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RESEARCH ARTICLE

Recent Developments on biodegradable polymers and their future trends

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ABSTRACT

Bio-based polymers are attracting increased attention due to environmental concerns and the realization that global petroleum resources are finite. Plastic is one of the major pollutants at present time around the world, which is used for daily use like packaging materials, carry bags, manufacturing of different types of materials etc. So, to replace the use of synthetic plastic as well as to reduce the increasing environmental pollution an alternative must be developed. This need of synthetic plastic can be fulfilled by use of bioplas tics. Bioplastics, such as Polyhydroxyalkanoates are polymers produced by bacteria among which Polyhydroxybutyrate (PHB) is one major group. The property of PHB is similar to synthetic plastics. So, it can be used as a suitable alternative to the present day conventional practices for sustainability. Several bacterial species like Actinobacillus, Azotobacter, Agrobacterium, Rhodobacter and Sphaerotilius have been under focus for their ability of converting organic waste to bacterial PHA. For industrial production of PHB, some bacterial species like *Bacillus spp., Pseudomonas spp., Aeromonas spp., Cupriavidus spp.* have been extensively used for their potential to produce PHB. Since the production of bio-plastic is expensive many techniques have been adopted for large scale production. But, to obtain PHB in large amount the selection of proper strains of bacteria, capable of producing or accumulating PHB is necessary. Marine ecosystem is one of the largest ecosystems on Earth and still required to be explored. So in this study, comparison of the production of PHB (Bio- Plastic) in Marine and Soil bacteria has been done to find out which one has the potency to accumulate more PHB.Bio-based polymers not only replace existing polymers in a number of applications but also provide new combinations of properties for new applications. A range of bio-based polymers are presented in this review, focusing on general methods of production, properties, and commercial applications. The review examines the technological and future challenges discussed in bringing these materials to a wide range of applications, together with potential solutions, as well as discusses the major industry players who are bringing these materials to the market.For bio-based polymers like PLA and PHA, additives are being developed to improve their performance, by blending with other polymers or making new copolymers.

Keywords: Bio-based polymers, Renewable resources, Biotechnologies, Sustainable materials

INTRODUCTION

Plastic is a major environmental pollutant in the environment. The accumulation of non-degradable plastic bags in the environment is one of the major causes of pollution now- a- days. Only 1 to 2% of plastic bags in the USA end up getting recycled. Approximately 380 billion plastic bags are used in the United States every year that is more than 1,200 bags per US resident, per year. Approximately 100 billion of the 380 billion are plastic shopping bags. Thousands of marine animals and more than 1 million birds die each year as a result of plastic pollution. The United Nations Environment Programme estimates that there are 46,000 pieces of plastic litter floating in every square mile of ocean. Often mistakenly ingested by animals, clogging their intestines which results in death by starvation. Plastics at present account for about 21% of all (paper, glass, tin plate. etc.) packaging materials. Packaging materials account for 25% of the total production of plastics in India, but in terms of consumption, they account for 52%. Plastic waste produced is around 2.0 million tonnes. Though plastics constitute only about 2.4 % (world average) of the total municipal solid waste, they are perceived as a major threat because of their long life and light weight. In India, plastic waste accounts for only 0.6% of municipal solid waste, whereas in urban areas of Kerala, it is as high as 4 - 6%. Plastic accounts for approximately 10% of solid waste (Heap, 2009) and contributes 80% of the wastes accumulating on ocean surface, land, shorelines etc. (Barnes et al., 2009).

Bio-plastics are bio-based, biodegradable plastics with almost similar properties to synthetic plastics. Biodegradation can be explained as a chemical process during which micro-organisms that present in the environment convert materials into natural substances such as water, carbon dioxide, and compost. The term bio-based means the material is partly derived from biomass (plants). Synthetic plastics remain in the environment for long time as they are resistant to degradation. Bioplastics are made from variety of sources like polysaccharides, lipids and also proteins. A few examples of protein used as substrates for bioplastic production are soy protein, wheat gluten, and rice and egg albumin. Plasticizer, which is a rupturing agent added with proteins to increase plasticity. The petroleum based conventional plastics are non-renewable where the feed stocks are reinforced by carbon fibres. Renewable resource feed stocks of plastics include polymers derived from microbial culture reinforced with natural fibres such as cellulose, jute etc. The accumulation of synthetic, petroleum derived plastics in the environment is a major cause of pollution. So the approach to produce plastic, which is an essential polymer used in our day to day life, using microbes (product of microorganisms) is a novel approach. It will reduce the environmental pollution as well as the use of petroleum to make plastic bags. So it can be said in one word that bio-plastic is eco-friendly.

