

## Bioplastics – a green choice for sustainable development

Nhựa sinh học - một giải pháp "xanh" cho phát triển bền vững  
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### Abstract

Nowadays, the overuse of conventional plastics has led to significantly negative impacts on the ecological systems. Hence, bioplastics have emerged as a visible solution to diminish such harmful effects caused by conventional plastics. This mini-review article briefly introduces and classifies main types of bioplastic materials. The understanding of bio-based content as well as biodegradability is also addressed. Lastly, three main classes of bio-based, biodegradable plastics which are thermoplastic starch (TPS), polylactic acid (PLA) and polyhydroxyalkanoate (PHA), are reviewed in terms of structure, properties, processing and applications.

**Keywords:** Bioplastics; bio-based; biodegradable; starch; polylactic acid; polyhydroxyalkanoate.

### Tóm tắt

Ngày nay, việc sử dụng quá nhiều các loại nhựa truyền thống đã dẫn đến những tác động tiêu cực to lớn đến hệ sinh thái. Vì vậy, nhựa sinh học đã dần trở thành một giải pháp hữu hiệu để giảm bớt những tác động tiêu cực đến từ nhựa truyền thống. Báo cáo tổng quan này sẽ giới thiệu ngắn gọn và phân loại các loại nhựa sinh học chính. Ngoài ra, phương pháp xác định hàm lượng gốc sinh học cũng như khả năng phân hủy sinh học cũng sẽ được đề cập. Cuối cùng, bài báo sẽ phân tích ngắn gọn về cấu trúc, tính chất, quy trình tổng hợp, gia công cũng như ứng dụng của ba nhóm chính của nhựa nguồn gốc sinh học và có khả năng phân hủy sinh học: nhựa nhiệt dẻo từ tinh bột (TPS), polylactic acid (PLA) và polyhydroxyalkanoate (PHA).

Từ khóa: Nhựa sinh học; gốc sinh học; phân hủy sinh học; tinh bột; polylactic acid; polyhydroxyalkanoate.

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## 1. Introduction

Over recent decades, plastics are prized for their lightness, durability, functionalities, versatility of designs and the ease of manufacturing [1]. With their intrinsic properties and a vast number of types, plastic materials have been widely applied in building and construction, electronics, automotive design, packaging, paints and coatings. Therefore, since the mass production of plastics began in 1950, there has been an 8.4% of compound annual growth rate in their production. Globally, an estimated 8.3 billion metric tons of plastics has been manufactured [2]. Over the years, expanding use of plastics has brought significantly negative consequences to the environment. It has been estimated that 348 million tons of plastics produced annually with only 7% is recycled, leaving the remaining of 93% dumped into oceans, landfills or subjected to incineration in municipal plants [3]. Indeed, of the mentioned amount of produced plastics, 40% is for single-use products, which are mainly composed of polyethylene (PE), polypropylene (PP) and polyethylene terephthalate (PET). It could be noticed that the persistence of plastics in the environment for up to a century causes contamination of marine and terrestrial organism as well as marine and sea-based food, which lead to the loss of biodiversity, living habitat and adverse health effects. Considering the side effects of micro- and nanoplastics, the leaching of toxic pollutants from fragmented plastic is assumed to negatively impact the environment and may affect the biological function of organisms. In fact, exposure to microplastics due to fragmentation of macroplastics or as a part of certain products could result in the wide spread of harmful pathogens and toxins in environment and the harassment of natural resources. Records show

that from 267 to 557 species of animals, including birds, fish, turtles, and marine mammals, have ingested plastic debris and been affected by ingestion and entanglement of marine litters [4]. In addition, cell damage, endocrine and metabolic disruption in human health are due to the ingestion (including marine food) and inhalation of microplastics particles and toxins. Recent studies suggest the possibility of micro- and nanoplastics penetrating secondary tissues, such as liver, muscle, and brain, attacking the immune system, causing immune toxicity, and triggering adverse effects like immunosuppression and abnormal inflammatory responses [5]. Another important concern with the explosive growth of plastics consumption is that the fossil source is finite while plastics sector would be account for 20% of total oil consumption by 2050 (World Economic Forum) [6]. Fossil resources require millions of years to generate while human consumes in the level of centuries.

