Valorization of Bio-based Waste Resources for the Production of High-quality Bioplastics

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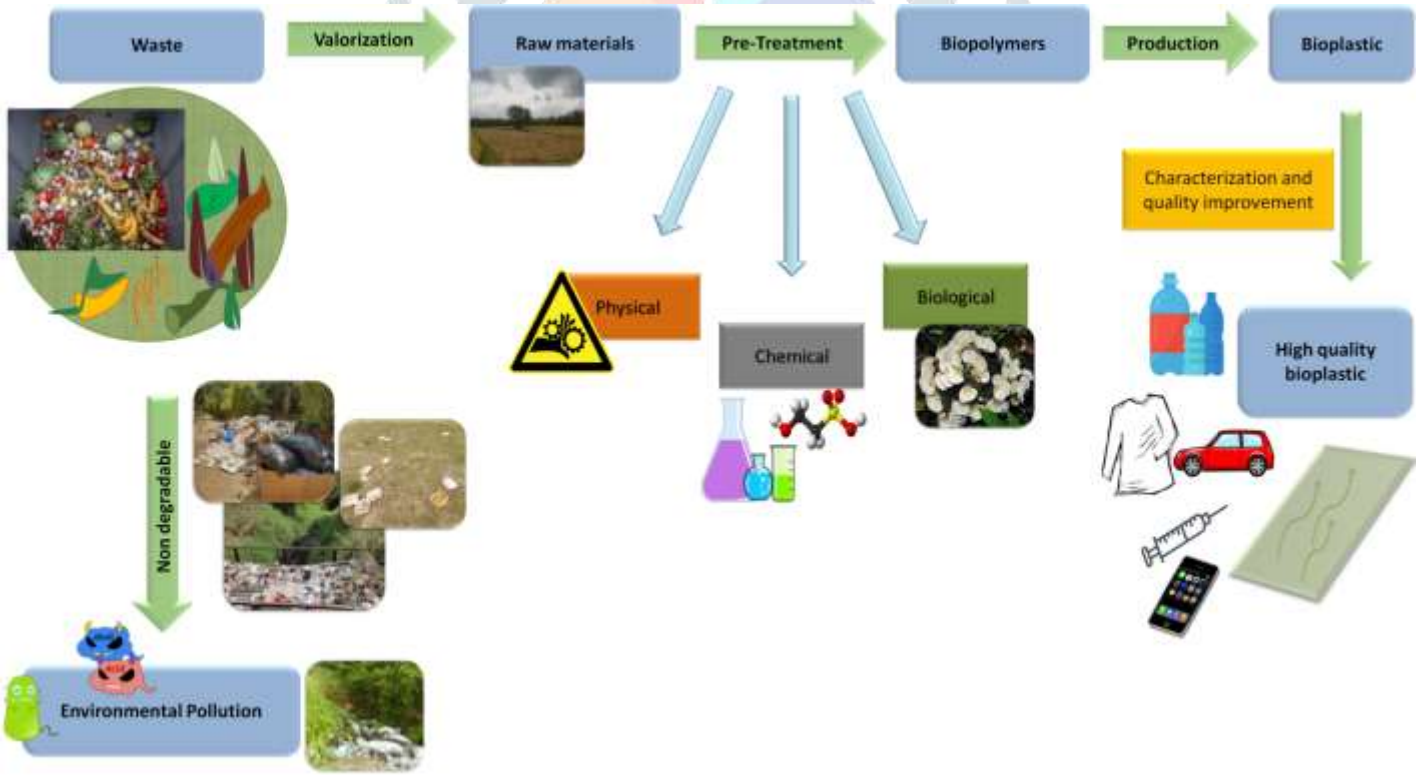
Graphical Abstract



Highlights

- This article reviews the problem of non-degradable plastics to the environment and their negative impacts on all living beings.
- Overview of various bio-based waste materials which have the potential to produce biodegradable plastic films.
- Bioplastic production was done in five distinct methods based on raw material origin.
- The analysis showed that the improvements of bioplastic properties have an impact on its quality and efficacy.

Abstract: The use of waste-based potential raw materials for degradable plastic has piqued interest due to the non-degradability of petroleum-based plastics. Various research studies have described and presented waste from many sources that can be used to synthesize bioplastic films, including starch-based, cellulose-based, protein-based, and oil-based waste. Bioplastic is produced either from biobased or biodegradable polymers or both. This review aims to present an insightful summary of the most recent advancements and development breakthroughs in bioplastic materials, concentrating on various applications. The quality and effectiveness of the bioplastics are analyzed concerning improvements in their mechanical, thermal, and processable properties. To bridge the gap between bioplastics and traditional plastics, improve bioplastic properties.

Keywords: Non-degradable plastics, waste-based raw materials, pretreatment, biopolymers, biodegradability.

1. Introduction

Waste generation is the most common emerging problem in the 21st century developing industries and growing population and urbanization (Patrício et al., 2021; Feo et al., 2019). Waste generation has been heavily dependent on economic growth and increasing day by day. The available waste management methods are not sufficient enough with the waste generation rate. Due to the high cost, unavailability of infrastructure and technology, waste materials are always disposed of improperly (Kumar & Samadder, 2017). The production of harmful toxic compounds, later on, leaching to groundwater bodies, minimize land available for human use and many more health-related issues can arise with this waste material problem. As the treatment processes for cleaning or removing non-degradable plastics from the environment is costly. The developing countries do not have proper waste management systems to eliminate waste or reduce waste.

Plastics, also denominated as polymers, are manufactured by transforming natural products or using primary chemicals typically coming from oil, natural gas, or coal. The monomers' greatest monomers, over 320 million applied to make plastics, are received from fossil hydrocarbons such as ethylene, the primary starting material for modern synthetic plastics and propylene. None of the often-used plastics is biodegradable (Geyer et al., 2017). Nevertheless, "plastics" contain a wide variety of semi-synthetic and synthetic polymers (Cucci et al., 2015).

- [CH2-CH2-CH2-CH2] n- Polyethylene

Semi-synthetic and synthetic varieties can cause significant stabilization problems due to production methods, additives, and treatments. Currently, reliance on petroleum-based polymers has expanded exponentially over the years, and it is about 322 million tons. Society utilizes synthetic polymers because numerous have highly acceptable qualities such as durability, versatility, resistance, chemical inertness, and the like. When bioplastics are not discharged accurately, they can contaminate batches of converted plastic and harm recycling infrastructure. Worldwide plastic production is expected to cross 300 million tons by 2015 (Bezirhan & Bilgen, 2019).

Plastic pollution in the environment is currently attracting worldwide attention. Improper unloading the use or disposal of plastic waste pollutes the environment. Plastic pollution in marine and freshwater ecosystems is relatively neglected compared to pollution in soil ecosystems. Plastic pollution in the soil environment and researched its effects on plastic waste, especially microplastics, on soil ecosystems. However, about 60% of the plastic has ended up in a landfill or the natural world, posing economic and environmental issues. The idea of waste energy recovery is one of the best practices adopted to manage plastic waste (Hidayah, 2018). When bioplastic is not discharged accurately, it can contaminate batches of converted plastic and harm recycling infrastructure. If bioplastic pollutes recycled PET (polyethene terephthalate, the most typical plastic utilized for water and soda bottles), the whole lot could be discarded and end up in a landfill. So separate recycling is essential to be capable of discarding biolistic suitably.

Industrial consumption (or primary waste) can usually be acquired from sizable plastic processing, manufacturing and packaging industries. Waste materials usually have suitable components for recycling. Although the volume of material is sometimes inadequate, the quantity grows as consumption and, therefore, increases. Commercial waste is often collected from workshops, artisans, shops, supermarkets, and wholesalers (Nkwachukwu et al., 2013).

Therefore, this review highlights the potential of waste resource processing techniques for producing bioplastics and their economic viabilities. Specifically, the recently published articles were critically reviewed to summarize the current knowledge about the novel and advanced techniques for waste valorization in producing high-quality bioplastics. Also, this review discusses the different methods to improve the bioplastic properties responsible for its quality and function corroborating its demand in a broad range of applications.

2. Waste as a potential resource

2.1 Problems associated with Food Waste, Agriculture waste, Municipal Solid Waste and Industrial waste

The industries overgrew, intending to fulfil the human needs of goods and services. Population in urban areas of India was 10.8% of the total population in1901 and reached up to 31.16% in 2011. From 1950 to 2015, the annual plastic production increased by around 160-fold to fulfil human needs (Z. Liu et al., 2021). With these

changes, the problem of waste comes to the platform as a huge problem (Figure 1). It has been reported that the heaps of garbage produced from cities, industrial zones and agricultural lands. Among them, the percentage of non-degradable materials is high (Figure2). Plastics using fossil fuel or petrochemical-based materials are common and have become a common problem in waste management (Figure 3). According to the estimations, plastic pollution due to medical waste can increase by 100% compared to the last decade (Wei et al., 2021). The Covid 19 pandemic is an accelerating factor for plastic pollution by medical waste. In China, municipal solid waste decreases by 30% in this public health emergency, while non-degradable medical waste increases by 370%. The total amount of medical waste in China was 207 kt from January 20 to March 31 of 2020. Medical waste output in Wuhan increased from 40 t/d to approximately 240 t/d, which exceeded the maximum incineration capacity of 49 t/d (Kleme et al., 2020). Due to the Covid-19 pandemic situation, single-use plastic packaging materials also goes high (Patrício et al., 2021). India uses 50% of the total single-use plastics produced in the world. Household plastic covers 10% of total waste production (Rajmohan et al., 2019). In the country Sweden, plastic household consumption increased by 23.8% between the years 1998-2007 (Eriksson, 2009).

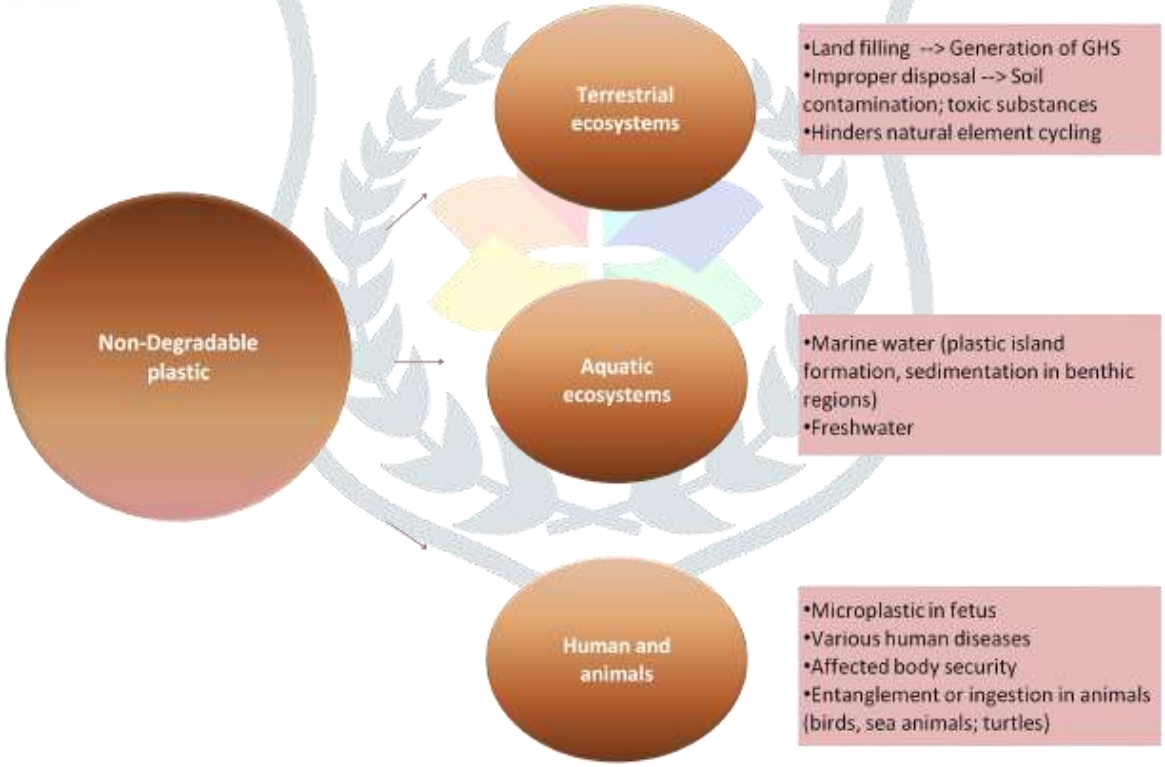


Figure 1: Problems associated with non- degradable plastics

Contaminating water resources with non-degradable plastics has become the top of the list of threats regarding the survival and stability of aquatic environments, mainly marine habitats (Figure 4) [Lotze et al., 2018]. The total global marine plastic percentage is 60-80% and may reach 90 to 95%, depending on the area, while more than 70% of floating substances belong to non-degradable plastics. Waste materials in coastal areas contain around 65% of synthetic polymer wastes, and 16.8% to 69.1% of them belong to single-use plastics (Gjyli et

