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Packaging material and need of biodegradable polymers: A review

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Abstract

In the coming anthropocenic age, during which the human activity has dominant influence on environment mainly plastic, there is high need of industrial application of biodegradable polymer and mainly in field of packaging. The United States commanding \$135 billion in the worldwide packaging industry (Flexible Packaging Association) and the flexible packaging industry is the second largest sector of the business. The key packaging materials are glass, metals, paper, plastic and laminates. Such huge application of non-biodegradable materials, creating greater environmental impact and incineration of plastics may generate toxic air pollution, thereby demand of biodegradable polymer is getting higher. Application of Biodegradable plastic in the same field are the one which fulfil all these functions without causing any threat to environment. It is classified under three categories: Natural (Starch, Proteins etc.), Synthetic (Produced from Petroleum resources (PHA) and Produced from microorganism (PBS, PES)), and Semi-synthetic (Starch-PLA Blend, Starch-PCL Blend etc.). The belief is that biodegradable polymer material will reduce need of synthetic polymer and reduces pollution. Thereby, producing positive affects both economically and ecologically by reducing expensive cost of recycling after use and by reducing pollution respectively.

Keywords: Blends, Synthetic Biodegradable Polymer, Polyesters, Packaging, Future Prospects.

Introduction

Now a day most of items are packed in materials^[1]. Packaging- a science, art and technology used to protect the product, distribution and storage of product, carry nutritional information and present the product to consumer^[2]. Consumption of packaging is higher in field of food packaging than the non-food and tobacco items according to Indian Brand Equity Foundation.

The current key materials for packaging is glass, paper, synthetic plastic, cardboards, co-extruded films, and laminates. Major segment of packaging in India and almost all world, is flexible packaging and laminates of different synthetic plastic.

Environmental impact of these persistent synthetic plastic as waste is raising general global concerns, Due to limitations of disposal methods and satisfactory landfills, their incineration causes generation of toxic air pollution, and expensive recycling methods, fuelling international drive to develop biodegradable packaging material which can fulfil all functions required for food from packaging material without causing any threat to environment^[3].

The other primary factors driving development of biodegradable packaging are increase in prices of crude oil, consumer demand increases, proliferation of consumer packaging, development of new applications of bio-plastic and increased economic viability.

Biodegradable polymers are classified under three categories:

1. Natural biopolymer e.g. PCL, PLA, PGA
2. Synthetically produces biodegradable polymer
 - a. Produced from microorganism (PBS, PES)
 - b. Produced from crude oil (PHA)
3. Semi-synthetic biodegradable polymer e.g. Starch blend (Starch+PCL, Starch+PLA)

This review study is to provide a brief outline of synthetic biodegradable polymer which can be used in the area of biodegradable packaging research and development to reduce the various harmful effects caused by excessive human involvement in every field.

Key Functions of a Packaging Material

An ideal packaging material has to perform the following functions

1. Keeping the product clean: It must keep the product clean and provide a barrier against dirt and other contaminants.

2. Prevent loss to product: Its design should provide protection and convenience in handling, during transport, distribution and marketing. In particular, the size, shape and weight of the packages must be considered.

3. Physical and chemical protection: It must provide protection to product against physical and chemical damage (e.g. water and water vapour, oxidation, light) and insects and rodents.

4. Provide necessary information so that the product is used correctly and have sales appeal [2, 4].

5. Influence the consumer decision

5a) Identification: Appearance of package in terms to identify brand easily.

5b) Attention: Packages colours and shapes with pictures on packages to attract the attention when consumers are less familiar with a brand.

5c) Communication: Package appearance can also influence the evaluation of the core product. The package supporting the view that brand identity and image can be enhanced if not created through packages [5].

Current Materials Usage in Packaging Industry

According to FICCI India, synthetic plastic is one of the materials which are used in packaging industry nearly about 50% as compared to other materials such as Metals, Glass and Paper-cardboard. Today the key determinants of demand of packaging are: shifting consumer trends, light weight, varied sizes, barrier properties, and shelf life of packaged products and transportation cost. [IBEF] Synthetic Plastic is only one that contains all the above properties but not eco-friendly. Yet other ones are eco-friendly or recyclable but doesn't have all the mentioned properties. So, let us review the today's scenario of polymer packaging industry.

Packaging Industry at Glance

The industry is expected to grow at a 10 year CAGR of over 15 % in India according to report published by FICCI. The large growing middle class, liberalisation and organised retail sector are the catalysts to growth in packaging industry. Earlier the rigid packaging is dominant, currently there is a shift of demand and demand is from rigid to flexible packaging. There are about 600-700 packaging machinery manufacturers, 95 % of them are in the small and medium sector located all over India [6]. Multi-layered laminated sheets of plastics mainly include PE, PP, PET, and PVC. Polyethylene and polypropylene account for ~ 62% of polymer usage in the flexible packaging industry. Flexible packaging is dominated by small & medium sized companies with maximum usage in processed foods segment. The Indian packaging industry is expected grow at

15%p.a. with flexible packaging growing at 17% p.a. and rigid growing at 14% p.a [7]

