

Circular and Bio-Based Solutions for the Ultimate Prevention of Plastics in Rivers Integrated with Elimination And Monitoring Technologies

Deliverable D.2.1

Mapping of alternative polymers

Deliverable information

Project profile

Document history

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Executive Summary

UPSTREAM project aims to improve the cleanliness and water quality of the rivers by deploying and demonstrating in 5 demo sites a suite of 15 advanced solutions to deal with pollution in terms of litter, plastic and microplastic in European rivers. This challenge is afforded by a consortium (22 partners from 11 countries), from top European Research and Technology Organisations (RTOs), specialized Small and Medium-sized Enterprises (SME) technology providers, a large company and completed by promoting a strong engagement of citizens and stakeholders.

The objective of *D2.1 Mapping of alternative polymers* is to provide an overview of the most promising bioplastics to substitute conventional petroleum-based plastics and thus reduce persistent plastic pollution. Specifically, their characteristics, main applications, and challenges to be overcome have been collected throughout this document.

This document aims to serve as a preliminary database of most promising polymers in terms of biodegradability and reduced end-of-life impact in the environment as alternativesto persistent Litter (L), Plastic (P) and MicroPlastic (MP) materials. The data provided in this document will be updated and embedded within the data platform to be developed within T1.5.

Deliverable Keywords: UPSTREAM, bioplastic, database, end-of-life, biodegradability.

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1. INTRODUCTION

1.1. PROJECT OVERVIEW

To address the current issue of litter (L), plastics (P), and microplastics (MP) in the European river systems, it is crucial to take actions along the whole water supply chain: prevention by design, release prevention after wastewater treatment plants (WWTPs), and remediation in the rivers themselves.

The overall objectives of UPSTREAM are to: (i) address pollution at every point in the path of L, P, and MP, (ii) to couple the technical demonstrations with analysis of circular (bio-based) value chains, environmental (biodegradability) and economic sustainability assessments, focusing efforts on knowledge co-creation and replication to accelerate the reduction of pollution in European rivers and (iii) to engage stakeholders at all levels – industry, government, and citizens.

To do that, the UPSTREAM project will deploy and demonstrate, in 5 demo sites, a suite of 15 advanced solutions that address the pollution from L, P, and MP in European rivers adopting 5 pillars – monitoring, prevention, reduction of L, P and MP at wastewater treatment plants and rivers, and valorisation of collected plastics. Besides, replication across Europe will be enhanced by means of a cascade funding initiative to join early adopters beyond the partnership to scale up the solutions quickly, contribute to the Mission objectives, and expand the water system knowledge base throughout Europe.

UPSTREAM represents a pan-European consortium with 5 demo sites across Europe, including 4 WWTPs (UK, ES, DE, IT), plus a testing area on the Danube in Serbia. The consortium (22 partners from 11 countries) is strengthened by top European Research and Technology Organisations (RTOs), specialized Small and medium-sized enterprises (SME) technology providers, a large company world leader in sustainable bioplastics development, Novamont, and completed by partners dedicated to creating a digital knowledge sharing platform and engaging with citizens and stakeholders.

1.2. CONTEXT AND DOCUMENT OVERVIEW

The definition of a "bio-based" product establishes a direct link between the biomass and the product (even if intermediate step(s) are necessary to obtain the biobased product from the biomass). The word "bio-based" is intrinsically related to the carbon source from which the product comes from: carbon-based monomers from renewable sources (i.e. Biomass) as specified in the European standard CEN/TC 411 - Bio-based products. The term "bio-based" therefore strictly refers to the origin of the raw materials (carbon source) which can be divided into two different main groups: renewable (such as byproducts from the agroindustry, agricultural sector, organic waste or sewage sludge) and fossil (such as crude oil, coal, natural gas...). To determine the renewable carbon content in bio-based polymers, it can be followed the test method EN16640:2017.

Usually bio-based materials (such as bioplastics) composed of biopolymers and often other raw materials, which have natural origin, are also biodegradable. Bio-based materials can be subdivided into 3 categories:

- 1. Materials obtained directly from biomass, such as wood, paper pulp, cellulose, starch and proteins.
- 2. Materials that can be made from building-blocks originated (i.e. *via* fermentation) from biomass, such as PLA.
- 3. Materials that are produced (*via* fermentation) by microorganisms, such as PHA.

Bio-based polymers and plastics have advantages over conventional counterparts as they can reduce the dependency on limited fossil resources and reduce greenhouse gas emissions or even be carbon neutral. Since the chemical composition of some of them is equal to the traditional materials (i.e. bio-PE), independently from the carbon origin, these products do not always fulfil the biodegradability standard criteria. Therefore, biobased plastics and polymers are not necessarily biodegradable, in the same way as conventional ones. Their production, replacing the traditional homologous polymer (i.e. bio-PE vs PE), can help the EU to meet its 2030 targets [1] of greenhouse gas emissions reduction. Moreover, bio-based plastics can significantly contribute to greater resource efficiency through a closed resource cycle and cascade use, especially if bio-based materials and products are being either reused or recycled (i.e. material recycling: mechanical and chemical or, if compostable, organic recycling into compost or digestate following the EU reg 1009/2019) and recovered for energy production (i.e. renewable energy in form of biogas/biomethane for the biodegradable materials and/or thermal energy for the not biodegradable ones) [2].

