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The Potential of Microalgae in Chitosan and Cellulose as Sustainable Materials for Edible Bioplastic Applications: A Review

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ABSTRACT

The demand for eco-friendly food packaging options with additional features to prolong shelf life has continuously increased since 1940. Bioplastic is gaining popularity as a viable replacement for plastics based on fossil fuels due to fluctuating oil prices. Therefore, technological innovation is required to resolve the issue. This study aimed to review the distinct characteristics of integrating chitosan and cellulose alongside exploring the capabilities of microalgae in bioplastic production. The potential techniques and applications for future development were also suggested. The unique growth yield of microalgae makes them a compelling option for producing bioplastics. Hence, utilizing microalgae for bioplastic production offers a significant opportunity to enhance moisture barrier capacity, alter the structural properties, and adjust the flow behaviour. Moreover, these materials can also serve as the key nanocomposites components in the food packaging industry. The potential for chitosan/cellulose/microalgae-based bioplastic and the key themes and obstacles for future research into these composites for bioplastic production were also reviewed.

1. Introduction

Plastic packaging is a critical tool in order to preserve the shelf life of packaged food products. Currently, non-biodegradable plastics are often used as packaging materials because of their low cost, ease of processing, and strong resilience to mechanical and chemical stress. Conventional plastics such as polypropylene, polyvinyl, and polystyrene exhibit strong mechanical and barrier properties, including tensile strength and flexibility, carbon dioxide and oxygen permeability, and odour transmission. However, because of their reduced efficiency for recycling, reliance on non-renewable resources, and restriction of biodegradability, these polymers pose significant environmental risks and cause major ecological issues [1,2]. Due to these restrictions, research has been concentrated on producing packaging materials from renewable resources in the past several years, and biopolymers have drawn a lot of interest in this area. Biopolymers used in packaging are

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generally safe since they do not include any toxic or dangerous compounds such as heavy metals or organic solvents for example toluene or chloroform. Additionally, because they have ability to decompose naturally, they release harmless byproducts into the soil. The discovery and application of biopolymer-based packaging materials to enhance the shelf life of fresh goods have drawn more attention throughout the past 20 years [3]. Nevertheless, the scientific community have been investigating the potential applications of biopolymer-based films for more than a century [4].

The edible films have been employed to enhance the gas and moisture barriers and preserve the product from harmful chemical, mechanical, and microbiological contamination. Additionally, they can improve sensory function and increase the shelf-life of foods, particularly perishable ones such as seafood [5]. This edible food packaging also has the advantage of being environmentally friendly because it is made of renewable resources. Based on the sources, biopolymers can be broadly classified into three classes: those derived from plant or animal biomass, such as protein and polysaccharides, that are directly extracted, polymers synthesized from microorganisms, and polymers formed chemically by using renewable monomers as precursors such as polylactic acid (PLA) [6]. Guar gum, for instance, is an annual legume plant called Cyamopsis tetragonoloba, a heteropolysaccharide with a mannose backbone [7]. The production of PLA from corn has been extensively studied for application in food packaging because PLA has excellent physical qualities, including as high strength, processability, thermoplasticity, and non-toxicity. Although biopolymers are abundant in nature, their application in edible food packaging is restricted because of their poor mechanical and barrier characteristics in comparison to conventional petrochemical packaging plastics. To address these issues, several physical and chemical techniques, including heat treatment, chemical modification, gamma irradiation, and the addition of different additives such as plasticizers and nanofillers have been proposed in the past in a way to alleviate the shortcomings of biopolymer-based packaging [8,9].

Protein interactions may readily cross-link via heat treatment to produce strong intermolecular covalent networks, compact molecular packaging, and decrease polymer mobility. A previous study by Xu *et al.,* [10] indicated the morphology structure of biofilm improved the tensile strength and water resistance by using soy protein, poly(vinyl alcohol) (PVA) and triglycidylamine (TGA) as crosslinkers in the formulation. The properties of soy protein and PVA enhance the toughness of the film by the interaction of hydrogen bonds in the matrices. Chemical modification of the biomolecules can potentially increase the functional qualities of biofilms. The mechanical tests indicated that the addition of extracted silk firoin and magnesium oxides to the cross-linked terephthaloyl thiourea-chitosan hydrogel might have improved the strength from 65.42 to 649.56 MPa, which met the conventional plastic standard of 24-302 MPa [11]. According to Saurabh *et al.,* [12], physical treatment such as subjecting biopolymers to radiation exposure, can induce the ordering of the polymeric chain which leads to enhanced mechanical strength.

Biodegradable films consist of the following based on their constituents: hydrocolloids, lipids, and composites. Hydrocolloids are hydrophilic polymers that can be synthetic or natural due to the presence of several hydroxyl groups. They are particularly good at producing films since they often gel texture in water, for example, proteins and polysaccharides [13]. Lipids such as waxes, essential oils, and fatty acids are molecules that are categorized as hydrophobic components [14]. Composites are materials composed of two or more constituents (lipid and/or hydrocolloids) that together have better properties than one part alone. Proteins such as casein, gelatin, wheat gluten and polysaccharides (starch, chitosan, and alginate) are frequently utilized either separately or in combination for the production of edible films [15]. Several species of microalgae such as Chlorella sp., Spirulina sp., Nannochloropsis oculate, and Nostoc sp., are capable of producing high-value functional chemicals such as pigments, carbohydrates, protein, lipids, vitamins, and minerals.

Microalgae are already served as food additives because of their biomass contains active components with anti-inflammatory, antioxidant, and coloring characteristics for example omega 6 fatty acids and pigments. Because of their simplicity in development, minimal feed and growth environment needs, and rapid growth capability, these microalgae are ideally suited for bioplastic production. Moreover, bioplastics provide superior mechanical properties such as tensile strength compared to materials sourced elsewhere.