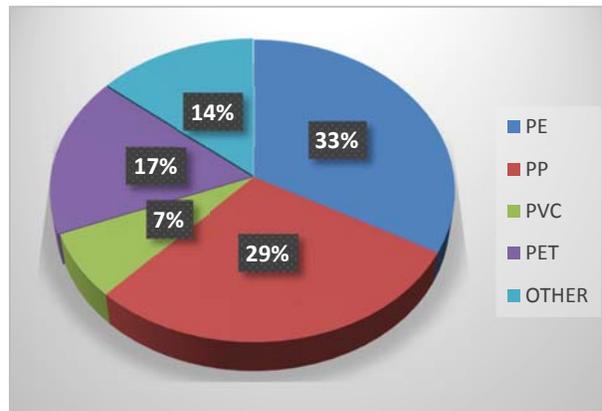


Fig.3: Consumption of different kinds of plastic in packaging

Flexible Packaging Films a Largely Growing Industry

In general, flexible films have the following characteristics:

- lower in cost
- good moisture and gas barrier property
- heat sealable
- wet and dry strength
- easy to handle and convenient for the manufacturer, retailer and consumer
- add lighter weight
- fit closely to the shape of the food, thereby wasting little space during storage and distribution [8].

In general, all the flexible films are made up of synthetic thermoplastic polymer such as Polyolefin, Polystyrene and PVC. Following is the percentage share of different material in packaging

Polyolefins Polyolefin is a collective term for polyethylene and Polypropylene, the two most widely used plastics in food packaging. Polyethylene and polypropylene both possess a successful combination of properties, including flexibility, strength, lightness, stability, moisture and chemical resistance, and easy process ability, and are well suited for recycling and reuse [9].

HDPE is High-density polyethylene is stronger, thicker, less flexible and more brittle than low-density polyethylene and has lower permeability to gases and moisture. It has higher softening temperature (121°C) and can therefore be heat sterilised. Since HDPE is cheaper to buy as raw material and process, it is used when clarity is not as great a factor.

LDPE is Low-density polyethylene is heat sealable, inert, odour free and shrinks when heated. It is a good moisture barrier but has relatively high gas permeability, sensitivity to oils and poor odour resistance. It is less expensive than most films and is therefore widely used for food storage bags [8, 10].

a) Polypropylene is a clear glossy film with a high strength and is puncture resistance. It has moderate permeability to moisture, gases and odours, which is not affected by changes in humidity. It stretches, although less than polyethylene. PP is used in rigid containers like baby bottles and cups and bowls [10].

b) Polystyrene Polystyrene is a brittle clear sparkling film which has high gas permeability [8]. Polystyrene, an addition polymer of styrene, is clear, hard, and brittle with a relatively low melting point. It can be mono-extruded, co-extruded with other plastics; injection moulded, or foamed to produce a range of products.

Foaming produces an opaque, rigid, lightweight material with impact protection and thermal insulation properties. Typical applications include protective packaging such as egg cartons, containers, disposable plastic silverware, lids, cups, plates, bottles, and food trays.

In expanded form, polystyrene is used for non-food packaging and cushioning, and it can be recycled or incinerated [9].

c) PVC (Polyvinyl chloride) PVC was one of the first polymers to be used in food packaging applications, and it replaced many traditional materials such as glass (used in bottles) and various forms of card and paper. PVC packaging for foodstuffs offers several

Advantages over the traditional materials:

- a) lightweight compared with glass, with the added benefit of reduced transport emissions due to the reduction in weight
- b) shatter resistant, as it would reduce the number of accidents with glass from broken bottles at home and outside
- c) excellent organoleptic properties: PVC-U imparts no taint or taste to foodstuffs
- d) high clarity and to allow high visibility of the product
- e) good barrier properties for the preservation of food
- f) good Cost: Performance ratio
- g) innovative product shapes and complex designs are achievable [11].

Application of PVC Bottles PVC's toughness combined with good clarity and resistance to oils and other substances makes it a suitable material for blow-moulded bottles for holding a variety of products

Flexible film PVC-P has found wide acceptance for use in the preservation of foods, both in the supermarket and the domestic kitchen. These materials have "stretch" properties and unique "cling," and hence they are suitable for the hand-wrapping of fresh produce.

Such films have high oxygen and water vapour transmission, keeping red meat "red" for longer periods on supermarket shelves. These materials can also be heat-sealed [11, 12].

d) PET Its primary packaging uses, however, are in metallized packages, such as those for potato chips and pretzels, as well as in microwave packaging, brick packs and medical packaging (such as sterile wrap and lidding material on pull-away packs).

It also can be found in tobacco wrap, cigarette wrap and labels. PET is used in these applications because it has good mechanical properties—such as toughness and stiffness—and good thermal properties, which mean it can withstand higher processing temperatures than some other resins [12].

e) Coextruded films Three layers blown film extrusion (also known as blow film co-extrusion) is a process of

simultaneously extruding in molten stage three polymers which adhere to each other through a common die to form an integral film of unique strength and barrier properties. The selection of layers depends upon end properties required as well as items to be packed. Some combinations of layers are as under:

Table 1: Some examples of combination of layers and their application

S. No.	Combination	Application
1.	LL/LD/LL	Heavy-duty bags
2.	LL/HD/LL	Industrial base food oil, backed confectionery, dry vegetables, dry unit, hydrogenated oil, lube oil
3.	LD/SCRAP/LD	Garbage bags.
4.	HD/LD/LD	Industrial base food oil, backed confectionery, dry vegetables, dry unit, hydrogenated oil, lube oil
5.	HD/LL/Primacor	Oil packaging