Figure 1. Most industrially relevant bioplastics [2]

In contrast, biodegradable plastics and polymers can be biodegraded by living organisms such as bacteria, eventually returning to the environment as CO₂ and water, closing the carbon cycle. Some biopolymers can be fully bio-based and not biodegradable (i.e. Bio-PE) while others are petroleum-based but biodegradable (i.e. PBAT). The main advantages of bioplastics are related to their characteristics to be both bio-based and biodegradable (i.e. PLA, Starch Blends...) [\(Figure 1\)](#page-9-0).

Bioplastics are commonly used for food packaging manufacturing, and they can be used in the very restrictive legislation in the field of food contact applications (Reg EU 11/2010). Compostability is an additional property attributed to biodegradable polymers offering end-of-life advantagesin applications where food contamination can be present (i.e. food packaging, tableware, cutlery, coffee pods) or where the materials are used to collect organic waste (i.e. waste bags, shopping bags, ultra-light fruit and vegetable bags), and its use can help the member state to increase the organic waste collection system in a qualitative and quantitative way improving its valorisation as a valuable soil improver (compost)[3]. Compostable materials are items that meet the international EN13432 standard for compostable plastics. EN13432-compliant materials (from food packaging to waste bags, shoppers or

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coffee pods…) are allowed to carry the seedling logo and can be collected with the organic fraction of municipal solid waste and be composted [4].

Biodegradability is strongly connected to specific environmental conditions: temperature, presence of microorganisms, presence of oxygen, macronutrients availability, and free water are the key parameters to take in account when a biodegradability assessment must be performed.

In the past, there was an increase in the number of products which appeared on the market for conventional plastics (such as polyethylene) which have been made degradable by means of additives (OXO additive). Currently, 'oxodegradable plastic' indicates plastic materials that include additives which, through oxidation, lead to the fragmentation of the material into micro-fragments or to chemical decomposition. Those additives have been banned from the EU market according to a final report published in August 2016 due to the creation of persistent microplastics [5].

Regarding end-products, there are already biobased plastics (or blends) that can serve as alternatives for several petrochemical plastics and can be used for different applications. Some of the most relevant end-products are summarized in the [Table 1.](#page-10-0)

Table 1. Overview of packaging applications made of fossil plastics and biobased alternatives and their intrinsic biodegradability

It is important to note that biopolymers developed on an industrial scale cover limited property windows when considered as standalone materials. It is difficult to foresee applications in which a single biopolymer could be used as it is. That is why biopolymers are combined and processed to obtain bio-materials with tailored characteristics based on specific final applications[. Table 2](#page-11-1) reports the main characteristics of the most relevant biopolymers.

The bioplastics market is rapidly expanding as industries seek sustainable alternatives to conventional plastics. Globally, they account for about 0.5% of the more than 400 million tonnes of plastic produced annually, reaching 2.18 million tonnes in 2023. However, global production capacity is set to increase significantly to about 7.43 million tonnes in 2028 [3]. The most popular biodegradable and compostable bioplastics on the market [\(Figure 2\)](#page-11-0) are polylactic acids (PLAs), starch-based bioplastics (or more generally: starch-contained polymer compound - SCPC), polybutylene adipate terephthalate (PBAT) and polyhydroxyalkanoates (PHAs family).

Global production capacities of bioplastics 2023 by material type

Regenerated cellulose films

Figure 2. Bioplastics production percentage in 2023 [2]

The European Commission published and adopted in January 2018 the European strategy for plastics as a part of the first circular economy action plan [7]. This strategy aims to manage plastics from a circular economy perspective. The action plan established concrete and ambitious actions, with measures covering the whole life cycle: from production and consumption to waste management and the market for secondary raw materials and a revised legislative proposal on waste. Related to plastic, the plan includes:

- 1) Reuse or recycle plastics effectively (by 2030).
- 2) Replace or phase out substances that hinder plastic recycling.
- 3) Develop and use innovative materials and alternative raw materials to produce plastics that are more environmentally sustainable than non-renewable raw materials.

Regarding the latter objective in particular, bioplastics, which have technical characteristics comparable to those of conventional plastics, are already successfully used in the production of items such as food packaging materials, disposable tableware, shopping bags and organic waste bags.

In the pursuit of greater environmental sustainability and achieving the goals identified in the European plastics strategy, biodegradable plastics from renewable sources are of particular interest, as their production and end-oflife management can lead to a reduction in environmental impact compared to conventional plastics. Indeed, instead of using fossil resources such as oil for their production, a theoretically carbon-neutral source such as biomass is used. Furthermore, end-of-life management should be significantly improved by recycling them in the composting process within the organic fraction of municipal solid waste. The need to pursue, at the European level, a resource-efficient and climate-neutral circular economy model, together with the ambition to achieve zero pollution and the need to protect and enhance biodiversity, triggered a general rethinking of the way some plastic products are produced, used and managed as waste (where recycling is favoured over other forms of management such as energy recovery and disposal).

Bioplastics are emerging in everyday life as alternatives to some of the conventional plastics (those most at risk of environmental dispersion where biodegradability, understood as the intrinsic property of the material, is to be considered an added value due to its non-persistence in the environment). In 2017, the European Commission requested ECHA to assess the scientific evidence for taking regulatory action at the EU level on microplastics that are intentionally added to products (i.e. substances and mixtures). As reported by the European Bioplastics Association, biodegradable plastics and polymers are gaining more significance as a potential solution. Although biodegradable and compostable plastics do –as all solid materials– produce small particles through abrasion when in use, they are not release persistent fragments like by conventional, non-biodegradable materials [8].

