Polyhydroxyalkanoates and Poly(lactic acid)'s Biodegradation

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The synthesis process for two types of biopolymer: PHAs and PLA. Synthesis of PHAs involves bacteria fermentation and extraction procedure. PLA is chemically synthesised via condensation polymerization of lactic acid (LA).

Keywords: polyhydroxyalkanoates ; poly(lactic acid) ; PHA-based composites ; PLA-based composites ; biodegradation

1. Introduction

Plastic pollution has been one of the major concerns in environmental issues due to the massive production of single-use plastic driven by consumer demand, inevitably producing voluminous plastic waste. Petroleum-based plastics are non-biodegradable, and most of the plastic wastes are discarded to the environment or accumulated in landfills. Conventional petroleum-based plastics are recalcitrant to microbial degradation and accumulate in the environment and food chains [1]. Moreover, incineration of petroleum-based plastic waste for energy generation leads to the release of greenhouse-related and toxic gases, namely dioxin, furans and polychlorinated biphenyls. On the other hand, recycling can serve as a progressive alternative to manage plastic waste; however, recycling only works to limited extent, owing to the complex composition of plastic, types of plastic, material fatigue and quality loss with every recycling process. It is estimated that 9% of the plastic waste has been recycled [2][3]. Furthermore, 99% of the plastic was derived from non-renewable fossil hydrocarbon (petroleum and natural gas) [4].

2. Polyhydroxyalkanoates (PHAs) and Poly(lactic acid) (PLA): Overview and Synthesis Process

2.1. Synthesis of PHAs

2.1.1. Fermentation

PHA is a bio-based polymer synthesized by various bacterial cells such as *Cupriavidus necator* and *Bacillus* sp. under specific growth conditions such as excess carbon sources and limited nitrogen sources [5][6]. PHAs were first discovered by Lemoigne in 1925 and are a family of naturally occurring biopolyester synthesized intracellularly by a great number of prokaryotes as storage polymers for carbon and energy sources [7][8]. Generally, PHAs can be synthesize via three pathways by utilization of various carbon sources: Pathway I is used predominantly in poly(hydroxybutyrate) (PHB) PHB-producing organisms, such as *C. necator* and *Bacillus* sp., while Pathways II and III are present in mcl-PHA producing *Pseudomonas* sp. [9][10][11]. Different monomers can be produced from various bacterial strains with different types of carbon substrate of the microorganisms and, therefore, co-polymer ratios can be tailored depending on its desired final product applications [8][12].

Research explores the utilization of various waste biomass as carbon sources for production of value-added products such as PHAs [13][14][15]. The aforementioned waste biomass requires pre-treatment to increase the corresponding monomer units for the ease of bacteria consumption. For example, molasses contain high amount of sucrose required to be hydrolyzed into its monomers glucose and fructose. An increment of 15% was observed for glucose and fructose content upon hydrothermal acid pretreatment of molasses [15].

Cupriavidus necator, formerly known as *Ralstonia eutropha*, is among the most studied PHB-producing strain due to its high PHB yielding capacity and it is mainly exploited at industrial scale [16][17]. By using a pure culture, this strain is able to accumulate intracellular reserves of PHB up to 80% of its cell dry weight (CDW) by consuming a broad range of carbon sources [17][18]. Typically, the molecular weight of PHB produced by wild-type bacteria is between 1.0×10^4 to 3.0×10^6 with a polydispersity index of about 2.0 [19]. To some extent, researchers have explored modifying the genetics of organisms to further enhance the PHAs with co-polymer production. High yield production of PHAs was reported by Kahar

et al. with the utilization of recombinant strain PHB⁻4/pJRDEE32d13 (a PHA-negative mutant harboring *Aeromonas caviae* PHA synthase gene, $phaC_{Ac}$) containing 5 mol% (*R*)-3-hydroxyhexanoate, P(3HB-*co*-5 mol% 3HHx), DCW of 128–138 g/L and a high PHA content of 71–74% (*w*/*w*). By utilizing wild-type strain *Ralstonia eutropha* H16, the CDW of 118–126 g/L and a high PHB content of 72–76% (*w*/*w*) were recorded [20]. The accumulated PHAs are then subjected to extraction procedures.

2.1.2. Biological Extraction Method of PHAs

Extensively used chemical extraction methods of PHAs in industrial applications have caused a downside such as increased production cost, degradation of polymers, and environmental issues upon disposal. Hence, more attention and initiatives are being given to the biological extraction method to overcome these drawbacks. The idea of bio-extraction involves the use of living organism to extract the polymer from the cells.

There are several biological approaches that have been investigated. For example, by feeding the cells containing intracellular PHAs to the animals and insects [21][22][23][24]. Chee et al. demonstrated that up to 90% of PHAs can be recovered upon feeding the insects. The undigested biopolymer was excreted as fecal pellets. Further simple purification that does not involve hazardous chemicals are required to remove the impurities from the fecal pellets which will result in PHAs that have similar properties with the solvent-extracted ones [22][24].