Waste Production By Packaging Industry And Need of Biodegradable Polymers

Around millions of tonnes of plastic waste including refuse sacks, carrier bags and packaging, are buried in landfill sites every year around the world. China generates about 16 million tonnes, India 4.5 million tonnes and the UK 1 million tonnes, of which more than 800,000 tonnes is Polyethylene waste. Conventional Polyethylene can take longer than a complete century to degrade [13]. Globally 3.2 million tonnes of the 26 million tonnes of the household waste produced annually is packaging. 150 million tonnes of packaging waste come from industry and commerce each year. People in the US throw away 2.5m plastic bottles every hour and out of which less than 3% are recycled. In the UK: of the 15 million plastic bottles used every day and much less than 3% gets recycled. Less than 1% of the billions of plastic bags used each year are recycled and the majority are used only once [14]. Increasing resource scarcity means it is more urgent than ever to be efficient with packaging and find solutions to deal with "Post-consumer" waste. Besides looking at 4 R's, find a permanent solution by expanding the research and development of Biodegradable polymers [15].

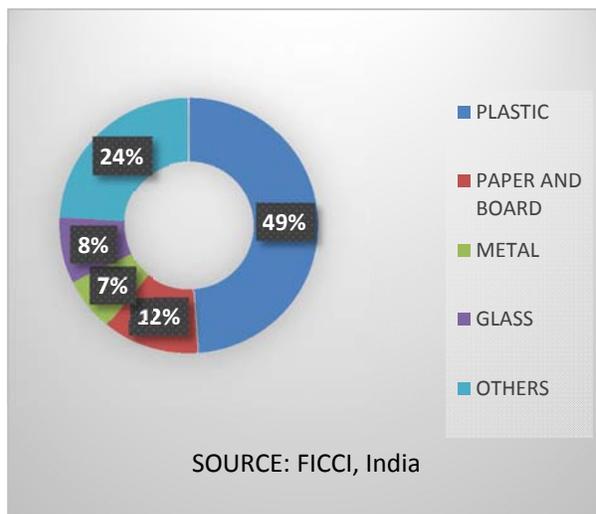
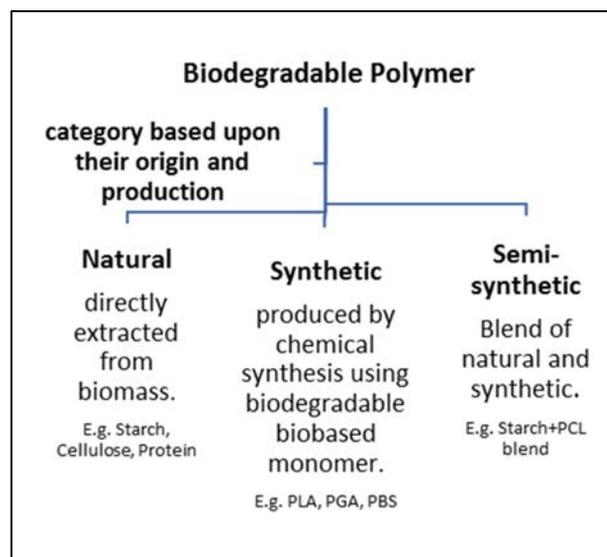


Fig.2: Consumption of different material in packaging

Biodegradable Polymers-A Today's Need

According to ASTM and ISO standards degradable polymers are those which undergo a significant change in chemical structure under specific environment conditions. The chemical changes occur in loss of physical and mechanical properties [16]. Biodegradable polymers were first introduced in 1980s. There are many sources of biodegradable plastics, from synthetic to natural polymers. Natural biopolymers are available in large quantities from renewable sources, while synthetic polymers are produced from non-renewable petroleum resources. Besides of their biodegradability, they have other characteristics such as air permeability, low temperature sealability, availability and low price. Several biopolymers such as starch, cellulose, chitosan, PLA, PCL, PHB etc. are used for packaging purposes. The new and current trend is to use blends of different biopolymers such as starch-PLA blends; starch-PCL blends etc. which are semi-synthetic in nature.



Synthetic Biodegradable Polymer

Synthetic Biodegradable Polymers are the synthetic ones with hydrolytically unstable linkage in backbone. This is commonly achieved by use of functional groups such as esters, amides, anhydrides and orthoesters. Their application in field of packaging is very much suitable. Mainly Aliphatic Polyesters are the dominants of this category.

Aliphatic Polyesters

Aliphatic polyesters are most extensively studied class of biodegradable polymers, because of their important diversity and its versatility. Mainly two routes i.e. Polycondensation and Ring opening polymerization leading to the development of synthetic polyesters. Polycondensation of bifunctional monomers preferentially yields low molecular weight polymers. Ring opening polymerization is preferred when high molecular polymers are desired. Most biodegradable polyesters are prepared via ring opening polymerization of six or seven membered lactones. The aliphatic polyesters are biodegradable in nature due to their hydrolysable ester bonds and almost all aliphatic compounds are the high molecular weight biodegradable compounds [17].

Aliphatic polyesters can be classified into two types according to the bonding of the constituent monomers.

Polyhydroxyalkanoates

Synthesized from hydroxyacids, HO-R-COOH
 E.g. Poly(glycolic acid) or Poly(Lactic Acid)

Poly (alkenedicarboxylate)

Synthesized from Polycondensation of Diols and dicarboxylate
 E.g. PBS, PES

Polyhydroxyalkanoates

Polyhydroxyalkanoates class mainly consist of PGA, PLA, and PCL.

