

## REVIEW ARTICLE

# Innovations in Starch Based Bioplastics: A Comprehensive Review of Methods and Applications

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**ABSTRACT:** The depletion of fossil fuel resources and their associated effects, such as greenhouse gas emissions, global warming, and environmental pollution, have prompted researchers to explore the bioeconomy in innovative ways. The concepts of sustainability and the circular economy have shifted the focus of research from mere development to development that also prioritizes environmental protection, known as sustainable development. Conventional plastics, derived from petroleum, can take hundreds of years to degrade naturally, thus harming the local environment. Bioplastics, which are biopolymers sourced from biological materials, offer the advantage of biodegradability in natural environments and are considered a virtuous replacement for conventional plastics. Starch, an abundant and low-cost biopolymer, is a promising material for bioplastics. However, starch-based bioplastics often fall short in practical applications due to limited mechanical and barrier properties. Effective solutions include copolymerization, the addition of fillers, plasticization, and chemical modification. This review focuses on starch-based bioplastics, enhanced with cellulose, polylactic acid, and polyhydroxyalkanoates, and their applications in food packaging, medical, and electronic sectors.

**Keywords:** Bioplastics, Starch, Food Packaging, Nanoparticles

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## 1. INTRODUCTION

History reveals that with the advent of a new era, new inventions and technologies emerge, and the research continues to develop and enhance them for the betterment of human survival. Humans have shifted away from environmental consciousness in the name of advancement, and as a result of these avaricious actions, the environment is currently deteriorating around the planet. With the introduction of industrial revolution in the 18<sup>th</sup> century, humans have destroyed nature in all the possible ways and now the nature reverts everything back, which in all scenarios will lead to havoc in the nearby future. One of the major culprits behind all these is petroleum-based plastics. It was in the year 1907 that the first synthetic plastic 'Bakelite' was

developed by Leo Hendrik Baekeland who used phenol and formaldehyde for its synthesis [1]. Since the onset of the 20<sup>th</sup> century, synthetic plastics have been widely commercialized to substitute products made from metal, paper, and glass. Versatile qualities like lightness, robustness, durability, stability, hydrophobicity, resistance to chemical and biological degradation, transparency and low cost, are all desired features which have favored them over all other products [2]. They are chemically and structurally moldable into a variety of shapes and strengths including fibers and thin films. Most commonly used synthetic plastics in the world are polyethylene (PE), polyvinyl chloride (PVC), and polystyrene (PS) [3]. But the challenge arises with their synthesis and disposal. Plastic being a petroleum byproduct leads to fossil fuel depletion problem. It has high molecular weight so its degradation in the natural environment is very slow and its complex structure allows a high environmental burden [4]. Due to this, they sustain in the environment for longer period of time mainly in water bodies, soil, and landfills. Exposure to weathering cause these plastics to breakdown into microplastics and nanoplastics. These micro

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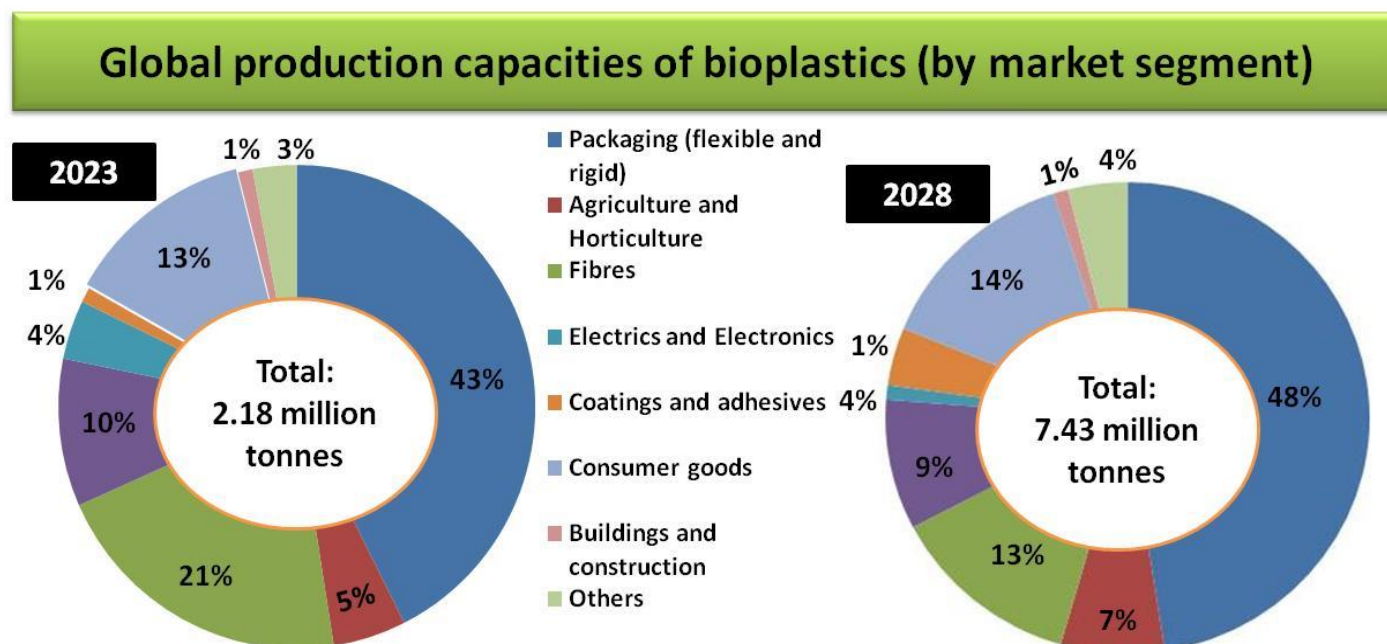
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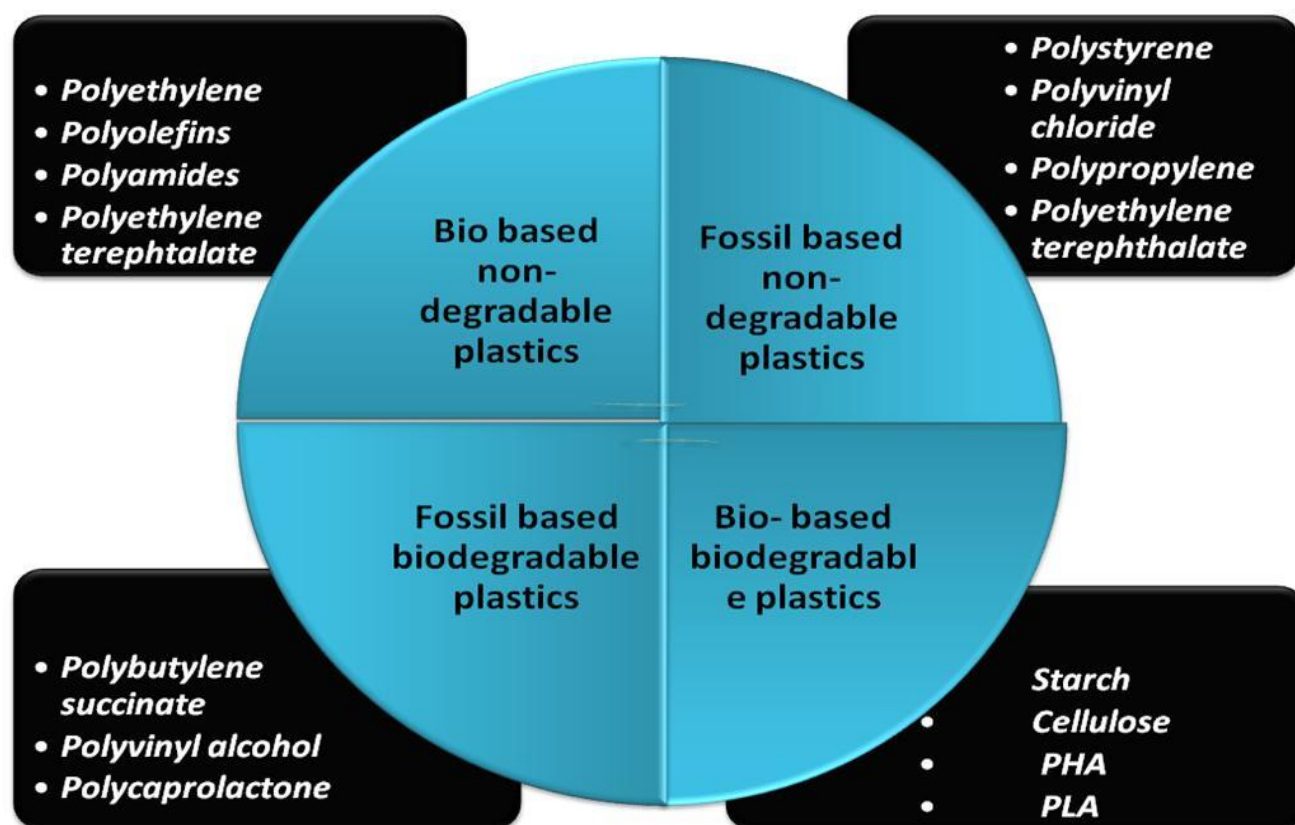
and nanoplastics are of great concern because of their penetration into the food web through aquatic to terrestrial ecosystem [5-9]. A study reveals that some traces of microplastics have been found in the placenta of a newborn baby which is actually a matter of concern [10]. More than 700 nm of these microplastics are found in human blood tested by 22 healthy donors [11]. These plastics are responsible for the death of more than 1 million seabirds and 10 million marine fauna globally [5, 12]. They also affect the concept of sustainability due to the overconsumption of fossil fuel resources and pose a serious threat to several ecological problems such as climatic change as the production of plastics are responsible for ~4% of global CO<sub>2</sub> emission [13] and the decline or disappearance of several biological diversity [14-16]. Plastic account for “25 %” of the total volume of landfills and create landfill problem [17]. These plastics are accumulated in the ocean. More than 5 trillion Plastic Pieces weighing over 250,000 tones afloat on sea and affect marine and nearby terrestrial flora and fauna [18]. The Great Pacific garbage patch (also called as Pacific trash vortex or North Pacific Garbage Patch) is a floating garbage patch in the Pacific Ocean which is visible from space. The researchers have claimed that the patch covers 1.6 million square km area entrapping a total of 45-130 thousand metric tons plastics [19]. Plastics have considerable effect on groundwater sources, as it has the ability to leach down the surface. Some studies estimated that during the Covid-19 pandemic, nearly 1.6 million tonnes of plastics were generated in a day in the form of disposal surgical face masks, face shields, syringes, medical gowns, hand sanitizer bottles, shoe covers, nitrile gloves [5,20]. Incineration of plastic

