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REVIEW ARTICLE

The 2G and 3G bioplastics: an overview

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Highlights

- 2G and 3G bioplastics as potential bioproducts in biorefineries.
- Re-use of liquid and solid wastes for biobased polymers' production.
- Raising number of innovative research and developments in 2G and 3G biopolymers.
- Bioplastics as a sustainable biorefineries in a circular bioeconomy.

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Biobased bioplastics;
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Lignocellulosic biomass;
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PHA, PLA;
Bacteria;
Microalgae.

Abstract: During the last decades an increase of biobased plastics was observed. This expansion of global biobased market is a reflection of the efforts of academic and industrial researchers, and production and marketing chain members. They have sought new possibilities to contribute to bioplastics´ large-scale production and commercialization cost-effectively. The main commercialized bioplastics are poly lactic acid (PLA), polyhydroxyalkanoates (PHAs), starch, cellulose and proteins plastics. The use of alternative raw materials has been implemented while applying innovations in production processes to reduce cost and decrease environmental impacts. In this review, the 2G and 3G bioplastics are presented as a promising solution to enable large scale and viable production of these biobased materials. 2G bioplastics are produced from lignocellulosic biomass and non-food edible oils, while 3G bioplastics are obtained from sugars or oils produced by micro-organisms (microalgae, bacteria, mushrooms, yeasts and others) or from municipal waste material.

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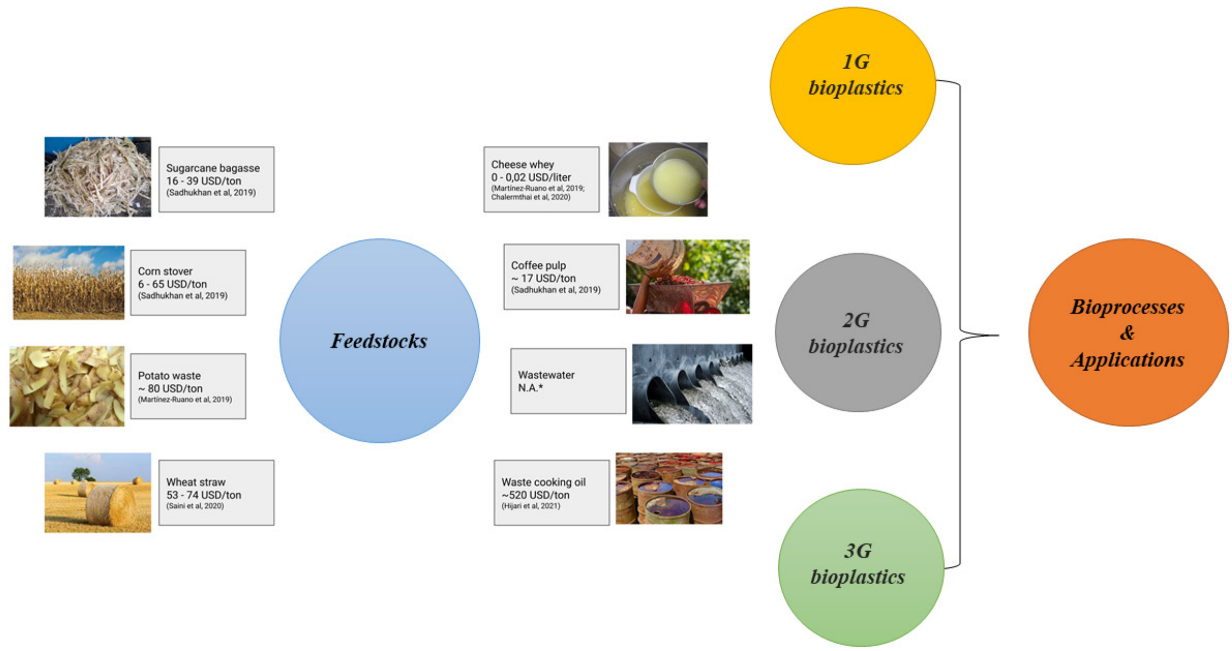
Graphical abstract