al., 2020). A study conducted related to tropical Atlantic region microplastics were observed in 68% of the plankton samples and 60% of the sand samples (Ivar & Costa, 2014). Sediment samples collected from the shore and sublittoral sediments of harbours of the Belgian coast observed microplastics with a diameter of less than 1 mm, including fibres, granules, pellets, and films. 391 microplastic particles per kilogram of dry sediment is the highest microplastic concentration reported in sediment samples from harbours. They are likely because of local anthropogenic activity, sediment trappings and river runoffs (Claessens et al., 2011). In the forms of industrial waste, electronic and other plastic-based residues, domestic waste, food packaging materials (which account for 38% of European plastic production), textiles world plastic production is sent to the marine ecosystem (Lithner et al., 2011). For instance, the United Kingdom produces 680 tons of microbeads per year and 0.01–4.1% of total microplastics present in the ocean belong to cosmetic microbeads (Fries et al., 2013). Accumulation of plastics in the ocean leads to the formation of plastic- islands. The largest plastic islands are observed in the North Pacific Ocean, and according to estimations, it contains 1.8 billion plastic pieces. It weighs about 80,000 tons and covers an area of about 1.6 million km2 (Segovia-mendoza & Nava-castro, 2020).

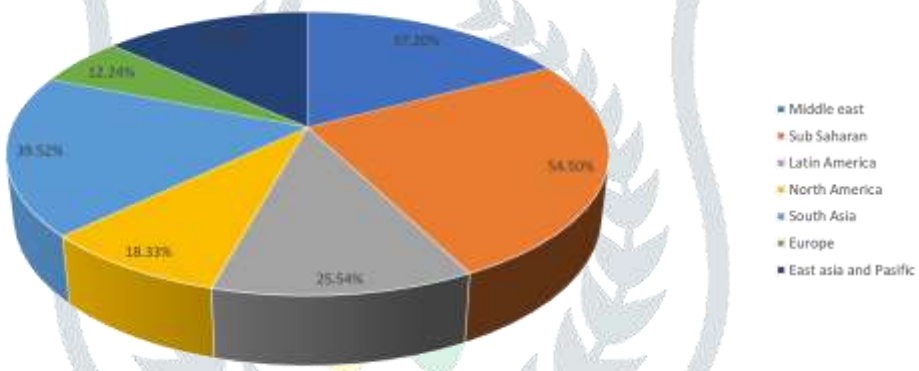


Figure 2: Increase of world waste generation in different regions 2016 to 2030

Moreover, sea birds, sea turtles and marine mammals face life-threatening effects due to the entanglement and ingestion of plastic debris. Plastic bags were swallowed by turtles slightly more often than other forms of debris, a total percentage of 44.7. Accordingly, 38.5% of Hard plastic, 7.73% of nylon, 5.1% of polystyrene, and 1.1% of rubber were contained in smaller quantities (Barletta & Costa, 2011). Plastic waste was found in the gastrointestinal tract of 40% of the seabirds along the southern Brazilian coast. *Twelve procellariiformes* (66%) were found to be contaminated, and two *Sphenisciformes* (22%) were found to be contaminated. However, *Charadriiformes* was not observed as contaminated *procellariiformes* ingesting large amounts of the products. Marine debris interactions are of particular concern for species that are recognized to be endangered, and 17% of all species, especially estuarine residents, are recognized to be threatened (Barletta & Costa, 2011). The small size of microplastics means that they have a high ratio between surface area and volume which improve the ability to allow transportation of pollutants (Teuten et al., 2009; Rowland et al.,

2007), The degree of microplastic particles promote the transfer of toxic substances to animals, the effects of this transfer, and the importance of this pathway in comparison to other pathways, is not yet completely understood (Walkinshaw et al., 2020). The effect on this accumulation cannot be predicted but can cause serious issues related to their lifecycle (Thompson, 2010).

Figure 3: Non-degradable plastic pollution in coastal sites

Freshwater plastic pollution (Figure 4) is a new research area, and most research works are done in European and American continents related to industrial zones and river pollution by industrial plastic effluents (Lebreton et al., 2017). Laurentian Great Lakes in the United States observed approximately 43,000 microplastics/km^2 of average abundance in 2012, and the downstream of two major cities observed more than 466,000 particles/km^2. Another study conducted in Yuyao city, China, observed a high bisphenol concentration in river waters (240–5680 ng L^{-1}) and aquatic organisms (116.13–477.42 ng g^{-1}) [Lin et al., 2017]. For the first time in human history, microplastics (MP) are reported in the human placenta, which can

be considered an over-the-level serious condition (Athapaththu et al., 2020). Antonio Ragusa et al. 2020 reported that four human placentas contained 12 MP fragments and were isolated for the first time using Raman Microspectroscopy (Ragusa et al., 2021). Microplastics were found in the fetal side, maternal side and in the chorioamnionitis membranes accordingly in 5MP, 4MP and 3MP. It indicates that once they entered the human body, these MPs can reach placenta tissues at all levels. The effect on the fetus is not identified yet, but it can harm the mother and fetus health (Fournier et al., 2020). Through the maternal surface, MPs can enter the placenta together with other exogenous materials. Further, they can invade the tissue in-depth via various transport mechanisms, which are still unknown (Tetro et al., 2018). The amount of transplacental passage is generally 5–10 m MPs, and it may be affected by various physiological and genetic factors.

Figure 4: Plastic pollution in freshwater ecosystem

The non-degradable plastic particles can negatively affect human health in many aspects, especially bisphenol and phthalates (Segovia-mendoza & Nava-castro, 2020). They can be inserted into the human body; for adults at the range of 1 to 2 μg/kg bw/d of a single compound and range of 0 to 4 μg/kg bw/d for children. Studies show that it increases allergic sensitization and IgE modulation mediated atopic disorders. The burning of plastic leads to significant health hazards such as heart disease, respiratory problems such as asthma and emphysema and causes rashes, nausea or headaches. When dioxins in plastics settle on crops and rivers, they inevitably reach our food and cause many health issues.

More miniature research works pay attention to plastic pollution of terrestrial ecosystems while emphasizing the adverse effects of plastic contamination on the soils. Most waste materials from municipal waste are sent to landfills, and these landfill areas are highly contaminated with non-degradable plastic particles (figure 5). Industrial soil samples from the Australian continent found 300–67,500 mg kg−1concentration of microplastics, and it can be varying with the site (Chae & An, 2018). Natural oestrogens were found in high concentrations in the leachate from tropical Asian countries (estrone: 0.127–1.00 mgl^{-1}; oestradiol: 0.002–0.0243 mgl^{-1}).In comparison, natural oestrogens were found in relatively low concentrations in leachate from a Japanese landfill site (Esteron: 0.05 $mgl^{-1:}$ Estradiol: 0.008 mgl^{-1}) (Rowland et al., 2007). Plastic particles can be ingested into the soil organisms and cause various harmful effects on their survival (Chem et al., 2018). Polyurethane foam microparticles is an accumulated pollutant in the invertebrate body and are identified in

earthworm *Eisenia fetida* (Gaylor et al., 2013). These plastic pollution act as a vector of hazardous chemicals across food chains (Chae & An, 2018).

Figure 5: Non-degradable plastic generation from various waste sources

At each trophic level, their concentration increased, and 129.8 ± 82.3 particles per gram of chicken faeces are the highest concentration reported (Lwanga et al., 2017). Landfilling is identified as the predominant source of GHS emission (Z. Zhao & Bian, 2020). Landfills of organic waste, especially food waste, is a global problem that is the highest fraction of municipal solid waste. 30% of the United Kingdom food waste production is sent to the recycling process of anaerobic digestion and composting (Moult et al., 2018). Landfills without greenhouse gas (GHS) capture facilities cause GHS to the environment, and the global percentage of such landfills is 20% (IPCC, 2006). Incineration of waste materials, especially Municipal Waste, dumping of organic waste, compost making can lead to GHS emission. Assumptions are made regarding the continuation of plastic pollution as if this continues by the year 2050, and conventional plastic will be responsible for 15% of total GHG emission in the world.

Plastic pollution causes significant damage to the dynamic equilibrium of the natural environment. Elemental cycles are hindered in various ways, especially the carbon cycle. Copepods play a significant role in trophic food chains and the carbon cycle as they take nutrition on phytoplankton and are prey for large species. The ingestion of plastics into the lantern fish has also been related to the toxicological and physical consequences. By the changes in the physical environment, plastic can also affect the biota. In beach sediments, substantial microplastic concentrations have been identified, and the potential to change physical characteristics like permeability and nutrient and water flow and sub-surface temperature that possibly cause sex determination in marine turtles is highly temperature-dependent (Villarrubia-gómez et al., 2017).

Moreover, it is suggested that plastic pollution is a risk to food security. Fisheries cover around 15% of animal protein consumption per person and show further growth (Béné et al., 2015). The derelict plastic debris used in the fishing industry has essentially become a fact for plastic contamination in fish. The trophic level and feeding strategy significantly affect the accumulation of toxic plastic debris in higher trophic levels (Walkinshaw et al., 2020). Predators at higher trophic levels get risks on their reproduction due to that toxic

plastic debris. As identified in the research, 60% to 80% of edible fish species are found with plastic contaminations. Other than common fish species, shellfish species were subjected to plastic contamination analysis and found that 0.2 to 5.36 microplastics g^{-1} in family Mytilidae. Crustaceans like Brown shrimp, *Crangon crangon* have high commercial value. Plastic contamination of these crustaceans' species was found as 0.68 ± 0.55 microplastics $gram^{-1}$ (Devriese et al., 2015).

2.2 Potential Waste Resource

The waste valorization process is used for analyzing the capability of industrial, agricultural and food production waste to produce bioplastic (Koutinas et al., 2014). Biorefinery, conversion of biomass feedstock to various biochemicals, biofuels, and bioenergy using novel technologies, is used to produce biopolymers (Cherubini, 2010). The European Circular Economic Strategy by European Commission (2008) for Plastics indicates that new materials and alternative feedstocks to develop plastics should be produced and used to show that they are more sustainable than petrochemical plastics. Various research, waste materials (By-products, Industrial effluents and Municipal waste) from food production, industrial waste and agricultural waste have been identified as potential raw materials for biopolymer production (Figure 6).

Figure 6: Types of potential waste sources

2.2.1 By-products (food and agriculture-related industries)

2.2.1.1 Starch-based By-products

Since 1950, several studies of starch-based bioplastics have been published. Varieties of starch from natural sources such as Wheat (studied maximum starch content of 74%), Sago, Corn, Potato, non-edible cashew nutshell extracts (Ashok & Rejeesh, 2017), Cassava and Rice derived from waste can be processed for further development and thermoplastic production (Mekonnen et al., 2013). Moreover, starch derived bioplastics represent 85% to 90% of the total bioplastics available in the market. By product 2,5-furan dicarboxylic acid (FDCA) of sugar beet processing (which consists of 30-50% fructose) can directly use as a monomer for bioplastic production further it can convert into poly ethylene 2,5-furan dicarboxylate, which equals to poly ethylene terephthalate (PET) produced in industries, in its structure. Biopolymer polyhydroxy butyrate (PHB) can be produced using outdated bakery waste consisting of 60% carbohydrates, as hydrolysate and seawater as solvent gain 80% of the recovery percentage (Koutinas et al., 2014). The powder form of organic fraction

of municipal waste (waste rice, vegetables, fruit skins and beans) constitutes 42.5% starch, and a maximum glucose yield of 498.5 g/ kg was obtained (Ebrahimian et al., 2020; Aithani & Mohanty, 2006). Cassava starch is studied for its potential as raw material. Oxidized starch observed 15.68% carbonyl content (Oluwasina et al., 2019) while it is higher than the oxidized starch content of Rice, Maize and Potato in percentages accordingly 0.48%, 0.73% and 0.95% (Xiao et al., 2011). Further, T. M. Ogunrinola et al. (2018) conducted a study on biofilm production using fresh cassava tuber from *Manihot emulate* TME1 variety reported amylase-amylopectin percentage ratio of around 40/60%. Due to low cost, ease of availability and natural origin potato peel (which contains 34.3 ± 2.7 of starch from the dry weight), waste is used for bioplastic production as a bioplastic with higher water absorption capacity than commercial bioplastics (Ogunrinola & Akpan, 2018; Bezirhan & Bilgen, 2019). Other than potato peels, potato starch and cornstarch is used in another study for biopolymer production and identified corn starch-based biopolymer is more advanced (R. Amin & Chowdhury, 2019). It is observed in Pacific regions, steam peeling losses are typically around 8% of the potato weight, resulting in approximately $1.01*10^9$ kg of potato peel waste by the year 2011 (Liang et al., 2014). Tapioca starch derived from agricultural waste Cassava tubers constitutes 15% of Amylose and 85% of Amylopectin (an essential component for bioplastic structure formation), which has excellent potential for bioplastic production. Malt-sprout is observed as a by-product of the brewing industry. It contains 89% organic material, 46% of carbohydrates, between 21% and 25% of proteins, depending on the variety, and is reported as having the potential to produce bioplastic films (Romano et al., 2017).