Poly (glycolic acid) (PGA) It is the simplest linear aliphatic polyester. It is highly crystalline, with a crystallinity of 45-55% and thus is not soluble in most organic solvents. It has a high melting point (T_m) 220-225 °C and a glass transition temperature of 35-40 °C. It is prepared by ring opening polymerization of a cyclic lactone, glycolide. It has excellent mechanical properties due to its high crystallinity [17, 18]. PGA has vital advantage of degradability through simple hydrolysis of the ester backbone in aqueous surroundings. The polymer is known to lose its strength in 1-2 months when hydrolysed and losses mass within 6-12 months [18].

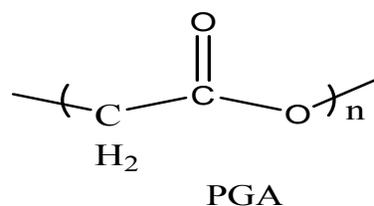


Fig 6: Structure of Poly glycolic acid (PGA)

Poly (lactic acid) (PLA) Poly (lactic acid) (PLA) also known as polylactide is a translucent aliphatic polyester made entirely from various natural sources found in great abundance like tapioca, corn, sugar beets wheat and etc. It is composed of repeating units of lactide, which is biorenewable and biodegradable. PLA is a semi-crystalline polymer. The degree of crystallinity is about 35-40%. It is a recyclable thermoplastic as the structure doesn't form crosslinks [19]. PLA is degraded by simple hydrolysis of the ester bond and does not require the presence of enzymes to catalyse this hydrolysis [20] but exists microorganisms, in environments, can degrade it.

Chemical Structure

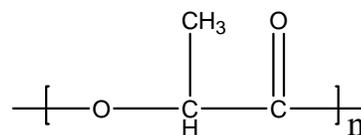


Fig 7: Structure of Poly (Lactic Acid) (PLA)

Different routes are used to obtain PLA. In general, there are three methods which can be used to produce high molecular mass PLA of about 1,00,000 Daltons:

- Direct condensation polymerization of D- or L-lactide acid.
- azeotropic dehydrative condensation and
- Polymerization through lactide formation, the ring-opening polymerization [17,21].

Currently, direct condensation and ring-opening polymerization are the most used production techniques. Ring-opening polymerization (ROP) of the lactide needs catalyst but results in PLA with controlled molecular weight and this route allows also the control of the final properties of PLA by adjusting the proportions of the two enantiomers [21]. Depending on monomer used and controlling reactions conditions, it is possible to control the ratio and sequence of D- and L-lactic acid units in the final polymer. PLA, a thermoplastic polymer having high-strength, high-modulus and a hard polymer whose hardness is similar to acrylic plastic, is not soluble in water and it is completely degraded under compost conditions; and it can be made from annually renewable proper resources to yield articles for use in either the industrial packaging field or the biocompatible/bioabsorbable medical device market [18, 20]. It is easily processed on standard plastics equipment to yield molded parts, film, or fibers. PLA can achieve excellent mechanical properties if high molecular weight PLA as comparison to Low or medium molecular weight [20].

PLA polymers are materials that have high potential for packaging applications and create a lot of interest in the packaging industry for its properties and earth-friendly biodegradability. PLA are

- resistance to oil-based products,
- seal ability at lower temperatures, and
- Can act as flavor or odor barriers for foodstuffs [18, 22].

The ratio between the two meso forms (L or D) of the lactic acid monomer decides the property:

a) 100% L-PLA results in a material with a very high melting point and high crystallinity.

b) If a mixture of D- and L-PLA is used instead of just the L-isomer, an amorphous polymer is obtained with a Tg of 60°C, which will be too low for some packaging purposes.

E.g. A **90/10% D/L copolymer** gives a material which can be polymerized in the melt, oriented above its Tg and is easy processable showing very high potential of meeting the requirements of a food packaging. The temperature of processing is between 60 and 125°C depending on the ratio of D- to L-lactic acid in the polymer [22].

Brittleness and poor thermal stability are the disadvantages of PLA. It can be plasticized to improve the chain mobility and to favour its crystallization. Plasticization is realized with oligomeric acid, citrate ester or low molecular polyethylene glycol. Furthermore, PLA may be plasticized with its monomer or, alternatively, oligomeric lactic acid and the presence of plasticizers lower the Tg. As outlined above, PLA offers numerous opportunities to tailor the properties of the finished material or package. PLA may be formed into blown films, injected molded objects and coatings all together explaining why PLA is the first novel biobased material produced on a major scale [22].

Poly(lactide-co-glycolide) (PLGA)

L-lactide and DL-lactide have been used for copolymerization with glycolic acid monomers (G). Different ratios of Poly (lactide-co-glycolide) have been commercially developed.

A copolymer with a monomer ratio of

- 25L: 75G monomer ratios - Amorphous Polymer
- 80L: 20G monomer ratios - semi-crystalline.

As the ratio of monomer L/G increases, the rate of degradation of the copolymer decreases [57]. Miller *et al.* have shown that the 50/50 Poly (lactide-co-glycolide) is very hydrolytically unstable and the resistance to hydrolytic degradation was found to be more pronounced at either end of the co-polymer composition range. The intermediate copolymers were found to be much more unstable compared to the homopolymers. Thus, 50/50 Poly (DL-lactide-co-glycolide) degrades in approximately 1–2 months, 75/25 in 4–5 months and 85/15 in 5–6 months [23].

