

バイオプラ最前線

A Survey of Biodegradable Plastics in the U.S.

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本号のバイオプラ最前線では、ニューヨーク州立大学シラキュース校環境科学・林学部のQ. Wang先生とChristopher T. Nomura先生に、アメリカにおける生分解性プラスチックに関する総説をお願いしました。C. T. Nomura先生はかつて、数年間に渡って理化学研究所で生分解性プラスチックに関する研究に携われた経歴をお持ちです。以下は寄稿いただいた総説の要旨部分の和訳です。

多くの石油系の合成プラスチックとは異なり、生分解性プラスチックは、廃棄後は環境内を容易に循環する分子(二酸化炭素や水)に分解されるように設計されている。

環境汚染や蓄積・残留性あるいは限りある化石資源の消費など、石油系プラスチックのもつ環境への負の影響に対する配慮から、米国では生分解性プラスチックの開発・生産ならびに利用に対する関心が高まってきている。生分解性プラスチックの潜在的な用途は、包装、農業あるいは医療などの分野である。生分解性プラスチックの利用は、石油系合成プラスチックの需要を削減し、より健全な環境を実現することができると考えられている。消費者や製造者の生分解性プラスチックに対する知識が増大するにつれ、この新しい材料から製造された製品の需要も増大すると期待される。

本総説では、米国における生分解性プラスチックの主な特性や事業的な展開について概要を述べる。

Abstract

Unlike many petroleum-based synthetic plastics, biodegradable plastics are designed to be degraded into molecules that easily cycle in the environment (CO₂, H₂O) after disposal. Because of the negative environmental impact of many petroleum-based plastics (pollution, accumulation in the environment, use of limited resource, etc.), there has been increasing interest in the U.S. for the development, use and manufacture of biodegradable materials. These biodegradable plastics have potential uses in

packaging, agriculture, medicine and other fields. The belief is that biodegradable plastics will reduce the need for petroleum-based synthetic plastics and that their use will lead to a healthier environment. As consumers and manufacturers become more knowledgeable about biodegradable plastics, it is expected that the demand for products made from these materials will increase. This paper provides a brief outline regarding properties of biodegradable plastics and their commercial development in U.S.

Introduction

Plastic materials are used worldwide today for a multitude of applications, ranging from high-tech application such as Kevlar in bulletproof vests to low-end applications such as garbage bags and shopping bags. More than 100 billion pounds of plastics are produced in the U.S. annually¹, most of which are derived from petroleum. Despite their multitude of uses, many petroleum-based plastics become a serious problem after their disposal. This is because most petroleum-based plastics are not biodegradable in the environment that are capable of causing considerable damage to ecosystems due to these properties. Incineration of plastics may generate toxic air pollution and disposal in landfill sites is limited. Furthermore, wastes generated from plastics that are used in food handling and packaging are often covered by food or other substances, which makes the recycling of these materials difficult and impractical. Thus, it is imperative to develop new plastics that are based on renewable resources and lead to biodegradable products especially for materials in single-use applications.

The American Society for Testing of Materials (ASTM) and the International Standards Organization (ISO) define biodegradable plastics as those that undergo a significant change in chemical structure under specific environmental conditions². Biodegradable plastics undergo degradation by the enzymatic action of microorganisms such as bacteria, fungi and algae. In order to be considered as a biodegradable plastic in the U.S., the plastic must pass specific tests from the ASTM. The protocols for these tests are outlined in the specific ASTM tests that include ASTM D6868, D5338, D6002, and D6400. Demand for biodegradable plastics in the U.S. is estimated to

expand by 16 percent per year and to 720 million pounds in 2012³. Target markets for biodegradable plastics include medical materials, packaging materials, disposable nonwovens, hygiene products, consumer goods, and agricultural tools⁴. In this paper, we have summarized progress, techniques, physical properties, environmental benefits, and markets of *biodegradable plastics* in the U.S., and we also describe what we view as highly promising biodegradable plastics.

Poly(lactic acid) (PLA)

Poly(lactic acid) (PLA) was first reported as a plastic by Carothers in 1932⁵ and was further developed by Dupont and Ethicon in 1954⁶. PLA is commercially produced from corn (**Figure 2**). Lactic acids are first made through bacterial fermentation using cornstarch as the feedstock. After fermentation, the lactic acids are transformed into cyclic lactide through dimerization using reactive distillation. The lactide ring is then polymerized to make PLA⁷. The chemical structure of PLA is shown in **Figure 1**.

