Bioplastics Production and Applications: A Mini Review

Chethan V Ghorpade[1] Swathi K S[2] Sai Charitha Shetty M M[3] Chetan C J[4] T P Krishna Murthy[5]
[1,2,3,5]Department of Biotechnology
[1,2,3,5]M S Ramaiah Institute of Technology, Bengaluru, Karnataka, India [4]Yangstofour Labs, St Mark's Road, Bangalore, India

Abstract— Ubiquitous contamination of plastics has raised growing concerns about its impact on livelihood and associated environmental risks. Globally, Food Packaging sector contributes a major share of the total plastic waste produced. Therefore, reducing plastics accumulation in the environment and the connected consequences dictates the essential need of each nation till presently. In line with this, biodegradable and/or edible plastics have been advocated as an alternative to conventional plastics and their production have shown marked increase with improved properties for its use in food and allied industries. Biodegradable plastics are the polymers of biological entities that helps to overcome the environmental impact of petroleum-based plastic, whereas, edible plastics are the one that are consumable by humans, animals and microbes without any noxious effects towards health. This review provides an insight into the sources and development of the bio-based plastics from the starch of both conventional and unconventional tubers. Optimization of process parameters for their efficient production, evaluation of physiochemical properties, moulding and application in food packaging sector are highlighted with emphasis on their antimicrobial property.

Keywords: Plastics, Petroleum-Based Plastics, Biodegradable, Edible Plastics, Bio-Based Plastics, Bioplastics

I. INTRODUCTION

Flexibility, durability and cheapness in plastics production are attributed for their increased usage. Extensive use of plastics and their improper dumping are the major reasons that are responsible for the elevation of plastic pollution in the ecosystem of plastics. Apart from polluting land and terrestrial animals, plastic imparts harm to marine organisms, thereby polluting water bodies. Plastic application in the field of agriculture and its leaching into the soil is reported to cause soil degradation. Thus, today, every country is making an effort to minimize plastic usage and accumulation to improve the environmental conditions [1].

Plastics are the wide range of synthetic and semi-synthetic materials that at some stage in manufacture (due to their plasticity), can be moulded, extruded, cast, spun or applied as coating. They are proven to be a versatile material with strong, inexpensive, lightweight, corrosion-resistant, and durable with high thermal and electrical insulation properties[2]. Parkesine was the first semi-synthetic plastic invented in 1862 in Europe and America as an alternative for the increased demand for ivory[3]. Bakelite is the first truly synthesized polymer by Leo Baekeland in 1907 and many other plastics were developed further over next few decades [2]. Invention of Nylon in 1938 with other synthetic fibres influenced the outcome of World War II, marking the beginning of a new plastic age. Statistics showed that, in 1950, an average of 1.7 kg of plastics was used by each person and by 2007, the annual consumption of plastic per capita rose up to 100 kg [3].

Conventional plastics have several advantages over metal and paper, but yet have adverse effects on human health and environment. Hence, in order to keep this industry rolling, the need for developing plastics with desired properties or raw materials is crucial. The magic terms in this context are ‘biodegradable’ and ‘edible’ plastics. Such new plastics serve as alternatives for the conventional packaging materials [3].

A. Biodegradable Plastics

Biodegradable refers to the ability of materials to break down and come back to nature within a short-time after disposal into the environment. typically, a year or less. Natural Polymers that are derived from agricultural products like starch, proteins, plant oils and cellulose are the major sources for the development of biodegradable polymer materials [1]. The ideal biodegradable plastic materials are one that is completely degradable into smaller molecules of CO_2 and HO_2 at the end. In case of commercial biodegradable plastics, the product mass should be composed of 50% of organic molecules and should not exceed the health limits in terms of heavy metals. In addition, product must possess an ability to degrade (>90%) within 6 months under controlled environmental conditions. Upon the synthesis of biodegradable plastics, various tests to examine the factors that are affecting their barrier and permeation properties, thermal and mechanical properties are carried out. A large number of analytical methods are also employed for the characterization and studying several other properties of plastics. Examples of plastics that come under the category of biodegradable plastics are Polylactic acid (PLA), Polyhydroxylalkonoates (PHAs), Polyhydroxybutyrate (PHB), Polycaprolactone (PCL), Starch and Cellulose. Studies showed that the drawbacks of these materials can be overcome by the formation of blends that enhances the properties of the matrix of the individual biodegradable plastics [4].

B. Edible Plastics

Edible plastics, as the name indicates, are consumable in nature which gives additional nutrition and calories to the consumer who consumes them. These plastics are synthesized from natural polymers *viz.*, polysaccharides, lipids and proteins (animal or plant origin) or combination of these components, thereby acting eco-friendly. The composition of edible plastics or films includes film-forming material, plasticizer, antimicrobial agents and additives [5]. They help in protecting the products from chemical, physical microbial and environmental changes [1] (Fig. 1). They can be eaten along with the product, thereby avoiding the need of extra packaging and disposal issues.

Edible plastics developed from proteins of plant origin includes wheat gluten, soy protein, sesame protein, corn zein and peanut protein, whereas, proteins of animal origin include gelatin, casein, milk whey protein, keratin, egg white protein, collagen, myofibrillar protein. Some polysaccharide materials that are tested edible in packaging include starch, cellulose, alginate, pectin, chitosan, carrageenan, pullan and gellan gum [1]. Various food manufacturing industries like the bakery industry, dairy industry, meat-poultry-sea foods industry and horticulture industry are using these plastics for packaging and protecting their products [6]. Thus, edible plastics have several advantages in improving the product safety, quality, stability and reducing the plastic waste as well as improving the convenience for the consumers by working as a wall between the food and the surrounding environment [1].