such as PVC leads to the release of harmful gases like dioxins and furans which are human carcinogens and this is widely used in food packaging [21]. Recycling alters the properties of plastic material and it is a time-consuming process. The sorting of a wide range of discarded plastic materials is very difficult. Furthermore, the existence of additives such as phthalate plasticizers and brominated flame retardants limits the use of the recycled material [22]. Some fossil-based biodegradable plastics like polybutylene succinate, polyvinyl alcohol, etc. are also an option but according to Neuling and Kaltschmitt (2017), if the present consumption rates of oil continue at the same pace, oil reserves will be consumed in the next 40-70 years and this plastic production consumes 6% of the total oil production [23]. By the year 2035 and 2050, the productivity of plastics is expected to reach 800 and 1600 million tons, respectively [24]. Due to all these drawbacks, there is an urgent need to produce a material that can be degraded naturally in the environment and has properties similar to conventional plastics [25].

Approach on ecofriendly material has caught attention of researchers all around the world and this has led to the development of a material, which is either bio-based, biodegradable or have both the attributes. Together, these are all considered bioplastics. The global market for bioplastic production is 2.18 million tonnes in 2023 and is expected to increase by 7.43 million tonnes in 2028 crossing the 2 % share of bioplastics in global plastic production [26] (Figure 1). The largest producers of bioplastics in the world are Asia and Europe [27].



**Fig. 1.** Global production capacity of bioplastics in 2023 and their expected growth in 2028 (redraw from the website of European bioplastics).



**Fig. 2.** Types of plastics based on their origin and biodegradability (redraw data from the website of European bioplastics).

The first bioplastics ‘Parkesine’ was demonstrated at the great international exhibition in London by Alexander Parkes derived from cellulose [28], and the first bioplastics from microorganism was Polyhydroxybutyrate (PHB) developed by Maurice Lemoigne in 1926 and the bacteria used was *Bacillus megaterium* [29]. Some bioplastics are produced directly from natural biopolymers including polysaccharides, lipids and protein while others are chemically processed from sugar and some of them are biologically generated by fermentation of sugar. Figure 2 represent the different types of plastics based on their origin and biodegradation.

These biopolymers are derived majorly from three routes first one is polymer derived from monomer, second one is polymer derived from microorganism and third one is polymer derived from bioresources [30]. These biopolymers have similar physicochemical properties to that of conventional petrochemicals-based plastics, with major advantages such as they do not scare crude oil, sustainable, biodegradable, non-toxic and so on (Figure 3).

Figure 4 represents the lifecycle of bioplastics. Various microorganisms and plants produce natural biopolymers, which are biocompatible and do not have adverse effects on biological systems. Biopolymers of bacterial origin are synthesized either as storage materials or as a defense mechanism. Bioplastics have been widely applied in fields such as medicine, packaging, electronics (including biosensors and capacitive materials), and textiles. Starch is one of the earliest biopolymers used in bioplastic production.

However, its weak mechanical, barrier, surface wettability, and thermal properties limit its commercial applications. This review specifically focuses on improving starch-based plastics by adding co-plasticizers, additives, copolymers, surfactants, and compatibilizers to enhance their suitability for various commercial applications.

## 2. STARCH: ORIGIN, COMPOSITION, AND PROCESSING TECHNIQUES

### 2.1 Origin and Composition

Starch is an organic chemical compound synthesized and stored by all green plants in the chloroplasts of leaves and serves as reserve food material. It is present in the different parts of the plants including seeds, roots, fruits, grains and tubers [31-32], especially in the endosperm storage tissue of cereal plant [33-34]. Starch molecule is denoted by formula  $(C_6H_{10}O_5)_n$ . Starch is basically a binary compound made up of two kind of glucose macromolecules i.e., the linear chain polymer amylose and branched chain amylopectin, which are D-glucose monomers (Figure 5), and serves as the primary storage polysaccharide in the vegetal cells. It is a white, granular, semi-crystalline organic chemical compound. Amylopectin contain the crystalline portion of the starch; the degree of crystallinity varies typically in different starch sources in the range of 15-45% [35].