PLA can be degraded in the environment biotically or abiotically⁸. In the absence of sufficient microbial activity or oxygen, hydrolysis is the primary way to break down PLA. Even at high humidity, it is uncommon for PLA to be degraded by fungi, mold or other microbes. This unusual property makes PLA suitable for applications where they are in direct contact with foods for extended time periods. Under aerated composting conditions, biotic process can break down PLA completely to CO₂ and H₂O within weeks. As an alternative to degradation of PLA, waste PLA can be recycled into lactic acids, which can be repolymerized into PLA.⁸

The properties of PLA are mainly determined by its molecular weight, the chain architecture (branch or linear), and

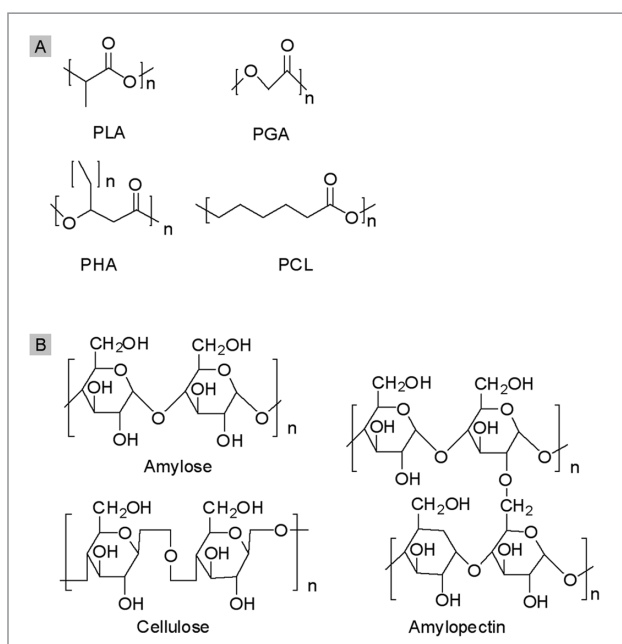


Figure 1. Structures of selected biodegradable polymers. A. Biodegradable polymers from polyesters. B. Biodegradable polymers from starch and cellulose.

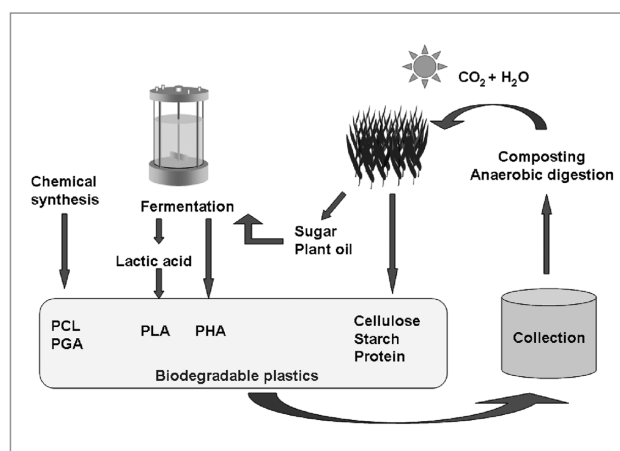


Figure 2. Production and degradation cycles for biodegradable plastics. Biodegradable plastics can be yielded from plant biomass directly, through fermentation processes, and from chemical synthesis. After usage, they will be biologically degraded by composting or anaerobic digestion to natural substances, such as CO₂ and water. New plant crops can use nutrients and CO₂ from the degradation of the biodegradable plastics to produce new polymer building blocks and starting materials.

the degree of crystallinity.⁷ The physical properties of PLA are listed in **Table 2**. PLA crystallinity and crystallization rate are controlled by the copolymerization of L- and D-lactide ratio in the polymer backbone. The greater the optical purity, the higher its crystallinity.⁴ The molecular weight, structure, and crystallinity of PLA also affect the mechanical properties, such as tensile strength, tensile modulus, and percent elongation to break.⁷ These variations in PLA properties provide the possibility to replace petroleum-based thermoplastics in many applications.