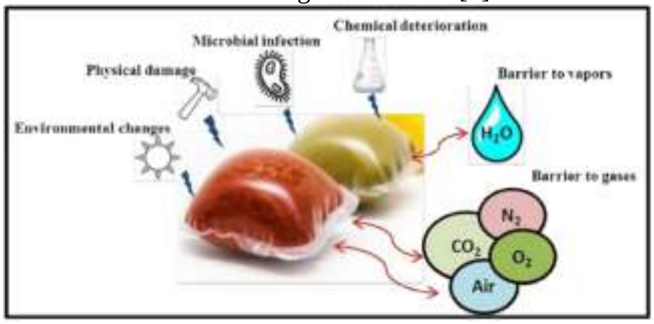

Fig. 1: Role of Edible Plastics in protection of products [1]

C. *Plastic Pollution during Covid-19 Pandemic Times*

Long before COVID-19, several tons of plastic ended up in the sea raising several threats to the environment. The main components for such a disaster include micro and nanoplastics contribute to the greater percentage of disaster in comparison to macroscopic plastics, which accounts to a very small percentage for the waste [7]. The health emergency caused by COVID-19 has highlighted the enhanced plastic use various reasons. To fight against this, large quantities of safe, inexpensive and easy to find materials were needed. The only choice available in that rush was PP and PE that come under the category of petroleum derived polymers as disposable materials. Though several biodegradable plastics were known to have been developed, they are not yet the first choice when there is a need for quick response during this pandemic. This is because these polymers are expensive, poorly known and difficult to handle and manage. In the absence of proper management plans, the massive use of such polymers, in addition to being a problem for the environment, may also be dangerous for consumers too [8].

II. BIOPLASTICS

A. *Biopolymers*

Biopolymers are natural polymers which are being synthesised in environment by the living cells. Biopolymers consist of n number of monomeric units that bond covalently to form large polymeric molecules by polymerisation [1]. Not all biopolymers are biodegradable, some of them are non-biodegradable. The use of such bio/natural polymers can be a better alternative to the use of conventional petroleum based synthetic polymers. In such way the environmental burdens like landfill waste/pollution and many other related environmental issues can be reduced [9].

1) *Classification of Biopolymers:*

There are 2 criteria to classify he biopolymers: 1) based on source and 2) based on degradability

1) Non-biodegradable and fossil based – Conventional plastics that are commonly found almost everywhere. Ex. PE, PP, PET [10].
2) Biodegradable and fossil based – Expensive and difficult to synthesize. Ex. PBAT, PCL [11].
3) Non-biodegradable and biobased – Obtained from the naturally occurring biopolymers but do not undergo natural degradation. Ex. Biobased PE, PET, PA, PTT [12].
4) Biodegradable and biobased – Obtained from the naturally occurring biopolymers and undergo natural biodegradation. Ex. PLA, PHA, PBS, Starch blends [13].

Edible biopolymers are obtained from the naturally occurring biopolymers and proven safe for animal and human's consumption without any adverse effects [14]. Their desired properties *viz.*, natural degradability, recyclability, and biocompatibility, overcome the limitations of non-biodegradability of conventional polymers. Additionally, these help in issues like reduction of contamination, producing eco-friendly products, enhancing materials recyclability, and provide better sustainability in environment [15].

Starch being the natural edible polymer, occurs in the form of granules in plant tissues. It accounts for about 80–90% of the polysaccharides that are present in the foods which are easily digestible [16]. Extraction of starch from potatoes, corn, carrot, beetroot, wheat, yam, tapioca and many other similar edible plant sources, makes them ideal for the synthesis of edible plastics (Table. 1). Many unexplored roots, shoots, tubers, rhizomes, and corns from plants are emerging as major alternative sources, which could replace many conventional starch sources like potato, corn or tapioca, and chemically modified starches [17]. Starch extraction from these sources depends mainly on type the plant source and the further applications of the extracted starch. Wet milling is the easiest and generally approached method. To facilitate the starch recovery and enhance protein removal some of the enzymes including cellulase, amylase and protease, could be used to ease purification and increase the quality of starch. several solutions can be used during the extraction in the form of steeping medium, for example, sodium chloride (NaCl), sodium metabisulphite ($Na_2S_2O_5$) etc. [18]. Starch when mixed with appropriate proportions of plasticizers and cross linkers, it can melt and casted into a thermoplastic [19].

Sl. No.	Source	Amylose (%)	Amylopectin (%)	Total starch (%)	References
1.	Banana	19±1.0	48±0.5	67±1.5	[20]
2.	Cassava	24±0.5	40±1.5	65±2.0	[21]
3.	Yam	17±0.5	41±2.5	58±3.0	[22]
4.	Sweet potato	28±1.0	24±1.5	52±2.5	[23]

5.	Corn	19±0.9	17±1.2	36±1.5	[23]
6.	Potato	7±1.5	13±1.2	26±1.3	[17]

Table 1. Starch content in different food/plant sources

B. Preparation of Bioplastics using starch

The preparation of bioplastic involves 3 main steps (Fig. 2):

- Extraction of starch from suitable source: extraction of the starch from the tubers follows isolation and purification of the extracted starch [24].
- Mixing to get bio-plastic composition: Preparation of the bioplastic composition by mixing starch, cross-linker and plasticizer in appropriate proportion [25].
- Moulding: moulding of the bio-plastic and drying process [26].

1) Extraction

The fresh tubers should be peeled off and washed carefully in water or in other solutions to remove soil and other particles referring to the previous method [27]. The tuber can be peeled, washed, chopped into tiny pieces and the pieces should be suspended in 0.1 % (W/V) $Na_2S_2O_5$ solution (sodium metabisulphite). The tubers later homogenised using a blender and then the mixture is suspended in 4 % NaCl (sodium chloride) solution. White coloured starch can be removed from the slurry by filtering through a micro sieve and was then dried in an oven in order to remove excess water at 48 h at 45 ˚C [24].

2) Mixing

The starch thus extracted is in the native form. So, during the mixing process a constant temperature of 80-90˚C must be maintained. The starch is dissolved in water and the cross linker is added and mixed well for 5-10 minutes depending on the addition of water. Note: the mixing speed and the temperature should be maintained constant. After mixing the mixture should be cooled to 45-55˚C and the plasticizer should be added and mixed well before moulding into bio-plastic [26]. Biopolymer mixture preparation can be achieved by referring to the previous method [1]. Starch in its native form is not an ideal thermoplastic, but upon treating with a cross linker (HCL) and plasticizer (like glycerol) at a temperature of 85 – 95 ˚C, it starts to form molten Thermoplastics starch. Native starch is composed of both straight chain (amylose) and branched (amylopectin). The branching in amylopectin is not suitable for the process as it disrupts the plastic formation, hence it should be treated with cross linker (HCL) helps to break the branches in amylopectin into a straight chain [28].