Kunasundari et al. [21] has patented the use of Sprague Dawley rats to partially purify the PHB granules. The rats were given freeze-dried cells of *C. necator* H16 as a sole diet source for 7, 14, and 28 days. The mortality rate is zero throughout the study. The rats with bacterial cells diet produced low-odor white fecal pellets with PHA content of 87–90 wt%, while rats in the control group excreted normal blackish fecal pellets and no PHA was detected. The melting temperature (T_m) and the degree of crystallinity (X_c) of the biological recovered polymer obtained is within the average range and no significant deviation from the PHB was obtained by solvents [21].

Murugan et al. [22] has reported on the biological extraction of the polymer by utilizing the intestine of mealworms as an alternative approach to minimize the usage of solvents and chemicals and applicable on an industrial scale. It was found that mealworms readily ingested the freeze-dried cells of *C. necator* and whitish fecal pellets containing PHA were excreted out [22][23]. The purity of biologically extracted PHA washed with water is about 89% [22].

2.1.3. Other Conventional Extraction Methods of PHAs

PHA is accumulated intracellularly in the form of granules in the bacterial cell cytoplasm [25]. Therefore, it is essential to break the cell wall in order to extract the PHAs. Effective PHA recovery from biomass components can be complicated and costly. It is estimated up to 50% of the polymer production costs is accounted from recovery process [19]. Numerous extraction and separation technologies have been developed on small scales as well as industrial scales. To facilitate cell disruption, a pre-treatment step is essential prior to the extraction by using chemical, physical or physicochemical means [19][26][27]. These processes include solvent extraction, chemical digestion, supercritical fluid disruption, enzymatic treatment, bead mill disruption, detergents, flotation techniques, use of gamma irradiation and aqueous two-phase system [26][27].

Table 1 shows the disadvantages and condition of conventional PHAs extraction methods together with the strains involved. Among the recovery methods available, solvent extraction by chloroform is the most preferred extraction method of PHAs. There are two steps involved in this solvent extraction method, first to break the cell wall using the solvent, and then to extract the polymer [19].

Table 1. Condition of conventional polyhydroxyalkanoate (PHA) extraction methods.

No.	Isolation Method	Strain	Condition of Extraction	Disadvantages	Purity (%)	References
1.	Solvent extraction	*C. necator*	Chloroform, ratio of cells to chloroform of 1 g:100 mL, stirring for five days	• Consumption of large volume of toxic and volatile solvents • Not environmentally friendly • High capital and operation cost • Difficulty in extracting PHA from solution containing more than 5% (*w*/*v*) PHB • Native order of polymer chains in PHA granules might be disrupted • Lengthy process • Low recovery	95	[19][26][27][28]
2.	Chemical digestion	Recombinant *E. coli*	Sodium hypochlorite, 30 °C, 1 h	• Low purity of PHA • Large volume of wastewater • Treatment needed to remove surfactant from wastewater • Severe degradation of the polymer	93	[26][27][29]
3.	Supercritical fluid disruption	*C. necator*	Supercritical CO_2, 100 min, 200 atm, 40 °C, and 0.2 mL of methanol	• Dependent on process parameters • Frequent need for clean up • Difficulties in extracting polar analytes • Difficulties in dealing with natural samples • Low recovery	89	[26][27][30]
4.	Enzymatic treatment	*Sinorhizobium meliloti*	*Microbispora* sp. culture-chloroform	• High cost of enzymes • Complex process	94	[26][27][31]

No.	Isolation Method	Strain	Condition of Extraction	Disadvantages	Purity (%)	References
5.	Bead mill disruption	*Alcaligenes latus*	Bead diameter of 512 µm, bead loading of 85%, 2800 rpm	• Require several passes • Long processing time • Various process parameters have to be controlled precisely	-	[26][27][32]
6.	Flotation techniques	*Zobellella denitrificans MW1*	Chloroform, 30 °C, 72 h, self-flotation of cell debris overnight at room temperature	• Consumption of large volume of toxic and volatile solvents	98	[26][27][33]
7.	Gamma irradiation	*Bacillus flexus*	Radiation doses of 10–40 kGy	• Length of irradiation time • High initial investment cost	-	[26][27][34]
8.	Aqueous two-phase system	*B. flexus*	Polyethylene glycol [PEG] 8000/phosphate, pH 8.0 and 28 °C, 30 min	• Dependent on process parameters • Issue of robustness and reproducibility • Absence of commercial kits to evaluate aqueous two-phase system at bench scale • Poor understanding of the mechanism	95	[26][27][34]

