



Bioplastics from Winemaking By-products in the Buildings Sector: A Feasibility Study on the Main Opportunities, Barriers and Challenges

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Abstract

Plastics from fossil source are third after steel and cement among the most widespread materials used in the buildings sector. Bioplastics are biopolymers that offer a sustainable alternative due to their biodegradability and compostability. The edible first-generation sugary-based feedstocks, having high costs that drive the market price even in presence of a large-scale production of bioplastics, should be partly replaced by 2030 with non-edible second-generation feedstocks based on recyclable organic solid agro-wastes according to “Green Deal” of the European Union. The winemaking wastes used as feedstock for the synthesis of biopolymer building blocks and reinforcing fillers could represent a suitable option to reduce biopolymer costs and increase their competitiveness in plastic market. Although bioplastic can solve more environmental issues, nonetheless the production cycle does not always respect the principles of sustainability overall during biopolymer recovery. The present feasibility study is aimed at taking the state of the art of bioplastics in the buildings industry for promoting winemaking co-products into a circular system. The literature data have been collected, consulted and empirically elaborated to find real and potential opportunities, barriers and challenges of developing wine wastes (e.g. wine shoots, grape pomace and wine lees) in the strategic market segment of bioclimatic architecture.

Keywords Agri-based circular economy · Bioplastic · Bioclimatic buildings design · Sustainability · Wine waste

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Introduction

Since the 1950s, about 8.3 billion tonnes (Bt) of plastics (only half in the last 13 years) from fossil source have turned into waste [1], where only 9% is recycled and 12% is incinerated with dangerous effects due to emission of toxic substances and global warming while the remaining 79% ended up in landfills or dispersed in the environment [2]. Plastic disposal too often is not carried out legally, being estimated that about 10 rivers in the world carry 90% of the plastic transported at sea; that correspond to 8 million tonnes per year (t yr^{-1}) [3]. If this trend will not be stopped or reversed in the next years, 12 Bt yr^{-1} of non-biodegradable plastic will be wasted by 2050 [4]. In particular, the packaging industry takes into account 98% of crude oil, but it recycles only 5% of plastic [5]. It is estimated that around 270 thousand tonnes of plastic-based wastes are floating in the oceans' surfaces and rivers [6, 7], and micro-plastics have reach the human food chains [8, 9] due to plastic ingestion and inhalation by marine animals [10, 11].

The current system of plastics production is mainly linear, being based on fossil source with low level of recycling [12]. For these reasons, it is necessary to use plastic material that meets the biodegradability requirement of the European Union (EU). These specifications are useful to define how and what bioplastic should be biodegraded into soil and water in the shortest period of time to be defined as a *biodegradable and compostable* material according to the bag's label defined by EN 13432. There is greater demand in bioplastics than the past supported by the increased availability of new feedstocks from the most varied agricultural sectors [13], as well as by the biofuel supply chains [14] and agri-food industry [15]. Utilization of organic solid waste for bioplastics production plays a key role in the transition from a linear economy model into an agri-based circular economy system [16]. Bio-based building blocks and reinforcing fillers are mainly obtained from edible first-generation sugary-based feedstocks that are economically unviable, being still too food competitive. The circular economy action plan (CEAP) of the EU is addressed to stimulating the transition towards a circular economy that will increase economic competitiveness, sustainable growth and new jobs [17]. The European bioplastics industry must be included in this transition process as major source of economic growth and jobs, since the European Commission (EC) has adopted new strategies on sustainable plastics management as a part of the CEAP [18].

The global market of bioplastic is already growing by about 20 to 100% per year, being expected that the EU will possess by 2021 around a quarter of the world's bioplastics production capacity [18]. The market share of the bioplastics industry employed 23,000 people in 2013, and some forecasts estimated that this number can grow up to 300,000 jobs by 2030 with the right incentives in Europe. However, the price of bioplastic is still too higher than conventional fossil-based plastics such as poly(ethyleneterephthalate) (PET) that still remains the most competitive in California (USA) [19]. For this reason, the production of bioplastics was only 2.1 million tonnes (Mt) (1.2 Mt of bio-based polymers and 0.9 Mt of biodegradable polymers), which represents only 0.6% of the total produced polymers [20, 21]. Nonetheless, the bioplastics market targets a number of industrial segments from food packaging and gastronomy [22] to consumer electronics, automotive and aircrafts [14, 15]. In these market segments, bioplastic is used to manufacture products for short-term use such as food catering products, as well as for durable applications such as mobile phone cover or interior components for cars. Bioplastic is much used to producing mulching film for covering greenhouses, tunnels and agricultural lands [14].

In order to make industrial processes for the bioplastics production that meet the principles of sustainability and bioeconomy, new targets will be imposed by the EC overall regarding the need of using inexpensive agro-based organic waste in place of expensive sugary-based substrate from food/energy crops (wheat, corn, rapeseed, sugar cane, etc.) [20, 21]. Despite the increased demand for biopolymers, the global market share of bio-composite in the buildings sector, bioclimatic architecture and residential construction is still too limited due to high manufacturing costs greatly associated to the materials used [23].

The purpose of this study is to focus feasibility for users and stakeholders to use inexpensive raw materials based on non-edible second-generation feedstocks to make bio-based materials in bioclimatic architecture. The paper has widely addressed its attention on potential use of winemaking by-products for the synthesis of biopolymer for building blocks and reinforcing fillers basing on the featured physicochemical characteristics of feedstock. Afterwards, it has considered an excellent case study of bioclimatic architecture whenever the potential use of winemaking by-products to make eco-friendly bio-composites is presented and discussed. Besides, some alternatives were explored in order to solve compatibility issues between wine wastes and other agro-wastes for making bio-composite materials and what alternatives could be used in place of hazardous solvents in biopolymer recovery which are nowadays less emphasized than in the past by regulatory bodies for biopolymer recovery.

Novelty Statement

Despite a very comprehensive handbook on grape processing by-products [24] and more else exhaustive review works dealing with the new trends of valorization of wine waste in bioeconomy were found [25–29], there are very few papers addressed on the feasibility study of using winemaking by-products as feedstock for the plastic industry in the buildings sector.