To address the abovementioned problems caused by conventional plastics, bioplastics have been investigated and applied gradually due to their numerous environmental benefits. Bioplastics are bio-based and/or biodegradable plastics, which are usually derived from renewable or biomass source, such as jackfruit, waste banana peels, organic waste, agriculture waste, newspaper waste, oil palm empty fruit bunch, sugar cane, corn starch, potato starch, rice straw, rapeseed oil, vegetables oil, cellulose from plants, cotton, and bacteria. Bioplastics have been used in an increasing number of markets including packaging, catering products, consumer electronics, automotive, agriculture/horticulture, toys, and textiles. A vast numbers of bioplastics have been commercialized and new ones are constantly emerging the market. Among numerous types which have been introduced so

far, the most common bioplastics with largest market share includes the bio-based polyethylene terephthalate (bio-PET), bio-based polyamides (bio-PA), bio-based polyethylene (bio-PE), starch blends, and polylactic acid (PLA) (**Figure 1**). As a part of “green economy” that aims to reduce environmental risks and achieve sustainable development, replacing traditional plastics by the more eco-friendly bioplastics undoubtedly would play a significant role. Currently, major plastics manufacturers have involved into industrial-scaled production of bioplastics. The main players in bioplastics industry include BASF, NatureWorks, Arkem, Novamont, Plantic, DSM, DuPont, Evonik Industries, Ecospan, Toray Industries and so on.

## 2. Classification of bioplastics

Bioplastics could be classified based on their original sources and biodegradability [7]. In terms of original source, “bio-based plastics” are plastics that derive from plant or biomass, which are non-fossilized and biodegradable organic materials that originate from plants, animals and micro-organisms. As a result, bio-based plastics can provide intrinsic zero “material carbon footprint”, which means that CO<sub>2</sub> releasing after decomposition would be consumed by the biomass plantation. Besides, “biodegradable plastics” are designated for plastics that undergo complete decomposition induced by micro-organisms in composting or anaerobic digestion processes within pre-determined time period. Hence, considering two terms simultaneously, there are three groups of bioplastics besides the conventional petro/fossil-based and non-biodegradable

plastics. The first group includes bio-based (or partly bio-based), non-biodegradable plastics, such as bio-PE, bio-based polypropylene (bio-PP), bio-PET, bio-PA, bio-based polytrimethylene terephthalate (bio-PTT) or bio-based polyurethanes (bio-PU). These bio-based plastics are synthesized from monomers which are derived from renewable resources e.g. starch, cellulose, oils, lignin etc. It should be noted that these bio-based polymers can be used to create plastic products that behave like conventional plastics. The second group is fossil resources-originated biodegradable plastics. This group consists of biopolymers based on polycaprolactone (PCL), polybutylene succinate (PBS) and polybutylene adipate terephthalate (PBAT). These polymers are normally used in combination with starch or other bioplastics to improve the mechanical properties as well as biodegradability. The last bioplastics group contains bio-based, biodegradable plastics. This is the most desirable group of bioplastics with main examples including PLA, polyhydroxyalkanoate (PHA) and thermoplastic starch (TPS). **Figure 2** describes the life-cycle of these bioplastics. First, the bioplastics are manufactured from raw materials that derived from renewable biomass feedstock. Subsequently, recycle or non-recycle products are made from these bioplastics. After service, the products undergo biodegradation process and release CO<sub>2</sub>, H<sub>2</sub>O and biomass, which would be consumed in photosynthesis for the plant growth. In this review, we focus to this bioplastics group, which are the most suitable candidates for the “green” concept of plastics.

