Bio-polyethylene Furanoate (Bio-PEF) from Lignocellulosic Biomass Adapted to the Circular Bioeconomy

Carolina Mónica Mendieta,* Giselle González, María Evangelina Vallejos, and María Cristina Area

There is a global trend to replace the production of conventional recyclable plastics with biobased ones, allowing a sustainable alternative adapted to the current concept of a circular bioeconomy. Forest-industrial and agricultural biomass wastes (lignocellulosic biomass waste, LCBW) produce severe problems in some developing countries because they are improperly disposed of or burned in the open air. Such wastes are attractive as a raw material to produce bioplastics due to their low cost. Furthermore, low-pollution processes can complete an economical and environmentally friendly approach. This review focuses on bio-polyethylene furanoate (PEF) production from LCBW as an alternative for polyethylene terephthalate (PET), one of the most widely used fossil-based plastic. The standpoint is based on the replacement of fossil-based monomers for the manufacture of PET, terephthalic acid (TPA), and ethylene glycol by two bio-based monomers, namely 2,5-furandicarboxylic acid (FDCA) and bio-ethylene glycol (Bio-MEG). This study describes the processes to obtain each bio-monomer, as well as the resulting polymers' performance aspects, biodegradability, environmental and economic considerations, and recycling.

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Contact information: IMAM, UNaM, CONICET, FCEQYN, Programa de Celulosa y Papel (PROCYP), Misiones, Argentina, Félix de Azara 1552, Posadas, Argentina;
** Corresponding author: caroo.mendieta@gmail.com*

INTRODUCTION

Fossil-based plastics are chemical products used worldwide. They are obtained from non-renewable oil by cracking hydrocarbons. They are polluting and usually non-biodegradable (Alauddin *et al.* 1995; Harding *et al.* 2007; Andrady 2017; Yadav *et al.* 2020). Usually, these plastics replace cotton, glass, or metal, since they are economical, highly available, and manageable for multiple purposes. Despite their great utility for industrial progress and in the food and health sectors (Mulder 1998; Andrady and Neal 2009; Gibb 2019; Yates *et al.* 2019; Sandu *et al.* 2020), many plastics are discarded immediately after use. Their accumulation has become a problem for the environment, the economy, and human health (Hopewell *et al.* 2009; Kunwar *et al.* 2016; Giacovelli 2018; Falappa *et al.* 2019; Okunola *et al.* 2019; Schmaltz *et al.* 2020). Besides, there is growing interest in greenhouse gas emissions during production, processing, and life cycle, as well as pollutants from the extraction of the raw material until the final disposition in land,

oceans, rivers, or lakes (Brandt *et al.* 2011; Royer *et al.* 2018; Shen *et al.* 2020). However, the production of pollutants is low in comparison with their accumulation, lack of recycling policies, and scarce biodegradation.

The weathering effect of the sun's UV radiation and the implementation of chemical additives such as d2w® for backbone degradation of the polymer generates smaller pieces of the material, called microplastics (Urbanek *et al.* 2018; Hale *et al.* 2020). These are more prone to biodegradation and mineralization than larger objects due to higher accessibility (Farzi *et al.* 2019). However, these small-sized particles of plastics (from less than 5 mm up to the order of microns), together with chemical additives such as bisphenol-A, can be consumed by animals and insects, hampering the health of both animal and human beings because of occlusions of the digestive tract and motor skills, among others (Alava 2020; Miller *et al.* 2020). Equally important, the reuse or recycling of already produced plastics is relevant to stop accumulating this material that could be exploited in several applications when possible. Non-biodegradable bioplastics can be a part-time solution for fossil plastics. However, if recycling policies are not improved, there could still be the problem of accumulation in landfills and water ecosystems.

Polyethylene terephthalate (PET) is one of the most produced plastics globally, about 87 MMT in 2022 (Chowdhury *et al.* 2018). China is the largest producer with more than 50% of the market share ("Global Polyethylene Terephtalate Market Report 2017 - By End-Use Industries, Products & Regions - Research and Markets | Business Wire" n.d.). PET is practical to mold, recyclable, and versatile, so it is the main material in most plastic products, such as bottles (Jankauskaite *et al.* 2008; Marathe *et al.* 2019). It is produced by polymerization of two fossil-derived monomers, terephthalic acid (TPA), obtained from catalytic reforming derived p-xylene, and monoethylene glycol (MEG), obtained from the ethylene derived from the thermal cracking of naphtha (Zhao *et al.* 1996; Eerhart *et al.* 2012; Li-Na 2013; Han 2019). This processing route produces significant amounts of CO_2, since 1 ton of PET generates 4 tons of CO_2, contributing to global warming (Movilla-Quesada *et al.* 2021). Currently, PET is made from fossil resources because it is more economical than bio-based production. Despite the fact that the plastic with some content of carbon 14 is considered biobased, in this work, the term "bio-based" refers to 100% biomass-based, measured by standards such as CEN/TS 16137 (Europe standard) or ASTM 6866 (United States Standard) (Taguchi *et al.* 2014).

It is estimated that plastics production will increase to 700 million and 1.8 billion tons in 2030 and 2050, respectively, where 9% of the plastic used is recycled, 12% is incinerated, and the remaining 79% is plastic waste that generates pollution problems in the environment (Más Azul 2020). In 2019, both the plastics production process and its incineration caused the release of 850 million metric tons of greenhouse gases into the atmosphere (Plastic & Health 2019). If production volumes continue on their current growth path, gas emissions could reach 1.34 (2030) and 2.8 (2050) gigatons of CO_2 per year, accumulating in the atmosphere over time (Muffett 2019). Figure 1 represents the problems caused by plastics during and after their useful life (Boehe 2011; Puma 2011; Clauser *et al.* 2021b; Ganesh *et al.* 2020).

For these reasons, it is required at a global level to adapt the production of sustainable plastics in a circular bioeconomy (Rosenboom *et al*. 2022), which seeks responsible consumption, the sustainable use of biodiversity, and the development of industry and innovation (Enguix 2019; Mendieta *et al.* 2021a). In addition, the balance between production and consumption by responsible use of resources is expected (Cuevas 2020).