2.2.1.2 Cellulose-based by-products

The high availability and characteristics of cellulose fibre in biobased waste materials make them useful for producing bioplastic (Brodin et al., 2017). Sugar beet pulp, lignocellulose containing by-product of the sugar production industry, consists of polysaccharides, including 22–24 wt% Cellulose, 30 wt% hemicelluloses. It can produce lactic acid and another process to Polylactic Acid (PLA), which is a well-known biopolymer. By-products left after, rinsing of sugar beet roots after the extraction of sugar, and sugar beet leaves also can be subjected to biopolymer production. Further, corn molasses, corncob molasses, sugar molasses can also be used to produce, and the highest yield ($0.95g\ g^{-1}$) is observed when cane molasses is used for producing Lactic acid. Getachew and Woldesenbet in 2016 identified that corncob, banana peel and sugarcane bagasse and Teff (*Eragrostis teff*) straw have cellulose and sugar content that can be used for biopolymer production. It is observed that sugarcane bagasse gives high lactic acid production (56%) compared to the other three residue materials (Getachew & Woldesenbet, 2016). Wasted banana peel material is considered a cellulose-based by-product in the food production industry. *Musa acuminata* peels collected from local markets are taken to laboratory-scale biopolymer production (Saad, 2016). Rice straw is a common agricultural residue in most Asian countries, and it does not require further separation from waste (Bilo et al., 2018). It is identified as a high Cellulose contented waste material that has percentage contents between 32 to 47% of Cellulose, 19 to 27% of Hemicellulose and 5 to 24% of lignin (Saha, 2003) other than rice straw, lignocellulosic biomass from orange and apple fruit juice extractions can be used for biopolymer formation (Kuster et al., 2017).

Tea waste, which is a by-product of tea brewing, is used to synthesize hydrophobic bioplastic material. Spent tea leaves, citric acid and water are the only ingredients used in this process, and it is a zero-waste procedure (M. Liu et al., 2020). At the milling stage of Tequila production, a cellulose base by-product known as Tequila bagasse is produced. The bagasse is fibrovascular bundles dispersed in an Agave stalk. It consists of cellulose, hemicellulose and lignin that cover about 85% of the total weight of Agave biomass and is a potential raw material for bioplastic production (Esteban et al., 2008). The empty fruit bunches of oil palm (EFBOP) is a popular waste material in Indonesia. It comes under cellulose-based by-product as it contains 42.7% of Cellulose in dry weight and produced in the crude palm oil industry (Larvae et al., 2020). Umi Kalsom quantified the biobased composition of EFBOP in 2017 as 50.4% of Cellulose, 21.9% of Hemicellulose and 10% of lignin. Cellulose and Hemicellulose polymers present in EFBOP are used as the Carbon source of PHB production on an industrial scale (Putra, 1997). In PHB (poly hydroxybutyrate) production, the cost for Carbon source is around 45% of the total cost. In 2010 Indonesia produced 25.32 million tonnes of EFBOP as a by-product of crude palm oil. So that EFBOP is considered a cost-effective waste raw material (Y. Zhang et al., 2013).

2.2.1.3 Protein-based by-products

Natural pure protein compounds are pretty expensive, and it is not profitable to produce biopolymers from them (Girotto et al., 2015). Proteins identified from Soy, corn zein, Wheat, cottonseed, sunflower, and more plant proteins can be used to make bio-based plastics. Potato fruit juice is a by-product of the Potato starch industry and is considered a protein-rich material that can produce bioplastics without using plasticizers. The technology used in its production is available in bulk quantities ranging from 0.7–7 m^3 per tonne tuber. Proteins with animal origin such as blood meal, gelatin and collagen, feather quill and Keratin, proteins from egg and whey, bone meal, and more can be used as raw materials for such bio-based plastics (Mekonnen et al., 2013). High-value protein derived from wasted food, shrimp and crab shell proteins, farm animal waste is identified as successful by-products possible for bioplastic production. A study conducted in Brazil has found that the food industry by-product of that species contains around 96.58% of high protein content desired region of 1.13% microfibrillar fish proteins can be derived from gilded catfish (*Brachyplatystoma rousseauxii*), and the optimal percentage identified is 0.79% of proteins. Glutamine is preferable to have bonds with glycerol and a good alternative for biopolymer film production (Araújo et al., 2018). Filleting scrapings and skin of king weakfish (*Macrodon ancylodon*) is used for another research work conducted in Brazil, and it also indicates that they can produce both gelatin films by using a protein concentration of 3% w/w) and microfibrillar films (Ali et al., 2014). Also, it is further developed into blended films (Araújo et al., 2018).

Agriculture based waste materials are produced at 1.3 trillion tons per year (Morone et al., 2019). Soy protein from the soybean oil production industry constitutes 91 wt% proteins. The presence of a high amount of Amino acid, Glutamine, and Aspartic acid (90% of total proteins in soy protein isolate) increases its super-absorbent quality (Cuadri et al., 2017; Li et al., 2019). Pae protein isolates from the discards of pea quality

selections constitute 89% wt% proteins. These pea proteins have an antibacterial quality that adds value to bioplastic production for food packaging purposes (Perez-puyana et al., 2017). Dilactic acid, an essential molecule of polylactic acid, is found in the wheat bran, and it consists of 19.7% proteinaceous compounds (Xiong et al., 2019). Chicken feathers are a more familiar, inexpensive, biodegradable, protein-based waste material with a creatine protein content of 90% (Ramakrishnan et al., 2018). Feather quills of 3 to 4 million pounds are produced in America and Canada every year and do not have a specific use of this by-product. One study conducted has revealed that feathers contain 7% of cysteine molecules that strengthen keratin proteins. This feature makes chicken feathers appropriate for polymer production and further process to thermoplastic resins (Quill et al., 2011). Protein-based biodegradable polymers are known as second-generation polymers, and they are used to produce 1.4% of total biomass production. The wasted cotton seed powder contains around 70% of cottonseed protein. World cottonseed production in the year between 2019 to 2020 is 44.95 million tonnes. The cottonseed protein is a by-product derived after oil extraction from cottonseed, a high-quality plant protein (Yue et al., 2020). Globulin proteins extracted from common beans containing 50% of whole proteins present in beans are observed in the bean Phaseolus Vulgaris cotyledons. They are used to make biopolymers combined with Fennel (*Foeniculum vulgare*) plant waste in Medeterrin regions (Ierro et al., 2007).

2.2.3 Municipal Waste

Waste from municipal areas contains several highly different waste materials, biodegradable or non-degradable and many more complex materials in different physical states. Among them, the organic fraction of municipal waste and can be used as feedstock for biopolymer production. This organic fraction may contain a large number of valuable compounds. These compounds can be derived via various biochemical or physical procedures. Also, it can be directly derived without pretreatments. The solid materials present in the organic fraction of municipal waste can be used as feedstock for biopolymer production (Russo et al., 2019). Research conducted focuses on urban organic waste like household waste and garden waste (Kepa et al., 2019). Household waste, containing 53% of food waste, can be considered as the primary source of food waste other than food production industry waste in cities. Household waste is 53% of food waste in municipal solid waste. With the use of a set of biochemical conversion steps, solid-state municipal waste can be converted to volatile fatty acids and further processed to PHA, a monomer of biodegradable plastics (Basset et al., 2016). Waste Office Paper (WOP) takes a higher fraction of organic municipal waste. The paper waste from the paper pulp industry containing 35% of lignocellulosic waste in MSW. It is very rich in cellulose carbon source. WOP has a potential to be used as a possible substrate in the production of biopolymers (Annamalai et al., 2018). More than 400 million tons of waste papers are manufactured annually, and only around 50-65% are recycled due to the limitations of recycling those paper fibres that have resulted in low-quality paper products, as well as the difficulty of mixing those papers with other waste products (Wang et al., 2012). Animal-related inexpensive feedstocks such as animal fats can be present in municipal wastes collected from slaughterhouses and food production industries. Also, from the household in small amounts from various

animal fat materials and using microbial fermentation methods, 102 g/L of PHA titer. Poultry, cattle, sheep and wild animals identified as potential sources of animal fats. It is a preferred feedstock for PHA production; the fatty acid ester fraction is used for this procedure (Riedel et al., 2015).

High salt concentrated wastewater from municipalities can be used for biopolymer production with the help of microalgae species. Giorgio Mannina et al. (2019) proposed that there is a potential to recover PHA from municipal waste directly. In the sewage treatment stages, sequencing batch reactor and feed batch accumulating reactor maximum 67% of PHAs have been recovered Evaluated waste activated sludge produced from the treatment of wastewater from a combined food and dairy industry PHB processing plant. Grain-based Jowar distillery spent wash (deproteinized) observed the yield of 42.3% PHB, followed by PHB yield of 40% from spent wash distillery based on filtered rice grain. By adding diammonium hydrogen phosphate (DAHP), PHB production increased to 67% when the spent wash was used on raw rice grain. With DAHP supplementation and removal of suspended particles by filtration, the same wastewater resulted in reduced PHB output (57.9%) [G. Chen & Chen, 2009].

A biobased polymer film is produced with antioxidant quality using Olive (*Olea europaea*) mill wastewater. Sources of waste carbon, including crude glycerol, high-strength possible substrates for reducing the total cost of PHB output, are salt-containing wastewater and wastewater-activated sludge (WAS) from urban areas. To obtain more evidence as a proof-of-method that it is possible to manufacture nonsterile PHB from synthetic waste and natural waste, further Ashby et al. (2011) reported 38% of PHB accumulation in a study conducted using crude glycerol with fatty acids and methyl esters (Strahan, 2011).

Three forms of organic waste were used for experiments: synthetic crude glycerol, high-concentrated synthetic wastewater, and genuine wastewater-activated sludge from municipals (Asiri et al., 2020). Activated sludge from fruit waste materials in the water treatment plant collected from the Lisbon pilot plant in Portugal is used to identify activated sludge possibilities to form biopolymers. It was observed that fruit waste-related activated sludge has fewer inorganic compounds than typical municipal wastewater (Luisa et al., 2020).

2.2.2 Industrial effluents

Effluents from the dairy industry have taken a smaller number of researchers to identify its potential to make biopolymers. The dairy industry makes effluent water and milk residues as major by-products. The wastes effluents from the cheese-making industry and the casein powder production industry can contain cheese whey. Furthermore, it has world-scale production of 190 × 106 tonnes per year. It contains many proteins, including Lactose. Compounds such as polyhydroxyalkanoates (PHAs) and polylactic acid (PLAs) can be made into bioplastics by bioconversion of Lactose present in whey permeate, and Lactose and glucose are detected as whey (contains 6.8% dry matter) sugars. So that, by volume, cheese whey containing 80– 90% of transformed milk can be considered a promising substrate to bioplastic production (Obruca et al., 2011).