Polycaprolactone

Polycaprolactone (PCL) is synthetic biodegradable polyester which is produced from crude oil and biodegradable thermoplastic polymer. It has a good resistance to water, oil, solvent and chlorine [24, 25]. PCL is a hydrophobic, semi-crystalline polymer; having a glass transition temperature (Tg) of –60 °C [17, 24] and melting point ranging between 59 and 64 °C, dictated by the crystalline nature of PCL which enables easy formability at relatively low temperatures [24]. A semi-crystalline linear polymer is obtained from ring-opening polymerization of ϵ -caprolactone in presence of tin octoate catalyst where the monomer is relatively cheap cyclic compound.

The number average molecular weight of PCL samples may generally vary from 3000 to

90,000 g/mol and can be graded according to the molecular weight. PCL crystallinity indirectly depends on the molecular weight i.e. increase in molecular weight tends to decrease in crystallinity [24].

At room temperature, PCL is a semi-rigid material has a modulus in the range of low-density polyethylene and high-density polyethylene, a low tensile strength of 23 MPa and a high elongation to break (more than 700). Enzymes and fungi easily biodegrade PCL. To improve the degradation rate, several copolymers with lactide or glycolide have been developed [17]. PCL has a wide range of application area, such as packaging, medical implant, and controlled drug delivery system, so the crystallization and the morphologic properties of PCL are very important. Crystallization is an important process because of the final properties of any polymer can be determined by the superstructural and micro domain morphology, and overall crystallinity. Additives can be used to control the crystallization properties of PCL [24].

Enzyme and fungi can easily biodegrade PCL. Hence, Due to the rapid degradation rate of PCL, several co-polymeric systems containing PCL have been investigated to improve the properties of the native polymer. Co-polymers of ϵ -caprolactone with DLlactide have yielded materials having high rate of degradation [18].

Polyhydroxyalkanoate (PHA)

Polyhydroxyalkanoates are natural, thermoplastic, aliphatic biogenic polyesters [44, 45] that can be naturally accumulated in microbial cultures and they differ in their properties depending on their chemical composition (homo-or copolyester, contained hydroxyl fatty acids).

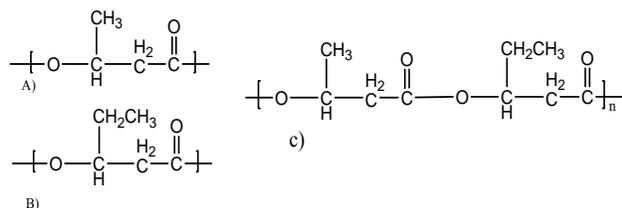


Fig 8: Structure of some Polyhydroxyalkonates A) PH3B, B) PHV, and C) PHBV

Properties Of Polyhydroxyalkanoates

- Water insoluble and relatively resistant to hydrolytic degradation.
- Poor Acid and Base resistant but good ultra-violet resistance.
- Soluble in chloroform and other chlorinated hydrocarbons.
- Biocompatible and hence suitable for medical applications.
- Sinks in water, facilitating its anaerobic biodegradation in sediments.
- Nontoxic.
- Less 'sticky' than traditional polymers when melted [26, 27].

PHA is a carbon-neutral and valuable polymer [28] that are bacterial polymer that could be produced from many renewable carbon sources by microorganisms usually under unbalanced growth conditions [29], making it a sustainable and environmental-friendly material. At present, PHA is not cost competitive compared to fossil-derived products.

Encouraging and intensifying research work on PHA is anticipated to enhance its economic viability in the future [28]. Due to mechanical properties of PHAs, they are suitable for replacements of petro-chemically produced bulk plastics (polyethylene, polypropylene etc.), but in contrast to these commodity plastics PHA through natural microbiological mineralization, are completely degradable to carbon dioxide and water [29].

PHB

Poly-3-hydroxybutyrate (PHB) is linear polyester of D (-)-3-hydroxybutyric acid is one of the biodegradable PHA (polyhydroxyalkanoates) [26, 30] and is a naturally occurring hydroxyacid (a linear polyester) which was first discovered in bacteria by Lemoigne in 1925. It is accumulated in intracellular granules by a wide variety of Gram-positive and Gram-negative organisms under conditions of a nutrient limitation other than the carbon source [26] mainly bacteria like *Alkaiogenes eutrophus* is used for production in which it acts as a storage material [31]. Poly-[(R)-3-hydroxybutyrate] (PHB) is a linear, isotactic, semi crystalline and has relatively high glass transition and melting temperatures. These aren't good features for packaging applications.

With the purpose to improve flexibility, PHB is synthesized with various co-polymers such as poly-(3-hydroxyvalerate) (HV) leading to a decrease of the glass transitions and melting temperatures). Current applications of PHB-based polymers or composites include the packaging industry, medicine, pharmacy, agriculture, food industry [28].

The characteristics of biodegradability and biocompatibility allow PHAs to be competitive in special market sectors. Both PHB homopolymer and PHBV copolyester have been

receiving commercial interest as a promising candidate for the large-scale production of biodegradable and biocompatible thermoplastics [19].

Properties of Poly(hydroxy butyrate)

- Eco-friendly, biodegradable
- decompose into water and CO₂ by microorganisms
- They are chain of 14 monomeric units.
- Usually molecular weight varies from 2-3×10³ kDa. This again depends on capacity of microorganism to store the produced polymer.
- Can be a good substitute for synthetic plastic
- Can be produced from non-replenishing sources [30].

