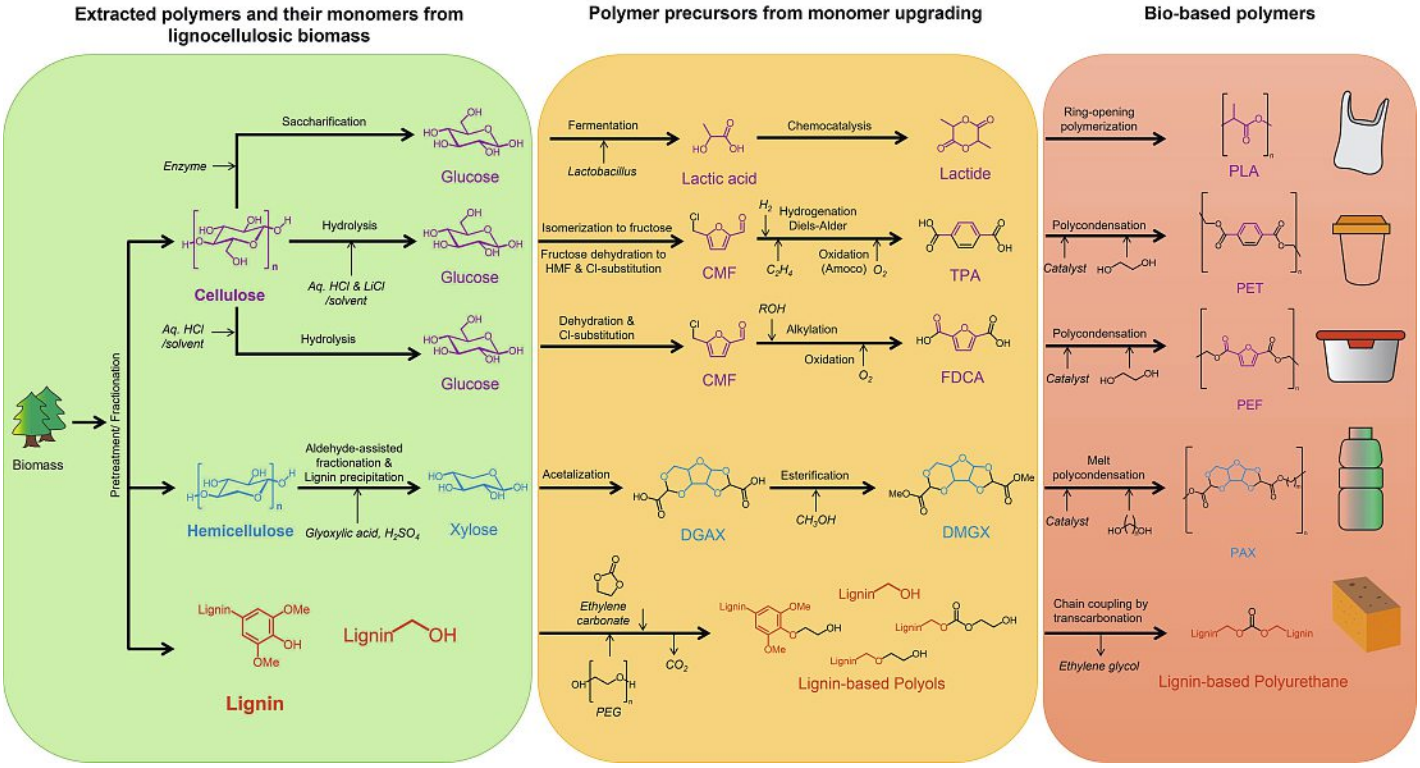


Fig 2. Representative piloted chemical processes for production of sustainable materials from lignocellulosic biomass.

most challenging steps of biomass conversion to polymer precursors, it often determines the overall material and energy efficiency, cost of production, and thus feasibility of the process at large scale.