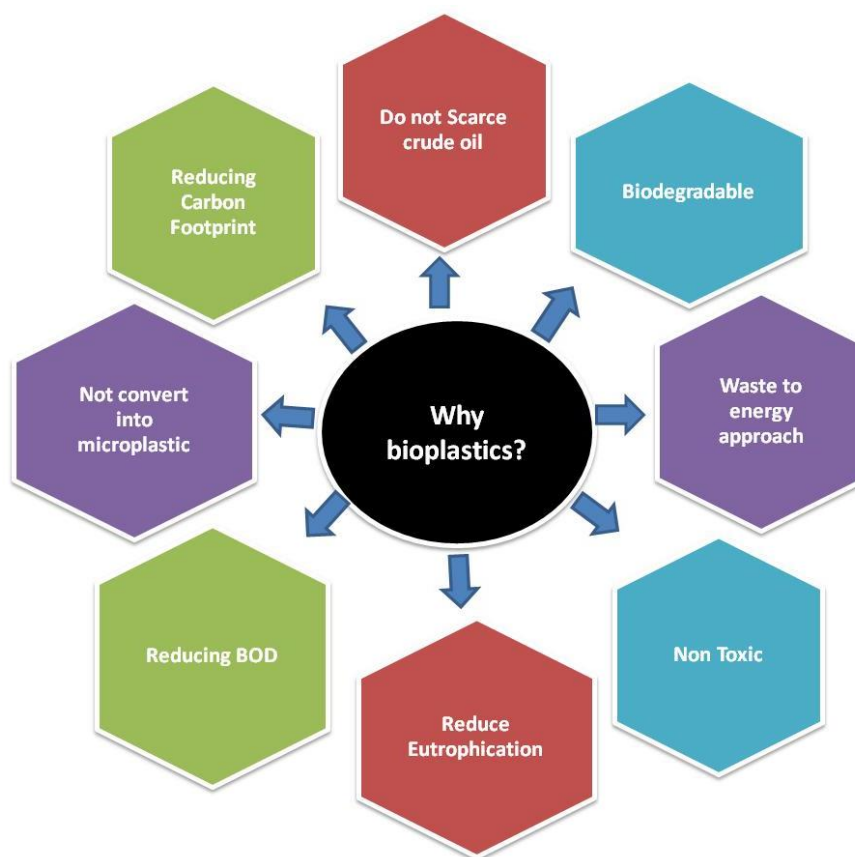


Fig. 3. Advantages of bioplastics.

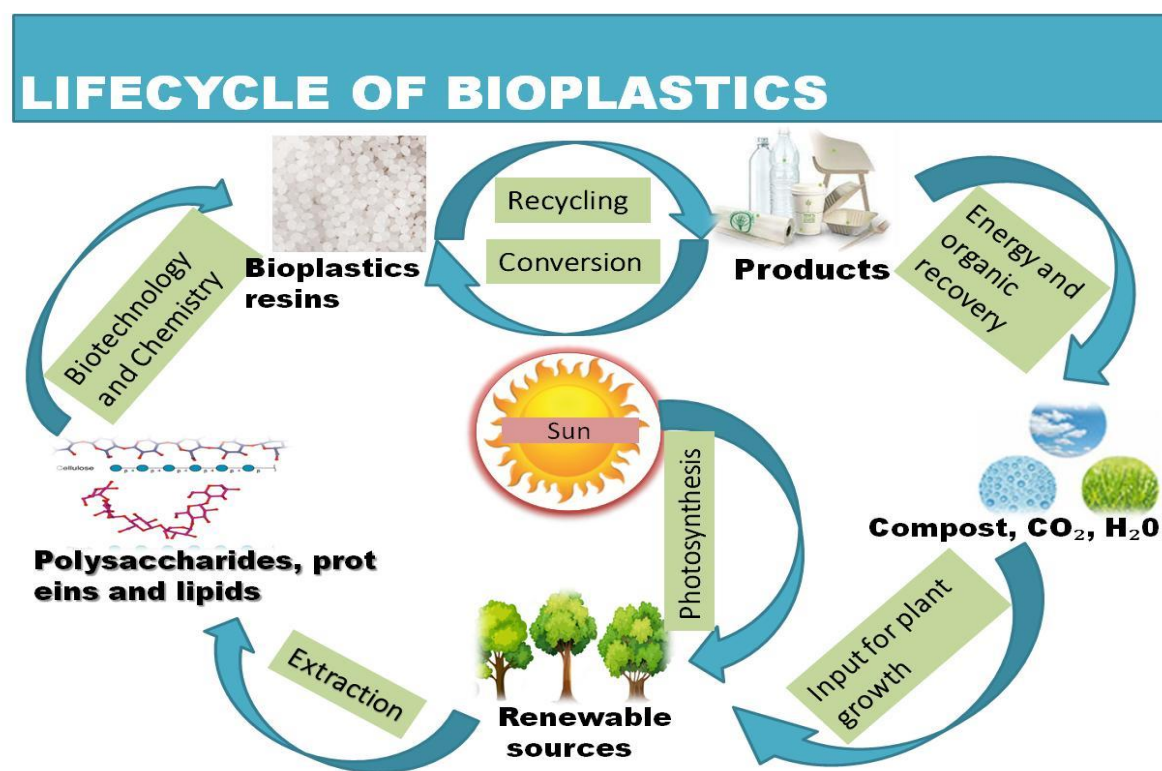
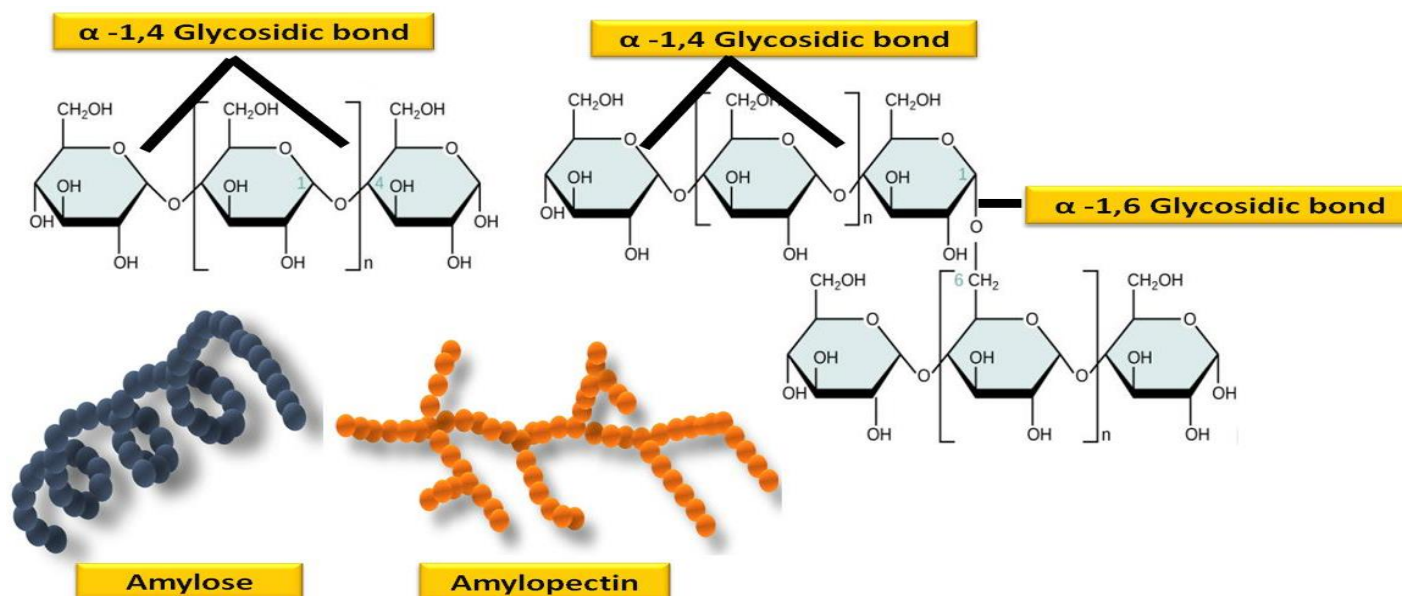


Fig. 4. Lifecycle of bioplastics.



**Fig. 5.** Representation of Amylose and Amylopectin in starch molecules. Reprinted with permission from ref. [41] Surendren, A., Mohanty A. K., Liu., and Misra, M. 2022. ‘A review of biodegradable thermoplastic starches, their blend and composites: recent developments and opportunities for single-use plastic packaging alternatives’, *Green Chemistry*, 24(220), pp. 8606-8636. Copyright@ Royal Society of Chemistry, and ref. [57] Sanyang, M. L., Ilyas, R. A., Sapuan, S. M., and Jumaidin, R. 2018. ‘Sugar palm starch-based composites for packaging applications’, *Bionanocomposites for Packaging Applications*, pp. 125-147. Copyright @ Springer Nature.