Since the large scale production of Bio-plastic in industry is very much costly so it has not been used extensively. During 20th century the bioplastics production was mainly dominated by the developed countries like North America, Japan, and Western Europe etc. On the basis of a study, it has been proposed thatin 2013, Brazil became one of the world's leadingbioplastics producers. In Japan, the demand of bioplastics reach a value six times more than 178000 metric tons in 2013. China also produce 100000 metric tons of bioplastics in 2013. The market of bioplastics is in the nascent stage in Southeast Asia. A research work carried out by BCC has revealed a fact that the bioplastics market value has reached 541 million pounds in 2007. By 2012, this value reached a level of 1.2 billion pounds. In 2008, a number of Biodegradable plastics like polylactic acid, resins, polyesters etc. accounted for about 90% of total bioplastics demand. Biodegradable plastics are environment friendly and can replace all plastics products available at this time. Production of bioplastics will definitely result in reduction in emission of CO_2 compared to traditional plastics. A fear of damaging already existing recycling projects by the bioplastics is one of the major concerns. The cost of production of bioplastics is also too high. This is one of the major problems related to bioplastics development. The cost is around 1.3 to 4 Euro per Kg now.

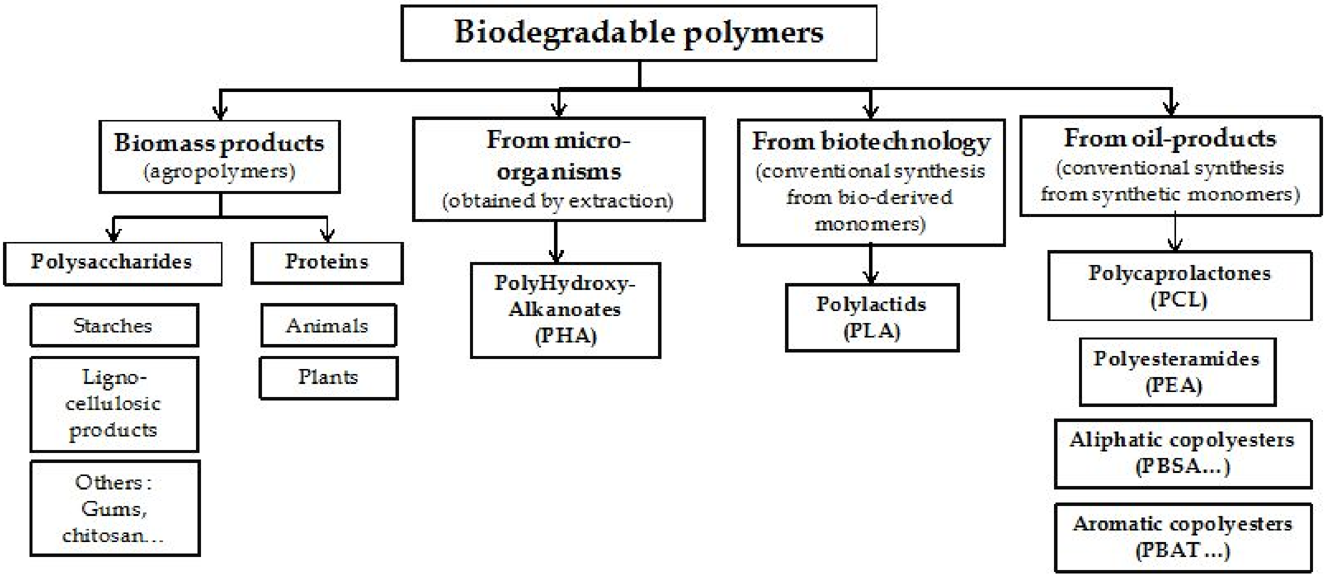
Classification and properties of bio-degradable polymers

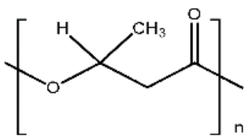
The biodegradable polymers can be classified according to their chemical composition, origin and synthesis method, processing method, economic importance, application, etc. Generally, biodegradable polymers are classified according to their origin into two groups: natural polymers which obtained from natural resources and synthetic polymers which produced from oil.