Fig. 2: Production of biopolymers from starch [17]

3) Moulding

In this step, the casting of bio-plastic occurs, in which it is casted to different shapes as required for the application. The major techniques including extrusion, thermoforming, solvent casting and injection moulding can be used to produce edible bioplastics. The shape and stability of the plastic such produced is majorly affected by the drying conditions [29].

Characterisation and Evaluation of Bio-plastics:

Evaluation of the bioplastics is very important to determine its application. Many factors influence the stability and strength of the bioplastics. Biodegradability, water solubility, thermal stability and mechanical properties determines the ability of bioplastics for suitable application. The surface properties can be measured and visualized with the help of X-ray Diffraction method (XRD), and microscopic evaluation. The major expected application is in the field of food packaging; hence the bioplastic produced should possess anti-bacterial properties.

4) Thickness

The thickness of the bioplastic can be determined by using a thickness gauge. It can be measured by keeping the sample (bioplastic) in between the stylus and the anvil, observing the value directly. The bioplastic should possess even thickness throughout the area, the variation in thickness may reduce the stability and strength of the bioplastic [30].

5) Mechanical strength

- Tensile strength,
- Young's modulus,
- Tensile load at break,

- Maximum extension,
- Elongation at break and
- Load at maximum extension

These properties can be determined using a universal mechanical testing machine. The samples should be cut into equal dimensions as required for the tests and weighed accurately [31].

6) Moisture Content

The moisture in the bioplastic can be measured by determining the weight loss of the bioplastics upon drying. The bioplastic can be cut into to a known dimension. The sample should be weighed and noted as wet weight. (The samples should be weighed accurately in a high accuracy weighing machine) The mass of the dry bioplastic should be recorded after drying at high temperature in an oven until it reaches a static dry weight. The bioplastic loses weight as the moisture content in the bioplastic is evaporated and it attains a constant weight at a certain point of time as all the moisture content is evaporated [30].

7) Degradability Test

The bioplastic must undergo natural bio degradation in order to overcome the current crisis over the synthetic plastics. As discussed, not all biopolymers are biodegradable hence it is important to determine the biodegradability of the material and shelf life of the material. The samples are to be cut into dimensions of 4 cm^2 and weighed accurately before taking to the test. One sample must be kept under soil and another sample in the container. Images of the samples should be captured both before and after the process just for ensuring visual inspection by Scanning Electron Microscope (SEM). The end weight of the bioplastics must be recorded and the percentage weight loss must be calculated [17].

8) Water Solubility Determination

The samples are to be cut into a known dimension, and the dry mass of the bioplastic should be accurately weighed and noted. Then bioplastics can be immersed completely in a known volume of distilled water and the solubility is observed at different temperatures. After a certain period of time the bioplastic must be taken out and dried in an oven at high temperature. The final weight of the bioplastic should be recorded and the percentage solubility has to be determined [31].

III. APPLICATIONS

Ncube et al., [32] reported that “Biopolymer global production has grown from just under 300,000 tonnes in 2009 to 2.11 million tonnes in 2019 and is predicted to grow to 2.42 million tonnes by the year 2024 and industry”. Among bioplastics, starch and its combinations form the highest share in commercial global production of bioplastics followed by PLA [33]. Bioplastics and its applications have been extensively studied in recent years as issues concerning plastic waste and their environmental impact has been increasing. Various studies and inventions have focused on developing sustainable alternatives which can successfully equivalent plastics in terms of strength and applications simultaneously having low impact on the environment. Such recent applications (Fig. 3) of bioplastics and related biopolymers are discussed below.

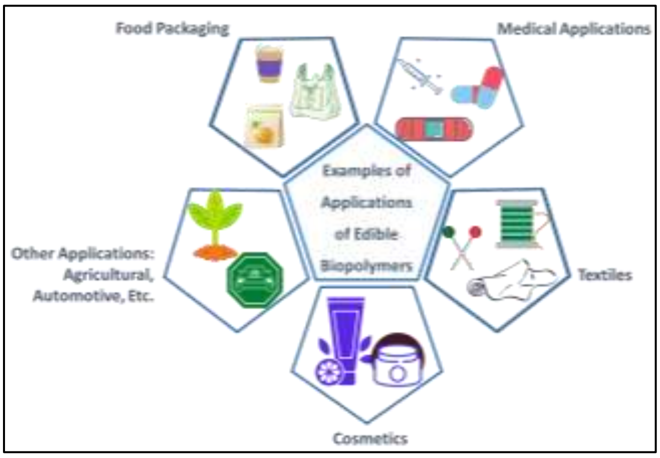

Fig. 3: Applications of Edible biopolymers in various fields

A. Food Packaging

Packaging applications in food products include primary packaging (direct contact with products), secondary and tertiary packaging used in transport of goods [33]. As packaging is the major contribution of bioplastics, which have been reported to account for 1.14 MT (million tonnes) in 2019, constituting more than 53% of total bioplastic products [32]. PLA is proposed to have all appropriate characteristics to replace synthetic plastic in the upcoming future; it has already been approved as a food contact substance by the United States Food and Drug Administration (FDA) [34]. Commercially PLA have been used to produce products like coffee capsules, cups, yogurt packaging, etc. which are available in markets [34]. Starch based plastics have seen their major function as edible thin coatings on food products to enhance their life on shelf storage [33]. Gelatin-based protein films offers protection of fresh meat and vegetable products. Biowastes of proteins from the dairy industry like sodium caseinate and whey protein concentrate were reported to be used for packaging applications of dairy products, like cheese, and other food products [35]. The film-forming ability of coffee by-product-derived polysaccharides could be directly applied to develop bioplastics. Pectic polysaccharides from coffee mucilage were used as starting material to develop biofilms with rigidity and water insolubility. Spent coffee grounds having polysaccharide rich extracts were used to yield heterogeneous light-brownish films with surface aggregates which is a drawback, but could be potentially improved by techniques like enzyme hydrolysis [36].

Starch extracted from any source; acetic acid from sources like white vinegar; citric acid from lemon juice can be proportioned to starch at ratio of 4:3; agarose and agaropectin mixture and plant-based glycerine; including 1.1% collagen and (<0.1%) thyme oil was employed used to develop a bio-plastic container (0.2 mm to 0.4 mm in material thickness). This embodiment was used to store sports nutrition supplements and is reported to be compatible with energy gels presently available, without any defect in taste or texture of the gels. This invention is expected to be useful in settings with limited fresh food access and proper waste disposal [37].

