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**Ajay Yadav**

Scientist (Food Technology),  
ICAR-Central Institute of  
Agricultural Engineering  
Nabibagh, Berasia Road,  
Bhopal, Madhya Pradesh, India

**S Mangaraj**

Senior Scientist (AS&PE),  
ICAR-Central Institute of  
Agricultural Engineering,  
Nabibagh, Bhopal, Madhya  
Pradesh, India

**Ranjeet Singh**

Senior Scientist (AS&PE),  
ICAR-Central Institute of Post-  
Harvest Engineering and  
Technology, PAU, Ludhiana  
Punjab, India

**Sanjay Kumar Das**

Professor and Head  
Department of Agriculture  
Processing and Food Engineering  
OUAT, Bhubneshwar, Odisha,  
India

**Naveen Kumar M**

Research Scholar, ICAR-  
Central Institute of Agricultural  
Engineering, Bhopal, Madhya  
Pradesh, India

**Simran Arora**

Simran Arora, Research Scholar,  
ICAR-Central Institute of Post-  
Harvest Engineering and  
Technology, PAU, Ludhiana  
Punjab, India

**Correspondence**

**Ranjeet Singh**

Senior Scientist (AS&PE),  
ICAR-Central Institute of Post-  
Harvest Engineering and  
Technology, PAU, Ludhiana  
Punjab, India

## Biopolymers as packaging material in food and allied industry

**Ajay Yadav, S Mangaraj, Ranjeet Singh, Sanjay Kumar Das, Naveen Kumar M and Simran Arora**

### Abstract

For last 50 years plastics are widely used for manufacturing of packaging materials because of their performance and ease in production. With the advent of food processing industries there is a great demand for petroleum based packaging materials for food applications. However increased use of plastics has created serious ecological problems to the environment because of their resistance to biodegradation. Biopolymers can be used as a solution to the problems posed by plastics as they easily degrade in the environment and also mimic the properties of conventional polymers. Biopolymers are polymers extracted from biomass, synthesized from bio-derived monomers and produced from microorganisms. There are different film formation methods for biopolymers like solution casting method, melt mix method, electro spinning method, thermo pressing and casting, extrusion blown film method. This review highlights the different kinds of biodegradable polymers, their characteristics with special emphasis on their market potential for food packaging applications.

**Keywords:** biopolymer, film properties, film production, packaging, modified atmosphere packaging

### Introduction

Today, polymers are an integral part of contemporary life because of their desirable properties including stability, resilience and ease in production. Worldwide production of plastics was approximately 322 million tons in 2015 which is 3.5% increase as compared to 2014 (European Bioplastics, 2016) [1]. In 2014-15, India produced 8.3 million tons of plastics (Government of India, 2015) [2]. At present, about 99% of all plastic materials are manufactured by the petrochemical industries (European Commission, 2013) [3]. In India, about 43% of annually produced synthetic polymers are utilized by packaging industry which is more than the world average of 39%. Production and processing of plastics are energy exhaustive processes; leads to increased emissions of greenhouse gases of enormous magnitude contributing to global warming. Moreover, plastics on burning release venomous emissions such as carbon monoxide, chlorine, hydrochloric acid, dioxin, furans, amines, benzene, 1,3-butadiene, and acetaldehyde which possess threat to environment as well as to public health (Smith, 2005) [4]. Waste generated from the plastics has been a pressing problem for many years because of their resistance to degradation (European Commission, 2013) [3]. In recent years bioplastics have emerged as an alternative to curb the menace caused by the plastics. The European Bioplastics Organization state that a plastic material is defined as a bioplastic if it is either biobased, biodegradable, or features both properties. The need of replacement for the petroleum based plastic with bio based polymers is impartial because producing conventional plastics consumes 65% more energy, unsustainable (due to environmental problems) and emits 30-80% higher greenhouse gases than bioplastics (Ahvenainen, 2003; Halley, 2002) [5, 6]. Biodegradable polymers are produced from renewable sources, are complete biodegradable and mimic the properties of conventional polymers like polyethylene (PE), polypropylene (PP), polyethylene terephthalate (PET) etc. Thus biopolymers in the form of packaging materials are key innovations that can help in reducing the environmental impact of plastic production (Maharana *et al.* 2009) [7].