A previous study by Deshmukh *et al.,* [16] reported that the combination of chitosan and Chlorella biomass showed improvement in terms of tensile strength, decreased water vapor permeability, and lower moisture content compared to chitosan film. To the best of the authors' knowledge, the literature on the combination between chitosan/cellulose/microalgae in bioplastics remains limited within the broader field of microalgae studies. Most articles published have focused on the use of agricultural waste and food waste for bioplastics [17-19]. The purpose of this review is to intrigue further interest in microalgae in chitosan-cellulose-based bioplastics by offering a topical overview of cutting-edge methods into bioplastics. Based on the overview, this state-of-the-art application of chitosan, cellulose, and microalgae in the development of food packaging is discussed. A particular focus is placed on the properties of films in terms of the impact of the additives on the mechanical, moisture barrier, and performance properties of the biopolymer's films will be highlighted. Lastly, their future challenges will also be reviewed.

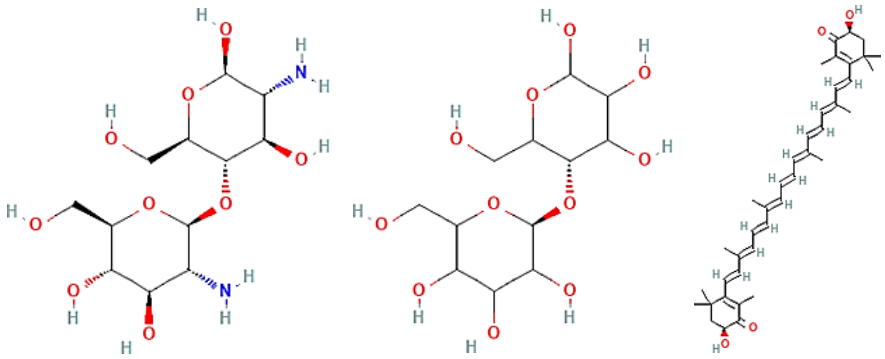
2. Edible Packaging Materials

Edible packaging should consist of at least two compositions: a biopolymers-based matrix that can form a cohesive structure and typically water as the solvent. Biopolymers are derived from biomasses such as polysaccharides, proteins, and lipids. The most promising biopolymers among these biomaterials appear to be chitosan and cellulose since they are biobased and biodegradable. Furthermore, they are some of the most common natural polymers and can be sourced from biowastes like corn, sugarcane, and seeds, promoting the principles of the circular economy.

2.1 Chitosan

Chitosan is a linear polymer synthesized for economical use through alkali deacetylation of chitin. It is often a copolymer of N-acetyl-D-glucosamine and D-glucosamine. Chitosan is mainly extracted from crustaceans, fungi, and algae waste using acid and alkali treatments. Temperature, incubation duration, alkali concentration, and determination of the final properties of the polymer produced are among the parameters that influence chitosan's deacetylation [20]. Because of its biological activity, chitosan and its derivatives have become extremely valuable in a variety of industries, including medicines, food, agriculture, cosmetics, and packaging. Chitosan offers a distinctive cationic composition that confers potent and broad-spectrum antibacterial capabilities compared to other neutral or negatively charged polysaccharides (Figure 1(a)). It has been shown that chitosan is stable, biodegradable, and biocompatible. It is soluble in weak acidic solvents such as formic, acetic, and diluted hydrochloric acid [21]. Numerous hypotheses have been proposed to explain the antibacterial activity of chitosan. Still, the most plausible one involves electrostatic interactions between the negatively charged surface of bacteria and the positively charged amino acid groups, NH^{3+} at a pH level below 6.3 (p*K*a of chitosan). This electrostatic interaction causes two types of interference: (i) it alters the permeability of the membrane wall, exacerbating osmotic imbalances within and hindering the proliferation of microorganisms, and (ii) it disrupts peptidoglycans structure in bacteria cell walls, leading to the leaking of the internal electrolytes such as potassium ion, as well as other

low molecular weight components including nucleic acid, protein, cellulose, and glucose. This theory was supported by recent study by Ardean *et al.,* [22] which showed that releasing soluble protonated glucosamine fractions caused the antibacterial activity in chitosan. However, chitosan has poor solubility in water at neutral pH, which can restrict its applications in certain contexts. Additionally, its mechanical properties may not be as robust as those of synthetic polymers (polyethylene, polystyrene, and nylon), limiting its use in high-stress environments.



(a) (b) (c)

Fig. 1. Molecular structure (a) Chitosan (b) Cellulose (c) Microalgae

2.1.1. Chitosan-based edible packaging

As reported in previous studies [23], chitosan-based biofilm exhibits superior mechanical properties and selective gas permeability for oxygen and carbon dioxide. However, the uses of chitosan films were limited due to their high-water vapor permeability. In the past, chitosan and other polymers were explored to address inadequate barrier properties. Cassava/chitosan-based composites showed better flexibility and water permeability than films made with any of the biopolymers. A pseudoplastic behavior was demonstrated by chitosan film and chitosan/starch film-forming suspensions. The water permeability of cassava film, chitosan film, and cassava/chitosan were 0.27 ± 0.04 g mm $m^{-2}h^{-1}kPa^{-1}$, 0.06 ± 0.007 g mm $m^{-2}h^{-1}kPa^{-1}$, and 0.17 ± 0.01 g mm $m^{-2}h^{-1}kPa^{-1}$, respectively [24]. The improvement in barrier characteristics can attributed to the interactions between the hydroxyl group of starch and amino acids groups found in chitosan.