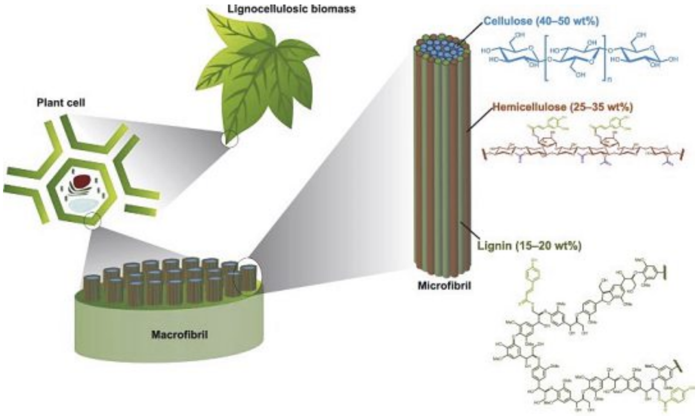


Fig. 3. Recalcitrant structure of biomass. Reused from ref. [29] with permission.

3.2.2 *Scalability of Processes for Cellulose-based Materials*

The major bottleneck in the conversion of lignocellulose to lactic acid arises due to the pretreatment which employs costly enzymes contributing nearly a quarter to the minimum lactic acid selling price.[50] Economic production of lactic acid from lignocellulosic biomass requires complete sugar conversion by co-fermentation of both glucose and xylose for economy at scale. However, only a few organisms can utilize xylose in addition to glucose.[15,16] Very recently, Läufer's group has reported a breakthrough to overcome these critical challenges in large-scale direct production of lactic acid from lignocellulose in a single bioreactor using consolidated bioprocessing.[17] This biological process omits the use of external enzymes by utilizing organisms capable of producing enzymes for hydrolysis of the polysaccharides in biomass and also fermenting both resulting glucose and xylose into lactic acid and useful by-products. Following this breakthrough, BluCon Biotech has attracted investor funding in 2023 to scale up this technology.[51] However, as presented in Table 1, although highly atom-efficient, this process still shows very slow rates of bioconversion (1 g L^{-1} h^{-1}) and lower product yields from realistic biomass feedstocks. Further work is needed to develop evolved strains as well as more efficient pretreatment methods for enhanced production of lactic acid from biomass.

We identified a few limitations that may be considered for large-scale applications of the CMF process in the long term. The key challenges in process development could include the lack of a continuous extraction system for CMF, the need for a highly expensive corrosion-resistant reactor due to use of HCl, degradation of xylose and lignin fractions in harsh acidic conditions, and poor quality of residual lignin with high content of chlorine.[46] Table 1 suggests that while the CMF process enables production of high performance bio-PET, the associated compromise on yields, BUE and degradation of side fractions of biomass to low-quality products (*e.g.* hydrothermal carbon from lignin) is currently unavoidable.

In the FDCA process, the technological development has entailed a number of challenges that should be addressed for continued growth of the PEF plastics in market. As shown in Table 1, the use of high-tech simulated moving bed reactors (SMB) for pretreatment of biomass, might be difficult to scale as it has very long residence time owing to the low operating temperatures.[26] Similar to Origin's CMF process, the FDCA recipe also may incur high cost of reactor to prevent corrosion due to using concentrated HCl. Besides, although FDCA attains a higher BUE of 59% than TPA and produces a high performing plastic (PEF), the lignin fraction still degrades to highly condensed solids. Another important factor in scaling issues is the long time of wait for demand for this new plastic to grow in the market thus making the technology development relatively risky.