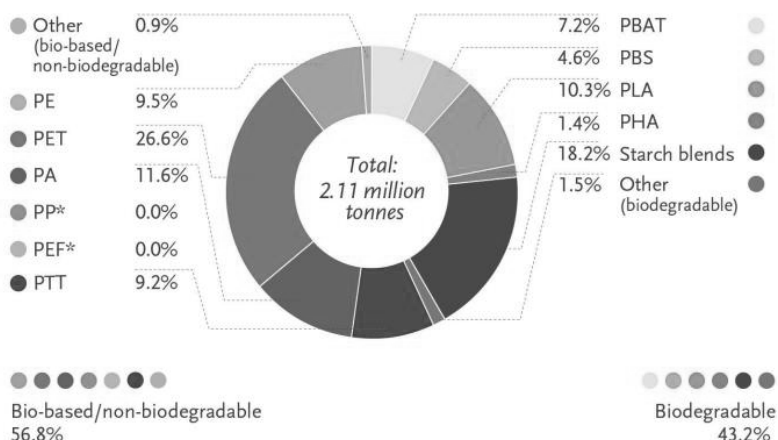


Figure 1. Global production capacities of bioplastics 2018 by material types (reproduced from [8])

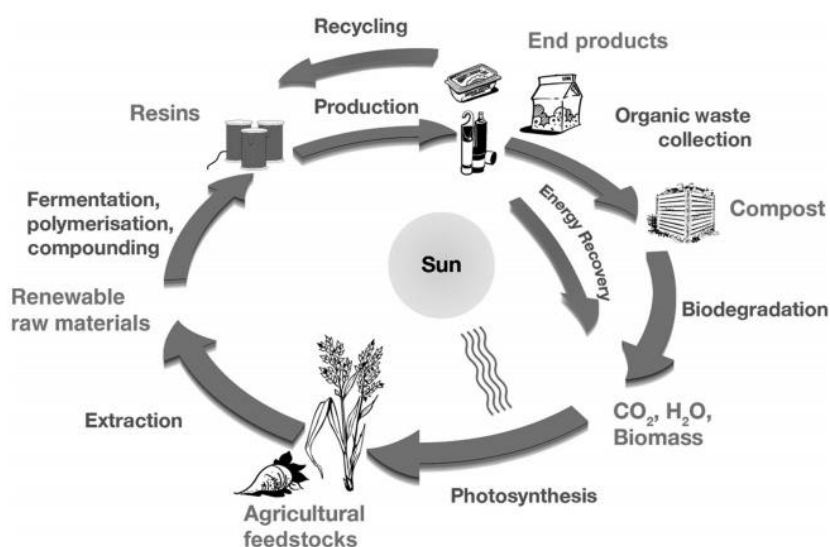
### 3. Bio-based content and biodegradation

To prevent misleading and false claims from some companies in bioplastics market, we should have a clear understanding about bio-based content and biodegradation, which are two pivotal parameters of bioplastics. From abovementioned point of bio-based origin of bioplastics, a transparent and accurate test procedure is on demand for determining “material carbon footprint” reduction degree obtained by using bio-based plastics. Generally, ASTM D6866 is widely used in bioplastics industry to measure the bio-based content of solid, liquid and gaseous samples using radiocarbon dating. In contrast to fossil sources, the biomass sources contain a well-characterized amount of carbon isotope ( $^{14}\text{C}$ ), which is quantified by ASTM D6866. The test protocol involves the combustion of sample to obtain  $\text{CO}_2$ , which then will be analyzed to measure  $^{14}\text{C}/^{12}\text{C}$ . The relative ratio of  $^{14}\text{C}$  on total organic carbon content can indicate the bio-based content of examined sample. If the bio-based origin is considered for the “begin-of-life” of bioplastics, thus biodegradability is representative for the “end-of-life” of bioplastics. Under the excretion of depolymerase enzymes from bacteria, fungi, algae, and protozoa in selected environments,