In another study [25], the monolayer crosslinked edible films were created by merging the antibacterial properties of chitosan with alginate emulsions, incorporating olive oil to serve as a hydrophobic barrier. The addition of other polysaccharides, such as starch and alginate, provided composite films that were water-soluble and sealable, simplifying the product wrapping process. Moreover, chitosan endowed the film with antibacterial properties, thereby increasing the shelf-life of packaged products. Efforts have been undertaken to enhance the chitosan film's physical properties, particularly in their mechanical strength, water permeability and water solubility by intermolecular interaction with crosslinking. Lu *et al.,* [26] investigated the novel composite film made from chitosan and bacteria cellulose, enhanced with varying concentrations of curcumin, aiming to extend the shelf-life of fruits. The results showed that 0.5% of curcumin exhibits excellent mechanical, moisture and hydrophobicity properties. The interaction of chitosan with nanocellulose can be used as active packaging for meat wrapping.

A study by Costa *et al.,* [27] demonstrated that a combination of 10 and 25 wt% cellulose nanocrystal (CNC) gave the highest antimicrobial activity with a tensile strength of 13 and 25.3 MPa, respectively. Chitosan films exhibited increased strength and enhanced resistance to stress, likely due to the strong hydrogen bond interaction between the nanocellulose and the polymer matrix.

Similar study has been reported with [28], indicated that nanocellulose extracted from pea pods enhance the tortuosity in the chitosan polymer matrix, leading to slower water vapor diffusion. Bioplastic films with an antimicrobial agent can be used in food packaging to minimize microbial loads, hence increasing the shelf life of foods. However, because chitosan-based films are naturally antibacterial, they extend the shelf-life of packaged products that do not include antimicrobial agents. In terms of food packaging applications, the US Food and Drug Administration (FDA) categorized chitosan as generally regarded as safe in 2001 and various research has been conducted as an alternative for chitosan film in the food packaging systems. Table 1 shows the summary of chitosan combined with other additives/fillers to produce the edible packaging.

Table 1
Summary of biopolymers for chitosan edible packaging in terms of mechanical and barrier properties

Biopolymers	Filler/additives	Plasticizer	Tensile strength (MPa)	Percentages of elongation (%)	Water vapor permeability (g mm m^{-2} h^{-2} kPa^{-1}	References
Chitosan	Cassava starch		3.2 ± 0.1	146 ± 9	0.17 ± 0.01	[24]
Chitosan	Bacteria cellulose/curcumin	-	86.31	40.68-65.67	19.08 ± 0.42-10.08 ± 0.23	[26]
Chitosan	Corn starch	Glycerol	5.3 ± 0.8	1.8 ± 7	0.19 ± 0.01	[24]
Chitosan	Nanocellulose	Glycerol	8.93-25.3	58.9	4.1 ± 0.45-5.6 ± 0.51	[27]
Chitosan	Crystalline nanocellulose/ Alginate	Glycerol	31 ± 2-43 ± 4	28 ± 4-55 ± 6	10.1-1.39	[25]
Chitosan	Cellulose nanocrystalline (Pea pods)	-	52.2-73.4	11.2-11.4	5.4×10^{-5}	[28]

2.2 Cellulose

Cellulose is the most common and sustainable biopolymer in the biosphere. The global market for it reached a value of USD 364 million in 2021 and is forecast to increase to USD 963 million by 2026 [29]. Cellulose is made of a liner chain of $\beta(1\rightarrow4)$ linked D-glucose molecules. A variety of cellulose derivatives are utilized to produce films with superior characteristics as compared to native cellulose films [30]. The method of isolation employed determines the structure and characteristics of the native which impacts the total number of hydrogen bonds, the length and distribution of the chain, the crystallinity, and the distribution of the functional groups within repeating units along the polymer chains [31] (Figure 1(b)). Particularly, the hydrogen bonding configurations inside and between cellulose fibers are thought to be the primary contributor of the materials in the physical and chemical characteristics. In recent years, cellulose has emerged as a novel nanomaterial that can serve as a reinforcement agent [32]. These celluloses are gaining popularity among scientists because of their exceptional properties, which include nanometric size, non-toxicity, high crystallinity, high surface area, and biodegradability. With the rising interest in nanomaterials, cellulose has also been manufactured with nanoscale sizes.

Subsequently, various forms of nanocellulose have aroused tremendous interest in a wide variety of uses, including cellulose nanocrystals (CNC), cellulose nanofibrils (CNF), and bacterial nanocellulose (BNF) [33]. Technically, CNFs are produced by mechanical homogenization of native cellulose that has already undergone chemical and enzyme treatment. Conversely, cellulose may be treated with strong acid to produce cellulose nanocrystals, which can hydrolyze the amorphous

portion of the fibres and leave only the crystalline portion of the original fibers. Lastly, BNF is devoid of hemicellulose and lignin since it is derived only from bacterial metabolism. This lowers the expenses associated with purification and the harm from chemical uses that which causes to the environment. One limitation of cellulose as a material is its relatively poor water resistance compared to synthetic materials. Additionally, cellulose can be prone to degradation when exposed to certain environmental conditions, limiting its durability in some applications. Furthermore, the processing of cellulose into usable materials can be energy intensive and may involve the use of chemicals that are harmful to the environment.

2.2.1 Cellulose-based edible packaging

Naturally, the various procedures may be made more efficient based on the specifications for the final chemical structure and the properties of the source material. Owing to the large number of hydroxyl groups on the fiber surface, nanocellulose is easily modifiable just like cellulose. It enables the production of a wide range of materials with changeable surface properties that are appropriate for several specialized purposes [34]. Cellulose inclusion into biopolymeric materials permits the fabrication of high-performance materials without affecting their biodegradability while increasing their mechanical resistance and moisture barrier properties. Nanocellulose-based films are commonly employed in food packaging because of their excellent oxygen and carbon dioxide barrier properties [35]. Extensive research has been conducted on chitosan-cellulose composite films. Cellulose and chitosan produce homogenous composite films, resulting in outstanding functional characteristics because of their similarity in fundamental structures [36]. Nanocellulose obtained from microcrystalline cellulose can be used as a reinforcing agent in the biofilm with the incorporation of agar [37].