3.2.3 *Scalability of Processes for Hemicellulose-based Materials*

The AAF technology for DMGX production has demonstrated solutions to some of the principal challenges in scaling up the production of lignocellulosic plastics. Firstly, as depicted in Table 1, DMGX can be produced as a stable, distillable intermediate at 83% yields and 97% BUE from birch wood *via* xylose, which is a remarkable improvement as compared to the CMF processes.[28] Further, a family of high performance poly(alkylenexylosediglyoxylate) (PAX) polyesters based on DMGX platform can be manufactured. Additionally, the AAF process also enables valorization of all side fractions of biomass to useful products (cellulose to glucose, lignin to chemicals) as shown in Fig. 4. Despite these highly scalable attributes, AAF process also needs to overcome several challenges for operating at industrial scale. Currently, the bottleneck of the AAF technology is the need to use glyoxylic acid which comes from fossil feedstocks at \$1 kg^{-1} and contributes significantly to the cost and environmental impact of this process.[28] In this regard, glyoxylic acid, if made renewable and at low-cost from ethanol or even CO_2, as demonstrated with the Volta process (Avantium), can enable the high volume production of PAX polyesters in the future.[52]

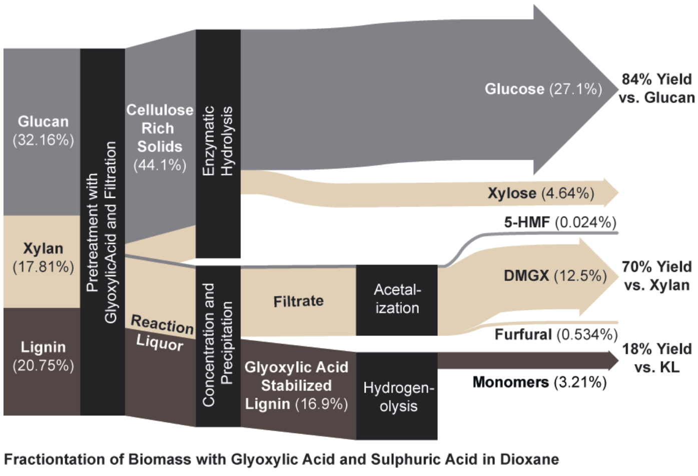


Fig. 4. Sankey diagram of mass flows in AAF process. Adapted from ref. [28] with permission.

3.2.4 *Scalability of Processes for Lignin-based Materials*

Some key challenges might prevail in the long-term application of lignin-based materials at large scale for different processes as outlined in Table 1.

Use of technical lignin extracted from Kraft pulping and organosolv processes has the benefit of merging with existing value chain of pulp & paper industry. However, lignin is recovered from these processes as a low-quality solid due to condensation reactions occurring in the harsh processing conditions with alkaline or organic solvents.[30,34] Unfortunately, the resulting lignin has limited opportunities for synthesis of high-performance materials and is mostly used as adsorbents, plasticizers, or additives in resins or for other applications.[31]

Table 1. Process development for production of sustainable materials from lignocellulosic biomass

