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Research Article

Perspective on the production, characterisation and physio-mechanical behaviour of bioplastics from different biomass feedstocks

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ABSTRACT

Due to the rapid growth in the world's population and industrialization, the demand for plastic has increased over the past few years. Synthetic plastics are produced from fossil fuels and their continuous use has significantly increased the global greenhouse gas (GHG) emissions making the carbon footprint of plastics high. These conventional plastics are not easily degradable, and their disposal poses an environmental threat to humans and aquatic organisms. Several innovations are ongoing to produce bioplastics from alternative materials that are renewable, easy to dispose of and biodegradable without compromising their physical and mechanical properties. The substitution of synthetic plastic with bioplastics from plants and animals' by-products will significantly decreases the amount of packaging waste generated. Numerous biomass feedstocks from polysaccharides, lipids and proteins are used in the production of bioplastics. However, the mechanical properties of bioplastics are continuously improved by using various reinforcement techniques to enhance their properties and increase their applicability. The influence of the type of feedstock, synthesis techniques, type and concentration of plasticizer and fillers blended with the feedstock on the physio-mechanical properties and degradation of bioplastics was reviewed in this study. The opportunities and challenges of bioplastics from biomass were also outlined.

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INTRODUCTION

The surge in population across the world and the industrial revolution have increased the global demand for plastics. The global annual plastic production over the years increased from 234 million tonnes (Mt) in 2000 to 460 Mt in 2019 as shown in Figure 1 [1]. Most of the petroleum-based plastics like polyethylene, terephthalate, nylon, polystyrene, teflon, polyethene and polyamides are known as synthetic plastics, and they have wide applications in the packaging and agricultural sectors. The main elements of synthetic bioplastics are carbon, hydrogen, nitrogen, oxygen, chlorine, and bromine and the degrees of crystallinity, chemical structures and molecular weights

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determine their rate of biodegradation [2]. Despite the non-biodegradability of synthetic plastics, their desirable properties such as lightweight, malleable, excellent thermal and rheological properties, and low cost of production have made them ubiquitous in the global economy over the years [3]. According to the OECD report [1], the continuous use of plastics has increased plastic waste from 156 Mt in 2000 to 353 Mt in 2019, about 9% of plastics were reused or recycled, 50% were dumped in controlled landfills, 19% were destroyed by burning while the residue 22% are dumped in uncontrolled sites, burnt in open pits, or enter the marine environment. The dumping and build-up of waste plastics in landfills and marine environment are the primary cause of ground and water pollution and poses a danger to the health of humans and aquatic animals. The production of synthetic plastic and its disposal contributes significantly to global greenhouse gas (GHG) emissions making the carbon footprint of plastics high. The average lifetime of synthetic plastic products is almost 10 years, therefore plastics buried in the ground pollute groundwater and cause danger to the well-being of living organisms. Alternative biodegradable plastics feedstock and proper management of plastics waste are necessary to reduce the GHG emitted into the atmosphere. The gradual degradation and incorporation of benzene into the food when synthetic plastics come in contact with food make synthetic plastics unsuitable and unsafe in the food packaging industries.

Innovations, excellent product designs, improved waste management, an increase in recycling, and the promotion of environmental and biodegradable polymers are needed to reduce the pollution and contamination from plastics. Biodegradable polymers are easily degraded by microorganisms (algae, fungi, yeast, bacteria, and insects) into biogases, biomass, and products with low molecular weight [4,5]. Environmental conditions like temperature, light, environmental pH, the presence of microorganisms, presence of oxygen and water influence the rate of bioplastics degradability [6,7]. Biodegradable polymers are commonly synthesized through the processes of fermentation that involve microorganisms and chemical modification of natural materials such as polysaccharides [5]. Several innovations are ongoing in the field of biological sciences and engineering to produce plastics tagged "bioplastics" which are synthesized from biomass, easy to dispose of and degrade without harming the environment. Bioplastics are synthesized from various biomass sources like lipids, proteins, and polysaccharides [8-10] as shown in Figure 2. The utilization of these bioplastics in the medical, packaging,

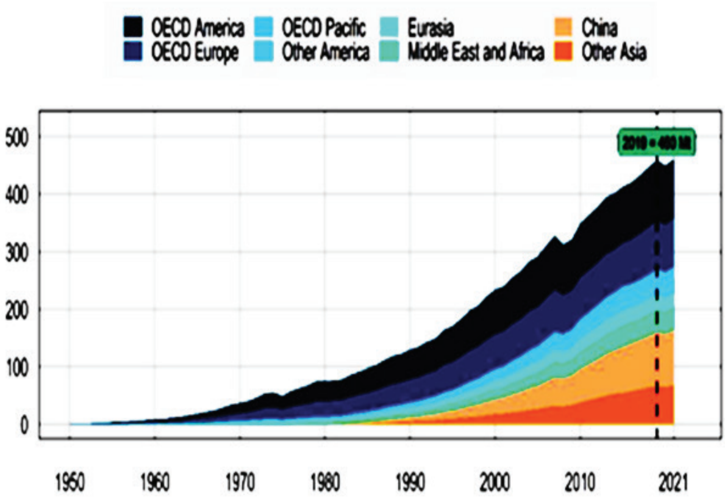

Figure 1. Global plastic use in million tonnes (Mt) from 1950-2021 [1].

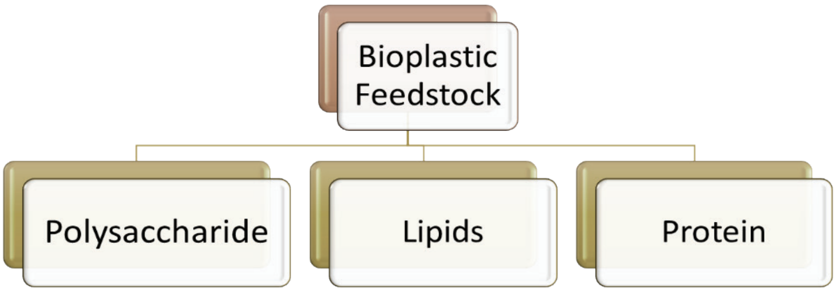

Figure 2. Bioplastic feedstock from Biomass.

electronics, construction, and agricultural industries will reduce energy consumed from fossil fuels and the emission of GHG, thereby making the environment suitable and safe for humans and aquatic animals. This review focuses on the techniques used in the production of bioplastics from polysaccharides, proteins, and lipids. The influence of various parameters and additives on the physical and mechanical properties of bioplastics considering the effect of micro-fibers and nanoparticles on the enhancement of the properties of bioplastics was discussed. Furthermore, the challenges and potential strategies required to improve the properties of bioplastics making them commercially sustainable were highlighted.