Among developing technologies, smart food packaging is an interesting emerging application, which involves functions for protection as well as for monitoring the food packaged and the surrounding environment. Such Smart food packaging application was reported in a gellan gum-based bioplastic composite with radish anthocyanins extract along with gelatin, where a change of colour from orange red-to-yellow was observed in range of pH 2 to12 [38].

In an embodiment, biodegradable and edible packaging material developed comprising gellan biopolymer and polyvinyl alcohol when analysed had tensile strength significantly higher compared to high molecular high-density Polyethylene (marketed film) and PLA. The films have also been reported to have biodegradation profile in appropriate limits, potentially large scale able, cost effective and can be developed with a variation in thickness corresponding to their intended application [39].

A hindrance in liquid food packaging (Ex: Milk or honey) is the tendency to adhere to the inside of the packaging leading to food wastage. As a solution to this drawback edible super-hydrophobic coatings have been reported studied where coffee and beeswax were fabricated through a spray coater nozzle into a micro/nano structured edible coating having super-hydrophobic characteristics [38].

B. Medical Applications

In the biomedical field, edible biopolymers have found to have potential to a large variety of applications from wound dressing, regenerative medicine, biomedical devices, 3D scaffolds for tissue engineering, and so on [33, 38]. Specific variants of biodegradable plastics have also been applied as surgical threads, surgical masks, cosmetic swabs, clamps, clips, bioresorbable implants, etc [40].

A delivery of β-carotene *via* sustained release has been observed in heat-treated soy glycinin stabilized Pickering emulsions, which were food grade gel-like applications. For applications as delivery agent to drugs having to target gastrointestinal tract, the hydrophilic and poorly soluble molecules are transported to the tract with soluble bioplastics as delivery vehicle which can disperse in acidic environment or enzyme secretion of the tract [38].

The biodegradability of bioplastics in living organisms, biocompatibility, low cytotoxicity, easy and safe disposability have seen potential applications in the areas of resorbable sutures, drug releasing gents, wound dressing products and many developments in pharmaceutical background [41].

Biopolymers have a great potential as diagnostic devices. There have been studies reported for biofilms integrated chemical signals where when in contact with the biological fluid, the signal prompts a controlled release. The biofilms have also been seen to act as a barrier, hence controlling the time between the loaded agent and media [38].

C. Textiles

Reichert et al., [42] reported that “The use of biopolymers in textile applications is possible *via* extrusion and is estimated at 100 kt per year”. Edible textiles have also been an emerging application where textiles are incorporated with edible biopolymers. Silk-like characteristics were shown by casein-based protein fibres which were also commercialized by the United States Department of Agriculture. A study also reported that the fibres could be loaded with flavours or bioactive ingredients to facilitate controlled delivery to target destinations like stomach, colon, etc. To develop antimicrobial features, herbal products (like essential oils, aloe vera etc.) have been incorporated into textile or packaging products [38]. One of the examples of applications of bioplastics in textile industry is PLA being used to produce furniture textiles and fabrics because PLA can compensate low moisture regain, smooth surface and other properties that of other traditional thermoplastics [41, 42]. Foamed PLA has been represented as a substitute to Polystyrene foam when applied as an insulator [41]. PLA yarn has been used to develop pyramid tea bags, which are also available in the market. BIOME Bioplastics (Biome Bioplastics Limited, South Hampton, United Kingdom) used bio-resins to develop a non-woven material termed BioMesh. ECOBIONET, a European union project focused on developing compostable nets (used in packaging fruits and vegetables) constituting Biodegradable polyesters, PLA, compatibilizers and plasticizers. Although PLA filaments and fabrics are internationally available on the markets, the capacity which they are found in are still narrowed [42].

D. Cosmetics and personal care

Recent years have seen emerging interest in applications of edible biopolymers in the field of cosmetics and personal care. They have been reportedly studied as thickeners, binders, conditioners, sensory materials, active ingredients, film-forming agents and so on [38]. The selection of biopolymer sources plays a major role, for example: animal sources are not often accepted in cosmetic applications [35]. PHAs have seen applications in cosmetology and everyday articles. Bio-on SpA, Italian based company, produced PHA microspheres as a biodegradable alternative to already present microspheres used in cosmetics, body washes and so on. Since production costs of PHAs are high, they are blended with other biopolymers like starch or PLA to make it cost efficient [34].

E. Other Applications

Biopolymers have been reported to have great potential in agricultural applications, biodegradable polymers that have superabsorbent properties have been studied for sustained release of specific micronutrients/pesticides for plants, saving water purposes and so on [35]. Nodax™ P(3HB-co-3HHx) and Mirel™ PHB have developed films for mulching, an agricultural practice [34]. PHA was used to produce a fishing gear which biodegraded in an aqueous environment [34].

Whole seaweeds (from genus *Eucheuma*, *Sargassum*, *Turbinaria*, or *Gracilaria)* were used as a raw material to produce bioplastics, which was also seen beneficial in saving cost. This invention has been claimed to have potential applications as an alternative to plastic-laminated paper (used in wrapping foods like burger or rice) plastic-aluminium composites (coffee sachets, cereal packaging etc.), or sheets for wrapping soap, sanitary napkins and so on but packaging liquid or semi solids is a constraint.

Protein-based bioplastics result in brittle structures with relatively low elongation property, soy-based composite materials have been proposed to have potential use in automotive industry products, due to the latter mentioned properties [35]. PLA has been reported to be popularly used in 3D printing and is also used to develop nanotubes blended with composites enhancing their mechanical properties for applications in automotive products [33].

IV. RECENT TRENDS

Nowadays, alternative green packaging focuses on newer approaches to enhance bioplastic's properties such as antimicrobial effects, biodegradability, packaging colour, tensile strength, elongation at break (EAB) and water resistance. These approaches involve incorporating bioactive compounds like vitamins, polyphenols, essential oils, plant extracts, amino acids or other composites into biofilm. In order to improve the mechanical properties other physical treatments like plasma, ultraviolet radiation, gamma irradiation or electron beams are also being assessed [43]. Recently many efforts have been made to transform bio-waste into bio-composite packaging, which could be a sustainable solution to reduce accumulating solid waste as well as an alternative renewable source for packaging materials [50].