Introduction

The circular bioeconomy is being increasingly studied and established as a sustainable way to produce different products from bioresources such as biofuels, bioplastics and other commercially important bioproducts (Brodin et al., 2017). Bioplastics are defined as naturally and chemically derived materials from renewable or oil-based resources that are biodegradable/compostable and have high recycling value (RameshKumar et al., 2020). Bioplastics emerged from the sustainability concept, where, the focus was reducing the petrochemical pollutants through the replacement of biodegradables products. This is true due to their "food basis" – actually named as first generation (1G) bioplastics, thus, they generate lower carbon emission rates, as well as fewer greenhouse gas emissions (Rattana & Gheewala, 2019).

Bioplastics production started from renewable sources, including corn, sugarcane, waste fats and oils. However, there is a growing interest in the use of lignocellulosic biomass and wastewaters, mainly non-foods crops, to produce bioplastics, which are generated in different agricultural and agro-industrial activities (NaturePlast, 2021). Different types of bioplastics are produced industrially, competing in performance and price with petrochemical-based plastics. The majority of bioplastics are produced from biomass where bacterial or algal fermentation is carried out using sugars from different sources to obtain building blocks molecules, which can be used as monomers for the production of second generation (2G) and third generation (3G) bioplastics, respectively (Figure 1).

Last developments and innovations show that biobased plastics seem to overcome the sustainability and waste disposal concerns. However, those that are mainly derived from agricultural plants (corn, wheat, potato and soybean) may pose a potential threat to food security, fertile land, and irrigation (Fabra et al., 2017). In fact, this scenario in Europe is being stablished. The European rapport about the French bio-economy (Netherlands Enterprise Agency, 2020), shows that they are observing how to improve this valuable chain through importation of raw biomasses. With this mission of increasing this segment in Europe, the "Bio-Plastics Club" was created in 2006, and nowadays, about 38,000 tons/year of bioplastics products are produced in France (Netherlands Enterprise Agency, 2020). The next generation biobased plastics obtained from produced sugars or oils by microorganisms such as microalgae, bacteria, mushrooms, yeasts, etc., the 3G bioplastics, are coming to replace the current biobased materials (Zhang et al., 2019).

The bioeconomy of bioplastics

The transition from a linear, fossil-based economy to a circular bioeconomy passes through the substitution of materials produced from non-renewable sources by those based on "green carbon". Additionally, life cycle and other environmental aspects are usually benefited by the use of materials of renewable origin, which can also be biodegradable. In this sense, bio-based materials, particularly bioplastics, represent an essential niche in a bio-based economy. In 2017, bio-based products had less than 5% market shares in many segments, including plastics and fuels (Wydra & Hüsing, 2017).

The first bioplastics arose in the 80/90's as biodegradable materials intended for packaging applications. However,