the bioplastics are degraded and converted into  $\text{CO}_2$ , water, mineral salts, and biomass. Such disposal environments could be under composting conditions, anaerobic conditions, soil conditions, or marine conditions [6]. In the current status of biodegradation standards, the most important norm for definition of compostable bioplastics is the European Standard EN 13432. The standard requires at least 90% disintegration of the examined samples after 12 weeks, 90% biodegradation ( $\text{CO}_2$  evolution) in six months, and includes the tests on ecotoxicity and heavy metal content. It has been the standard for biodegradable packaging designed for treatment in industrial composting facilities with enhanced temperature, presence of moisture and anaerobic digestion. There is currently no international standard specifying the conditions for home composting of biodegradable plastics. Nevertheless, home compostability labels generally require at least 90% degradation in 12 months at ambient temperature. For biodegradation in soil, the TUV Austria-Belgium certificate adapted from EN 13432 standard requires at least 90% biodegradation in two years at ambient temperatures after burying in the selected soils. Lastly, there have been several standards

related to biodegradation of plastics in marine conditions, however, no obvious requirements for conditions or timeframes are given. There is a certification that demands a biodegradation degree of at least 90% in six months after immersing samples in sea water. Besides the “real” biodegradable plastics, the formulations of conventional synthetic polymers such as PE, PP, PET, with the presence of metal-based catalysts are sometimes claimed to be

“biodegradable”. In fact, those plastics belong to “oxo-fragmentable” plastic class that employs inorganic metal salt additives to accelerate the decomposition process. However, it was concluded that oxo-degradable plastics do not satisfy the industrial/home composting biodegradation requirements [9]. Therefore, bioplastics consumers should be aware of this fact to distinguish oxo-degradable plastics from bioplastics.



**Figure 2.** Life-cycle of bioplastics (reproduced from [1]).

## 4. Bioplastics materials

In this section, we discuss TPS, PLA and PHA, as the most important candidates for bio-based and biodegradable bioplastics [1]. Particularly, starch is the dominant raw material for bioplastics that directly extracted from biomass, among other polysaccharides and proteins such as cellulose, chitin/chitosan, collagen, wool, soy, and gluten. PLA is the most widely used synthetic bioplastics which are synthesized from bio-derived monomers, besides bio-PE and polyglycolic acid (PGA). PHA is the representative for emerging bioplastics which are produced directly by natural/genetically modified organisms. The structure, properties, processing and applications of these bioplastics are addressed in following sections.

### 4.1. Starch

Starch is one of most promising raw materials for bioplastics due to its abundant and cheap resources, and biodegradability. Starch exists as granular form and is a carbohydrate polymer of D-glucose organized in two major constitutes including 20-25% amylose – a linear or sparsely branched polymer with molecular weight of 0.2-2 million and 75-80% amylopectin – a highly multiple-branched polymer attached on amylose starch with much higher molecular weight of 100-400 million [10]. Worldwide, main sources of starch include corn (82%), wheat (8%), potatoes (5%), and cassava (5%). Thermoplastic starch (TPS) is termed for destructured starch that possesses processing ability similar to

conventional thermoplastics. The native starch is hardly usable due to its brittleness and melting point higher than thermal decomposition temperature. Hence, with the applications of heat, mechanical shear force and/or the addition of plasticizers, in the presence of water, its granular state is disrupted, its native crystalline structure is disappeared and the melting temperature is lowered down below the degradation temperature, allowing conventional processing such as extrusion, injection molding, blow molding, and injection compression molding [11]. A suitable plasticizer should be small molecule, polar, hydrophilic, highly compatible, has high boiling point, and should be added at an appropriate concentration depending on desired properties. Water and glycerol are considered to be most effective plasticizers among a vast range of plasticizers including sucrose, fructose, glucose, glycols, urea, amides and, amino acids [12], [13]. Properties of interest from TPS including glass-transition temperature, tensile properties, gas barrier properties, moisture sorption, retrogradation, and surface tension, are governed by starch sources, additives, processing parameters, and chemical treatment [11]. Generally, TPSs can be produced in form of pellets thus then can be utilized for extrusion machine. TPS pellets melt and travel from the hopper, through the screw, to the barrel chamber and finally reach the die before extruding out. After being blown or molded into designed shape, TPSs will be solidified. TPS containing at least 70 wt% starch can be extruded into sheets for thermoforming of food packaging trays. Other components of TPS could be plasticizers, polymeric property modifiers and fillers.