Natural biodegradable polymers

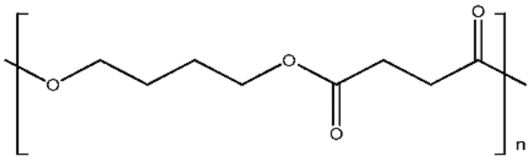
Biopolymers are polymers formed in nature during the growth cycles of all organisms; hence, they are also referred to as natural polymers. Their synthesis generally involves enzyme catalyzed, chain growth polymerization reactions of activated monomers, which are typically formed within cells by complex metabolic processes.

Biopolymers produced directly by natural or genetically modified organisms, ***Microbial polyesters***. The microbial polyesters are produced by biosynthetic function of a microorganism and readily biodegraded by microorganisms and within the body of higher animals, including humans.

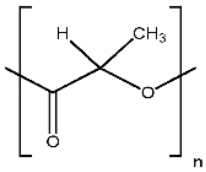




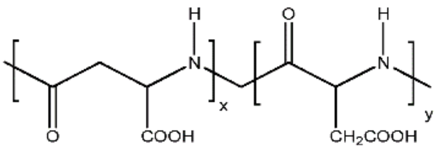
Polyhydroxybutyrate



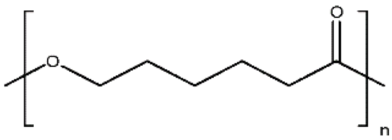
Polybutylenesuccinate



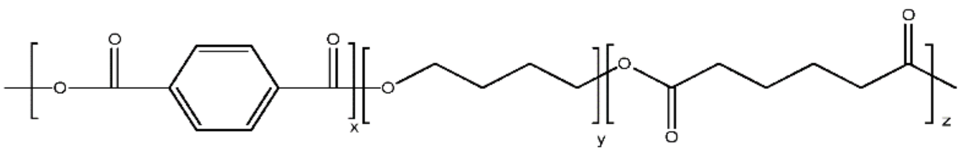
Polylactic Acid



Thermal Polyaspartate (TPA)