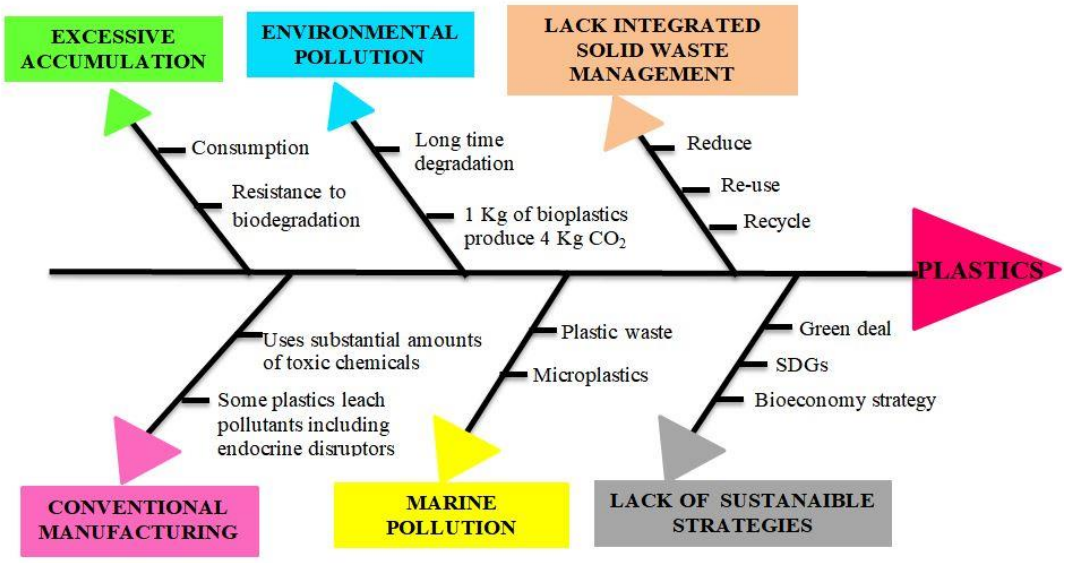

Fig. 1. The problem of plastics explained with an Ishikawa diagram

Biomass is a great candidate for production of oxygenated high-value compounds (Xin *et al.* 2020). Especially, lignocellulosic biomass waste (LCBW) is a sustainable raw material to produce bioplastics, since it is an economical, renewable, and available carbon source (Ezgi Bezirhan Arikan and Havva Duygu Ozsoy 2015; Brodin *et al*. 2017; Andreeßen and Steinbüchel 2019a).

This alternative can reduce emissions of greenhouse gases (GHG), decrease the dependence on fossil resources (Chen and Patel 2012; Clauser *et al*. 2021c), and close the carbon cycle, since the CO_2 generated can be fixated by the raw material through photosynthesis (Zhang and Peng 2017a). Second-generation biobased products (2G) are preferred over first-generation biobased products ones (1G) because they do not compete with food and feed, are highly availability, and have a low cost (Mendieta *et al*. 2021a).

Fully biobased PET is not produced yet. Partially biobased PET is obtained from bio-MEG by 1G bioethanol conversion and further transesterification with fossil-based TPA. Estimations of its global production accounted for 7 million tons in 2020. The high demand for biobased PET motivated the development of commercial processes for fully biobased PET and has generated investment by companies such as Coca-Cola, Ford, Nike, and others. The production of partially biobased PET from 2G bioethanol requires pretreatment of the lignocellulosic raw material, implying high energy requirement in the process, together with the use of chemicals (Chen *et al.* 2016).

On the other hand, Avantium is one of the companies inducing the transition towards bio-based plastics with the "YXY building blocks" to replace fossil-based polyesters, such as PET, with furanic polyesters. For example, the commonly used terephthalic acid (TPA) can be replaced with 2,5-furandicarboxylic acid (FDCA) in conventional plastic bottles, fibers, and textiles, among others (De Jong *et al.* 2011).

Enzymatic studies have shown that PEF films degrade 1.7 times faster than PET ones, but further studies in the soil under accurately controlled conditions are necessary to assess and compare both biodegradabilities (Loos *et al*. 2020). Oriented PEF bottles are compatible with existing recycling equipment, can be mechanically recycled at up to 5 wt% together with PET, and have outstanding barrier properties. Moreover, the O_2 barrier property was six times greater than PET, whereas the CO_2, and H_2O barrier properties were

twofold more. PEF has a higher glass transition temperature (T_g), lower melting temperature (T_m), and lower Heat Deflection Temperature (HDT), higher tensile strength, lower elongation to break, and higher density than PET (De Jong *et al.* 2012). In addition, PEF has better performance than PET in three-dimensional (3D)-printing and could be recycled for the same use several times (Kucherov *et al.* 2017).

The production route for PEF adapts well to a circular bioeconomy (Kim *et al.* 2022), whose main objective is the reinsertion through the recycling process at the end of their life cycle (Coppola *et al.* 2021; Dahmen *et al.* 2019; Gatto and Re 2021; Orejuela-Escobar *et al.* 2021). In addition, replacing plastics with biopolymers can help reduce food waste and prevent the accumulation of plastics in the soil (Rendón-Villalobos *et al.* 2016; Shamsuddin and Isah 2018; Su *et al.* 2018; Ribba *et al.* 2020). A comparison of both PET and PEF processes is shown in Fig. 2.