STARCH-BASED BIOPLASTICS

Polysaccharides are macromolecular polymers obtained from biomass like plants, microorganisms, and algae [11]. They are categorized according to the class of monosaccharide, their physiological properties and building blocks [12]. Pure starch is a carbohydrate that is usually white in colour, odourless, nontoxic, not soluble in cold water or alcohol and contains a high amount of glucose and they are extracted from polysaccharides. Branched amylopectin and linear and helical amylose are the types of molecules present in starch [13]. The gelatinization, viscosity, tensile properties, and functional and chemical properties of bioplastics are influenced by the percentage content of amylopectin and amylose in the starch [14-16]. Almost 50% of bioplastics produced for commercial packaging are starch-based. Starch is favourable in the production of bioplastics because they are cheap, renewable, sustainable, plentiful, low-cost of production, biodegradable and possesses suitable tensile properties [17,18].

Extraction of Starch from Polysaccharides and the Preparation of Bioplastics

The extraction of starch from polysaccharides such as yam, maize, rice, potatoes, cassava, corn etc. involves the milling of the seeds and tubers with distilled water using a high-speed blender to obtain paste. The paste is filtered using a filtering cloth to get the crude starch filtrate liquid. The filtrate liquid is let to sediment at room temperature for the pure starch to settle. The supernatant is poured away, and the pure starch obtained is dried using an oven at 70 °C to evaporate the remaining water. The starch is sieved with a strainer for better homogeneous size [19-22]. Despite starch being abundant in nature and cheap, it cannot be used as packing polymers without additives due to its high-water solubility and poor mechanical properties. These limitations need to be overcome before starch can be considered a sustainable feedstock in bioplastics production. Additives such as plasticizers are added to starch to boost its mechanical properties and lower the water solubility of bioplastics. The commonly used plasticizers are sorbitol, formamide, urea, xylitol, and glycerol. Glycerol is mostly used because it is from natural sources, and they are edible and safe to use for packaging without any form of contamination with food.

Bioplastics are usually produced using thermomechanical and casting methods. The thermomechanical method consists of the extrusion, injection moulding, thermomoulding and compression moulding methods [23,24] as shown in Figure 3. The extrusion technique involves the use of an extruder with a spindle barrel and electric heating system to extrude the dough like bioplastic and the injection moulding is a closed mould process involving the injection of bioplastic polymer into the mould under elevated pressure and temperatures. The thermomoulding technique is usually employed to produce line-walled and lightweighted products from preextruded plastic sheets while the compression moulding technique involves the application of mechanical pressure on the heated polymeric material placed in the lower side of the mould. The casting method is commonly used in the production of starch-based bioplastics [25-28]. Thu and Aye [29] used the casting method to produce bioplastic film from dent corn

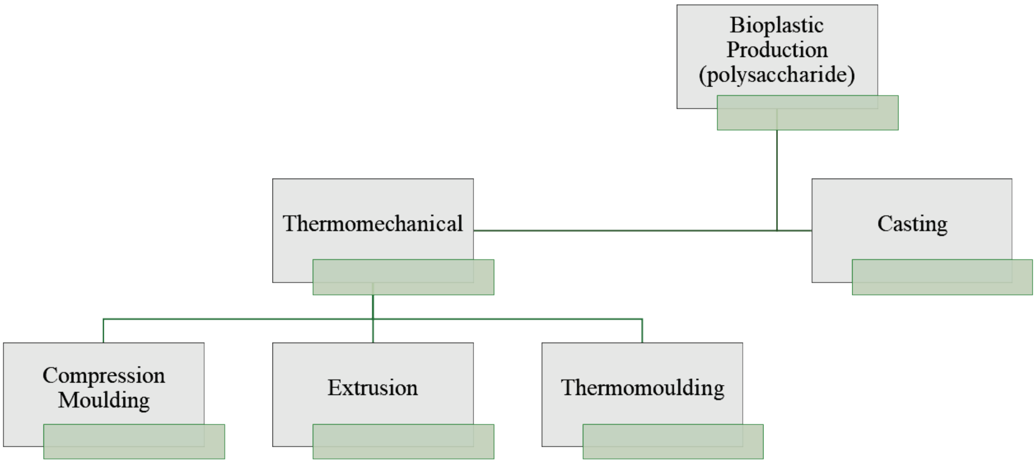

Figure 3. Bioplastic production methods for polysaccharide.

starch using 0.5 g of glycerol and 20 mL of distilled water. The gelatinized mixture was transferred and pasted on a substantial steel plate and allowed to dry in the oven for 90 minutes at 50 °C. It is very important to ensure that air bubbles are removed and not formed in the bioplastic film when using the casting method. The presence of air bubbles affects the characterization and properties of the bioplastic film. Gustafsson et. al [30] employed compression moulding and solution casting methods to produce bioplastic film from apple pomace blended with glycerol. In the compression moulding, the glycerol is blended with apple pomace powers before compression to produce a 3D object while in the solution casting, the blend of apple pomace powder, glycerol and citric acid was heated to 70 °C while stirring the mixture on the magnetic stirrer and later poured on a non-stick plate. Santana et al [31] and Nguyen et al [32], produced bioplastics from jackfruit starch using the casting method. Cassava and sugar fiber hybrid composite bioplastic film was produced by Edhirej et al. [33] employing the casting technique with fructose as the plasticizer. Most of the literature reviewed showed that starch-based bioplastics were mostly produced using the casting method. Further studies should be conducted to investigate the impact of production techniques on the characterization and physical mechanical properties of bioplastics.