Polycaprolactone



Aliphatic-aromatic resin/polymer

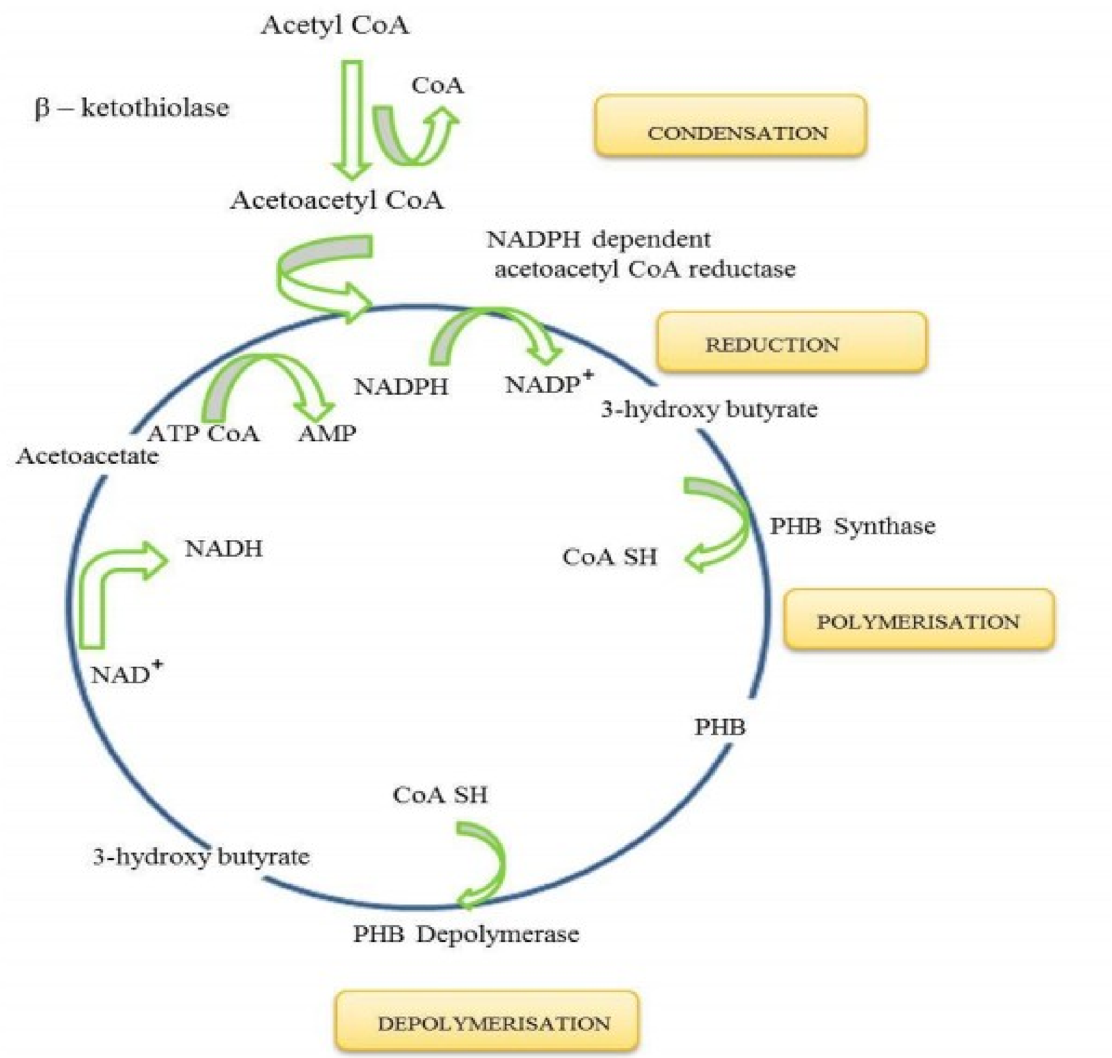


Fig. 1: The cycle representing the synthesis and degradation of poly-hydroxy-butyrate (PHB) Microbes as the bioplastic producers.

Bioplastic: its synthesis and degradation by microbes

Bioplastics are biomass based biodegradable plastics, it includes different types of plastics such as cellulose based, starch based, some aliphatic polyesters like Polylactic acid (PLA), Poly hydroxyl butyrate (PHB) etc. Poly-3- hydroxy butyrate (PHB) is 100% biodegradable and it is produced from various renewable sources. It has similar physical properties with polypropylene. Due to this character, PHB is being able to attract the vision of researchers towards its study and production. Another reason for gaining priority is that use of these biodegradable and bio- based plastics will definitely reduce the pollution caused by CO_2 emission from plastic wastes. Poly-3-hydroxybutyrate (PHB) is a polymer of 3- hydroxybutyrate and are intracellular granules produced by prokaryotic organisms as energy and carbon storage during starvation.

Poly-3-hydroxybutyrate is included in the family 'Polyhydoxyalkanoates'. Accumulation of Poly-3-hydroxybutyrate in most of the microorganisms takes place in the presence of excess carbon and limited nitrogen sources.

Biochemical studies have revealed two different pathways for synthesis of PHB.

(1) In organisms like Azotobacter beijerinckii and Zoogloea ramigera, a three-step metabolic pathway is seen. The first step is catalyzed by enzyme ketothiolase, which condenses acetyl coenzyme A (acetyl-CoA) to acetoacetyl-CoA. This intermediate is then reduced to D-(-)-P3-hydroxybutyryl-CoA by an NADPH-dependent acetoacetyl-CoA reductase (Nishimura et al., 1978; Schubert et al., 1988). The last step is catalysed by the

enzyme PHB synthase and cause head-to-tail polymerization of the monomer to PHB.

(2) In Rhodospirillum rubrum PHB synthesis is carried out through five-step synthetic pathway. An NADH-dependent acetoacetyl-CoA reductase enzyme catalyzes the formation of L-(+)-3-hydroxybutyryl-CoA, which is then converted to D-(-) -P-hydroxybutyryl-CoA by two stereospecific enoyl-CoA hydratases before polymerization.