The key for research lies in its properties like biodegradable, durable, temperature resistant, non-pollution causing polymer. This simple human friendly polymer belongs to a huge class of biopolymers by PHA and well known for their physico-chemical properties as natural polymers. Their properties resemble that of synthetic plastics however the most attracting property of them is production and degradation which is contradictory to synthetic plastics [28, 30].

PHBV

Among PHA class, the main biodegradable microbial polymer studied is a copolymer of hydroxybutyrate and hydroxyvalerate (HV). It was first synthesized by ICI in 1983. It can be produced by adding propionic acid to nutrient feedstock supplied to bacteria. Mixed carbon source is also used [17].

Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) is a potential environment-friendly substitute for traditional plastics. Many groups have been reported about the structure, mechanical properties and biodegradability of PHBV as biodegradable plastics [32]. PHBV is a highly crystalline polymer with a melting point of 108 °C and glass transition temperature in the range -5 °C to 20 °C. The pure copolymer is also brittle, less than PHB. Elongation at break is lower than 15% and elastic modulus is 1.2 GPa. [17].

By changing the hydroxyvalerate unit content, properties of copolymer can be modified.

- The melting point of the copolymer, as well as the glass transition temperature and the crystallinity decrease as the hydroxyvalerate unit content increases.
- The impact strength increases and the tensile strength decreases with an increase of the HV units [17, 33].

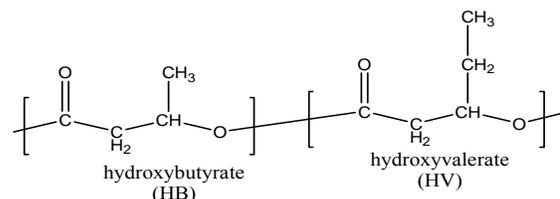


Fig 9: Structure of PHBV

Some of the properties of the PHBV range span those of polypropylene to polyethylene. But PHBV properties can also be enhanced by adding normal polymer additives such as natural plasticizers, fillers, colorants, etc. [33].

The PHBV's biodegradability, apparent biocompatibility, and its manufacture from renewable resources are the main

keys of their wide applications. Primary application areas in which these features meet some market needs are

- (i) disposable personal hygiene: PHBV could be used as the sole structural material or as part of a degradable composite;
- (ii) packaging: PHBV could be used for films, blow-molded bottles, and as a coating on paper; and
- (iii) Medical: potential in reconstructive surgery and controlled release fields due to PHBV's biocompatibility coupled with its slow hydrolytic degradation [33].

Polybutylene succinate (PBS)

Polybutylene succinate (PBS), produced from two monomers i.e. butanediol and succinic acid, is one of the biodegradable thermoplastic polyesters [34]. PBS is a white crystalline thermoplastic with density of 1.25g/cc melting point (T_m) in the range of 90-120 °C and a low glass transition temperature (T_g) of about -45 to -10 °C. It has generally excellent mechanical properties and processability [35]. However, low molecular weight and low stiffness and high cost restrict its applications [34]. Like other aliphatic polyesters, it is thermally stable up to approximately 200 °C and because of its good biodegradability; it has attracted intense attention from industry. It can also substitute for PET, PP, and PS and replace Polyolefins [35]. In addition to its applications in textile industry and medical fields, PBS is a promising candidate to produce disposable packaging [34]. PBS is biodegradable aliphatic polyester with properties that are comparable to Polypropylene [36].

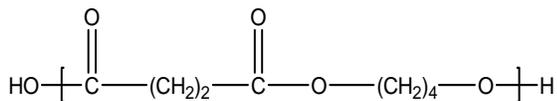


Fig 10: Structure of Polybutylene succinate

PBS technically produced biodegradable polyester is important due to

- Succinic acid can be a biogenic monomer i.e. monomer based on renewable resources.
- Rate of hydrolysis of polysuccinates is higher than those polyesters produced from aliphatic dicarboxylic acid
- Melting temperature and Heat distortion temperature of PBS is highest [35].

Polymer blending is often performed in order to improve the physical properties.

- a) In the case of copolyester (PBSA), polyester tensile strength decreases with the introduction of the secondary component (adipate), exhibiting a tendency similar to that of the other physical properties. E.g. PBS is the polyester with the highest tensile strength, while the copolymers PBSA (80/20) and PBSA (60/40) shows improved elongation.
- b) In case of PBSU was found to be miscible with Poly(vinylidene fluoride), poly(vinylidene chloride-co-vinyl chloride), poly(ethylene oxide) (PEO) and poly(vinyl phenol). On the other hand, PBSU was found to show no miscibility with PHB, poly(3-hydroxybutyrate-co-hydroxyvalerate) and poly(caprolactone) [37].

PES

Poly(ethylene succinate), PESu, synthesized by the polycondensation of an aliphatic diacid (i.e. succinic acid)

and an aliphatic diol (i.e. 1,2-ethanediol) is a biodegradable aliphatic semi-crystalline polyester. Aliphatic polyesters have poor physical and mechanical properties [38] but PESu shows very attractive properties including, melt processability, chemical resistance and mechanical properties similar to that of high density polyethylene and polypropylene [39]. Poly(ethylene succinate)- melting point 103–106 °C, respectively, and they present controllable biodegradation rate and high processability [40].