Currently, more efforts are in act to gain a better understanding of the origin and creation of microplastics and their effect upon release into the environment, in order to minimise their environmental impacts. Recent scientific studies have been looking into the option of biodegradable materials to be a potential solution for microplastic accumulation, because in natural environments ubiquitous microbes present are able to metabolise biodegradable polymers that accidentally reach them in the form of small fragments [9]. Biodegradable polymers have also the advantage that they do not erode into permanent secondary microplastics upon degradation, thus, the residence time is considerably lower for biodegradable polymers compared to conventional plastic materials. In this way, biodegradable plastics can help to minimise environmental impacts and reduce the accumulation of plastic particles in different environmental habitats.

Biodegradable bioplastics, if certified in accordance with UNI EN 13432:2002 for packaging (UNI, 2002) or UNI EN 14995:2007 for other materials (UNI, 2007), have the advantage of being suitable to be valorised, at the end of their life, in the organic waste treatment system and organically recycled together with the organic fraction of the municipal solid waste (OFMSW). Currently, the virtuous end-of-life path for compostable bioplastics is to be organically recycled by their inclusion and biological treatment (aerobic and/or anaerobic) with the OFMSW. Furthermore, the use of bioplastics in selected applications - aimed at reducing our dependency on traditional feedstock as well as at reducing the plastic pollution of sea and soil is an essential tool towards meeting the ambitious sustainable development goals described in the "Resolution adopted by the United Nation General Assembly" on 2015 [10].

Within WP2 objectives, D2.1 aims to do a comprehensive study in the state of the art for bioplastics and their biodegradability commonly used at industrial scale in order to have a better knowledge in the different alternatives for the UPSTREAM purpose.

2. LIST OF ALTERNATIVE POLYMERS

2.1. PLA: Polylactic acid

2.1.1. Material Overview

Polyesters derived from hydroxy acids are one of the large family of linear polymers which form the basis of many industrially biodegradable bioplastic grades relevant in everyday applications. In detail, the most common polyester derived from a hydroxy acid is polylactic acid (PLA), a thermoplastic, compostable, and renewable biopolymer.

Since its discovery by the chemist Wallace Hume Carothers [11] from the experimental station of DuPont in 1932, PLA has found applications in a multitude of relevant industrial applications (i.e. packaging, textile, films, hygiene absorbents, 3D printing and more). The development of PLA grades came in the last 30 years thanks to both progresses in polymerization technology and ecological chemical products that has enabled the production of PLA grades with high molecular weight and thus suitable characteristics to be processed in various technologies [12][13][14].

Nowadays, the production of PLA mostly follows the scheme reported in [Figure 3,](#page-13-4) where the plant-derived sugars (e.g. starch from corn and potatoes, sucrose from beets and sugar cane) are fermented into lactic acid which is prepolymerized into a linear oligomer with a low molecular weight. This lactic oligomer is converted in six-membered cyclic dimers of lactic acid (lactide) than, after purification, are subjected to ring-opening polymerisation to yield a high-molecular weight PLA.

Figure 3. Schemes of the production process of PLA through lactic acid fermentation, lactide formation and ring-opening polymerization

The possibility of sourcing lactic acid, its technology of purification and the ring-opening polymerization permit to obtain high-molecular weight PLA that are well established and required at industrial level. Indeed, the global capacity of PLA production has reached more than 300 kt/y, and the main producers are: Natureworks (Blair US) and Total-Corbion (Rayong, Thailand).

2.1.2. Most common applications

PLA was first produced in the 1700s, and its first biomedical application was in the repair of mandibular fractures in dogs. However, because of its high cost and low availability, the use of PLA was firstly limited to medical applications, but currently high molecular weight PLA can be processed by extrusion, injection moulding, blow moulding, electrospinning and thermoforming. PLA also offers similar thermal, mechanical, optical and barrier properties compared to commercially available polymers, such as polystyrene (PS), polypropylene (PP), and polyethylene terephthalate (PET), thus widening its use for a range of other applications [15].

PLA is extremely versatile, and can be moulded into several structures, including nanoparticles, films, scaffolds, rods, sutures and micelles. Its industrial uses include both durable consumer goods and perishable goods, such as flexible films and rigid packaging, cutlery, cold drink cups, bottles, clothing and staple fibres, injection moulded products and extrusion coating [16].

The biodegradability and biocompatibility of PLA support its use for several medical and biomedical applications. The innovative features highlight the role of PLA in the biomedical field, in particular, for tissue engineering by improving healing rates and, as well as for implants and prosthetics [16].

A revolutionary application of PLA currently under investigation is 3D printing. 3D printers will greatly expand the possibilities in biomedical engineering, enabling numerous applications for PLA, which will be widely used as a temporary extracellular matrix for tissue engineering [17].