Besides TPSs, starch can also be blended with other bioplastics to obtain starch-

complexes with various functionalities and/or reduced cost. In blending formulations, starch acts as a weak link in the matrix and provides sites for biodegradation attack at lower starch concentration (i.e. < 60%). At concentrations exceeding 60%, significant enhancement in biodegradation occurs. Hence, starch-based materials are suitable for applications that required the rapid biodegradation and minimized dispersion to environment. Numerous starch-based blending systems with polycaprolactone (PCL), polyvinyl alcohol (PVOH), polybutylene succinate (PBS), polybutylene succinate adipate (PBSA), ethylene-acrylic acid copolymer (EAA), ethylene-vinyl alcohol copolymer (EVOH), polyolefin, and aliphatic-aromatic polyester have been reported [14]–[16]. Among them, the combinations of starch with biodegradable polymers such as PCL, PLA, PBS or PBSA are of interest for bioplastics to maintain biodegradability [17]. Particularly, blending starch with the aliphatic polyester PCL provided dimension stability and reduced water sensitivity while adding PLA increased water adsorption resistance and mechanical strength [18], [19]. Additionally, the blending with PBS or PBSA improved the mechanical and rheological properties compared to parental starch [20]. Furthermore, starch-complexes are amongst the most commercially exploited bioplastic materials, on the basis of their cost reduction and functionality enhancements. These starch-based complexes have found wider industrial applications ranging from extrusion, injection moulding, blow moulding, film blowing and foaming. Finally, starch nanocomposites have been developed by incorporating inorganic additives, such as clay, into starch matrix in order to enhance mechanical and barrier properties [21]–[23]. Currently, TPSs have been commercialized for

drug delivery capsules, golf tees, cutlery, plates, and food containers. To overcome intrinsic limitations of TPSs including low mechanical properties, poor water vapor resistance and retrogradation, more studies in modification and optimization of starch-based formulations with other polymers, nanoclays, fibers, should be carried out [17].

#### 4.2. Poly (lactic acid)

Poly(lactic acid (or polylactide) has gathered significant attention in environmental, biomedical and pharmaceutical applications as well as bioplastics for everyday consuming goods. As one of the biggest alternatives to petro-based polymers, PLAs are usually made from starch/sugar-rich crops such as corn, potatoes, beets, sugar canes, but it can also be derived from petrochemicals. However, for bioplastics concept, the corn-derived carbohydrates such as glucose are most frequently used as raw material for PLAs. From carbohydrate sources such as corn, glucose is produced and lactic acid is synthesized by bacterial fermentation of resulting glucose. Afterward, PLAs can be synthesized by polycondensation of lactic acid or ring-opening polymerization (ROP) of lactide, resulting in three typical optical isomeric forms, which are poly(L-lactide) (PLLA), poly(D-lactide) (PDLA), and poly(DL-lactide) (PDLLA) [24]. While condensation polymerization results in low molecular weight PLAs with increased susceptibility to biodegradation, ROP-synthesized PLAs usually possess high molecular weight with low susceptibility to microbial attack. Moreover, ROP-synthesized PLAs also exhibit enhanced mechanical properties, higher glass transition temperature and degradation temperature [25]. In final PLA structure, the ratio and sequences of D- and L-units drastically affect the crystallinity, thermal and mechanical properties. Nevertheless,

among three optical isomeric forms, PLLA is the most widely manufactured material due to the low production cost. PLA-based bioplastics can be manufactured with the same processing for petro-based polymers. However, the processing temperature should be low and processing time should be short, to prevent any premature thermal degradation. Moreover, the water absorbance should be minimized in prior to processing. In terms of properties, PLAs provide high transparency, surface gloss, and good chemical resistance to fats and oils [1].