PLA and PLA derivatives have a wide range of usage, including medical materials, fiber in woven or nonwoven products, packaging materials, and molded or extruded consumer products. In initial medical application, lactide and glycolide copolymers were used to make absorbable surgery sutures. PLA and PLA copolymers are currently designed to replace metal and other non-absorbable polymers as therapeutic aids in surgery, and they also can be used for drug delivery.^{7,9} PLA is also suitable for commercial fiber products. Fabrics

made from PLA provide a silky feel, durability, and moisture will be quickly wicked away by them, keeping the wearer dry and comfortable.¹⁰ PLA also has a great opportunity to replace less environmentally friendly materials for packaging, including bags, film, thermoformed containers, and short-shelf-life bottles.⁴ This is because materials made by PLA are tough and puncture resistant but are completely degraded within 4-6 weeks under composting condition. Another application for PLA is to replace polyethylene to coat paper.¹¹

NatureWorks LLC is one of the largest facilities to produce PLA today, with annual production is 140,000 tonnes in 2006 (**Table 1**).¹² LACTEL absorbable polymers are produced by DURECT from glycolide, lactide, and ϵ -caprolactone monomers for the drug delivery and the medical device industries.¹³ DaniMer Scientific LLC is another facility in the U.S. that makes resins based primarily on PLA for coating, injection molding, thermoforming, packaging, film, additives, hot melt adhesives, and wax replacing polymers.¹⁴

Company	Polymer base	Location
Earthshell	starch + limestone	Santa Barbara, CA, USA
NatureWorks LLC	PLA	Minneapolis, MN, USA
(Cargill + Teijin)		Blair, NE, USA
DURECT	PLA, PCL	Birmingham, AL, USA
DaniMer Scientific LLC	PLA	Bainbridge, GA, USA
Metabolix, Inc.	PHA	Cambridge, MA, USA
Meredian, Inc.	PHA, PLA	Bainbridge, GA, USA
Tepha, Inc.	PHA	Lexington, MA, USA
Anderson Development Company	PCL	Adrian, MI, USA
Dow plastics	PCL	Midland, MI, USA
Eastman	Cellulose acetate	Kingsport, TN, USA
DuPont	Plant-derived polyethylene terephthalate (PET)	Wilmington, DE, USA

Table 1. Biopolymers produced by US producers

Polycaprolactone (PCL)

Polycaprolactone (PCL) is a thermoplastic biodegradable polymer (**Figure 1**) synthesized from crude oil, which is derived from the ring opening polymerization of 3-caprolactone using a catalyst like stannous octanoate. The ring-opening polymerization of PCL was first studied in 1930s¹⁵, and the biodegradable property of this polymer was first discovered in 1973.¹⁶ PCL has a very low glass transition temperature (**Table 2**) ($T_g = -60^\circ\text{C}$) and melting temperature ($T_m = 60^\circ\text{C}$), and good water, oil, solvent, and chlorine resistance.^{4,16} Since it is flexible and hydrophobic, and it is commercially available in large quantities, PCL can be blended with starch and cellulose to lower the costs and to gain better properties. Copolymers of PLA and PCL with the proper compositions and sequences could have better flexibility, hydrophilic/hydrophobic balance,

Polymer	Crystallinity (%)	T_g ($^\circ\text{C}$)	T_m ($^\circ\text{C}$)	Tensile stress (MPa)	Elongation at break (%)
PLLA *	37	50-65	173-178	33	5
sc-PLA *	60	65-72	230	45	10
P3HB *	72	10	177	40	6
P(3HB-co-7%3HV) *	51	5	135	25	25
P(3HB-co-10%3HHx) *	35-40	-1	127	21	400
P4HB *	20-30	-51	60	50	1000
PCL *	67	-60	60	4	800-1000
PGA *	45-55	35-40	225-230	-	15-20
CA (Eastman) *	-	189	230-250	32-77	5-15

* : **PLLA**, poly-L-lactic acid; **sc-PLA**, stereocomplex of PLLA and PDLA at 1:1; **P3HB**, poly-3-hydroxybutyrate; **P(3HB-co-7%3HV)**, copolymer of 93% 3-hydroxybutyrate and 7% 3-hydroxyvalerate; **P(3HB-co-10%3HHx)**, copolymer of 90% 3-hydroxybutyrate and 10% 3-hydroxyhexanoate; **P4HB**, poly-4-hydroxybutyrate; **PCL**, polycaprolactone; **PGA**, polyglycolic acid; **CA(Eastman)**, cellulose acetate produced by Eastman Chemical Company.

Table 2. Physical properties of major biodegradable polymers

and impact strength than the homopolymer of PLA.¹⁷ The two leading producers of PCL in U.S. are Anderson Development Company and Dow plastics. The company DURECT also produces copolymers of PLA and PCL for medicine delivery (**Table 1**). The degradation of PCL is slower than other biodegradable plastics making it suitable for long-term drug delivery extending over a period of more than one year.¹⁶

Polyglycolic acid (PGA)