This study is focused on three aspects: the subject under consideration, the motivation why the present study is performed and the detailed scope of the study. Although the aforementioned literature outlines the bioplastic market as a pillar for the green economy, nevertheless biopolymers were still produced at lower amount and do not represent always a suitable option, being still too expensive and food competitive. Nonetheless, the current state of the art of using bioplastics from cheaper raw materials seemed to be insufficiently investigated in the buildings industry despite this market segment covers a strategic sector that needs increasing amount of eco-friendly bio-composite material at lower cost. On the other hand, the use of winemaking co-products could represent a profitable option to reducing the production costs and to increasing the amount of building blocks and reinforcing fillers [30]. Therefore, it will be economically beneficial if the final price of biopolymers is taken into account in the life cycle cost analyses for the buildings sector. By this way, the price would be decreased almost proportionally because bio-composites would have more attractiveness due to the enhanced technological properties [31–33].

The novelty of the work is established through a critical analysis of related issues showing that most of the European's projects are still addressed to sustainable biomaterials from integrated biorefinery waste streams based on starchy derivatives of maize, sugar beet and sugarcane rather than on winemaking by-products. Literature reports overlook the buildings industry sector, although this sector employs about 1.5 million people in Europe accounting revenue of more than 355 billion euro per year [34]. The motivation to perform this work is based on the assumption that the global market for bioplastics is predicted to grow

continuously over the next decades. The production capacities of bioplastics are predicted to increase from around 2.11 Mt in 2019 to approximately 2.42 Mt by 2024 according to market sharing given by the European Bioplastics in collaboration with the nova-Institute. Moreover, requirements of eco-friendly biomaterials at low cost in bioclimatic architecture are increased in accordance with the guidelines, recommendations and economic incentives (white/green certificates) to improving the programs of energy efficiency and/or energy saving in the residential buildings sector [35]. The question that the paper tried to answer is based on the assumption that the portfolio of application resulted to be diversified in the EU because the market share of buildings sector and bioclimatic architecture is increased. In fact, bioplastic is potentially usable in the buildings structures and structural design more than in the past since it represents the second manufacturing sector in demanding for plastic material, only preceded by food packaging [36]. The scientific hypothesis that justifies the present study is based on assumption of using solvents less toxic for human and environment in biopolymer recovery for making bioplastic more safe and economically competitive, while maintaining high material purity and industrial productivity of refined bio-composite [37, 38].

Methodology

The methodology used to perform this study is entirely based on an empirical approach by consulting the updated references from several databases to drawing a feasibility study focused on potential use of biopolymers from winemaking by-products in the buildings sector.

In order to identify and prioritize the literature consulted on which this review is based, the most appropriate references were selected within the EU framework since the “Green Deal” was a challenge for the “Italian National Agency for New Technologies, Energy and Sustainable Economic Development” (ENEA). Overall, I focused the present literature data basing on the most recent scientific findings (up to 2021) by searching it with the common databases by inputting the following keywords: “agri-based circular economy”, “biopolymer”, “bioclimatic buildings design”, “bio-composite”, “sustainability” and “winemaking waste”. I selected the most relevant papers and books overall in English language into Scopus database with high scholar rating focusing on the circularity principles of bio-based economy. On the other hand, I searched the main European projects from websites of Google Scholar and IEA (International Energy Agency) – Bioenergy Task 42 – while technical reports, deliverables and regulatory bodies were given from the EU and Bioplastics websites. A strong help was done by the Research Gate platform and direct contacts with the corresponding authors of the most relevant literature. At the end of searching works, a total of 62 references were selected and customized for organizing and structuring the paper in order to give a feasibility study for users and stakeholders that could be interested to converting winery waste into bio-composite for buildings sector.

Results and Discussion

Concepts, Definitions, Legislation and Directives

Like plastics from fossil source, bioplastics are polymeric chains to confer particular requirements. However, it is quite usual to use the terms *plastic* and *polymer* as synonyms. A polymer

is defined as “a molecule of high relative molecular mass, the structure of which essentially comprises the multiple repetitions of units derived, actually or conceptually, from molecules of low relative molecular mass” [39]. The term *biopolymer* identifies a group of materials with the proper properties to be fully biodegraded in the environment, being originated from agri-based biomass waste. Instead, the term *biomass* is defined by the European Directive 2009/28/EC as “The biodegradable fraction of products, waste and residues of biological origin from agriculture activity including vegetal and animal substances, forestry and related industrial products, fisheries and aquaculture, as well as the biodegradable fraction of industrial and municipal waste”. Such definitions have been received by the Italian legislation (Italian Legislative Decree No. 387 of 2003). The term *biodegradable* indicates “a material that, thanks to its chemical structure, is able to undergo a process in which microorganisms such as bacteria and fungi metabolize the polymer chains into water, carbon dioxide (or methane), and biomass under aerobic (or anaerobic) conditions” as defined by EN 13423 (*industrial compostability*), EN 17033 (*degradation on soil under weather conditions*) and ASTM D6691 (*biodegradability in marine environment*). **Bio-based labels** should be conformed to the EU standards EN 16640 and EN 16785. These standards specify the calculation method that should be adopted for determining the bio-based (carbon) content in monomers, polymers, plastic materials and products based on the C_{14} content measurement or C_{14} method and elemental analysis, respectively. **Compostability labels** should be certified according to the standards EN 13432 and EN 14995 to be called a material as *compostable*. The Seedlings’ logo is a reliable label for industrial compostability which addresses the decision on purchasing and disposing a product or packaging. Biodegradability does not necessarily include a condition of biocompatibility or environmental sustainability [40]. The regulatory bodies measured biodegradability through the quantity of carbon dioxide (CO_2) produced at the fixed time (EN 14046, ISO 14855) whose 90% level of CO_2 should be considered as an acceptable limit for the biodegradable mass during a period of 180 days (EN 13432). The attitudes of biodegradability and compostability of bioplastic widely vary with temperature, stability and oxygen content of the polymeric chains [15]. Some bioplastics are both biodegradable and compostable and considered as a viable option to produce high-quality compost in a real farming system [41].