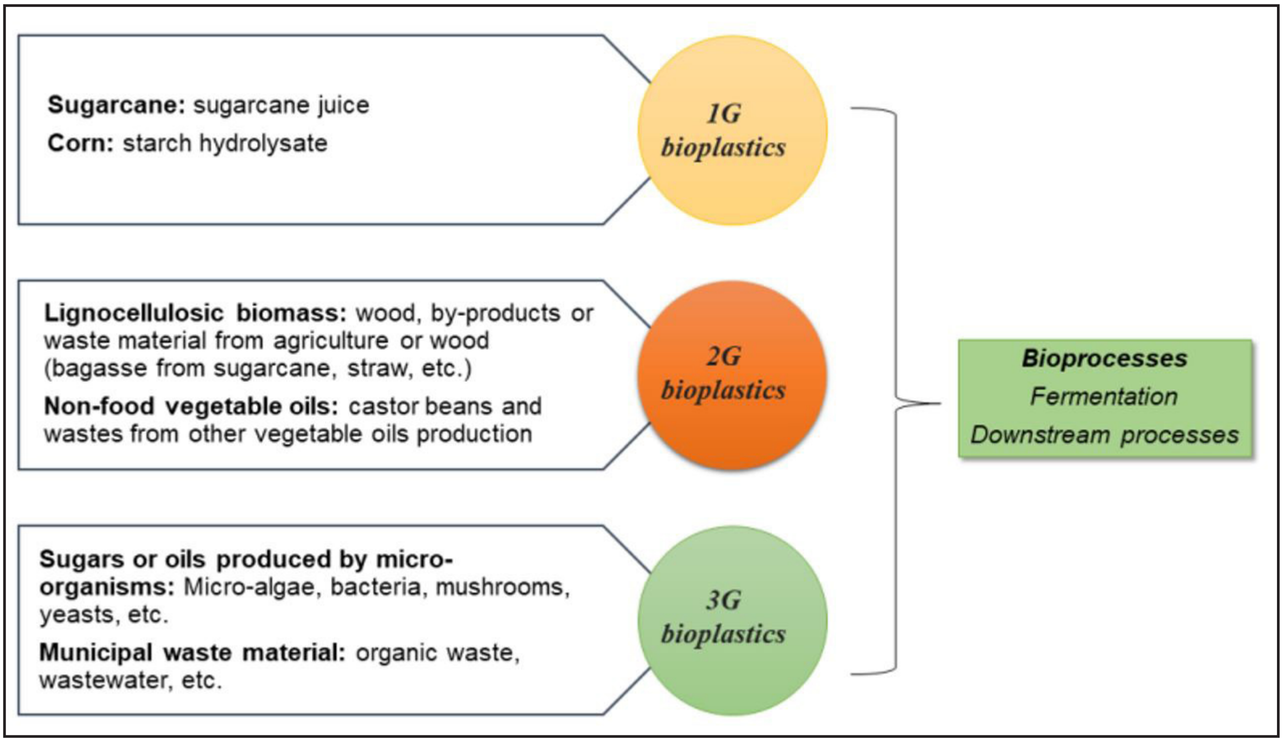


Figure 1. Classification of bioplastics according to the source. Modified from (NaturePlast, 2021).

technological and economic issues limited their competitiveness with fossil-based materials. In the first decade of the 00's, plastics of renewable origin with chemical structures identical to those of fossil origin were developed, and this increased their range of applications. However, their competitiveness was in great part dependent on the oil prices (nova-Institute, 2018; Wydra et al., 2021).

Wydra et al. (2021) applied Multi-Level-Perspective's transition pathways to the niche of bioplastics in the developing bioeconomy. The following types of transition pathways were identified: *transformation* and *technological substitution* – here, the cost competitiveness of bioplastics would increase as a result of high oil prices associated with fast technological progress; *reconfiguration* (1: eco-regime) – this pathway would depend on the development of a governmental circular economy strategy and a climate protection policy; *reconfiguration* (2: eco-innovation-regime) – this pathway would result from the embrace of an anti-pollution policy focused on plastics, like the reduction of polymeric microparticles and marine pollution. Finally, the *dead end* or stagnation of this niche could be a consequence of bioplastics causing the same pollution problems as fossil-based plastics; of bioplastics suffering from decreased feedstock availability or feedstock competition with other segments resulting in increased production prices; or of CO_2-based plastics being prioritized over bioplastics due to their increased capacity to mitigate climate change. In this sense, the transition to a circular bioeconomy in the segment of materials depends on complex arrangements of several interconnected factors, that should be carefully addressed in the development of a consistent bioeconomy strategy.

Techno-economic analysis of bioplastics

Despite the great diversity of low-cost feedstocks, such as illustrated in Figure 2, the flow market of bioplastics is still low. In addition, a techno-economic analysis has not been carried out for most of them. Despite several advantages of microbial bioplastics over synthetic plastics, the main bottleneck for their production is their non-cost effectiveness. The major production costs are related to the fermentation process, carbon sources, yield and productivity of the process, downstream processing and other details involved. So, commercialization of bioplastics is still limited in comparison to synthetic plastics. For an economical production of 2G and 3G, the definition of suitable feedstock and efficient producing strains is determinant for competitiveness of biobased products (Naresh Kumar et al., 2020). Another important point would be the integration of bioplastics production in a biorefinery producing chain, where some benefits such as energy consumption, feedstock transport and common use of some equipment could enable the process with positive impact over final costs.