Moreover, another study conducted shows that combining whey proteins (composed of 50–55% Beta-lactoglobulin and 20–25% alpha-lactalbumin) with egg albumin or natural latex rubber can process more thermostable bioplastic films (Sharma & Luzinov, 2013). Another alternative reported for the production of

milk bioplastics is supplanted formaldehyde with acid, such as vinegar. Nevertheless, it limits the bioplastic production from sour milk to the demonstration level only due to ethical considerations (Je et al., 2020).

Waste from oil mills wasted cooking oil from the household, vegetable oils, animal-related oils, fats containing materials from the places like slaughterhouses are considered identified raw waste materials for biopolymer production (Talan et al., 2020). It is a low-cost waste material that can produce poly3-hydroxybutyrate-co-3-hydroxyvalerate with the help of microorganisms. This Olive mill wastewater is rich in many carbon sources such as lipids, volatile fatty acids and some carbohydrate types (Dionisi et al., 2005). One study conducted to analyze PHA production by using microorganism *H. mediterranei*, and zero cost did not require pretreatment of Olive mill wastewater and gave a high yield of PHA (Alsafadi & Al-mashaqbeh, 2017). Possible substrates in the production of PHAs are vegetable oils (Alias & Tan, 2005). Waste streams that are even cheaper than purified oils from oil mills or used oils can also be used (Fernández-hernández et al., 2014; Mumtaz et al., 2010). There is a potential to transform this wasted resource into a helpful biomaterial because deep-frying is common.

Waste rapeseed oil has been successfully used with valerate monomers to develop PHAs (Obruca & Marova, 2010). Further, it is advantageous to use wasted oil to produce biopolymer as it does not affect the molecular properties of the final biopolymer. Waste cooking oil (corn oil) can be used as the carbon source for PHB production using the microorganism Pseudomonas aeruginosa. 45% of higher production of 1.98 g/l is observed at an optimum corn oil concentration of 3%. (Gatea & Hayder, 2017) Epoxidized soybean oil from waste cooking oil is used as a plasticizer in Poly Lactic Acid biopolymer (Mekonnen et al., 2013). Medium-chain-length PHA can be produced from wasted rapeseed oil. Globally, Rapeseed oil is one of the most efficiently produced types of vegetable oil, and the wasted type is highly cost-effective for biopolymer production (Przybyłek & Ciesielski, 2011).

Leather making industry produces industrial effluent containing many solid and liquid chemical and biological compounds. These wastes have a high content of the structural fibrous protein collagen and keratin (Sundar et al., 2017). They are hydrolyzing waste through a promising alternative acid or enzymatic hydrolysis to obtain protein hydrolysates containing necessary peptides and amino acids that can be used as novel applications. By reacting enzyme waste of collagen proteins from the manufacture of leather and edible meat product casings with dialdehyde starch, hydrogels for use as biodegradable packaging materials are produced (Langmaier et al., 2008).

Wastewater from the paper and pulp industry (PPI) is one of the potential raw material for PHA production, according to the studies on the use of PPI wastewater for PHA production (Jiang et al., 2012; Mannina et al., 2020). One study conducted by Queiro's et al., 2014, using complex feedstock material from the paper pulp industry containing lignosulfonates, phenolic compounds, sugars, and acetic acid, hardwood-spent sulfite liquor (HSSL), was directly used for the production of PHA without the use of initial acidogenic fermentation (Chakraborty & Mohan, 2019). The application of low-cost crude glycerol phase (CGP) from biodiesel production observed the same polyester composition and volumetric productivity, suggesting that CGP as

feedstock was feasible. CGP contains up to 60% glycerol and many impurities such as water, methanol, traces of hydroxide, fatty acids and esters (Hájek & Skopal, 2010). No negative impact is observed in the efficiency or polyester properties when comparing CGP to the pure glycerol process (Hermann-krauss et al., 2013). Other than CGP, Jatropha biodiesel residues are used as raw material to produce biopolymer PHAs (Shrivastav et al., 2010). Crude levulinic acid derived from seaweed was proposed as co-feed, containing formic acid, sugars, and dissolved minerals (Bera et al., 2015).

3. Biopolymers

3.1 Biopolymer Types

It is observed that the raw material and the polymer biodegradability are the two distinct criteria that define bioplastic. Hence, three main types of biopolymers have been identified (Figure 7).

Figure 7: Types of Biopolymers

3.1.1 Type A

These biopolymers are produced either by biological systems, including animals and plants and synthesized from the biological feedstock. Biodegradable biopolymers include 1) Synthetic polymers from renewable sources 2) produced by microorganisms; 3) Naturally available polymers that are biosynthesized by different routes in the biosphere (starch or proteins). The most utilized bio-based biodegradable polymers are starch and PHAs.

3.1.2 Type B

Type B biopolymers are produced from biobased resources but are biodegradable. These biopolymers comprise 1) Synthetic polymers from renewable sources and 2) Naturally occurring polymers like natural rubber or amber.

3.1.3 Type C

Petrochemical-based materials make these biopolymers; synthetic aliphatic polyesters produced from crude oil are biodegradable and compostable. Partially fossil fuel-based polymers like polybutylene succinate (PBS) and specific "aliphatic-aromatic" copolyesters are discovered to be degraded by microorganisms (Niaounakis,

2015). Polyvinyl alcohol (PVA) and PBS are petrochemical-based biopolymers. Fossil-based non-degradable polymers include polystyrenes (Jõgi & Bhat, 2020).

Common bioplastics in the present market regarding production and ability to renew are starch plastics, cellulose esters, and protein-based plastics (Figure 8). Bio-originated plastics recognized to succeed in the sustainability and environmental difficulties caused by synthetic plastics. Recent advancements in using bacteria for the synthesis of bioplastics. The usage of natural compounds recovered from waste materials known to establish a significant improvement towards biopolymers in the past decade (Mekonnen et al., 2013).

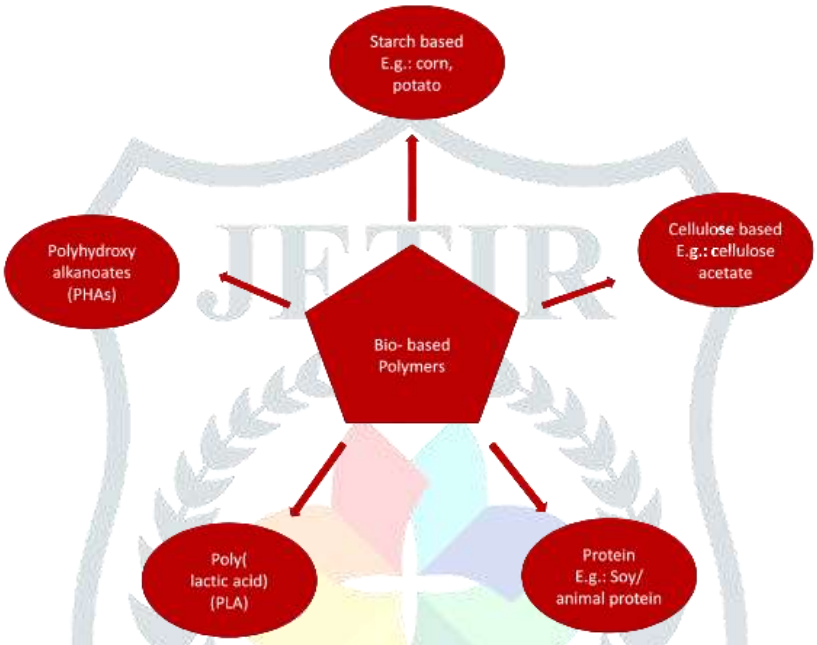


Figure 8: Types of bio-based polymers

3.2 Classification of Biopolymers

Biopolymers are classified into two extensive categories, particularly biodegradable and nonbiodegradable biopolymers (Figure 9). Apart from that, biopolymers are divided based on their origin as bio-based or fossil fuel-based (Riedel et al., 2015). Non-degradable bio biopolymers exist more than biodegradable biopolymers. Biopolymers are also classified as thermoplastics, thermosets (Ebrahimian et al., 2020), or elastomers based on their reaction to heat. Further, they classified it based on composition as blends and composites. The biopolymers can be categorized into chemical synthesis types such as PLA acquired by polymerization of lactic acid produced by fermentation of starch materials; microbial production type known to comprises PHAs; poly(3-hydroxybutyrate) (P3HB) [Ishii-Hyakutake et al., 2018] and natural types like cellulose acetate, esterified starch, starch-modified PVA (Niaounakis, 2015).

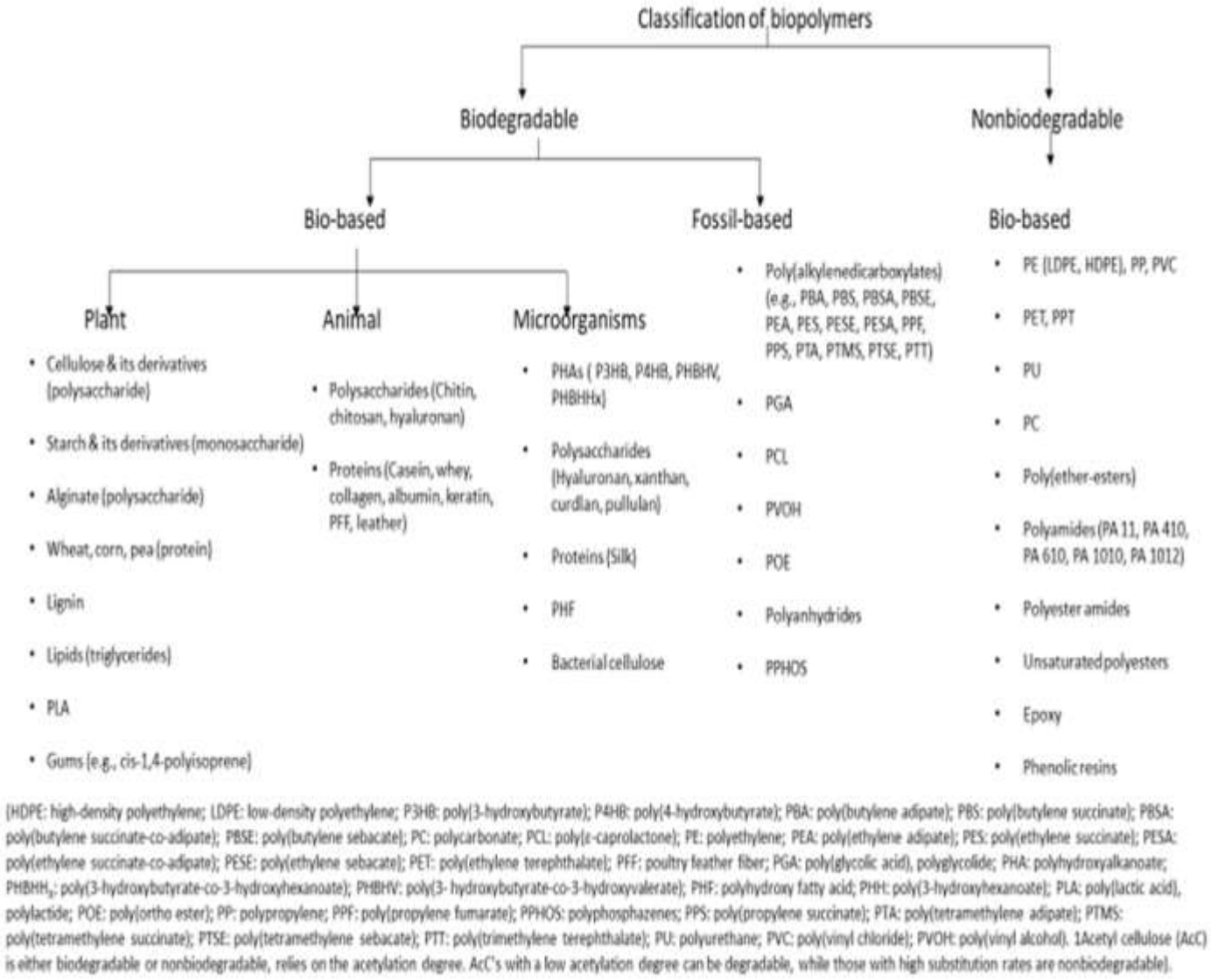


Figure 9: Classification of biopolymers

3.2.1 Biodegradable Natural polymers

Natural polymers are observed to be obtained by microbial fermentation or in vitro production by enzymatic actions. According to the wide variety of polymers' chemical structure as living organisms synthesized, they are categorized into three major classes: polysaccharides, proteins, and polyesters (Hassan et al., 2019).