Microbes have been reported to be the potent producers of PHB due to their high adaptability in various extreme environmental conditions. Out of these, Bacillus spp., Pseudomonas spp. and Vibrio spp. are found to be more efficient for PHB production due to their higher stability and reproducibility under environmental stress.

Many types of bacteria, such as Bacillus spp., Pseudomonas spp., Cupriavidus spp., and Aeromonas spp., have been studied for their use in industry for efficient capacity to produce PHA. Some bacterial species like Bacillus megaterium, Ralstonia eutropha have gained more attraction from the researchers. The PHB production from Bacillus megaterium has been reported to be around 84%.Several bacterial species like Actinobacillus, Azotobacter, Agrobacterium, Rhodobacter and Sphaerotilius have been under focus for their ability of converting organic waste to bacterial PHA. For industrial production of PHB, some bacterial species like Bacillus spp., Pseudomonas spp., Aeromonas spp., Cupriavidus spp. have been extensively used for their potential to produce PHB.

The optimum conditions for bioplastic synthesis and influence on changes of parameters

PHB are lipid intracellular lipid granules which are formed by bacteria under stress conditions like limitations of nutrients such as nitrogen, phosphorus, oxygen etc. and in excess of carbon. Generally, in the production of PHB along with both presence and absence of nutrients other factors like initial culture pH, culture temperature, rate of agitation.

Effect of culture pH:

Metabolic processes require specific pH to occur and slight change in pH affect the processes and make those critical. It has also shown that the production of PHB is maximum at pH 7.0.

Effect of culture temperature:

Temperature also play a major role in PHB production. The PHB production is maximum at 30˚ C.

Effect of Agitation rate:

Agitation rate also determines the growth of potent bacterial strains and PHB production. Proper agitation prevents the clumping of cells into large mass and thereby helps in the growth. Agitation facilitates each cell to utilize the nutrients available in the culture media. The rate of agitation should be in between 150-200 rpm and if it exceeds 200, the production decreases because of excessive shear force due to agitation.

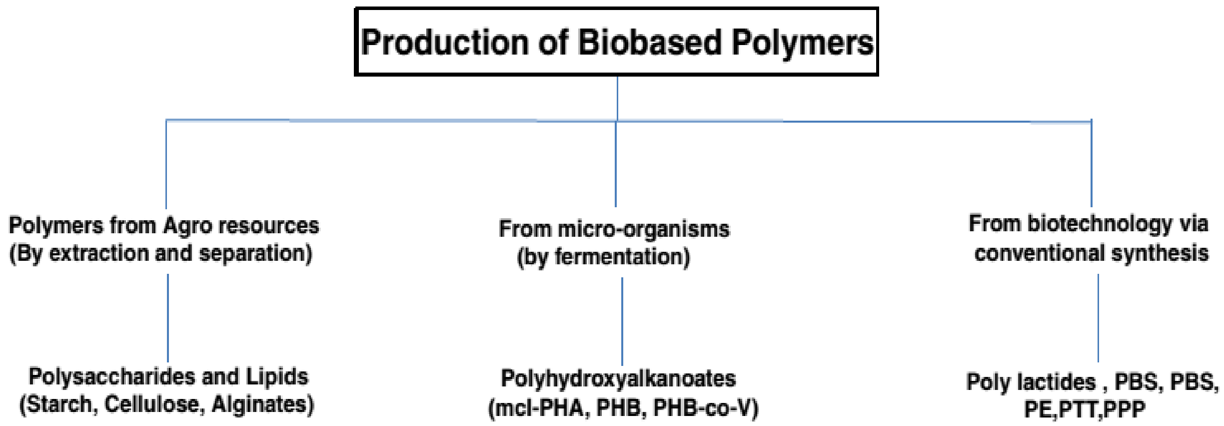


Fig. 2: shows the overview of the types and production of bio-based polymers.

Blending of substrates with PHB (bioplastics) to reduce the cost of production

The cost of industrial production of bioplastics is very high in comparison to synthetic plastics now-a-days and basically the cost of production depends on the cost of biomass for fermentation, but at the same time its production in large scale is also essential. So to reduce the cost to some extent blending of PHB can be performed with other polymers. According to a study, if the ratio of starch blendingto PHB is maintained at 30:70 % it would be advantageous to reduce the cost of PHB.