The 2G bioplastics production

Nowadays, 2G bioplastics take part in the green circular economy with a fundamental role with the use of 5C-6C sugars for their production. Earlier, 1G bioplastics were usually derived from carbohydrate-rich plants such as corn, sugarcane, or sugar beet, which were strictly considered as food biomass with a critical competition with the agro-industrial market and increasing the food costs (Wellenreuther & Wolf, 2020). As a result, alternative biomasses have been exploited for the so called 2G bioplastics that are mainly derived from processes' wastes, which are composed of pentoses and bioconverted into new revetments and packages. As alternative substrates, corn stover, sugarcane bagasse, wheat, and other crop residues have been exploited. Substrates considered as non-food are utilized, such as wood lignin, recycled paper, and wastewater, which depending on its composition, can be considered as biomass for 3G bioplastics (Ögmundarson et al., 2020). Figure 3 presents different substrates that are used for 2G bioplastics production.

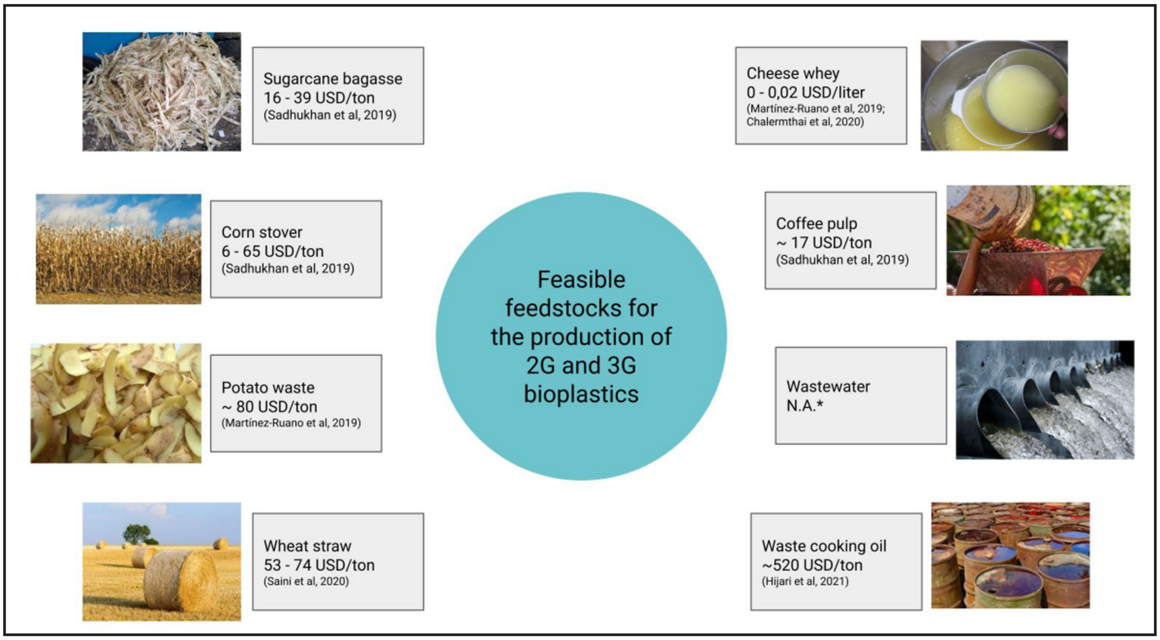


Figure 2. Feasible substrates for bioplastics production and their average market prices.