3.2.2 Based material classification

1) Sugar-based Biopolymers: These biopolymers are fabricated by blowing, vacuum forming, injection moulding and extrusion. Polylactides are produced from Lactose obtained from crops like maize, Wheat, and potatoes (Hassan et al., 2019).

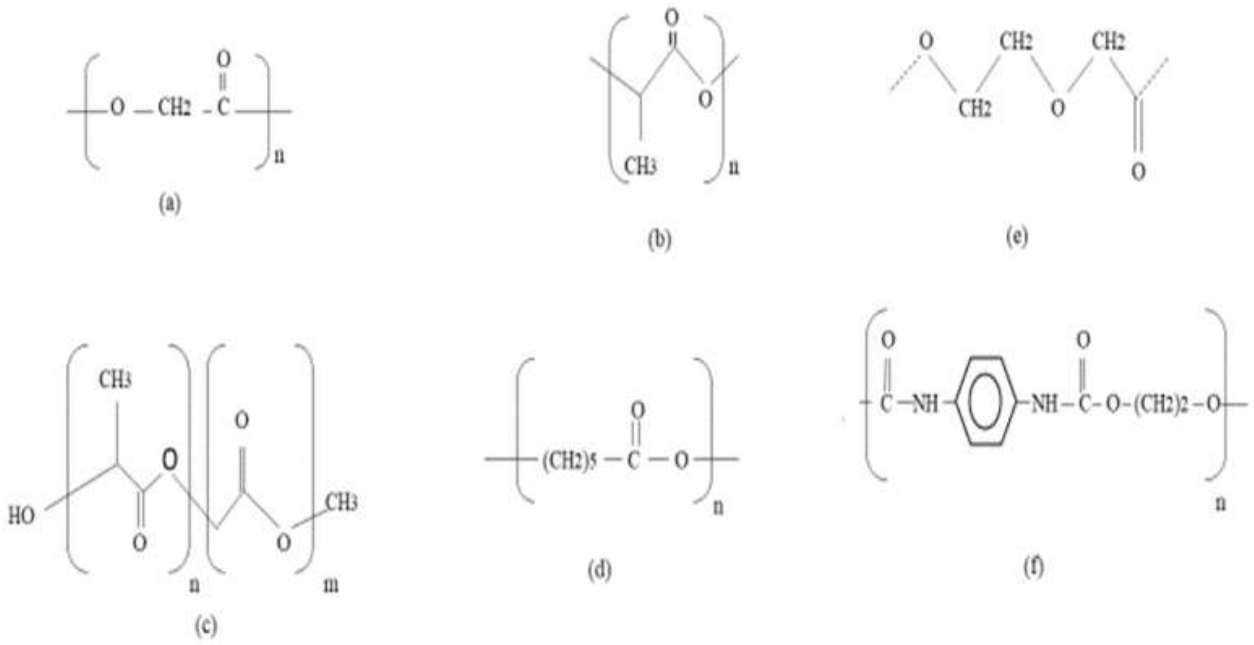
2) Starch-based Biopolymers: Starch-based biopolymers are acquired by melting starch in Wheat, barley, and maize (Basiak et al., 2018). Dextrans generated by starch hydrolysis, which is a combination of low molecular-weight carbohydrates, enzymatically synthesized by immobilized *Enterococcus faecalis,* Esawy dextransucrase onto biopolymer vehicles (Hassan et al., 2019). The mechanical characteristics of

thermoplastic starch (TPS) vary differently with temperature and humidity. Therefore, synthetic polymers and biodegradable polymers have mixed TPS (X. Zhao et al., 2020).

3) Cellulose-based Biopolymers: This polymer is constituted with glucose and is originated from natural resources, plant cell walls, microorganisms, marine animals (tunicate), and invertebrates (Nechyporchuk et al., 2016; Koutinas et al., 2014). These biopolymers are employed in packing cigarettes, CDs, and confectionery (Hassan et al., 2019).

4) Biopolymers based on Synthetic materials: These biopolymers are used to produce aliphatic-aromatic copolyesters obtained from petroleum. They are reported as completely compostable and biodegradable even though manufactured from synthetic components (Hassan et al., 2019).

3.2.3 Biodegradable synthetic polymer



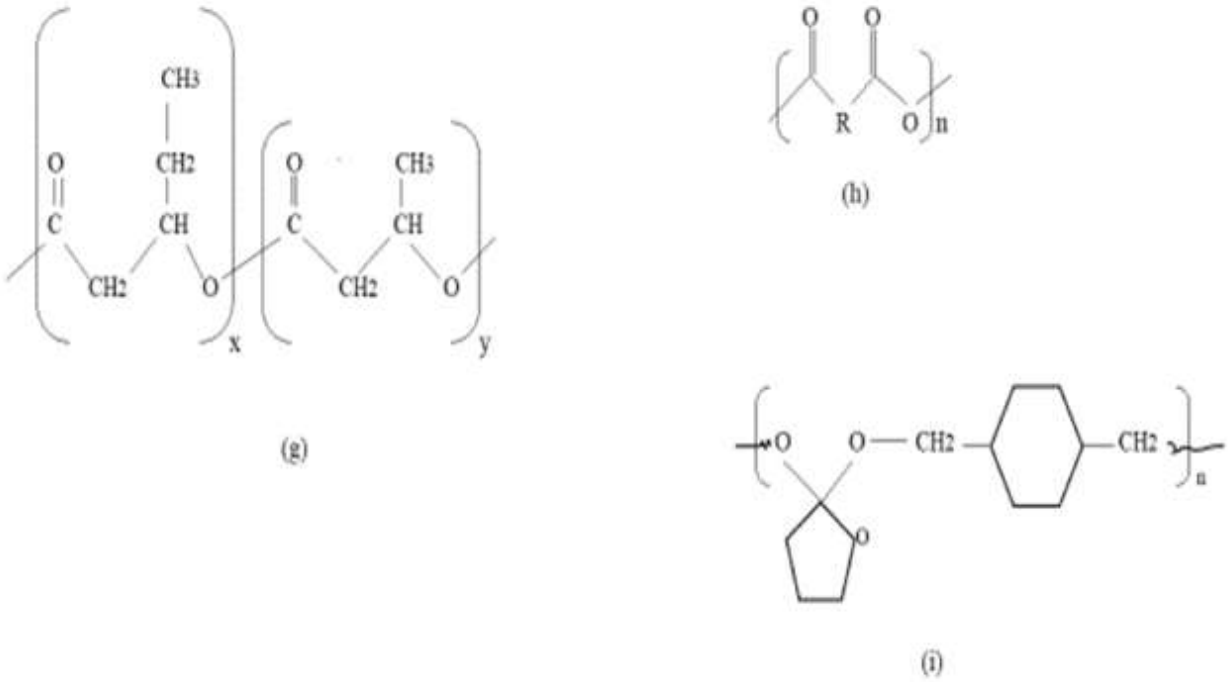


Figure 10: Biodegradable synthetic polymer

1) Polyglycolide Acid (PGA): PGA is a polymer that shows great Crystallinity of 45-55% and high tensile modulus, and significantly lower solubility in organic solvents. (Figure 10a) The glass transition (Tg) and melting temperatures (Tm) observed are 361°C and 2251°C, respectively. Gunatillake et al., 2009 reported that if stannous octoate is used as a catalyst, a temperature of about 1751°C is required for 2–6 h glycolide polymerization. Nowadays, PGA has primarily been employed in medical applications to produce resorbable sutures due to its great fibre-forming capacity (Yamane et al., 2014; Hassan et al., 2019).
2) Polylactide Acid (PLA): PLA is aliphatic polyester with 37% crystallinity (Figure 10b). It has a 60–65°C Tg and about 175°C Tm. (Gunatillake et al., 2006) described semi-crystalline PLA have around 50–70 MPa tensile strength, 3 GPa tensile modulus, and elongation at a break of 4%. Excellent mechanical properties have been achieved byenantiomeric PLA stereo complexation, and the enhancement is ascribed to the formation of stereo complex crystallites that function as intermolecular cross-links (Hassan et al., 2019)
3) Polymer of (lactide-co-glycolide) (PLGA): PLGA (Figure 10c) is considered as a copolymer of hydrophilic PGA and hydrophobic PLA. It is reported that the monomer ratio of 80 lactide/20 glycolide along with copolymer is semi-crystalline, and with the increase of the ratio of monomer lactide/glycolide, the degradation rate of copolymer's decreases (Niaounakis, 2015). PLGA copolymers belong to the few biodegradable polymers with approval of the Food and Drug Administration (FDA) for human medical applications (Gunatillake et al., 2006) such as drug delivery and tissue regeneration treatments since it degrades non-toxic compounds (Niaounakis, 2015).

4) Polycaprolactone (PCL): A rigid, flexible, semi-crystalline polymer (50% crystallinity) with high solubility (Figure 10d). It is observed to have relatively low Tm (60 °C) and Tg (−60 °C). Its ability to increase the polymer chains' mobility makes it a suitable plasticizer. Biocompatibility and its "in vivo" degradation enable its usage in the delivery of bioactive agents like drugs and enzymes (Niaounakis, 2015; Hassan et al., 2019). With a primary molecular weight of approximately 50,000, it is observed that the total degradation time is around 3 years for PCL (Gunatillake et al., 2006).
5) Polydioxanone (PDS): PDS is a transparent, non-toxic, semi-crystalline, and synthetic polymer of many ether-ester units (Figure 10e). The ring-opening step of polymerization of p-dioxanone manufactures PDS. The polymer is treated at the lowest possible temperature to inhibit depolymerization (Niaounakis, 2015). It is used to produce a monofilament suture with a Tg of -10 to 0°C and around 55% crystallinity (Hassan et al., 2019).
6) Polyurethane: It is generally developed by diisocyanates polycondensation with alcohols and amines (Figure 10f). Polyurethanes used for medical applications have unique mechanical properties, high biocompatibility, and structural versatility feasible to reach the demands of a broad range of medical implant applications such as cardiac pacemakers and vascular grafts (Gunatillake et al., 2006). For biocompatibility, polyurethane connects to the membrane and does not stimulate immune reactions quickly (Hassan et al., 2019).
7) Polyhydroxybutyrate (PHB), polyhydroxy valerate (PHV) and copolymers: Both PHB and PHB-PHV can be manufactured into various shapes and structures such as films, sheets, and fibres (Figure 10g). Both are also observed as soluble in a large number of solvents. Polymers like PHB, PHV, and copolymers with short biodegradation time are obtained from microbial fermentation. PHB has excellent potential in the gas barrier and degrades D-3 hydroxybutyric acid in vivo collectively with low toxicity. It regulated drug release, modified artificial skin, used sutures, and examined it as a substance for bone pins and plate development. Bioactive ceramics are also combined to make them become possible biopolymers (Hassan et al., 2019).
8) Polyanhydride: Polyanhydrides exhibited best-controlled release properties and biocompatibility in vivo (Figure 10h). Poly [carboxyphenoxy propane- sebacic acid] (PCPP-SA) the most widely studied polyanhydride. Polyanhydrides have only a few mechanical characteristics that limit their supporting applications, such as in orthopaedics. For example, poly[1,6-bis(carboxyphenoxy) hexane] reported having 1.3MPa Young's modulus, which is less than the modulus (40-60 MPa) of human cortical bone (Gunatillake et al., 2006). One potential carrier for gentamicin to treat osteomyelitis was observed by a polymer made by mixing sebacic acid and erucic acid dimer into a 1:1 ratio (Hassan et al., 2019).
9) Poly (orthoesters): A biodegradable polymer generated and examined for applications such as burns treatment, drug delivery in ocular, management of postoperative pain and orthopaedic treatments (Gunatillake et al., 2006). Poly (orthoesters) erosion in aqueous ecosystems is prolonged due to its hydrophobic properties. The degradation rate for these polymers, pH sensitivity, and flexibility in the

Tg are regulated using different levels of chain diols(Hassan et al., 2019; Heller et al., 2002)have reported developing poly(orthoesters) families with various mechanical characteristics and degradation rates.