### Production and application statistics of bioplastics

The global market for biodegradable polymers reached 206 million pounds at an average annual growth rate (AAGR) of 12.6% in 2010 and is expected to rise further by several folds in next ten years (Auras *et al.* 2005) [8] According to a study by Institute of Bioplastics and Bio

composites (2016), bioplastics production increased from 1.6 to 2.0 million tons during the period 2013-2015, biobased non-degradable polymers had the share of 0.9-1.3 tons and biodegradable plastics 0.6-0.7 million tons and it may attain 1.7 million tons by 2020. The shares of biobased non-degradable and biodegradable plastics were 63.7% and 36.3%, respectively. Majority of biodegradable plastics are made up of PLA (10.9%), biodegradable polyesters (10.8%), biodegradable starch blends (9.4%) and PHA (3.6%). In bioplastics production, Asia contributes 63.1%, North America 13.5%, Europe 13.0% and South America 10.0%.

Mostly biodegradable bioplastics are used for flexible packaging and non-degradable bioplastics are used for rigid packaging. The future of bioplastics focuses on the market for compostable, semi-durable and durable bioplastics used in consumer and industrial applications (European Bioplastics, 2016) [9]. Biodegradable polymers can be used for modified atmospheric storage (MAP) of fruits and vegetables instead of conventional polymers. Fig.1 represents the global scenario of bioplastics application in different sectors at present and a future estimation.

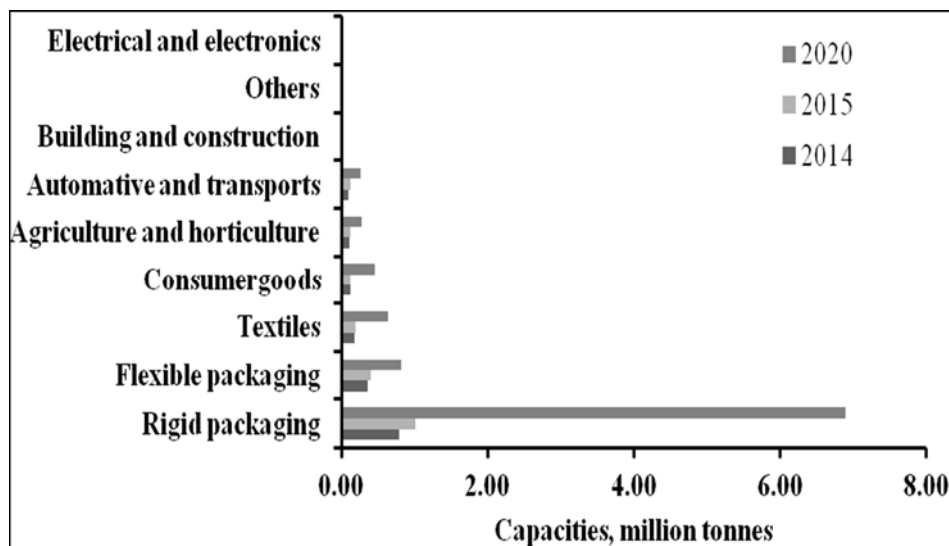


Fig 1: Application of bioplastics in different sectors (European Bioplastics, 2016) [9]

### Biopolymers and their potential as a packaging material

Packaging is an integral component of the food processing sector. Food packaging is a combination of art, science and technology of enclosing a product for achieving safe transportation and distribution of the products in wholesome conditions to the users at least price (Robertson, 2012) [10]. Most of the conventional packaging materials are products of petro chemicals like PVC, PET, polystyrene (PS), polypropylene (PP), polyamide (PA) (Averous and Pollet, 2012) [12]. The only problem with synthetic polymers is their resistance to degradation in the environment (Webb *et al.* 2013) [11]. According to ASTM standards D-5488-94d, biodegradable is defined as capable of undergoing decomposition into carbon dioxide, methane, water, inorganic compounds and biomass. With increased awareness on sustainability, the packaging industries around the globe are looking for biopolymers as the replacement of synthetic polymer. Biopolymers may be defined as the polymers that are biodegradable by the enzymatic action of microbes. In last two decades a lot of research has been done on biopolymers for food packaging applications (Averous and Pollet, 2012; Mangaraj *et al.* 2015) [12, 13].

Based on the researches biopolymer based packaging materials may be divided into three main groups based on their origin and production as shown in Fig. 2 (Averous and Pollet, 2012) [12].

**Group 1:** This constitutes polymers which are directly extracted or removed from biomass. Certain polysaccharides such as starch, cellulose and proteins like casein and gluten constitute this category. All these are, by nature, hydrophilic and somewhat crystalline and create problems while processing. Besides, the performances are also poor especially in relation to packaging of moist food products. However,

their excellent gas barrier properties make them suitable for their utilization in food packaging industry (Averous and Pollet, 2012) [12].