Physical-Mechanical Behaviour of Starch-Based Bioplastics

Natural starches have some limitations relating to thermal stability, water absorption and mechanical properties [34]. To overcome these challenges, certain additives made from natural or synthetic materials are blended with starch to make them thermoplastics and enhance the material processing, properties, functional behaviour and widening their applications [35]. The resistance of bioplastics to moisture is improved through the addition of synthetic polymers, cross-linked agents, slats and lignine while the flexibility of starch is improved through the blending of plasticizer [36]. The interaction of the polymer-polymer hydrogen bond is reduced by the plasticizer, thereby reducing the intermolecular binding sites starch in granules' crystalline region and enhancing the interfacial adhesion [37]. The required properties of the bioplastic can be achieved by varying the quantities of the additives such as glycerine, water, fillers, and eco-friendly polyesters.

Thet [8] studied the effect of glycerine (0.25g - 1.0g), and water (6 ml - 10 ml) content on the biodegradable characteristics of plastics produced from corn starch. The viscosity and the thickness of the plastic produced were observed to be proportional to the content of the glycerine. The volume of water in the sample affected the viscosity, number of bubbles and translucency of the plastic produced. The most suitable condition for biodegradable plastic produced from corn starch was corn starch of 1.5 g, glycerine of 0.5 g, distilled water of 7 ml and 1 ml of vinegar. The product at these conditions had no bubbles, good tensile strength, and less drying time. The degradability test showed that bioplastics can degrade within 28 days when buried in the soil or soaked in water. To further enhance the properties of corn starch bioplastics, Marichelvan et. al [20] added rich starch to the corn starch. The higher amylose concentration of the corn and rice starches motivated the authors to combine them. The quantities of water, glycerol, citric acid, and gelatine used in the preparation process were constant but the quantity of rice to corn starch was varied. The mechanical resistance against rupture of the bioplastics impregnated with rich starch was 1.5 times higher than that of a non-impregnated counterpart. Samples with the ratio of rice to corn starches of 7:3 had an elongation of 6.8%, Young's modulus of 0.183 GPa, and maximum tensile strength of 12.5 MPa. The thickness of the bioplastics obtained was 250 microns. Comparing the thickness of the bioplastics with other works [38,39], the higher thickness was attributed to the presence of the corn starch but the influence of concentration of glycerol on the bioplastic thickness was not considered in this study. The biodegradability of the sample achieved after 15 days was 48.7%. Potatoes and corn starch were used as feedstock to produce bioplastics with glycerol and acetic acid as additives by Al Balushi et al. [19]. The bioplastics with higher content of potato starch had lower flexibility and durability but they were soft and transparent while those with higher corn starch were rough with low transparency, but the durability and flexibility were enhanced. The degradation rate of bioplastics is influenced by the proportion of corn starch and potato starch in each sample. The low durability and flexibility of the potato-based bioplastics can be attributed to the percentage of amylose content in the starch. According to Young [40], the amylose of potatoes is lower than that of rice and corn starches. Ceseracciu et al. [14], stated that the tensile property of the bioplastic is proportional to the amylose content in starch.

The low durability and non-adhesion between glycerol, water and starch affect the application and properties of the bioplastics respectively. To overcome this challenge, citric acid and sodium bicarbonate were blended with jackfruit seed starch by Nguyen et al. [32] to enhance their mechanical and chemical properties. The citric acid strengthens the adhesion between the glycerol, starch, and water and effectively inhibits starch retrogradation due to its strong hydrogen bonding interaction with starch while the sodium bicarbonate was added as a preservative to strengthen and make the bioplastic more durable. The study observed that the bioplastics produced were softer and more flexible when the ratio of the glycerol increased but increasing the citric acid concentration in the bioplastic improved the hardness and decreased the plasticity of the bioplastics. The maximum tensile strength of 5.15 MPa was recorded with a starch glycerol ratio of 3.5:1 but micro air bubbles were found inside the bioplastic thereby affecting its mechanical properties. Jiugao et al. [41] investigated the influence of citric acid on the properties of thermoplastic starch plasticized by glycerol and stated that the citric acid decreased the shear velocity and enhanced the fluidity of thermoplastic starch but decreased the tensile stress. The water resistance is improved at high relative humidity, thereby elongating the durability of the bioplastic.

The oil content in starch-based plants has been observed to influence the mechanical properties of bioplastics. To further enhance the applicability of bioplastics from rice, the oil and fiber fraction in rice bran was extracted by Alonso-Gonzalez et al. [42], the mechanical, functional, and microstructural properties of rice bran-based bioplastics with sorbitol and glycerol as plasticizers were investigated. The result showed that the viscoelastic moduli of the defatted rice brain bioplastic were higher and had better tensile properties, but the vapour uptake capacity decreased. There was no significant improvement from the fiber-free rice bran-based bioplastics. Considering the influence of plasticizer type, the glycerol-plasticized bioplastics showed a lower glass transition temperature and higher plasticizing efficiency with higher deformability. While the bioplastics from sorbitol plasticizer exhibited high-stress values and better stiffness. The blending of starch with plasticizer improves the flexibility and workability of the bioplastics but the hardness is reduced [43]. The high susceptibility to humidity and retrogradation process has limited the application of bioplastic from starch [44].

Physical-Mechanical Behaviour of Micro-Natural Filler in Starch-Based Bioplastics

The poor mechanical properties and high moisture absorption of starch-based bioplastics have limited its application. One of the foremost innovations adopted in addressing these shortcomings is the blending of starch-based bioplastics with less hydrophilic polymers known as fillers. Fillers are more economical because they are less expensive, and they act as a primary load-bearing component. Fillers such as cellulose from plants are used to improve the mechanical properties, decrease elongation capacity, increase Youngs' Modulus, and enhance permeability to gas and resistance to water [45]. Natural fillers were added in the production of bioplastics by Shafgat et al. [26] to improve the chemical and physical characteristics of bioplastics produced from banana peels, rice, and corn starches. The bioplastics produced were reinforced using waste agricultural products from potato peel powders and sawdust as fillers. In this study, the content of plasticizers (glycerol, sorbitol) and fillers in the bioplastics produced were varied. The ratio of banana peel paste, corn and rice starches solution were blended in the ratio of 40:30:30 respectively. The absorption of water by bioplastics was reported to be influenced by the nature of the filler and its content. Introducing plasticizer into the bioplastics enhanced the rate of biodegradation while the addition of fillers (5% w/v) reduced the biodegradation rate of plasticized bioplastics and enhanced it in the unplasticized bioplastics. Bioplastics with glycerol had the least Youngs Modulus and tensile strength while those with sorbitol had the highest Youngs Modulus and tensile strength. These properties increased proportionally with the content of the filler in both the glycerol and sorbitol bioplastics.