3.3 Process of bioplastic production

3.3.1 Microorganisms

Biodegradation of biopolymers occurs through enzymes and chemical breakdown connected with various microorganisms (aerobic and anaerobic bacteria and fungi) and their secretion products (Xu & Yang, 2012). Biodegradation by microbial action observed in almost every waste source. However, biodegradation's degree relies upon the conditions such as humidity, temperature, and air content of the waste. Many enzymes can degrade polymers comprising hydrolyzable groups such as esters, amides. In order to degrade substrate polymers that are aliphatic polyesters, aromatic poly (ester amide)s or partly aromatic poly(ester urethane)s (Niaounakis, 2013), microbial enzymes are used. For instance, cutinase from *Humicola insolens*, lipases from *Aspergillus niger, Candida antarctica* (lipase component B), and *Mucor miehei* (Lipozyme 20,000 L)

3.3.2 Pretreatment

Waste is an excellent primary feedstock for bioplastic production, and it is necessary to pretreat feedstock to improve and modify the biological and physiochemical characteristics. The commonly used pretreatment technologies on bioplastic production include physical, chemical, and biological treatment (Tsang et al., 2019). Moreover, the pretreatment choice considers the feedstocks, enzymes, and organisms' compatibility (H. Chen et al., 2017). Successful conversion methods lead to the incomplete or total liberation of monomers in the waste resource with the polysaccharides, proteins, and lipids accessibility to subsequent enzymatic hydrolysis and fermentation (Kim & Kim, 2018). Different techniques are combined into a single process to achieve better bioplastic performance. For instance, bioplastic production with physical and acid treatment (heating at 121°C for 60 min supporting 2% sulfuric acid conversion) of industrial food waste was reported obtaining the maximum 3-hydroxyvaleric acid (3 HV) with 22.9% mole fraction (Elbeshbishy et al., 2011). The pretreatment's central scope involves substrate size reduction, extracting smaller chemical composites, and excluding inert substances irrelevant for subsequent steps. As waste composition varies significantly, influential parameters like temperature, pH, and hydraulic retention time need to be managed precisely for the maximum results (Strazzera et al., 2018).

3.3.2.1 Physical pretreatment methods

This pretreatment is performed using mechanical and thermal conversion methods (Tsang et al., 2019). It involves mechanical crushing/milling, Microwave and Ultrasonic processing, high strength electron radiation system, and heating for increasing surface area, biological conversion, separation rate, and fermentable substrates (H. Chen et al., 2017; Tsang et al., 2019). For example, during laboratory-scale PHA production,

PHB generated by utilizing Jambul seeds dried at 60°C to diminish the moisture content, and later seeds are crushed into fine particles. Lignocellulosic wastes are pretreated through a steam-explosion method at 160 °C-260°C with 0.7- 4.8 MPa (Tsang et al., 2019). More advanced processes include several steps, such as stripping, pulping, metal separation, scattering, screw pressing, disc-screening, and sieving (Bernstad et al., 2013).

3.3.2.2 Chemical pretreatment methods

Acid pretreatment involves inorganic acids (sulfuric, nitric, hydrochloric, phosphoric acids) and organic acids (formic, acetic, propionic acids) (H. Chen et al., 2017). This treatment enhances the availability of feedstocks to enzymatic hydrolysis while producing cell growth inhibitors in low quantities (Monavari et al., 2011). Alkali pretreatment principally depends on lignin's solubility within alkali solvent. NaOH, $Ca(OH)_2$, KOH, and NH4OH are involved in alkaline pretreatment (H. Chen et al., 2017). This pretreatment completely hydrolyzes ester linkages combining cellulose and lignin for lignin solubilization (Alvira et al., 2010). Chemical pretreatment also includes oxidation pretreatment, Ionic liquid (IL) pretreatment, and organosolv pretreatment. Oxidation indicates significant biomass degradation via the oxidant, and it comprises wet oxidation, ozonolysis, and photocatalysis. An IL (green solvents) is a novel type of cellulose solvent, and no explosive or toxic gases are produced. Organosolv pretreatment efficiently degrades lignin and mainly applied osmosis for breaking internal cellulose, hemicellulose linkages.

3.3.2.3 Physicochemical pretreatment

The steam explosion is a hydrothermal pretreatment of high-temperature steam that lasts for seconds to minutes before being withdrawn from the reactor. The primary mechanism of this process is increased pressure steamed through the fibres and discharged as air from the closed pore, causing mechanical cracks in the fibre. Pretreating feedstock in anhydrous ammonia or liquid form with temperature between 90–100°C and pressure of 1–5.2MPa using AFEX is a mixture of steam explosion and alkali treatment. The pressure and ammonia immediately ejected, causing fast temperature fluctuations, damage to the cellulose structure and enhanced surface area, and refined enzyme accessibility. AFEX is applicable for the pretreatment of agricultural waste with high cellulose content. CO_2 explosion pretreatment, SO_2 explosion pretreatment, and electrical catalysis pretreatment are the other physicochemical pretreatment techniques. (Elbeshbishy et al., 2011) studied combined techniques, including heat ultrasonication of heat, ultrasonication of acid (1 N HCl, 3.0 pH,24 h, 4°C), and ultrasonication of base (1 N NaOH, 11.0 pH, 24 h, 4°C). Alkaline ultrasonication was reported the highest increase of soluble chemical oxygen demand value for 30% and 40% of soluble protein.

3.3.2.4 Biological pretreatment method

Biological pretreatment mainly utilized some bacteria and fungi to degrade lignin. They produce enzymes that disintegrate lignin during the process. The white-rot fungi have the most beneficial lignin breakdown ability

(Baruah et al., 2018). Single-cell protein by-products have achieved their pretreatment of cellulose with reduced energy. White-rot fungi including *Phanerochaete chrysosporium, Ceriporiopsis subvermispora, Pleurotus ostreaus, Cyathus stercolerus,* and *Pycnoporus cinnarbarinus* have been studied on a variety of biomasses with strong delignification capability. Even though a long cycle is required, this method has benefits, such as moderate conditions, lesser energy utilization, and pollution. However, a long cycle is required (H. Chen et al., 2017).

3.3.2.5 The recent advancement in pretreatment for Bioplastic production & degradation.

The synergistic impact of $Al(NO_3)_3$ and automated grinding is the most profitable among all other pretreatment strategies. The alkali-acid pretreatment method by microwave can enhance fermentable sugar content, and it has a significant impact on lignin extraction. The alkali pretreatment supported by ultrasound disrupts hydrogen bonds within lignocellulosic biomass (LCB) molecules and reduces their crystallinity. Moreover, radiation and chemical processes combined improve the degradation rate considerably more than single radiation or chemical process utilization (H. Chen et al., 2017).

Thermal and chemical bioplastic pretreatment was firstly investigated by Benn, N. & Zitomer, D., 2018 applying biochemical methane potential (BMP) assays on both the untreated bioplastics. The pretreatment was conducted for 3-48 hours at 35–90°C temperature and alkaline pH from 8-12. This process with PHB and PLA raised the BMP rate by more than 100%. Bioplastic pretreatment under 55°C and pH 10 conditions for 24h led to rapid and more total anaerobic co-digestion of the product. Studies on IL pretreatment have enhanced the recovery of hemicellulose and lignin from solutions following cellulose removal due to its eco-friendly characteristics. Genetic modification, adaptive procedures, and evolutionary engineering are hopeful alternatives in steam explosion pretreatment. Use of Electrical catalysis pretreatment for cellulose hydrolysis process with titanium-based electrode material (SnO_2 and CeO_2) with electric catalytic oxidation characteristics is also beneficial. The improved genetic engineering microorganisms will perform a crucial part in the LCB biological pretreatment (H. Chen et al., 2017).

3.3.3 Bioplastic production methods

Based on the origin of materials using and technologies used for bioplastic production methods are grouped into five main types (Figure 11).

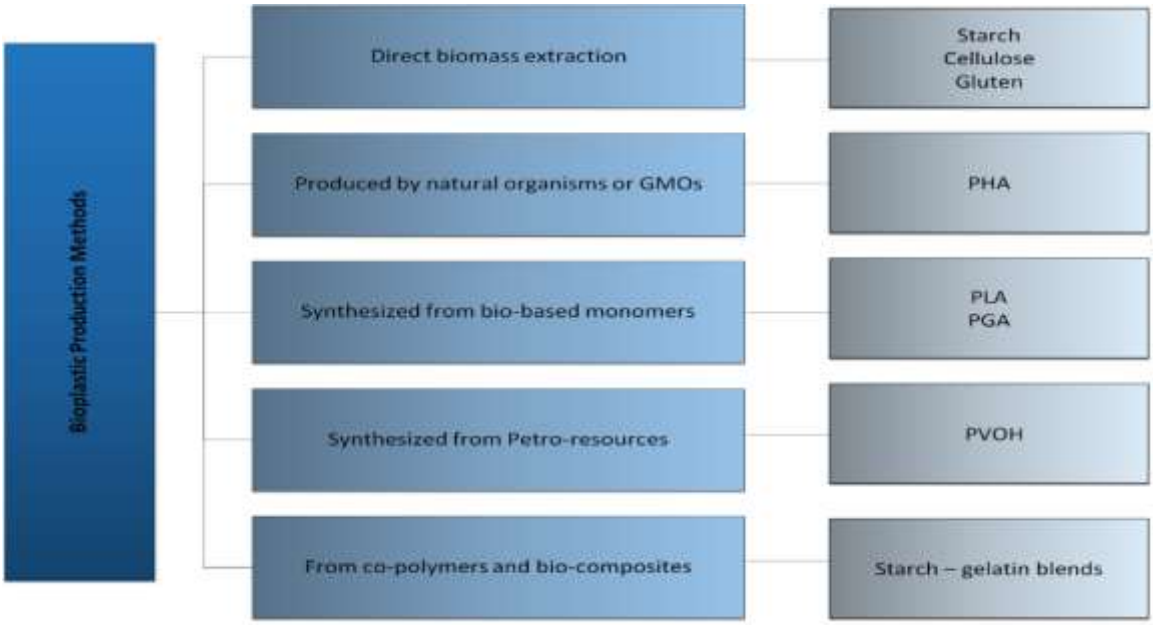


Figure 11: Bioplastic production methods

3.3.3.1 Bioplastics by direct biomass extraction

These bioplastics are produced by direct extraction of natural polymers like polysaccharides and proteins (Imre et al., 2019). Most of those bioplastics produced by direct extraction methods require additives or blending with other polymers to enhance their properties (Jõgi & Bhat, 2020). The blending with nanoparticles or additives and many chemical and physical techniques reduces poor material properties (Bilo et al., 2018). For example, Cellulose based bioplastic has attractive physical characteristics like a high Young's modulus of 114 GPa (for a single fibril), 89% of high crystallinity, high polymerization rates, and high specific surface area (Jõgi & Bhat, 2020).

3.3.3.2 Bioplastics provided by natural or GMOs

PHA: PHAs provided by natural or GMOs are degradable polymers (Geueke, 2014). Several microorganisms generate PHAs as carbon sources, usually under nutrient depletion (Dietrich et al., 2019). The PHA biosynthesis process is demonstrated in Figure 12. Around 250 types of natural PHA-producing bacteria have been observed, but only a less number of bacteria have been employed in PHA production on a large scale, including *Cupriavidus necator*, *Bacillus megaterium*, *Pseudomonas oleovorans*, and *Alcaligenes latus* (Tsang et al., 2019).