**Group 2:** This includes polymeric materials which are synthesized by a classical polymerization procedure such as aliphatic aromatic copolymers, aliphatic polyesters, polylactide, aliphatic copolymer (CPLA), using renewable bio-based monomers such as poly (lactic acid) and oil-based monomers like poly-caprolactones. A good example of polymer produced by classical chemical synthesis using renewable bio-based monomers is polylactic acid (PLA), a biopolyester polymerized from lactic acid monomers. PLA may be formed into blown film, injected mould objects and coating, all together explaining why PLA is first novel bio-based material produced at commercial scale (Averous and Pollet, 2012) [12].

**Group 3:** Polymers which are produced by microorganisms or genetically modified bacteria constitute this group. Till date, this group of bio-based polymers consists mainly of the polyhydroxy-alkanoates, but developments with bacterial cellulose and other polysaccharides are also in progress (Weber, 2000) [14].

### Starch based biopolymers

Starches are low cost polysaccharides, abundantly available and one of the cheapest groups of biodegradable polymers. It is also known hydrocolloid biopolymer. Various kinds of starches like potato, cassava, rice, corn and tapioca are used for the preparation of biopolymers (Ezeoha and Ezenwanne, 2013) [17]. Starch is usually used as a thermoplastic. It is plasticized through destructuration in presence of specific amounts of water or plasticizers and heat and then it is extruded. Starches are poor resistance to moisture and their

mechanical property restricts their uses. To improve these properties starches are blended with various biopolymers and

certain additives. The list of research carried out on starch based biopolymers is given in Table 1.

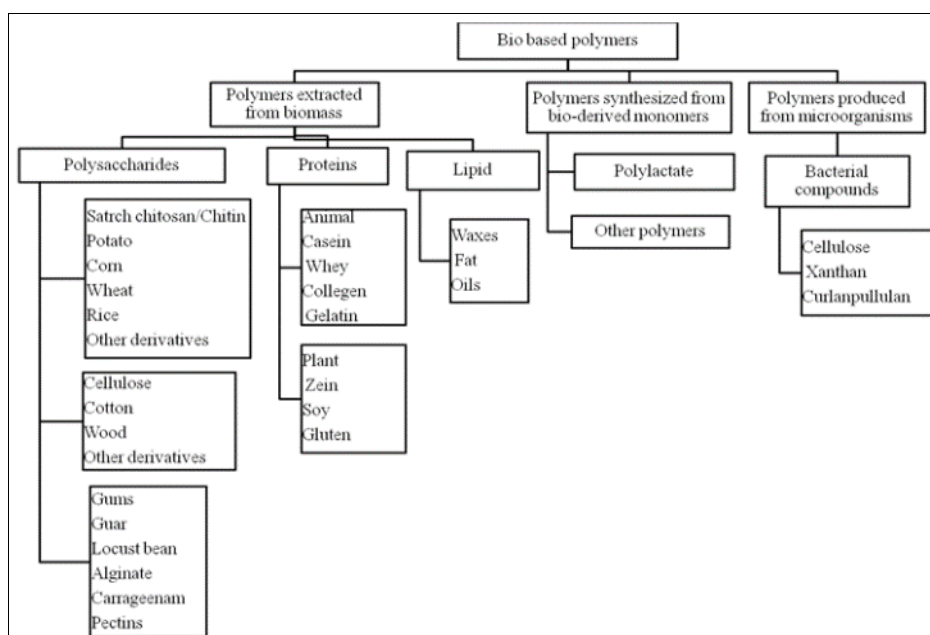
**Table 1:** The list of researches has been carried out on starch based biopolymers

Type of starch blend	Film formation method	Film characterization	Observation	Reference
Starch blended with chitosan and gelatin	Solution casting method	FTRI, SEM	Have highest density and low moisture uptake. Exhibited smooth surface with no visible pores and less agglomerates.	(Aini, 2010) [18]
Wheat starch based antimicrobial films incorporated with lauric acid and chitosan	Solution casting method	FTIR, XRD, SEM	Exhibited a growth inhibition for <i>B. subtilis</i> but not effective for <i>E. coli</i> .	(Salleh <i>et al.</i> 2009) [19]
Corn starch blend with PVOH	Solution casting method	SEM	The biodegradability of films made from corn starch was high in soil and compost and by enzymes also.	(Azahari <i>et al.</i> 2011) [20]
Thermoplastic starch and chitosan	Melt mix method	SEM, FTRI	Incorporation of chitosan resulted in increase of tensile strength (up to 85%).	(Tome <i>et al.</i> 2012) [21]
PVA/Oxidized Starch (OS) Fibres	Electrospinning technique	SEM, FTIR, XRD and DSC.	The average diameter of fibres was lowered by changing the solution concentration and weight ratio (PVA/OS). The fibres were irregular and interspersed with shuttle shape bead at the weight ratio (PVA/OS) below 1:3.	(Wang <i>et al.</i> 2011) [22]
Rice starch/flour	Casting	SEM analysis	Preparing edible films from rice flour is a new alternative for using this raw material, which is sometimes much cheaper than commercial starches.	(Dias <i>et al.</i> 2010) [23]
Rice starch/ polyvinyl alcohol (PVA)	Casting	SEM, FTIR	Films made up of rice starch and PVA with a ratio of 2:8 showed highest TS. Films made up of rice starch and PVA and sugar with a ratio of 1:8:1 showed highest TS and E <sub>b</sub> .	(Parvin <i>et al.</i> 2010) [24]