PLA is commonly blended with other polymers for different applications. Below it is shown the most typical application for each PLA-based blend [18]:

2.1.3. Biodegradation

PLA is a fully biobased bioplastic, derived from renewable monomer precursors through polymerization processes, and it is biodegradable under industrial composting conditions. It has recently been shown to be biodegradable in cold environments [19]. However, PLA's biodegradability in cooler (soil, freshwater, home composting...) or mesophilic conditions (20-45°C) is relatively slow. This slow rate is due to PLA's need for an abiotic pre-hydrolysis process, which requires heat and moisture. During this process, PLA undergoes depolymerization, leading to the formation of oligomers and lactide monomers that can then be rapidly biodegraded by microorganisms. The rate of hydrolysis, and consequently biodegradation, is significantly faster in high-temperature and high-humidity environments typical of industrial composting conditions

2.1.4. Challenges to overcome.

Some of the limitations of PLA compared to fossil-based plastics, are low thermal stability and fragility. To try to overcome the limitation of PLA related to its hardness, researchers are currently trying to blend PLA with other stereo complexes such as PLLA/PDLA, which are the most common types, both synthesized by ring-opening (ROP). This blend improves the mechanical and thermal properties of PLA-based materials, besides their resistance against hydrolysis, due to the strong interaction between the sequences of L-lactyl units and D-lactyl (the two different forms of lactic acid used in PLA).

PLA has a lower melting point compared to many conventional plastics. This can limit its use in applications requiring high heat tolerance, such as hot beverages or sterilization [20].

Even though PLA is more cost-competitive than other biopolymers (such as PHA), its cost is still higher than other conventional alternatives. Anyway, as its production increases, operational costs decrease, allowing PLA to be used in a great deal of products.

2.2. PBAT: Polybutylene adipate terephthalate

2.2.1. Material Overview

Poly(butylene adipate-*co*-terephthalate) (PBAT) is a member of the aliphatic-aromatic polyester family. It is a synthetic statistic polyester obtained by polycondensation reactions starting from 1,4-butanediol, adipic acid and terephthalic acid. PBAT is characterized by a relatively low degree of crystallinity which makes it tough, and soft, with high deformability and good hydrolytic resistance. Based on these characteristics, PBAT is typically used in the design of soft materials with good film-forming properties. PBAT has been industrially available since the 1990s thanks to early development by Eastman under the Easter Bio trademark and soon after by BASF SE with Ecoflex materials. I[n Figure 4,](#page-15-4) the scheme of macromolecular structure related to PBAT is represented.

Figure 4. Schematic macromolecular structure of PBAT (aliphatic-aromatic polyesters)

Regarding the aliphatic-aromatic polyester family, the Novamont group has developed proprietary technology for the production of PBAT.

2.2.2. Most common applications

PBAT are a class of soft biopolymer (Elastic modulus (E) < 150 MPa) with high deformability and low glass transition temperature, has good crystallization and thermal stability; as a result, it has good processing stability to be used alone or blended with other materials through conventional manufacturing process such as extrusion, intrusion and blowing film, etc. However, as a biodegradable and flexible biopolymer, is usually designed for film extrusion and extrusion coating applications.

Anyway, PBAT based products (compounded with starch, PLA, or other additives) have been widely used in many applications such as packaging, mulch film and bags, etc. for their good quality, satisfactory performance and competitive cost [23].

Some examples of these products are clinging wrap for food packaging, compostable plastic bags for gardening and agricultural use, and water-resistant coatings for other materials, such as in paper cups.

2.2.3. Biodegradation

PBAT is biodegradable polyester under aerobic conditions. Cutinases from bacterial strains such as *Thermobifida fusca* can degrade PBAT into terephthalic acid (TPA) and contribute to the complete decomposition of PBAT. These bacterial strains are commonly found in mature compost. In freshwater environments, PBAT has been found to not be readily biodegradable [24] but as a polyester, PBAT undergoes hydrolysis in aquatic environments, so fragment generation is a matter to consider when PBAT degrades in water, especially after exposure to UV light [23]. Under UV or sunlight, PBAT undergoes oxidation, leading to both crosslinking and chain scission, which promotes the formation of fragments from the polymer. According to Xin-Feng Wei *et al*. (2021), fragmentation and subsequent

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biodegradation occur more readily for PBAT than for conventional non-biodegradable LDPE in aquatic environments [25].

PBAT is one of the main biopolymers suitable for home composting or soil biodegradation because is usually based on easily biodegradable monomers that characterised its amorphous aliphatic-aromatic polyester macromolecular structure.

2.2.4. Challenges to overcome

PBAT can be more challenging to process during manufacturing compared to traditional plastics. These challenges can be related in finding the right processing conditions to achieve desired shapes and properties or creating the most appropriate blend with other biopolymers to gain the most adequate level of performance required by the specific final application

PBAT can have lower mechanical strength than conventional plastics. This might limit its use in applications requiring high durability or load-bearing capacity.

Currently, PBAT is still more expensive than traditional plastics (i.e. PE) due to lower production volumes and more complex processing needs, the content of renewable monomers, the production sites located mainly in Europe and USA where manpower, energy and environmental protection laws determined an intrinsic increase of cost [26].

2.3. PBS(A): Polybutylene Succinate

2.3.1. Material Overview

Poly(butylensuccinate) (PBS) and its aliphatic copolyesters are semicrystalline polymers belonging to the polyester family. They are known for their good overall biodegradation behaviour and thermal characteristics, making them suitable for various applications. This family of polyesters is obtained mainly by the polycondensation of 1,4 butandiol and succinic acid (and other monomers in case of copolyesters). Although PBS and its copolyesters were first investigated by Carothers in 1931, significant interest in their biodegradability and advancements in polycondensation processes only emerged in the 1990s [27]. These advancements have allowed for the production of materials with suitable molecular weights and physicochemical properties.