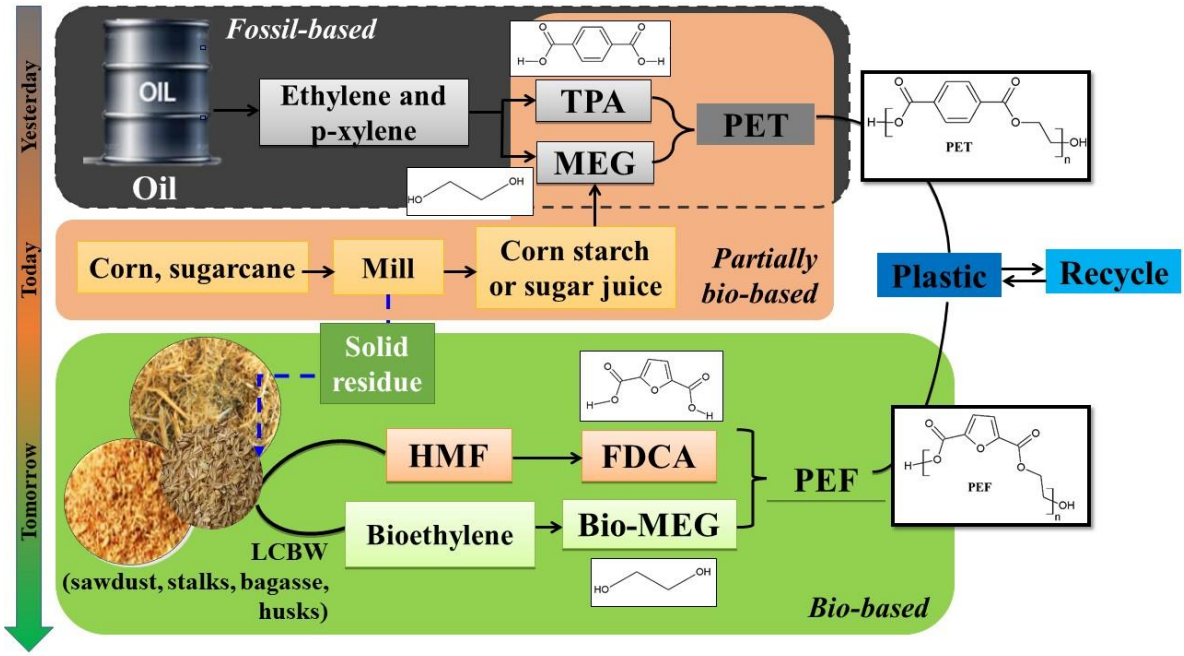

Fig. 2. Routes for bioplastics production from LCBW (p-xylene: paraxylene; MEG: monoethylene glycol monomer; TPA: terephthalic acid; HMF: 5-hydroxymethylfurfural; FDCA: 2,5-furandi-carboxylic acid; PET: polyethylene terephthalate; PEF: bio polyethylene furanoate; LCBW: lignocellulosic biomass waste). Based on references (Wilson *et al*. 2018; Volanti *et al.* 2019; Loos *et al.* 2020)

Whether using oil or biomass as raw material, both materials need to be subjected to a prior process to reach the platform compounds from which arise the structural monomers of bioplastics. For example, in the case of obtaining TPA and MEG, reforming and cracking reactions systems have to be firstly involved (Busca 2021). On the other hand, biomass needs to be subjected to a fractionation/pretreatment process to separate the structural polymers (cellulose, hemicelluloses, and lignin) and release the sugars from carbohydrate fraction, which could be the platform molecules of the FDCA and Bio-MEG (Kim *et al.* 2020a; Mendieta *et al.* 2021a).

This review focuses on PEF production from LCBW as an alternative for PET, which is one of the most heavily utilized fossil-based plastics. The standpoint is the replacement of fossil-based monomers by two bio-based compounds, such as FDCA and

bio- Bio-MEG. This work raises the possibility of producing PEF from lignocellulosic biomass wastes to give this residue an added value, generating jobs and promoting integral sustainability in the regions of high LCBW generation. It presents the obtaining of the monomers from LCBW, the polymerization of the FDCA and Bio-MEG monomers, and the techno-economical concerns of the proposal.

ROUTE FOR PEF PRODUCTION

PET and PEF production

The polymerization of bio-monoethylene glycol (Bio-MEG) and 2,5-furan dicarboxylic acid (FDCA) produces polyethylene furanoate (PEF) (Hwang *et al.* 2020a; Loos *et al.* 2020). FDCA can be obtained from 5-hydroxymethylfurfural (HMF), obtainable from fructose and glucose (Song *et al.* 2020). The difference between MEG and Bio-MEG is the raw material. However, the TPA and FDCA present different chemical structures, which leads to other properties of the resulting material (Eerhart *et al.* 2012; Loos *et al.* 2020).

First, the FDCA or FDCA diester undergoes transesterification with the MEG at 160 to 180 °C for 1 to 2 h, resulting in 99% conversion and producing methanol as a byproduct, which could be separated by evaporation. The FDCA generates a colored product, so the diester of FDCA, *e.g.*, dimethyl 2,5-furandicarboxylate (dmFDCA), is preferred. A polycondensation, at 230 to 240 °C and pressures below 1 mbar at melt conditions, follow the transesterification. The process ends when the obtained polymer has a number average molecular weight not less than 10,000. A subsequent solid-state polymerization (SSP) is carried out for greater molecular weights, where elevated temperatures below the melting point are applied. In SSP, the catalyst system Sn(IV)/Sn(II) leads to a higher molecular weight than Ti catalysts. Also, dmFDCA is more reactive than dimethyl terephthalate, allowing lower temperatures and reaction times than PET SSP. Conventionally used catalysts for obtaining PET (with Mn, Co, and Ge) generate a colored PEF (De Jong *et al.* 2012). This effect could be due to chromophores generated in the process.

The synthesis of PEF via ring-opening polymerization (ROP) has been proposed, replacing the polycondensation. In this process, dmFDCA and MEG are subjected to prepolymerization to obtain short linear PEF oligomers, further depolymerized to cyclic oligomers under dilution in a high boiling solvent (*e.g.*, 2-methylnaphthalene or 1,2-dichlorobenzene). Later, the purified cyclic oligomers are polymerized by ROP to reach the PEF. The catalyst is a solid powder of cyclic stannoxane. Thus, it is possible to increase the reaction conversion (>95%) and obtain bottle-compatible PEF (high molecular weight, > 30 kg mol^{-1}) without the presence of colored products in shorter reaction times (Rosenboom *et al.* 2018).