**Polylactic acid (PLA)**

PLA is one of the biopolymer that has gained lot of attention in recent years because of its economic and commercial viability during processing. Poly (lactic acid) (PLA) belongs to the family of aliphatic polyesters made up from alpha-hydroxyacids, including polyglycolic acid or polymandelic (Donald, 2001) [25]. The polylactic acid (PLA) is obtained from the controlled depolymerization of the lactic acid monomer obtained from the fermentation of sugar feedstock, corn, etc., which are readily biodegradable. PLA is becoming a growing alternative as a green food packaging material because it was found that in many circumstances its performance was better than synthetic plastic materials (Auras, 2005) [8].

Properties that make PLA a good food packaging material are their high molecular weight, water solubility resistance, good process ability i. e. easy to process by thermoforming and biodegradability (Averous and Pollet, 2012) [12]. PLA has the tensile strength modulus, flavor and odor barrier of polyethylene and PET or flexible PVC; the temperature stability and process ability of polystyrene; and the printability and grease resistance of polyethylene. Processed PLA comes in the form of films, containers and coatings for paper and paper boards. Although PLA seems to be potential biodegradable polymer to be utilized in packaging of various food products, it exhibits certain limitations in unmodified form, viz. it is more brittle and degrades easily at substantial temperature rise. The list of patents on starch and PLA based biopolymers is shown in Table 2.



**Fig 2:** Different categories of bio-based materials (Weber *et al.* 2002) [16]

**Table 2:** Patent searches in Starches and PLA based Biopolymers

Patent No.	Topic	References
EP 2712889 A1	Starch-based biodegradable material	(Law <i>et al.</i> 2014) [26]
US 8188185 B2	Biodegradable packaging film made from TPS/PLA blend	(Wang and Funk, 2012) [27]
US 8133558 B2	Poly lactic acid blown film and method of manufacturing	(Tweed and McDaniel, 2012) [28]
US 20110135912 A1	Biodegradable packaging materials with enhanced oxygen barrier performance	(Xu, 2012) [29]
US 8263197 B2	Poly lactic acid shrink films and methods of casting same	(Tweed <i>et al.</i> 2012) [30]
US 6987138 B2	Biodegradable poly lactide resin composition	(Tokawa and Raku, 2006) [31]

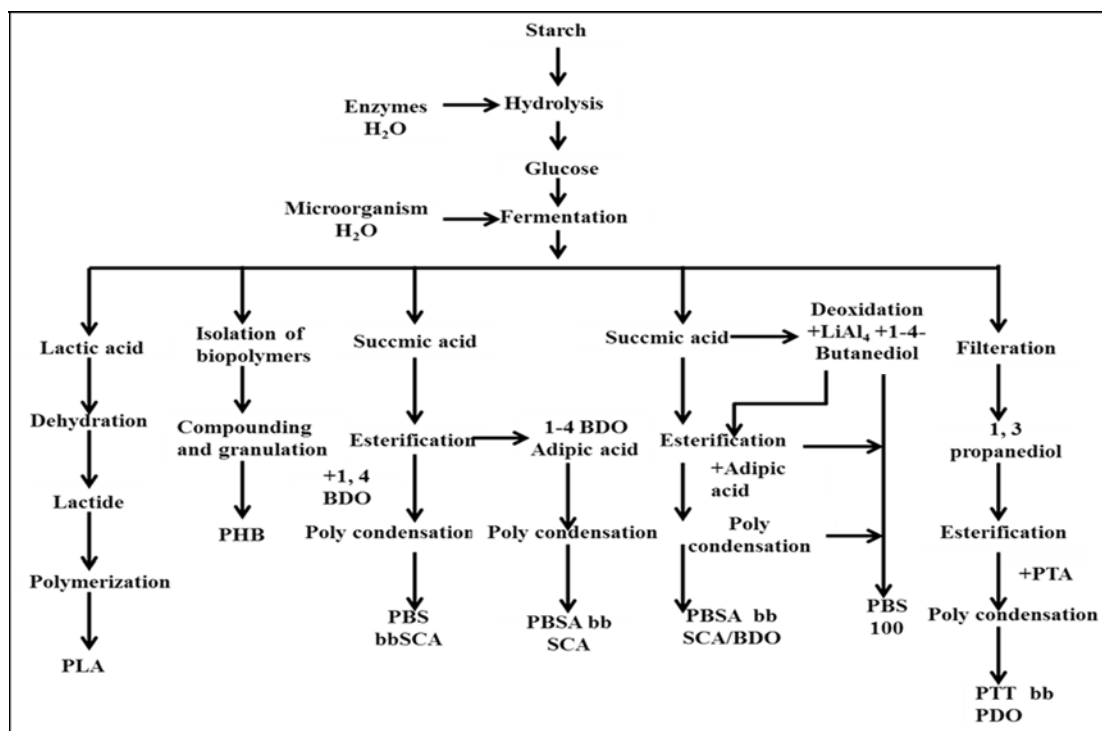
### Polyhydroxyalkanoates (PHA)

