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Bio-based and biodegradable polymers - State-of-the-art, Challenges and Emerging Trends

--Manuscript Draft--

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Abstract:	Frontiers of bio-based and biodegradable polymers are constantly expanding in a view to achieve sustainability. Hence, designing sustainable bioplastics made of either bio-based or biodegradable polymers opens up opportunities to overcome resource depletion and plastic pollution. This review presents a broad perspective on state-of-the-art technologies in bioplastics manufacturing along with the challenges underlying its production, application and post-consumer waste management. Recent scientific advances are catalysing the sustainable design of bioplastics to overcome the present challenges of plastic waste and emerging end-of-life options are contributing to circular economy. As research insights on developing sustainable bioplastics are rapidly evolving, its production and waste management approaches are not only confined to those discussed in this review.
Author Comments:	

5th December 2019

To

The Editor,

Current Opinion in Green and Sustainable Chemistry

Elsevier Publications

Dear Fabio Arico,

Sub: Submission of revised manuscript- COGSC-D-19-00022-Reg.

Thank you for reviewer's comments and we have addressed comments made by the Reviewer 2 and specific changes made in the manuscript are highlighted in Red in the revised version. All the editorial corrections suggested were incorporated in the revised version. Hope the revised manuscript meet the requirements COGSC for publication.

Reviewer 2:

The article highlights effectively currently available strategies for production and management of the most common types of bio-based plastics and partially bio-based ones. Although it deserves to be published on "Current Opinion in Green and Sustainable Chemistry", as it stands the manuscript requires some extensive revision as listed below and in the attached PDF file

Page 1: when discussing the yearly production volumes of PHAs compared to PBS and PBAT, please add some relevant updated figures on production.

Authors Response*: We have included the updated figures on global production volumes to compare PHA with PBS and PBAT as per recent European Bioplastics market report with relevant citation.*

Page 2, Figure 1: this figure is unclear, in particular the bottom part displaying "innovative bioplastics". Please clarify: is it a list of applications? Are these applications specific for the different types of bioplastics? If so, this should be highlighted in the figure.

***Authors Response**: Figure 1 has been modified in the revised manuscript. We intend to mention that there are emerging avenues of developing novel bioplastics by synergistically combining bio-based and biodegradable polymers for various applications.*

Page 5, Table 1: this table is adapted from one of the references reported? If so, it should be added in the table caption.

***Authors Response**: Yes, information mentioned in Table 1 was adapted from reference [17] and the same reference is included in Table caption.*

Page 7: when discussing the BBI competitive co-funding scheme from the European Commission, please provide some relevant, updated figures on the program. Additionally, please avoid discussing ongoing project (e.g. AgriChemWhey, which is due for completion in 2021).

***Authors Response:** Relevant updated figures regarding BBI consortium has now been included revised manuscript and as per reviewer's suggestion, any information related to the ongoing projects has been removed.*

Page 7: when discussing CO_2 chemical conversion strategies for the production of polycarbonates, please add some figures. Although it is a CO_2 valorization strategy, it will have limited impact on CO_2 mitigation compared to synthesis of CO_2 based inorganic materials.

***Authors Response**: We have added relevant figures on CO_2 content in bio-based polycarbonates and the expected production demand of plastics made from 70 % direct CO_2 have been included in the revised manuscript to highlight the significance of CO_2 upcycling strategies.*

Conflict of Interest and Authorship Conformation Form

Please check the following as appropriate:

- ○ All authors have participated in (a) conception and design, or analysis and interpretation of the data; (b) drafting the article or revising it critically for important intellectual content; and (c) approval of the final version.
- ○ This manuscript has not been submitted to, nor is under review at, another journal or other publishing venue.
- ○ The authors have no affiliation with any organization with a direct or indirect financial interest in the subject matter discussed in the manuscript
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Bio-based and biodegradable polymers - State-of-the-art, Challenges and Emerging Trends

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Abstract:

Frontiers of bio-based and biodegradable polymers are constantly expanding in a view to achieve sustainability. Hence, designing sustainable bioplastics made of either bio-based or biodegradable polymers opens up opportunities to overcome resource depletion and plastic pollution. This review presents a broad perspective on state-of-the-art technologies in bioplastics manufacturing along with the challenges underlying its production, application and post-consumer waste management. Recent scientific advances are catalysing the sustainable design of bioplastics to overcome the present challenges of plastic waste and emerging end-of-life options are contributing to circular economy. As research insights on developing sustainable bioplastics are rapidly evolving, its production and waste management approaches are not only confined to those discussed in this review.