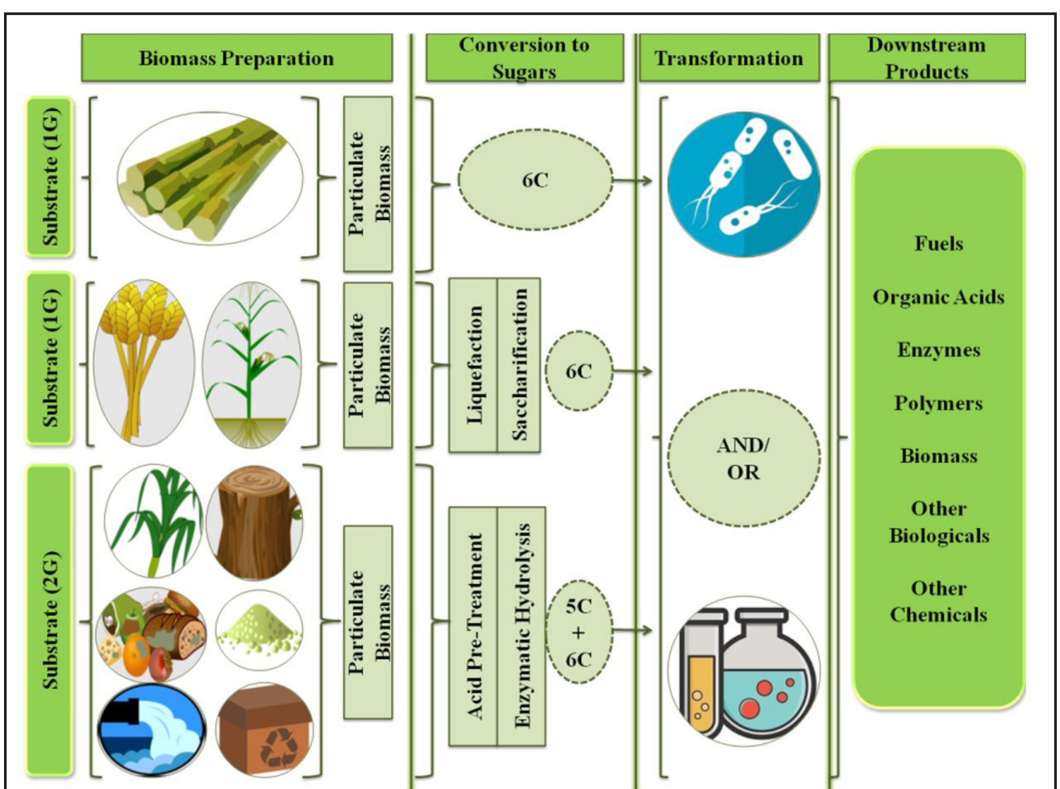


Figure 3. Substrates and treatment for the production of 1G and 2G bioplastics

Regarding sustainability, this tendency is eco-friendlier and more economic with biomass availability all over the year and with efficient logistics, but with certain challenges to overcome. To surpass the new challenges, it is necessary to provide engineering microorganisms, which are able to convert pentoses into new biobased plastics. Another aspect to consider is the inclusion of supplementary process steps that have high demands of water and chemical solvents, which will require final product purification (Changwichan et al., 2018).

The three most important 2G bioplastics in the industry point of view are: poly-lactic acid (PLA), polyhydroxyalkanoates (PHA), and polyhydroxybutyrate (PHB). Some examples of substrate and treatment for 2G bioplastics production are presented in Table 1. Different chemical solvents and enzymatic hydrolysis are employed.

The 3G bioplastics production

The use of microalgae biomass as an alternative feedstock to petrochemical and plant-based resources in a 3G bioplastic production is encouraged due to its high biomass productivity, and the eco-friendly ability to absorb greenhouse gases (GHGs) in its autotrophic complex (Elrayies, 2018). Microalgae are photosynthetic microorganisms with simple cell structure, that requires light, CO_2, water, and micronutrients for growth (Mustafa et al., 2021). Their bioactive composition consists of lipids (8-70%), proteins (40-70%), carbohydrates (11-56%), and carotenoids (10-14%) (Devadas et al., 2021; Muhammad et al., 2021). Their potential contribution to the global bioeconomy include a wide range of bioproducts, such as biofuels, biofertilizers, animal and human nutrition,

Table 1. Substrates and methods for 2G bioplastic obtention.