Polyhydroxyalkanoates (PHAs), a family of bacterial polyesters, are formed and accumulated by various bacterial species under unbalanced growth conditions. These polymers are produced in nature by bacterial fermentation of sugar and lipids. Structurally, PHAs comprise simple macromolecules composed of 3-hydroxy fatty acid monomers. In 2008, approximately 55115.57 tons of PHAs were commercially produced. PHAs have thermo mechanical properties similar to synthetic polymers such as polypropylene (Galego *et al.* 2000) [32]. PHA polyesters are biodegradable, biocompatible and can be obtained from renewable resources (Steinbuche and Fuchtenbusch, 1998) [33]. They have several desirable properties such as petroleum displacement and greenhouse gas minimization apart from their fully biodegradable nature (Tripathi *et al.* 2014) [15]. Applications of PHA as a biodegradable packaging include bottles, containers, sheets, films, laminates, fibers and coatings. Over 100 monomers and copolymers can be developed from PHAs. PHAs exhibit good tensile strength, printability, flavor and odor barrier properties, heat sealability, grease and oil resistance,

temperature stability and are easy to dye, which boosts its application in food industry. [15] For example, Metabolix, a US-based company, produces “Metabolix PHA”, which is a blend of polyhydroxybutyrate (PHB) and poly (3-hydroxyoctanoate) that has been approved by the FDA for production of food additives and making packages that maintain all the performance characteristics of non-degradable plastics (Sasikala and Ramana, 1996) [34].

### Pathways for synthesis of biopolymers

Renewable sources like agriculture feed stocks (starches) act as a precursor for the synthesis of various biopolymers through enzymes and microbial fermentation. A schematic flow chart for the synthesis of biopolymers is shown in Fig. 3. Commercial manufacturing and application of Biofilms Starch, PLA and their blends are commercially manufactured with different trade names especially in the developed countries. Some of the company’s manufacturing biodegradable films and their commercial applications are shown in Table 3-4.

**Fig 3:** Schematic flowchart for synthesis of different biopolymer from starches (European Bioplastics, 2016) [1]**Table 3:** List of companies manufacturing biodegradable films

Starch based film (Trade name/Company /Country)	PLA based (Trade name/Company /Country)	PLA/starch based (Trade name/Company /Country)
Plantic (Plantic Technologies, USA)	Ingeo (Nature Works, USA)	Ecovio® (Nature Works, USA)
Solanyl (Rodenburg Biopolymers, Japan)	PURAC (PURAC Co., Thailand)	Bio-Flex® (BASF, Germany)
Bioplast (Biotec, USA)	BIOFRONT (Teijin, Japan)	Plantic (Plantic Co., USA)
Biopar (Biop, Germany)	HiSun (Revoda, Canada)	Biolice m(Limagrain, France)
Mater Bi (Novamont, USA)	Pyramid (Tate & Lyle, Denmark)	Compole (Japan corn starch, Japan)