To decrease bioplastics' moisture absorption and improve its mechanical properties, Maulida [21] reinforced cassava peel starch with microcrystalline cellulose (MCC) of particle size 50 µm using varying content between 0 to 6% dispersed in water. The sorbitol was used as the plasticizer and the concentration varied between 20- 30 % wt/v to the starch. The increase in the content of MCC improved the tensile strength and this was credited to the excellent interfacial adhesion that formed a strong 3D hydrogen bond network between the starch matrix and the microcrystalline cellulose fillers. Due to the hydrophilic nature of water, cellulose is less sensitive to water than starch. Therefore, increasing the concentration of MCC (0-6%) led to a decrease in the water uptake. The maximum tensile strength of 9.12 MPa was observed at a microcrystalline cellulose content of 6% and 20% of sorbitol. The combination of MCC particles with the starch matrix led to agglomerates that led to deflections in the samples. Abdullah [46] also investigated the effect of microcrystalline cellulose reinforcement derived from high-quality wood pulp on the mechanical, physical, and biodegradable properties of bioplastics derived from cassava starch using glycerol, potato dextrose agar, and salt agar as additives. The thickness and the density of the bioplastics produced increased as the content of the microcrystalline cellulose increased. Increasing the content of microcrystalline cellulose enhanced the hydrophobic properties of bioplastics. The elongation reduced while the thermal stability and Young's modulus increased with an increase in the microcrystalline cellulose. Due to the chemical similarity between the starch and the cellulose, the adhesion of the microcrystalline cellulose starch interface increased the tensile properties of the bioplastics. In summary, the presence of natural cellulose micro-fiber in the matrix of the bioplastic enhanced the physical-mechanical properties of the starch-based bioplastics. The effect of the size of the filler and concentration used as reinforcement in bioplastics needs to be adequately investigated to understand how it affects the characterization and properties of the bioplastics.

Physical-Mechanical Behaviour Nanoparticles in Starch-Based Bioplastics

Polymer researchers have deployed nanotechnology in the enhancement of packaging material's properties from starch without compromising the biodegradability of the materials. For significant enhancement of the properties of bioplastics, low concentrations of nano-sized additives were blended with starch-based bioplastics to advance their mechanical, thermal, optical, and physicochemical properties [47]. Carbon nanotubes, nano-calcium carbonate and nano-silica are mostly used as nanofillers. In this technology, the polymer matrix of the bioplastic is reinforced with nano-dimensional particles as an alternative to the conventional micro-dimensional fillers earlier discussed. The large surface possessed by nanoparticles promotes better interfacial interactions with the polymer matrix [48]. The use of nanosized particles at a lower concentration between 1- 5% produces an additional homogeneous dispersion of the particles, increases the specific surface of reinforcement, and possesses better mechanical, physical, and thermal properties than the micro-sizes reinforcement [49-51].

Nano clay is an advanced biodegradable polymer with improved barrier and mechanical properties. The most used clay is montmorillonite (MMT), and the type of MMT-polymer interaction influences the behaviour of the nanocomposite material. Using the solvent casting method, Calambas et al. [47] produced films from achira starch/polyvinyl alcohol (PVA) reinforced with MMT nano clay at varying concentrations (0.5 - 5% w/v) and glycerol. The starch/polyvinyl alcohol to nanoclay ratio used in this study was 3:2. The percentage of elongation, Youngs' Modulus and tensile strength enhanced with sonicated nano clay of 0.5% w/v. the mechanical properties were worse in the 1.0 and 1.5% w/v when compared with films without nano clay. Nano clay concentration above 1.0% w/v saturated the polymer matrix thereby influencing the physicochemical properties of the nano bioplastic. It was concluded that only 0.5% w/v of nano clay can be successfully mixed with matrix starch/PVA for use as biodegradable packaging. Apart from the concentration of nano clay used, Chen and Evans [52] reported that the type of plasticized used also limits the application of bioplastic. The percentage elongation of the nanocomposites prepared from glycerol plasticized starch with varying content of sodium montmorillonite and hectorite increased from 47 to 57% while the tensile strength increased from 2.6 to 3.3 MP for 5% sodium montmorillonite. Sariadi and Raudah [48], reported that the increase in plasticizer content negatively affected the reinforcing influence of the clay on the starch, thereby causing an increment in the elongation percentage and a reduction in the tensile strength. The maximum tensile strength was observed at 0.6% concentration of nano clay and 25% glycerol. Park et al. [53] stated that the high levels of glycerol plasticizers limit the applications of bioplastics in certain areas.

Using inorganic fillers such as metals and metallic oxides, the thermal and mechanical properties of composite bioplastics from corn starch blended with titanium dioxide (TiO_2) nanoparticle was analysed by Amin et al. [54]. The addition of TiO_2 nanoparticles decreased the elongation of the bioplastic from 88 to 62 and enhanced the tensile strength from 3.55 to 3.95 MPa. The mechanical strength was enhanced but with less flexibility. From the TGA analysis, the composite bioplastic had higher thermal stability when compared with starch bioplastic, but the rate of biodegradability was higher for the nano starch bioplastic. The higher biodegradability of the nano-based bioplastic was due to the antimicrobial properties of TiO_2. This makes bioplastics suitable for packing in the pharmaceutical and food industries.