PGA is the simplest linear aliphatic polyester (**Figure 1**). PGA can be synthesized mainly through two ways. The first way is polycondensation of glycolic acid, and the second way is ring-opening polymerization of glycolide, which is the cyclic dimer of glycolic acid.^{18,19} PGA is a rigid thermoplastic with high degree of crystallization (45-55%), high T_g (220-225°C), and high T_m (35-44°C) (**Table 2**). Because it is highly crystalline, it is not soluble in most of organic solvents, including acetone, dichloromethane, chloroform, ethyl acetate, and tetrahydrofuran.¹⁹ PGA was used to make the first totally absorbable synthetic sutures by Dexon in the 1960s. Copolymers of lactic acid and glycolic acid has been commercial biodegradable polymers to be used as biomaterials, sutures, wound dressing, and drug delivery systems since the 1970s.¹⁷ PGA can be degraded into glycolic acid by random hydrolysis and by certain enzymes, and then glycolic acid can be broken down into CO₂ and H₂O by tricarboxylic acid cycle²⁰.

Polyhydroxyalkanoates (PHAs)

Polyhydroxyalkanoates (PHAs) are polyesters (**Figure 1**) that can be produced by many bacteria as an intracellular carbon and energy storage source.²¹ These biopolymers can be formed via metabolic transformation of various carbon sources, and they can be accumulated to high levels in bacteria (up to 95% of cell dry weight). PHAs have garnered much interest as an alternative for conventional plastics because of their biodegradability and biocompatibility, and also because they can be produced from renewable sources.²² The first PHA was discovered in *Bacillus megaterium* in 1926,²³ and now, more than 100 different monomers have been reported as PHA constituents.²⁴ The most common structure of the repeating units of PHA polymers consist of 3-hydroxyalkanoates. These 3-hydroxyalkanoate harboring PHAs are divided up into three main classes based on the number of carbon atoms within each of the repeating monomers: (i) short-chain-length (SCL) PHAs consist of monomers with chain lengths C3-C5; (ii) medium-chain-length (MCL) PHAs consist of monomers with chain lengths C6-C14; (iii) long-chain-length (LCL) PHAs consist of

monomers with chain lengths greater than 14. The chemical and physical properties of PHAs are dependent on the monomer composition. Polymers composed solely of SCL monomers generally have thermoplastic properties, while polymers composed of MCL monomers generally have elastomeric properties.²¹ The physical properties of SCL PHAs and SCL-co-MCL PHAs are listed in **Table 2**. These monomers can also form copolymers that have properties that are either similar to SCL PHAs or MCL PHAs dependent on the mole ratios of these monomer units. PHA copolymers with about 95 mol% of SCL monomers and about 5 mol% of MCL monomers have properties similar to the bulk plastic polypropylene.²⁵ The monomeric composition of PHA polymers are determined by the organisms producing the PHAs, the carbon source on which cells are grown, and how that carbon source is metabolized in the cells. The polymer composition and production can also be changed by genetic modification of PHA producing strains. PHAs can be produced by recombinant bacteria strains and recombinant eukaryotes.

In late 1980s, ICI Zeneca had developed a pilot plant to produce PHAs by microbial fermentation under the trade name Biopol. However, the commercial production of PHAs has remained relatively small scale because it has been economically uncompetitive to conventional petroleum-based plastics such as polyethylene and polypropylene. Therefore, efforts to produce PHAs by transgenic plants have also been pursued as an alternative. In 1994, Sommerville and coworker transferred the PHA synthesis genes from *Ralstonia eutropha* into the transgenic plant *Arabidopsis thaliana* leaves, and the PHA production reached 14% of the plant dry weight. In 1996, Monsanto bought all the patents for making PHAs from ICI Zeneca. However, Monsanto believed PHA-producing plants should not be commercialized unless PHA production of more than 20% was achieved. Therefore, the company stopped further development of PHA, and sold the patents to Metabolix, Inc. Metabolix, Inc. continues to pursue the commercialization of PHAs by both transgenic plants and microbial fermentation.⁴ Today, Archer Daniels Midland (ADM) company has joined Metabolix, Inc. to form the subsidiary, Telles, to produce PHAs under the trade name Mirel (**Table 1**). The products include PHAs used for compost bags, durable goods, consumer retail packaging, agriculture, and polymers for marine environments.²⁶ These bulk-commodity applications are limited for PHAs due to the relatively low price for petroleum-based plastics like polypropylene and polyethylene. On the other hand, the application of PHAs in the medical applications might not be hampered by the cost, because the material properties rather than cost receive the top priority. Tepha, the sister company of Metabolix, is actively pursuing the development of PHAs for medical application, including surgery sutures, meshes, tissue scaffolds, and surgical films.²⁷