**Table 4:** Biopolymers use in commercial packaging

Rigid Packaging			Flexible packaging		
Conventional plastic materials	Alternate Bio based materials	Items that can be manufactured	Conventional plastic materials	Alternate bio based materials	Items that can be Manufactured
PET	Bio-PET	Bottles for carbonated beverages	PS/PP	PLA/Blends of PLA/Bio-PET	clear films for fruits and vegetables
PP	Bio-PP	Cups for yoghurt	PS/PP	PLA/PHB	Coffee capsule/pouches
PET, PP,PS	Bio-PP, PLA	Trays	PS	PBAT	Shrink films
PE,PP	PLA blends	Caps	PE,	Bio-PE	Stretch films
PET, PS	Cellulose, starch blends	Cutlery	PE, PP, PS	Blends of PLA/PHA/PBAT	Shopping/waste bags

### Application of biofilm for MA Packaging

Modified atmosphere packaging is food preservation techniques in which the O<sub>2</sub> concentration is reduced and CO<sub>2</sub> concentration is increased to reduce the overall metabolic processes, there by extend the shelf life of the commodities. It is an economical and simple technique for extending the shelf life with preserving quality of fruits and vegetables. Researchers have successfully used petroleum based film for enhancing the storage life of various commodities (Mangaraj *et al.* 2015) [13]. In this study some relevant research work carried out on modified atmosphere packaging of fruits and vegetables using biodegradable polymers are listed and presented in Table 5.

### Methods for manufacturing biodegradable films

Manufacturing of biopolymers is a multi-step process that requires proper skill and thorough understanding of behavior of bio polymers during processing. Biopolymers can be processed into varieties of products (packaging films, laminated paper, films, trays, cups cutlery items) depending upon the processing route (cast films, blow moulding, coextruded films). The fundamental step of processing of any

biopolymer involves melting the biopolymer mix followed by casting, extrusion, blow molding, depending upon the material to be made.

### Life cycle assessment of biopolymers

LCA is an instrument to measure the sustainability and performance of a material with environment. Life cycle of biopolymer mimics the LCA of a biomass (Rossia *et al.* 2015) [42]. Like biomass, biopolymers are degraded into carbon and water upon degradation by the action of enzymes and microbes. LCA of biopolymer is shown in Fig. 4. Biodegradation rate of different biopolymers is shown in Fig 5. Biodegradation rate is calculated as the CO<sub>2</sub> released during analysis, divided by the theoretical CO<sub>2</sub> contained in the sample; <sup>Ed</sup>K is a parameter used to quantitatively evaluate the potential biodegradability of biodegradable polymers in the natural environment. Natural soil samples are inoculated into bioreactors, and rates of biodegradation of reference materials are determined over a 2-week period. Starch and polyethylene are used as reference materials to define the <sup>Ed</sup>K values of 100 and 0, respectively. Values are determined using the ISO 14852 method, detecting the evolved carbon dioxide as an analytical parameter.

**Table 5:** Showing researches on biodegradable films used in MAP of fruits and vegetables

S. No	Commodity	Biodegradable material	Observation	Reference
1.	Sweet cherries	PLA	<ul style="list-style-type: none"> <li>This study investigated the changes in quality of sweet cherries during MAP with polyethylene and biodegradable films, film 1 (25 µm thickness; O<sub>2</sub> TR 3000 cm<sup>3</sup> m<sup>-2</sup> day<sup>-1</sup> atm<sup>-1</sup>) and film 2 (25 µm thickness O<sub>2</sub> TR 900 cm<sup>3</sup> m<sup>-2</sup> day<sup>-1</sup> atm<sup>-1</sup>).</li> <li>More CO<sub>2</sub> concentration was observed in film 1 (6.9%) after 15 days of storage.</li> <li>A slight increment was observed in storage under MAP condition (for both films 1 and 2).</li> <li>Film 2 had higher permeability to CO<sub>2</sub> and barrier to O<sub>2</sub> and it resulted good quality of fruits in terms of color, acidity and firmness.</li> </ul>	(Giacalone and V. Chiabrando, 2013) [35]
3.	Plum Tomato	PLA and polyolefin films	<ul style="list-style-type: none"> <li>Plum tomato (cultivar <i>Iride</i>) was packaged in three different plastic films, a commercially available polyolefinic film and two biodegradable films, and stored at market conditions (15 °C and 75% RH).</li> <li>Results showed that the use of packaging films with high barrier properties speeded up the quality decay kinetics of the investigated produce.</li> <li>The biodegradable films with an appropriate permeability coefficient could be advantageously used to prevent contamination from both microorganisms and insects, without reducing the shelf life of the produce.</li> </ul>	(Muratore <i>et al.</i> 2006) [36]
4.	Mushrooms ( <i>Agaricusbisporus</i> L.)	PVC film, Paper, paper coated with wheat gluten.	<ul style="list-style-type: none"> <li>The effect of different packaging films (paper, PVC, paper coated with wheat gluten) on quality of mushrooms during the storage (20 °C and 80% relative humidity) was investigated.</li> <li>It was observed that mushroom stored in wheat gluten coated paper offered more shelf life (3 days) and it retained the product color, texture and unbroken veils as compared to that stored in PVC film and the shelf life in PVC film was limited to one day.</li> <li>This shelf life extension of mushroom was attributed to storage at medium CO<sub>2</sub> (9.5 kPa) and low O<sub>2</sub> (2.5 kPa) partial pressure, without</li> </ul>	(Guillaume <i>et al.</i> 2010) [37]