1. Introduction and market growth

21st century is thriving with tremendous economic growth but at the same time facing an irrecoverable ecological damage. Plastic pollution is recently being highlighted as global crisis at every stage right from its production to disposal and incineration [1]. Bioplastics constituting both naturally and chemically derived materials from renewable or oil-based resources are being designed to feature minimal carbon footprint, high recycling value and complete biodegradability/compostability [2, 3]. In order to ascertain no competition with food and agriculture resources, recent advancements are emerging to develop next-generation bioplastics derived from renewable waste streams, microbial/microalgal cells and biomass which eventually fosters carbon neutral infrastructure for bioplastics production and management [4, 5]. Moreover, sustainable production and recycling mechanisms for bioplastics are considered to have huge compliance with the policies/actions set by United Nation's Sustainability development goals (UN SDGs) and European circular economy strategy [6].

The global bioplastics production capacities are difficult to estimate and are usually based on forecast due to continuously emerging range of bio-based and biodegradable polymers and rising interests on investing in bioplastics sector. Recent report published by Nova-Institute has predicted that global bioplastics production capacity growing at a considerable pace from around 2.11 million tonnes in 2018 to 2.62 million tonnes in 2023 [7]. Europe ranks top in the research and development of bioplastics and stands next to Asia as major hub for bioplastics production and consumption [8]. With many innovative bioplastics entering the market segments for diversified applications, industries are interested in expanding the production capacity. Acute relevance to sustainability and circular economy has been indeed influencing the bioplastics industry to achieve substantial growth, technological maturity with multiple production routes.

2. Progress and trends in commercial bioplastics

Naturally occurring polymers like cellulose derivatives, thermoplastic starch (TPS) and their blends stands highest in terms of production capacity as these materials are replacing plastics particularly in flexible film packaging sector [9, 10]. Recent bioplastics market update shows that polylactic acid (PLA) receives greater attention from both academia and industry due to its technological advances productivity and functionality [11, 12]. PLA is a known for its versatility featuring excellent barrier properties thus gaining value to replace polystyrene (PS) and polypropylene (PP) in packaging and other challenging applications [13]. Next to PLA, polyhydroxy alkanoates (PHA) receives interest as evidenced by greater number of international patents [14]. However, in terms of global production capacity, PHA stands next to poly (butylene adipate-coterephthalate (PBAT) and polybutylene succinate (PBS). As per recent market report, current global production of PHA is about 25,320 tonnes, which accounts to 1.2 % as against PBAT and PBS holding 13.4 % and 4.3 % respectively [15]. Polycaprolactone (PCL) and PBAT are fossil-based polymers but tend to biodegrade, signifying that biodegradability is not always dependent on its source of origin or the polymer building block. Schematic representation shown in Figure 1 clearly demarcates various technological approaches specific to different classes of bioplastics.