Furthermore, amylose molecules contain a central amorphous region and its periphery contains alternative units of amorphous and crystalline region [36-37]. Starch molecules contain 30% crystalline and 70% amorphous region [38]. Amylose consist of an linear unbranched chain of glucose monomers interconnected by  $\alpha$ (1-4) glycosidic bonds and amylopectin is made up of linear and branched-chain, combined together by  $\alpha$  (1-4) and  $\alpha$  (1-6) glycosidic bond. About 72–75% and 25–28%, amylopectin and amylose respectively, is the average concentration contained in starch [39]. Amylose and amylopectin have an average molecular weight of around 0.2 – 2 and 100 – 400 million Da [40]. The variability of these macromolecules differs from one source to other source. Different sources of starches are corn, rice, potato, cassava, quinoa, lentils, millets, taro, tapioca, cashew nuts, etc.

## 2.2. Processing techniques

Starch is a potential candidate in this race as it is abundantly present in the nature. For the synthesis of starch-based bioplastics, it is extracted from its source and heated with water until the solution became viscous and then dried to form a thin plastic film. Starch when heated is disrupted due to water and it started with the amorphous region of the

starch. The absorption of water by amorphous regions within the granules destabilizes their crystalline structure, resulting in the loss of birefringence. Upon continuous heating, granules tend to swell to greater extents, and the crystallites melt, resulting in an increased molecular motion that eventually leads to the complete separation of amylose and amylopectin and when it is dried, a neat film is formed due to the entanglement of disordered polymer. The semi-crystalline structure of starch is lost as the starch granules swell and burst, ultimately leading to the leaching out of amylose from the granule forming a network that holds water and increasing the mixture’s viscosity. This process is termed as gelatinization. It starts with the amorphous region that is predominantly held by the amylopectin in which the outer branches are hydrogen bonded to form crystallites unravel [47]. Thus, it makes the starch to become a paste and increases further in viscosity. The molecules can undergo significant changes when exposed to gelatinization temperatures, provoking the breakage of hydrogen bonds between starch components from the supply of heat in an aqueous solution. The preparation of starch-based films can be classified into dry and wet processes. The dry process includes molding and extrusion and the wet process includes casting (Figure 6) [48]. The wet process is usually done at laboratory scale and extrusion techniques are usually done at industrial scale. Extrusion is more reliable than casting and

thus give any shape to the desired material through polymer melt such as films, tubes and sheets.

### 2.3. Properties

Plastics can be characterized on the basis of their properties (mechanical, barrier, thermal, morphological and rheological properties) of their constituent polymers, as well as their biodegradation rate. Every source has a different type of starch-based on their ratio of amylose and amylopectin, degree of crystallinity, granular size, and structure, molecular weight, transition temperatures, and density [49-50] (Table 1). Numerous investigations have been carried out on starch granules to examine their physical and chemistry characteristics. The morphological and structural properties of the starch granules can be analyzed by the numerous characterization techniques such as Fourier transform Infrared spectroscopy (FTIR), X-ray diffraction (XRD), and X-ray scattering (XRS), scanning electron microscopy (SEM), light microscopy and atomic force microscopy (AFM) [41]. Undoubtedly, starch is a widely available, cheap, and easily biodegradable natural resource. But, starch-based bioplastics have poor physical properties such as rigidity, brittleness, fragility, low mechanical strength, water vapour

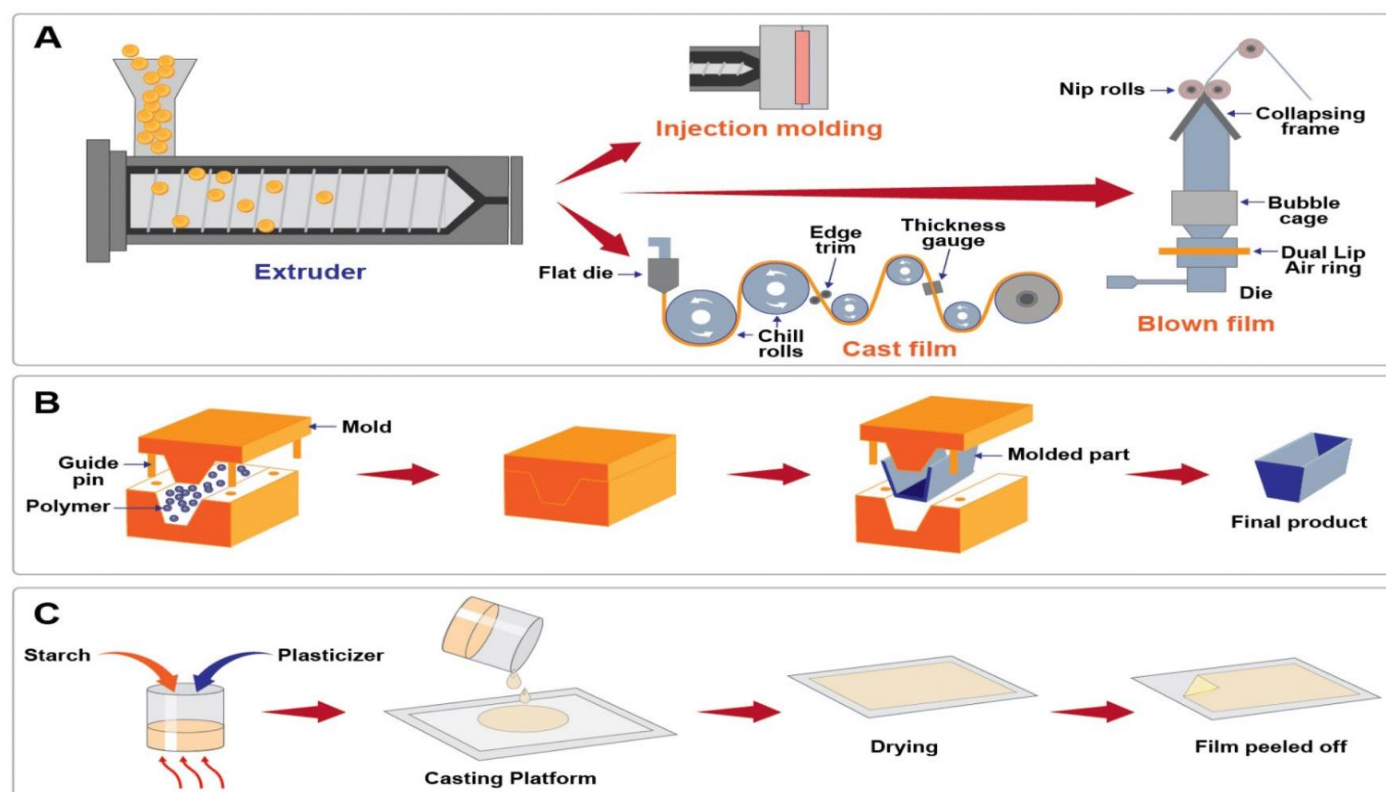
permeability (WVP), high gas permeability.

## 3. IMPROVEMENT IN STARCH BASED BIOPLASTICS

Bioplastics synthesized directly from starch have some limitations like low tensile strength and water instability. However, in order to overcome with the limitations and impede their practical applications, the addition of copolymers, fillers or additives to the starch-based bioplastics providing high water resistance and mechanical strength [31]. Different types of bioplastics have different properties depending on their polymer molecular weight, transition temperatures, degree of crystallinity, and density.

### 3.1. Plasticization of starch

The starch based bioplastic film are usually very brittle in nature and for the formulation of flexible bioplastics, plasticizers are added in the starch [51-52]. The most common examples of plasticizers used are water and polyols such as glycerol, glycol and sorbitol, citric acid, and nitrogen-containing compounds such as urea and amines [53].



**Fig. 6.** Different processes of bioplastic production. Reprinted with permission from ref. [41] Surendren, A., Mohanty A. K., Liu., and Misra, M. 2022. 'A review of biodegradable thermoplastic starches, their blend and composites: recent developments and opportunities for single-use plastic packaging alternatives', *Green Chemistry*, 24(220), pp. 8606-8636. Copyright@ Royal Society of Chemistry.