PES is prepared either by ring-opening copolymerization of succinic anhydride with ethylene oxide or by polycondensation of succinic acid and ethylene glycol. It can be used for most application in which polyethylene is currently used and yield strength of PES is higher than polyethylene [41].

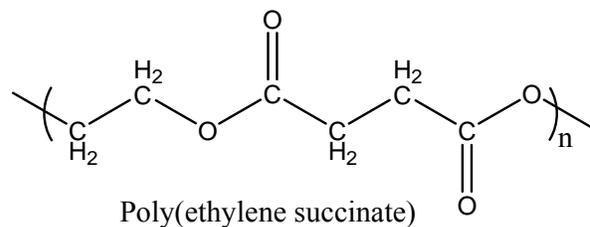


Fig 11: Structure of PESu

Dimitrios *et al.* concluded that the biodegradation rates of the polymers decreased following the order PPSu>PESu>PBSu where PPSu, PESu, and PBSu is Poly(propylene succinate). Poly(ethylene succinate) and Poly(butylene succinate) respectively [42].

Natural-Synthetic Polymer Blend

This category mainly consists of blends of aliphatic polyester with starch. Due to various disadvantages, starch is unsuitable for many applications. These include:

- 1) brittleness in the absence of suitable plasticizers,
- 2) hydrophilic nature of starch and poor water resistance
- 3) deterioration of mechanical properties upon exposure to environmental conditions like humidity, and
- 4) Soft and weak nature of starch in the presence of plasticizers.

Most of the synthetic polymers are hydrophobic and thermodynamically immiscible with hydrophilic starch, thus mixing result in phase incompatibility and poor mechanical properties. Ideally, starch and the second polymer should be covalently bonded through existing functional groups or by introduction of new functional groups [43, 44].

One means to improve the characteristics of starch films is through the preparation of composites or blends with other materials such as fibers, nano clays or biodegradable polymers. Studies on starch/polyester blends have focused on the incorporation of starch in the polyester matrix to reduce production costs and maintain the biodegradability [43].

Starch and Poly(vinyl alcohol) Polyvinyl alcohol (PVA) is a synthetic polymer, non-toxic, flexible, soluble in water and biodegradable. PVA exhibits excellent mechanical and barrier properties and good biocompatibility and high thermal stability. It is also compatible with starch [45, 46]. The physical properties of PVA, such as resistance, water solubility, thermal characteristics and gas permeability vary

with the degree of crystallinity, which is highly dependent on the degree of hydrolysis and average molecular weight of the polymer [45]. PVA can be blended with starch. It can be completely degraded by a soil bacteria *pseudomonas* but degradation of pure PVA is very slow under the presence of plasticizer glycerol. It is the largest synthetic water-soluble material produced in the world and biodegradable in nature and it is well suited for making blends with starch. Thus, starch/PVA blend is the most popular biodegradable polymer but it still lacks in mechanical strength and is hydrophilic in nature [46, 47]. The blend with 70% PVA and 30% starch shows the best mechanical strength. On addition of Nano-filler the mechanical strength has increased significantly in all the samples. Sample with 70% starch shows maximum increase in strength compared to the unfilled sample [48]. The PVOH content has an important impact on the rate of starch degradation increasing the amount of PVOH will decrease degradation rate [47].

Starch and PLA

Starch is abundant in plants such as maize, wheat, potato, rice, and pea, and is thus cheap and readily available. Physical treatment of native starch, i.e., disruption of its granular structure, produces thermoplastic starch (TPS) which unfortunately has poor mechanical properties and is unsuitable for use as a polymer matrix. Thus, it is predominantly used as cheap filler for biodegradable polymers. Alternatively, PLA (produced from the chemical treatment of native starch and sugar) is naturally biodegradable and compostable. PLA has excellent mechanical properties and can be processed using ordinary equipment, making it a promising biocompatible replacement for several petrol-based plastics [49]. Jozsef Gabor Kovacs *et al.* concluded that high starch content in starch-PLA blend leads to fast degradation which further leads to shorter shelf life of product [43].

Starch-PCL Blend

Aliphatic polyesters are usually biodegradable thermoplastic polymers with good processability, thermal stability, excellent mechanical properties, good water resistance, and dimensional stability. To achieve improved properties of blends, the compatibility between natural polymers and aliphatic polyesters has to be enhanced, and considerable progress has been made in this regard [50]. Among all biodegradable polymeric materials that are nowadays commercially available, blends based on destructured starch and hydrophobic synthetic polymers, such as aliphatic polyesters, have received increasing industrial attention for using as commodity plastics [47]. Polycaprolactone (PCL) is biodegradable synthetic Polymer. For improving further the processing and mechanical properties, PCL is usually modified by blending with resins or the type of starch used in the polymer blend significantly contributes to the blend's resulting properties [51].

Starch-PHB blend and Starch-PHBV Blend

It was shown that poly(hydroxyalkanoate)s can form miscible blends with polymers which contain an appropriate functional group i.e. capable of hydrogen bonding or donor-acceptor interactions. The properties of blend films with various proportions of starch are identical. A single glass transition temperature is obtained for all the samples, which are semi-crystalline. The tensile strength was optimum for a

PHB/starch ratio of 70/30 (% wt/wt). In this particular case, an advantageous cost reduction and an improvement of mechanical properties compared to pure PHB are obtained.