A. Antioxidant and Antimicrobial Effects

Phenolic compounds have been observed to play an important role in antibacterial properties of Biopolymers. The review [44], reports the potential of pomegranate rind as effective antioxidant and antimicrobial additive in bioplastics because of its high phenolic content (PC) as PCs like ellagic acid has the capability to chelate vitamins, minerals or carbohydrates, making them unavailable to microbes [44]. Microbial spoilage was observed to be delayed due to inherent antimicrobial properties of coffee phenolic compounds. Phenolic-extracts acquired from coffee pulp when added to chitosan-based formulations developed yellow biofilms with increased antioxidant and antimicrobial properties [36].

Vegetable wastes or a surplus of edible plants (ex: carrots, radicchio, parsley, cauliflower, spinach and so on) were used as a starting material to develop biofilms. These waste materials compositions according to preferred embodiment were approximated to have 62% (mol) cellulose, up to approximately 35% pectin, up to 15% hemicellulose and up to 30% of other components such as polyesters or fat materials. The developed biofilm solution was casted in suitably shaped moulds of glass/ceramic material/non-adhesive plastic material and were dried to obtain the bioplastic, which then could be subjected to waterproofing treatments, colouring, etc. or used to develop composites on substrates like paper or fabrics. In order to increase the hydrophobicity, the soluble constituents (like sugars) on the film were dissolved by immersing the film in water for some amount of time depending on its degree of hydrophobicity. The objective of the invention was to obtain environmentally sustainable bioplastic films without any toxic residue in the final edible product and simultaneously keeping the antioxidant properties of the starting material intact. Moreover, the process of the invention is easily scalable, without the need of large consumption of energy or expensive reagent [45].

A study found that extract from pomegranate grind embedded in chitosan nanoparticles showed strong antimicrobial effect on *L. monocytogenes* species in combination with biodegradable zein biofilm when used for meat preservation. The lysing and precipitation of cell membranes of bacteria through the mechanism of the rind phenolics are responsible for the antimicrobial activity of hydrolyzable polyphenols within the extracts. While incorporating the rind powder in protein films made of mung bean and biopolymers based on starch, it enhanced their antimicrobial activity against salmonella [44]. By-product from mango (peel powder) has also been used to develop edible biofilms with or without antioxidant extract from fruit kernel [46].

Chitosan and pomegranate rind extract combination in edible coatings has shown to lower total aerobic plate count and prevented the increase of total volatile basic nitrogen in ice stored pacific white shrimp and also slowed the increase of peroxide value and TBARS (lipid oxidation detection assay) responsible for spoilage, in Nile tilapia fillets [44].

Incorporation of nanoparticle fillers like ZnO nanoparticles confer antimicrobial activity to bioplastics. The addition of ZnO allowed the bioplastic to be used in horticulture applications as the bioplastic was shown to hold ZnO nanoparticles in a great capacity and release them in a sustained manner [47].

B. Biodegradability

Studies show that addition of fillers to biofilms led to improved biodegradation rate which may be due to the huge surface contact area that the filler provides allowing degrading agents to enter the biopolymer. However, incorporation of natural fillers like rice husk flour have been seen to enhance the biodegradation of bioplastics. The biodegradation of the polymer depends on the chemical and physical characteristics of fillers [48]. Bio composites formed from a combination of wine lees and PHB had an enhanced biodegradation rate of PHB both in soil and marine water. Though grape seeds-derived antioxidants delay the PHB biodegradation, controlling the biodegradation rate is possible with a balanced composition of wine lees and grape seeds [49].

C. Packaging colour

Anthocyanin flavonoids in pomegranates have the potential to increase the redness in edible bioplastics hence decreasing its transparency which in turn results in reduced oxidation resulting from light exposure. While also improved sensory and texture qualities was seen when pomegranate rind extract was added into chitosan layer for shrimp that subdued melanosis browning. In one study, the browning and ripening process of guava fruit was delayed by decreasing the rate of respiration when 1 w/v rind extract was added to chitosan. Pomegranate flesh extract having comparatively greater anthocyanin content than non-edible pomegranate rind extract was shown to have a higher sensitivity to colour changes due to pH than the latter. Conversely the visual aesthetics of food when added to edible coatings can be affected by anthocyanins [44]. Edible biofilms conferred a good barrier to radiation with 600 nm light spectrum, due to the carotenoids and anthocyanins present in the mango peel powder incorporated in the films [46].

D. Plasticizing, Strengthening, and EAB and other effects

Blending of starch with plasticizers, fillers, fibres or other polymers, have been observed to enhance the physicochemical properties. Glycerol plasticized bioplastic is reported to show highest degree mobility of the biopolymer chain among other plasticizers. Bioplastics made of Starch/PLA blends are used to develop electronics devices, pharmaceuticals and more, and when coating these blends with beeswax improves its material strength properties and lowers water vapor permeability making it applicable in biodegradable food tray applications. Starch/PVC blends have applications in the areas of biomedical and clinical field, as Water-soluble laundry bags and also can be a potential replacement of polystyrene [18].

Keratin extracted from waste of chicken feathers was incorporated along with ginger starch to obtain a bio-composite film, where smoothness and tensile strength increased significantly but transparency and solubility decreased with increase in keratin level [50]. Wine by-products used as filler contents have shown to increase elastic modulus which could be due to the fact that wine fillers have higher intrinsic stiffness than those of the biopolymer matrices. A study showed that wine lees' (elastic modulus calculated as around 4.2–7.3 GPa.) was able to increase the elastic modulus of various biopolymers like PBS, poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) (PHBH), and poly (3-hydroxybutyrate-co-3- hydroxyvalerate) (PHBV). However, addition of wine filler content was shown to decrease tensile strength, but physical treatment (like autoclave treatments etc.) and/or chemical treatments (like enzymatic treatments) or addition of coupling agents could possibly help overcoming this constraint [49].

Pomegranate seed oil when incorporated into an edible biofilm constituting k-carrageenan and whey protein showed a plasticizing effect and lowered the tensile strength of the film. In an edible film developed from *Chlorella* sp. Pomegranate seed oil was incorporated to increase the shelf life of fruits and to stimulate the emulsifying and plasticizing effects. Whereas other studies reported the use of pomegranate rind extract and essential oil to decrease the flexibility, tensile strength (TS), and EAB of films [44]. SPI (Soy protein isolate, a by-product from soybean oil production), glycerol and zinc oxide (ZnO) nanoparticles in varying concentrations were used to develop a bioplastic. It was observed that when temperature increased, the interaction of nanoparticles with biopolymers led to improvement in mechanical properties of the film whereas this wasn't consistent after a limit [47].