**Table.1.** Physical and Chemical properties of starch from different botanical sources.

Source	Amylose (%)	Amylopectin (%)	Crystallinity (%)	Granular size and shape	Protein (%)	Fat (%)	Density (g cm <sup>-3</sup> )	Moisture content (%)	Ref.
Wheat	18.10	72	36-39	12.37 and Polyhedral, Lenticular	0.20	0.27	1.11	9.70	[41,42]
Corn	28	78	43-48	5.2 and Polyhedral	0.27	0.29	1.4	10.45	[42,43]
Potato	17.8	76-84	23-53	7.14 and Ellipsoidal	1.35	1.0	-	15.6	[41,44]
Cassava	16-21	81.4	13	5-25 and Semi-spherical	1.35	1.0	1.5	8-10	[44,45]
Sago	30	77	23.09	6.9	0.19	0.10	0.76	13.9	[46]
Rice	22	72	38	< 20 and Polyhedral	0.33	0.34	1.282	11.24	[41]

Higher flexibility of the bioplastics is due to an increase in the interstitial volume of the polymeric matrix due to the plasticizer effect [54]. Therefore, it decreases the glass transition temperature. The plasticization process involves different physical and chemical interactions such as, starch granule expansion, gelatinization, water diffusion and polymer melting. Thus, it increases the molecular mobility, water vapour permeability, elongation and hydrophilic degree of the bioplastics [55]. Bioplastics plasticized with glycerol showed lower resistance capacity and higher elongation than sorbitol [56], and are a better contender in the search for effective bioplastic.

### 3.2. Thermoplastic starch and its blends

Native starch-based films are inherently brittle due to their high glass transition temperature (T<sub>g</sub>) [41]. The brittleness further increases during the starch retrogradation process, resulting in poor melt processing capabilities. Therefore, to produce thermoplastic starch (TPS), native starch must be modified through processes such as oxidation, esterification, and etherification. These modifications involve reacting the free hydroxyl groups of glucose monomers with functional groups such as acid anhydrides, epoxy, ethylenic, and chloro compounds [58-59]. Studies have shown that bioplastics made with starch containing 100% amylose yield better results compared to those made from native starch, which contains both amylose and amylopectin. One method to achieve 100% amylose starch is through transgenic modification of plants [38].

With these chemical modifications, native starch can be applied in various sectors, including pharmaceuticals, the food industry, and drug delivery systems (Figure 7) [59]. However, plasticization alone cannot render native starch-based polymers thermoplastic unless copolymerized with a strong polymer or chemically modified. Gelatinization induces thermoplastic properties in starch for bioplastic formation through the addition of plasticizers. This process

alters the original configuration of starch by swelling and transforming it into a melted gel similar to conventional thermoplastics [54]. The manufacturing of bioplastics from thermoplastic starch involves depositing the gelatinized filmogenic solution onto a non-adherent surface. The solution is then heated and dehydrated in an oven, facilitating retrogradation due to increased intramolecular interactions between polymers as the volume of the polymeric matrix reduces [54].

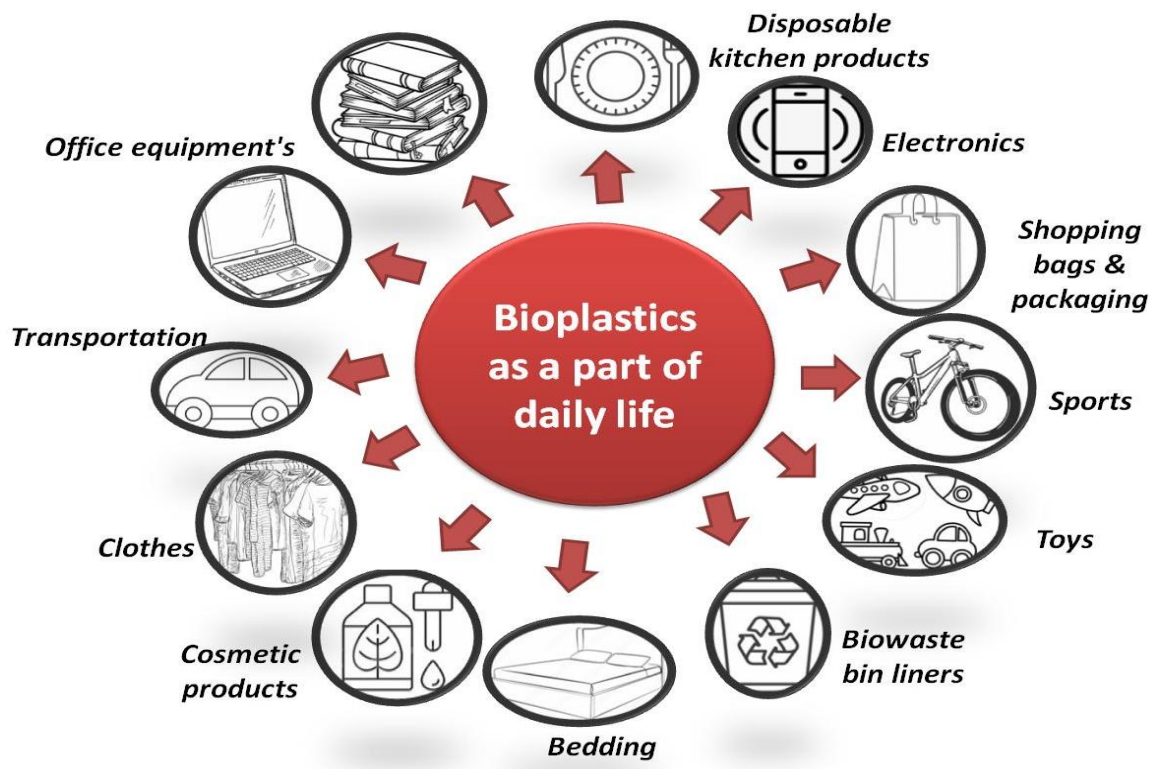
### 3.3. Copolymerization with other polymer

Some bioplastics can be produced by blending thermoplastic starch with polylactic acid (PLA), polyhydroxyalkanoates (PHA), polybutylene adipate-co-terephthalate (PBAT), polycaprolactone (PCL), and poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) and also lignocellulosic waste as a reinforcing agent [60], and have the potential to be applied on an industrial level through injection molding, foaming, film blowing, blow molding and extrusion [61-64].

#### 3.3.1. Cellulose@starch blend

Cellulose is the most abundant polysaccharide found in nature and has a high degree of crystallization and polymerization [65]. Cellulose-based bioplastic contains excellent mechanical strength, biodegradability, and flexibility. This range varies with different feedstock. It is mostly present in lignocellulosic waste such as wood, rice, wheat, stubble, straw, etc. The cellulose-based bioplastic can exhibit a tensile strength of 400-1000 MPa and high roughness of 30 MJ m<sup>-3</sup> due to the presence of hydroxyl groups on fibrillated cellulose [66].

Cellulose-based nanopapers can show an optical transmittance of about 70% in visible spectra and mechanical stress of 223 MPa (Mega Pascal) [67].



**Fig. 7.** Applications of bioplastics in various fields.

Apart from this, cellulose-based nanopapers produce a smooth surface with high gas barrier properties and low thermal conductivities (oxygen permeability of less than  $40 \text{ cm}^3 \mu\text{m m}^{-2} \text{ day atm}$ ). Derivate of the cellulose that has been used in bioplastic synthesis was cellulose nanocrystals (CNC) [68], cellulose nanofibres (CNF) [69], cellulose acetate butyrate [70], cellulose acetate [71] and bio-PE [72]. However, cellulose has no plasticity feature and hence its use in the production of bioplastic requires some modification. Moreover, it is very challenging to remove cellulose from its source. Various pretreatment methods are required to separate cellulose from lignin and hemicellulose. On the other hand, starch is very cheap, abundantly available biopolymer, therefore blending of starch with cellulose resulting into a cost-effective and having better properties than starch based bioplastic film.