Biomass Fraction (Source)	**Process (Industry partners)**	**Conditions**	**Solvents, Catalysts, Reagents**	**Target Platform Chemical**	**Yield**	**BUE**	**Other Fractions of Biomass**	**Target Material Performance**
Cellulose (Wheat straw, recycled paper)	Consolidated bioprocessing (BluCon Biotech)	70 °C, 14 days Batch reaction	Water, *Caldicellulosiruptor*	Lactic acid	49–63% (up to 11.9 g L^{-1})	100%	Lignin → *Solid residue*	PLA: Hard, brittle
Cellulose (Cardboard)	CMF process (Origin Materials)	Pretreatment: 125–175 °C, 10–60 min Upgrading: 80–120 °C Periodic extraction in biphasic reactor	Water, toluene or alkylbenzene, Conc. HCl, LiCl salt Water and Dichloromethane, HCl	CMF	47%	42%	Hemicellulose → *Furfural and degradation to solids* Lignin → *Condensed solids (hydrothermal carbon)*	Bio-PET: Hard, tough
Cellulose (Pine wood)	FDCA process (Avantium)	Pretreatment: Stage 1. 25 °C, 16 h Stage 2. 30 °C, 24 h SMB reactor Upgrading: 90°-100 °C Biphasic counter current reactor	Water and Dichloromethane, Conc. HCl	CMF	30%	59%	Hemicellulose: *100% to C_5 sugars (1.3 wt% in pre-hydrolysate stream)* Lignin → *Condensed solids*	Bio-PEF: Hard, tough
Hemicellulose (Birch)	Aldehyde assisted fractionation (Bloom Biorenewables)	60 °C, 48 h	1,4-Dioxane, Glyoxylic acid, H_2SO_4	DMGX	70%	97%	Glucan: *84% to Glucose* Xylan: *70% to DMGX* Lignin: *18% to Aromatic chemicals*	Hard, tough
Lignin (Pulp mill waste)	Alkali-O_2 oxidation of Kraft lignin (VTT)	70 °C, 30 min	Kraft lignin, oxidized white liquor, NaOH, O_2	Oxidized Kraft lignin (LigniOx)	89%	–	–	Cement plasticizer
Lignin (Pine wood)	Step-1. Reductive catalytic fractionation	235 °C	Methanol, Ru/C, 30 bar H_2	Lignin oligomers	46% Lignin oligomers (high Ph-OH content& low M_n)	–	Glucan: *99.7% in pulp* Xylan: *91.5% in pulp* Lignin: *41.5% in pulp*	Epoxy thermosets, heat-resistant
Lignin (Beech)	Step-1. Acetone Organosolv FABIOLA™ process Step-2. Polyol synthesis from lignin (Soprema)	140 °C, 2 h 110°–130 °C, 4-6 h	50% Aqueous acetone, H_2SO_4 FABIOLA™ Lignin, polyethylene glycol, ethylene carbonate	Lignin-based polyols	79.4% Lignin (full conversion of Ph-OH groups)	–	Glucan: *76% to glucose* Xylan: *87.3% to C_5 sugar monomers*	Rigid PUR foams
Lignin (Beech)	Step-1. AAF-lignin Hydrogenolysis	250 °C, 3 h Batch depolymerization	Ethanol, Ru/C, 10 bar H_2	Lignin oligomers with polyol functionality	50% Lignin oligomers	–	–	Rigid PUR foams
	Step-2. Polyol synthesis for PUR foams (Bloom Biorenewables)	<150 °C, 5h	Lignin oligomers, propylene carbonate					

Despite the major accomplishment of the RCF process for extracting lignin as a highly reactive, uncondensed substrate for making performance materials such as BPA replacement in a cost-effective and efficient way, the technology also suffers from a few issues from the scalability aspect. Firstly, incomplete conversion of the hemicellulose fraction of biomass can be observed.[53] The process employs low boiling point solvents (*e.g.* MeOH) under highly pressurized conditions, increasing the cost of reactor material. In addition, this process may involve complicated operation in flow-through reactors of separated sections for solid catalysts and wood consuming more solvent.[39] In this regard, use of a multi-pass system in flow reactor can be helpful.[54] Recyclability of solvents, side products and catalyst deactivation are also very important economic factors to be considered in scaling-up this technology.[55]

As depicted in Fig. 4, the AAF process is efficient at stabilizing lignin polymers and hemicellulose-derived sugars by acetal protection while also producing a highly digestible cellulose pulp.[28] Due to formation of stabilized lignin, high yields of lignin monomers and oligomers can be obtained from this process. With the vast potential of this technology in biomaterials development, including PU foams from lignin oligomers, Bloom Biorenewables, a spin-off company from EPFL, has been performing pilot runs to evaluate feasibility of the AAF process at 250 L scale with the help of HEIA ChemTech at Fribourg, Switzerland. Nevertheless, in addition to cost and sustainability concerns for the use of glyoxylic acid, hurdles of recyclability of solvent, side products and catalysts still need to be overcome for further growth of this technology.[41]

3.2.5 *General Outlook for Industrialization of Bio-based Sustainable Materials*

Industrialization of biomass fractionation requires addressing many general roadblocks besides technical challenges. The transportation and handling of the solid feedstock, the competition with oil refineries, the effect of governmental policies, the integration with existing supply and value chain – all of which can attribute to critical challenges in further growth of any biorefinery.