Depending on the featured physicochemical properties, biodegradability ratio, microorganisms and feedstocks used, bioplastics can be classified in the following categories [30]: biodegradable polymers from fermentable biomass such as poly(L-lactic acid) (PLA), poly(hydroxyalkanoates) (PHAs) and poly(3-hydroxybutyrate) (PHB); biodegradable polymers of petrochemical origin such as poly(butylene adipate/terephthalate) (PBAT), poly(butylene succinate) (PBS), poly(butylene succinate adipate) (PBSA), poly(methylene adipate/terephthalate) (PTMAT) and poly(ϵ -caprolactone) (PCL); and non-biodegradable biopolymers of petrochemical origin such as poly(ethylene) (PE). The most recent studies were addressed towards biopolymers produced by microbial fermentation using growth substrates from plant waste, forestry residues, food waste, agro-industrial waste and biofuel chain co-products derived from a circular economy system overall based on the biorefinery approaches [14]. Such biomass contains nutrients to allow growth and multiplication of microbial cells (overall bacteria, but also yeasts are nowadays most studied). PHA and PHB make up the main stocks of nutrients for bacteria and yeasts under unfavourable growth conditions [42] that are synthesized from about 300 bacteria species belonging to 90 genera [13]. Accumulation of metabolites for cell survival along the time, including high value-added lipids, occurs when microbes are deprived of some macro-micronutrients with the presence of abundant carbon

sources in fermentation substrate. Afterwards, some reserve molecules such as polyphosphates, lipids and overall PHAs were accumulated in the cells whenever PHA takes the form of white granules (from 0.2- to 0.7- μm diameter) [43]. Production of PHAs from inexpensive second-generation feedstock is accepted as a viable strategy that does not compete with the food sector, although the scaling up to industrial scale remains still a challenge. The main characteristics of PHAs are thermo-plasticity, biodegradability, biocompatibility, independence from fossil sources, fragility and elasticity, molecular design ability and poor permeability to gases and vapours. Biodegradability is linked to the presence of enzymes in the environment (overall soil and water), but it does not mean that the process is always triggered and how long it takes. Viscosity must be low sufficiently to allow the removal from the finished product, while boiling temperature of the process must allow to achieve and maintain a proper viscosity without excessive energy requirement when applied up to industrial scale. Another key factor is due to conversion ratio in fermenter between the amounts of substrates added and PHAs produced. For instance, if 100-g PHAs are theoretically produced in 50 h from a litre of substrate ($2 \text{ g l}^{-1} \text{ h}^{-1}$), a fermenter of 20 m^3 capacity will be needed to produce 10 t of PHAs in 250 h. But, if the real productivity is reduced to 50%, a fermenter of 40 m^3 (un-published data) will be required. Some studies demonstrated that an increase in productivity of 60% (from 1.98 to $3.2 \text{ g l}^{-1} \text{ h}^{-1}$) leads to a decrease in costs up to 10% [44]. Further studies encourage players and stakeholders of making bioplastics from inexpensive sources with high recycling potential to reducing the total processing costs up to 30% [45].

Potentiality of Winery By-products for Bioplastics

Table 1 summarizes some commodities and their main application fields coming from integrated biorefineries to producing bioplastics using diversified feedstocks of plant waste, food waste, agro-industrial waste, biofuel co-products and unavoidable agri-based wastes where fermentation substrates are currently used to produce bioplastics thanks to biological action of selected bacteria. Particularly, winery industry is one of the agri-food sectors that produce a waste surplus, although legislation tends to define it with the term *co/by-product* for allowing the reuse of such biomass waste in new productive cycles rather than be wasted. Wine global production was 29.2 million hectolitres (Mhl) in 2018, where Europe was the leading producer (57% of the total production) followed by the Americas (26%), Asia (7.5%), Australia (5.4%) and Africa (4.5%). The top five wine producers include Italy (18.8% of the total production) followed by France (16.6%), Spain (15.2%), USA (8.2%) and Argentina (5%) [46, 47]. In Italy, the annual production of wine grapes amounts to an average of 53 million q corresponding to 45 Mhl of wine [46, 47].

Authors estimated that the winemaking supply chains generate approximately 0.3–0.5 kg of wine by-products per litre of wine, but should be taken into account the by-products generated during the pruning seasons [47, 48]. Figure 1 shows the winemaking flowcharts of both red and white wine with associated solid wastes that are currently generated. The main co-products from wine industry can be classified in the following categories [30]: (a) wine shoots, the non-wood lignocellulosic agricultural residues discarded during vine pruning; (b) grape stalks (or stems, rasps), the woody-herbaceous structure of the bunch of grapes that can constitute between 1.4 and 7% by weight of the fruit [49]; (c) grape pomace and grape peels, the solid residues from pressing that are homogeneous mass whose appearance, consistency and chemical composition depend on the agronomic practices of wine grape (climate conditions, soil features, cultivar, pruning technique, ripening stage, etc.) and winemaking technique

Table 1 Commodities and their main application fields from plant waste, forestry residues, food waste, agro-industrial waste and biofuel co-products from a circular economy system based on biorefinery approaches (source: [13, 14])

Feedstock	Raw material	Processing	By-product	Commodity and main application field ^(*)
<ul style="list-style-type: none"> - Processing residues from sugary industry: molasses, beet-top, beet-pulp, etc. - Sugary co-products from the bio-ethanol chain: sugar beet bagasse and pulp, cropping residue (roots, tubers, rhizomes, bulbs, etc.) - Co-products and residues from sugarcane and cassava bagasse, starchy materials from potato, tapioca, corn stover, wheat straw, wheat bran, lignocellulose/hemicellulose hydrolysates, Jerusalem artichoke, beet molasses, rye flour, sweet sorghum, carrot processing waste and molasses spent wash - Unmarketable sugary fruit and vegetable: potatoes, apples, peaches, strawberries, carrots, beets, etc. - Vegetable processing leftovers - Crop waste: potatoes, corn and sugarcane stalk 	Starch, glucose, fructose, sucrose	Fermentation by bacteria	<p>Succinic acid</p> <p>Lactic acid Poly(L-lactic acid) (PLA)</p> <p>Acetic acid</p>	<p><i>Bioplastics</i>, food additive, surfactants, detergents, solvents, textiles, pharmaceutical drugs</p> <p>- <i>Food packaging and beverages</i>, <i>polymers, textiles</i></p> <p>- Biomedical applications in the fields of suture, bone fixation material, drug delivery and tissue engineering</p> <p>- <i>Production of poly(ethylene terephthalate) (PET) for food packaging, textiles and films</i></p> <p>- Production of food additive, industrial chemicals</p> <p>- Production of ethyl acetate as solvent, glues, fibres</p> <p><i>Biodegradable plastics for films and packaging, fibres, coatings, foams, medical and pharmaceutical applications</i></p> <p>- <i>Bioplastics for food packaging and biodegradable films</i></p> <p>- <i>Bioplastics for structural composites in bioclimatic buildings</i></p>
<ul style="list-style-type: none"> - Lignocellulosic biomass from forestry residues, wood pulp and wood waste (wood chips, wood shavings, aspen chips, olive brush residues, etc.) - Crop residues (branches, foliage, etc.); crop waste (corn stalks, rice husks, cobs, wheat straw, etc.) 	Cellulose, hemicellulose	Chemical-mechanical treatment, electro-spinning	<p>Poly(hydroxyalkanoates) (PHAs)</p> <p>Poly(3-hydroxybutyrate) (PHB)</p> <p>Cellulose nanofibres</p>	<p><i>Biodegradable plastics for films and packaging, fibres, coatings, foams, medical and pharmaceutical applications</i></p> <p>- <i>Bioplastics for food packaging and biodegradable films</i></p> <p>- <i>Bioplastics for structural composites in bioclimatic buildings</i></p>
<ul style="list-style-type: none"> - Oleaginous co-products from the biodiesel chain: oil-less seed cakes/meals/pellets of edible oilseed of oleaginous crops (rape, sunflower, soybean and palm oil) and non-edible oilseed of <i>Brassica carinata</i>, <i>Jatropha</i>, Indian mustard, <i>Crambe abyssinica</i>, camelina and canola 	Protein, amino acid, carbon	Chopping, drying, cubing, pelletizing	<p>High-value proteins, essential amino acids and pure carbon</p>	<p>- Livestock feed for ruminants (cattle) and monogastric species (poultry and pigs)</p> <p>- <i>Bio-composite material polymeric and thermoplastic from soybean meal</i></p>