Titani and Haryanto [55], combined zinc oxide (ZnO) and titanium oxide (TiO_2) nanoparticles as fillers in the production of bioplastic from 40 grams of tapioca starch mixed with 10 g corn starch. The bioplastics produced without nanoparticles had the presence of air bubbles and the addition of TiO_2 and ZnO eliminated the air bubbles. The addition of nanoparticles also improved the mechanical properties and elongation of the bioplastic. The antimicrobial property of nanoparticles caused the nano-bioplastics to degrade longer when compared with bioplastics without nanoparticles. Rahman [56] investigated the effect of copper oxide (CuO) nanoparticle concentration on the elongation and tensile strength of potatoes-based bioplastic. The nanoparticles varied from 0.5 % to 4% and the highest tensile strength of 6.84 MPa was observed at 5% concentration whereas the tensile strength of 4.1 MPa was reported for bioplastic without nanoparticles. The tensile strength was below that of the starch sample when the nanoparticle content was increased beyond 1%. This observation was attributed to the agglomeration of CuO on the sites of CuO and the functional group (OH, CH, CO) interaction in the starch. Above 0.5% concentration, the elongation decreases as the nanoparticle's concentration increases. The elongation was lower than that of starch-based bioplastics above 2% concentration.

The increase in the concentration of inorganic nanoparticles reduced the elongation of the bioplastic. Therefore, to overcome this limitation, organic nanoparticles were used as reinforcement [57]. The starch nanoparticles are smaller in size and possess a large surface area which makes them suitable for use as fillers in the production of bioplastics. The organic nanofiller concentration, characteristics and filler-matrix interaction impact the properties and characterization of the bioplastics [58]. Starch nanoparticles were used as nano-fillers to produce a bioplastic film from rice using sorbitol [59]. The flexibility of the film reduced while the tensile strength increased as the content of rice nanoparticles increased. The tensile strength of the bioplastic without the rice nanoparticle fillers was reported as 7.12 MPa while the elongation at break had a value of 53.46%. The enhanced tensile strength of 12.86 MPa and flexibility of 2.48% were recorded when a 30% concentration of rice nanoparticles was added to the starch matrix. The effect of cellulose nanofiber (CNF) and cellulose nanocrystal (CNC) reinforcement on pumpkin starch (PS)-based composite film properties was studied by Zhang et al. [60]. The nanocomposite was prepared using the casting method with 1% or 2% concentration of CNC, 10% or 15% concentration of CNF and 30% glycerol. The agglomeration of the CNC at a higher concentration in the PS matrix led to a decrease in the degradation of CNC/PS as the concentration of cellulose nanocrystals increased. The thermal stability of cellulose nanofiber nanocomposites was better than that of cellulose nanocrystal nanocomposites while the tensile strength results showed that cellulose nanocrystal had better-reinforcing capacities than cellulose nanofiber. The high elastic modulus of cellulose nanocrystals is the reason for the enhancement. The maximum tensile strength of 30.32 MPa was obtained in nanocomposite films with 1% cellulose nanocrystal. The mechanical property of the nanocomposite was influenced by the functional and crystallinity (related to its apparent rigidity and modulus) groups of nanocellulose. Table 1 shows different bioplastics feedstock, additives, and their properties.

PROTEIN-BASED BIOPLASTICS

The increase in agricultural and food waste has advanced the production of protein-based bioplastics. Proteins are heteropolymeric chains containing different amino acids. Proteins occur in nature, and they are made up of a long chain of amino acids [61]. Protein-based biopolymers are extensively utilized as edible films because of their improved mechanical properties when compared with polysaccharides [62]. The bioplastics produced from protein possess antimicrobial and antifungal properties thereby preventing the contamination of food and drugs [63]. Covering food with protein-based bioplastics prevents the loss of moisture and flavour, thus, increasing the shelf life and controlling the exchange of gases [64]. These properties make protein-based bioplastics suitable for packaging in the food and drug industries.

Protein-Based Bioplastics Synthesis Methods

For the polymerization of protein to produce bioplastic, the primary amino acid structure of the protein serves as a repeat unit. The protein is denatured to break down the secondary bonds leaving the primary bond in the amino acid backbone [65]. In the denaturing process, force is applied to ensure that the plasticization process is complete, insufficient force can result in particle plasticization causing some protein to return to its coiled state [66]. The plastics produced after complete plasticization have similar properties to the protein used. Heat in the form of UV treatment can also be applied in the denaturing of protein [67]. Examples of protein used in the production of protein-based bioplastics are albumin, wheat gluten, whey, keratin, corn zein, bloodmeal, soy protein, keratin, collagen, animal protein, fish myofibrillar protein and gelatin [62,68]. The source of the protein is a major factor that affects the characteristics and properties of protein-based bioplastics. Albumin obtained from the egg white of a hen was used to produce bioplastic using glycerol as the plasticizer. The bioplastic obtained from albumin exhibited a better rheological property when compared with that obtained from wheat gluten plasticized with glycerol [24].

The common techniques used in the production of bioplastics from protein are injection moulding, compression moulding, casting, extrusion, and hot-press moulding as shown in Figure 4. The casting method involves the dissolution of the protein in a solvent with a plasticizer. The mixture is then poured into the mould with the desired cavity. The solution is allowed to dry with the solvent evaporating. The physical and mechanical of the bioplastic produced are dependent on the drying temperature. The tensile strength and Young's Modulus increased with the increase in drying temperature [69]. Protein-based bioplastic was prepared by Kayserilioğlu et al. [70] from cotton seed protein using the hot press moulding after denaturing the protein and formaldehyde, glyoxal or glutaraldehyde was used as the crosslinking agents. In the extrusion technique, shear stress and heat treatment were applied to the protein causing the protein to undergo complex association and dissociation. The characteristics and properties of the bioplastics are dependent on the screw speed, temperature gradient within the extruder, the type and concentration of additives and plasticizers used, the size and shape of the die which affects denaturing of the protein, and aggregation of proteins [71]. Alias and Ishak [72] prepared protein-based bioplastics from fish waste using the extrusion and compression moulding methods. The impact of heat treatment and processing technique on the thermos-mechanical properties of bioplastics produced from rice, albumen, wheat gluten, and albumen/gluten blends using glycerol as the plasticizer showed that the casting method provided bioplastics with higher thermosetting properties when compared with the thermos-mechanical method [24]. Injection modelling is the most popular process used in the production of protein-based bioplastics [73]. The piston injection moulding method was used to produce bioplastics from the dough-like blend of pea and glycerol [74]. With shorter mixing time, lower energy consumption and intermediate mixing speed of 30 rpm, the bioplastic produced had higher elongation and enhanced tensile properties along with better homogenization. Increasing the injection mould temperature improved the tensile stress and elongation, but there was no significant improvement in Young's modulus, and the water uptake capacity was hindered [75].