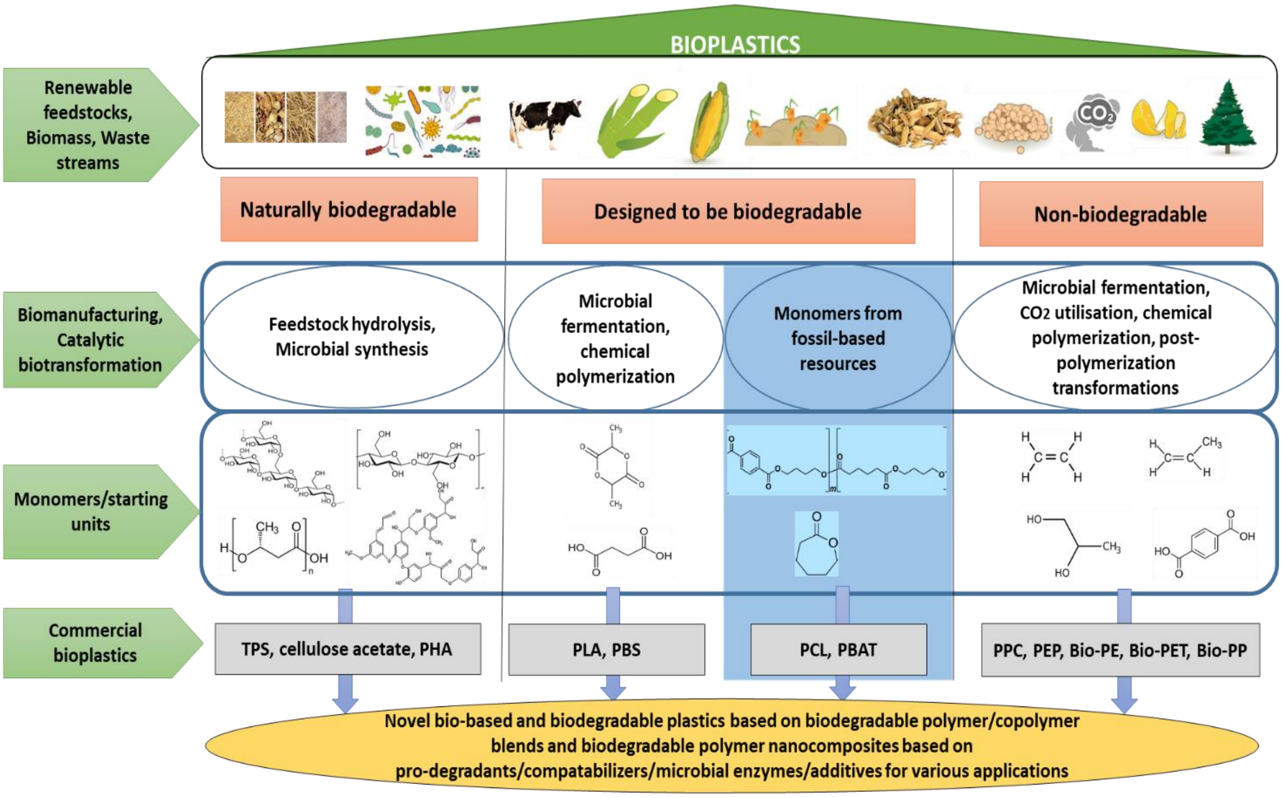

Figure 1. Schematic representation of technological approaches in producing commercial bioplastics (*Shaded in blue-Biodegradable polymers derived from oil-based resources*)

Majorly used commodity polymers like polyethylene terephthalate, polyamide and polypropylene have also been manufactured from bio monomers derived glucose fermentation or lignin fermentation which facilitates the resurgence of Bio-PET, Bio-PA, Bio-PP respectively [16]. Growing interest in novel bioplastics constituting two or more existing biodegradable polymers would eventually result in second-generation bioplastics, thus offering advantage of developing scalable counterparts to synthetic plastics [17]. Hence, the goal would be to design novel composites comprising only of bio-based building blocks having specific desired functionalities suitable for applications and at the same time completely biodegradable and recyclable building blocks having specific desired functionalities suitable for technological applications [18]. For example, in a recent work, synergic blends of PLA and PCL were highlighted as completely biodegradable (in domestic composting conditions) alternatives to conventional, petrochemical based plastics [19]. Emerging bio-based polymers like Poly (Ethylene 2, 5-Furandicarboxylate) (PEF)/ poly (trimethylene terephthalate) (PTT) and polypropylene carbonate (PPC) produced from bio-based furan monomers and alcohols/epoxides respectively are characterized by excellent thermal and barrier properties comparable to its petroleum analogues [20]. For instance, blending PEF with PLA or PHA would ultimately contribute to superior functional and biodegradable properties enabling practical application in packaging applications [21]. The trade-off between biodegradability and functionality brings huge research scope on blending and compatibilization of various bio-based polymers to push its performance efficiency and versatility [22, 23]. Table 1 shows the widely known bio-based and biodegradable and their respective starting materials and feasible end-of-life options [18, 24].

Table 1: Commercial bioplastics including both biodegradable and non-biodegradable polymers, its production source, capacity and end-of-life options (Adapted and modified from [18])