Table 1 (continued)

Feedstock	Raw material	Processing	By-product	Commodity and main application field ^(*)
Crude glycerine from the biodiesel chain of edible and non-edible oleaginous crops	Carbon	Oil trans-esterification, bio-refining	Bio-glycerine	<ul style="list-style-type: none"> - Food, paper, <i>bioplastics</i>, rubber, lubricants, soap, cosmetics, toiletries, surfactants, pharmaceuticals, fertilizers and textiles from palm oil - <i>Bioplastics or food packaging and biodegradable films</i> - Food and beverage industries; personal care and oral products; medical and pharmaceutical application and nitro-glycerine; livestock feed rations for pigs; <i>biomaterial applications including chemicals, monomers, plasticizers</i>; hydrogen generation and polyester production; phytosanitary application (additive, conjugate, synthesis of carbamates, carrier of antibiotics)
- Cooking oil - Exhausted engine oil - Animal fat	Medium- and long-chain of fatty acid units	Refining	Lipids, triglycerides, di-glycerides, mono-glycerides	<i>Biodegradable plastics used in films and packaging, fibres, coatings, foams, medical and pharmaceutical applications</i>
Residues from winery industry (see Table 2 for more details)	Fructose, lipid, polyphosphates, polyphenols, anthocyanins and tartaric acid	Fermentation by bacteria	PHAs Lactic acid	<i>Bioplastics for structural composites in bioclimatic buildings</i>
Spent coffee grounds	Lipid	Fermentation by bacteria	PHAs	<i>Biodegradable plastics used in films and packaging, fibres, coatings, foams, medical and pharmaceutical applications</i>
Olive pomace	Oil	Anaerobic fermentation	PHAs	
Rapeseed waste	Oil	Fermentation by bacteria	PHAs	
Wheat straw hydrolysate	Protein	Fermentation by yeast	PHAs	
Giant reed stem hydrolysate	Protein	Fermentation by yeast	PHAs	

(*) Bioplastics and their application fields were *italicized* in the last column

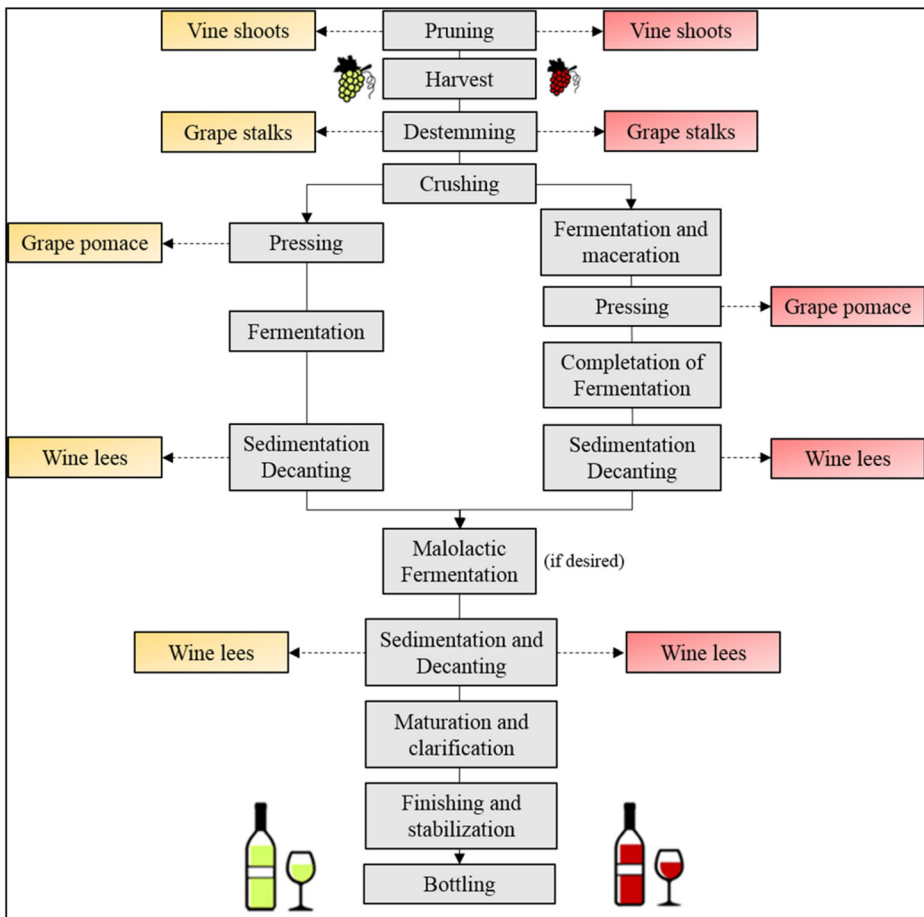


Fig. 1 Flowchart of winemaking process for producing red and white wine with associated solid wine co-products (taken from [30] with the permission of the author)