### 3.3.2. PLA@ starch blend

PLA is linear aliphatic thermoplastic polyester produced through two routes either through chemical synthesis or microbial synthesis. Generally, microbial synthesis is preferred over chemical synthesis due to its high purity. PLA is produced from the microbial fermentation of carbohydrates to lactic acid and its subsequent polymerization. PLA characteristics are similar to low density polyethylene (LDPE), high-density polyethylene (HDPE), polystyrene (PS) and polyethylene terephthalate (PET). The bacterium mainly used

is *lactobacillus*. The basic monomer of PLA is 2-hydroxy propionic acid which is also known as lactic acid. PLA is synthesized through polymerization of D-, L-, and DL- lactic acid monomer or by ring-opening polymerization of lactide and is classified into poly-D-lactic acid (PDLA), propionic acid which is also known as lactic acid. PLA is poly-L-lactic acid (PLLA), poly-DL-lactic acid [73]. PLA has excellent properties like high tensile strength, high transparency, water vapour barrier properties, ultraviolet resistance, and hydrophobicity [73]. PLA monomers can be semi crystalline or amorphous in their solid state on the basis of their enantiomer's composition [74]. Because of its antimicrobial property, excellent tensile strength, stiffness, and high transparency, PLA is very suitable to be applied in food and beverage industries [75], and it is also included in Generally Recognized as Safe (GRAS) by the Food and Drug Administration (FDA) [76]. The main drawbacks of PLA are its high cost, brittleness, slow crystallization rate, and low resistance to heat [77-78]. Studies have shown a steady increase in PLA-based bioplastics by blending with other biodegradable material like starch. This has ultimately made it cheaper and improved its slow degradation rate. Research has claimed that adding plasticizer in further PLA blended starch enhances properties such as crystallization, ductility, and toughness.

### 3.3.3. PHAs@ starch blend

PHA are polyester, produced by numerous groups of



microorganisms as lipid inclusions intracellularly in the form of granules for energy storage [79], and then harvested using solvent extraction techniques by using solvents such as chloroform, methylene chloride, or propylene chloride. PHA is stored by various species of gram-positive and gram-negative bacteria. These bacteria store PHA within their cytoplasm in the size range of 0.2 to 0.5  $\mu\text{m}$  [80]. The simpler form of PHA is (PHB). PHB and its copolymers have similar characteristics to that of conventional plastics such as polypropylene (PP) [81]. PHAs can be accumulated up to 90% (dry weight) within the cells. Studies showed that 90 genera of microbial species accumulate PHA under both aerobic and anaerobic conditions. Approximately 150 different monomer components of PHA contain branched, straight, aromatic, saturated, and unsaturated structures [80]. Bacteria can be categorized into two classes with respect to the production of PHAs. In the first class, bacteria are kept in a nutrient-deficient environment (such as phosphorous, nitrogen, oxygen, or magnesium) with excess carbon source to accumulate PHAs, and they do not accumulate PHAs during the growth phase. The second class of bacteria accumulates PHAs during the growth phase and does not require any nutrient limitation [17]. For example, bacteria *Ralstonia eutropha*, *Pseudomonas oleovorans* and *Pseudomonas putida* accumulate PHA during nutrient limiting condition while recombinant *Escherichia coli* accumulate PHA without nutrient limiting conditions [80].

Structural variations in monomers constituting PHAs, leads to differentiation in their properties and chemical composition as homo or copolymers. Good resistance to moisture and excellent barrier properties to gases is shown by PHB which is very similar to PP, PET and PVC. PHAs are insoluble in water, resistant to hydrolytic attack, resistant to UV, and sink in water which facilitates anaerobic biodegradation in sediments. In addition, they are biocompatible, biodegradable, behave as piezoelectric materials, and also have chiral molecules [80-82]. The type, composition, environmental conditions, and the type of microorganisms (different microorganisms produce different PHA-depolymerase to degrade PHAs) affect the PHAs' degradability [80, 82-84]. Their glass transition temperature and melting temperature varies from  $-50$  to  $4$   $^{\circ}\text{C}$  and  $40$  to  $180$   $^{\circ}\text{C}$ , respectively [80]. The type of polymer produced and the composition of the monomeric unit describes the different thermos-degradation temperature, tensile strength, Young's modulus, water vapor, and oxygen transmission rate for different PHAs [81]. PHA is synthesized using solvent extraction technique and some lipid residue remain attached with the biopolymer after extraction process and high level of purification is necessary in order to use any material in real world application and this lipid residue cause foul smell to the packaged material and destroy the product [85]. This can be improved by chemical method [86]. But, it turn out to be costly and high fermentation cost. Moreover it is brittle and has low thermal properties. Therefore, blending of PHA with plasticized starch is one of the method to improve the properties of both the polymers.

### 3.4. Additive as nanocomposite or other biocomposite

Undoubtedly, blending starch with other polymer improves some of the properties, but still there are some drawbacks to impede their practical application. i.e. in case of PLA@Starch (Starch being hydrophilic in nature and PLA being hydrophobic have a lack in compatibility. Thus, for obtaining efficient compatibility, cross linkers, essential oils and organic acids can be added) [41]. And in case of PHA@Starch (Numerous studies have been investigated on blending PHA with starch, most of them resulted in low mechanical property films, thus inorganic and natural fillers can be introduced). Moreover, integration of nano fillers such as metal based nanoparticles, carbon based nanoparticles, and nanocrystals improves the mechanical, optical, thermal and barrier properties. (Table 2)

## 4. APPLICATIONS OF BIOPLASTICS IN DIFFERENT SECTORS

### 4.1. Popular Applications of Bioplastics in the food packaging field

The need for the development of an eco-friendly material has asserted the researchers to initiate their efforts, mind, and time in process of building a suitable material for food packaging. The bans on "single-use plastics" by many governmental bodies have led to the adoption of a suitable material and in this initiative bioplastics, at the present time are front runners. Packaging has been an integral part of food packaging and in today's world, there exists no food commodity which doesn't require packaging. This packaging is present in various forms and varies for different food items [95]. For proper food packaging, it must follow these four common functions i.e., containment, protection, convenience, and communication, thus dependency of these features vary accordingly from product to product. Without these above mentioned features, product handling would be inconvenient, inefficient, and messy and would lead to a dissatisfied consumer experience.

Moreover, the main function of food packaging is to extend the food shelf life of any commodity by preventing them from the attack of micro-organisms, moisture, gases, dust, compression, and odors (Figure 8) [96]. The food that is in direct contact with plastic must follow the guidelines of widely acknowledged categories of GRAS (Generally Recognized as Safe), accepted by the EU (European Union) and FDA (Federal Dietary Allowances) [97]. It has been estimated by the EU, plastic waste comes around 60% of post-consumer plastic waste [26]. This comes mainly through food packaging.