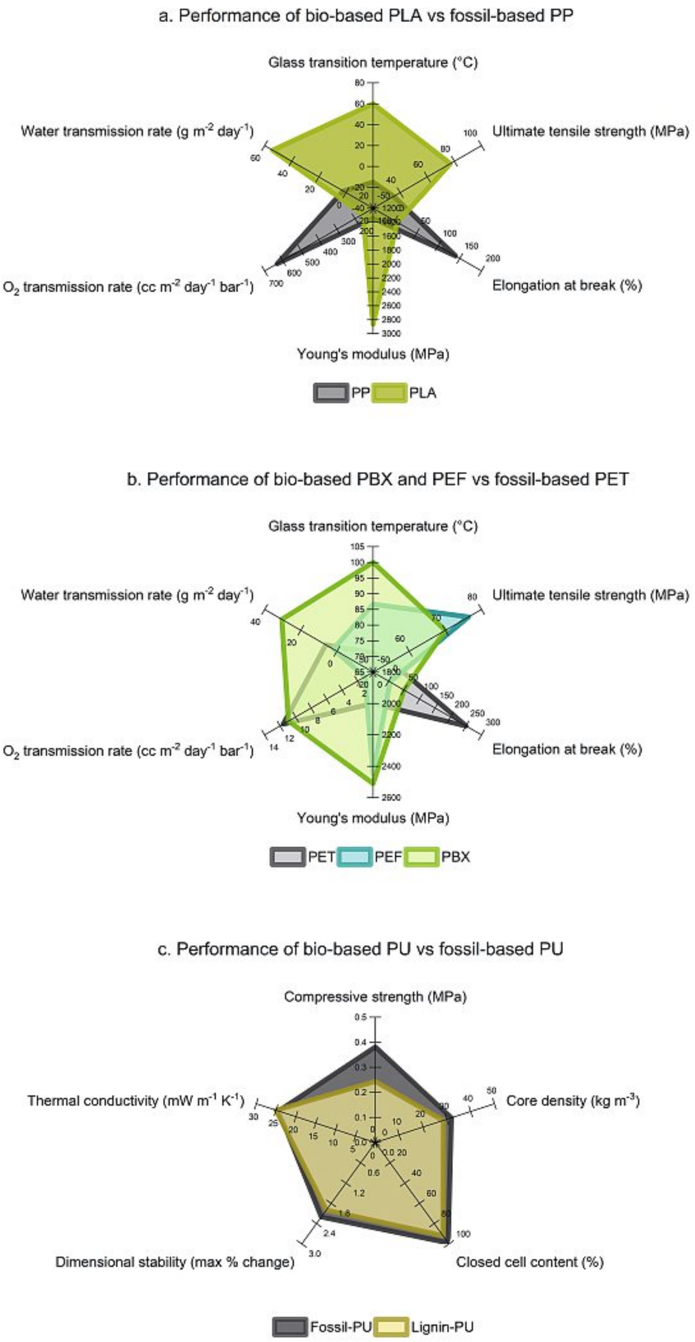
If the process is versatile enough, a licensing model can be used to expand industrialization across the globe. For example, modular plants can be constructed to valorize wood in Sweden, Norway, and Finland, straw in Eastern Europe, corn stover in central USA and bagasse in India. Further, government subsidies to develop biorefining technologies would be greatly welcome as well as favorable international policies and the participation of oil producers and distributers could accelerate these efforts. Lastly, process integration with current supply and value chains could also help rather than reinventing the wheel (*e.g.* lignin as an additive in wood-binding resins).[37]

3.3 *Performance*

With the objective of designing a new sustainable material from biomass to prepare drop-in ready replacements of current fossil-based products, it becomes imperative that the properties of the bio-based material be characterized and benchmarked against a chemically equivalent petro-based polymer.