Hoppe *et al*. (2018) gave evidence of the presence of oligomers PEF and highlighted them as potential migrants to food when in contact with the material. Further studies of this aspect should be carried out. Concerning GHG emissions, a report of PEF produced by polymerization of FDCA from corn-starch and MEG (fossil or biobased) indicates that PEF production could reduce the Non-Renewable Energy Use (NREU) by 40 to 50% and GHG emissions by 45 to 55%. These reductions are higher than for other bioplastics such as polylactic acid or polyethylene and can increase if the replacement of PET for PEF reaches the fibers and film industry. However, the work emphasizes the necessity of a similar study

for PEF based on lignocellulosic materials, since starch is considered food (Eerhart *et al.* 2012).

Bio-monoethylene Glycol (Bio-MEG) from Biomass

Firstly, in this process, the LCBW has to go through a pretreatment to separate the lignin from the carbohydrates that compose cellulose and hemicelluloses to increase the enzymatic accessibility (Vallejos *et al.* 2017). Lignin is an inhibitor for the microorganisms employed in subsequent processes, since it obstructs the accessibility to the substrate (Kruyeniski *et al.* 2019). Once the polysaccharides are isolated, a Simultaneous Saccharification and Fermentation (SSF) strategy for 2G bio-ethanol production proceeds, which is preferred because it allows high substrate loading (Olofsson *et al.* 2008; Mendieta *et al.* 2021b, 2022). Afterwards, the dehydration of 2G bioethanol generates 2G bioethylene (Mohsenzadeh *et al.* 2017; Mendieta *et al.* 2021a). Following this, the 2G bioethylene goes through a catalytic epoxidation/oxidation, commonly using silver (Ag) as a catalyst for the formation of bioethylene oxide (Bio-EO) (López 2014). Finally, the hydration of ethylene oxide in the presence of an acid catalyst leads to the formation of bio-monoethylene glycol (McClellan 1950; Kandasamy *et al.* 2019a). This process is illustrated in Fig. 3.

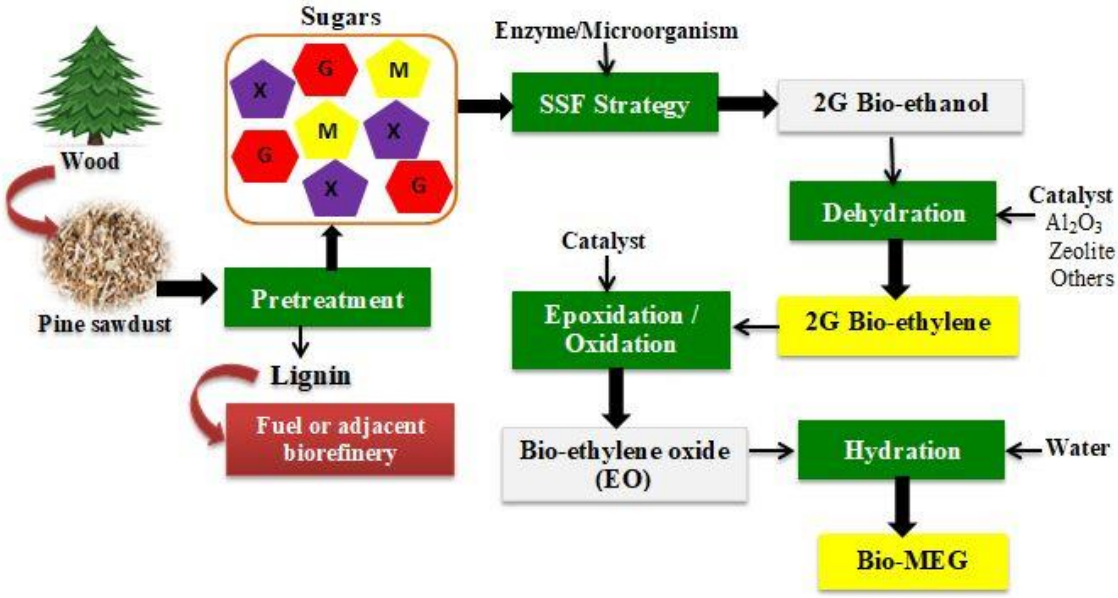

Fig. 3. Route for bio-ethylene glycol production from LCBW

MEG from biomass is currently produced from 1G ethanol using a completely new process for the one-step production of bio-ethylene oxide from bio-ethanol. The cooled reactor is sized for air-based bioethanol oxidation, producing ethylene oxide in a single step. Then, the product is separated from the gas phase effluent through absorption in a hydro-alcoholic solution (Salusjärvi 2019; Ripamonti *et al.* 2021). On the other hand, EO production in fossil-based industries is generally carried out in fixed-bed reactors using an ethylene oxidation mechanism in a stream of air or oxygen with the help of a silver-based catalyst in the gaseous phase (Montrasi *et al.* 1983). EO is an industrial organic derivative that is widely used for producing MEG, non-ionic surfactants, alcoholic ether, and other oxygenated chemicals. Since 2013, it has had a demand of more than 20 million metric tons with a 6 to 7% annual growth rate (Ghanta *et al.* 2012a; Lu *et al.* 2016a).

CO_2 is the main byproduct, so it is necessary to maintain the ethylene conversion in a range of 10 to 15% to minimize the products that generate the combustion. The achieved selectivity of EO higher than 90%, so the resulting yield can be near 9 to 13.5% (Ghannadzadeh and Meymivand 2019). Safe and more environmentally friendly technological developments are needed, adopting sustainable alternatives to minimize CO_2 formation in the process (Faria *et al.* 2020). Safety is a relevant factor in conventional EO production due to the generation of explosive mixtures in the reaction between ethylene, EO, and air in the gas phase. Currently, researchers achieve EO production while mitigating impact factors on the environment, safety, and health (Ghanta *et al.* 2012b).