Biodegradable polymers				
Polymer Name	**Source/Feedstock**	**Production capacity (Kt/Year)**	**Trademark/Company**	**Sustainable end-of-life options**
TPS, Cellulose, Cellulose acetate, Starch blends	Biomass, agro-residues, lignocellulosic derivatives	384	Mater-Bi/Novamont, Agrana starke, NaturePlast, Indochine Bio Plastiques	HC, IC, AD
PLA & PLA blends	Lactic acid from dairy whey, corn starch or organic residues	225	Ingeo/NatureWorks, Luminy®/Total Corbion, Lacty/Shimadzu Cor. Vyloecol/Toyobo, Danimer Scientific	IC, MR, CR
PHA, PHB, PHO	Volatile fatty acids, glucose/glycerol from fermentation of municipal solid waste or any carbon feedstocks	30	Minerv-PHA^{TM}/Bio-On, PHBH$^{TM/}$Kaneka, TephaFlex®/Tepha, Nodax^{TM}/Danimer Scientific, AirCarbon®/Newlight Technologies	HC, IC, AD, CR
PCL	Chiral hydroxy acids, lactones	--	CAPA^{TM}/Perstorp (Ingevity)	HC, IC, CR

PBS	Succinic acid, 1,4-butanediol	97	GS PLA®/Mitsubishi Chemical, Bionolle$^{\text{TM}}$ 1000/Showa Denko K.K., Skygreen®/SK Chemicals, Succinity, BioPBS$^{\text{TM}}$/PTT MCC Biochem	CR, ED
PBAT & PBAT blends	Terephthalic acid, adipic acid hydroxymethyl furfurals (HMFs), butanediol	152	Ecoflex®/BASF, Wango/Zhuhai Wango Chemical Co., Ecoworld/JinHui ZhaoLong, Eastar Bio/Eastman, Origo-Bi®/Novamont	IC, CR
Bio-based and non-biodegradable polymers				
Bio-PE	Bioethanol from sugarcane	200	Braskem	MR
Bio-PET	Furan dicarboxylic acid from HMFs	560	PlantBottle$^{\text{TM}}$/Coca Cola	MR, CR, ED
Bio-PTT	1, 3-propanediol	45	DuPont Corterra$^{\text{TM}}$, Sorona®/ Shell Chemicals	MR, CR
Bio-PEF	HMFs	-	Synvina/Total-Corbion	ED
Bio-PP	Isobutanol	-	Technoform, LanzaTech	MR
Bio-PA	Volatile fatty acids, HMFs	-	Evonik VESTAMID® TERRA, Dupont, FKuR, BIOFED	MR, CR
Bio-polycarbonates	Bioethanol/dialkyl carbonate/epoxides and carbon-dioxide	-	Asahi Kasei Corporation, Saudi Aramco Converge®	CR

*- Emerging options in bioplastics waste management with either limited or no evidence on technology commercialisation

AD-Anaerobic Digestion; MR- Mechanical Recycling; CR-Chemical/catalytic Recycling; ED-Enzymatic depolymerisation; IC- Industrial composting HC- Home composting

3. State-of-the-art technologies for bioplastics innovations and production

The current bioplastic sustainable production model relies on design and development of novel valorisation protocols of renewable resources derived from urban, agricultural and food wastes. Approaches to develop monomers and biodegradable polymers from biomass feedstock received great attention in chemical industries by leveraging on the innovative biocatalytic transformation and synthetic chemistry [25, 26]. Sustainable bioplastics materials are currently under development, and innovation relies either on developing completely new types of polymers or drop-in substitutes derived from renewable resources. Advancements in industrial biotechnology offer various chemo-enzymatic or bio-catalytic synthetic routes for converting biomass or renewable feedstocks into high-value building blocks or monomers [27]. Additionally, engineering of consumer grade bioplastics based on monomers derived from waste residues represents a sustainable production value chain which accounts for establishing circular bioeconomy. Growing global demand for bio-based and biodegradable polymers prompted investments in research to promote and establish large scale production of bioplastics. Bio-based industries (BBI) consortium in partnership with European Union (EU) is investing about 3.7 billion on large scale flagship projects to encourage new technologies for production of bio-based monomers and polymers from waste biomass/renewable feedstock's [28]. As one of the specific impacts of BBI's programme is to replace at least 30% of fossil-based raw materials with bio-based and biodegradable ones by 2030, potential scope for bioplastics manufacturing processes is foreseen in the coming decade [29].

Bioplastics production by utilising greenhouse gases like carbon dioxide is one of the sustainable carbon upcycling approach which is gaining huge attention [30]. Recent report by Nova Institute has highlighted the projected estimation of directly converting 70 % CO_2 for bioplastics manufacturing [31]. Breakthrough research in areas of selective copolymerization process has resulted in the commercial production of polycarbonates constituting about 30-50 wt.% of waste CO_2 [32]. CO_2 upcycling efforts are constantly evolving for meeting the predicted demand of producing 450 million tonnes plastic by 2050, which are completely made from renewable carbon [31]. This CO_2 recycling approach holds benefit of being easily retro-fitted in the fossil-fuel based polymer manufacturing infrastructure thus exerting both economic and environmental benefits. Indeed, lesser dependence of agro-feedstocks, monomer extraction/transformations and complex pre-treatments are considered as highly advantageous against bio-resources derived polymers [33].