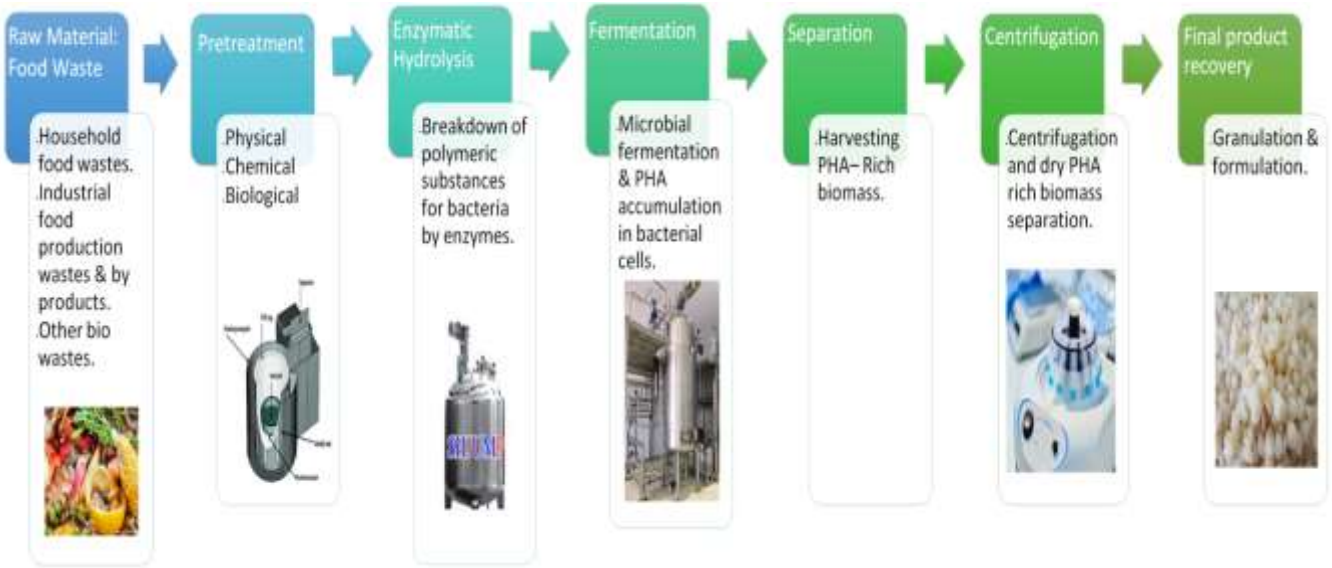


Figure 12: PHA biosynthesis process

Besides pure cultures, mixed microbial cultures and genetically engineered bacterial strains are employed (Albuquerque & Malafaia, 2018; Tsang et al., 2019). For example, recombinant *E. coli* generates 3HB from soybean oil. The microbial cultures for industrial PHA production are carried in batch and fed-batch reactors (Jõgi & Bhat, 2020). The cultivation process is basically about bacterial cell growth till proposed cell mass concentration and nutrient-limited growth with a carbon source (Tsang et al., 2019). Bioplastics production by natural or GMOs is briefly displayed in Figure 13.

Figure 13: Bioplastics provided by natural or GMOs

3.3.3.3 Bioplastics synthesized from naturally occurring monomers

PLA: PLA is produced by industrial polycondensation of lactic acid or lactide ring-opening polymerization (Jem & Tan, 2020; Castro-Aguirre et al., 2016). PLA's are generated by carbohydrate source, which is converted into dextrose, followed by fermentation into lactic acid and polycondensation (Figure 14) [Peelman et al., 2013]. Melt processing is the principal procedure applied for PLA mass production for the medical and packaging industries (Castro-Aguirre et al., 2016).

Figure 14: Bioplastics synthesized from bio-based monomers: PLA production process

PGA: PGA is synthesized from glycolide by ring-opening polymerization under the impact of metal salt catalysts at a low concentration (Yamane et al., 2014). The molar mass of the PGA polymer is determined by time, temperature, catalyst concentration, and chain transfer agents. PGA-based materials are immune to most organic solvents but are still comparatively sensitive to hydrolysis (Song et al., 2011).

3.3.3.4 Bioplastics synthesized by fossil fuel-based materials

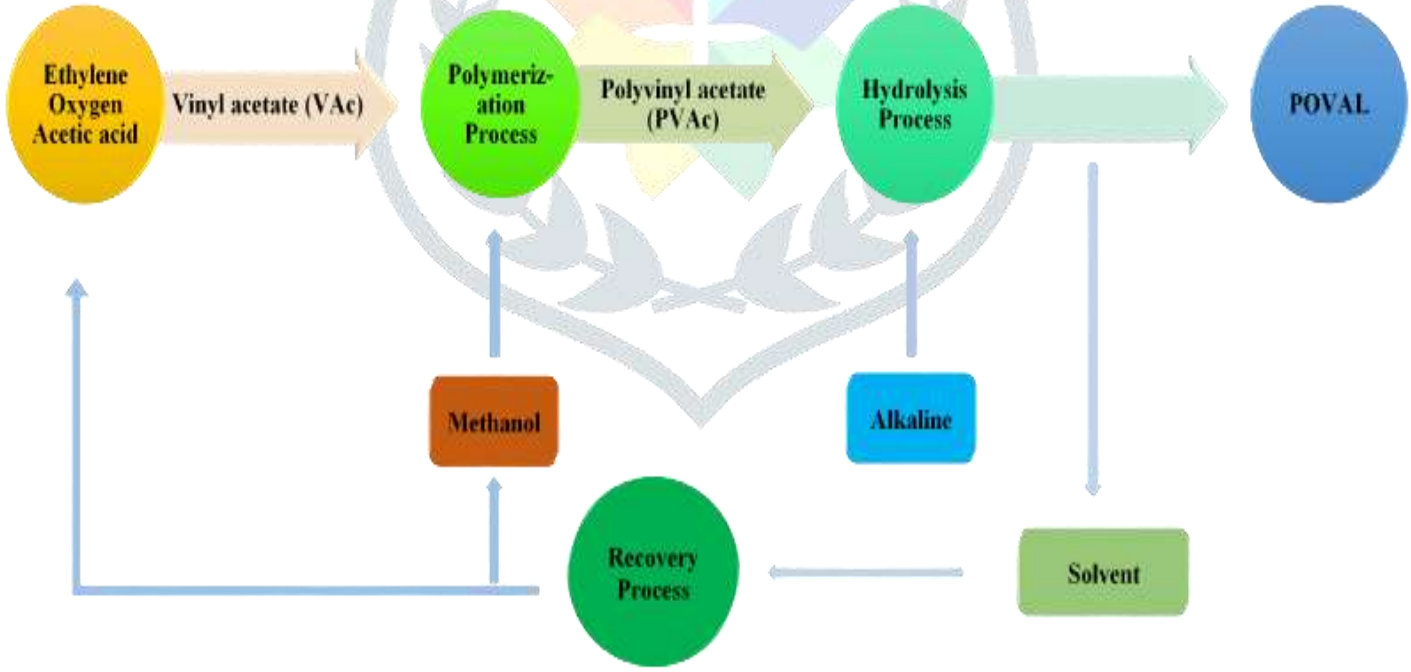


Figure 15: Production process of PVOH (POVAL)

Bioplastics generated from Petroleum-resources (e.g., water-soluble synthetic polyvinyl alcohol (PVOH/PVA) are costlier than conventional petrochemical plastics. Hence, they have limited application as a packaging material and are often mixed with Cellulose or starch (Alashwal et al., 2020). The polymerization

of vinyl acetate can produce PVOH to polyvinyl acetate (PVAC) and following hydrolysis (Figure 15) [Song et al., 2011]. PHOV is observed to have excellent biocompatibility and biodegradability (Jõgi & Bhat, 2020).

3.3.3.5. Bioplastics from copolymers and bio-composites

Most bioplastics are used as copolymers to enhance the properties of the materials, such as biodegradability and cost-effectiveness. One of the best bioplastic combinations in food is gelatin and potato starch combined with suitable plasticizers, including glycerol and sorbitol. The final product has almost high mechanical features than conventional plastics such as PVC (Podshivalov et al., 2017).

3.4 Analytical methods

3.4.1. Surface morphology of bioplastic

The microstructure of the bioplastic was studied through a Scanning Electron Microscopy (SEM) [Muralidharan et al., 2020]. It involves the characterization of sample surfaces and their cross-sections. Bioplastic samples are cut into 9mm^2 sections by a keen razor knife. Then film sections submerged in a 6% glutaraldehyde-0.1M phosphate buffer with 6.8 pH in a room temperature for 48h. Following being rinsed by phosphate buffer for 1h, and sample strips evaporated in an ethanol graded series (15-100%). Then blocked by an epoxy resin and formed conductive including a 60:40 gold-palladium compound stored using a Hummer sputter coater with 400 Å thickness. For bioplastics subjected to bacterial degradation, samples are rinsed by distilled water, dehydrated, and then examined through SEM (Ierro et al., 2007). Alternatively, samples are preconditioned in a desiccator and sliced into 1cm x 1cm-sized samples mounted over Aluminium stubs applying the both-sided sticky tape. The samples are sputter-coated with a slim gold layer. Then samples are examined in 10 kV accelerating voltage. Finally, samples are observed at 500x to 2500x magnification levels (Muralidharan et al., 2020).

3.4.2 Mechanical Properties.

3.4.2.1 Thickness

The bioplastic film thickness adapted in 25 °C, 46% relative humidity (RH) for 72h estimated by an electronic digital micrometre with ± 2 µm sensitivity. Film strips set between the micrometre clamps and lessened the gap until the minimum resistance was measured. Mean thickness was calculated using average measurements in five sites (Ierro et al., 2007). This average value is applied to determine the bioplastics' cross-sectional area (de Azevedo et al., 2020).

3.4.2.2 Tensile strength (TS), elongation at break (EAB), and modulus

These characteristics are estimated using the Instron universal electromechanical testing equipment (Figure 16) [de Azevedo et al., 2020]. Bioplastic samples are sliced into pieces with 10-11 mm breadth and 100 mm in length by a sharpened razor knife. Then pieces equilibrated at 50 ±5% RH and 23 ±2 °C for 24h in an

environmental chamber. All the sample strips located within the pneumatic clamps in Instron are set earlier at 90 mm initial standard reading, and the pieces are then pulled at a 30 mm min-1 rate till sample collapse happens. Estimations of the weight (N) and deformation (mm) employed in evaluating the tensile strength and the elongation at break. Lastly, the modulus was discovered from force versus deformation measures by a simple extension (Ierro et al., 2007).

Figure 16: Universal testing machine - 3382A – Instron

3.4.2.3 Dynamic Mechanical Analysis (DMA).

Ullah et al. (2011) used Perkin-Elmer dynamic mechanical analyzer in measuring bioplastics' dynamic mechanical characteristics in the tensile form at a 1 Hz oscillatory frequency with 0.05 mm deformation while heating. Analyses held on rectangular specimens' dimensions of about 11 × 6 × 0.8 mm (length × width × thickness). A temperature scan at 0-160 °C was conducted at a 2 °C/min heating rate. Then specimens equilibrated at 0% RH in a desiccator for 2 weeks with P_2O_5 (Dubey et al., 2018). described that DMA's bioplastic mechanical strength was estimated by NETZSCH DMA apparatus. The scan was done at -50 to 300 °C in 5°C /min and 1 Hz tension.

3.4.3 Spectral characterization

3.4.3.1 Fourier transformed infrared spectroscopy (FT-IR) Analysis

FTIR (Figure 17) exhibits various conformational bands related to bioplastic. A study by Zhang et al., 2020, the exterior functional groups of bioplastic films was recognized by FTIR spectrum read at room temperature

with 2cm^{-1} resolution, varying from 400cm^{-1} to 4000cm^{-1} per sample. FTIR microscope spectrometer (Hyperion 2000) altered by germanium attenuated total reflection (ATR) microscope. A mercury cadmium telluride (MCT) detector is also employed. Sample cut into small parts and equilibrated at 0% RH in a desiccator containing P_2O_5 for 14 days before FTIR examination. Then spectra gathered in the 4000-650cm^{-1} frequency range, following the exact environmental requirements (Ullah et al., 2011). Liu et al. (2020) conducted FTIR in Shimadzu IRAffinity-1S spectrophotometer, including SpecacQuest ATR associate. Results were obtained in LabSolutions IR at %Transmittance method including Happ-Genzel Apodization, average scans of 128 by 400-4000 cm^{-1}. Spectrum was normalized through multipoint baseline improvement and softening (M. Liu et al., 2020).

Figure 17: Fourier transformed infrared spectroscopy

3.4.4 Thermal analysis of bioplastic films

Differential Scanning Calorimeter (DSC), Thermogravimetric analysis (TGA), and also differential thermal analysis (DTA) employed to investigate the composition and crystal formation of the bioplastic films supporting the N_2 with a 10°C min^{-1} heating speed from 30-800°C (C. Zhang et al., 2020).

3.4.4.1 Differential Scanning Calorimeter (DSC)

The thermal properties such as the glass transition (Tg) and melting (Tm) temperature of the biopolymers are estimated using a DSC (Figure 18). Dubey et al. (2018) operated extracted polymer samples in -20 to 500°C temperature range at 10°C min^{-1} heating rate in a DSC applying DSC 204 F1 Phoenix apparatus including Netzsch software. The native trimming hydrolysate powder (TH), PVA, and control sheets like TH-CA and PVACA were used (Muralidharan et al., 2020) for thermal analysis. Each sample was held at 25 °C before sampling for DSC. Then a 5 mg unit cut from the samples and put in the sample pan. Samples examined at 10 °C/min heating rate from 0 to 300 °C to discover the bioplastic films' heat stability.