A methodic study of EO production using catalysts based on titanosilicates of different topologies obtained selectivities of 90 to 100% ethylene oxide (Lu *et al.* 2016b). On the other hand, Lee *et al.* (2010) evaluated an EO production mechanism using a liquid phase process employing a homogeneous catalyst methyl trioxorhenium (MTO) and aqueous hydrogen peroxide (H_2O_2) as an oxidant in methanol/water reaction medium under mild process conditions. An increase of pressure (50 bar) allows the condensation of the ethylene into the liquid medium and the dissolution in the organic solvent. This mechanism enables a catalytic process under a completely homogeneous liquid phase to eliminate CO_2. EO is then produced at 48% yield and 90% selectivity at 40 ºC and is recovered through distillation because of its low boiling point (10.8 ºC), giving an advantage to the recycling of the catalyst.

Fossil-based monoethylene glycol (MEG) is a diol with several applications, such as in the production of plastics (PET) and methanol (Kandasamy *et al.* 2019b). For MEG production at the industrial level, EO is thermally hydrated without the aid of a catalyst at a temperature of approximately 200 °C (Yue *et al.* 2012). High MEG production requires a large amount of water (20 to 25 mol water/mol EO) (Nexant 2010). In this process, diethylene glycol (DEG) and triethylene glycol (TEG) are generated as byproducts (Eq. 3 and 4 in Fig. 4) in small amounts, since EO reacts faster with ethylene glycols compared with water (Chemicals-Technology 2020).

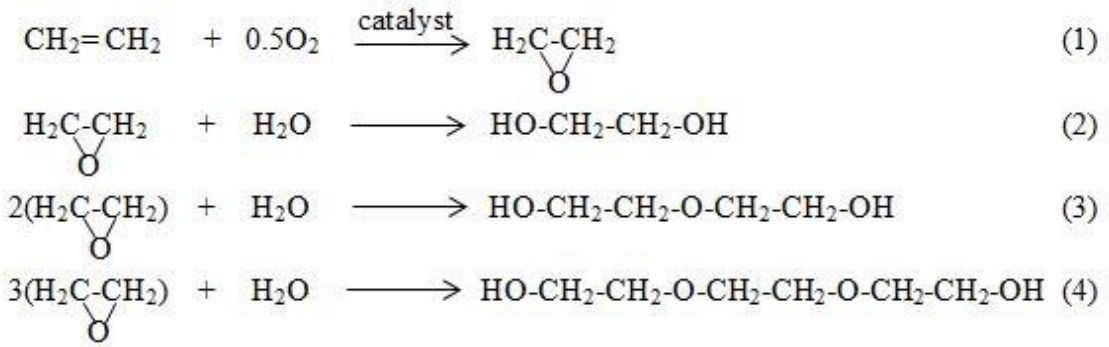

Fig. 4. Scheme of the reaction mechanisms for obtaining EO, MEG, and byproducts (DEG and TEG) from ethylene. Based on references (Othmer and Thakar 1958a; Yue *et al*. 2012)

Using an excess of water can maximize the EO conversion towards MEG up to 90%, and both DEG and TEG are easily separated by distillation (Othmer and Thakar 1958b). Although it is a simple and straightforward method, its drawback is the energy consumption required during distillation due to the amount of used water and the formation of ethylene glycols (Altiokka and Akyalçin 2009). For this reason, different catalysts capable of optimizing MEG selectivity and reducing the reaction temperature and the excess water

required in the process have been studied (van Hal *et al.* 2007a). The yield of MEG from cellulose can be calculated through Eq. 5, considering that the weight of the catalyst does not vary during the reaction, and the cellulose conversion was calculated based on the solid weight difference before and after the reaction (Xi *et al.* 2014).

$$MEG\ yield\ (\%) = \frac{moles\ of\ produced\ EG}{initial\ moles\ of\ glucose} * 100 \quad (5)$$

Shvets *et al.* (2005) developed a mathematical model for the EO hydration process in a fixed bed catalytic reactor, adequately describing the determined parameters: reaction rate, product distribution in the reactor, swelling, and catalyst deactivation. They reached an EO conversion higher than 95% and more than 98% ethylene glycol selectivity. Van Hal *et al.* (2007) studied amine and bifunctional compounds as catalysts for the catalytic hydration of EO to MEG, with selectivity for its production by employing reaction mechanisms catalyzed by acids and bases (van Hal *et al.* 2007b). The selectivity to MEG using amines as catalysts was: 92%, 90%, and 93% using ethylenediamine (EDA), diethylamine (DEA), and hexamethyleneimine (HMA), respectively, whereas 86% was possible using bifunctional compounds such as ethylene diamine tetraacetic acid (EDTA) with sodium derivatives (van Hal *et al.* 2007b).

From Biomass to 2,5-Furandicarboxylic Acid (FDCA)

FDCA is very stable, has a high melting point (342 ºC), and is insoluble in most solvents. It can be obtained through chemical, biological, and electrochemical conversion, preferably using heterogeneous catalytic systems. The reaction media strongly affects the mechanism. FDCA is usually obtained from sugars such as fructose (FRU), since biomass is more recalcitrant. After a partial delignification of LCBW, cellulose and hemicelluloses from the biomass complex structure are hydrolyzed to C6 sugars. Hemicellulosic C5 sugars can also produce a furanic molecule (furfural), but the use of this pathway achieves low FDCA yields, together with the generation of byproducts. Glucose (GLU) derived from cellulose must be isomerized to FRU to achieve higher FDCA yields (Zhang and Peng 2017; Deshan *et al.* 2020). After that, FRU is dehydrated to reach 5-hydroxymethyl furfural (HMF). The yields of HMF production can be calculated using Eq. 6.

$$HMF\ yield\ (\%) = \frac{moles\ of\ produced\ HMF}{initial\ moles\ of\ fructose\ or\ glucose} * 100 \quad (6)$$

The oxidation of the HMF alcohol or aldehyde produces DFF (2,5-diformylfuran) and HMFCA (hydroxymethylfurancarboxylic acid), respectively, and continues with the latter intermediates to FFCA (5-formylfurancarboxylic acid), finally being converted into FDCA (Boldyreva *et al.* 2019; Deshan *et al.* 2020b). The conversion yield can be lowered by the generation of products of polymerization/ degradation called humins. However, they can be adsorbed by activated carbon and subsequently transformed into more activated carbon when regenerating by burning with O_2, with the additional benefit of avoiding catalyst clogging. Additional energy consumption can be met by the energy integration of the processes (Kim *et al.* 2020a). Hydrophilic medium with acid sites favors the formation of HMF from carbohydrate dehydration, whereas hydrophobic medium with metal sites favors FDCA from HMF oxidation, as can be seen in Fig. 5 (Zhang and Peng 2017b; Deshan *et al.* 2020b).