The effect of mechanical and water vapor properties has been significantly enhanced with the addition of nanocellulose reinforcement (up to 3 wt%) with 52.8 MPa and 0.97 g mm m-2 h-2 kPa-1, respectively. This improvement is attributed to the strong interaction between the homogeneously dispersed nanosized nanocellulose, which has a high surface area. Salim *et al.,* [38] reported that 2% of chitosan in cellulose nanocrystals isolated from garlic skin waste improved the mechanical, optical, and antibacterial properties of chitosan matrix. Furthermore, incorporating chitosan and a concentration of 40% cellulose nanocrystal inhibited the growth of Gram positive and negative bacteria. A previous study by Yanti *et al.,* [39] demonstrated the bacterial cellulose based on sago liquid waste combined with carboxymethyl cellulose on the development of food packaging. The result showed the optimal mechanical strength by adding 1% CMC and 1% glycerol with tensile strength and percentages of elongation of 17.47 MPa and 25.60%, respectively as mentioned in Table 2.

Moreover, the use of bacterial cellulose in this film as a meat sausage packaging was found to sustain the quality of the sausage for up to 6 days at ambient temperature. A similar study has been reported by using bacterial cellulose with the incorporation of polyvinyl pyrrolidone carboxymethyl cellulose (PVC-CMC) and guar gum stated the biofilm shows improvement in moisture barrier, optical, mechanical strength and hydrophobic properties. Thus, with this combination, the films can withstand more strength before breaking, making it suitable for food packaging with optimum stabilization efficacy [40]. In other studies, the evaluation of the physical and mechanical performance of cellulose nanofibrils from Euterpe oleracea Mart. with chitosan reduced the water solubility (28.11% to 17.91%) and water vapor permeability (75.20% to 51.93%) which advantage over other packaging applications [41]. Such films were shown to be helpful in increasing the shelf-life of packaged bioplastics by maintaining an optimal environment inside the packaging. Although

the cellulose has great film characteristics, its high cost makes it impractical for commercial usage in the production of the biodegradable films.

Table 2
Summary of biopolymers for cellulose edible packaging in terms of mechanical and barrier properties

Biopolymers	Filler/ additives	Plasticizer	Tensile strength (MPa)	Percentages of elongation (%)	Water vapor permeability (g mm m^{-2} h^{-2} kPa^{-1}	References
Cellulose nanocrystalline	Agar	Glycerol	48.7 ± 2.1-38.4 ± 2.6	15.4 ± 2.3-17.8 ± 2.5	1.18 ± 0.16-1.32 ± 0.07	[37]
Cellulose nanocrystalline (garlic skin)	Chitosan	-	36.75-63.75	12.58-10.34	-	[38]
Cellulose nanofibrils (Euterpe oleracea mart.)	Chitosan	Glycerol	8.18 ± 1.77-9.73 ± 1.14	0.02-0.01 $mm.mm^{-1}$	47.30 ± 0.87-44.48 ± 0.14	[41]
Bacterial cellulose	Guargum/ Carboxymethyl cellulose/ Polyvinyl pyrrolidone	Glycerol	25.9 ± 1.1	21 ± 3	0.7 ± 0.56	[40]
Bacterial cellulose	Carboxymethyl cellulose	Glycerol	7.76 ± 10.82-17.47 ± 1.17	21.82 ± 1.08-29.67 ± 1.04	-	[39]

3. Integration of Microalgae in the Polymer Matrix

The industrial of microalgae has been well-established and bioproducts derived from microalgae are available on the market as dietary supplements, cosmetics, as well as feed for aquaculture. To be economically feasible, it is better to utilize a sustainable biorefinery model composed of the continued use of the remaining biomass, which is still rich in high-value compounds after the recovery of the core product. Microalgae have been studied as potential bioplastic packaging materials [42], [43]. Microalgae are typically unicellular aquatic microorganisms that may be grown in a variety of environments, including freshwater, wastewater, and saltwater due to their remarkable tolerance to environmental stress. The growth rates, biomass output, and nutritional value of microalgae in terms of pigments, protein, carbohydrate, lipid, and fatty acid production may all be impacted by varying growing conditions [44]. The primary variables that might affect the growth of microalgae are nutrient content, followed by temperature, light intensity, salinity, pH, and carbon dioxide concentration [45]. The limitation of microalgae as a material is the harvesting and processing of microalgae can be energy intensive and costly, posing challenges for economical and sustainable utilization. Furthermore, ensuring consistent quality and purity of microalgae biomass may be difficult due to variations in environmental conditions and contamination risks.

Figure 2 shows an overview step processing in the microalgae which has a significant impact on the overall production of biomass and bioproducts. Microalgae constitute an excellent feedstock for the production of bioplastic because of their high photosynthetic conversion efficiency and capacity to grow at the rate that is acceptable in nonarable terrain. On the other hand, most of the species have been investigated for the synthesis of bioplastic, primarily using polyhydroxyalkanoates (PHA), which are naturally obtained by microalgae [46]. There is less information available about the use of microalgae in edible biofilms compared to seaweed/carrageenan-based edible packaging. Edible

packaging was formed by combining microalgae with other biopolymers. This formulation might be accomplished by using the whole cell of microalgae by extracting the bioactive components such as proteins and lipids from microalgae strains such as Spirulina platensis or Chlorella vulgaris [47]. Figure 1(c) shows the molecular structure of microalgae which offered a good hydroxyl group to form a compact structure with other biopolymers such as chitosan and cellulose.