			condensation.	
5.	cabbage tomatoes, sweet corn and blueberries	Laminate of chitosan – cellulose and polycaprolactone.	<ul style="list-style-type: none"> <li>The effect of temperature (10-25 °C) on gas permeability coefficients (O<sub>2</sub>, CO<sub>2</sub> and N<sub>2</sub>) of biodegradable laminates was evaluated.</li> <li>This study reveals that gas permeability coefficients increased with increasing temperature.</li> <li>The biodegradable laminate was found suitable as a packaging material for storage of fresh produce.</li> </ul>	(Makino and T. Hirata, 1997) [38]
6.	Mango	Chitosan based film	<ul style="list-style-type: none"> <li>The effects of chitosan based biodegradable film and LDPE film on quality of mango stored at a temperature 27±1 °C and 65% RH were studied.</li> <li>The mango fruits were kept in carton boxes and top surfaces covered with biodegradable film (MAP1) and LDPE (MAP2) and were compared with control (samples kept as such).</li> <li>MAP2 fruits showed lower weight loss (3.5%) compared to MAP1 (7.5%), which was attributed to reduction in transpiration of water vapour in LDPE films.</li> <li>The pH of the fruits was 4.06, but as the fruits ripened, the pH of control fruits increased to 6.73 on day 12, which was higher compared to MAP1 (5.04) and MAP2 (5.79) fruits.</li> </ul>	(Srinivasa <i>et al.</i> 2002) [39]
7.	Green pepper	PLA	<ul style="list-style-type: none"> <li>The effects of PLA based biodegradable packaging, LDPE film packaging and perforated LDPE packaging on the microbial and physiochemical quality of green pepper were compared. Study was conducted at 10 °C for 10 days.</li> <li>The levels of coliform bacteria were increased by less than 1log CFU/g (0.2 log CFU/g) in the biodegradable film packaging, whereas it was 2.3 log CFU/g in LDPE film package, and less than 1log CFU/g (0.9 log CFU/g) in the perforated LDPE film package, after 7 days storage period.</li> <li>The weight loss values for green pepper at the end of 7 days period were 2.46±0.6%, 0.38±0.1%, and 1.59±0.4% for PLA, LDPE, and perforated LDPE film packaging, respectively.</li> <li>The O<sub>2</sub> and CO<sub>2</sub> concentrations at 7 days of storage were 11.6±4% and 5.6±2% for PLA film, and 14.8± 3% and 2.3±1% for LDPE film, respectively.</li> </ul>	(Koide, 2007) [40]
8.	Fruit salad	Cellulose based NatureFlex™ NVS INNOVIA Films and VC999 BioPack PLA coated with a barrier of pure silicon oxide (SiO <sub>x</sub> )	<ul style="list-style-type: none"> <li>In this study minimally processed apple and pear mixed salad samples were packed under MAP in PP containers and sealed by different polymers (BOPP Propa fresh™ P2GAF, Amcor Agrifresh film, Biodegradable Nature Flex™ NVS INNOVIA Film and VC999 Bio Pack PLA film coated with a barrier of pure silicon oxide (SiO<sub>x</sub>)) and stored at 4.0±0.5 °C up to 10 days.</li> <li>The mass losses as evaporated water permeated through VC999 Bio Pack PLA lidding film was observed to be the highest (0.46%) compared to other materials.</li> <li>CO<sub>2</sub> content in containers sealed by BOPP Propafresh™ P2GAF film, Amcor Agrifresh film, Nature Flex™ NVS INNOVIA Film pouch and VC999 Bio Pack PLA film achieved 18, 13, 15 and 9%, respectively. There was no increase in CO<sub>2</sub> (1-3%) content for containers sealed with PP and this was maintained during storage period of 10 days.</li> <li>It was concluded that biodegradable films are good alternatives to conventional polymers for packaging fruit salads.</li> </ul>	(Krasnova <i>et al.</i> 2012) [41]