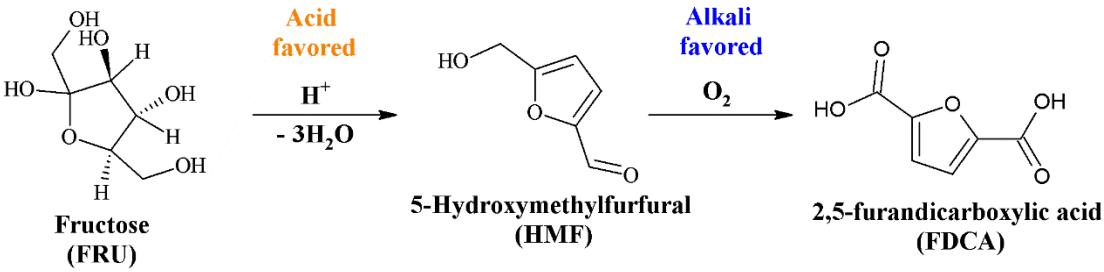

Fig. 5. Scheme of the reaction mechanism for obtaining FDCA from fructose. Adapted from (Zhang and Peng 2017a)

Multiple oxidants (O_2, H_2O_2, t-BuOOH, among others) can be used to achieve FDCA. O_2 is the most available and inexpensive but it needs high pressures to ensure the diffusion in the reaction media (Deshan *et al*. 2020b). Figure 6 shows the process to obtain FDCA from LCBW, and Table 1 presents the results of productions reported in the literature.

Table 1. Previous Results of FDCA Production

Precursor	Experiment Conditions	Results	Reference
Sugars	Separated heterogeneous catalytic systems + continuous removal of water	Up to 85% FDCA yield	Deshan *et al.* 2020
FRU	Two-phase environment (water+organic)	< 70 % FDCA	Klushin *et al.* 2016
FRU	FRU→HMF: 15 wt% loading, dehydration in gamma-valerolatone (GVL)/water. Result: 70% HMF yield HMF→FDCA: 7.5 wt% loading, GVL/water (5:5 mass ratio) using Pt/C (heterogeneous catalysis), 110ºC, 40 atm. Result: 93% FDCA yield	65% FDCA	Motagamwala *et al.* 2018
Cellulose	Cellulose→GLU: 4.8 wt% solid loading, GVL/water (4:1) + 5mM H_2SO_4, 0.5h, 157-217 ºC. Result: 71% GLU yield. GLU→HMF: 3 wt% loading, GVL/water (4:1) + 0.2M HCl + 0.1 M NaCl, 1h, 140 ºC, 20 atm. Result: 62% HMF yield. HMF→FDCA: 7.5 wt% loading, GVL/water (5:5 mass ratio) using Pt/C (heterogeneous catalysis), 110ºC, 40 atm. Result: 93% FDCA yield. + heat integration, currents separation, and crystallization purification of FDCA	>41% FDCA (theoretically)	Kim *et al.* 2020
Jerusalem artichoke rhizomes	1º step: Na_2SO_4 10 H_2O – methyl isobutyl ketone treatment, 2 h, 85°C. Result 40% HMF 2º step: aqueous NaOH/$KMnO_4$ followed by separation. Result: 89% FDCA yield after separation	35% FDCA yield	Boldyreva *et al.* 2019

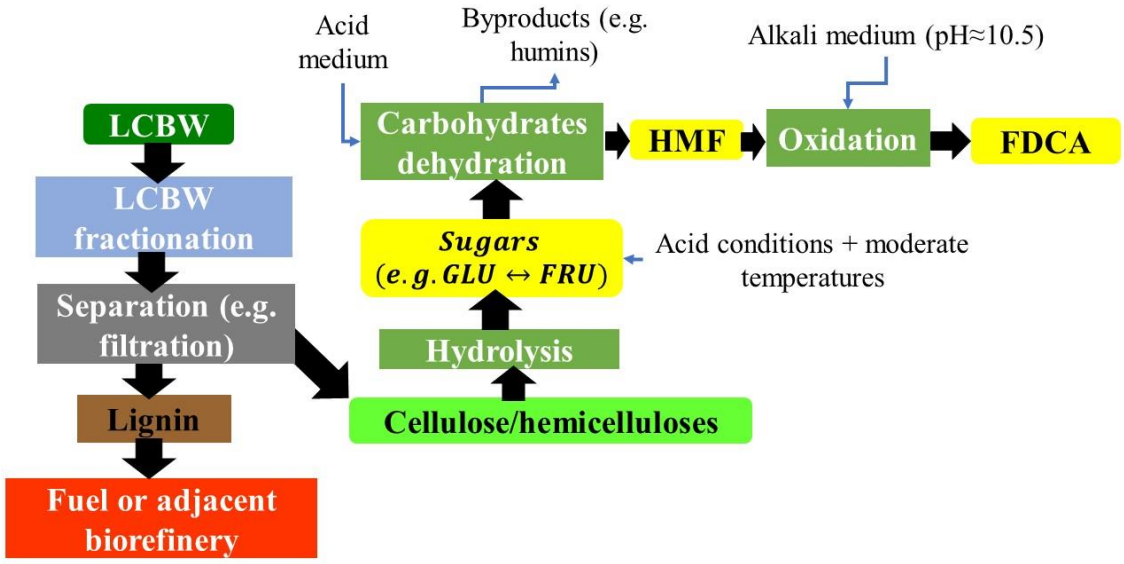

Fig. 6. Route for 2,5-furandicarboxylic acid (FDCA) production from lignocellulosic biomass waste (LCBW)

Kim *et al.* (2020) modeled a process to obtain FDCA from cellulose with heat integration, streams separation, and purification by crystallization of FDCA, improving its yield by 17%. Boldyreva *et al.* (2019) highlighted the importance of maintaining the pH at 10.5 in their process because of the formation of intermediates at lower values and the excessive consumption of reagents at higher values.