In a study, the extract from mango kernel incorporated into biopolymer was reported to increase water vapour permeability [46]. Rice straw (agricultural waste) was used to develop a bioplastic that could be utilised to procure paper sheets and edible thin-biofilms or to drive shape memory effect [51]. Addition of fillers like sawdust and potato peel powder showed to decrease moisture content in banana peel bioplastics based on starch and solubility in alcohol and water was observed to be reduced. It has also been observed that absorption of water increases with increasing amounts of cellular based fillers due to intramolecular hydrogen bonding [48]. By-products from the use of crude coffee have potential to be used as additives (Coffee husks, coffee silverskin or spent coffee grounds) for plastics. Crude spent coffee ground (SCG) powder incorporated to corn starch-based biopolymer resulted in bioplastics with increased tensile strength. Adding crude SCG to cellulose-based formulations resulted in high light resistance. elasticity, stretchability, and water resistance were observed to be increased with addition of coffee silverskin into potato starch-based biopolymers. Incorporation of coffee husk into formulations based on corn-starch and chitosan led to films with higher water resistance, tensile strength properties [36].

V. Limitations and Future Prospects

Proper treatment of edible biofilms during storage and transportation is a pre-requisite as they are sensitive to different environments. The water content of the edible plastics also affects its efficiency. The increase in the water concentration reduces the oxygen barrier effect but increases the action of antioxidants in the plastic [52]. In addition, biofilms also require an outer covering to keep it safe for consumption and protect it from contaminants. As the films are prepared from natural polymers, they have poor mechanical strength. Edible biofilms are water soluble which is both boon and bane. If the film is too soluble, it cannot be detained in a humid climate and can even break down if it is kept cold. For further studies, nanoparticles can be added to the films in order to increase their mechanical properties and nutritional features of food. Biofilms can be produced in a continuous process at industrial level with efficient energy and in a more economical and sustainable manner.

VI. Conclusion

Plastic is an essential commodity for human society as it is applied in every sector including packaging, textiles, transportation, buildings and construction, industrial machinery, electrical and electronics. But the waste generated by plastics is causing a threat to the ecosystem. Statistics showed that 10.5 million ton of plastics produced in India generated 6 million ton of plastic waste that causes a major problem for solid waste management. Hence, there is a need to find alternatives for conventional plastics sources with different raw materials and improved properties. Biodegradable and edible polymers are found to be the suitable alternatives as they can be degraded and consumed, thereby reducing the waste generation and helps to have a shift from carbon footprint. Preparation of starch-based biofilms is seen as a best alternative as it is in toxic, renewable and exploration of unconventional plants as sources of starch helps to replace many conventional starch sources. Once the biofilm is formed, it has to be characterized to meet the standards and applications. Major tests that could be carried out include thickness measurement, moisture content evaluation, degradability test, solubility test and test to check its mechanical, thermal and chemical properties. Characterized biofilms from various sources can be applied specially for packaging with improved properties like antimicrobial effects, biodegradability, tensile strength, packaging colour, water resistance, elongation at break and so on. To conclude, biopolymers consisting of both edible and biodegradable films draw innovations towards a sustainable society and environment that helps every nation's dream of a clean country.