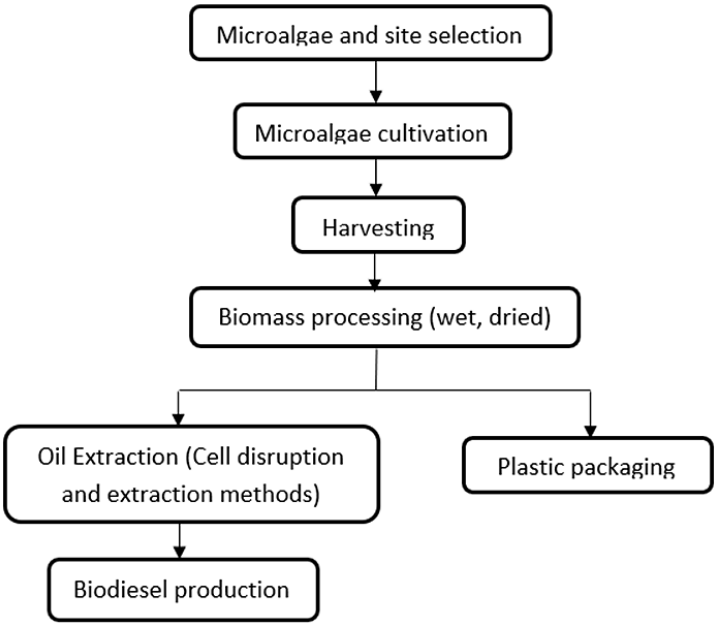


Fig. 2. An overview process of the sustainable product from microalgae

The most popular method for enhancing and altering the physical polymer characteristics is blending. A homogenous phase of the polymers is achieved by mixing multiple polymers together. Bioplastic-based microalgae and other biomass feedstock have been documented in a number of research. Despite this, starch, glycerol, and wheat-gluten are frequently needed as seen in Table 3. A comparative study was conducted by Fabra *et al.,* [48] to examine the impact of three strains of microalgae- Spirulina, N. gaditana, and Scenedesmus on the physicochemical and structural characteristics of thermoplastic starch biocomposite films. N. gaditana proved to be the most successful of the three strains of microalgae in producing biodegradable films based on corn starch by reducing the stiffness of the corn starch matrix. N. gaditana improved the oxygen and water vapor permeability of the polymer matrix meanwhile Spirulina and Scendesmus only increased water vapor permeability when added to the biocomposite. The addition of starch to Ulva armoricana sp. and polyvinyl alcohol has resulted in a 40% decrease in the percentage of polyvinyl alcohol which improved the compact of the blended solution [49].

Ciapponi *et al.,* [50] investigated the bioplastic composites by using wheat gluten and microalgae (A. platensis). Three different plasticizers and their impact on the characteristics of the biocomposite were examined. The mechanical properties of the biocomposite were improved by the presence of microalgae as a filler. The wheat gluten/A. platensis blending plasticized with 1,4-butanediol showed an increase in tensile strength with 3.3 MPa to 4.9 MPa meanwhile Young's modulus was 36.5 MPa to 273.1 MPa compared to the native wheat gluten, respectively. There have been attempts to use the chitosan found in microalgae as a source of bioplastic, in addition to combining them with plasticizers. For example, chitosan, glycerol, and defatted Chlorella biomass were characterized and the results showed an increasing value from 5 to 25 wt. % in tensile strength. In addition, increasing

the content of biomass resulted in a decreased in light transmittance and reduced the water vapor permeability by more than 60% [16].

Table 3
Summary of biopolymers for microalgae edible packaging in terms of mechanical and barrier properties

Microalgae	Filler/additives	Plasticizer	Tensile strength (MPa)	Percentages of elongation (%)	Water vapor permeability (g mm m^{-2} h^{-2} kPa^{-1}	References
N. gaditana	Corn starch	Glycerol	11.7 ± 1.2	2.1 ± 0.3	1.68 ± 0.16	[48]
Spirulina	Corn starch	Glycerol	13.5 ± 1.1	1.2 ± 0.1	1.66 ± 0.15	[48]
Scenedesmus	Corn starch	Glycerol	10.8 ± 1.2	0.9 ± 0.1	1.58 ± 0.24	[48]
Spirulina platensis	Wheat gluten	1,4-butanediol	4.2 ± 0.6-4.9 ± 0.9	>22.2 ± 7.8-82.1 ± 10.5	20.3	[50]
Ulva sp.	Plantago ovata seed	Polyethylene glycol	44.58 ± 2.1-46.62 ± 2.4	148.7-203.7	-	[49]
Chlorella	Chitosan	Glycerol	19.6-67.4	<40	<2.6	[16]

4. Edible Film-Forming Methods

Edible films are utilized as packaging materials to mitigate the environmental impact of plastic petroleum-based. Additionally, there is a growing trend in consumer desire for environmentally friendly and renewable packaging materials. It is important to comprehend the chemical characteristics of the biopolymers and additives that make films and to modify them accordingly when producing films for particular uses. There are two distinct methods for obtaining edible films from edible materials: solvent-casting and extrusion methods, which are also known as wet and dry processes, respectively.

4.1 Solvent-Casting

Edible film production might be achieved by using the same technology used for conventional polymers. The two primary techniques that have been employed are the wet process (solvent-casting) and dry process (extrusion). Four steps are involved in solvent-casting methods: (i) production of a film-forming solution by dissolving biopolymers in a suitable solvent such as acetic acid, ethanol, and water; (ii) adding bioactive components or additives agents and plasticizers; (iii) pouring the solution into molds (trays, acrylic, or silicon); (iv) finally the cast solution was dried at room temperature or under controlled conditions using the microwave or hot air oven to facilitate the simple removal of solvents and peel off the film as illustrated in Figure 3. The process of air-drying edible film is a crucial step in enhancing the intramolecular interaction between the polymer chain and producing the excellent morphology of biofilm [51]. The structural and physical characteristics of the film have been negatively impacted by the quick-drying technologies used during the casting process. The effectiveness of air-drying and drying temperatures in the production of edible film has been the subject of several research [52-54]. The produced edible film needs to be uniform and devoid of errors (mechanical effect or non-consistency). The most crucial aspects of edible films are their thickness, mechanical, transparency, moisture content, thermal stability, oxygen, and water vapour permeability as well as the biological properties [55,56]. Cohesive biopolymer matrix chains and plasticizers are employed to produce an excellent edible film with high mechanical strength,

moisture, thermal stability, and morphology structure properties [57]. As a result, edible film may have commercial applications for controlling postharvest losses in vegetables and fruits and reduce microbial contaminations in products [58].