TECHNO-ECONOMIC AND ENVIRONMENTAL CONSIDERATIONS FOR BIO-PEF PRODUCTION

To be competitive with fossil economies and make the costs viable on a pilot scale, the biorefinery platform for the production of PEF requires the optimization of the process variables for obtaining Bio-MEG and FDCA (Hwang *et al.* 2020c; Yang *et al.* 2021). Bio-MEG studies involved technical and economic viability, competitiveness, environmental benefits, and comparison with MEG from coal and oil. The process design and modeling used in the techno-economic analysis indicated that the total production cost of Bio-MEG is 20% higher than the total cost of production of carbon-based MEG and 43.3% higher than the cost of producing petroleum-based MEG (Yang *et al.* 2019; Zhao *et al.* 2021a).
Energy integration strategies of the whole process (heat, energy, and mass), are necessary (Clauser *et al.* 2021a) to achieve economic benefits in the Bio-MEG production. In process integration (PI), energy integration is a strategy applied in chemical plants to promote economics and sustainability through heat recovery and efficient use of energy, water, and other resources. The following methodologies are, for example, Heat Exchanger Networks (HENs) and pinch analysis (optimization methodologies). Besides, Mass Integration (MI) is a method for reducing water consumption, inputs, wastes, and other streams which can be recovered, improving the process profitability and environmental impacts (Klemeš 2013;

Linnhoff and Hindmarsh 1983). Savings for heat and cooling demand of 31.5% and 39.5%, respectively, are estimated (Becerra *et al.* 2017; Nitzsche *et al.* 2016). In the stages where separation methods such as distillation or separation by chemical reactions are required, the implemented technologies represent more than 70% of the energy consumed in the chemical process (Kumar *et al.* 2020; Parvatker and Eckelman 2020), which directly influences the costs of bio-ethanol for bio-ethylene production through the biochemical route (Hackl *et al.* 2015a).

In a bio-ethylene refinery, the heating services for both ethanol and ethylene can be reduced from 131 MW to less than 80 MW approximately if the flue gas is integrated with the ethanol dehydration reactors (Arvidsson 2011). An integration alternative widely used involves a systematic approach to take advantage of all the heat content in the different stages and optimize it (Hackl *et al.* 2015b; Valderrama *et al.* 2020).

The generation of GHG is a big concern to consider in the production of bioplastics. Nowadays, only a fraction of plastics is recycled. In addition, the current amount of recycled material in plastics commodities do not reach 30%. The remainder is incinerated or landfilled, and the majority gets disposed of in an uncontrolled environment. The production of Bio-MEG derived from LCBW allows reducing greenhouse gas (GHG) emissions compared to production from coal and oil in 51% and 69.5%, respectively (Zhao *et al.* 2021b).

The challenge of FDCA production lies in the generation of humins, isomerization of co-products of glucose, and dehydration of fructose. Separating them by adsorbents such as activated carbon, the resulting stream can reach a high concentration of HMF (Motagamwala *et al.* 2018). Applying purification methods such as heat integration, streams separation, and crystallization can increase FDCA productivity by up to 17%, reducing capital investment by 52.6%. A minimum selling price (MSP, minimum selling price) of $1,366/t is reached for FDCA, while the oil-based TPA is $ 1,445/t. The sensitivity analysis shows that FDA production's utmost cost drivers are the raw material, steam price, and discount rate (Kim *et al.* 2020b). Another study considered that the FDCA's main price drivers were feedstock price and scale economy. In 2012, De Jong postulated that by means of producing TPA at 50 Mt/y at 1100 €/t, at a scale greater than 300 kt/y, the price of FDCA could be lower than 1000 €/t (De Jong *et al.* 2012).

Currently, the production of non-biodegradable bioplastics derived from renewable materials represents 57% of the market, with Bio-PET in the first place, followed by bio-based polyamides and bio-PE (Andreeßen and Steinbüchel 2019b; Mendieta *et al.* 2019). This alternative for the production of bioplastics (bio-PET) contains in its structure the highest proportion of petrochemical raw material (80%) and the remaining 20% corresponding to biomass, terephthalic acid (TPA) based on petroleum, and bio-MEG from renewable sources (Hwang *et al.* 2020b). Many researchers have obtained bio-PET by substituting fossil-based terephthalic acid with a biological-based one, achieving reductions in the range of 25 to 58% in greenhouse gas (GHG) emissions, depending on the renewable material implemented to produce bio-TPA (Semba *et al.* 2018). However, the alternative of a bio-PEF from 100% LCBW is of interest due to its already mentioned characteristics (Eerhart *et al.* 2012).

The PEF production using fossil-based MEG could lower the non-renewable energy usage (NREU) by 40 to 50% and greenhouse gas (GHG) emissions by 45 to 55% in a cradle to the grave system compared with PET, which are lower values than comparing PET with

other bio-based plastics, such as polylactic acid or polyethylene. Percentages could be even higher if Bio-MEG is used (De Jong *et al.* 2012).

As the variety of bioplastics is very wide (Di Bartolo *et al.* 2021), its life cycle depends on the type of material for which it was created (Walker and Rothman 2020), and its degradability significantly influences the chemical and physical structure it presents (Strapasson *et al.* 2005; Gautam *et al.* 2007; Pathak and Navneet 2017). The degradation process for non-biodegradable bioplastics depends on several factors, such as UV radiation, temperature, humidity, pH, among others, whereas for degradable bioplastics, the microbial activity is also significant (Kjeldsen *et al.* 2019; Ruggero *et al.* 2019; Folino *et al.* 2020; Meereboer *et al.* 2020). Degradation of polymeric materials is usually studied by the photo-oxidative process, exposing them to ultraviolet radiation in the presence of oxygen *(Qin et al.* 2003). This process is widely applied to study the chemical degradation of polyethylene because it is a highly used polymer in the world (Trozzolo and Winslow 1968; Torikai *et al.* 1990).