The PHAs from *C. necator* can be extracted by using chloroform as a solvent, with a ratio of cells to chloroform of 1:100 and stirring for five consecutive days [28]. Then, the separation of PHB is achieved by solvent evaporation or subsequent precipitation of the polymer from the non-PHB cell biomass in a non-solvent such as methanol or ethanol [19][26]. Chloroform extraction is not complex as compared to the other methods and very effective to separate the PHB granules [25]. Moreover, highly purified and high molecular weight of PHB can be recovered without the degradation of PHB molecules [23][25]. However, there are various disadvantages arising from the solvent extraction method. Solvent extraction involves the use of large volumes of toxic and volatile solvent [26][27] in order to modify the cell membrane permeability and to dissolve the polymer [19][23]. The PHB recovery from biomass using the solvent pre-treatment method has however raised an unfavourable concern to the environment and high consumption of solvent consequently increase the cost of the extraction process [19][26][27].

The chemical digestion method has also been commonly used in extracting the PHB from recombinant *Escherichia coli*. The biomass was mixed in sodium hypochlorite and treated at 30 °C for 1 h [29]. The purification method, however, possesses many drawbacks such as severe degradation of the PHB molecules, a large volume of wastewater produced, and treatment being needed in order to remove the surfactant from wastewater [26][27].

2.2. Advantages of Biological Extraction Method

Biological recovery of PHB granules accumulated in cells is preferably applied in a laboratory or on an industrial scale as compared to the conventional chemical extraction method. By utilizing the intestine of insects such as mealworm as a biological method to partially purify the PHB, the undesired drawback of the chemical extraction method can be avoided. The bio-extraction of PHB using insects is more relevant to employ at a large scale due to easy rearing and require

minimal resources and space [6]. By utilizing the digestive system of insects as a green tool to extract the PHB from cells, this initiative can eventually reduce the production cost of PHB due to less solvents or chemicals being utilized.

This bio-extraction of PHB from cells is an alternative approach towards a green and sustainable method with the goal of minimizing the consumption of toxic solvents and strong chemicals [6][22]. The PHB polymer can be further purified in an eco-friendly method, by using water, sodium hydroxide or low concentration of surfactants such as sodium dodecyl sulphate and sodium dodecylbenzenesulfonate for pretreatment rather than chloroform and other harmful solvents [6][22][23].

It is interesting to note that the molecular weight (M_w) of the biologically recovered PHB is comparable to the chloroform extracted PHB. This indicates that the biological extraction process did not degrade the molecular weight of PHB granules and was touted as the preferable extraction method [22][35]. In authors opinion, the usefulness of biological extraction method will be the subject of interest for many researchers due to the high PHA purity obtained and lesser amount of chemicals involved.

2.3. Synthesis of PLA

Poly(lactic acid) or polylactide (PLA), a biodegradable thermoplastic polyester, has gained attention as compared to other polyesters due to its potential to replace conventional petrochemical-based polymers [36] Good processability, sustainability and eco-friendly characteristics make PLA a favourable biopolymer, and it has thus gained attraction in fields such as packaging, textile, automotive composites, and biomedical application [37][38][39][40].

PLA is synthesized by direct condensation polymerization of lactic acid (LA) or by ring-opening polymerization (ROP) of lactide acid cyclic dimer, known as lactide [41][42]. Lactic acid is an organic acid which occurs naturally and can be produced by chemical synthesis or fermentation. Due to the prospects of environmental friendliness and using renewable resources instead of petrochemicals, interest in the fermentative production of lactic acid has increased. The carbon source for microbial production of lactic acid can be molasses, sugar cane bagasse, or whey, etc. [43][44]. In the polycondensation process, LA monomers are linked together through the reaction between the -OH and -COOH groups by removal of by-products, resulting to low molecular weight polymer [45].

Ring-opening polymerization (ROP) of lactide is used to produce a high molecular weight of PLA. First, the water is removed in a continuous condensation reaction of aqueous LA to produce low molecular weight prepolymers. Through internal transesterification, the prepolymer is then catalytically converted by ‘back-biting’ reaction to the lactide and purified. Three potential forms resulting from the production of cyclic lactide: D,D-lactide (called D-lactide), L,L-lactide (called L-lactide) and L,D- or D,L-lactide (called meso-lactide) [42]. The ratio and sequence of D and L-lactic acid units in the final polymer can be controlled depending on the monomer used and controlling reaction conditions [38].

However, PLA suffers certain drawbacks such as poor toughness, slow crystallization rate, and low heat distortion temperature [46][47]. The application of PLA could be extensive if its performance was enhanced to achieve suitable properties. Various approaches have been proposed, for instance, blending with other polymers, plasticizer [36][48][49][50], reinforcing materials in micro- (natural fibres, particles) and nanosize (nanoclays, carbon nanotubes, nanoparticles or nanocrystals) [43][46][51][52][53]. This review is mainly focused on blending PLA with polymers such as PHA, PLA/poly(butylene succinate) (PBS) and other polymers which are discussed further in the following sections.

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