OBJECTIVES

1. **Isolation and screening of bacterial species capable of producing PHB from organic wastesand marine environment sources.**
2. **Extraction of produced PHB in the potent isolates**
3. **Characterization of PHB produced by the isolates by FTIR analysis.**

(Characterization of the two potent PHB producers was performed by various methods like Gram staining to find out whether they are Gram positive or Gram negative, Scanning Electron Microscopy to find out their morphology, different biochemical tests to find out their sources for growth and development and Antibiotic susceptibility test to find out their sensitivity towards an antibiotic.)

4. **Phenotypic and genotypic characterization of the PHB producing isolates.**
5. **To compare the PHB producing capability of the isolates.**
6. **To deduce the genetic mechanism of PHB production in the isolates.**

Biodegradability of Polymers

The bio-degradation of polymers is a very complex process and can occur in a number of ways. According, to the American Society for Testing of Materials (ASTM) and the International Standards Organization (ISO) the degradable plastics undergo a significant change in chemical structure under specific environmental conditions. These changes result in a loss of physical and mechanical properties, as measured by standard methods. Biodegradable plastics undergo degradation due to the action of naturally occurring microorganisms such as bacteria, fungi and algae. Plastics may also be designated as photo-degradable, oxidatively degradable, hydrolytically degradable, or those that may be composted. Between October 1990 and June 1992, the confusion as to the true definition of "biodegradable" led to lawsuits regarding misleading environmental advertising. Thus, it became evident to the ASTM and ISO that common test methods and protocols for degradable plastics were needed. There are three primary classes of polymer materials that gains the attention of material scientists currently. These polymer materials are usually referred to in the general class of plastics by consumers and industry. Their design is similar to that of a composite, where a polymer matrix can be plastic material or resin, it forms a dominant phase around a filler material. The inorganic or organic filler reinforced in polymer to enhance its mechanical properties and reduce material cost. *Hibiscus sabdariffa* (*Hs*) commonly known as Roselle (cellulose rich) fiber when reinforced in phenol-formaldehyde (PF) resin increased the physico-chemico-thermo-mechanical strength of the composite as seen in Figure 1-3 below.