The major advantage of the solvent casting method of biofilm is its flexibility of manufacture without the need for specified equipment and inexpensive cost. This wet procedure improves the particle contact between materials, making the particle more homogenous and resulting in fewer shortcomings [54]. Lower temperatures during processing phases might also be advantageous, as most food-processing materials cannot be moulded at higher temperatures without causing an irreversible structural change in the morphology of materials. The key disadvantages of using solvent-casting methods are: (i) the shape of the film is restricted (usually only simple sheets and tubes can appear; (ii) there is a chance that toxic solvent will be trapped inside the polymer; (iii) denaturation of proteins and other molecules introduced into the polymers by using solvents or high temperature occur; films can be vacuum-dried to eliminate the toxic solvent; (iv) limits on the quantity of the film can be produce; (v) casting required a long period of drying, which is impractical in commercial production; and (vi) converting film production from lab-scale to pilot scale presents the most problems since a variety of factors such as interaction of speed and temperature can affect the quality and inhibit continuous advancement for commercial scales [54]. The limitation of solvent casting in bioplastic preparation by involving microalgae, chitosan, and cellulose lies in the choice of solvent is critical in ensuring compatibility with the biopolymer components (microalgae, chitosan, cellulose) and achieving desired material properties. Some solvents may not effectively dissolve all components or may cause undesirable interactions, leading to compromised bioplastic quality or performance. Therefore, selecting appropriate solvents and optimizing casting parameters are crucial steps in mitigating the limitations associated with solvent casting in the preparation of biofilm involving microalgae, chitosan, and cellulose.

4.2 Extrusion

The basis for extrusion is the thermoplastic behavior of polymers that are heated above their glass-transition temperature and plasticized. This method is referred to as the “dry method” since it may function without water or any other solvent. However, plasticizers are required to increase the flexibility of the film. Plasticizers such as glycerol, polyethylene glycol, or sorbitol are frequently added through extrusion in amounts ranging from 10% to 50% w/w [59]. As the ingredients reached the kneading zone, the density, strain, and temperature of the mixture rose as well. Furthermore, a wide range of forms that are not achievable with the solvent casting method can manufactured with this method. This process modified the morphological structure of materials while enhancing the physicochemical characteristics of materials. Generally, three zones make up the process of extrusion: (i) the feeding zone; (ii) the kneading zone; and (iii) the heating zone at the machine’s final component or exit [60] as illustrated in Figure 3.

The process of making extruder-based biofilm involves both mechanical energy (specific mechanical) and heat energy (extruder barrel temperature) [61]. Several scholars have reported on the impact of screw speed in rotation per minute (rpm) on specific mechanical speed [62,63] The modification in screw speed during extrusion affects the features of starch-based edibles such as shear stress, shear rate, and homogeneity as well as the duration period, allowing for the addition and removal of additives for example agent reinforcement or stabilizers. A study by Nguyen *et al.,* [64] investigated the formation of thermoplastic from starch and alginate with reinforcing filler of spent coffee grounds and the results showed in reducing the water vapor permeability from (10.50 ± 1.07) $\times 10^{-13}$ to (9.03 ± 1.69) $\times 10^{-13}$ $g.s^{-1}m^{-2}$ Pa^{-1} and 67.6 ± 6.1 MPa for tensile strength which

concluded that these materials have a substantial potential for use in food packaging. The extrusion process depends on several variables that affect the end products, including the moisture content of the feed, diameter, pressure, energy input etc. Co-extrusion is a process that can be used to develop multi-layer films and provide flexibility in achieving the desired film qualities [60]. In addition to enhancing the characterization of produced film and capability, the multilayer also contributed to the design structural of the multilayer biofilm. Different physical or chemical properties can play a role in the development of various malformations in multilayer films. The optical and thermal performance may be hampered by these deformities. Significant physicochemical features have changed because of thermomechanical modifications made to the starch-based films during extrusion. Extrusion works best at the rpm of 80 to produce consistent thermoplastic starch-based in films and shapes although it also produces fractured starch materials more quickly at 40 and 120 rpm [65]. Thus, this method has been used extensively in industries to produce edible food packaging. Other than that, extrusion is frequently combined with additional processing techniques for example molding and thermos-pressing to produce the end products.