adopted for those specific geographical area where the wine is produced [50]; (d) grape seeds; and (e) wine lees, where the latter consists of sediments fermented, tartaric acid and died cells [40]. The main solid winemaking by-products are represented by vine shoots, grape stalks, grape pomace (grape peels/grape seeds, about 1:1 weight ratio) and wine lees. Grape pomace represent the most abundant fraction of by-products generated during winemaking processes (20–25%), followed by wine lees (2–6%) and grape stalks (3–5%) [30]. In Italy, the potential amount of by-products can be estimated as 8 million q of grape pomace (corresponding to 15% of the grapes undergo to winemaking) and 2.25 Mhl of wine lees (corresponding to 5% of the wine produced) [51]. Winery wastes generally were further reused in distillation, but more frequently destined to landfilling and incineration, or more simply land-spread. Grape stalks were usually wasted in land-spread (76% in Italy, 55% in Spain, and 40% in France), disposed in landfill (50% in Greece), or incinerated (36% in France). Grape pomace was commonly used in distillery (100% Italy, 90% France, 30–60% Spain) or wasted by land spreading (50% in Spain), while in Greece they are mainly discarded to landfill (67%) or sold as animal fodder (33%). Finally, vine shoots were usually spread on land or burned in the field, while wine lees

were distilled [30]. In Italy, winemaking by-products are overall delivered in distillery (Italian Law No. 82 of 2006). Alternative to distillation should be carefully in the European scenario. For instance, wine wastes should not be used as fertilizer or amendment of soil without pre-treatments and conditioning due to their low pH, high organic matter and high concentrations of macronutrients that can inhibit the germination of seeds [52]. Moreover, landfilling is strongly discouraged since wine wastes affect the soil erosion and decrease the groundwater quality, grape stalks and marc were currently most used for the extraction of antioxidant compounds based on phenolic substances by using solvents [53], grape pomace cannot be used as animal feed due to its high amount of polyphenols that bond with proteins [26, 54], and distillation of wine by-products has been stopped by the regulatory bodies (Reg. No. 1308 of 2013—European Decree in the matter of wine waste disposal rules) [24].

The aforementioned issues lead winery wastes to be carefully reconsidered as inexpensive non-edible second-generation feedstock to be recycled into bioplastic thanks to its own lignocellulosic structure and high carbon content. Despite wine waste cannot still be recycled into bioplastics due to legal limits (Italian Ministerial Decree No. 11/27 of 2008), the most recent literature showed more else potentialities of wine by-products to be recycled into bioplastics. In fact, wine waste can be reused as a raw material in microbial substrate for biopolymer synthesis through the production of ethanol, lactic acid, PHAs and succinic acid [30]. Figure 2 shows the hypothesized pathways for the valorization of solid winemaking by-products as a feedstock to producing bio-based polymers. The reuse of such co-products into biopolymers is allowed by high content of fermentable carbon sources for microbial feeding. Meanwhile, the recycle of wine residues into bioplastics is hindered by antioxidant phenolic-

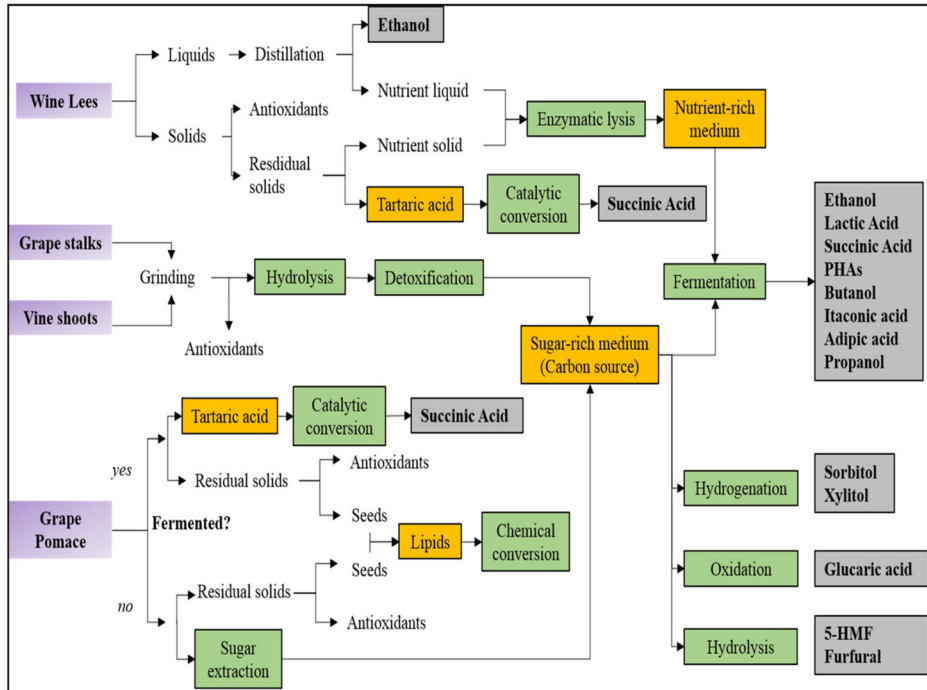


Fig. 2 Hypothesized pathways for the valorization of solid winemaking by-products as a feedstock for the production of bio-based polymers (taken from [30] with the permission of the author)

based substances (polyphenols, flavonoids and anthocyanins) and antimicrobial tannins that can inhibit cellular growth and PHA accumulation in the cells. For these reasons, such molecules should be previously removed from the substrate. These high value-added substances can be readdressed towards other profitable market segments, being currently used to make edible protective films and novel food coatings to prolong shelf-life of minimally processed fruit and vegetable [22] or other applications in the food sector (tannins), although the latter is hardly removable from the raw material. Authors estimated that the extraction of natural antioxidants (polyphenols, flavonoids and anthocyanins) from grape pomace by using the supercritical CO₂ extraction technique for the production and storage of PHAs in the cells of a selected strain of *Cupriavidus necator* has produced weight surplus up to 0.2–0.25% of biopolymer when compared to the raw biomass without extraction, thereby reaching a production of about 1200 t yr⁻¹ of biopolymer [55]. The literature has showed that there are still divergent evidences about the positive correlation between the amount of pomace used in fermenter (input) and biopolymer produced (output) [40]. However, thanks to the high contents of carbon in grape pomace and tartaric acid in wine lees, potentially convertible into PHAs and succinic acid respectively (Fig. 2), winemaking co-products represent a viable feedstock to make biopolymers, thereby making it an optimal fermentation substrate after polyphenol, flavonoid, anthocyanin and tannin removal.