One development case of PBS synthesis is the Bionolle grades produced by Showa High Polymer. These were obtained via melt condensation polymerization followed by a chain-extension method using diisocyanate as the chain-coupling agent, to increase the molecular weight [28]. Similarly, in 2003, Mitsubishi Chemicals, introduced PBS grades (tradename GS Pla) with high molecular weight through direct melt polycondensation. This was followed by Anqing He Xing Chemical Co with its own direct melt polycondensation method, and Xinfu Pharmaceutical (China) with one-step polymerization technology [29].

Nowadays, Mitsubishi Chemical Corporation (MCC) and TT Public Company Limited (PTT) have funded the joint venture PTT MCC under the BioPBS brand with a declared production capacity of 20 kt/y. The physical properties or the biodegradation rate can be adjusted by controlling the copolymerization and molecular weight of the PBS chains [30].

Figure 5. Chemical structure of (a) succinate acid, (b) 1,4-butanediol, and (c) poly(butylene succinate) (PBS)

2.3.2. Most common applications

PBS is a versatile material used to produce various packaging solutions, including food packaging, beverage containers, and cosmetic containers. It can be processed into films, bags, or boxes, offering an eco-friendly alternative to traditional plastics.

PBS is suitable for disposable products such as tableware, cutlery, and medical devices due to its biodegradability and processability [31].

Research is exploring the use of PBS in various other fields beyond packaging and disposables. Potential applications including [30]:

- Agriculture: Mulch films, seedling pots
- Fishery: Fishing nets for reduced environmental impact after use
- Construction: Components with biodegradability for temporary applications
- Electronics: Biodegradable housings for electronic devices

2.3.3. Biodegradation

PBS biodegradation is widely proven under aerobic composting conditions. PBS and PBSA copolymers are considered biodegradable in water following the standardised guideline for chemicals OECD 301, and PBSA also meets the 10-day window criteria, so it can be considered readily biodegradable in water [32]. Furthermore, the properties and the biodegradation rate of PBS can be tailored via copolymerization with several types and contents of comonomer units to meet various requirements.

2.3.4. Challenges to overcome

PBS shows promising properties as bio-based and biodegradable plastic, but there are still hurdles for its widespread use.

Like PBAT, PBS can be more challenging to process during manufacturing compared to traditional plastics. Finding the right equipment and processing conditions to achieve desired shapes and properties can be an obstacle [33]. PBS can have lower mechanical strength than some conventional plastics limiting its use for specific applications that require high durability or load-bearing capacity.

Similarly to PLA and PBAT, decreasing the cost of the material would make it more competitive in relation to conventional plastics [32].

2.4. PHA: Polyhydroxyalkanoates

2.4.1. Material Overview

Polyhydroxyalkanoates (PHAs) are a family of biodegradable polyesters naturally synthesized by numerous microorganisms as intracellular carbon and energy storage. PHAs can be produced through bacterial fermentation using various carbon sources, and their properties can range from brittle, highly crystalline thermoplastics (e.g. poly(3-hydroxybutyrate), PHB) to amorphous, rubber- or glue-like elastomers (e.g. medium-chain-length PHA, mcl-PHA), depending on the number of carbon atoms in their monomer units.

The most common PHA from direct fermentation is poly(3-hydroxybutyrate) (P3HB) also simply referred to as poly(hydroxybutyrate) (PHB). As mentioned before, the chemical structure of the PHAs obtained by fermentation depends on the mix of carbon feedstocks fed during the accumulation stage, the metabolic pathways that the bacteria use for the conversion into precursors and the substrate specificities of the enzymes involved. Through careful selection of these parameters, it is possible to obtain a wide range of PHA copolymers [\(Figure 6\)](#page-18-3).

Poly(3-hydroxybutyrate) - PHB

Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) - PHBV

Poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) PHRH

Poly(3-hydroxybutyrate-co-4-hydroxyhexanoate) P3HB4HB

Figure 6. Comparison of the macromolecular structures of polyhydroxyalkanoate obtained via fermentation: PHB, PHBV, PHBH, and P3HB4HB

These biopolymers are one of the most promising sustainable alternatives to traditional plastics due to their similar mechanical properties and complete biodegradability, including home composting, soil and marine environments.

Among these, while the homopolymer PHB was the first to be investigated, the copolymers poly(3 hydroxybutyrrate)-*co*-(3-hydroxyhexanoate) (PHBH or PHBHH) and poly(3-hydroxybutyrrate)-co-(3 hydroxyvalerate) (PHBV or PHBHV) are the ones adopted in the few industrial applications developed up to now.

2.4.2. Most common applications

Over the last decade, several partnerships and collaborations have been developed aiming to increase the commercial versatility of PHAs. The main applications reported for this biopolymer comprise: flexible and rigid packaging, coating applications, consumer goods (e.g., single-use containers for cosmetic and hygiene materials), durable goods, compostable bags for solid waste and agriculture/horticulture bags and films (e.g. mulch films).

Moreover, the unique biocompatible and biodegradable properties of PHAs make them suitable to be used in highvalue applications in the medical field, such as in bone plates, surgical sutures, patches, and as delivery systems for slow release of hydrophobic drugs [34]. In addition, PHAs have demonstrated to be promising in a variety of other fields, including biofuels, raw material for paints, waterproof coating for paper and fibre material [35].