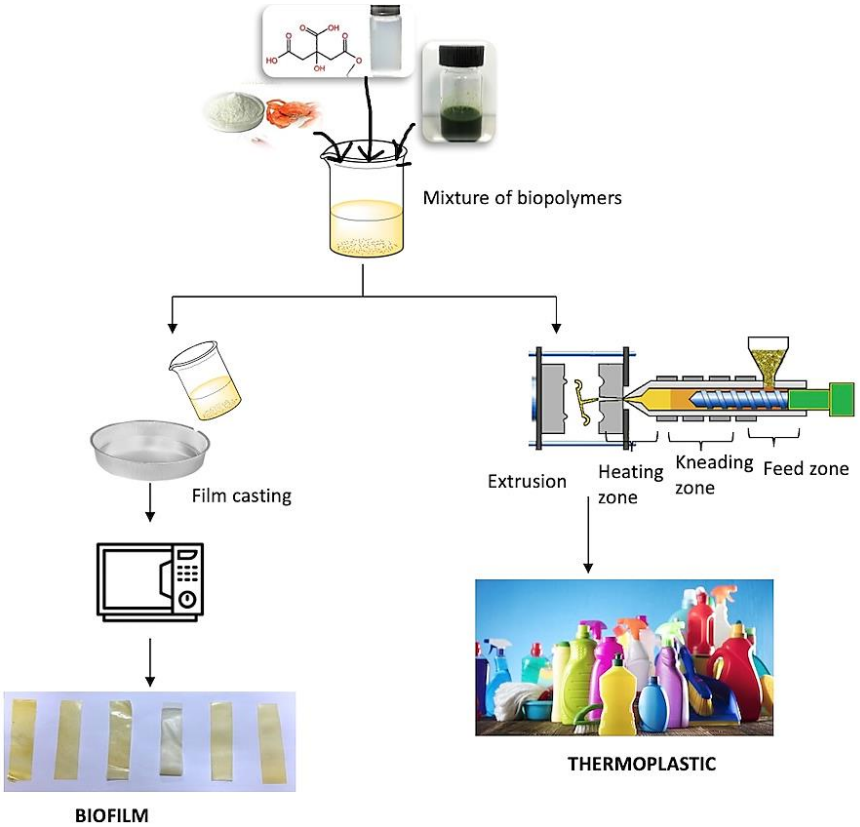


Fig. 3. The illustration of film formation by solvent-casting method (left) and extrusion process (right)

The primary benefits of the dry extrusion method film over the casting method are its shorter processing times and lower energy usage, as well as its improved mechanical and transparency properties, which include tensile strength, elongation, and optical, respectively. Other benefits include the absence of solvents, ease of handling the high viscosity polymers, a broad range of processing conditions from 0 to 500 bars of pressure and 70 to 500°C of temperature and improved control and degree of mixing [60]. In addition to being more effective at controlling the mechanical characteristics of edible food packaging, extrusion film production can produce a large variety of forms that the solvent casting method approach is unable to produce. The disadvantages of the

extrusion method were only a few polymers with low moisture content and temperature tolerance can be used under standard extrusion circumstances. Besides, the cost of using this procedure is further impacted by greater upfront and ongoing maintenance costs for specialized equipment. On the other hand, the potential challenges may include difficulties in achieving homogenous dispersion of the biopolymer components such as microalgae, cellulose, and chitosan, as well as maintaining their structural during the extrusion process. Additionally, the thermal sensitivity of some biopolymers such as cellulose may pose some constraints on processing parameters such as temperature and shear rate, impacting the quality and properties of the final bioplastic.

5. Challenges of Bioplastic Composite as Food Packaging

Currently, the rate of global population expansion is exceptionally high. Instead of growth rapid, it also surpasses the pace of production and the enhancement of societal well-being [66]. The effects of constant pressure on agriculture and marine resources to meet the demands of global food security have been detrimental, resulting in problems with biodiversity preservation, degradation of soil, the alarming destruction of natural resources, and rising global temperature [1]. Many strategies have been studied, one of them is packaging, which is essential to extending the shelf life of foods and keeping them fresher for longer [67]. Due to recent disturbing warnings about the harmful ecological implications of routine usage of conventional plastic packaging, the continued quest for biodegradable including edible packaging materials has received attention and increased by year [68]. However, leftover biomass from food processing could be utilized in order to avoid competing with food production. Industrial biowastes are constantly produced, and they have rich sources of biopolymers and other bioactive components [18]. Edible food packaging helps to preserve food quality, extend shelf life and provide nutritional value and sensory features to food products. For example, remarkably fresh food would need packaging with an efficient water barrier, whereas ready-to-eat food may be wrapped with highly water resistant films. Therefore, in order to have the most appropriate application of edible packaging, it is necessary to conduct studies on different formulations and evaluate their properties.

As this review has extensively covered edible packaging offers many advantages over conventional plastics, but there are still certain drawbacks that prevent its to be commercialization at this time. The limitations have included cost, production scale, allergic responses, moisture barrier problems, and religious restrictions on the use of animal-based films. For example, barrier concerns have been addressed by cross-linking/agent reinforcement (chemical, physical, enzymatic) blending with hydrophobic biopolymers, and using food byproducts to lower the cost of production [69,70]. Over the last decade, the compatibility of the hydrophobic polymer matrix and hydrophilic fibers have been the biggest obstacle in the production of bioplastic over the past decade. This led to the nonuniform dispersion of fibers within the matrix and poor mechanical properties respectively. Detailed knowledge of the molecular structure and interfacial interaction between the matrix and the fibers, as well as the link between structure and properties, would be a significant advancement in this research.

One intriguing option is microalgae-based bioplastics. Microalgae may be produced on nonarable soil, in wastewater or salty and can recycle valuable nutrients such as phosphorus and nitrogen in the agricultural system. This condition would significantly minimize the demand for mineral fertilizers. Strategies have been offered to reduce the total production costs, such as using wastewater for the cultivation of microalgae, which lowers the expense of nutrient and freshwater supply [71]. Presently, the focus of microalgae usage is on the bioproducts separately, which can produce a variety of products from a single microalgae source ought to be more profitable. For

example, the utilization of residual microalgae biomass after lipid extraction for the bioplastic composites might possibly lower overall the production of bioplastic-based microalgae by countering biodiesel production costs, given the microalgae lipids are utilized as an alternative for fossil fuels [72,73]. The removal of water and nutrients saves USD 2342.8 per hectare annually to produce 109 MT of microalgae [44]. A number of studies have shown a methodical technique to make bioplastic composites with a combination of non-biodegradable plastics and microalgae such as polyethylene (PE), polypropylene (PP), and polyvinyl chloride (PVC) [74,75]. However, only a few strains such as Spirulina and Chlorella have been studied and subsequent research ought to investigate distinct varieties of microalgae strains. The identification of more microalgae strains also helps microalgae-based bioplastics compete with bacterial-based bioplastics and even with conventional plastics.