Bioplastics to Buildings Sector

The buildings sector is the third in demand of bioplastics after steel and cement [56]. The main requirements of bioplastics, before being employed in this manufacturing sector, are the following: durability and resistance to decay for windows and doors, thermo-acoustic insulation to increasing energy performance, lightness for reduction of transport and labour costs, propensity to further reusing into new productive cycles by composting due to its biodegradability in soil, and low maintenance costs for hygienic coatings in civil homes and hospitals.

Regarding the mechanical properties, winery wastes could improve the stiffness of bio-composites and decrease their tensile strength, meanwhile heat resistance, biodegradation rate and vapour and/or water permeability could be increased using wine fillers. The following three projects were selected within the EU framework to building blocks and reinforcing fillers in bioclimatic architecture. The first EU-funded project, BIOREFINE-2G, has developed in Italy novel processes to convert pentose-rich side-streams into biopolymers for bioclimatic architecture (<http://www.biorefine2g.eu/>). The second EU-project has developed in Spain new bio-based PHB and PBS from lignosulfonates, a category of lignocellulosic sugar waste, for demanding fire-resistant materials (<http://www.brigit-project.eu/>). The third EU-funded project, NANO3BIO, has developed in Germany microbial consortia of selected strains of fungi, bacteria and microalgae to produce environmentally friendly chitosan that will serve as a raw material for many applications including bioclimatic architecture (<http://www.nano3bio.eu/start/>). Table 2 summarizes the main opportunities of wine by-products to be converted into bio-based composites basing on chemical composition of the feedstock, processing technology to producing lactic acid and PHAs, type of bio-composite obtained and physical and thermogravimetric properties. In brief, from the table results, wine shoots, grape pomace and wine lees are better winery wastes to be converted into bio-composites.

An excellent case study whose biopolymer was widely used in bioclimatic buildings is given by the University of Stuttgart (Germany)—Institute of Building Structures and Structural Design (ITKE)—that studies biopolymer application for innovative bio-composite materials.

Table 2 Winemaking by-products converted into bio-composites on the basis of composition of the raw materials, technological process for the production of lactic acid and poly(hydroxyalkanoates) and technological features of the refined bio-composites (source: [30])

Wine by-product	Composition	Lactic acid		Poly(hydroxyalkanoates) (PHAs)		Bio-composite	Physic-thermogravimetric parameter
		Technology	Parameter	Technology	Parameter		
Vine shoot	- 34% cellulose	- <i>Lactobacillus pentosus</i>	LA: 21.8	—	—	PHBV (5–20)	Mixture of particles and long fibres
	- 20–30% hemicellulose	- Acid hydrolysis and CaCO ₃ detoxification	Y _{LA} : 0.77 P: 0.844				
Grape pomace (marc)	- 20–27% lignin	- Batch fermentation					
	- 1.25% tannins	- <i>Lactobacillus rhammosus</i>	LA: 31.5				
	- 5% proteins	- Acid hydrolysis and CaCO ₃ detoxification	Y _{LA} : 0.93 P: 1.312				
	- 3–4% ashes	- Two-stage sequential batch fermentation					
		- <i>Lactobacillus acidophilus</i>	LA: 32.7				
		- Acid hydrolysis and CaCO ₃ detoxification	Y _{LA} : 0.72 P: 1.363				
		- Two-stage sequential batch fermentation					
		- <i>Lactobacillus pentosus</i>	LA: 43.0				
		- Acid hydrolysis and delignification	Y _{LA} : 0.68 P: 0.253				
		- Saccharification and fermentation					
		- <i>Lactobacillus pentosus</i>	LA: 7.2				
		- Acid hydrolysis	Y _{LA} : 0.71 P: 0.476				
		- Batch fermentation					
	- 10.5% cellulose	- <i>Pseudomonas resinovorans</i>					
	- 6.1% hemicellulose	- Enzymatic hydrolysis	PHA: 21.3				
	- 34–41% lignin	- Batch fermentation	Y _{PHA} : 23.3 Y _{sub} : -				
	- 10% proteins	- <i>Pseudomonas putida</i>	P: 0.05				
	- 1–2% soluble polyphenols (strongly depends on if pomace is fermented or not)	- <i>Pseudomonas putida</i> KT2400	PHA: 21.8 Y _{PHA} : 77				
		- Water extraction	Y _{sub} : -				
		- Fed-batch fermentation	P: 0.10				
		- <i>Cupriavidus necator</i>	PHA: -				
	- 8–9% ashes	- Dephenolization	Y _{PHA} : 68				

Table 2 (continued)

Wine by-product	Composition	Lactic acid	Poly(hydroxyalkanoates) (PHAs)	Bio-composite		
	Technology	Parameter	Technology	Parameter		
	Technology	Parameter	Biopolymer (wt.% range)	Physic-thermogravimetric parameter		
	<ul style="list-style-type: none"> - 15% condensed tannins (insoluble) - 20% pectin substances - 30% polysaccharides 					
	Tartaric acid (50–75 kg/ton)					
Wine lees	<ul style="list-style-type: none"> (A) Solid phase - Yeast and bacteria - Cellulose, hemi-cellulose and lignin - Proteins - Organic salts (tartrates) - Pectin substances (2–6%) - Pigments (1.2%) 	<ul style="list-style-type: none"> - <i>Lactobacillus casei</i> - Alkaline hydrolysis assisted by microwaves - Batch fermentation - <i>Lactobacillus casei</i> - No treatments - Batch fermentation - <i>Lactobacillus rhamnosus</i> - No treatments - Batch fermentation 	<ul style="list-style-type: none"> - Anaerobic digestion - Two fed-batch fermentation - <i>C. necator</i> Enzymatic hydrolysis Batch fermentation - No bacteria are used - Enzymatic hydrolysis - Fed-batch fermentation 	<ul style="list-style-type: none"> Y_{sub}: 0.27 P: - PHA: 8.3 Y_{PHA}: 63 Y_{sub}: - P: 1.363 PHA: 30.1 Y_{PHA}: 71.3 Y_{sub}: - P: 0.56 	<ul style="list-style-type: none"> PHBV (10–30) PHBH (10–30) PHB (10–30) PBS (10–30) 	<ul style="list-style-type: none"> Tiny particles PS: 25 D: 1.36–1.43 T_{deg}: 268 Res: 40 Ef: 4.2–7.3
Grape stalk	<ul style="list-style-type: none"> - Organic acids (tartaric, lactic and acetic) C) Tartaric acid (100–150 kg/ton) - 20–30% cellulose - 15–20% hemicellulose - 17–26% lignin - 6–16% condensed tannins (insoluble) - 6% proteins - 6–9% ashes - 1–3% soluble polyphenols 					
Grape peel	<ul style="list-style-type: none"> - 5–12% proteins 					