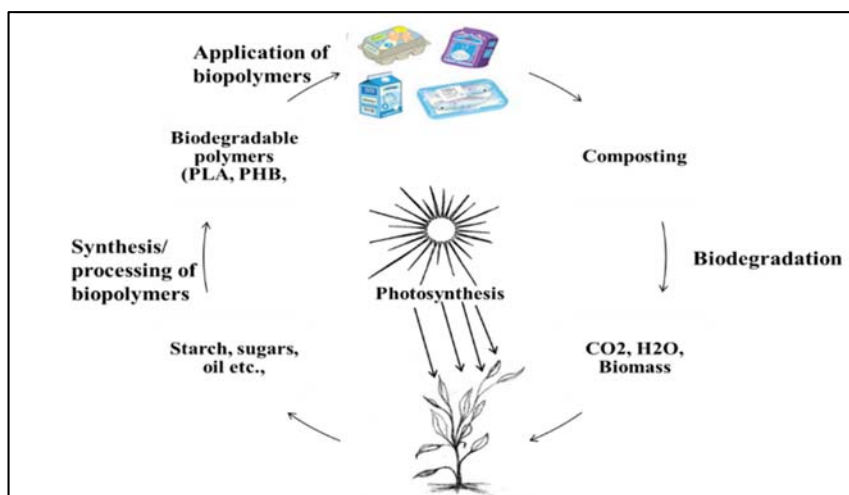


Fig 4: Life cycle assessment of biopolymers

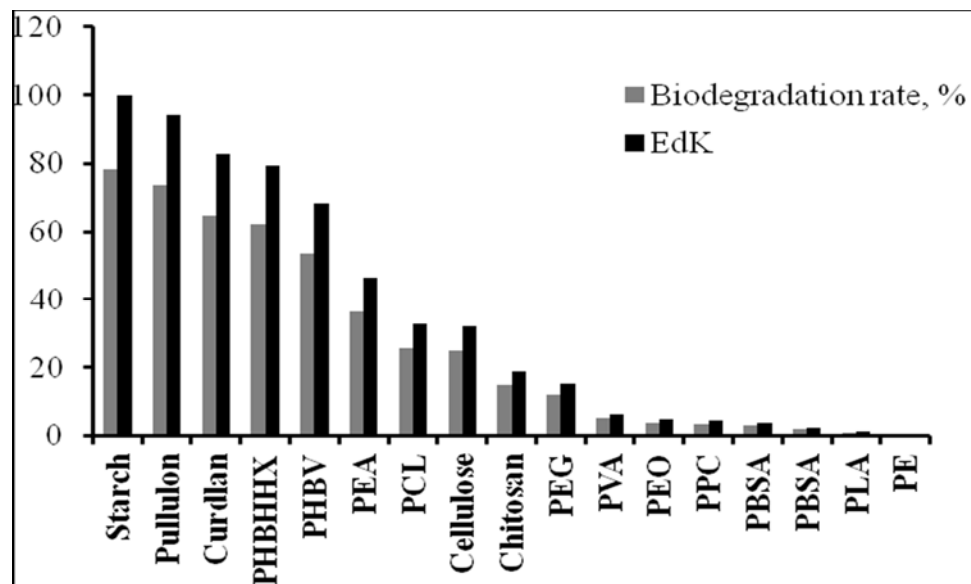


Fig 5: Biodegradation rate and <sup>EdK</sup> values of different biodegradable biopolymers (Webb, 2013)<sup>[11]</sup>

### Conclusions

Biopolymers help in reducing the environmental impact of plastic production, processing and in a way lead towards green economy. As biodegradable films are made from renewable feed stocks, agricultural waste, there is a great opportunity for research work in harnessing this economic opportunity. But still, the biodegradable polymer at present only replaces about 1% of the plastics. Therefore, need of the hour is that basic and applied researches have to be more focused on improving the performance (physiochemical, thermal properties), reducing the cost and improving ease in production of biopolymers. In context of food packaging the biodegradable packaging can be used for modified atmosphere packaging of high value products and niche merchandises like organic foods. However before adopting any bio based packaging for food there has to be proper studies on the interaction between food components and biopolymers during processing and storage. Future researches have to be more focused on adding value to the packaging materials, i. e. use of nanotechnology, smart sensors, etc., which will not only maintain the integrity but also communicate the information about the product to the consumers. Through biodegradable polymers there is a greater potential in better utilization of agricultural waste, addressing the problems of shortage of fossil fuel, health hazards, solid waste management and environmental issues as possessed by plastics.

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