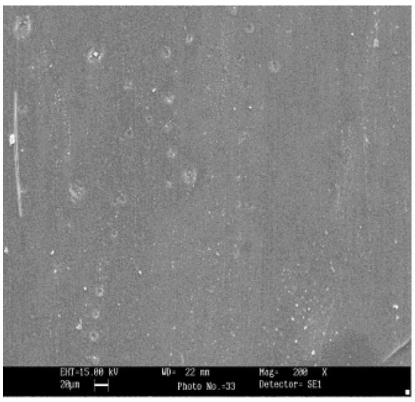


Figure 1: SEM of P-F resin

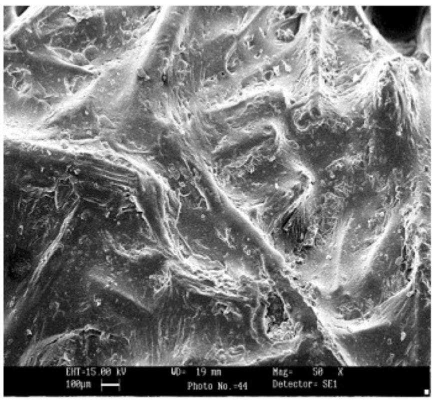


Figure 2: SEM of *Hs*-reinforced-PF composite

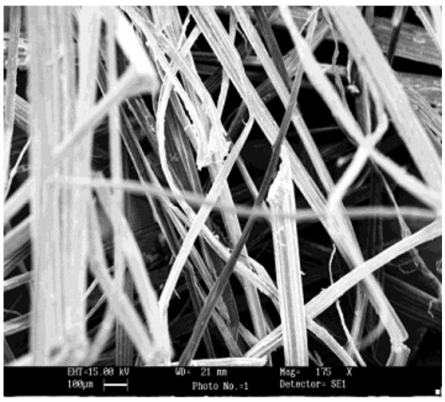


Figure 3: SEM of *H. sabdariffa* fiber

Conventional plastics are resistant to biodegradation, as the surfaces in contact with the soil in which they are disposed are characteristically smooth. Microorganisms within the soil are unable to consume a portion of the plastic that in turn, causes a more rapid breakdown of the supporting matrix. This group of materials usually has an impenetrable petroleum based matrix, which is reinforced with carbon or glass fibers. The second class of polymer materials under consideration is partially degradable. They are designed with the goal of more rapid degradation than that of conventional synthetic plastics. Production of this class of materials typically includes surrounding naturally produced fibers with a conventional petroleum based matrix. When disposed of, microorganisms are able to consume the natural macromolecules within the plastic matrix. This leaves a weak material with rough and open edges. Further degradation may then occur. The final class of polymer materials is currently attracting a great deal of attention from researchers and industry. These plastics are designed to be completely biodegradable. The polymer matrix is derived from natural sources such as starch, cellulose or microbially grown polymers and the fiber reinforcements are produced from common crops such as flax or hemp. Microorganisms are able to consume these materials entirely, eventually leaving carbon dioxide and water as by-products. Materials must meet specific criteria set by the ASTM and ISO in order to be classified as biodegradable. In general, the likelihood of microbial attack on a material is dependent on the structure of the polymer. When examining polymer materials from a scientific standpoint, there are certain ingredients that must be present in order for biodegradation to occur. Most importantly, the active microorganisms (fungi, bacteria, actinomycetes, etc.) must be present at the disposal site. The organism type determines the appropriate degradation temperature, which usually falls between 20 to 60oC. The disposal site must be rich in oxygen, moisture and mineral nutrients, while the pH must be neutral or slightly acidic (5 to 8). Biodegradation of materials occurs in various steps. Initially, the digestible macromolecules join to form a chain and experience a direct enzymatic scission. It is followed by metabolism of the split portions, leading to a progressive enzymatic dissimilation of the macromolecule from the chain ends. Oxidative cleavage of the macromolecules may occur instead, leading to metabolization of the fragments. Either ways, eventually the chain fragments become short enough to be converted by microorganisms. Biodegradable polymers that are derived from plant sources begin their lifecycle as renewable resources, usually in the form of starch or cellulose. Innovative polymer research and development and leads to large scale production by plastic converters. The biopolymers are formed into the specific end products and used by consumer. Ideally, the biopolymer will be disposed in a bio waste collection and later composted. This process will ultimately leave behind carbon dioxide and water that are environmentally friendly by-product.

In an aerobic composting environment, biodegradable polymers are expected to go through complete mineralization to CO2 and H2O:

$$\text{Polymer} + O_2 \rightarrow CO_2 + H_2O$$

Applications of Biodegradable Polymers

Research and development to introduce a biodegradable polymer is based on the design of materials with a conceptual application. It planned to replace an existing material or complement one. Biopolymers are mainly useful in medicine, packaging, agriculture and the automotive industry. Biopolymers that may be employed in packaging continue to receive more attention than those developed for others. All levels of government, particularly in China and Germany, are endorsing the widespread application of biodegradable packaging materials in order to reduce the volume of inert materials disposed of landfills, scarcity of space. It is estimated that 41% of plastics are used in packaging, and that almost half of that volume is used to package food products. BASF, a world leader in the chemical and plastic industry, is working on further development of biodegradable plastics based upon polyester and starch. Eco-flex is a fully biodegradable plastic material that was introduced to consumers by BASF in 2001. The material is resistant to water, grease, hygienic for disposable wrapping, decompose in normal composting systems. Consequently, Eco-flex has found a number of suitable applications as a packaging wrap. Environmental polymers (Woolston, Warrington, UK) has also developed a biodegradable plastic material, known as depart, the polyvinyl alcohol product is designed for extrusion, injection molding and blow molding. Depart features user-controlled solubility in water that is determined by the formulation employed. Examples include hospital laundry bags which are washed away, allowing sanitary laundering, wrap for disposable food service items, agricultural products and catheter bags. The renewable and biodegradable characteristics of biopolymers render the innovative use as garbage bags, disposable cutlery & plates, food packaging, shipping materials, agricultural products and studies. These may be developed even from renewable biopolymers such as starch. The starch material is treated by an acetylation process, chemical treatments and post-extrusion steaming. Mechanical properties of the material are adequate and effective biodegradability is achieved. Use of a clear plastic mulch cover immediately following seeding increases the yield of spring wheat if used for less than 40 days. Therefore, plastic films that begin to degrade in average soil conditions after approximately one month are ideal as crop mulches. Agricultural applications for biopolymers are not limited to film covers. Containers such as biodegradable plant pots and disposable composting containers and bags are areas of interest. The pots are seeded directly into the soil and it gets breakdown as the plant begins to grow. Fertilizer and chemical storage bags that are biodegradable are also applications that material scientists have examined. From an agricultural point of view, biopolymers that are compostable are important, as they may supplement the current nutrient cycle in the soils where the remnants are added. The medical world is constantly changing, and consequently the materials employed by it also see recurrent adjustments.