4. Sustainability and end-of-life options for bioplastics

On a global trend, plastic production from fossil-based resources and plastic waste incineration together accounts to about 400 million tonnes of CO_2 every year [34]. Replacement of fossil-based plastics with bio-based/biodegradable will certainly reduce carbon footprint at production level. However, assessing its sustainability aspects in terms of end-of-life management is vital to exert bioplastics as an environmentally friendly alternative. Not all bio-based polymers are deemed biodegradable and in contrast some of the biodegradable polymers could also be produced from fossil-based raw materials. Indeed, popularly known bioplastics families like PHB, PCL and starch and their blends are proven to be biodegraded in both managed and specific unmanaged environments [19], however, failing to manage their disposal would result in uncontrolled biodegradation adding to existing plastic pollution [35]. Hence, it is of utmost importance to practice specific end-of-life management considering the properties and processing conditions of each bioplastics rather than a generic waste management plan. Life cycle analysis (LCA) is an indispensable tool to gauge and quantify the benefits or impacts of any bioplastics, subjecting to the boundary conditions and assessment considerations [36].

Despite being resources-efficient and derived from renewable bio-based feedstocks/residues, it is crucial to look closely into environmental impacts of bioplastics waste. Disposal of bioplastics waste in landfill certainly contributes to similar management problems that of conventional plastic waste. Hence, advocating best end-life management of post-consumer bioplastics waste is needed to achieve lower carbon footprint [37]. Sustainable management of bioplastics waste is highly challenging as some of the bioplastics are designed to only biodegrade in specific managed conditions thus creating huge ill-effects when disposed in non-ideal environments like soil, fresh water and marine. Indeed, scientists aim at developing bioplastics that could achieve complete and quicker biodegradation in any environment as per ASTM and ISO standards [38]. However, most of the reported biodegradability of various biodegradable polymers was demonstrated at lab scale and it is essential to establish biodegradation of the commercial bioplastics and their blends at appropriate industrial scale [39, 40].

Recycling is considered as the most preferred option to manage bioplastic waste as similar to conventional plastic waste [41]. However, recycling can either be mechanical, chemical/catalytic and organic depending on whether the bioplastics is of biodegradable and/or if the considered polymeric material biodegrades only in managed conditions. The distinct recycling options shown in Figure 2 represents the state-of-the-art on closed-loop management of post-consumer bioplastics waste. Prime challenges in recycling of post-consumer bioplastic waste is attributed to its heterogeneity, low market volumes, diverse sources and high potential for plastics waste contamination. These challenges indicate a clear need for more efficient chemical and biochemical processes to valorise the bioplastics waste into perpetually reusable high-value end-products. Implementing combined recycling and recovering concepts including extraction of high-value chemicals/monomers via chemical recycling, solvent extraction [42] and cogeneration of biofuel and volatile fatty acids through anaerobic digestion [43] would certainly create positive impact towards a circular bioeconomy. Perhaps, some of the management approaches would not directly recycle back the bioplastics into its starting monomer. However, it is worthwhile to invest on valorisation of post-consumer bioplastic waste and provide incentives for recycling or energy recovery for contributing to circular bioeconomy and sustainable management of bioplastics waste [44].

Figure 2. Schematic representation of recycling strategies for sustainable management of bioplastics and contribution towards SDGs

6. Future outlook

Rationally designing the bioplastics to impart desired functionality and recyclability [45, 46] and utilising unaccounted biomass as a valuable resource would together establish a sustainable production value chain for bioplastics. Despite some of the bioplastics production technologies are lacking the scalability and productivity comparable to petroleum based routes, governmental regulations and consumer pressure has been fostering the bioplastics industry to adopt and implement sustainable production routes. Circular bioeconomy is also gaining global momentum which in turn triggered wide range of stakeholders to leverage the synergistic potential of bioplastics manufacturing and upscaling/recycling strategies [47, 48]. Innovations in fundamental redesigning of bioplastics with improved economics for recycling will pave a way for the next generation of sustainable bioplastics.

Acknowledgements

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