One of the most widely commercially available PHAs are polyhydroxybutyrate (PHB) and poly(3-hydroxybutyrateco-3-hydroxyhexanoate) (PHBV), both of which are short-chain PHAs [36] and have the potential to substitute PE in some film application.

2.4.3. Biodegradation

The biodegradation pathways of PHAs are similar to those of starch, as approximately 90% of the starch-degrading organisms can also degrade short-chain PHAs [38]. Under controlled composting conditions by following standardized guidelines UNE EN ISO 14855-1 PHAs with multiple types of chemical structures can be biodegraded [39]. When evaluating PHA biodegradation in aquatic environments, according to standardized guidelines, such as UNE EN ISO 14851 and 14852, PHAs are inherently biodegradable [40][41].

PHA and PHB biosynthetic polymers can be blended with other synthetic polymers such as PVA, PCL and PLA, enhancing not only their mechanical properties but also their biodegradability potential [42].

The biodegradability of polymer blends containing PHA/PHB is strongly dependent on the phase structure, surface and volume ratio of the blend. Generally, the amorphous part of the PHA chain degrades faster than the crystalline part, so the mineralization by microorganisms is influenced by the crystallinity of the polymer. This means PHAs that with lower crystallinity such as PHBV, are more quickly mineralized than PHB copolymers.

A typically used blend for PHA/PHB biopolymer is PVA-PHB, as PVA is a versatile macromolecular compatibilizer [43]. However, the crystallinity level of PHB could negatively affect the PVA biodegradation rate [37].

PHAs with longer repeating units exhibit improved biodegradation behaviour, partly due to their lower glass transition temperature (T_g) compared to PLA. PHAs can be biodegraded by microorganisms in a wide range of conditions including industrial composting plants, home-composting bins, soil, anaerobic digestion plants and even in water at specific temperatures [37] along with other biodegradable bioplastics such as aliphatic and aliphaticaromatic polyesters.

Another common blend is PLA-PHB, which improves their ductility, toughness and thermal stability [44].

2.4.4. Challenges to overcome

PHA are biopolymers with significant potential to replace conventional plastics, offering a more sustainable solution due to their biodegradability and production from renewable resources. However, the journey towards their widespread adoption is not without challenges. One of the primary hurdles is the high production cost and consequent market price, which stems from the expensive carbon substrates used in the fermentation process (accounting for 30-50% of the total production costs) and the energy-intensive bioproduction process using pure cultures currently used for commercial polymer production [45].

To address these issues, research is focusing on utilizing cheaper, renewable feedstocks and improving bioprocessing techniques to enhance yield and reduce energy consumption. For example, studies utilizing mixed microbial cultures have demonstrated cost reduction through less energy-intensive processes and the successful utilization of waste materials and agri-food residues as substrates [46] [47] [48] [49].

PHAs, especially the homopolymer PHB, may exhibit poor mechanical properties compared to their counterparts, which limits their application in industries that demand high-performance materials [50]. To overcome this issue, research efforts are exploring various strategies such as blending PHAs with other biopolymers or additives to enhance their mechanical properties. Additionally, the use of PHA copolymers, such as PHBV, has shown potential in producing materials with improved flexibility and strength [51].

Another challenge is the scalability of PHA production. While the pilot-scale processes have been successfully demonstrated using a cheaper feedstocks approach [46] [47] [48] [49], translating these to industrial scales poses some difficulties in maintaining the same level of efficiency and quality. This challenge is compounded by the need for precise control over operating conditions to ensure good yields, high productivity, and consistent polymer properties when the feedstock composition varies [46]. Moreover, the current purification processes for PHAs can be environmentally taxing and cost prohibitive. Advancements in downstream processing, such as developing environmentally friendly and cost-effective purification techniques, are being investigated to further lower production costs and minimize environmental impact [52].

Furthermore, the integration of PHAs into existing manufacturing infrastructure is a significant barrier. Most current industrial processes are optimized for traditional plastics, and significant investment is needed to modify equipment and processes to accommodate PHAs. This transition also requires a shift in consumer behaviour and perception, as the market must be educated about the benefits of PHAs over conventional plastics.

Despite these challenges, the future of PHAs looks promising. As society increasingly prioritizes sustainability, PHAs offer a viable solution in the quest to reduce plastic pollution and dependence on fossil fuels. Through ongoing innovation and collaboration among scientists, industry, and policymakers, the hurdles associated with PHA usage can be overcome, paving the way for a greener and more sustainable future.

2.5. Starch-based polymers

2.5.1. Material Overview

Starch is a natural and renewable polysaccaride used by plants, including roots, stalks, and crop seeds, as an energy reserve. It is the second most abundant biomass material found in nature. Industrially, starch is extracted from agricultural raw materials such as grains (corn according for 75% and wheat 8%), or tubers (potatoes, 45% and cassava 12.5%) [53]. Considering the global production of corn (1147 Mt/y), wheat (734 Mt/y), potatoes (368 Mt/y) and cassava (277 My/y), it is possible to understand why starch is such an abundant and low-cost renewable raw material [54]. Additionally, starches are used in a variety of applications, including livestock feed, processed food, industrial chemicals (such as in bioethanol fermentation), pharmaceutical excipients, and paper industry additives, among others.