Figure 18: Differential Scanning Calorimeter from NETZSCH-Gerätebau GmbH

3.4.4.2 Thermogravimetric analysis (TGA)

The thermal degeneration temperature of the bioplastics such as PHB is estimated from TGA (Reddy et al., 2019). The temperature at specific weight loss of bioplastic is used as the decomposition temperature (Td) to assess the thermostability (Venkateswar Reddy et al., 2014). The recovered polymer sample is subjected to TGA with the TG–DTA system in TG 209 F1 instrument. Analysis was conducted over 25°C to 500 °C at a 10 °C min^{-1} heating rate supporting a N_2 atmosphere (Dubey et al., 2018).

3.4.5 Molecular mass determination

According to Hermann-krauss et al. (2013), the average number molecular mass (Mn) and molecular mass dispersity index, Pi (Mw/Mn) is measured through gel permeation chromatography (GPC) within $CHCl_3$ solvent at 1 mL/min flow rate in 35 °C, employing a Spectra-Physic 8800 solvent transfer method with two PLgel 5 µm MIXED-C ultra-high-efficiency column and the refractive index detector (Shodex SE 61). A 10µL sample in CHCl3 at a 1% w/v concentration was inoculated. Polystyrene with small molecular mass diffusion was applied in creating a calibration curve.

3.4.6 Film Water Vapor Permeability (WVP)

A gravimetric test estimates the film water vapour permeability according to the ASTM E9618 procedure. The film area having access to vapour transmission was observed as $10cm^2$ (Ierro et al., 2007). The water vapour transmission speed through the film was determined by the straight portion of the diagram achieved by plotting the weight increment of the Silica gel cups used in this experiment cup as a function of time. It was thought that a constant state was attained once the regression analyses made by utilizing the last four data occurred in $r^2 \geq 0.998$ (Ierro et al., 2007).

3.4.7. Transparency and Opacity

According to Azevedo et al. (2020), colour investigations of bioplastic were discovered using Hunterlab colourimeter Miniscan EZ. The transparency and opacity of bioplastic were examined using rectangular pieces of the biofilms, sized 0.8 x 30 mm, that adhered to the interior wall of a quartz cuvette used to remain vertical to the light beam to record transmittance spectrum between 200-800 nm UV-VIS spectrophotometer using a blank as a control (de Azevedo et al., 2020).

3.4.8 Biodegradability Tests

In the degradation of biofilms in the vivo environment(soil), the three different types of bioplastic films kept for UV sterilization (30 min at 15 cm from UV lamp) and fully coated with a layer (5-6 cm) of clay and loam mix soil. The observed soil-water contents at field capacity (in situ) and -1.5 MPa were similar to 27.6 and 13.6% by weight, individually, and the density was equal to $1.25tm^{-3}$. The soil was stimulated by water scattering around 10% of the soil's total weight. Within 7 and 21 days of incubation period at 30°C, soil microflora was completely estimated by microbial counts on PCA (Ierro et al., 2007). A biodegradation assay of the starch-based bioplastics was performed by preparing 4 x 10 cm sized pieces and pressed into voile covers with four parts of each sample per cover. They were then immersed in 20 cm of the soil pitch. Samples were examined every 48 hours for 14 days. After that, the frequency of measurements was lessened to 7 days. The total investigation period was 40 days. The measurements were taken according to the bioplastic area and completed when the samples reduced their area and degraded (de Azevedo et al., 2020).

3.4.9 Antibacterial activity

The bioplastic films (trimming-based) were examined for in-vitro antimicrobial activity using the agar overlay method described in Kemme & Weiland, 2018. The process connects the disc diffusion method, and according to the Kirby-Bauer susceptibility test, the method is modified from seeded lawn overlay spot assay, and action was examined by observing growth inhibition zones (Muralidharan et al., 2020).

3.4.10 Static water contact angle

Liu et al. (2020) described the static water contact angle estimated by the rame-hart Model with 500.1 mL drops of Milli-Q water found on every type of Tea waste bioplastic. Contact angles estimated 10 times over 10 s, at both left and right Θ values equalized to which results in a one mean contact angle (M. Liu et al., 2020).

4. Properties of bioplastic

4.1. Biodegradability

Bioplastic biodegradability relies on the Physiochemical arrangement of the biopolymer. For instance, aromatic polyesters are highly sensitive to microbial degradation, and aliphatic polyesters degradation

occurred through hydrolyzable ester linkages. Moreover, medium pH, O_2 content, temperature, and moisture like environmental requirements significantly impact biodegradability. Usually, biopolymers' biodegradation includes polymer decomposition through breaking hydrolytically and enzymatically susceptible linkages. Many naturally synthesizing polymers undergo enzymatic degradation by reacting to hydrolase enzymes such as protease, urease, esterase, and environmental degradation of artificial polymers abiotic hydrolysis (Jõgi & Bhat, 2020).

An ideal environmentally biodegradable material maintains its structural integrity, free from the environmental conditions, for a defined time with minimum or no degradation, and experiences accelerated degradation after the bioplastic has gained its desired functional outcome or completion of functional purpose. The degree of disposable plastic degradation relies on various factors, including chemical alteration of the biopolymer backbone, morphology or structural changes, modifications to the polymer composition, choice of production requirements, structure of the polymeric substance, and substance after-treatment. The stereoisomerism content, overall crystallinity, molecular weight, and accessible volume are all factors that influence the morphology or structure of polymers. Raising the molecular weight of biopolymers decreases their degradability. For instance, high molecular weight (Mn > 4000) PCL degraded more gradually by Rhizopus delemar lipase than in low Mn PCL. Polymer composition modified by contaminants or joining several additives. Material of the polymer altered by modifying its exterior area, porosity, and density. Plasticizers are also applied to modify and regulate the disposable bioplastics degradation rate (Niaounakis, 2013)

4.2. Tensile strength (MPa)

Tensile strength is the highest pull that can be achieved until the bioplastic film can endure before breaking up or the maximum pressure that the material can accept or measurement to discover the amount of force gained to obtain the highest pull in each film widen or elongate. The tensile strength of bioplastic films is highly affected by the combined plasticizer concentration. When the plasticizer concentration increases, the lower tensile strength is observed (Sofiah et al., 2019; X. Zhao et al., 2020). Moreover, hemicellulose plasticization transesterification, including vinyl laurate, improved tensile elongation of hemicellulose laurate to around 200%. Bioplastic tensile elasticity reduced as thickness diminished and as temperature raised, bioplastics' intended flexibility enhanced as temperature increased. As the temperature decreased, the plastics' mechanical properties improved (Dianursanti et al., 2018). The mechanical performance of biopolymer also improves blending with other polymers (X. Zhao et al., 2020).

4.3. Elongation

Elongation is the ratio between the maximum length of the sample before the collapse and its initial strength (Dianursanti et al., 2018)or the difference in material length emerging from tensile force (Zhou, 2016). Elongation Percent is a change in the maximum length of the film before being separated (Sofiah et al., 2019).

Plasticizer and compatibilizer can increase the elongation of the bioplastics (Dianursanti et al., 2018). Sofiah et al. 2019 observed that elongation of bioplastic film produced by adding glycerol and sorbitol ranges from 6.0606% - 15.1515% and 3.0303% 15.15152%, respectively. Hence, the higher the plasticizer concentration in bioplastic production, the higher the percent elongation of bioplastics.

4.4. Modulus of elasticity: Young's, shear, and bulk modulus

Modulus is the standard ratio of a distinct stress type to a particular pressure (Sofiah et al., 2019). Modulus is also defined as the measurement of a bioplastic's resistance to flexible deformation. If a bioplastic has a large modulus of elasticity, it prevents deformation and is recognized as a rigid body. If a bioplastic has a low modulus of elasticity, it enables deformation and is recognized as flexible or non-rigid.

4.5. Opacity & Transparency

The opacity of the bioplastic is the ratio of the daylight amount imitated from a material to the incident light amount transmitted by the material (Gonzalez-Gutierrez et al., 2010) described that during the production of highly permeable starch or protein-based bioplastics, proteins form a web of narrow aggregates and the ending gel seems translucent. Also, heat processing was recognized to damage starch granules, appearing in enhanced transparency. Therefore, thermo-mechanical processing is an acceptable method to achieve highly translucent (38% transmittance) albumen or starch-based bioplastics with high elasticity. Gelation was recognized in modifying the starch semi-crystalline arrangement into an amorphous structure, significantly improving transparency.

4.6. Water vapour transmission rate (WVTR)

The water vapour amount is transferred by a specimen within a fixed time, supporting the controlled temperature, age, and RH (Sofiah et al., 2019). Low water vapour permeability (WVP) indicates that bioplastic can restrict moisture transfer among food and the environment. Hence, bioplastic with low WVP is the most suitable for food packaging (Azmin et al., 2020). In packaging, WVPR protects food against degradation by water (Sofiah et al., 2019).

4.7. Oxygen transmission rate (OTR)

OTR is an O_2 quantity transferred via the substance within a particular time, temperature, RH, plus pressure. Support food protection against oxidation and prevents microorganism growth in food packaging (Sofiah et al., 2019).

4.8. Thermal properties

Thermal properties of biopolymer are essential, which give information regarding thermal and oxidative stability of biopolymer, phase and phase transfers occurring in polymer and their shelf life under definite conditions (Mumtaz et al., 2010). Poly (3-hydroxybutyrate-3-hydroxy valerate) [PHBV] thermal properties

are enhanced by chemical modification, grafting, the inclusion of alternative polymers and nanofillers like nanocellulose, nano clay, nano metal, and nanocarbon materials (X. Zhao et al., 2020).

4.8.1. Decomposition temperature (Td) & Thermostability

The study allotted by (M. R. Amin et al., 2019) declared that the solid surface interaction developed the rate of nanocomposite crystallinity within TiO_2 nanoparticles and the series's amorphous region. Starch bioplastics' Td was improved by adding oxide nanoparticles. The Td of composite bioplastic is on top of starch bioplastic. It demonstrates that starch bioplastic enhances Td once supplementing TiO_2 (nanoparticles), and higher heat resistance was observed in composite bioplastic resembling starch-based bioplastic. For example, the attachment of PVP units into PHBV backbones developed thermal resistance, and hydrophilicity restrained the crystallization method, plus reducing PHBV plasticity. The 30% rise in PLA heat deformation temperature is observed when improving the degree of crystallinity (X. Zhao et al., 2020).

4.8.2. Melting temperature (Tm) and Glass transition temperature (Tg)

In examining biopolymer biodegradability, it is also necessary to study the polymer's melting temperature (Harmaen et al., 2016). Tg is a temperature in which the polymer losses its glass-like presentation and becomes rubbery and more flexible (Mumtaz et al., 2010). As higher the biopolymers' Tg, the higher their barrier characteristics. Tg signifies the consistency and miscibility of constituents within a biopolymer. Recognizing a single Tg for every plastic implies that each ingredient offered over the bioplastic structure has been adequately expanded and consistent within all (M. R. Amin et al., 2019). Amin et al. (2019) discovered that the maximum 297°C (Tm) in starch bioplastic and 303°C (Tm) in composite bioplastic, and composite bioplastic Tg observed was about 66.8°C which higher than starch bioplastic Tg (57.2°C). The antiplasticizers (TiO_2) blending with starch bioplastic improves the Tg.

Conclusion

Due to the high number of problems raised with non-degradable plastics, the world's focus centred on degradable plastics as they show excellent biocompatibility, biodegradability and many advantageous mechanical properties. Bio-based waste materials from various origins have been identified as profitable, readily available and efficient raw materials for producing these biodegradable plastics. Biopolymers originated from fossil fuel-based resources comprising polymers that are being biodegradable. Numerous bioplastics have material properties similar to conventional plastics giving more value through decreased carbon footprint and environmentally favourable waste-management choices like biodegradation and industrial composting. Cost-effectiveness narrows the bioplastic production and usage. Few waste valorization methods are presently executed on a broad scale for waste management and production of bioplastic, although a future investigation is led to the genetic engineering, pretreatment methods, and biorefinery principles. The bioplastics were chemically and mechanically characterized by various analytical methods, including uniaxial

tensile tests, dynamic mechanical analysis, and bioplastic surface, characterized through contact angle measures, SEM, and confocal microscopy.

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