Crystalline PLLA or PDLA have mechanical properties comparable with synthetic polymer such as polystyrene. Stereocomplexation between PLLA and PDLA can improve overall mechanical strength of PLA [26]. Degradation of PLAs relies on the hydrolytic cleavage of ester bonds and the enzyme-catalyzed decomposition. Chemical hydrolytic degradation of PLAs proceeds via bulk, core-accelerated or surface-erosion, regarding to the thickness of the materials. On the other hand, microbial biodegradation of PLAs is considered to be rather slow, which normally takes 508 months to complete degrade PLLA into lactic acid. Mechanical properties as well as degradation behavior of PLAs can be tuned by controlling molecular structure, higher ordered structures, material morphology, blending, or surface treatment. For final use as bioplastics, current limitations of PLAs including low mechanical properties at temperature excess glass transition temperature ( $\sim 55^{\circ}\text{C}$ ) and slow degradation should be solved. Blending PLA with fast biodegradable polymer such as starch could improve the degradation rate of PLA. To improve thermal/hydrolytic degradation resistance of PLA, crystallinity should be increased during processing by the use of nucleating agents or crystallization-accelerating agents such as talc,

montmorillonite, nanostructured carbon, amide, hydrazide, other biodegradable polymers or even PLA stereocomplex [27]–[33]. Moreover, composites or nanocomposites of PLA could be a visible option to modify the mechanical performance and barrier properties for industrial applications. A variety of PLA-nanocomposites using layered silicate, sepiolite, halloysite nanotube, nanocellulose, carbon nanotube, graphene, silver nanoparticles, zinc nanoparticles, titanium oxide nanoparticles, layered double hydroxide nanoparticles, silica, POSS, and Fe<sub>3</sub>O<sub>4</sub> were reviewed elsewhere [34]. Besides, PLA-based composites reinforced by natural fibers as well as blending system with other polymers such as starch, glucosamine, PHB, spidroin, soy protein have been reported [35]–[40]. Lastly, beside mechanical properties, stereo-complexation of PLA can also be deployed to enhance the hydrolytic and thermal degradation resistance, or gas barrier properties [26]. Currently, industrial PLAs production has mainly focused on alternating petro-based polymers such as PP or PET for packaging material, automotive materials, and chassis for electrical appliances. Future investigations and optimizations of PLA-based bioplastics should be devoted to some challenges such as reducing production cost, enhancing mechanical properties as well as hydrolytic/thermal degradation resistance, and reducing the utilization of edible raw materials.

#### 4.3. Polyhydroxy alkanates (PHA)

PHAs are the most widely investigated and applied polyesters that synthesized within bacteria and produced by bioengineering. PHAs are linear polyesters comprising hydroxyl acid monomers connected by ester bonds. Due to the range of possible monomers that can be polymerized *in vivo* by bacteria, the microbial

PHA family allows the versatility to adjust mechanical properties for specific applications [41]. The variability in structure and extensive material properties compared to other bioplastics such as PLA or TPS have made PHAs the most potential materials for future bioplastics. Since the first discovery of poly(3-hydroxybutyrate) P(3HB) in *Bacillus megaterium* by Maurice Lemoigne in 1925, PHA family has broadened with a vast range of more than 150 monomers and molecular masses ranging from 50,000 to 1,000,000 Da [42]. A broad range of microbial genera including gram-positive eubacteria, gram-negative eubacteria, archaea, and cyanobacteria with over 300 species can biosynthesize PHAs. Firstly, the biosynthesis of PHAs starts from prized substrates of high nutritional value such as glucose, starch or edible oils. Under unfavorable growth conditions with imbalanced nutrient supply, the bacteria can accumulate the polymers in their cytoplasm as an intracellular carbon and energy storage compound, in the shape of granules. The biosynthesis of PHA pathways is catalyzed by the enzyme PHA synthases, which bound to the surface of PHA granules [43]. Structure and properties of PHAs rely on their monomer content, which could be controlled by type of micro-organisms, media ingredients, fermentation conditions, modes of fermentation, and recovery process [44]. As PHAs are stored within the cells, several methods should be applied to recover the polymers, including solvent extraction, floatation, digestion, and supercritical fluid extraction [45].