2G Bioplastic	Substrate	Method	Reference
PLA	Sugar beet pulp	Acid and enzymatic hydrolysis, fermentation	Alves de Oliveira et al. (2020)
PLA	Starch from potato waste	The cradle-to-factory gate scope	Broeren et al. (2017)
PLA	Seaweed Ulva spp.	Glucose obtained from hydrolysis, fermentation	Helmes et al. (2018)
PLA	Cheese whey	Enzymatic hydrolysis and fermentation	Liu et al. (2018)
PLA	Coffee waste and coffee pulp	Acid and enzymatic hydrolysis, fermentation	Pleissner et al. (2016)
PHA	Coffee waste – oil extraction containing fatty acids	Organosolvent extraction, evaporation and fermentation	Bhatia et al. (2018)
PHA	Cellulose from rice husks	Alkaline and enzymatic hydrolysis, fermentation	Heng et al. (2017)
PHA	Cheese whey	Acidogenic fermentation	Israni et al. (2020)
PHA	Municipal wastewater	Carbon-rich residuals conversion from industrial/agricultural process into volatile fatty acids for fermentation	Morgan-Sagastume et al. (2016)
PHA	Pineapple peel solution	Fermentation	Penkhrue et al. (2020)
PHA	Chicken feather waste	Fermentation	Pernicova et al. (2019)
PHA	Waste cooking oil	Fermentation	Sangkharak et al. (2021)
PHA	Palm oil	Fermentation	Thinagaran & Sudesh (2019)
PHA	Wastewater sludge	Pyrolytic pre-treatment, fermentation	Vogli et al. (2020)
PHB	Waste paper from municipal solid waste	Enzymatic hydrolysis pre-treatment and fermentation	Al-Battashi et al. (2019)
PHB	Lignocellulosic biomass	Hot water pre-treatment and fermentation	Yin et al. (2019)

as well as bioremediation and nutrient recycling through wastewater treatment, and carbon fixation (Mendonça et al., 2021). Furthermore, microalgae can be produced in areas unsuitable for agriculture in different photobioreactors. In comparison with terrestrial plants cultivation, microalgae has higher concentrations of chlorophyll per unit area of production, resulting in a CO_2 removal that is 10 to 50 times higher, which represents 1.83 kg of CO_2 per kg of algal biomass (Mendonça et al., 2021; Raeesossadati et al., 2014). In addition, its biomass productivity is 5 to 10 times faster than conventional food crops. Therefore, microalgae are a potential feedstock to biopolymer industry.

Chlorella sp. and *Spirulina* sp. are the main microalgae species employed for bioplastic production. Researches focused on optimization of blending composition (Table 2), with the use of additives, together with microalgae biomass, to reach biomaterials with higher quality than conventional plastics (Rai et al., 2021; Zeller et al., 2013). One alternative route is the intracellular accumulation of biomolecules, such as PHA, PHB and starch, and its subsequent extraction (Kartik et al., 2021). The PHA synthesis in microalgae metabolism occurs naturally, however, media conditioning with nutrient deficiency, such as phosphate and nitrogen depletion, can increase drastically its concentration.

Table 2. Summary of algae-based bioplastic production.