Table 2 (continued)

Wine by-product	Composition	Lactic acid		Poly(hydroxyalkanoates) (PHAs)		Bio-composite	
		Technology	Parameter	Technology	Parameter	Biopolymer (wt.% range)	Physic-thermogravimetric parameter
Grape seed	- 2–8% ashes						D: -
	- 2–5% lipids						T _{deg} : 192
	- 1–2% soluble polyphenols (strongly depends on if pomace is fermented or not)						Res: 29,2
	- 1–70% soluble sugars (strongly depends on if pomace is fermented or not)						Ef: -
	- 60% dietary fibres						
	- 98.5% insoluble dietary fibres						
	- 1.5% soluble dietary fibres						
	- 4–6% pectin substances						
	- 48% dietary fibres						
	- 11.5% proteins						
Grape seed	- 13–15% lipids (oils, fatty acids)						Aggregates of particles
	- 5–8% polyphenolic compounds						PS: 750
	- 60–70% carbohydrates						D: -
							T _{deg} : 237
							Res:31
							Ef: -

PBS, poly(butylene succinate); PLA, poly(L-lactic acid), PHBV, poly(3-hydroxybutyrate-co-3-hydroxyvalerate); PHBH, poly(3-hydroxybutyrate-co-3-hydroxyhexanoate); PHB, poly(3-hydroxybutyrate); LA (g/L), lactic acid production concentration (g LA per litre of reactor); YLA (g/g), lactic acid yield with respect to the carbon source used; P [g/Lh], lactic acid volumetric productivity; PHA (g/L), PHA concentration (g PHAs per litre of medium); YPHA (%), PHA accumulated within the microbial cell; Ysub (g/g), PHA yield with respect to the used carbon source; P (g/Lh), PHA volumetric productivity; wt.%, weight fraction of by-product with respect to the processed grapes; PS, particle size (µm); D, density (kg/m³); Tdeg (°C), degradative temperature; Res (%), Young's modulus; Ef (GPa), intrinsic elastic modulus

ArboSkin Pavilion was funded by the European fund for regional development. This experimental pavilion was built with envelopes made of bio-polymeric materials that consist of a series of triangular pyramidal modules of various sizes which combine among them to create a double-curved surface self-supporting. The double-curved skin is formed by linking the pyramids together with bracing rings and joists to create load-bearing walls (Fig. 3). Researchers of the ITKE designed the freeform facade to demonstrate the structural properties of a new bioplastic envelop for use in the construction industry. The bioplastic used in the ArboSkin Pavilion project is called “Arboblend” and is an optimal design produced by combining different biopolymers such as lignin—a by-product of the wood pulping process and wine industry—with natural reinforcing fibres made of waxes and resins. The final results are biomaterials composed by extrudable granules converted into sheets completely biodegradable, thermo-formable, pierceable, laminable and printables. The project team of ITKE explained that “Thermoformable sheets of bioplastics will represent a resource-efficient alternative to oil-based plastics, glass, or metal in the future, as they combine the high malleability and recyclability of plastics with the environmental benefits of materials consisting primarily of renewable resources”. Furthermore, thermo-formable sheets of bioplastics allowed to obtain high-durability materials with good fire behaviour. Waste material from manufacturing processes can be re-granulated and brought back into the production process, while the sheets can be further re-granulated or subjected to composting at the end of their life cycle (Fig. 4). Again, the international patent of high-density panels in biopolymer fibres from agri-food waste for 90% of their weight comes from the ITKE’s project team. These panels have a high degree of elasticity. They are completely recyclable and compostable since they are made of biopolymer from starch of rice mixed with bio-composite from winemaking by-products. In particular, the high content of silicates present in rice starch conferred an excellent reaction to fire (DIN 4102-B1) to the refined material [57]. The panels can be further recycled for many times or undergo composting at the end of their life cycle [58].



Fig. 3 Spiky modules of the ArboSkin Pavilion in [Stuttgart](#) (Germany) made of bioplastic containing over 90% renewable materials (freely downloaded from the Institute of Building Structures and Structural Design of the University of Stuttgart)

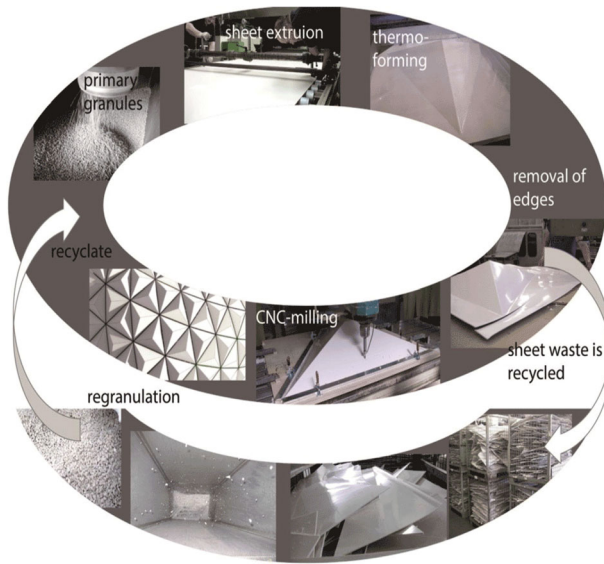


Fig. 4 Scheme of producing and recycling bioplastics of the ArboSkin Pavilion (freely downloaded from the Institute of Building Structures and Structural Design of the University of Stuttgart)

Although this scenario could implement the bioclimatic architecture since the life cycle of biopolymers resulted to be longer than other application fields, nevertheless works on life cycle assessment (LCA) were not featured. However, the main drivers that should be carefully considered in LCA are composition of fermentation substrate, temperature and time of biopolymer recovery, viscosity and boiling temperature of the refined biopolymer, productivity ratio and choice of the extraction solvent. At the current state, the production costs of bioplastic remain still too prohibitive when using alternative solvent to chloroform in biopolymer recovery [37, 38]. Thus, the global cost should be still substantially reduced to increase the competitiveness of winery wastes towards the new niches of market.