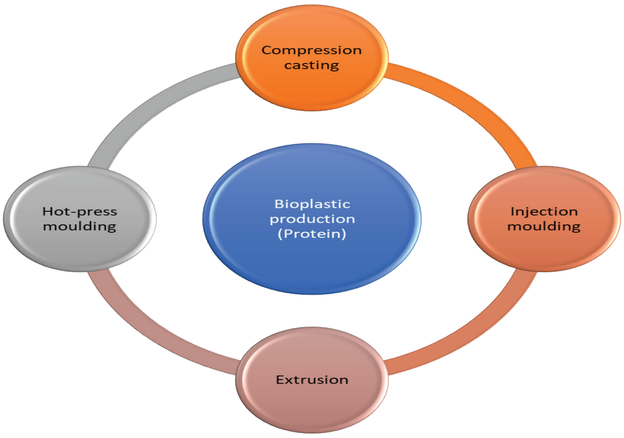

Figure 4. Bioplastic production technique from protein source.

Crosslinking is a chemical process of creating covalent bonds to link two or more polymers. Various crosslinking methods are used in the preparation of bioplastics from proteins. The three most common methods are physical crosslinking which uses physical agents, chemical crosslinking which uses chemical agents and enzymatic crosslinking which uses enzymes [76-78]. In the production of wheat gluten films, 1-Ethyl-3-(3-dimethaylaminopropyl) carbodiimide and N-hydrooxysuccinimide were used as cross-linking agents to improve water sensitivity and enhance the tensile strength of the wheat. Due to the innate toxicity of these crosslinking agents, they are not used in food packaging applications [79]. The characteristics of protein-based bioplastics such as antibacterial resistance, antioxidant properties, biodegradability, and mechanical properties make them suitable for use in the packing of food and drug delivery, but the low percentage of elongation causes them to break as they cannot reach the 150-500% elongation of polyethene or PVC plastics.

Physical-Mechanical Behaviour Protein-Based Bioplastics

Protein can be added to starch to overcome some of the challenges with starch-based bioplastics earlier discussed. Omrani-Fard et. al [80] added protein into potato flour to improve the starch-based bioplastics properties. The protein enhanced the tensile strength and improved the flexibility and elasticity of the starch-based bioplastics [23]. Albumen was introduced into corn/potato starch to further enhance the properties and application of bioplastics. The influence of thermo-mechanical, thermoplastic formation processes on the optical and mechanical properties of the albumen-starch samples containing 0-30 wt.% starch was investigated. The outcomes of the investigation were compared with the results obtained for wheat gluten-based bioplastics. The tensile strength and elongation at break of the albumen/starch bioplastic showed a monotonous and rapid decrease as the content of starch increased while the gluten-based bioplastic had a higher elongation at break, but the tensile strength was low. It was reported that the inclusion of starch granules in the protein matrix created heterogeneities in the matrix that operated as stress concentration points generating cracks and causing the values of elongation and strength to be lower. The work highlighted that certain starch is incompatible with protein and it was more pronounced for the corn starch and the thermo-mechanical properties of the bioplastics are mainly influenced by the protein matrix. The addition of starch to albumen bioplastics within the concentration and temperature investigated has no significant impact on the thermo-mechanical properties. The albumen/starch-based bioplastic properties were greatly affected by the processing method. To improve the compatibility of protein with starch, sodium bisulfite was used as a reducing agent along with glyoxal and L-cysteine as crossing linking agents to process the rice protein concentrate used in the development of rice protein-based bioplastics with high biodegradability and excellent thermo-mechanical properties. Thermoplastic and injection moulding techniques were used to develop rice protein-based bioplastics. The optimum glycerol concentration, mixing time and process conditions were obtained in this study from the calorimetric and rheological measurements. The specimen produced degraded within 21 days.

Using the hot-press moulding technique, the influence of crosslinking on the mechanical strength, water absorption resistance and thermal stability of cotton seed protein bioplastics using glycerol, aldehydes, and urea as plasticizers [70]. Due to the increase in free volume and reduction in structural integrity, an increase in the content of the glycerol in the cotton protein led to a decrease in the α-relaxation and denaturation temperature. At low temperatures, a higher value of storage modulus was reported at lower concentrations of glycerol. Perez-Puyana et al. [74] introduced heat treatment as an additional physical crosslinking to enhance the mechanical and functional properties of the bioplastics from pea protein. The heat treatment process enhanced the mechanical and elastic properties of pea protein while lowering the critical strain and water uptake capacity. The pea-based bioplastics exhibit excellent antimicrobial properties, but the heat treatment modified the protein structure, worsening the antimicrobial character. The antibacterial, viscoelastic, and thermal properties of bioplastics produced from whey, soy and albumin were investigated by [65] and rubber latex, glycerol, and water were used as the plasticizer. The thermal properties of the soy and whey bioplastics have similar degradation temperatures occurring between 50 and 60 °C and this was lower than that of albumin bioplastics. The albumin or whey films plasticized with glycerol had the best antibacterial properties while the albumin and whey showed similar viscoelastic properties. The soy bioplastics presented a larger range of properties based on plasticizers. Alias and Ishak [72] prepared protein-based bioplastics from sardine by-product, mackerel by-product and mackerel fillet powder investigating the effect of various plasticizers (polyethylene, triethylene and glycol) on the thermal stability and mechanical of the bioplastics. The incorporation of plasticizer increased the elongation at break but lowered thermal decomposition and tensile strength. The type of plasticizer does not significantly influence the mechanical properties, but the processing of the protein was affected by the type and concentration of plasticizers. Due to the large protein miscibility, high solubility in water and low molecular weight of glycerol, the bioplastics produced using glycerol had the highest enhancement in mechanical properties but the lowest decomposition temperature. Rezaiyan Attar [81] using glycerol and sorbitol plasticizers at varying concentrations studied the opacity, surface properties, and thermal and mechanical, of bioplastics produced from Vicia villosa protein isolate. As the plasticizer concentration increased, the tensile strength decreased concurrently with an increase in elongation at break and water vapour permeability. The film plasticized with sorbitol exhibited higher film solubility and tensile strength when compared with glycerol-plasticized films, but the water vapour permeability was low. The moisture content for sorbitol plasticized films was lower than those of glycerol.