REFERENCES

[1] Hassan, M., Bai, Jun, Dou, D.Q., (2019). Biopolymers; Definition, Classification and Applications, Egypt. J. Chem. (2019) https://doi.org/10.21608/ejchem.2019.6967.1580.
[2] Thompson, R.C., Swan, S. H., Moore, C.J., & vom Saal, F.S., Our plastic age, Philos. Trans. R. Soc. Lond., B, Biol. Sci. 364(1526) (2009) 1973–1976. https://doi.org/10.1098/rstb.2009.0054.
[3] Filiciotto, L., Rothenberg, G., Biodegradable Plastics: Standards, Policies, and Impacts, Chem. Sus. Chem. 14(1) (2021) 56–72. https://doi.org/10.1002/cssc.202002044.
[4] Din, M.I., Ghaffar, T., Najeeb, J., Hussain, Z., Khalid, R., Zahid, H., Potential perspectives of biodegradable plastics for food packaging application-review of properties and recent developments. Food. Addit. Contam. Part A. Chem. Anal. Control. Expo. Risk. Assess. 37(4) (2020) 665–680. https://doi.org/10.1080/19440049.2020.1718219.
[5] Dhumal, C.V., Sarkar, P., Composite edible films and coatings from food-grade biopolymers, J. Food. Sci. Technol. 55(11) (2018) 4369–4383. https://doi.org/10.1007/s13197-018-3402-9.
[6] Pascall, M.A., Lin, S,J., The Application of Edible Polymeric Films and Coatings in the Food Industry, J. Food. Process. Technol. 4(2) (2013). https://doi.org/10.4172/2157-7110.1000e116.
[7] Padervand, M., Lichtfouse, E., Robert, D., Wang, C., Removal of microplastics from the environment. A review. Environ. Eng. Sci. 18(3) (2020) 807–828. https://doi.org/10.1007/s10311-020-00983-1.
[8] Gorrasi, G., Sorrentino, A., Lichtfouse, E., Back to plastic pollution in COVID times, Environ. Chem. Lett. 19(1) (2021). https://doi.org/10.1007/s10311-020-01129-z.
[9] Abdel-Shafy, H.I., Mansour, M.S., Solid waste issue: Sources, composition, disposal, recycling, and valorization, Egypt. J. Pet. 27(4) (2018) 1275-1290. https://doi.org/10.1016/j.ejpe.2018.07.003.
[10] Yousfi, M., Soulestin, J., Vergnes, B., Lacrampe, M. F., Krawczak, P., Morphology and mechanical properties of PET/PE blends compatibilized by nanoclays: Effect of thermal stability of nanofiller organic modifier, J. Appl. Polym. Sci. 128(5) (2013) 2766-2778. https://doi.org/10.1002/app.38450.
[11] Sousa, F.M., Costa, A.R.M., Reul, L.T.A., Cavalcanti, F.B., Carvalho, L.H., Almeida, T.G., Canedo, E.L., Rheological and thermal characterization of PCL/PBAT blends, Poly Bull. 76(3) (2019). https://doi.org/10.1007/s00289-018-2428-5.
[12] Siracusa, V., Blanco, I. Bio-Polyethylene (Bio-PE), Bio-Polypropylene (Bio-PP) and Bio-Poly(ethylene terephthalate) (Bio-PET): Recent Developments in Bio-Based Polymers Analogous to Petroleum-Derived Ones for Packaging and Engineering Applications, Polymers. 12(8) (2020) 1641. https://doi.org/10.3390/polym12081641.
[13] Ghaffarzadegan, R., Khoee, S., Rezazadeh, S., Fabrication, characterization and optimization of berberine-loaded PLA nanoparticles using coaxial electrospray for sustained drug release, DARU. J. Pharm. Sci. 28(1) (2020). https://doi.org/10.1007/s40199-020-00335-y.
[14] Ali, A., Ahmed, S., Recent Advances in Edible Polymer Based Hydrogels as a Sustainable Alternative to Conventional Polymers, J. Agric. Food Chem. 66(27) (2018) 6940-6967. https://doi.org/10.1021/acs.jafc.8b01052.
[15] Shit, S.C., Shah, P.M., Edible Polymers: Challenges and Opportunities, J. Polym. (2014). https://doi.org/10.1155/2014/427259.
[16] Sadeghizadeh-Yazdi, J., Habibi, M., Kamali, A.A., Banaei, M., Application of edible and biodegradable starch-based films in food packaging: A systematic review and meta-analysis. Current Research in Nutrition and Food Science. 7(3) (2019) 624–637. https://doi.org/10.12944/CRNFSJ.7.3.03.
[17] Datta, D., Halder, G., Effect of media on degradability, physico-mechanical and optical properties of synthesized polyolefinic and PLA film in comparison with casted potato/corn starch biofilm, Process SAF Environ. 124 (2019) 39–62. https://doi.org/10.1016/j.psep.2019.02.002.
[18] Diyana, Z.N., Jumaidin, R., Selamat, M.Z., Ghazali, I., Julmohammad, N., Huda, N., Ilyas, R.A., Physical properties of thermoplastic starch derived from natural resources and its blends: A review, Polymers. 13(9) (2021a) 5–20. https://doi.org/10.3390/polym13091396.
[19] Nagar, M., Sharanagat, V.S., Kumar, Y., Singh, L., Development and characterization of elephant foot yam starch–hydrocolloids based edible packaging film: physical, optical, thermal and barrier properties, J. Food. Sci. Technol. 57(4) (2020) 1331–1341. https://doi.org/10.1007/s13197-019-04167-w.
[20] Moongngarm, A., Chemical compositions and resistant starch content in starchy foods, Am J Agr Bio Sci. 8(2) (2013) 107–113. https://doi.org/10.3844/ajabssp.2013.107.113.
[21] Souza, A.C., Benze, R., Ferrão, E.S., Ditchfield, C., Coelho, A.C.V., Tadini, C.C., Cassava starch biodegradable films: Influence of glycerol and clay nanoparticles content on tensile and barrier properties and glass transition temperature. LWT-Food. Sci. Technol. 46(1) (2012) 110–117. https://doi.org/10.1016/j.lwt.2011.10.018.
[22] Andrade, L.A., Barbosa, N.A., Pereira, J., Extraction and properties of starches from the non-traditional vegetables Yam and Taro, Polimeros. 27(2), (2017) 151–157. https://doi.org/10.1590/0104-1428.04216.
[23] Zhu, X., He, Q., Hu, Y., Huang, R., Shao, N., Gao, Y., A comparative study of structure, thermal degradation, and combustion behavior of starch from different plant sources, J. Therm. Anal. Calorim. 132(2) (2018) 927–935. https://doi.org/10.1007/s10973-018-7030-4.