The main thermomechanical properties typically used for comparing the performance of sustainable materials with fossil-based polymers include glass transition temperature (T_g), ultimate stress (σ_B) and elongation at break (ε_B), and Young's modulus (E) *etc*.[56] The thermal analysis obtained from dynamic scanning calorimetry gives an idea about the working temperature regime of the material. Furthermore, a dynamic mechanical analysis of the polymer can be used to test how the polymeric material may soften during the glass transition phase.[13] Mechanical properties from stress-strain curve are highly important to determine the applications for the newly synthesized bio-based polymers. For example, a polymer that continues to elongate after reaching maximum stress is considered tough, such as PET, while the one that does not is brittle, such as PLA.[13] Depending on the application, important properties such as O_2 and CO_2 barrier, water resistance, UV blocking, antimicrobial and antiviral activity, adhesive and bonding strength, and flame retardancy can be used to analyze the performance of a new bio-based material.[34,57]

For main applications in food packaging, the performance of cellulose-based PLA was compared with a common fossil-based plastic, polypropylene (PP). Fig. 5(a) shows that bio-based PLA usually has a significantly higher T_g, σ_B, E, and lower O_2 transmission rates, indicating that it can act as a heat-resistant, hard material with high barrier to gases leading to longer shelf-life.[13] Thus, PLA is suitable for food packaging. However, its lower ε_B and higher water transmission rates suggest that PLA is rather brittle and might be more prone to degradation in aqueous environment.


a. Performance of bio-based PLA vs fossil-based PP

b. Performance of bio-based PBX and PEF vs fossil-based PET

c. Performance of bio-based PU vs fossil-based PU

Fig. 5. Comparison of performance of bio-based materials with fossil-based commercial materials.

To characterize materials made from cellulose and hemicellulose, bio-PEF and a plastic from the PAX polyester family, namely poly(butylene xylosediglyoxylate) (PBX), have been selected and compared with PET for performance as shown in Fig. 5b.[25,28] Firstly, bio-based PEF and PBX exhibit excellent thermomechanical and gas barrier properties compared to PET, including higher T_g, σ_B, E, and lower O_2 transmission rates, suggesting that these biomaterials could potentially be alternatives in the market for fossil-based PET.[13,28,58] The resemblance of the structural rigidity of PEF with PET can be attributed to its 2,5-furandicarboxylic acid unit which is very similar to terephthalic acid, the monomeric unit of the latter polymer. Furthermore, PBX can show enhanced heat resistance and toughness compared to PEF but has lower barriers to oxygen and water. The higher glass transition temperature of PAX polyester is attributed to the rigid, tricyclic structure of its constituent DMGX monomer thanks to acetalization of the natural xylose sugar derived from biomass.

For evaluating lignin-based materials, an example of lignin-based PU foam has been compared with commercial fossil-derived PU foam (see Fig. 5(c)). Duval *et al.* synthesized lignin-based polyurethane rigid foams which had very similar compressive strength, core density, dimensional stability and thermal conductivity to the reference, making it a great alternative green insulation material for the bio-based market.[38] In this work, the enhanced performance of lignin as a substrate for PU foams was attributed to full conversion of its phenolic OH groups to aliphatic OH of high reactivity by reacting with ethylene carbonate.

With the powerful and tunable chemistry of biomass, unlimited designs of high-performance materials can be created if we know how to manipulate biomass structure with the right recipes of organic synthesis. To strategically achieve performance with high BUE in materials though, it is now necessary to develop structure–property–performance relationships for the production of sustainable materials. This kind of study can reveal key insights into effective design of new polymers from biomass and intensify process development for sustainable materials.

3.4 *Sustainability*

Sustainability is the key metric for ensuring environmental neutrality of future materials. Making a high-performing polymer is no longer enough to be competitive in the circular economy; it has to be either biodegradable or recyclable.[59] Furthermore, the production methods also need to be environmentally-friendly as well as displaying no significant toxicity or health hazards. In this regard, two major aspects where attention is required while designing a new product are: end-of-life strategy and environmental impact of the process itself.