Economical & Ecological Aspect Of Biodegradable Packaging

From the industry, the advantage of using biodegradable polymers derived from renewable feed stocks due to their low cost. At a first glance, biodegradable polymers lead to be a win-win opportunity for the economy and the environment. However, it is the situation with environmental issues; a sharp look at the cost-performance ratio of biodegradable polymer must be taken in order to make reasonable economic decisions [52]. According to Leaversuch. cost is always at higher side for synthetically derived biodegradable plastic materials when they are directly compared with their conventional counterparts. As the case with any new material, manufacturers and Businessman must expect a minimum of two years of losses before a profit is returned [53]. Salmoral reported a number of major chemical companies and innovators are gaining interest in developing biopolymer technologies used to manufacture products from renewable resources [54]. Tharanathan reported that synthetic plastics will never be totally replaced by biodegradable materials. However, he believes that in niche markets where the development is possible, there exists an opportunity for manufacturers to find a large profit [55].

All manufactured products, including their packaging, have impacts on the environment that includes:

- a) Consumption of non-renewable resources (e.g. materials and energy);
- b) Generation of air emissions in production, transport and use that contribute to air pollution, Including ozone layer depletion and global warming;
- c) Generation of waterborne emissions that contribute to pollution of waterways; and
- d) Production of solid waste requiring disposal in landfill.

Some primary packaging products have relatively high recycling rates:

- Aluminium Cans - 68%
- Glass Containers - 42%
- Steel Cans - 40%
- HDPE Milk Bottles - 51%
- PET Containers - 30%
- Packaging Paper - 77%.

Governments are also taking interest in the environmental aspects of packaging, seeking to reduce waste and increase recovery and recycling rates. Yet, governments are also increasingly interested in the communication aspects of packaging, either because of the perceived lack of appropriate information (e.g. front-of-pack nutrient labelling on food products) or because of the perceived power of on-pack communication and branding to promote goods where they might wish to see sales restricted (e.g. on cigarettes) [55].

Future Aspect of Biodegradable Packaging

Biodegradable polymers are a part of the larger overall plastics market. Typically, bio- plastics are either bio-based or biodegradable, although some materials are both. The biodegradable polymers market is still young and very

small, but the numbers are off the charts in terms of expected demand growth and potential for these materials in the coming years. Food packaging, dishes and cutlery constitute a major market for the product because these materials can be composted with the food waste without sorting, which is a huge benefit to the waste management effort and to reducing food waste and packaging disposal in landfills. Increasing legislation and consumer pressures are also encouraging retailers and manufacturers to seek out these biodegradable products and materials. Packaging industry has nearly 66% shares in bio-based biodegradable plastic market which is more than above other ones.

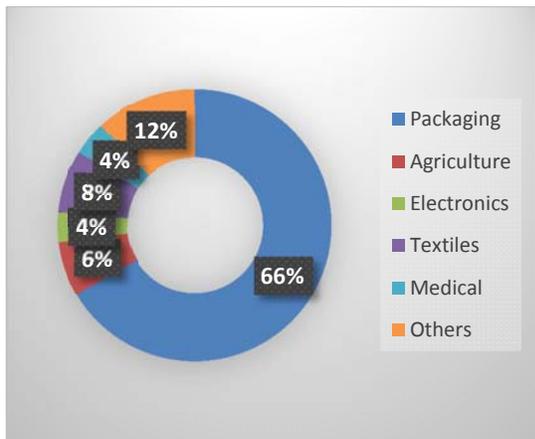


Fig 12: Bio-Based Biodegradable Plastics Market (By Applications, 2016)

It is expected that the growth of bio-based plastic is high in 2020 in almost complete world with the highest growth in Europe followed by USA then South America and then Asia.

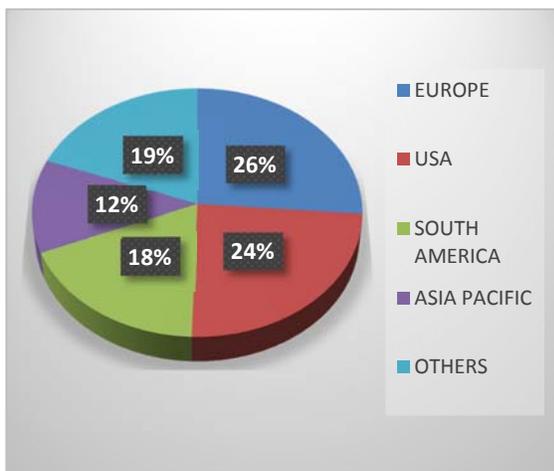


Fig 13: Bio-based plastic-2020

The rapid urbanization and the expected dramatic growth of Indian middle class from 50 million to 583 million by 2025 will increase the overall consumption of packed products enormously. The Indian retail market is already the fifth largest retail destination globally and has been ranked the second most attractive emerging market for investments. Moreover, foreign direct investment in Multi Brand Retail will fuel the packaging industry in the coming years [FICCI, IBF, ^[14].

Conclusion

Man life style at the dawn of 21st century is so much dependent on the use of plastics mainly in the packaging sector. Every day consumers make decisions about what to buy, and how to buy it, based on a number of factors—value, quality and convenience. An increasing number of consumers are also trying to evaluate the environmental consequences of purchasing certain products and the packages in which they are contained. As per the future perspective, the concept of biodegradable packaging becomes an extremely important global objective. Hence, the above review describes the different biodegradable materials which can play role directly or indirectly in development of biodegradable packaging.

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