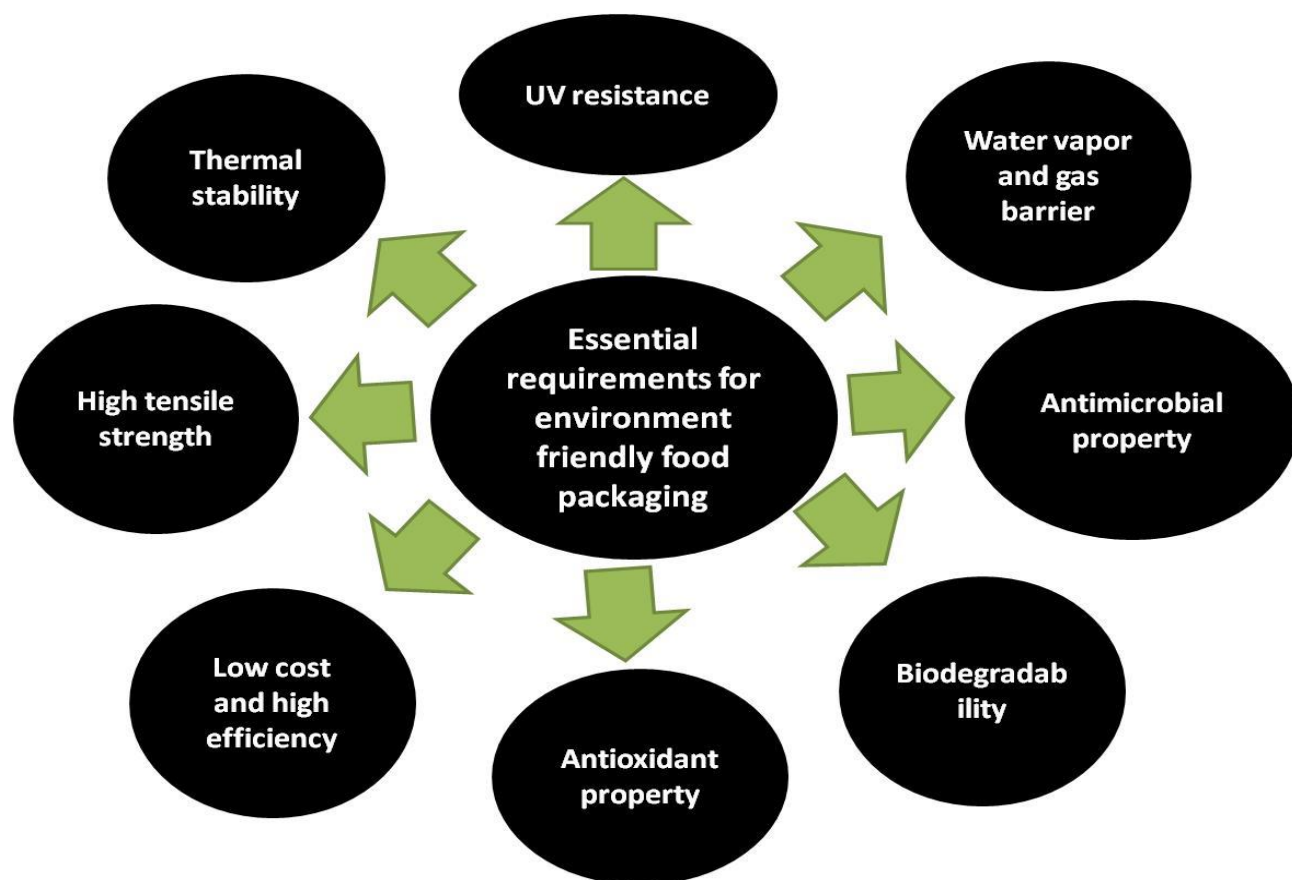
The market for bioplastics in packaging has shown a steady increase as 65% of the total bioplastic production in 2018 was employed for food packaging [98]. It is estimated that in the year 2030, the projected bioplastic market value would grow to \$324,000 million [99]. The food packaging is done either with films (edible or non-edible) or by coatings. A film can be defined as a thin stand-alone solid sheet. On

the other hand, coating is applied directly to the surface of a product, in its liquid form by brushing, spraying, dipping, or panning [100]. Edible films and coatings are the constituent of edible polymers, like polysaccharides, lipids, proteins, and food-grade additives, and hence must be non-toxic, fully digestible, and biodegradable. This makes them an integral part that can be used for covering drugs and food products

and can be consumed as it is [101]. Some of the examples of edible films and coatings are sugar coatings on drug pills, and gelatin films on soft capsules [102], coatings for cheese and fruits, edible sachets for premixed ingredients [103], and bio-based foams and hydrogels [104]. Non-edible films or coating is used in the various types of packaging products like plastic bags, plastic wraps, mulching films, etc.

**Table 2.** Starch based bio-composite films and their characteristics.

S. No.	Biofilm name/ Composite Film	Plasticiser used	Characteristics of biofilm	Ref.	
1.	Starch microparticle/Malic Acid (MA-SM)	-----	Increase tensile strength (12% starch concentration) The films showed greater hydrophobicity with increase in starch content and ester group in malic acid.	[87]	
2.	Starch/Nanocellulose	Glycerol	Tensile strength (4.68 MPa with starch + 2% CNF).	[88]	
3.	Wheat Gluten/Potato Starch (20% wt) Wheat Gluten/Corn Starch (20% wt)	Glycerol	Potato starch composite showing greater strength than corn starch biocomposite.	[89]	
4.	Corn and Rice starch TPF	Glycerol, acid	Gelatin-Citric acid	Greater tensile strength of 12.5 MPa.(7g rice starch) Higher rice starch conc. showed less water absorption and reduced water solubility.	[90]
5.	Starch (Rice/Corn/potato) + Agave Fibers	Glycerol	GSSM (Gene Site Saturation Mutagenesis) synthesized films showed a storage modulus of 2743 MPa and 874 MPa for Corn starch and potato starch respectively as compared to the values of 1409 MPa and 603 MPa for conventional made corn and potato starch films. GSSM processes decreased 18% release of CO <sub>2</sub> .	[91]	
6.	Starch (Yam Starch) + Bentonite	Glycerol	Increase in tensile strength and young's modulus with increase in bentonite concentration with values of 4.063 ± 0.12 MPa and 72.65 ± 11.42 MPa, respectively.	[92]	
7.	Rye Starch/nanorod-ZnO	Glycerol and liquid sorbitol	Decrease in WVP with higher conc. of zinc nanoparticles. Better suitable films for food packaging due to excellent UV shielding.	[93]	
8.	Potato Starch	Glycerol	Higher concentration of glycerol (30% wt) showed higher thermal stability due to formation of more hydrogen bridges with starch. Biodegradability was seen more in higher concentration of glycerol used.	[94]	



**Fig. 8.** Essential requirement for environmental friendly food packaging.

Packaging sector consumes 43% of the overall bioplastic production [30]. This leads to serious issue because packaging usage is for short span. Amylose-amylopectin ratios have a massive effect on the resulting film and this affects its use in food packaging applications [100]. Researches on starch-based films for different types of food packaging have shown promising results and are suitable alternative compared to conventional plastics. A study demonstrated that replacement of PVC films can be done through two biodegradable films based on starch i.e., one blend of starch and polyester and other blends of 3 biodegradable/biobased polyesters [105-106]. Ifezue (2009) illustrated that Mater-Bi® (a blend of starch with biodegradable synthetic polymers like PCL or PVOH) has superior mechanical properties and is suitable in packing whole fresh celery as compared to perforated LDPE, PLA, and Ecoflex [107]. Some commercial application of starch based bioplastics are shown in Table 3.

One study conducted by Soukoulis et al. in 2017 demonstrated the blending of starch with protein to form biofilms and examined its effect on *Lactobacillus rhamnosus* [108]. Rice and corn was used as feedstock for starch and bovine skin gelatin, sodium caseinate and soy protein was used as feedstock for proteins. The study was based on compositional, physiochemical and auxiliary

characterization of biofilm. The results showed an enhanced shelf life of the biofilm using 6 log practical CFU/g, and 15-24 days of extended shelf life. Another study focused on chitosan/starch blend where the starch used was modified starch and was used to test the physiochemical features of the biofilm. The formation of biofilm was done by casting technique which implied the usage of chitosan, waxy, oxidized, and acetylated corn, and their respective blends [109] Gomes et al., 2016 in his study for the preservation of cherry tomatoes after harvest, worked on *S. burchelli* phosphate starch blended edible films that were coated on them [110]. The coated tomatoes were put in a temperature of  $10 \pm 2$  °C, relative humidity of  $80 \pm 5\%$ , and a timespan of 21 days. The parameters checked during this experiment were thickness of film, solubility in water, and the permeability by water vapour. The results showed that the water permeability increases, water solubility reduces. The study established that consumable films were affected with glycerol concentrations. The results reported greater permeability of water vapour for the conservation of cherry tomatoes. Adjouman et al., (2018) examined the edible films made of 4 g cassava starch, soyabean lecithin (0-5% w/w), shelled nut oil (5-10% w/w), and glycerol (25-30% w/w), on water vapour porousness, temperature and relative humidity of 25 °C and 75% respectively [111].