The end-of-life strategy strongly influences the market readiness of a sustainable material. It is becoming increasingly necessary to assess cradle-to-gate environmental impact of new sustainable materials in industry. Cradle-to-gate refers to the carbon impact of a product from the moment it is produced in a biorefinery to the moment it enters the store. This kind of cradle-to-gate measurement of carbon impact of a new product encourages manufacturers to design materials that can easily be recycled or composted, while avoiding landfill as an end-of-life option altogether.[60]

With that said, it is important to realize that recycling is a very under-developed technology for most plastics. Although recycling has increased since the 1980s, the recycling of non-fiber plastics is still as low as 18% and almost no textiles are recycled.[4] Currently available methods for recycling of polymers derived from biomass and related checkpoints are illustrated in Fig. 6.

Mechanical recycling is the most common form of recycling which breaks down waste plastics into smaller size and new shapes by mechanical force and heat. This process always reduces the product quality, and the properties generally do not match with that of virgin plastics due to presence of impurities and decreased molecular weight caused by the conditions of thermomechanical processes during recycling. Therefore, this type of process should be reserved for mono-materials that are chemically made up of the same monomeric units, require low heating energy for melting and processing, and also have a system for highly efficient sorting and contamination-free waste collection such as the PET bottles. Mechanical recycling can also be a suitable technique for circular use of rigid, durable polymers having crosslinked structures with amides or aromatic backbones which are less susceptible to enzymatic or chemical attack.

In contrast, chemical recycling offers several advantages for a more robust, efficient recycling of bio-based materials.[61] This type of recycling strategy is highly suitable for the bio-based materials that might be disposed as a mixed waste, and are chemically heterogeneous or multilayered such as polyesters, polyamides, polyurethanes, and epoxy resins. Using water or solvents (*e.g.* methanol), catalysts and heat, the waste plastics