Environmental and Economic Issues

Biopolymers are currently obtained from starchy-based by-products in fermentation substrate, whenever starchy and sugary crops such as maize, sugar beet, sugarcane and potatoes were removed from the food chains to producing biopolymer rather than food [59]. The global production of bioplastics accounted to 2.11 Mt which required approximately 0.79 million hectares (ha) of lands in 2019. In order to grow the needed feedstock for bioplastics production, were required lands of around 0.02% of the global agricultural areas that correspond to 4.8 billion ha [18]. This scenario causes land consumption of about 15.7 million ha that correspond to 0.3% of all agricultural areas of the world [60, 61]. In addition, since the increasing amount of carbon to feed bacterial cultures needs further sugar addition, should be provided an additional input of sugar in fermentable substrate whose cost is strictly related to crude oil price. Therefore, the proper choice of cheaper substrates deeply determines the incidence of the total cost of production followed by the equipment costs [59].

The main driver of biopolymer production is the productivity ratio, defined as the volume of product obtained in the unit of time. Since the latter is a constant factor, products and

equipment vary consequently with the related costs. In order to rationalize the natural resources, the limiting factor of using winemaking by-products for making bio-composite materials is the lower productivity of biopolymer than starchy-based substrates [62]. Another limiting factor concerns potential incompatibility between wine co-products and other agro-based by-products. Removal of phenolic-based substances and tannins could be a suitable option to solve this issue by avoiding growth inhibition or slowdown of microbial cultures.

The last critical issue of biopolymer production, but no less important, depends on the PHA and PHB granule recovery from bacterial cells and their further purification. Conventionally, by using solvents that allow granule collection by precipitation process is the most used technique. The remaining solid residue, composed by dead cells and residual biomass, can be further recycled into new fermentation cycles, thereby continuing to feed the production cycle of PHB from wine lees [40]. This phase characterizes and limits quality of refined biopolymer. In order to use quantitative reasoning by comparing appropriate benchmarks, a series of risks and issues related to toxic solvent that can compromise large-scale production of bioplastics should be taken into account. The current use of flammable volatile solvents is linked to purity degree of the biopolymer, a quality factor that could compromise its own performance. In fact, if the most used solvent category belongs to chlorinated hydrocarbons (such as chloroform) that are highly toxic, flammable, expensive and overall un-recyclable and hazardous for environment, on the other hand, alternative molecules can affect the purity degree of biopolymer. Alternatives to use of chloroform were found in literature. Authors focused the issues related to recovery of PHBs from *C. necator* that significantly increased total processing costs [38]. The adoption of efficient, economical viable and eco-friendly extraction solvents for PHB recovery from the cells is increasingly needed for improving the industrial production on larger scale. They tested an array of non-halogenated organic solvents such as dimethyl sulfoxide, dimethyl formamide, ethylene carbonate, propanol, methanol, hexane and acetic acid to recover PHB with different incubation temperatures and times, here considered as the main variables that should be carefully monitored during the bioplastic production cycle. These authors observed that the highest recovery percentage (98.6%) and product purity (up to 98%) were correlated to ethylene carbonate-based extraction carried out at 150°C for 60 min. Molecular weight of the recovered PHB (average 1.3×10^6) was not significantly ($p > 0.05$) related to the solvent type or the extraction variables. The melting point of PHB extracted using ethylene carbonate was 176.2°C with an enthalpy of fusion of 16.8% and a corresponding degree of crystallinity of 59.2%. Other sustainable alternatives for biopolymer recovery can be recorded. Another study deals with use of non-halogenated organic solvents, thereby obtaining high degrees of purity of refined biopolymer. The same authors developed an efficient and eco-friendly method for recovering PHB from *C. necator* by testing two non-halogenated molecules [37]. They found that butyl acetate and ethyl acetate were both suitable solvents for the biopolymer recovery. But, butyl acetate had a higher recovery percentage (96%) and product purity (up to 99%) than ethyl acetate. In addition, PHB recorded the highest molecular weight (average 1.4×10^6) in biopolymer recovery when compared to chloroform. This study showed that butyl acetate is noticed as a good alternative to chloroform in recovering PHB. In addition, temperature and time of extraction, viscosity and boiling temperature of the refined biopolymer driven the whole process and should be taken into account in relation to the choice of the most proper solvent.

Conclusion

The environmental issues related to plastic disposal are focus that the EU has imposed by 2030 with the “Green Deal” whose more research is addressed on new biopolymers from agri-based biomass. In view of the crucial need across the world for sustainable materials, agro-waste deems special importance for their reutilization, being employed for several practical purposes. The cogent need to replace unrenewable plastics from fossil source with biodegradable and compostable biomaterials is a fundamental challenge in the bioplastics industry. The paper covers this theme providing some feasibility results that can help user and stakeholder to recycle agro-wastes into the buildings sector according to the sustainability and circular economic principles. In this sense, winemaking by-products can be truly an ideal substrate, even if combined with other treatments to obtain antioxidant substances and tannins for food packaging. The spreading use of winery waste in Europe (mainly in Italy) could reduce land consumption, so freeing up economic resources. Moreover, in order to maintain high level of productivity by using overall wine shoots, grape pomace and wine lees for microbial fermentation, some opportunities in minimizing the production cost of feedstock were drawn. The use of winery by-products as reinforcing fillers for making bio-composites in bioclimatic architecture is a suitable way since wine wastes are inexpensive (e.g., wine lees costs about 0.045 €/kg and grape pomace 0.022€/kg), abundant (especially in the EU and Italy) and easily processed with relatively high temperature without significant efficiency loss. Winemaking by-products could be a promising source of raw material for the buildings sector that needs higher performance of the bio-composite materials than other applications. In order to maintain a good degree of purity of the refined biopolymer, should be used eco-friendly solvents in biopolymer recovery for safer handling of chemicals that, having costs comparable to chloroform, could obtain more savings. Nonetheless, the common use of chlorinated solvents still remains a critical issue which has not been fully solved. Finally, LCA studies are still needed to investigate in-depth the relationships among the costs sustained of using an adequate fermenter capacity and the savings obtained by employing wine shoots, grape pomace and wine lees when compared to other non-edible and inexpensive second-generation feedstocks.

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Code availability Not applicable

Declarations

Additional declarations Not applicable

Ethics approval Not applicable

Consent to participate Not applicable

Consent for publication Not applicable

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