**Table 3.** Commercial applications of bioplastics in packaging fields.

Source	Product	Company	Ref.
Corn based packaging	Organic tomatoes	Iper supermarkets (Italy), Coop Italia	[30]
Corn starch trays	Milk chocolates	Cadbury Schweppes, food group Marks and Spencer	[30]
TPS starch	fresh produce and meat	EverCorn, INC, Novamont, Plantic Technologies	[30]
Starch-based	trays	Cadbury Schweppes	[30]

The results showed evidently that a concentration of 25% w/w, 5% w/w and 5% w/w for glycerol, shelled nut oil and soyabean lecithin respectively showed optimal WVP.

In other recommendation, iron yam and maize starch with lemon oil as plasticizer was used in biofilm production to analyze the physiochemical properties and antimicrobial of the starch based film. The results showed a reduction in water vapour permeability, dampness reduction, solvency and malleability quality. Also, due to presence of lemon oil, the antimicrobial characteristics improved which ultimately nourishes the flavor of the packaging material [112]. In order to improve the shelf life of food or to improve its sensory properties, some treatments can be employed such as coating biofilm with edible material [113], modified atmosphere packaging [114], plasma treatment [115], chemical treatment [116], and active and intelligent packaging [117]. And also by treating biopolymer with antioxidants, antimicrobial agents.

#### 4.2. Popular Applications of Bioplastics in the Biomedical Field

Biodegradable polymers have always given the priority to be used in the biomedical field from last 50 years. These biopolymers are used in the area of tissue engineering including implants and three dimensional scaffolds, as pathway to controlled drug release, cell culture technology, wound healing and for therapeutic devices [118-120]. Different bio-based and synthetic polymers are being in existence to encapsulate and carry hemoglobin. Natural polymers can be used as an encapsulating agent of Hemoglobin based oxygen carriers because of their biocompatibility and biodegradable nature [121].

In medical sector, starch can be used as a binder, drug delivery vehicle, tissue engineering scaffold, capsule or tablet glidant and muco-adhesive drug delivery [122-125]. Native starch has very limited applications in biomedical sector due to their poor solubility and large size. Polysaccharides plays very promising role to be used in drug delivery system but the problem starts when comes to raw material cost, thus researchers have focused on starch due to their low cost and abundancy, therefore decreases the overall cost of the product. Starch alone may not be a right vehicle for carrying drugs, biomedicine etc.

Blending of starch with other polymer can be an alternative to be used it at medical applications e.g chitosan has antioxidant and antimicrobial property but can't be used

in drug delivery system due to poor solubility and high swelling ratio, thus blending chitosan with starch improves overall solubility, mechanical strength and resistance to UV and high temperature [126]. Starch nanoparticles can have a potential to be used in delivery of anti- cancer drug by tuned its hydrophilicity [127].

For the drug delivery system, biodegradable polymer should be synthesized in such a way that can self- assembly into nano-carriers. There are 2 major requirement to be used a material in a drug delivery system: safety and performance. Safety relates with the in vivo toxicity and material should not pose any threat to the immunological system. In order to use nano-carrier in a drug delivery system to deliver as a smooth carrier in the therapeutic load of a tissue specifically, it must have high surface charge ratio, low molecular weight, less aggregation of nanoparticles, amphiphilic nature and colloidal stability.

Out of all, PHA have most potential application in medical field. But the synthesized materials still lacks the quality to be applied in commercial applications and high level of purity is needed for the approval by drug administration and medical field. PHA with certain modifications has the capacity to be used in tissue engineering because of their biocompatible nature. In case of starch, modifications can be applied. One among all is cationic starch which can be produced by the etherification process and this cationic charge may provide antimicrobial property and thus used in medical field and it can be induced by adding a functional group into starch backbone chain like imino, amino, phosphonium or sulfonium. Thus, used for gene delivery because of easily digestible amylase bond [128].

#### 4.3 Bioplastics in electronic Industry

The massive and continual use of electronics all over the world leads to serious disposal problem due to its non-biodegradable nature and huge mass. Mankind generated approximately 54 MT of electronic waste until 2019 [129-130]. From the overall plastic production industry 15% contributes to the electronic sector [131]. The PCB's (Printed Circuit Board) used in electronic sector have excellent electrical, mechanical, chemical, thermal properties. The precursor used in the formation of PCB are fossil based plastic i.e. polyethylene terephthalate, polyether ether ketone, polyimide etc [129]. Therefore in order to support the concept of sustainability and green electronics there is a need

to develop a biomaterial or biodegradable polymer or their blend which has property required to be used in the electronics sector [132]. The properties required in this sector are different from that used in packaging or medical industry. PLA and its blends are found to be good use in medical and packaging industry but the same does not applied to be used in electronics industry because the material requires exceptional mechanical and electrical properties. The use of metal in the process also leads to some problem at the time of end of their life. For the conductive polymers we can use polythiophenes, polypyrroles, polyanilines and are considered as a greener option [133]. We can also use carbon based conductive material such as carbon nanotubes, quantum carbon dots, graphene oxide are thermally and electrically stable can be considered as greener options [129]. These carbon based particles can't beat metals but can effectively to be used as a filler with other polymer material. Bioplastics in electronic items can be used in mobile phone parts, speakers, mouse, air conditioners, refrigerators parts etc. Starch can be used as a raw material in printing inks due to excellent gelling properties [134-135]. Chen et al. (2019) reported the rheological characteristics of rice, potato and corn starch and their application in 3D printing and evaluated that suspension of starch had excellent extrusion processability and mechanical strength and hold high dynamic moduli and yield stress [136].

Bioplastics have limited applications in electronic and electrical industries due to the requirement of the thermal and electric properties as that of critical metals. They have usually low thermal and electrical property and most of them are considered as electric and thermal insulators and in order to make them conductive, blending is necessary, cellulose nanofibers, carbon nanotubes and graphene nanotubes can be incorporated which ultimately enhances thermal, electrical and mechanical property. Blend of PHA with PLA after coating with CNT or pyrolyzed lignin through which electronic bioplastic film can be formed by using methods such as inkjet printing, molding, extrusion, plasma exposure and dip coating. The exposure of biopolymer to argon plasma helps in improving the hydrophobicity ultimately enhances its property to be used electronic industry.

## 5. CONCLUSION

Undoubtedly, bioplastics are an efficient alternative to synthetic plastics which causes numerous harmful effects to the natural environment through various ways. For the replacement of plastics, a material possess properties such as good mechanical (tensile strength, elongation at break, young modulus), optical (UV barrier), thermal (heat tolerance), barrier (water vapour permeability, oxygen permeability), antibacterial, antifungal, antioxidant, cost effective, good efficiency, biodegradable nature. Due to abundancy and low cost native starch comes out to be a better contender in this field as it reduces the overall product cost, but it has some limitations. This can be accomplished by plasticization, copolymerization and blending. For the

thermal resistance, plasticization is an efficient technique. Plasticization improves the intermolecular adhesion between starch molecules. Likewise, copolymerization starch with cellulose, PLA and PHA improves degree of crystallinity, mechanical strength, hydrophobicity. And for the solubility, fillers and nucleating agents can be added. Furthermore, addition of waste serves several advantages i.e. valorization of waste and promote circular economy. Everyone in this world is aware with the deadly effects caused by plastic pollution, so it is duty of governmental and non-governmental agencies to invest much more on bioplastic through research and innovation in order to commercialize it at the wider scale.

## CONFLICT OF INTEREST

The authors declare that there is no conflict of interests

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