Biomass species	Product	Process description	Reference
S. platensis	*S. platensis*-PVA blend film	6% MAH and 30% Glycerol; tensile strength at 27.7-28.26 kgf/cm^2 and 59.17-66.0% elongation	Dianursanti et al. (2018, 2019)
S. platensis	*S. platensis*-wheat gluten blend thermoplastic	30% microalgae: wheat gluten, compression mold at 120 ºC, 40 bar, 10 min, glycerol or 1,4-butanediol as plasticizers	Ciapponi et al. (2019)
***Spirulina* sp.**	*Spirulina*-Polybutylene succinate (PBS) composites	Melt blending at 130 ºC, 6 min, with 6% MAH-grafted PBS and 15-50% *Spirulina* biomass	Zhu et al. (2017)
Chlorella	*Chlorella*-PVA blend films	Ultrasonic *Chlorella* treatment, solvent casting at 80 ºC, with glycerol and citric acid; 15.3 kgf/cm^2 tensile strength and 99.63% elongation	Sabathini et al. (2018)
Chlorella vulgaris	*Chlorella*-PVA blend film	Compression mold at 120 ºC between *Chlorella*:glycerol and MAH grafted PVA; 42.25 kgf/cm2 tensile strength and 13.0% elongation	Dianursanti & Khalis (2018)
***Scenedesmus* sp.**	PHB	Glucose, N, P, Fe, and salinity concentration optimized by Taguchi design; 0.83–29.92% (w:w) dry weight final accumulation	García et al. (2020)
Chlorogloea fritschii	PHB	5% (v:v) inoculum in 150 mL BG-11 medium, pH 7.5, 32 ºC, 100μmol/m^2/s light, CO_2 supply at 160 rpm; 51% (w:w) substrate conversion yield	Monshupanee et al. (2016)
Botryococcus braunii	PHB	60% of sewage wastewater at pH 7.5, 40 ºC; 247 mg/L of PHB	Kavitha et al. (2016)
***Spirulina* sp. LEB 18**	PHB	Adapted culture with 8.4 g/L $NaHCO_3$, 0.05 g/L $NaNO_3$, and 0.5 g/L KH_2PO_4; 30.7% (w:w) dry weight	Coelho et al. (2015)
***Chlorella* sp. and *Scenedesmus* sp.**	PHB, lipids and bioethanol	Hybrid pretreatment and fermentation with wastewater treatment inoculum; 0.43 g PHB/g dry cell weight; 76.17% sugar utilization	Naresh Kumar et al. (2020)
Chlorella pyrenoidosa	PHB and biodiesel	27.0% (w:w) dry weight, after 14 days under optimized conditions	Das et al. (2018)

The amount of nutrient supplementation, light variations and reaction conditions also influence biopolymer accumulation levels (Costa et al., 2019). Genetic modifications in microalgae species were also conducted to increase the production of PHAs (Kaparapu, 2018). Some recent works applied the biorefinery concept to convert different fractions of algal biomass into high value-added products (Costa et al., 2019). In this context, the microalgae biomass is integrally exploited as a substrate in conventional biorefinery processes, such as pre-treatment and microbial fermentation techniques.

The main 2G and 3G bioplastic market and producers

The global bioplastics market size was valued at USD 8.3 billion in 2019 with a production capacity of around 4 million tons (European Bioplastics, 2018; nova-Institute, 2018). A compound annual growth rate (CAGR) of 16.1% from 2020 to 2027 (Grand View Research, 2020), is expected, which is driven by the packaging end-use industry. Other industries such as consumer goods, automotive and transportation, agriculture and horticulture, and textile, among others also employ bioplastics and biopolymers (Markets and Markets, 2021). Actually, biobased polymers are mainly produced from renewable resources, which can compete with human or animal food. However, bioplastic manufacturing represents only approximately 0.02% of the global agricultural land (NaturePlast, 2021).

Based on region, the bioplastics market has been segmented into APAC, Europe, North America, RoW. The major players are NatureWorks (Italy), which leads the group of manufacturers, Braskem (Brazil), BASF (Germany), Total Corbion (Netherlands), Novamont (Italy) and others (Markets and Markets, 2021). The main commercialized bioplastics are PLA, PHAs, starch plastics, cellulose plastics and proteins plastics (Zhang et al., 2019). PLA, for example, is obtained from corn by NatureWorks™ and BioPE is produced from sugarcane by Braskem, in Brazil.

Recent patents and innovation on 2G and 3G biobased bioplastics

A search of patent databases and innovations during the last 20 years was carried out where the most relevant results and information are presented in Figure 4. The analysis shows that the number of published patents about biobased bioplastics has increased steadily since 2010 (Figure 4a). That tendency is consistent with the rise in global awareness of the worldwide negative impact of petroleum-based plastics and the necessity to reduce reliance on these materials. Besides, actions were taken by the major players and countries in the market, with the USA, South Korea, and China topping the list, which may have boosted the number of published documents (Figure 4b).

The predominant bioplastics cited in the analyzed documents are PHA and PLA (Figure 4c), both produced at least as 2G biopolymers. Nowadays, PLA has a consolidated market, with applications in several areas such as biomedical, cosmetics, and pharmaceutical. Thus, most of the documents involving PLA in the original database were about formulated products and applications rather than the production process or use of different raw materials. On the other hand, the large-scale production of PHA is an in-development and rising technology, mainly because the reduction of the production costs is still needed, and thus, the application of alternative feedstocks must be reinforced.