Starch-based bioplastics

Starch is an inexpensive and annually renewable material with availability just second to cellulose. The most important industrial sources of starch include corn, potato, tapioca and rice. In the last decade, the price of corn and potato starch dropped significantly in Europe and The U.S. The low price and availability of starch make it an attractive alternative to petroleum-based plastics. Starch is totally biodegradable in a wide range of environmental conditions, and CO₂ generated by incineration and degradation of starch products can be fixed by starch producing plants. Thus, the use of starch will not contribute to increases in global warming.²⁸

All starches contain two major components: amylase, a mostly linear α -D(1,4)-glucan, and amylopectin, and α -D(1,4)-glucan which has α -D(1,6) linkages at the branch point. The ratios of these two components vary based on the starch sources, and provide a natural mechanism for regulating starch material properties. Starch occurs naturally as discrete granules, and they are extremely hydrophilic. The melting point of native starch is higher than the thermal decomposition temperature, so native starch has poor thermal stability and needs chemical modification to improve its properties. There are three main ways for starch to be used effectively in plastics and materials. The first way is to use starch as a filler in traditional petroleum-based plastics, such as polyolefins. Generally, starch is added at fairly low concentrations (6-15%) to these petroleum-based plastics. Commercial products based on this technology were first sold by Ecostar and ADM companies²⁹. However, for these petroleum-based plastics supplemented with starch, only the starch component is biodegradable. The second way is to blend starch with biodegradable synthetic polymers, such as PLA and PCL, to increase flexibility and resistance to moisture for the starch component. Blends with more than 85% starch are used for foaming and injection molding. The third way is to use thermoplastic-like starch (TPS). Starch is not a true thermoplastic, but in presence of plasticizer (water, glycerin, and sorbitol), high temperature (90-180°C), and shearing, it can be melted and fluidized like other thermoplastics.^{4, 28} Starch-based bioplastics take up about 80% of the global market of bioplastics. The two leading producers are the Italian company Novamont, and the German company Biotec.²⁸ Earthshell is an American company that produces packaging material from starch and limestone (Table 1).

Cellulose

Chemically modified plant cellulose, like cellulose acetate (CA), is used in a wide range of applications. Plastic-grade

CAs were formerly classified as nonbiodegradable materials. However, the studies in mid-1990s proved that CAs with a degree of substitution of less than 2.5 are biodegradable. A decrease in the degree of substitution of CAs from 2.5 to 1.7 leads to a large increase of their biodegradability⁴. CA films usually take several years to degrade in many water and soil samples³⁰, but under appropriate composting conditions and using certain CA-degrading microbes, CA can be degraded within several months³¹. CA with an average degree of substitute 2.5 is commercially produced by Eastman Chemical Company (Table 1) with the trade name CA398-30,^{4, 31} which is used as a biodegradable plastic itself or blended with other polymers.³²

Biobased, nonbiodegradable plastics

Polyamide (PA) 11 or Nylon 11 is derived from vegetable oil. PA11 is produced by Arkema under the trade name Rilsan. PA11 is made from a renewable resource, castor bean, but is not biodegradable. PA 11 exhibits similar properties as other polyamides, such as high service temperature, exceptional chemical resistance, and used in high performance applications.³³

Polyethylene can be made from ethanol, which can be produced by fermentation of sugars derived from sugar cane and corn. Bio-based polyethylene is virtually identical to traditional polyethylene. It is not biodegradable, but it can be recycled. Producing polyethylene from ethanol also can reduce greenhouse gas emission³⁴.

Perspectives and conclusions

The demand for biodegradable plastics continues to grow and is influenced by several factors including: the continued increase of crude oil prices, consumer demand for more environmentally friendly products, and increased restrictions on the use of nondegradable plastic products. To avoid disrupting the ecosystem, plastics needed to be recycled, without creating chemical or biological imbalances. Biodegradable plastics from renewable sources allow cyclic process to be done over short time intervals (usually less than one year) and fit nicely with naturally occurring carbon cycles in the environment. Another benefit of producing plastics from renewable resources is the reduction of fossil fuel derived CO₂.

The production of monomers and polymers by enzymes, microbes, and plants represents a cleaner and more sustainable process. The progress of techniques, especially genetic engineering, will allow microbes and plants to produce biodegradable polymers or their monomers more efficiently from inexpensive biobased and renewable carbon sources. On

the basis of economic and environmental considerations, the commercialization of biodegradable plastics will keep increasing in the market. It has been estimated that the production of biobased plastics will increase from 360,000 tons in 2007 to about 2.3 million tons by 2013. Potential for up to 90% of total plastic consumer replaced by biopolymers is technically possible.³⁵

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