The effect of heat treatment and processing techniques on the thermos-mechanical properties of bioplastics produced from wheat gluten, albumen, rice, and albumen/gluten blends using glycerol was studied [24]. Protein denaturing occurred during the thermos-mechanical treatment, and this led to an increase in the degree of crosslinking between the molecules. A deduction in the values of the linear viscoelasticity functions was observed in the protein/glycerol film and this was caused by the hygroscopic characteristics of the films. The blend of albumen and rice reduced the concentration of protein and thermosetting temperature giving rise to linear viscoelastic moduli values like that of LDPE and HDPE. Apart from the mechanical and thermal treatment, the concentration and characterization of the glycerol, the source of protein affected the characteristics and properties of the films. Ramos et al. [82] investigated the properties of whey protein concentrate and whey protein isolate with varying concentrations of glycerol. The Young's Modulus and tensile strength decreased as the concentration of glycerol increased making the bioplastic film weaker for whey protein concentrate and whey protein isolate. Evaluating the mechanical and oxygen barrier properties of the films, the whey protein films exhibited better properties when compared with other proteins from soy protein isolate, wheat gluten, and corn zein and polysaccharide-based films. However, the percentage elongation for the whey protein film is lower and water vapour permeability is higher than that of synthetic polymer films.

The continuous reliance on edible protein for bioplastic feedstock will put pressure on the protein food chain and compete with fresh water and arable land. To address this issue, films were produced from single-cell protein obtained from dry and dead unicellular microbial biomass, which are cultures of industrial and agricultural by-products using the compression moulding technique [83]. Using glycerol, the effect of compression temperature and time on the physical-mechanical properties of single-cell protein-based films and it was reported that the pressing temperature and time influenced the properties. The thermal stability and flexibility of the films were enhanced while the Youngs' Modulus and tensile strength increased with pressing temperature between 110 to 130 °C due to network strengthening and aggregate formation. As the pressing temperature increases, the properties become poor. The values of oxygen permeability of the bioplastics obtained were better than those of LDPE and HDPE.

LIPIDS-BASED BIOPLASTICS

Fatty acids derived from plant oil and animal fats are considered as a viable feedstock for the synthesis of bioplastic and a potential alternative to synthetic plastics. Lipids are usually gotten from animals, plants and insects that contain fatty acids glycerides, phospholipids, fatty alcohol, and terpenes, and they are hydrophobic materials that retards the movement of moisture within food due to their non-polar nature [84]. Films produced from lipids are usually edible, possess moisture-entrapment property and gives bioplastics a glossy appearance when used as a coating [62].

Lipids-Based Bioplastics Synthesis Methods

High-quality lipids need to be extracted from oil seeds and animal fats to produce bioplastics with excellent properties. Lipids can be extracted using the mechanical or chemical method. Using the chemical method, Kadioglu et al. [85] extracted oil from corn using surfactants instead of hexane. Lipids were extracted from lyophilized biomass by washing them with a methanol solvent mixture and chloroform in a ratio of 4 to 1 respectively [86]. To extract the lipids, the Soxhlet apparatus was used while the rotary vacuum evaporator was used to evaporate the solvent from the lipid samples. A hydraulic press is used to extract oil from seeds in the mechanical method.

Physical Mechanical Properties of Lipids-Based Bioplastics

Lipids are usually used as coatings in bioplastics, Chiumarelli and Hubinger [87] melted stearic acid and carnauba wax in cassava starch plasticized with glycerol. The film produced had good barrier properties and enhanced physical, mechanical and thermal properties.

The influence of sonication and oil content on the properties of Mesquite seed gum and palm fruit oil emulsion edible films was studied by Rodrigues et. al [88]. Bioplastics films were produced by blending the palm fruit oil nano-emulsion with the mesquite seed gum using glycerol as the plasticizer. The casting and evaporation techniques were used in producing the film. The authors reported that the water solubility, tensile strength, and water vapour permeability of the film decreased as the palm fruit oil increased. The sonication process improved the tensile property by further promoting a uniform droplet dispersion. Palm oil and epoxidized palm oil were incorporated into starch-based bioplastics to study the effect of the oils on the water resistance and mechanical properties of bioplastics. At a lower oil concentration of less than 3% wt, the tensile strength and elongation at break enhanced. The strengthened interactions between the hydroxyl groups of starch/glycerol and the carboxylic groups of fatty acids in the oils were the reasons behind the enhancement. The formation of discontinuous zones was reported at higher concentrations because of the phase separation that occurs between the oil and the starch. The interaction between the oils showed lower bonding when compared with the starch and oil interaction. This interaction affects the mechanical properties of the bioplastic. The presence of epoxy groups in the epoxidized palm oil improved its compatibility with the starch matrix while the compatibility with the palm oil is lower. The authors recorded a little reduction in water uptake and solubility for both oils when compared with starch-based bioplastics [89]. There are limited resources in the literature on the production of bioplastics from lipids. More research should be conducted on the use of lipids and blending them with starch or protein since they possess reduced water uptake.

Table 1 gives the summary of different bioplastic's sources, additives and properties for polysaccharide, protein, and lipid.

OPPORTUNITIES AND CHALLENGES OF BIOPLASTIC FROM BIOMASS SOURCES

The bioplastic production techniques are still evolving with each type of bioplastic from different biomass feedstocks having different production techniques and exclusive properties. The starch-based bioplastic with micro-sized filler and nanoparticles has great potential, but its commercial use is still limited. First-generation feedstocks that are edible are mostly used in the production of bioplastics. Hence, this has contributed to the high cost of production of bioplastics when compared with the cost of synthetic plastics production. The use of bioplastics from biomass must be sustainable without creating food insecurity or competing with available arable lands. Researchers need to source alternative feedstocks that are nonedible second and third generation feedstocks as shown in Figure 5. Legislative and financial incentives should be given to companies and individuals who manufacture, trade, and advocate the use of bioplastics.