[24] Kumar, S., Pattanayak, S.K., Pandey, K.M., Bio-plastic from Yam-An Ecofriendly Prospective, (2021) 397-405. https://doi.org/10.1007/978-981-15-7711-6_41.
[25] Tanetrungroj, Y., Prachayawarakorn, J., Effect of dual modification on properties of biodegradable crosslinked-oxidized starch and oxidized-crosslinked starch films, Int. J. Biol. Macromol., 120 (2018) 1240–1246. https://doi.org/10.1016/j.ijbiomac.2018.08.137.
[26] Princey, J. M., Nandhini, A., Abinaya, E., Preparation and Characterization Studies of Amorphophallus paeonifolius and Manihot esculenta as a Bioplastic using Glycerol and Agar-Agar as Plasticizer, Orient. J. Chem. 36(6) (2020) 1205–1211. https://doi.org/10.13005/ojc/360627.
[27] Georges Amani, N., Buléon, A., Kamenan, A., Colonna, P., Variability in starch physicochemical and functional properties of yam (*Dioscorea* sp) cultivated in Ivory Coast, J. Sci. Food Agric. 84(15) (2005) 2085-2096. https://doi.org/10.1002/jsfa.2160.
[28] Raquez, J.M., Nabar, Y., Srinivasan, M., Shin, B.Y., Narayan, R., Dubois, P., Maleated thermoplastic starch by reactive extrusion, Carb Polymers. 74(2) (2008) 159-169. https://doi.org/10.1016/j.carbpol.2008.01.027.
[29] do Val Siqueira, L., Arias, C. I. L.F., Maniglia, B.C., Tadini, C.C., Starch-based biodegradable plastics: methods of production, challenges and future perspectives, Curr. Opin. Food Sci. 38 (2021) 122-130. https://doi.org/10.1016/j.cofs.2020.10.020.
[30] Marichelvam, M. K., Jawaid, M., Asim, M., Corn and rice starch-based bio-plastics as alternative packaging materials, Fibers. 7(4) (2019) 32. https://doi.org/10.3390/fib7040032.
[31] Syafri, E., Kasim, A., Abral, H., Asben, A., Effect of precipitated calcium carbonate on physical, mechanical and thermal properties of cassava starch bioplastic composites, Int. J. Adv. Sci. Eng. Inf. Technol. 7(5) (2017) 1950–1956. https://doi.org/10.18517/ijaseit.7.5.1292.
[32] Ncube, L.K., Ude, A.U., Ogunmuyiwa, E.N., Zulkifli, R., Beas, I.N., Environmental impact of food packaging materials: A review of contemporary development from conventional plastics to polylactic acid-based materials, Materials. 13(21) (2020) 1–24. https://doi.org/10.3390/ma13214994.
[33] Narancic, T., Cerrone, F., Beagan, N., O'Connor, K.E., Recent advances in bioplastics: Application and biodegradation, Polymers. 12(4) (2020) 920. https://doi.org/10.3390/POLYM12040920.
[34] Boey, J.Y., Mohamad, L., Khok, Y. Sen, Tay, G.S., Baidurah, S., A Review of the Applications and Biodegradation of Polyhydroxyalkanoates and Poly(lactic acid) and Its Composites, Polymers. 13(10), (2021) 1544. https://doi.org/10.3390/polym13101544.
[35] Álvarez-Castillo, E., Felix, M., Bengoechea, C., Guerrero, A., Proteins from Agri-Food Industrial Biowastes or Co-Products and Their Applications as Green Materials, Foods. 10(5) (2021), 981. https://doi.org/10.3390/foods10050981.
[36] Oliveira, G., Passos, C.P., Ferreira, P., Coimbra, M.A., Gonçalves, I., Coffee by-products and their suitability for developing active food packaging materials, Foods. 10(3) (2021) 683. https://doi.org/10.3390/foods10030683.
[37] Elizabeth, W.M., Biodegradable Edible Bio-plastic Container. WRIGHT MONICA ELIZABETH OP - US 201862699879 P 20180718. (2020). https://patentscope.wipo.int/search/en/detail.jsf?docId=WO2020018480&_cid=P12-KO8B0G-44749-1.
[38] Kouhi, M., Prabhakaran, M.P., Ramakrishna, S., Edible polymers: An insight into its application in food, biomedicine and cosmetics, Trends Food Sci Technol. 103 (2020) 248–263. https://doi.org/10.1016/j.tifs.2020.05.025.
[39] Banerjee, R., Kapoor, M.S., Biodegradable and Biocompositible Packaging Material and a method of preparation thereof. https://patentscope.wipo.int/search/en/detail.jsf?docId=IN300645391&tab=NATIONALBIBLIO&_cid=P10-KP7RJH-79265-1.
[40] Sikora, J., Majewski, Ł., Puszka, A., (2020). Modern biodegradable plastics-processing and properties: Part I, Materials. 13(8) (2020) 1–28. https://doi.org/10.3390/MA13081986.
[41] Fredi, G., Dorigato, A., Recycling of bioplastics waste: a review, Adv. Indus. Eng. Poly. Res. 28 (2021) 2551–2571. https://doi.org/10.1016/j.aiepr.2021.06.006.
[42] Reichert, C.L., Bugnicourt, E., Coltelli, M.B., Cinelli, P., Lazzeri, A., Canesi, I., Braca, F., Martínez, B.M., Alonso, R., Agostinis, L., Verstichel, S., Six, L., de Mets, S., Gómez, E.C., Ißbrücker, C., Geerinck, R., Nettleton, D.F., Campos, I., Sauter, E., … Schmid, M., Bio-based packaging: Materials, modifications, industrial applications and sustainability, Polymers, 12(7) 2020 1558. https://doi.org/10.3390/polym12071558
[43] Mironescu, M., Lazea-Stoyanova, A., Barbinta-Patrascu, M.E., Virchea, L.-I., Rexhepi, D., Mathe, E., Georgescu, C., Green Design of Novel Starch-Based Packaging Materials Sustaining Human and Environmental Health, Polymers. 13(8) (2021) 1190. https://doi.org/10.3390/polym13081190.
[44] Ko, K., Dadmohammadi, Y., Abbaspourrad, A., Nutritional and bioactive components of pomegranate waste used in food and cosmetic applications: A review, Foods. 10(3) (2021) 657. https://doi.org/10.3390/foods10030657.
[45] Giovanni, P., Ilker, B., Athanasia, A., A process for the preparation of biodegradable plastic films, European patent office. (2019) https://patentscope.wipo.int/search/en/detail.jsf?docId=EP277548644&tab=NATIONALBIBLIO.
[46] Dilucia, F., Lacivita, V., Conte, A., Del Nobile, M.A., Sustainable Use of Fruit and Vegetable By-Products to Enhance Food Packaging Performance, Foods. 7 (2020) 857.
[47] Jiménez-Rosado, M., Perez-Puyana, V., Sánchez-Cid, P., Guerrero, A., Romero, A., Incorporation of Zno nanoparticles into soy protein-based bioplastics to improve their functional properties, Polymers. 13(4) (2021) 1–14. https://doi.org/10.3390/polym13040486.
[48] Shafqat, A., Al-Zaqri, N., Tahir, A., Alsalme, A., Synthesis and characterization of starch based bioplatics using varying plant-based ingredients, plasticizers and

natural fillers, Saudi J. Biol. Sci. 28(3) (2021) 1739–1749. https://doi.org/10.1016/j.sjbs.2020.12.015.
[49] Nanni, A., Parisi, M., Colonna, M., Wine By-Products as Raw Materials for the Production of Biopolymers and of Natural Reinforcing Fillers: A Critical Review. Polymers. 13(3) 2021 381.
[50] Oluba, O.M., Obi, C.F., Akpor, O.B., Ojeaburu, S.I., Ogunrotimi, F.D., Adediran, A.A., Oki, M., Fabrication and characterization of keratin starch biocomposite film from chicken feather waste and ginger starch, Scientific Reports. 11(1) (2021) 1–11. https://doi.org/10.1038/s41598-021-88002-3.
[51] Skendi, A., Zinoviadou, K.G., Papageorgiou, M., Rocha, J.M., Advances on the valorisation and functionalization of by-products and wastes from cereal-based processing industry, Foods. 9(9) (2020) 1243. https://doi.org/10.3390/foods9091243.
[52] Bonilla, J., Atarés, L., Vargas, M., Chiralt, A., Edible films and coatings to prevent the detrimental effect of oxygen on food quality: Possibilities and limitations, J. Food. Eng. 110(2) (2012) 208-213. https://doi.org/10.1016/j.jfoodeng.2011.05.034