To process starch, its native form must undergo significant structural modifications through various chemical, physico-chemical, and mechanical treatments. These modifications can alter its hydrophilic properties, swelling behaviour, rheological characteristics, and both physical and chemical properties.

Starch can be converted into a thermoplastic material through a combination of heat, water, and shear. This process, known as plasticization, involves the use of water and/or plasticizers (such as glycerol) to reduce the intermolecular forces between starch molecules, allowing them to flow and be shaped under heat and pressure. The resulting thermoplastic starch (TPS) can be further processed through melt extrusion or moulding, where it becomes a versatile material that can be blended with other polymers to enhance its properties. TPS blends exhibit improved mechanical properties, such as increased strength and rigidity, making them suitable for a range of industrial applications, including packaging, agricultural films, and disposable items.

TPS is distinguished from native starch by its ability to be processed at elevated temperatures and pressures, which makes it more flexible and mouldable. This transformation into a thermoplastic form is crucial for its use in various commercial and industrial applications.

The starch-based polymers family can be processed like traditional plastic polymers in film blow machines. By adjusting their constituents, such as by adding other polymers or additives, it is possible to modify the mechanical and technical properties of the polymer to suit various end uses (e.g. transparent bags, waste bags, shopping bags, water-resistant food packaging, gas barrier food packaging, and more). Depending on the raw materials used to produce the polyesters, it is possible to modify the renewable carbon content, producing a remarkably high renewable final starch-based material.

2.5.2. Most common applications

The most common applications are based on film blow technology (bags, mulch, flexible food packaging among others). A typical application, where compostable bioplastics give the best results, is the waste bags to collect the organic fraction of municipal solid waste or shopping bags that can be re-used to collect organic waste at the household level.

2.5.3. Biodegradation

Starch based polymers are biodegradable under various conditions, including industrial composting, following the principal international standards described in the previous sections. They can also biodegrade in home composting and soil environments, depending on the specific chemical composition of the final material produced from the TPS. Additionally, some materials in this category have the capability to biodegrade in fresh or marine water.

2.5.4. Challenges to overcome

To address a wider range of applications, the mechanical and technical properties of starch-based polymers need to be improved. At the same time, new processes must be developed to replace the raw materials derived from fossil sources, which are used in conjunction with TPS processing, with agricultural ones or, even better, with second generation feedstocks or organic wastes. New different polymers and compounds will require an increase of efforts to fully satisfy the end-of-life properties (biodegradability, ecotoxicology and more) in diverse and less common natural environments to ensure that accidental dispersion (littering) of biodegradable items will not represent an environmental issue.

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3. RESULTS FROM COMPLEMENTARY R&D PROJECTS

3.1. PROJECT ESTELLA: Lignin-based epoxy resins [55]

- Material Overview: The bio-based epoxy resin is produced using lignin and epoxide chemicals. The epoxy resin is produced from renewable resources such as sawdust.
- Applications: The epoxy resins are under test for window frames and scooter platforms (ex., scooter decks).
- Biodegradation: The biodegradation is under study.
- Challenges to overcome: The challenge to overcome is the development of a chemical recycling process for epoxy resins to extend the life cycle of the material.

3.2. PROJECT GREEN-LOOP: Lignin as an additive for improved flame retardancy [56]

- Material Overview: The main role of lignin in product formulation is to improve the fire behaviour of the end product. The most important characteristics of kraft lignin (and thus a potentially renewable material for the development of flame-retardant lignin-rubber composites) are its high thermal stability, its ability to form char and its aromatic structure with numerous reactive hydroxyl groups.
- Applications: Lignin-enriched rubber composites could be used in construction industry in a variety of intended uses in walls or floors. The identified intended uses are: i) flooring for use indoors, ii) wall lining for use indoors, iii) roofing and iv) wall covering for use outdoors; these are often made as continuous layer extending over the roof.
- Biodegradation: The biodegradation is under study.
- Challenges to overcome: The challenge to be overcome is the compatibility of lignin with the components of the composite formulation based on recycled rubber in order to improve the mechanical properties of the material

3.3. PROJECT PROPLANET: Natural biopolymers and synthetic coating compounds [57]

- Material Overview: Textile coatings from different natural polysaccharides (agar, alginate, chitosan, cellulose etc.) with the addition of natural/modified natural/synthetic compounds, e.g. alkyl ketene dimer (AKD), starch, modified starch, wax.
- Applications: Application on various textile substrates with the aim to modify the wettability of the material on different liquids.
- Biodegradation: Biodegradation studies not performed yet.
- Challenges to overcome: Achieving durability to washing, oleophobic.

3.4. PROJECT REMEDIES: Natural biopolymer-based coatings for zero-waste cosmetics [58]

• Material Overview: Coating as water soluble packaging to protect the dewatered shampoo and shower gel formulations consist of alginate, stearic acid, glycerol, ethyl alcohol. The product is a biopolymer-based solutions, where the dewatered cosmetic product in form of balls is placed to create a coating.

- Applications: Zero-waste cosmetics involving shampoo, soap, shower gel, conditioner, as a plastic or other material type packaging replacement.
- Biodegradation: Biodegradation of the seaweed-based material in the form of foils was performed in soil and wastewater treatment plant active sludge and reveal biodegradation using OxiTop[®] for the natural biopolymer coating (foil) in 120h (5 days).
- Challenges to overcome: Lower the water vapor transmission rate through the coating, upscaling the technology, and upgrading to other applications beyond cosmetics.