According to Zimmermann et al. [92] some existing bioplastics, especially starch- and cellulose-based plastics contain some level of toxins like those in synthetic plastics. These toxins are often from some of the chemicals used as additives or the production process employed in the production of bioplastics. Bioplastics' production cost is higher than those of synthetic plastics, therefore continuous innovative research is necessary to reduce production costs and replace

Table 1. Summary of bioplastic feedstock, additives, and properties

Biomass source	Additives	Bioplastic properties	References
Corn starch	Glycerol water	Good tensile strength Less dying time	[8]
Corn starch and rice starch	Water Glycerol citric acid gelatine	Improved tensile strength. Citric acid enhanced adhesion between the glycerol, water, and starch	[20]
Cassava peel starch	microcrystalline cellulose, sorbitol	MCC improved the tensile strength	[21]
Achira starch	Polyvinyl alcohol MMT nanoclay Glycerol	Enhanced percentage of elongation, Youngs' Modulus, and tensile strength at lower MMT nano clay concentration	[47]
Corn starch	titanium dioxide	TiO_2 nanoparticles decreased the elongation and enhanced the tensile strength.	[54]
Tapioca starch	combined zinc oxide (ZnO) and titanium oxide (TiO_2) nanoparticles	Improved mechanical properties and elongation of the bioplastic. Antimicrobial property of nanoparticles caused the nano-bioplastics to degrade longer.	[55]
Whey protein	Glycerol	Lower percentage elongation Higher water permeability Tensile strength decreased with concentration of glycerol	[82]
Orange by product	glycerol	High water vapour permeability Poor overall tensile property	[90]
Cassava starch	Glycerol Palm oil Epoxidized palm oil	The addition of oil slightly reduced the water uptake and increased the tensile strength.	[89]
Rice straw	Trifluoroacetic acid	Enhanced tensile strength and elongation. Higher water absorption	[91]

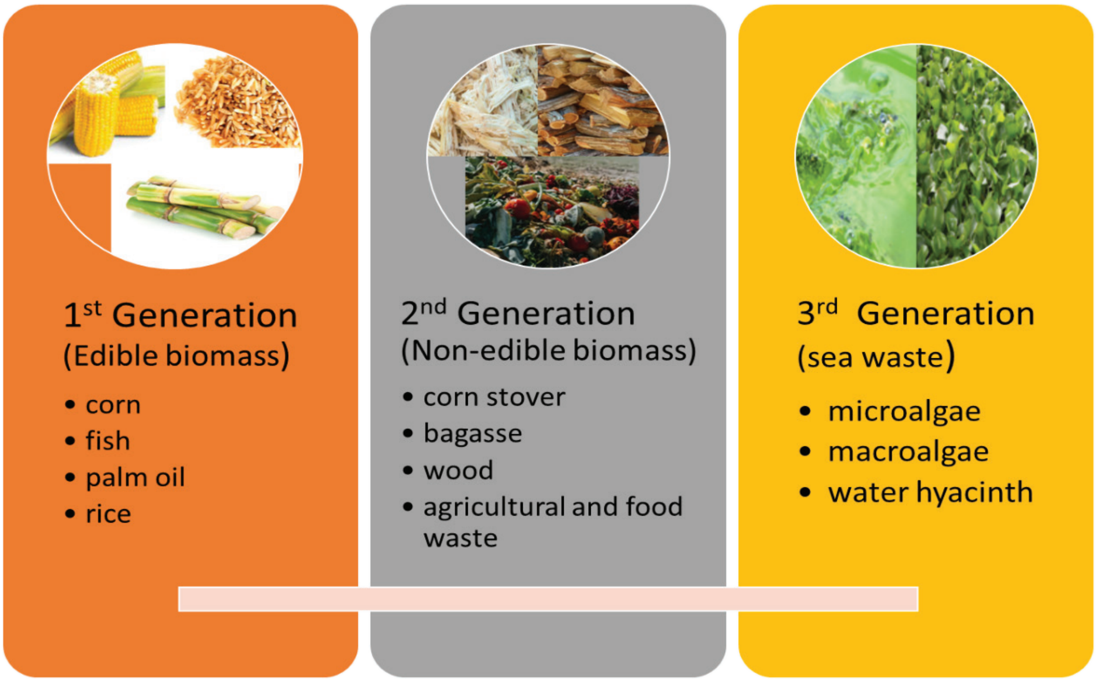

Figure 5. Biomass generational sources for bioplastic production.

toxic additives with natural additives that are safe for human consumption. The flexibility and elongation of bioplastics from starch were reported to be low by several researchers and this limitation is hindering the application of bioplastic as packages in grocery and other retail stores. The flexibility of bioplastics can be enhanced through further modification of the fillers or production process. Jayathilaka et al. [58] stated that there are limited resources that investigate the optimal concentration of nanoparticles that can be used as reinforcement in bioplastics for optimal characteristics and properties. Therefore, the influence of micro and nano filler's size and concentration needs to be further investigated to understand how the size and concentration of fillers concerning dispersion and homogeneity affect the characterization and optical properties of bioplastics.

Bioplastics are prone to thermal degradation, and they are likely to degrade when subjected to elevated temperatures. The biodegradation of bioplastics by biological agents is strongly dependent on certain environmental conditions, which differ from one location to another. These environmental conditions need to be met to fully take advantage of the biodegradability of bioplastics. For sustainable waste management of bioplastic waste, the biodegradability of bioplastics from different feedstocks, and compositions using various additives/plasticizers needs to be further investigated considering that the rate of degradation is dependent on the environmental conditions and microorganism activities.

AUTHORSHIP CONTRIBUTIONS

Authors equally contributed to this work.

DATA AVAILABILITY STATEMENT

The authors confirm that the data that supports the findings of this study are available within the article. Raw data that support the finding of this study are available from the corresponding author, upon reasonable request.

CONFLICT OF INTEREST

The author declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

ETHICS

There are no ethical issues with the publication of this manuscript.

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