Generally, depending on the number of carbon atoms in the monomers, PHAs can be classified into three groups with short-chain-length (scl-PHA, 3-5 carbon atoms), medium-chain-length (mcl-PHA, 6-14 carbon atoms) and long-chain-length (lcl-PHA, more than 14



carbon atoms) PHAs. Among the three groups, lcl-PHAs are uncommon and less studied. 3-hydroxyvalerate (3HV) and 3-hydroxybutyrate (3HB) are representatives for short-chain PHAs while 3-hydroxyhexanoate (3HHx), 3-hydroxyoctanoate (3HO), 3-hydroxydecanoate (3HD) and 3-hydroxydodecanoate (3HDD) are examples for medium-chain PHAs. The final composition of PHAs is determined by the carbon substrates and the bacterial strain used. PHAs can be polymers, random copolymers or block copolymers. The side chain can be saturated or not, can be branched or aromatic, can be halogenated or epoxidized. A variety of functional groups can be introduced to PHA side chains via chemical modification, enabling the easy and precise modulation of polymer structure to yield desirable properties [46]. Moreover, physical modification via blending with a range of natural polymers such as starch, lignin, cellulose derivatives or synthetic polymers such as PLA and PCL could be considered as a simple yet effective approach to tailor the properties of PHAs [46]. PHB, the most used PHA, possesses good resistance to moisture, excellent gas barrier properties, good hydrolytic degradation resistance, good UV resistance, solubility in chloroform, biocompatibility and biodegradability. Besides, the thermal and mechanical properties vary depending on the particular structure and composition of PHAs. Although extensive range of PHAs have been produced but low productivity and high cost still remain as the big challenges for industrial applications of PHAs. Only P(3HB), poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (P(3HB-co-3HV)) and poly(3-hydroxyhexanoate-co-3-hydroxyoctanoate) (P(3HHx-co-3HO)) have relatively high productivity, however, the production cost still approximately 15 times more expensive than petro-based polymers such as polypropylene

(PP) [47]. In order to reduce the production cost of PHAs, the cost of carbon substrates for microbial growth should be cut down, the downstream process should be eco-friendly and cost-effective, and the bacterial growth condition should be optimized [48]. Addressing these challenges, the use of agro-industrial wastes as carbon feedstock materials could be a viable strategy to cut down the raw material price for industrial production of PHAs. Thereafter, the use of thermo-separating polymers like ethylene oxide or propylene oxide in aqueous two phases extraction process (ATPE) could be an eco-friendly, economical and industrially viable downstream process for isolation and recovery of PHAs. Lastly, genetically modified gram-positive bacteria can be considered as a promising approach to enhance accumulation of a single structure of PHA, reduce the cost of PHAs production for industrial scale but still maintain medical grade quality of produced PHAs.

## 5. Conclusion

Recently, bioplastics have emerged the plastics market as a promising green solution to reduce the dependence on cost-rising sources of fossil or petroleum, the CO<sub>2</sub> emission and the harmful environmental consequences caused by the overuse of conventional plastics. Among numerous bioplastic material types that have been introduced and commercialized, TPS, PLA and PHA are the representatives for fully bioplastics which are extracted/synthesized from renewable resources and possess biodegradability after product use. However, several big concerns should be taken seriously for the future development of these bioplastics. Those are the weak mechanical properties and low water resistance of TPS, hydrolytic/thermal degradation and slow biodegradation of PLA, and very high production cost of PHA. Besides

blending and compositing of these bioplastics with other materials, we believe that the optimization of material/process or the utilization of novel cost-effective processes as well as cheap alternative feedstock materials would also positively contribute to solve the abovementioned challenges. Furthermore, new material platforms that generated from agro-industrial wastes with controlled structure and well-tuned properties would be of great interest to bring bioplastics closer to a promising and sustainable future.

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