3.5. PROJECT BRILIAN: Circular Future for rural areas [59]

- Material Overview: thermoplastic starch coming from rejected potatoes, blended with other bio-based polymers such as PBS, PLA and PBAT; biopolyesters derived from bio-based building blocks from low-input oleaginous crops; proteins from rapeseed cake.
- Applications: Bio-based products for agriculture, shrink film, bio-based cosmetics ingredients; vegan proteins; and bio-based adhesives.
- Biodegradation: in soil biodegradation, industrial composting.
- Challenges to overcome: mechanical properties of the shrink film (flexible and resilient), end users' acceptance towards innovative products that may have higher prices compared to conventional alternatives.

3.6. PROJECT NAUTILUS: Bio-based polymers for antifouling paint formulations [60]

- Material Overview: biobased alternative polymers such as PLA and PHA.
- Applications: antifouling paint formulations to be applied in marine vessels.
- Biodegradation: The biodegradation of the developed biopolymers is under study for freshwater environments as well as the biodegradation of the final paint formulations in marine environments.
- Challenges to overcome: the development of antifouling paints using biobased polymers as a matrix while maintaining the properties of the paint at the same level as the acrylic-based ones; enhancing their antifouling capacity while reducing the biocide used. Other additional challenges: the solubility of the biopolymers and the viscosity required to obtain the final paint formulation characteristics; the durability of the paint; the scalability of PHA production and the associated costs.

3.7. PROJECT ATLANTIDA: Biodegradable alternatives for microplastics intentionally added in detergent and cosmetic industrial applications [61]

- Material Overview: biodegradable polymers with rheological and opacity properties, biodegradable filmformers, biodegradable microcapsules with abrasive and exfoliating properties and biodegradable microcapsules with fragrance, bio-based packaging and biodegradable detergents.
- Applications: biodegradable microcapsules and polymers for the detergent and cosmetic industry.
- Biodegradation: The biodegradation of the developed biopolymers and the biobased alternatives is under study for freshwater environments.
- Challenges to overcome: the development of complex matrices (biopolymers and microcapsules) while maintaining the properties compare to the fossil-based ones; the solubility of the biopolymers and the viscosity in the final formulations; providing and demonstrating the added value to the final product.

4. CONCLUSIONS

Bio-based plastics and biopolymers, particularly those derived from renewable resources like PBS, PLA, PBAT, TPS, and PHA, have emerged as promising alternatives to conventional plastics:

- PLA (Polylactic acid): Derived from renewable resources like corn starch, PLA is biodegradable and compostable (mainly at industrial level), making it suitable for packaging, rigid items (cutlery), textiles, and 3D printing.
- PBAT (Polybutylene adipate terephthalate): Depending on the monomer's origin, it can be varied the renewable carbon content promoting the production of high renewable PBAT. It offers good flexibility and composability, finding applications in flexible packaging and agricultural films. Combining PBAT with other bioplastics (i.e. PLA) is possible modify the mechanical properties and increase the possibility of final use.
- PBS (Polybutylene succinate): Known for its good mechanical properties, processability and biodegradability, PBS is used in various applications, including packaging, automotive parts, and medical devices.
- PHA (Polyhydroxyalkanoates): Produced by fermentative microbial route, PHAs have a wide range of properties, including biodegradability and biocompatibility, making them suitable for medical implants and packaging. Very high production costs and difficulties related to the industrial scale production processes, make challenging the competition over conventional plastics
- Starch-based biomaterials: Starch, a natural polymer, can be processed into various bioplastics, offering biodegradability and composability. The resulting biobased thermoplastic starch (TPS) blends have enhanced properties including strength resistance and rigidity becoming relevant for industrial applications.

These materials offer several advantages, including:

- Sustainability: Bioplastics are produced from renewable sources, reducing dependence on fossil fuels and mitigating environmental impacts associated with traditional plastic production.
- Biodegradability: Many bioplastics can biodegrade under specific conditions, minimizing environmental pollution derived by fragmentation of conventional plastics (microplastic production).
- Compostability: Certain bioplastics can be processed to produce items (i.e. compostable waste bags) that can be used as a tool to promote the correct management of organic waste (from the collection to the valorisation into compost), returning valuable nutrients and carbon to the soil (compost).
- Versatility: Bioplastics have a wide range of applications, from food packaging to waste bags including medical implants, offering high versatility in various industrial sectors.

While bioplastics offers immense potential, there are still opportunities to optimize their performance and expand their applications:

- Cost: Currently, bioplastics production is generally more expensive if compared to conventional plastics.
- Performance: In some applications, bioplastics may not yet match the performance of conventional plastics.
- Infrastructure: The infrastructure for the production and processing of certain bioplastics (i.e. PHA) is still in development.

Future research and development efforts should focus on:

• Cost reduction: Developing more efficient and cost-effective production processes.

- Performance enhancement: Improving the mechanical properties and processing characteristics of bioplastics.
- Expanding applications: Identifying new applications for bioplastics in various industries.
- Life cycle assessment: Conducting comprehensive life cycle assessments to evaluate the overall environmental impact of bioplastics comparing it with the production of conventional fossil plastic polymers (including their end-of-life management).

In conclusion, bioplastics offers a sustainable and environmentally friendly alternative to conventional plastics. Continued research and development efforts will help to overcome current challenges and accelerate their adoption in various sectors.

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