Recycling plastic waste is an alternative approach to reducing the volume of urban solid waste (MSW) (Arutchelvi *et al.* 2008; Ghatge *et al.* 2020; Montazer *et al.* 2020), generating savings both in the use of fossil fuels and the energy required in its production (Shent *et al.* 1999), and there are different ways to do it. For example, the thermo-mechanical process has existed since the 1970s to process plastic waste and produce new materials with similar characteristics and properties (Vu *et al.* 2020). It is the most applied process on thermoplastic materials, using equipment such as screw extrusion, injection, and blow molding, among others (Grigore 2017; Lamberti *et al.* 2020). Plastic is a material prone to stress, fractures, and defects, among others, which drastically decrease its life cycle. It has first to be sorted to reduce contamination of particles and compatibility issues, which lead to problems in the structure. It is a challenge, as there are a variety of plastics with different properties, but processes such as Near Infrared (NIR) sorting, float/sink operation, or melt filtration can facilitate this task (Schyns and Shaver 2021). Afterward, the material is thoroughly washed and granulated. The final application of the recycled material has to be known to homogenize the properties and color. Finally, the pellets are extruded to get the plastic pellets commercialized as raw material for the plastic industries. Efficient collection systems can allow PET recycling (Vollmer *et al.* 2020).

On the other hand, chemical recycling aims to depolymerize or degrade plastic waste into other usable materials or smaller units (monomers), which can also be raw materials for new products (Payne *et al.* 2019; Xia *et al.* 2021; Zhu *et al.* 2016). Breaking the chemical bonds requires energy, which is higher in polyolefins (about 400 °C) and lower in functionalized polymers such as PET (about 100 °C).

The main benefits of chemical recycling compared to mechanical recycling are lower process costs, which is typically due to decreased energy consumption. However, a higher polymer degradation implies higher overall costs (De Castro *et al.* 2006; Liu *et al.* 2018; Shogren *et al.* 2019). Nevertheless, there is a lack of commercial processes that meet the demand of recycling necessity, which may be due to the limitations of each recovery mechanism. Their combined use could solve this disadvantage. Chemical recycling involves solvolysis, dissolution/precipitation, or pyrolysis. Depending on the type of polymer to be treated, they present advantages and disadvantages (Hopewell *et al.* 2009; Niaounakis 2013).

Dissolution/precipitation involves the dissolution of polymers in solvents (or combination of solvent and antisolvent), followed by the filtration of impurities and finishing with the desired polymer precipitation. It is considered chemical recycling

because of the chemistry involved in the solvent choice, even if bonds cleavage is not evidenced (Vollmer *et al.* 2020). It implies a higher CO_2 emission avoidance (65 to 75 wt% of plastic). Besides, additives can be removed by filtration to obtain higher purity. Bio-based solvents are suggested to increase the process sustainability, highlighting γ-valerolactone for PET (Walker *et al.* 2020; Chen *et al.* 2021). In addition, the separation of the solvent from the polymer remains a challenge, since its presence in plastic is considered impurities and can affect the polymer's properties. Solvolysis allows the monomer recovery by breaking the polymer chains through a reaction of the functional groups of the backbone with a solvent or solvent system. It could be an alternative for PEF (a polyester) recycling, since it only applies to pure mixtures of polyamides and polyesters (Vollmer *et al.* 2020).

Pyrolysis is suitable for polyolefins, polystyrene (PS), and polymethyl methacrylate (PMMA). Their contamination with other plastics and non-plastics results in operational problems. Moreover, it is considered the least preferred alternative, since it produces different multi-product phases, which segregation increases the overall cost. However, catalysts development with energy harness in an integral biorefinery could be interesting to obtain high yields of monomers. Another recycling alternative is biological recycling or plasma-assisted pyrolysis, but these and other alternatives remain on a laboratory scale (Vollmer *et al.* 2020; Schwarz *et al.* 2021).

The recycling processes can be divided into four categories hierarchically ordered by the recovery potential of the exploited plastic in a closed-loop. They are primary or where the recycled material has the same properties as the original, such as mechanical recycling; secondary, or open-loop, in which the product has lower quality compared with the original; tertiary, including plastic to feedstock and plastic to monomer; and quaternary or incineration with energy recovery. The optimal recycling process depends on the plastic or polymer.

A better way to decrease waste is to reuse it. However, it is not always possible due to deterioration or hygiene reasons. So polymers must be mechanically recycled until the quality of the material become poor. Bio-PET and bio-PE maintain their mechanical properties during a few cycles. Their monomers are recovered through a chemical route and can be re-polymerized, contributing to the circular economy. Challenges include the need for better plastic waste collection schemes and improving chemical recycling infrastructures to reduce the production costs of new biopolymers and improve their mechanical capacity (Lamberti *et al.* 2020).

(Schwarz *et al.* 2021) performed a life cycle analysis (LCA) of the 15 most demanded polymers in Europe, concluding that recycling can reduce up to 73% CO_2 of plastic processing emissions (or 200 MT equivalent).

CONCLUDING STATEMENTS

Polyethylene furanoate (PEF) production from biomass is promising but needs further research. This article reviews bio-based ethylene glycol (Bio-MEG) and 2,5-furandicarboxylic acid (FDCA) as monomers for the subsequent production of PEF. Based on the exposed, the aspects needing research are:

- Bio-MEG and FDCA production from lignocellulosic biomass waste (LCBW) instead of commercial sugars from food resources or fossil-based chemicals.
- Multi-functional catalysts selection for the post-fractionation reactions.

- Organic solvent selection for the oxidation of ethylene. Gamma-valerolactone (GVL) has proved to solubilize FDCA, so it could be used for ethylene and 5-hydroxymethylfurfural (HMF) oxidation, decreasing the capital costs of the process.
- Biodegradation essays to determine PEF biodegradability and microplastics generation.

Nevertheless, the best alternative for PEF and PET is to decrease the accumulation of both materials.

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