Another idea to make greener bioplastic is from agricultural waste. This approach enables the production of market products while reducing waste volumes. Chitosan and cellulose are an excellent biopolymer that has been shown to increase the shelf life of fresh vegetables, seafood, and meat products. Other biopolymers such as carrageenan, starch, and alginate are blended with chitosan to form a biocomposite film and coating. Chitosan and cellulose matrices offer a good carried for active materials such as nanofillers, essential oils, fruit extracts, and other phytochemicals. As a biopolymer, chitosan extends the shelf life of a variety of foods and other agricultural products while also lowering the usage of conventional plastics and harmful additives that support a healthy environment and diet. Overall, the benefits and possibilities of bioplastics obtained from renewable sources such as edible crops and biowastes have been extensively studied. However, there is much more to investigate when it comes to exploiting the potential of microalgae in cellulose and chitosan for bioplastic production.

6. Case study of the Utilization of Microalgae-Based Bioplastics in Industrial

Products composed of plastics or polymers are essential to many aspects of modern society. Plastics generated from petroleum-based compounds, such as polypropylene (PP), polyethylene (PE), and polyvinyl chloride (PVC) are used to make the majority of commodity products that are currently on the market. Due to the widespread usage of these plastics over time, CO_2 emissions and the persistence of non-biodegradable elements in soil and water have a significant negative influence on the environment. Drinking cups made from bio-based polylactic acid (PLA) contribute to an annual global warming potential of 12.05 and 3.04 million tons of CO_2 equivalent, respectively, in comparison to cups made from polyethylene terephthalate and polypropylene cups [76]. Similar evaluation of animal/plant-based packaging materials is also required to raise consumer awareness and support in large-scale commercial manufacturing. According to the International Union of Pure and Applied Chemistry (IUPAC), any polymer sourced from biomass is classified as a bioplastic. The typical size of microalgae cells is around 50 m, allowing them to blend effectively with conventional plastics. Nannochloropsis sp. and Spirulina sp. mixed with corn starch were blended with plastic petroleum based by using a twin screw extruder to produce films and blends [77].

Zeller *et al.,* [78] conducted a study comparing the blending properties of Spirulina and Chlorella sp. with polyethylene. The authors found that Spirulina demonstrated superior blending properties at polyethene ratios of 50% and 65% compared to Chlorella. This is attributed to Spirulina's higher content of hydrophobic and non-polar amino acids. According to Shah *et al.,* [79], the tensile strength and elongation at break of Chlorella-polyvinyl alcohol biofilm prepared with ultrasonic homogenizer rise to 15.3 MPa and 99.3%, respectively. SEM and FTIR analysis revealed cross-linkages between polyvinyl alcohol (PVA) and Chlorella sp., resulting in fewer pores and smoother surface due to improvement in homogenization. Polyethylene with 10% Chlorella biomass demonstrated

mechanical properties of 22 MPa, in contrast to 20 MPa in the absence of microalgae biomass. Polypropylene was grafted to maleic anhydride at 180℃ for the bioplastic composite processing. Chlorella with polypropylene-maleic anhydride blend exhibited a tensile strength of 32 MPa. The properties of PVC-Chlorella composite material were investigated by Rahman *et al.,* [43]. PVC and Chlorella biomass were molded together in this investigation at an 80/20 ratio of PVC to Chlorella. The resultant bioplastic has a tensile strength of 20 MPa at a molding temperature of about 180℃ and a pressure of 4.4 MPa. Overall, it was evident that a great deal of research has been done on the varieties, benefits, and possibilities of bioplastic made from microalgae blends with conventional polymers as mentioned in Table 4. However, there is still needed to explore about producing bioplastics utilizing microalgae especially incorporation with cellulose and chitosan [80]. The field of microalgae-based bioplastics blends with multiple source biomass, especially chitosan and cellulose must continue to advance through research and advancement. Every stage of the research process is essential to ensure the best values of each property for bioplastic derived from microalgae, chitosan, and cellulose at the lowest possible cost. The incorporation of biowaste byproducts into bioplastics may increase the biodegradation in soil for these composite films [81]. As a result, more research is required to determine the efficacy of the novel bioplastic film in real-world food packaging applications.

Table 4
Summary of strains studied in microalgae blend with conventional polymer

Biomass strain	Products	Additional materials added	Tensile strength (MPa)	References
Spirulina sp.	Bioplastic and thermoplastic	Polyethylene + glycerol	8.5	[78]
Chlorella sp.	Bioplastic	Polyvinyl alcohol	35.1 ± 4.3	[79]
Chlorella sp.	Bioplastic and thermoplastic blend	Polypropylene + maleic anhydride	32	[43]
Chlorella sp.	Bioplastic and thermoplastic blend	Polyvinyl chloride	30	[43]
Chlorella sp.	Bioplastic and thermoplastic blend	None	22	[43]

7. Conclusions

Biopolymers-based byproducts are renewable, low-cost, and biodegradable raw materials generated from biomass of non-edible resources wastes such as rice, wheat, corn, microalgae, legumes, crustaceans, and wood plants. This review leads to the conclusion that food packaging materials can improve the properties and structure by the addition of these biopolymers-based byproducts. The innovations for edible food packaging based on residual biomass might contribute to enhancing environmental sustainability and food security by minimizing the use of food resources and limiting the widespread use of persistent fossil fuel-based plastics. The presence of polysaccharide byproducts derived from chitosan and microalgae was the main factor in the pH-sensitive, antioxidant, and antibacterial properties. Nevertheless, the intermolecular interactions between the hydroxyl groups of microalgae and amine groups of the chitosan that constitute the films impeded the mechanical and barrier properties of these films. Plant byproducts derived from woods such as cellulose showed the potential reinforcing capabilities to enhance the barrier and optical characteristics of the films because of the content of high lignin and hemicellulose in the woods. Ultimately, these innovative biopolymer-based byproducts such as chitosan, cellulose, and microalgae have the potential to be transformed into sustainable, inexpensive, biodegradable, and food packaging options.

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