Another major and important application for biopolymers is in controlled release delivery of drug. The bioactive material releases medication at a rate determined by its enzymatic degradation.The automotive sector is responding to societal and governmental demands for environmental responsibility. Bio-based cars are lighter, making them a more economical choice for consumers, as fuel costs are reduced. The attraction of biopolymers in all of these areas is their derivation from renewable sources, slowing the depletion of limited fossil fuel stores. From the viewpoint of industry, the greatest advantage of using biopolymers derived from renewable feed stocks is their low cost. At a first glance, biopolymers appear to be a lucrative opportunity for the economy and the environment. However, as is the situation with environmental issues, a closer look at the cost-performance ratio of biopolymers must be taken in order to make sound economic decisions. One sector in which economic benefits exist from the use of biopolymer materials is in the automotive industry. With respect to fiber reinforcements, widely employed traditional glass fibres are abrasive and quickly wear down processing equipment. The texture of flax fibres is less coarse, prolonging the life of processing equipment. Natural fibres are advantageous over synthetic ones because they are less expensive and more readily available.

Work continues in the development of the biodegradable polymers due to ever increasing demand and effective use in various applications. The biopolymer industry has been enhancing its arena to a point where it is economically competitive with the conventional plastic industry. Synthetic plastics are produced on a large scale, while for the most part biopolymers are currently produced on a small scale.

Company	Location	Brand name	Production/planned capacity (kton/year)
Bio-on	Italy	Minerv	10
Kaneka	Singapore		10 (by 2013)
Meredian	USA		13.5
Metabolix	USA	Mirel	50
Mitsubishi Gas Chemicals	Japan	Biogreen	0.05
PHB Industrial S/A	Brazil	Biocycle	0.05
Shenzen O'Bioer	China		
TEPHA	USA	ThephaFLEX/ThephELAST	
Tianan Biological Materials	China	Enmat	2
Tianjin Green Biosciences	China	Green Bio	10
Tianjin Northern Food	China		
Yikeman Shandong	China		3

The inexpensive nature of the renewable resource feed stocks is encouraging researchers and industry officials to invest time to further develop these processes. As the production of biopolymers expands, so too will the services associated with it. As a general summary, it may be stated that time will lead to greater economic strength for the incorporation of biopolymer materials into society.

The fig. shows the production or the planned capacity of the world's largest Bioplastics producing companies.

CONCLUSIONS

There are unlimited areas where biodegradable polymer materials may find its use. The sectors of agriculture, automotives, medicine, controlled drug release and packaging all require environmentally friendly polymers. Due to biodegradation tailored to specific needs, each industry could create its own ideal material. The various modes of biodegradation are also a key advantage of such materials, because disposal methods may be tailored to industry specifications. Environmental responsibility is constantly increasing in importance to both consumers and industry. For those who produce biodegradable plastic materials, it is a key advantage. Biopolymers limit carbon dioxide emissions during creation and degrade to organic matter after disposal. The most promising processes are those that employ further development of biopolymer materials using renewable natural resources. Biodegradable plastics containing starch and cellulose fibres appear to be the most likely to experience continual growth in usage. Microbially grown plastics are scientifically sound and a novel approach but the infrastructure needed to commercially expand their use is still costly and inconvenient to develop. It is the right time for scientist and researchers to explore the hidden potential of natural wealth existing as polymer and fiber, to utilize them and develop biodegradable polymer for the development of science and technology while sustaining the pollution free environment.

To improve the properties of biodegradable polymers, a lot of methods have been developed,

such as random and block copolymerization or grafting. These methods improve both the biodegradation rate and the mechanical properties of the final products. Physical blending is another route to prepare biodegradable materials with different morphologies and physical characteristics.

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