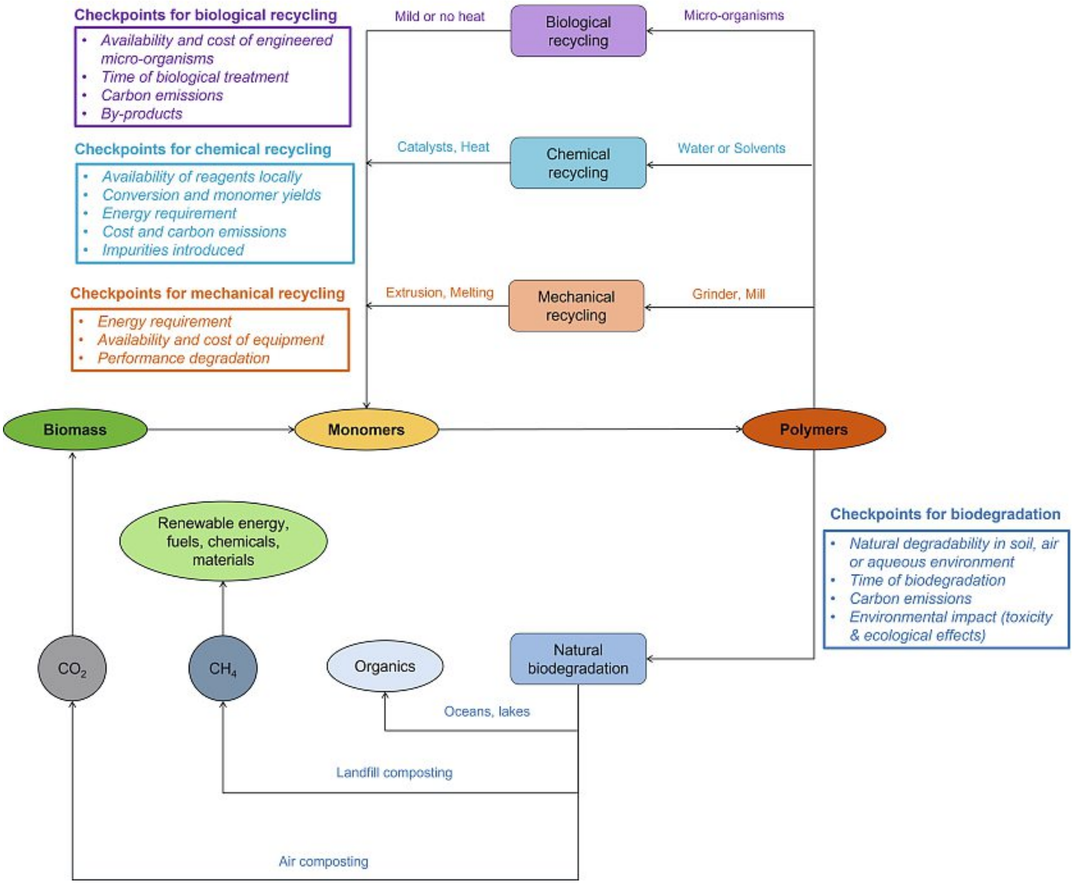


Fig. 6. End-of-life strategies for sustainable materials produced from biomass.

can be depolymerized back to the constituent monomers which can subsequently be separated, purified and then repolymerized to the original biomaterials.[28,61] Furthermore, a special type of chemical recycling, termed ‘upcycling’, can be applied to construct new polymeric materials by reacting depolymerized monomers from a plastic waste with external reagents. For example, an aminolytic upcycling method can depolymerize PET using amino-alcohols by organocatalysis and subsequently produce poly(ester-amides).[62] However, the success of this recycling process depends on using locally available reagents, conversion and monomer yields of recycling reaction, cost of recycling and any emissions.

Biological recycling is similar in its overall scheme to chemical recycling. Instead of using chemicals and heat, this strategy relies on using micro-organisms or enzymes to break down polymers into their initial monomers. Hence, wherever possible, this method should be implemented for a greener recycling path.[63] Nevertheless, the process has a few downsides which requires to be considered such as cost and low versatility of engineered micro-organisms, long time of biological depolymerization, and generation of by-products.[59]

Biodegradation is an effective, greener end-of-life strategy for easily hydrolysable ester materials, such as PLA, when disposed in nature. Through hydrolysis in aqueous environment, aerobic or anaerobic digestion in landfill composting, the biomaterials could break down into organic compounds, CO_2 or CH_4, respectively, which can be further reused to make renewable bioproducts.[64] However, generation of CH_4 is not desired because it is a greenhouse gas and thus must be recycled. Typically, PLA degrades relatively rapidly (6–9 weeks) in an industrial setting and in >1.5 years in the ocean. Biodegradation must be performed in a well-controlled setup preferably as the products can ensue unintended leakage of pollutants and microplastics into the environment.[65]

Sustainability of the production method of a new bio-based material is undoubtedly an important factor for being competitive in the emerging bioeconomy that is trying to shift to more natural products. This could mean finding alternative bio-based raw materials, greener reagents, minimizing consumption of energy, utilities and generation of underutilized side products and wastewater while developing a process without compromising heavily on the yield and quality of targeted bio-based material. Borregaard has marketed wood-binding resin products where they used lignin as an alternative to BPA for its natural phenolic structure and high content of reactive hydroxyl groups. Life Cycle Analysis (LCA) performed at different levels including raw materials, manufacturing, distribution, use, and end-of-life can be a great tool for producers and decision makers in order to choose the most sustainable design options for the materials.

4. Conclusions and Perspectives

In this review, different production methods of sustainable materials from lignocellulosic biomass were analyzed. Major checkpoints to help overcome technical challenges arising from cost, scalability, performance and sustainability were provided and guidelines were established for successful entry of a new sustainable material in the market.

Besides technical aspects, consumer behavior is an important factor that can help accelerate the journey to reach ‘net-zero’ emission goal. It is also up to governments and companies to run awareness campaigns and highlight their work by marketing, communication and advertisement to encourage and include the public into the mission to shift to sustainable materials.

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