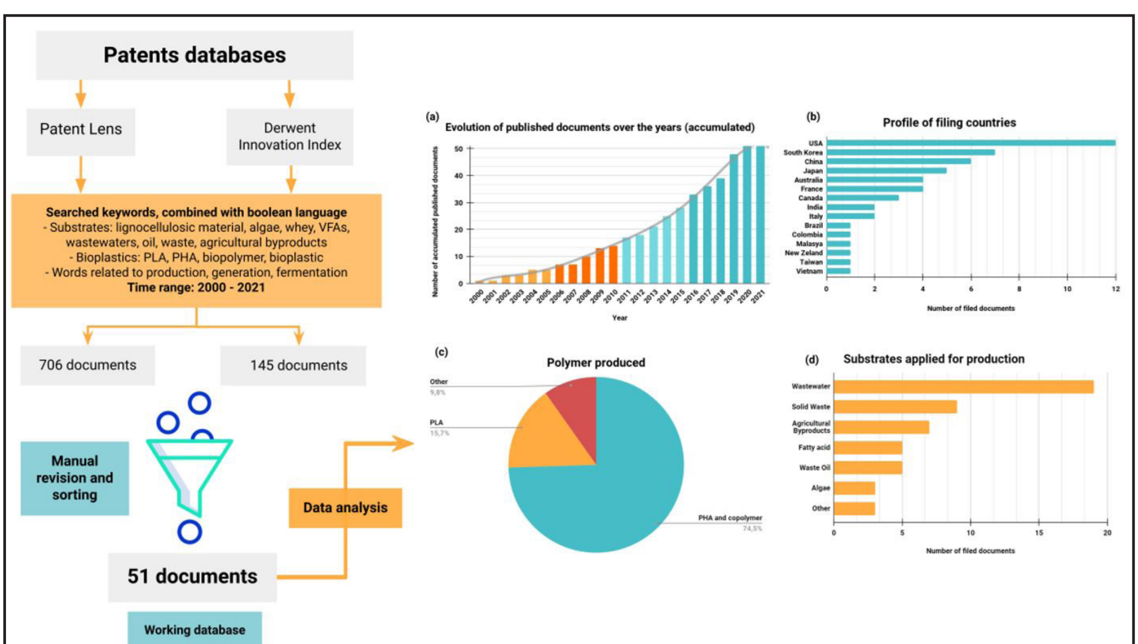


Figure 4. Patents and innovation on 2G and 3G bioplastics in the last 20 years.

Regarding 2G bioplastics, some of the residues cited for its production were lignocellulosic materials (i.e., wasted agricultural straw, industrial rice residues, wood cellulose, and others) as well as slurry, organic waste, cheese whey, crude glycerol, and wastewater of various origins (Anderson & Anderson, 2016; Goyal et al., 2019; Cuervo, 2020). Among these substrates, wastewater was the most recurrent in the analyzed documents (Figure 4d). These effluents are generated worldwide in large quantities, and their treatment and disposal are an issue that requires urgent addressing. This residual material is used as substrate for bioplastics production because of the immediate accessibility of nutrients (while, for example, lignocellulosic materials may need several pre-treatments for sugar release). Finally, examples of algae biomass as feedstock for 3G bioplastics were found, but to a lesser extent (Jin et al., 2013; Yamada et al., 2020).

Conclusions

Over the years chemical-based plastics are still in the domain of the market due to their durability and large-scale production, which makes the 1G biobased plastics production more expensive. However, the new generation of biobased materials, the 2G and 3G, are promising alternatives, since they are mainly produced from renewable resources, which can compete with human or animal food with reduced land use. New research and innovation are being developed where PLA and PHAs are the main produced bioplastics, which can be produced from lignocellulosic biomass, non-food vegetable oils, sugars oils produced by microorganisms and municipal waste materials. Until now, the biobased products' market price prospection is low, but, as it was elucidated; their base-substrates are cheap and have great potential to integrate the green circular economy.

Conflict of interests

The authors declare that they have no conflicts of interest.

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